DEVELOPMENT OF INFLATABLE AND BACKUP SEALS FOR SODIUM COOLED FAST BREEDER REACTORS: AN APPROACH FOR UNIFICATION AND STANDARDISATION OF ELASTOMERIC SEALING

By

N. K. SINHA

Enrolment No. ENGG02200804005

Indira Gandhi Centre for Atomic Research, Kalpakkam

A thesis submitted to the

Board of Studies in Engineering Sciences

In Partial fulfillment of requirements

for the Degree of

DOCTOR OFPHILOSOPHY

of

HOMI BHABHA NATIONAL INSTITUTE

February, 2016

Homi Bhabha National Institute

Recommendations of the Viva Voce Committee

As members of the Viva Voce Committee, we certify that we have read the dissertation prepared by N. K. Sinha entitled "Development of inflatable and backup seals for Sodium Cooled Fast Breeder Reactors: An approach for unification and standardisation of elastomeric sealing" and recommend that it may be accepted as fulfilling the thesis requirement for the award of Degree of Doctor of Philosophy.

M. Sai Baba 11-11-16

Chairman – Prof. M. Sai Baba Date:

Guide / Convener – Prof. P. R. Vasudeva Rao Date: 11/11/16

Co-guide ---- Date:

Examiner – Prof. Anil K. Bhowmik Date: 11/11/2016

Member 1- Prof. A. K. Bhaduri Date: 11/11/2016

Member 2- Prof. T. Jayakumar (Retd.) Date: 11/11/2016

Final approval and acceptance of this thesis is contingent upon the candidate's submission of the final copies of the thesis to HBNI.

I/We hereby certify that I/we have read this thesis prepared under my/our direction and recommend that it may be accepted as fulfilling the thesis requirement.

Date:

Place:

11/11

Co-guide (if applicable) **Guide**

STATEMENT BY AUTHOR

This dissertation has been submitted in partial fulfillment of requirements for an advanced degree at Homi Bhabha National Institute (HBNI) and is deposited in the Library to be made available to borrowers under rules of the HBNI.

Brief quotations from this dissertation are allowable without special permission, provided that accurate acknowledgement of source is made. Requests for permission for extended quotation from or reproduction of this manuscript in whole or in part may be granted by the Competent Authority of HBNI when in his or her judgment the proposed use of the material is in the interests of scholarship. In all other instances, however, permission must be obtained from the author.

28/10/16
N. K. Sinha

DECLARATION

I, hereby declare that the investigation presented in the thesis has been carried out by me. The work is original and has not been submitted earlier as a whole or in part for a degree / diploma at this or any other Institution / University.

28/10/16
N. K. Sinha

List of Publications arising from the thesis

Journal

1. Deposition of thick and adherent Teflon-like coating on industrial scale stainless steel shell using pulsed DC and RF PECVD, A. Satyaprasad, S. K. Nema, N. K. Sinha, Baldev Raj, *J. Appl. Surf. Sci.*, **April 2010**,*256/13,* 4334-4338.

2. Development of polymeric applications for sodium cooled fast breeder reactors: chronicles of inception, progress and achievements, N. K. Sinha, Baldev Raj, *Nucl. Eng. Des.* **October2010**, *240/10*, 2925-2947.

3. Optimization of material and production to develop fluoroelastomer inflatable seals for sodium cooled fast breeder reactor, N. K. Sinha, Baldev Raj, *Nucl. Eng. Des.* **March 2011**, *241/3*, 739-751.

4. Development of fluoroelastomer backup seal: snapshots of a novel effort in rubber engineering for a critical application (Part I), N. K. Sinha, Baldev Raj, R. Mukhopadhyay, *Sealing Technol.* **April 2011**, *2011/4*, 10-13.

5. Development of fluoroelastomer backup seal: snapshots of a novel effort in rubber engineering for a critical application (Part II), N. K. Sinha, Baldev Raj, R. Mukhopadhyay, *Sealing Technol.* **May 2011**, *2011/5*, 9-12.

6. Development of fluorocarbon rubber for backup seals of sodium cooled fast breeder reactor, N. K. Sinha, R. Mukhopadhyay, D. Dhupia, S. Das Gupta, Baldev Raj, *J. Mater. Des.* **December 2011**, *32/10*, 5141-5153.

7. Unification of reactor elastomeric sealing based on material, N. K. Sinha, Baldev Raj, *Nucl. Eng. Des.* **February 2012**, *243*, 404-418.

8. Basis of property limits for inflatable seal fluoroelastomer, N. K. Sinha, Baldev Raj, *Nucl. Eng. Des.* **March 2012**, *244*, 100-108.

9. Design optimization of backup seal for sodium cooled fast breeder reactor, N. K. Sinha, P. Ghosh, A. Saha, R. Mukhopadhyay, Baldev Raj, S. C. Chetal, *J. Mater. Des.* **April 2012**, *244*, 396-407.

10. Mechanical behavior of fluoroelastomer considering long term ageing, N. K. Sinha, R. Mukhopadhyay, Baldev Raj, *Nucl. Eng. Des.* **January 2013**, *254*, 89-96.

11. Development of inflatable seals for the rotatable plugs of sodium cooled fast breeder reactors: a review Part I- Key areas, N. K. Sinha, Baldev Raj, *Kerntechnik* **November 2013**, *78/5*, 380-399.

12. Development of inflatable seals for the rotatable plugs of sodium cooled fast breeder reactors: a review Part II- R & D necessities and development across the world, N. K. Sinha, Baldev Raj, *Kerntechnik* **December 2013**, *78/6*, 463-476.

13. Leakage limits for inflatable seals of sodium cooled fast breeder reactor, N. K. Sinha, Baldev Raj, *Nucl. Eng. Des.* **January 2014**, *266*, 194-198.

14. Choice of rotatable plug seals for prototype fast breeder reactor: Review of historical perspectives, N. K. Sinha, Baldev Raj, *Nucl. Eng. Des.* **September 2015**, *291*, 109-132.

Chapters in books and lectures notes

1. N. K. Sinha: Design, development, production, validation and performance of organic components to ensure safety of nuclear applications: a finite element analysis perspective, In: S P. Dharne, S. Hajela, R. R. Sahaya (Eds.), Thematic Volume Evolved from *(Selected works of)* Peacefulatom 2009: Reactor Systems and Applications, BARC, DAE, Mumbai, January 2011, pp. 131-141.

2. N. K. Sinha, S. C. Chetal, Baldev Raj: Simplification and unification of elastomeric sealing for fast breeder reactors based on Fluorohydrocarbon rubber. In: Mendez- Vilas, A. (Ed.), Fuelling the Future: Advances in Science & Technology for Energy Generation, Transmission and Storage, Brown Walker Press, Boca Raton, 2012, pp. 445-451. Selected works from accepted abstracts and presentations made in, 'The Energy and Materials Research Conference (EMR 2012)', held at Torremolinos, Spain during June 20-22, 2012

Conferences

1. N. K. Sinha: Design, development, production, validation and performance of organic components to ensure safety of nuclear applications: a finite element analysis perspective, in: Proc. Int. Conf. on Peaceful Uses of Atomic Energy- 2009 (Peacefulatom 2009), Vol. 1, BARC, DAE, Trombay, 2009, pp. 186-187.

2. N. K. Sinha, S. C. Chetal, Bladev Raj: Simplification of elastomeric sealing of fast breeder reactors based on a few material formulations, analysis procedures, coating technique and manufacturing processes. Book of Abstracts, The Energy Materials Research Conference – EMR 2012, Torremolinos, Spain, June 20-22, 2012, (June, 2012) p. 303.

3. N. K. Sinha, S. C. Chetal, Bladev Raj: Unification of elastomeric sealing applications of fast breeder reactors based on peroxide cured fluorohydrocarbon rubbers. Book of Abstracts, EMR 2012, 2012, (June, 2012) p. 306.

4. N. K. Sinha, S. Suresh Kumar, A. Babu, Baldev Raj: Quality control strategy for critical elastomeric seals of sodium cooled fast breeder reactor. Book of Abstracts, International Conference on Rubber and Rubber- like Materials (ICRRM 2013) – IIT Kharagpur, West Bengal, March 6-9 2013, (March, 2013) pp. 169-170.

Others

1. **Presentation-International Conference:** N. K. Sinha: Design, development, production, validation and performance of organic components to ensure safety of nuclear applications: a finite element analysis perspective. *Presentation at Peaceful Uses of Atomic Energy – 2009 (Peacefulatom 2009), New Delhi, September 29- October 1, 2009.*

2. **International Magazine:** N. K. Sinha, Baldev Raj: PFBR backup seal development, Nucl. Eng. Int. March (2011) 36-38.

3. **Presentation- International Conference:** N. K. Sinha, S.C. Chetal, Baldev Raj: Simplification of elastomeric sealing of fast breeder reactors based on a few material formulations, analysis procedures, coating technique and manufacturing processes. *Presentation at The Energy Materials Research Conference – EMR 2012, Torremolinos, Spain, June 20- 22, 2012.*

4. **Presentation- International Conference:** N. K. Sinha, S. Suresh Kumar, A. Babu, Baldev Raj: Quality control strategy for critical elastomeric seals of sodium cooled fast breeder reactor. (Posters I and II). *Posters presented at International Conference on Rubber and Rubber- like Materials (ICRRM 2013), IIT Kharagpur, West Bengal, March 6- 9, 2013.*

28/10/16
N. K. Sinha

DEDICATIONS

Dedicated to my wife

Late Mrs. Krishna Sinha

and

my Parents

ACKNOWLEDGEMENTS

This research from Fast Reactor Technology Group (FRTG) of Indira Gandhi Centre for Atomic Research (IGCAR) is a result of theoretical, experimental and numerical studies carried out at various organisations (apart from IGCAR) in continuation of the program of *Special Elastomeric Component* development which makes it difficult to acknowledge a large number of individuals separately, involved in various roles and capacities.

This quest owes its inception and progress to a large extent to my former Guide Dr. Baldev Raj (*Former Director, IGCAR*) without whose encouragement and continuing support till now, the search would never have been materialised. Dr. G. Vaidyanathan (*Former Director, FRTG)* provided the initial administrative impetus and the drive to carry forward an endeavour of multidisciplinary nature, continued by Dr. P. Kalyanasundaram (*Former Director, FRTG)*, Dr. K. K. Rajan (*Former Director, FRTG)* and Shri P. Selvaraj (Director, FRTG). I am indebted to the previous and the present Doctoral Committee on the whole and to Dr. K. S. Viswanathan (*Former Dean, HBNI*), Dr, T, Jayakumar (*Former Director, MMG, IGCAR*), Dr. M. Sai Baba (AD, HRD, IGCAR) and Dr. A. K. Bhaduri (Director, IGCAR) in particular for rich interactions at various times. Special thanks to Dr. P. Chellapandi (*Former CMD, BHAVINI*) with whom there were enlightening technical exchanges which inspired the thinking to no end. There has been priceless administrative support with the necessary interpretations from Dr. G. Sasikala (Dean Academic, Engineering Science, HBNI, IGCAR) during the times of delays in thesis completion due to health reasons. Finally, deep sense of gratitude to my present Guide Dr. P. R, Vasudeva Rao (*Former Director, IGCAR*) who motivated the shaping of thesis to this form with pointed queries, insightful questions, constructive criticisms and enriching discussions amidst very busy schedule.

I do owe this work also to the broader realm of my family and friends, my daughters Ms. Koel Sinha and Ms. Urmika Sinha in particular.

I take this opportunity to express sincere gratefulness to the following for the profound support and encouragements received in countless forms.

Inspirations and support: Late Dr. Placid Rodriguez, Former Director, IGCAR; Shri S. B. Bhoje, Former Director, IGCAR; Dr. Anil Kakodkar, Former Chairman AEC.

Colleagues, early years of development (IGCAR): Late Shri K. V. Kasiviswanathan, Shri R. Veerasamy and Shri Sulekh Chander:

Invaluable discussions (IGCAR): Shri S. C. Chetal, Shri P. V. Kumar, Dr. S. L. Mannan.

Colleagues, later years (IGCAR): Shri A. Babu, Shri Jose Varghese, Shri Ramachandra Aithal, Shri R. Veerasamy, Shri S. Raghupathy, Shri S. Suresh Kumar, Dr. S. Venugopal, Shri Utpal Borah.

Collaborating Institutes & Individuals: Prof A. K. Bhowmik, Former Head, RTC, IIT Kharagpur; Dr. C. R. Jagga, Former Professor, ITMMEC, IIT Delhi; Shri G. S. Anand, MD, ASP Sealing Products Ltd., New Delhi.; Prof. K. K. Kar, Dept. of Mechanical Engineering, IIT Kanpur.; Prof. N. Sivaprasad, Dept. of Mechanical Engineering, IIT Madras.; Prof. P.I. John, Former Meghnad Saha Chair Professor, IPR, Gandhinagar.; Prof. K. Kaw, Former Director, IPR; Dr. R. K. Singh, Former Additional Director, DMSRDE, Kanpur.; Dr. R. Mukhopadhyay, Director & CEO, HASETRI, Rajasthan.; Dr. S. Mukherjee, Head, FCIPT/IPR, Gandhinagar; Dr. S. K. Nema, FCIPT/IPR, Gandhinagar; Shri S. Singha Roy, Former Maint. Supdt., MAPS, Kalpakkam and Former PD, Jaitapur, NPCIL; Prof. S. G. Dhande, Former Head, CAD Lab., Director, IIT Kanpur.

28/10/16
N. K. Sinha

CONTENTS

Synopsis

Climate change effects from GHG emission is shaping wide measures of decarbonisation in the energy sector, responsible for about two- third of anthropogenic emission across the globe. The aim of confining the average global temperature rise with respect to the pre- industrial age to 2 ^{0}C by the end of this century necessitated key reforms in the power sector where growth of variable renewable power source (solar, wind, hydro, bio) and the steady baseload generation capacity of nuclear is expected to play complimentary roles. The projected global growth of nuclear to 930 GWe by 2050 is a downscaling from the earlier growth estimation because of Fukusima Daiichi accident on March 11, 2011 which has brought in safety at the forefront with life extension of plants to 60 y and economy of NOAK plant in long term as the basic objectives of sustainability. The pre- 2050s scenario is expected to be dominated by Gen III LWRs, being refined in an evolutionary fashion towards the intended objectives, which are likely to give way to the Gen IV SFRs during the latter part of this century in order to maximise the utilisation of uranium fuel (by recycling and closed fuel cycle) to 80% from 1% in the PHWRs for energy security and sustainability. Nuclear power is expected to see significant growth in India, China, USA, Middle East, South Africa, Poland, Turkey, Republic of Korea, UK, Russia in attaining the objective during the coming decades. Design simplification and standardisation along with harmonisation of codes and standards have been a common measure towards maximising safety, reliability and economy for sustainability. Sealing of NPPs preventing release of hazardous activity and fluids is an important, common design element where simplification and standardisation could bring in far reaching effects. This discourse addresses simplification, unification and standardisation of critical elastomeric sealing for FBRs (transiting towards SFRs) by taking material as cornerstone, FEA as facilitator and design as an all-encompassing base with cover gas seals as the representative for critical elastomeric sealing and the 500 MWe INDIAN- SFR (being readied for criticality) as the presentative model for MOX fuelled FBRs. The approach is subsequently extended to other Indian nuclear genres such as metallic fuelled FBRs, PHWRs and AHWR with a potential to encompass various reactor designs across the globe.

The whole research, covered through 8 Chapters in this thesis, has been carried out by combining literature review, data extrapolation (to minimise experiment and analysis), experiments, analysis and design where the past developments on *Special Elastomeric Component* with fluoroelastomer inflatable and backup seals played pivotal roles. T_s, E_b, tear strength, CS and potentially harmful volatile release from fluoroelastomers (such as HF) have been used as the common material failure parameters (contribution towards the seal failure parameters of leakage, 10^{-3} scc/s/m-seal-length, and friction, 1000 N/m-seal-length, excepting HF release) with a FOS based novel approach (on T_s and E_b) used to assess failure by synergistic ageing along with CS. The uniqueness of elastomers which make FEA based seal sizing, manufacture, coating etc. largely compound specific has been utilised to arrive at a streamlined and effective scheme of simplification, unification and standardisation which runs as a common thread throughout this dissertation in parallel with design. Past developments of thermal and fast reactors indicated necessity of putting the seals on a common foundation with minimisation of number of elastomeric compounds and using high temperature elastomers for life maximisation as temperature determines other ageing degradation rates from radiation, oxygen etc. However, the material-sizing-manufacture-coating correlations, failure parameters and FOS based approach for assessment of synergistic ageing, concerted use of predictive techniques (FEA, Arrhenius and WLF) and data extrapolation for minimisation of experiments and analysis adopted in this thesis towards arriving at a scheme of 1 synchronised replacement of cover gas elastomeric seals during reactor life of 60 y is new in international context

The cover gas elastomeric seals of INDIAN- SFR was simplified by classification based on size or manufacturing process (large, > 0.5 m, extrusion; small, ≤ 0.5 m, moulding) and further by geometry and design (O- ring, V-ring, Lip seal) to arrive at 8 representative categories along with INDIAN- SFR RP inflatable seal as the 9th special category. Performance of inflatable seal is not governed by CS based failure. Classification and categorisation were carried out till there is no more envisaged change of seal elastomeric formulation with category. The approach was to identify 1 representative seal for each category (operating under the most demanding requirements) and identify 1 corresponding representative elastomeric formulation (defined by material specification) which is applicable to all other seals in the category. Simplification, unification and standardisation was carried out, for minimum R & D effort and maximum safety as well as reliability (along with economy) towards 1 synchronised replacement in 60 y, by exploiting the uniqueness of elastomer where unification by standardisation of 6 complimentary modules (i.e. FEA based seal sizing, manufacture, coating, FEA based life assessment, life assessment by Arrhenius and WLF, Quality control) are seal compound specific with all the standardised 7 modules (material as the 1st module) contributing towards R & D (the 8th module) standardisation. Founding the entire application of ~ 700 m long elastomeric cover gas sealing of INDIAN- SFR (as model) on a single generic elastomer was therefore the first step. The findings from end- 1970s to the early phase of the first decade of new millennium that γ radiation dose (in isolation) is inconsequential in influencing the performance of cover gas seals and thermal ageing is the determining criterion for failure of nuclear elastomeric sealing applications indicated that one of the high temperature elastomers (maximum temperature rating ≥ 175 ^{0}C) should be chosen as the common generic elastomer for all the cover gas seals. Experience in developing elastomeric cover gas sealing in general and inflatable seals of FBR RPs in specific from 1950s to 1990s indicated that this choice should be from MQ or FKM rubbers capable of continuous operation at > 200 ^{0}C. The development of bisphenol cured, Viton® A-401C based compound (S- 1) for the inflatable seal of INDIAN- SFR RPs with 10 y of life *(prior to this research)*, amenable to hot feed extrusion and autoclave cure, limitations of MQ rubber (highest permeability amongst elastomers and one of the lowest RT strengths which accounted for a large part of international R & D for FBR RP inflatable seals) resulted in the choice of FKM rubber as the common generic elastomer for the cover gas seals of INDIAN- SFR, used as a model.

Abstracting the functioning of INDIAN- SFR cover gas seals by one set of operating requirements (10y- 70kPa- 120 ^{0}C- 23 mGy/h- 100km- 1000 N/m-seal-length, 10^{-3} scc/s/m-seal-length), with common sets of materials (carbon steel, SS, aluminium- bronze) and fluids (air with CO_2, ozone and moisture; argon, sodium aerosol, xenon, krypton) in contact, was used as the basis for identifying a common FKM rubber formulation across categories. INDIAN- SFR represents MOX fuelled FBRs in the capacity of cover gas seals because of the proximity of its representative conditions to the maximum of other MOX fuelled FBRs (10 y ---- 100 kPa ---- 120 ^{0}C ---- 0.1 Gy/ h, along with the materials and fluids in contact), built till date. Total debarring of liquid lubricants in the cover gas seals to avoid positive reactivity coefficient of voids (post- Fukusima approach in particular) is another key area of commonness in unifying material, FEA procedures, manufacture and friction- minimisation means (such as coating). The combination of inflatable and backup seals in the INDIAN- SFR RPs is most important and hence represents the dynamic and static classes of cover gas sealing because of the proximity of design operating requirements (inflatable- backup: 10y- 10y, 25kPa- 25 kPa, 120 ^{0}C- 110 ^{0}C, 23 mGy/h- 23 mGy/h, 100km- 0 km, 1000 N/m-seal-length- 0 N/m-seal-length, 10^{-3} scc/s/m-seal-length- 10^{-3} scc/s/m-seal-length) to those of the abstracted conditions. Backup seal (secondary barrier) is not exposed to mist and does not wear because of static operation. In addition, inflatable seal as primary barrier in INDIAN- SFR RPs influence reactor economy (minimisation of RP diameter and height), safety (reducing moment and stress, improving alignment convenience and accuracy of ARDM and IVTM with minimum size), reliability, operability and life (enhanced life with reduced stress) in one of the most profound ways as replacement involves shutting down of the 500 MWe reactor for at least 4 weeks. Largest surface area to volume ratio of the thin-walled (2 mm) primary inflatable seal (in direct contact with radioactive argon cover gas laden with mist in a hanging plug), coupled with tensile stress field during static and dynamic operations and a number of potential stress raisers in the design, poses maximum demand on resistance to synergistic ageing (temperature + radiation + mist + air) and cracking of seal material due to vulnerability of elastomers to low cycle fatigue by rubbing during intermittent rotations. This implies that unification by standardisation should start with maximising the life of inflatable seal elastomer and then tailoring the formulation for simultaneous maximum life for other cover gas seals. Independence from CS based failure means that the FOS or the margin of safety on unaged T_s and E_b of inflatable seal material (determined at design temperature of 120

^{0}C) should be maximised. Principal- tensile- true stress- strain field in seals exceeding T_s and E_b (determined at design seal temperature) is taken as the inception of cracking failure and leakage therefrom. The FOS accounts for deviations from batch-to-batch material property variations, inter/intra- laboratory measurements and moulded specimens to extruded seals transition along with increased stress- strain from pressure surge (2.1 MPa) during CDA in certain cases, such as backup seal. Possible erosion of initial FOS due to synergistic ageing and enhanced ageing rates from strain is the other complimentary half. Data extrapolation indicated that life of S- 1 and the INDIAN- SFR RP inflatable seal made of it is limited to 10 y with a FOS of 2 as synergistic ageing effects from radiation beyond 2 kGy could bring down the E_b outside of elastomeric behaviour zone. With a FOS of 2 the limiting principal stress and strain for S- 1 at 120 ^{0}C are 1.63 MPa and 40%. Maximising FOS with an improved fluoroelastomer grade and different extrusion technology for better quality seals was the key to the whole process along with the bisphenol cured, Viton®A-401C based compound (S-4) developed for backup seal, supported by its sizing and life assessment by FEA, Arrhenius and WLF methodologies which form core of this thesis. The approach was to standardise each of the 7 modules for unification starting with material which provided the ingredients for standardisation of the 8th module i.e. R & D.

A minimum life of 10 y was ascertained for S- 4 (obtained by tailoring S-1 for static use with much lower filler content) and backup seal made of it by using Arrhenius and WLF methodologies on accelerated aged specimens in air (32 weeks, 140/170/200 ^{0}C) with a CS of 80% as failure criteria. This was consistent with the life obtained by using parallel failure modes based on T_s and E_b using a FOS of 2 which defined the limiting principal stress (0.7 MPa) and strain (42.5%) in seal at a design seal operating temperature of 110 ^{0}C. Synergistic ageing studies by data extrapolation indicated that outstanding thermal stability of fluoroelastomer allows the FOS to be preserved for additional degradation by strain and CDA pressure surge at any point of time during design life. An interim unification scheme indicated that S- 1 and S-4 and its 2 variations (corresponding to $\leq$ 0.5 m diameter moulded static and dynamic seals) could cover most of the cover gas sealing applications.

Assessment of life using another failure mode (i.e. HF release during seal operation in reactor) by data extrapolation showed consistent results with 10 y of achievable life for S-1 and S-4 considering γ dose induced volatile release (operating temperature induced release nil for any duration) which indicated achievability of standardisation and unification of the INDIAN- SFR cover gas seals for 10 y of life by S-1, S-4 and their variations. The change of INDIAN- SFR RP inflatable seal elastomeric formulations to peroxide cured, 50:50 blend of Viton® GBL200S and 600S made of APA (S- 2) based on production of ~ 2m diameter test seals through experiments in a commercial cold feed extruder with continuous, pressureless cure (by MW and HAT) resulted in a material paradigm shift which improved seal life by maximising the FOS to 4 and above. Cold feed extrusion and continuous cure in addition provides improved seal with one end joint (per seal) and better uniformity of cure as well as properties along seal circumference with provisions for process-automation (by gear pump, online feedback etc.) which could ensure performance- reproducibility across production batches. Conventional fluoroelastomers (such as bisphenol cured S- 1 and S- 4) derive a large part of their RT strength from ionic interactions which are lost at elevated operating temperature resulting in two- third to three- fourth drop in T_s and E_b on average. The peroxide cured, 50:50 blend of S- 2 made of APA significantly enhances strength- elongation retention at elevated temperature (while reducing modulus) because of minimal ionic interactions at RT which increases the unaged FOS beyond 4. Compatibility with mist and resistance to permeation are better in this elastomer compared to the bisphenol cured counterparts. The increase in FOS to beyond 6 considering the usual, lower operating temperature of inflatable (100 ^{0}C) and backup (90 ^{0}C) seals takes the radiation resistance to 10 kGy or more under synergistic ageing which implies 50 y of life ideally (synergistic ageing – mist – strain – CDA) and feasibility of 1 replacement during reactor life of 60 y by conservative assumptions of design in consideration of various overall reactor- boundary conditions. Shaping of other design elements in parallel for standardisation of the individual and combining them to a totally unified scheme take a form of simplification- unification- standardisation at the end of this research which is described in subsequent paragraphs. Backup seal made of S- 4 and inflatable seal designed by FEA (depicted in this discourse) withstood various pre-commissioning conditions in the INDIAN- SFR for the past 6 y which (along with the scaled- rig tests) validates the constituting elements of simplification, unification and standardisation.

The aim of synchronised cover gas seal replacement (1 in 60 y) for MOX fuelled Indian FBRs (transiting towards SFRs) represented by INDIAN- SFR could be realised by standardising the material module with peroxide cured, FKM compound S- 2 made of APA (INDIAN- SFR RP inflatable seal compound) and its 4 main variations (2 for static and dynamic sealing, each) without changing the blend ratio (50:50) of Viton® GBL 200S and 600S. This is facilitated by FEA based sizing, life assessment and design (using T_s and E_b with a FOS) with a uniform approach of plane-strain (initial approximation) and axisymmetric (finalisation) conditions and Mooney- Rivlin material model (normal operation) for normal operating conditions and 3- term Mooney- Rivlin as well as Ogden constitutive relations for special sealing as well as accidental conditions involving strain in excess of 50 %. Arrhenius and WLF methodologies complete the circle for life assessment and design using CS based failure criterion (specimens under strain) in a complimentary fashion which does also provide a mode of cross- verifications. The manufacturing module is similarly standardised with 2 processes (cold feed extrusion and continuous cure; injection moulding) for small ($\leq$ 0.5 m) and large (> 0.5 m) diameter cover gas seals with dedicated extrusion and moulding machines (1 each), supported by 2 PECVD based Teflon- like coating procedures corresponding to large- small diameter cover gas seals with 2 limits of adhesion strength ($\geq$ 4.5- 7 MPa) and thickness (5- 10/ 3- 5μm) requirements. The standardised quality control scheme (along with the standards) arrived based on development of inflatable and backup seals corresponding to large diameter, extruded dynamic and static barriers respectively (supported by the material and coating specifications) provide foundation for future implementation of unification by tailoring S- 2 to 4 major variations for the cover gas seals. Future R & D standardisation comprises of the 7 standardised elements of design for implementation along with an approach of design options as well as R & D minimisation (prior to implementation of the 7 modules) based on survey, applicable to all the cover gas seals. The whole scheme of standardisation and unification provides a standardised design framework for the cover gas seals of Indian and global SFRs alike for sustainability which could result in a design code while working in smooth interfacing with the commercial FEA codes such as ABAQUS, ANSYS and MARC.

This scheme of simplification, unification and standardisation is applicable equally to metallic fuelled FBRs as operating requirements of their cover gas seals are marginally different. The post- Fukusima safety scenario demanded renewed emphasis on BDBE with qualification of severe accident mitigation systems and emergency preparedness for severe accident management (decay heat, core degradation, sodium- water reaction) at the fore, in particular from multiple external events (flood, earthquake etc.) at multi- plant sites. The current Gen IV SFR safety philosophy of making CDA from core meltdown an impossibility by innovative design and then to ensure that energy release is minimum even if the impossibility occurs, has generated room for elastomeric cover gas barriers to exploit their advantages (simplicity, economy, compactness, resilience, low contact pressure, low or zero assembly load, surface conforming capacity, tolerance absorbing ability etc.) along with provisions for accessibility, maintainability and replacement of components while retaining the advantage of welded joints with elastomers of lowest permeability. This approach has similarity with that during the latter half of EFR conceptualisation (1988- 1993), carried out as part of Western European (WE) collaboration between 6 nations (Belgium, France, Germany, Italy, Netherlands and UK). The usage of fluoroelastomer (with one of the lowest permeability amongst elastomers) as cornerstone in the present scheme creates the provision of extending simplification- unification- standardisation to MOX and metallic fuelled, Indian and global FBRs as well as SFRs alike.

The APA FKM formulation based on a blend of peroxide cured Viton® GBL 200S and 600S has natural applicability for unifying the PHWR elastomeric barriers by varying the blend ratio (from the 50:50 for FBRs), operating at a maximum temperature, pressure and γ radiation range of 200 ^{0}C, 10 MPa and 10- 100 kGy with usual replacement span varying from 3 months to 5 y. Higher operating temperature (300 ^{0}C) of the critical elastomeric sealing for AHWR expands the material selection further to grades of perfluoroelastomers with silicone rubber as an option. EPDM with lower maximum temperature rating (149 ^{0}C) compared to MQ and FKM rubbers (> 200 ^{0}C) was the most popular generic elastomer during the decades of 1970s to 1990s (followed by fluoroelastomers)- primarily because of compatibility with hot water and steam. Earlier generation fluoroelastomers fell short on this count as unused metal oxide and hydroxide and unsaturation (double bonds) in the bisphenol cured product make them vulnerable to polar fluids with propensities of macromolecular chain scission (softening) and crosslinking (embrittlement) from various counts, accompanied by release of harmful volatiles such as HF. Peroxide cured fluoroelastomers made of APA has removed this limitation resulting in applicability and unification in the domain of thermal reactors. A closer look at the operating requirements of INDIAN- SFR, PHWR and AHWR provides an indication that a variety of 10- 12 well characterised compounds (mostly based on peroxide cured blend of APA fluoroelastomers) could encompass the entire panorama of critical elastomeric sealing applications of Indian nuclear genre.

Complete nuclear characterisation of S-2 and its 4 variations comprising of mist compatibility in long term, permeability, processability (curing characteristics and rheological behaviour), extrudability, tribology, determination of physico- mechanical properties (stress- strain, modulus, T_s, E_b, hardness, CS etc.at various strain rates and specimen conditioning, RT and elevated operating temperatures) on unaged, short- term aged and long- term aged (at least 100 weeks, up to 225 ^{0}C) specimens along with sizing, life assessment and design by FEA, Arrhenius and WLF methodologies form the essential gamut of future work ideally. The unchanged blend ratio (50:50) in S-2 and its 4 variations for the cover gas seals of FBRs transiting towards SFRs indicates similar FOS, fluid compatibility and processability which signify that choosing 2-3 representative compounds out of the 5 from static and dynamic domains (including S-2) for complete characterisation may be sufficient. Similarly, representative seals could be chosen from 5 categories (extruded large diameter static O- rings, moulded small diameter dynamic O- rings, moulded small diameter dynamic V-ring/lip seals, large diameter extruded dynamic inflatable seal and large diameter extruded static backup seal, derived from the initial 9 categories) for sizing, design and life assessment implementations by FEA, Arrhenius and WLF methodologies using the qualified and designated compounds. Performance validation of the representative seals in scaled down test rig with provisions for accelerated ageing (up to 100 weeks and 225 ^{0}C) of test seals is an essential part of the whole process. Assessment of the potential effects of cumulative release of HF (induced by γ dose) from the cover gas seals on reactor performance in long term forms an important part of future research, as also the determination of strain effects on ageing and that of transition from moulded specimens to moulded/ extruded products by using specimens extracted from seals. Attaining the aim of 1 synchronised replacement of cover gas seals in reactor life of 60 y is influenced significantly by another key failure parameter i.e. CS. This research shows that attaining 1 replacement in 60 y based on FOS maximisation could be a trivial issue if the cumulative effect of HF release on long-term reactor performance is ascertained. Demonstration of 10 y of life based on a CS threshold of 80% and several improvement suggestions on this count in tailoring of S- 2 notwithstanding, there is significant scope in future research to maximise CS resistance further (by finer compositional variations in the 50:50 blend) towards attainment of 1 replacement in 60 y. Feasibility of this approach is substantiated by the operating records of FBTR which indicates 15- 20y of life for FKM and MQ static O – rings. Production and installation of inflatable seals (made of S- 2) with PECVD Teflon- like coating in INDIAN- SFR RPs, R & D completion on PECVD Teflon- like coating for the inflatable- seal- mating- steel- shells and development of adhesion-less end joining as well as automated glue dispensation technique are some of the future research.

Demonstration of the possible effects of low molecular weight volatile (such as HF) release induced by γ dose during long- term reactor operation is key to the implementation of unification in the PHWR domain along with validation of extrusion resistance of the seals. For AHWR the future research could be maximising the temperature rating of peroxide cured perfluoroelastomers for life maximisation and unification along with explorations by silicone rubber.

A minimum gain of 10 fold in R & D effort, cost and time for implementation of unification in FBR domain is indicated by comparing the 5 representative compounds for the INDIAN- SFR cover gas seal model and the corresponding 21 compounds (at least) for FBTR along with the reduction of basic development span from more than a decade internationally to 2.5- 5 y for the INDIAN- SFR and FBTR inflatable and backup seals. These efforts could be reduced substantially by further reduction of representative compounds and seals resulting in at least 20- fold gain. This research demonstrated that results from predictive methodologies (FEA, Arrhenius and WLF) are practically independent of specimen conditioning and strain rate for elastomers with low filler content (static elastomeric seals) which implies feasibility of quality control and design with a single set of specimen data (determined at a strain rate of 500 mm/ min) and further substantial potential gains in R & D effort apart from seal- performance- reproducibility- benefits.

The gains in operating cost (15% out of the total cost) is expected to be substantial which is indicated by 827 seal replacements (out of ~ 5000) per year in FBTR vis-à-vis 1 envisaged synchronised replacement in 60 y. Standardised predictive techniques, R & D and quality control maximises the safety and reliability of the reactor on this count.

This research has applied a phenomenological (rather than molecular) approach with experiments, analysis, data extrapolation and design as a novelty in Pasteur's Quadrant (application of science rather than applied science) to provide a comprehensive scheme for simplification, unification and standardisation of critical elastomeric sealing of Indian FBRs (transiting towards SFRs) and other nuclear genres towards attaining the goal of 1 synchronised replacement during a reactor life of 60 y.

List of Figures

List of Tables

Nomenclature

List of Symbols

α_i	Temperature dependent material parameters
α_T	Shift factor
A, B	Constants
A	Deformed cross-sectional area
A_B	Bulge area
A_L	Load area
A_0	Original cross-sectional area
a, b	Material constants
Ɓ	Shape factor
$C_{ijkl}/C_{ijk}/C_{ij}$	Tensor of material or calibration constants
C_{AB}	Green Deformation tensor
C_B	Compression set
c_{ij}	Cauchy's strain or Cauchy deformation tensor
δ_{ij}	Kronecker delta
D_i	Material compressibility
Є	Engineering strain
$\varepsilon/\varepsilon_i/$	Scalar/vector form of infinitesimal strain
ε_{ij}	Cauchy's infinitesimal strain tensor
É	Finite strain, Lagrangian measure
Ē	Activation energy
E_0	Young's modulus
E_b	Elongation at break
E_c	Effective compression modulus
$E_{AB}(E_{ij})$	Finite Lagrangian of Green's strain tensor
é	Finite strain, Eulerian measure
e/e_i	Scalar/vector form of true strain
e_{ij}	Eulerian finite strain tensor or Almansi's strain tensor for finite strain
Ƒ	Force
F	Deformation gradient matrix (superscript c for compressibility)
$F_{i,A}\,(x_{i,A})$	Deformation gradient tensor
$\boldsymbol{f}$	Force vector on ſ
ḟi	Body force vector
$f(\lambda_j)$	Interpolating function
G	Shear modulus
φ	Elastomer compression coefficient
γ	Engineering shear strain
H	Mean pressure function
H_{ijkl}	Tangent stiffness matrix or Hessian
$I/\ I_i$	Scalar/ vector form of strain invariants
$\mathbf{I}_A$	Base vector for undeformed configuration
$\mathbf{i}_i$	base vector for deformed configuration
$J^c(J^{el})$	Elastic volume ratio (superscript c for compressibility)
K(T)	Reaction rate
K_c	Compression spring rate
$\boldsymbol{\Lambda}_{\underline{N}}$	Stretch ratio in original (undeformed) configuration along $\underline{\mathbf{N}}$
λ	Stretch or extension ratio in current configuration
λ_i	Principal stretch ratios
$\bar{\lambda}_i$	Deviatoric principal stretches

λ_j	Principal stretch ratio corresponding to σ_j
λ_n	Stretch ratio along **n**
ƛ and G	Lamé constants
ΔL	Change in gauge length
L	Deformed gauge length
L_0	Initial gauge length
$\boldsymbol{\mu}$	Coefficient of friction
μ_i	Temperature dependent material parameters
M_H	Maximum torque
M_L	Minimum torque
M_x	Modulus or stress at x% strain
ν	Poisson’s ratio
N	Number of terms in W
$\underline{N}$ (X-Y-Z)	Original (undeformed or unstrained) configuration
n	Number of experimental data points
$\underline{n}$ (x-y-z)	Current (deformed or strained) configuration
p	Hydrostatic component of stress tensor (infinitesimal strain)
ƥ	Hydrostatic pressure for infinitesimal strain
Ŕ	Volumetric expansion with temperature change
R	Universal gas constant
$\sigma/\sigma_i/\sigma_{ij}$	Cauchy or true stress in scalar/ vector/ tensor form
σ_j	J^{th} principal true stress from experimental data
σ'_{ij}	Deviatoric stress tensor
σ_{ut}	True, uniaxial tensile stress
ʃ	Closed surface in a continuum
$S/S_i/S_{ij}$	Engineering or Biot stress in scalar/ vector/ tensor form
$S_{(n)}(S_{kk})$	Traction or stress normal to surface (along **η**)
$\theta/3$	Mean stress
τ ($S_{(t)}$)	Engineering shear stress
T	Absolute temperature
$\mathbf{T}^{\eta}$ (T_i^{η})	Traction or stress vector on a limiting surface (normal vector **η**)
T_0	Reference temperature
T_s	Tensile strength
t	Elastomer block thickness
t_i	Final specimen thickness
t_n	Spacer bar thickness
t_o	Initial specimen thickness
t_1/ t_2	Time corresponding to ageing temperature, T_1/ T_2
t_s2	Scorch time
t'_x	Time for x% torque increase
t_x	Threshold time to reach a specific stage (x) of compression set
u/u_i	Scalar/ vector form of displacements
W	Strain energy density function
$X_i(X_A)$	Undeformed coordinates
x_i	Deformed coordinates

List of Abbreviations

A	Air
A1	Axial
ACM	Polyacrylate rubber
AHWR	Advanced Heavy Water Reactor
AI	Atomics International
APA	Advanced polymer architecture
AR	Abrasion resistance
ARDM	Absorber rod drive mechanism
ARP	Aerospace Recommended Practise
AS	Aerospace Standard
ASTM	American Society for Testing and Materials
ASTRID	Advanced Technological Reactor for Industrial Demonstration
AU EU	Polyurethane elastomer
a	Annum
BDBE	Beyond design basis event
BES	Backup elastomeric seal
BMFT	Federal Ministry for Research and Technology of the Federal Republic of Germany
BN-350/600	Bystrieneytrony (Fast neutrons)
BOR-60	Bystrij Opytnyj Reactor (Fast Experimental Reactor);
BS	British Standard
BSR	Bearing support ring
BWR	Boling Water Reactor
CANDU	CANadian Deuterium Uranium
CD	Conceptual design
CDA	Core disruptive accident
CDD	Cumulative design dose
CDFR	Commercial Demonstration Fast Reactor
CDT	Cumulative design travel
CEA	Commissariat a l Energy Atomique
CO, ECO	Epichlorohydrin elastomers
CoP	Conference of parties
COMP	Maximum compressive principal stress
CP	Control plug
CR	Chloroprene rubber
CR	Cumulative rubbing (able only)
CRBRP	Clinch River Breeder Reactor Plant
CS	Compression set
CS	Carbon steel (Table only)
CSM	Chlorosulfonated polyethylene elastomer
CSR	Compression stress relaxation
CSRDM	Control and safety rod drive mechanism
CV	Convection barrier
cpm	Cycle per minute
D	Dynamic
DAE	Department of Atomic Energy
DBA	Design basis accident
DBE	Design basis event
DCP	Dicumyl peroxide

DCP1	Design contact pressure
DD	Design drag
DFBR	Demonstration Fast Breeder Reactor
DFR	Dounreay Fast Reactor
DG	Double gasket
DL	Design life
DLO	Diffusion limited oxidation
DMSRDE	Defence Materials Stores Research and Development Establishment
DP	Design pressure
DP1	Dynamic part
DSRDM	Diverse safety rod drive mechanism
DT	Design temperature
EBR-II	Experimental Breeder Reactor II
EFR	European Fast Reactor
ELR	Typical E_b range
EOR	Elastomeric O Ring
EPDM	Ethylene propylene diene methylene
ESS	Elastomeric static seal
ET	Elevated temperature
EX	Excellent
FBTR	Fast Breeder Test Reactor
FBR	Fast Breeder Reactor
FCR	Flex cracking resistance
FE	Fair- to excellent
FEA	Finite element analysis
FEF	Fast extrusion furnace
FFIM	Failed fuel inspection module
FFKM	Perfluoroelastomer
FFTF	Fast Flux Test Facility
FG	Fair- to good
FH	Fuel handling
FIC	Fluids in contact
FKM	Fluorohydrocarbon rubber
FMQ	Fluorosilicone rubber
FOAK	First of a kind
FOS	Factor of safety
FR	Flame resistance
FT-IR	Fourier transform infrared spectroscopy
Gen	Generation
GD	Good
GE	Good-to excellent
GHG	Greenhouse gas emission
G	Glued
GS	Gap sealed
HASETRI	Hari Shankar Singhania Elastomer and Tyre Research Institute
HAT	Hot air tunnel
HDR	Typical hardness range
HFP	Hexafluoropropylene
HTR	High temperature range
I	Inward
IF	Inflatable
IFS	Inflatable seal
IFTM	Inclined fuel transfer machine
IGCAR	Indira Gandhi Centre for Atomic Research
IHX	Intermediate heat exchanger
IIR	Butyl rubber
IIT	Indian Institute Technology
IMR	Impact resistance

IPR	Institute for Plasma Research
INDC	Intended nationally determined contribution
INDIAN- SFR	Indian Sodium- cooled Fast Reactor
ISO	International Organisation for Standardisation
IVTM	In vessel transfer machine
KNK-II	Kompakte Natriumgekuhlte Kernreaktoranlage (Compact Sodium Cooled Nuclear Power Plant
LEV	Level
LL	Liquid lubricant
LM	Liquid metal
LMS	Liquid metal seal
LMFBR	Liquid Metal Fast Breeder Reactor
LRP	Large rotatable plug
LTC	Leak tight cell
LWR	Light Water Reactor
MDR	Moving die rheometer
MIC	Materials in contact
MOX	Mixed oxide
MP	Maximum pressure
MQ	Silicone rubber
MT	Medium thermal
MV	Main vessel
MW	Microwave
NBR	Acrylonitrile-butadiene rubber
NO	Normal operation
NOAK	N^{th} of a kind
NOS	Number of seals (Table only)
NR	Non reinforced
NT	Normal temperature
NPP	Nuclear power plant
O	Outward
OC	Operating cycle
OR	Ozone resistance
OXR	Oxidation resistance
P	Primary seal
P1	Purge
PEC	Prova Elementi di Combustible
PECVD	Plasma enhanced chemical vapour deposition
PF	Poor-to fair
PFR	Prototype Fast Reactor
PG	Poor-to good
PHWR	Pressurised Heavy Water Reactor
PLBR	Prototype Large Breeder Reactor
PNC	Power Reactor and Nuclear Fuel Development Corporation
PPA	Parallel to plug axis
PSP	Primary sodium pump
PTFE	Polytetrafluoroethylene
PWR	Pressurised Water Reactor
PR	Poor
phr	Parts per hundred parts of rubber
QC	Quality control
R	Radial
Rec.	Reciprocating
RA	Reactor assembly
RCB	Reactor containment building
RF	Radiofrequency
RI	Reinforced

RP	Rotatable plug
RR	Rubber ring
RR	Radiation range
RRO	Range of rotary oscillation
RS	Roof slab
RSOM	Relative speed of motion
RT	Room temperature
S	Secondary seal
S1	Static
SAE	Society of Automotive Engineers
SFR	Sodium-cooled Fast Reactor
SNR 300	Schneller Natriumgekühlte Reaktor
SP	Static part
SPX- 1	Super Phenix 1
SRF	Semi reinforcing furnace
SRP	Small rotatable plug
SBR	Styrene-butadiene rubber
SEES	Spring energised elastomeric seal
SS	Stainless Steel
SS	Stress softened (Figures only)
SS1	Static seal
SSE	Safe shutdown earthquake
STR	Steam resistance
SUR	Sunlight resistance
TAIC	Triallyl isocynanurate
TBC	Teflon based coating
TENS	Maximum tensile principal stress
TFE	Tetrafluoroethylene
TGA	Thermogravimetric analysis
TLC	Teflon like coating
TMR	Top and middle ring
TR	Tear resistance
TS	Top shield
TSR	Typical T_s range
UA	Unaged
UKAEA	United Kingdom Atomic Energy Authority
UPDN	Level mismatch of 2 mm
UN	United Nations
USDOE	United States Department of Energy
UTM	Universal testing machine
VD	Vibration damping
VDF	Vinylidene fluoride
VTSR	Validatory test in scaled rig
W	Weeks
WAR	Water resistance
WDTRS	Westinghouse Development Test Requirement Specification
WE	Western European
WER	Weather resistance
WLF	William Landel Ferry
WTR	Working temperature range
WW	World War
y	Year
3- D	Three dimensional

Chapter 1

Introduction

1.1 Outline

An energy revolution takes shape as climate change effects (flood, rain, storm, Arctic polar ice melt, ocean acidity etc.) from anthropogenic greenhouse gas (GHG) emission compels reformation of energy sector, responsible for about two- third of emission across the globe [1]. Power sector is assuming growing importance as a subset of energy due to increasing electricity demand from population growth, urbanization, improving living standard and better health where power from nuclear and renewable (solar, wind, hydro, bio etc.) sources are expected to assume key complimentary roles because of very low CO_2 emission compared to fossil fuel[1] and steady base load generation capacity of the former [2-8]. This thesis in 8 Chapters takes top- down and bottom-up approach through the vistas of energy- power- nuclear- reactor- Fast Breeder Reactor (FBR)- seal- material to address an area of nuclear power (simplification- unification- standardisation of critical elastomeric sealing) where imbibing the global energy trend could bring major improvements in some of the main goal areas of Generation (Gen) IV technology (i.e. sustainability, safety, reliability and economy) through an evolutionary route.

The introduction is kept unusually long in order to capture the wide diversity of perspectives at the beginning itself. Capturing multidisciplinary domains in entirety with centuries of design, R & D and operation in non- nuclear and nuclear areas as background alike necessitated a style of narration which touches upon key elements of subsequent topics in advance, so as to provide a more complete picture. Design and simplification- unification- standardisation run as common, complimentary and parallel threads throughout where literature data extrapolation is used as one of the main tools of design to minimise experimentations and analysis. This proposition has been

[1] Nuclear: 17 t/GW-h; Biomass: 46 t/GW-h; Hydro: 18 t/GW-h; Solar: 39 t/GW-h; Wind: 14 t/GW-h; Coal: 1041 t/GW-h; Natural Gas: 622 t/GW-h [7]

shaped by cross- interactions of important elements belonging to the domains of energy (energy- power- nuclear- reactor- FBR- seal- material) and design (8 elements) with material as pivotal. Chapter 2 provides motivation and objectives followed by literature review and problem definition (Chapter 3). Chapter 4 describes experimental and analysis methodologies. Results and discussion have been distributed in 3 Chapters (5-7) in order to, i) Demarcate FBR (5-6) and Pressurised Heavy Water Reactor (PHWR) as well as Advanced Heavy Water (AHWR) reactor (7) unification, ii) Demarcate identification of pivotal elastomeric formulation (1st element of design, 5) for life maximisation along with quality control (7th element, 5) and overall unification of all the elements taking the identified formulation as origin (6), and iii) Delineating interfaces and continuity to highlight material as the most important common element of unification. Chapter 8 presents conclusion and future research.

This dissertation primarily aims at betterment of sustainability, safety, reliability, and economy of mixed oxide (MOX) fuelled, sodium cooled FBRs (transiting towards the Gen IV Sodium- cooled Fast Reactor or SFR) by simplification, unification and standardization of critical elastomeric sealing which attempts to embrace the entire nuclear domain by taking FBR as central and expanding it subsequently to the genres of PHWR and AHWR. Material is taken as cornerstone, finite element analysis (FEA, 2nd and 5th elements of design) as facilitator and design as an all- encompassing base towards the objective of 1 synchronised replacement of seals during reactor life of 60 years (y). The 500 MWe, sodium cooled, MOX fuelled, 2-loop, pool type Indian Sodium-cooled Fast Reactor (INDIAN- SFR) nearing its first criticality is the Phase II demonstration prototype of Department of Atomic Energy (DAE) which is taken as a representative model (Fig. 1.1).This is due to the fact that most of the FBRs built till date were MOX fuelled, usually cooled by argon- blanket- covered (Cover Gas) liquid sodium held in main vessel (MV), where elastomers were mostly used for the low temperature-pressure-radiation sealing applications of cover gas [9- 10].

Cover gas seals, designed to prevent release of radioactive argon cover gas to Reactor Containment Building (RCB) and ingress of RCB air into the cover gas space [10] through

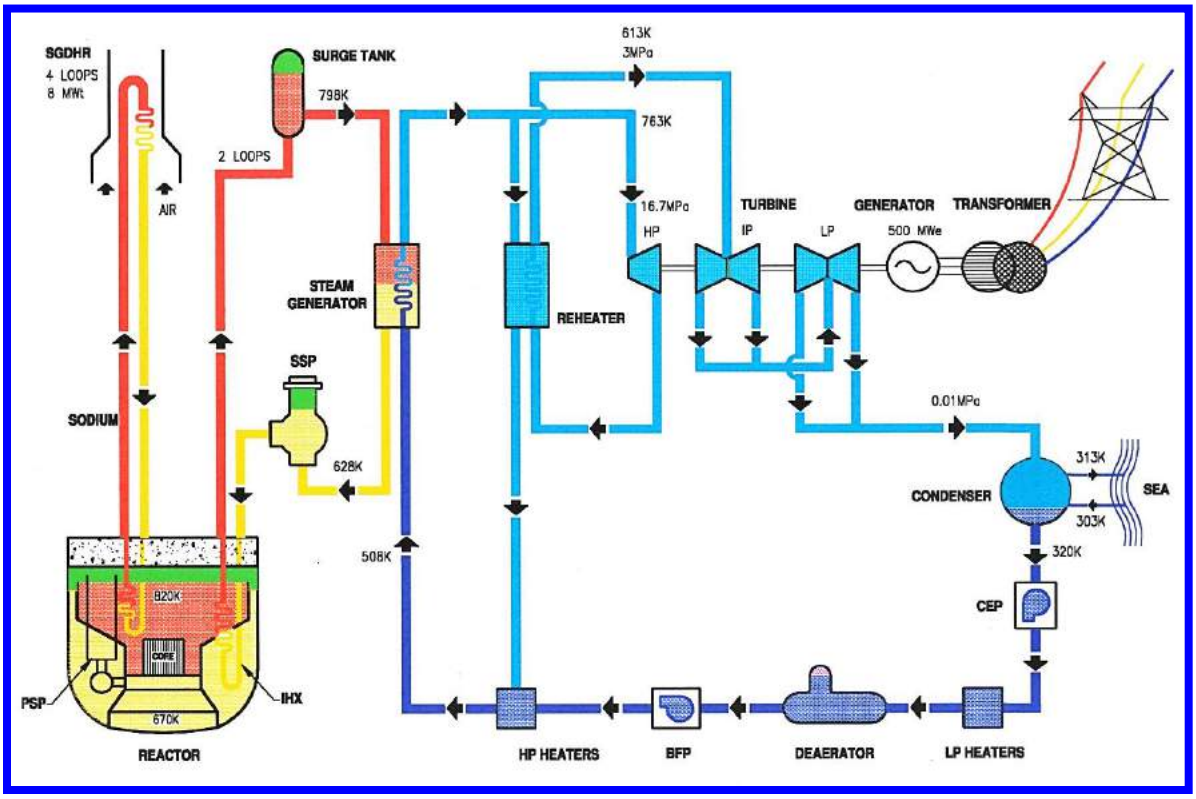


Fig.1.1. Schematic of INDIAN- SFR with Reactor Assembly, cooling and conversion circuits.

component penetration openings on top shield (TS), are representative of critical elastomeric sealing. Nucleating point of research is uniqueness of elastomers which makes part- sizing, manufacture, coating etc. largely rubber compound specific. Elastomeric- failure- criteria (contributing towards seal failure by leakage and friction) comprising of limiting stress- strain[2], release of HF, compression set (CS), tear strength etc., used to assess and maximise seal life, is the other common thread along with simplification- unification- standardisation and design. Figure 1.2 shows an elevation of INDIAN- SFR Reactor Assembly (RA) with various component penetrations on TS.

Feasibility of maximising TS seal life concurrently for synchronised replacement is aided [10] by commonness of low maximum differential pressure (100 kPa), temperature (120 ^{0}C) and γ dose rate (0.1 Gy/ h) at barrier locations across FBRs (experimental, demonstration and commercial; common seal life: 10y) during normal operation and fuel handling conditions along with a set of common fluids (air, argon, sodium aerosol, xenon, krypton) and materials (carbon steel, stainless steel or SS,

[2] Determined by a factor of safety or FOS on tensile strength or T_s and elongation at break or E_b.

aluminium- bronze) in contact. The INDIAN- SFR is representative of MOX fuelled FBRs (transiting towards SFR) in the capacity of cover gas sealing. This owes to the proximity of common representative sealing conditions of the former to those of the later including materials and fluids in contact i.e. 10 y ---- 70 kPa ---- 120 ^{0}C ---- 23 mGy/ h ---- 1000 N/ m-seal-length (maximum rotational drag for dynamic seal) ---- 100 km (maximum cumulative rubbing of dynamic seal during 10 y life) ---- 10 $^{-3}$ scc/ s/ m- seal- length (allowable leakage during normal operation).

The definition of elastomer (used interchangeably with synthetic rubber or rubber) could be elaborated based on American Society for Testing and Materials (ASTM).

i. It must not break when stretched approximately by 100%.
ii. It must retract to ≤ 110% of initial length within 5 min from release after holding on the stretch for 5 min.

1.2 Perspectives

1.2.1 **Energy:** Relevance and importance of evolutionary route and simplification-unification- standardisation

The world has already consumed about two- third of CO_2- emission- ceiling (~2900 Gt, 1870- 2100), required to limit the mean-temperature- rise of earth (from GHG emission) within the agreed goal of 2 ^{0}C by the end of this century in order to avoid climatic catastrophe [2-3]. This calls for consolidation of resources, knowhow and efforts across geographical barriers with a unified and standardised approach. Wide diversity of

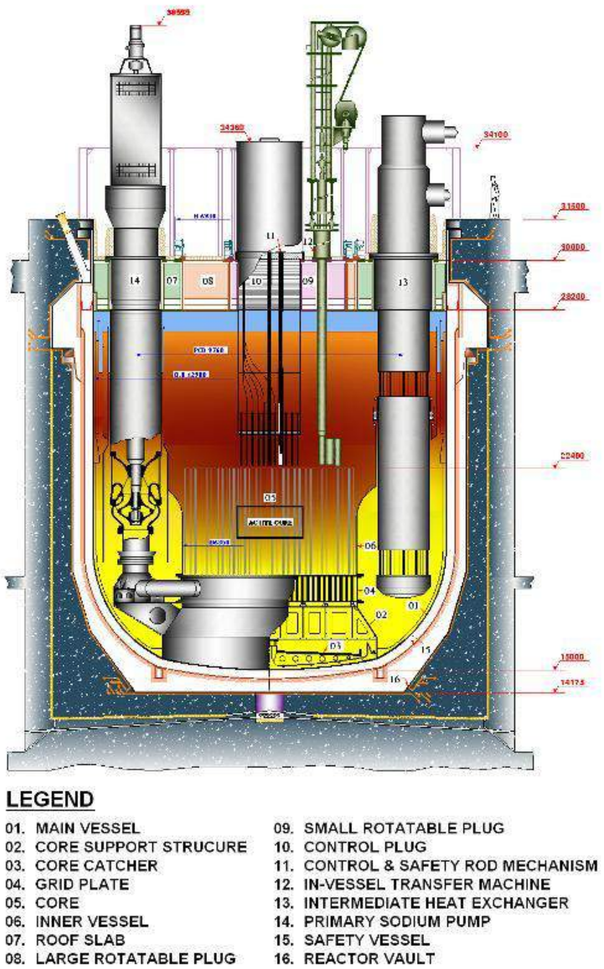


Fig.1.2. Various component penetrations on top shield of the present INDIAN- SFR Reactor Assembly.

societal, infrastructural, technological and economic preparedness suggests that changes should be evolutionary if a common agreed global strategy is to be devised. Conference of Parties (CoP) 21, the recently held (December 2015) 196 nation United Nation (UN) climate summit at Paris concluded that emission reduction volunteered by 150 countries through Intended Nationally Determined Contributions (INDCs) could at the most attain a global CO_2 emission rate of 55 Gt/ annum(a) by 2030 vis-à-vis 30 Gt/a [1], required to meet the short- term boundary condition of 50% - emission - reduction by 2050 (with respect to 2009) towards the 2 ^{0}C limit [5].

Smooth and effective transition to the new energy scheme through an evolutionary route with exchange, sharing and joint- ownership by inter- national cooperation necessitates continuation of the existing resources, knowhow and efforts for energy security with improving efficiency and standardisation as the first step (while new infrastructures, knowledge base, skills and funding structures are introduced) in order to ensure natural interfacing with the new paradigms for effective decarbonisation. Simplification by classification and categorisation (grouping) of the existing for standardisation and unification is fundamental to the process as number of elements to be dealt with is reduce for better safety, reliability and economy. The post- CoP21 approach of reaching peak CO_2emission by 2020 for subsequent reduction [1, 5] is a reflection where infrastructural and technological preparedness imply complimentary roles for nuclear and renewable power sources. New materials for better performance and material security are essential ingredients for sustainable energy security with low carbon footprint [4- 6, 8, 11].

Nuclear power is poised for a significant growth from the current generation of 372 GWe (11% of global electricity, 443 Nuclear Power Plants or NPPs) to 930 GWe by 2050 (17%) in pursuance of the 2 ^{0}C limit, Fukushima Daiichi accident (11 March 2011) and the preceding financial crisis (2008- 2010) notwithstanding [5- 6]. 66 reactors are presently under construction in 15 countries (Gen III Light Water Reactors or LWRs, about 50%) while there are plans for another 164 in 27 countries over the next 8- 10 y [6]. Existing capacity is shared mostly by LWRs (~ 82%) and [5-6] PHWRs (~ 11%; mostly in India, 4.46 GWe, and Canada). PHWR is the second technology of choice (> 7%) in the ongoing constructions [5- 6] also after LWRs (> 85%) with all being built in India [12- 17].

This treatise imbibes the current trend to provides a framework for short- term evolutionary change towards long term objective of economy and sustainability taking safety as the first priority [11]. Design- operation data from INDIAN- SFR and Fast Breeder Test Reactor (FBTR) and R & D results from development of inflatable as well as backup seals for the rotatable plugs (RPs) of INDIAN- SFR and FBTR are utilised. The R & D was a part of *Special Elastomeric Component* development at Indira Gandhi Centre for Atomic Research (IGCAR) which involved more than 15 Indian Agencies [9- 10]. Experiments and analysis carried out as part of this thesis has been described while others are referred.

1.2.2 **Nuclear Power:** Evolutionary changes by simplification and harmonisation for long term economy taking safety as the first priority

Emergence of safety as a priority amongst Gen IV objectives following the Fukushima Daiichi accident, status of thermal reactors and SFR technology, and the necessity for a step- jump in grid capacity addition rate (from 5 GWe/a to 20 GWe/a) towards 930 GWe by 2050 necessitated specific methodologies in evolutionary route for longer term economy or affordability of power (by minimising capital and electricity generation cost) by design simplification and harmonisation (of codes as well as standards) across countries [5]. Envisaged progress in FBRs towards Gen IV and that of the Gen III LWRs till 2050 by simplification and harmonisation of the existing state of knowledge as well as preparedness[3] provides an indication about the route and methodology.

Gen II commercial power LWRs continuing since mid- 1960s with life extension transited towards advanced LWRs (Gen III) from mid- 1990s. These are giving way to the Gen III+ evolutionary design towards the Gen IV technology, represented by 6 systems i.e. SFR, Lead- cooled Fast Reactor, Gas- cooled Fast Reactor, Molten Salt Reactor, Very High Temperature Reactor and

[3] Where material is the key in terms of stress-strain, ageing, life extension, FBR clad to maximise fuel burn up, FBR clad spacer wire for negative void coefficient of reactivity etc. [8, 11]

Supercritical Water- cooled Reactor [11]. FBRs transiting towards SFRs have a clear edge in Gen IV technology because of > 400 reactor- years of operating experience [11, 18- 22]. SFR technology is epitomised by the French Advanced Technological Reactor for Industrial Demonstration (ASTRID) for which detailed-design-completion is scheduled on 2019. Russian Federation is going ahead with a large SFR (BN- 1200; could be deployed by 2030) after the criticality of 800 MWe BN- 800 (June 2014) while the Chinese design of a 1000 MWe prototype is in progress [5]. Japanese Sodium- cooled Fast Reactor (1500 MWe) and Kalimer (600 MWe) are the Japanese and Korean SFR designs [20]. Present Gen IV objective is to attain safety and economy equivalent to the GEN III (similar to the post- 1980s outlook) with targeted fuel burn up, availability and life of 200 GWd/ t, 90 % and 60 y [5, 20]. Future Indian FBRs are in consonance with Gen IV safety philosophy as life and capacity factor of at least 60 y and 85 % are aimed from 40 y and 75 % for INDIAN- SFR [14].

SFRs are likely to emerge as one of the mainstays of nuclear power during the second half of this century globally (by series- commercial- construction) from safety, economy and sustainability perspectives because of maximum fuel utilisation, minimum waste and its storage duration, much better thermodynamic efficiency (40% compared to 30% for LWRs), much higher fuel burn up (100- 200 GWd/ t; LWR: 55 GWd/t; PHWR: 7 GWd/ t) etc. [14].The 2050 reactor mix is expected to be dominated by Gen III (with some Gen II, mostly under construction now) with countable presence from PHWRs and FBRs transiting towards the Gen IV SFRs. Evolutionary changes could be seen from the efforts to adequately utilise the existing and also to improve the efficiency of utilisation at the same time by simplification, harmonisation and trained manpower for streamlined and time-bound results. Nuclear power capacity addition is therefore being carried out in faster and larger steps of 1000- 1700 MWe Gen III LWRs (which does also bring in economy of scale) in order to bring in step- jump in grid capacity addition rate. The Indian progress towards attaining 63 GWe by 2032 [13- 16] while contemplating for 100 GWe by 2050 is an equivalence in analogy, with reactors worth 7.7 GWe under construction [17] and preparatory works for another 28 GWe of imported

LWRs (24 nos., 1000- 1700 MWe) nearing completion [15- 16] Life extension of the existing Gen II LWR fleet to at least 60 y is another example of ensuring continued generation of economic low-carbon power for sustainability by energy security [5]. Safety upgrades of LWRs during post-Fukushima period completes the circle of activities for smooth interfacing with the new energy scheme through the subset of nuclear power using evolutionary route.

Post- Fukushima safety upgrades corresponding to demonstration- qualification of emergency preparedness and mitigation abilities have increased the cost of the already pricy Gen III LWRs by about 20 %, similar to the premiums of life extension. Ensuring enhanced safety culture across regulators, operators (utilities), industries (vendors) and supply chain (to be supported by enhanced quality consistency) is part of the process. Design simplification and harmonization of codes as well as standards across various roles and countries are therefore being integrated for a broader reach of standardization in order to meet the longer- term objective of economy (similar to peaking of CO_2 emission by 2020 for subsequent reduction) while imbibing lessons from First- of- a- kind (FOAK) plant towards the economic and standardised N^{th}- of- a- kind (NOAK) through the shortest learning curve (with trained manpower), taking safety as a priority [5]. This is applicable for the FBRs transiting towards the Gen IV advanced SFRs as well, illustrated by this dissertation. Initial estimations on INDIAN- SFR cost- breakup (capital/ operating/ fuel- cycle: 75/ 15/ 10 %) is indicative of the proportion of price components in demonstration or commercial FBRs [12].

1.2.3 **Energy and Nuclear Power:** Material and energy security, energy efficiency, reliability, economy and sustainability

Material security is an essential ingredient of energy security, required to ensure implementability of design simplification and harmonisation (or simplification- unification-standardisation) towards economy with reliability (redundancy + diversity + independence) which results in sustainability. From another perspective, material security and simplification- unification-standardisation are complimentary halves of consolidating resources, knowhow and efforts as part of an evolutionary process. Material security does also include material efficiency i.e. minimum material input for a product of particular functionality which enhances economy. Energy efficiency is about maximising performance with same power (rate of energy) input which enhances material efficiency (fuel for power) and security along with economy e.g. better thermodynamic efficiency of FBRs vis-a-vis LWR. Energy conservation (i. e. reduced energy consumption) improves material security and economy. Enhancing these three factors (sometimes called energy efficiency together) results in less GHG emission and improved sustainability. Evolving trends of FBRs transiting towards SFRs and the Indian AHWR are examples of material and energy efficiency [23- 25].

Closing of fuel cycle by converting (breeding) ^{238}U isotopes (99.3% of natural uranium) to the fissionable ^{239}Pu in SFRs and multi- recycling of plutonium could enhance uranium utilization up to 80% (thermal reactors: 1%) for maximisation of material and energy security as well as economy towards enhanced sustainability in nuclear domain. Transmutation of trans-uranium or minor actinides in the process brings down long-lived radioactive isotopes to < 0.1% which reduces the footprint of deep geological repositories (> 200 times) and duration of storage (< 300 y) substantially for significantly improved sustainability. These are further strengthened by diversity in Indian context due to the potentials in thorium cycle [14].

One of the aims in choosing 6 representative systems for Gen IV technology were recycling of spent fuel in a symbiotic fashion using cross-cutting mode which converts one fuel form to another for material security and sustainability of nuclear power [22]. This does also add to reliability because of the usage of multiple resources for the same purpose. An excellent complimentary example from energy domain on widening fuel resource (diversity, material security and sustainability) while meeting the objectives of decarbonisation, utilization and waste minimization simultaneously could be sequestering the emitted CO_2 and combining it with H_2 using heat and catalyst to produce polymers and various other chemicals which, by a recent estimation, consume at least 6 % of global energy mostly from oil [26].

The necessity for consolidation of right resource for the right end-use (at the right time) with simplification- unification- standardisation has brought in concerted exchange, sharing and ownership of knowhow, material technology and financial resources. This could be seen from research drives across Europe and America in order to arrive at a minimum number of representative materials for the Gen IV technology using necessary characterising- qualification techniques and by dividing the systems into a few representative domains i.e. pressure boundary, reactor internals and heat conversion system. Impressive strides in recent times could be found in convergence towards oxide dispersion strengthened ferritic- martensitic and advanced austenitic SS for fuel clad, wrapper and other applications [27- 34]. One of the key examples of similar search in the broader energy domain is consolidation of rare earth metal such as dysprosium and neodymium (permanent magnets for wind turbines and electric vehicle motors) or developing substitutes for them [35].

Indian thorium is of the best quality in the world. There is an envisaged scenario of 600 GWe of nuclear power by 2080- 90 with acceleration of the DAE Phase III- program on reaching FBR capacity of 220 GWe [14]. Both China and India are considering PHWR designs for a thorium fuel cycle as thorium (^{232}Th) could sustain thermal breeding by acting as a fertile host in MOX fuel (UO_2-ThO_2), to be converted into fissionable ^{233}U with nearly constant conversion ratio in a wide spectrum of neutron energy. The cycle produces negligible plutonium and the waste contains low amount of α emitter. In this context the Indian design of 300 MWe, light water cooled (boiling), heavy water moderated, MOX fuelled AHWR based on lightly enriched uranium and thorium fuel cycle assumes significance with heat removal from core by natural circulation (pump and motor eliminated), negative void coefficient of reactivity, average fuel burn- up of 64 GWd/t, efficiency of 39 % and life of 100 y [23- 24].

The concept of simplification, unification and standardization pursued in this proposition aims at reinforcing Indian nuclear domain as the first objective with 1 synchronised seal replacement during 60 y life of Indian SFRs in specific and similar approach for other reactor genres in general.

Minimising and synchronising seal replacements are synonymous to enhanced reliability and safety of reactors with predictability which is strengthened by lower operating cost[4] (15% of reactor cost, Section 1.2.2), improved capacity factor[5] (towards 85%, Section 1.2.2) and better material efficiency/security[6] implying significant potential cumulative gains in economy and sustainability if adopted universally across genres and countries with a standardised quality control (for repeatability and reproducibility, 7th element of design) and R & D scheme (8th element) as suggested by the outcome of this dissertation.

1.2.4 **Nuclear Power:** Safety as the first priority encouraging elastomeric sealing and simplification- unification- standardisation

The Fukushima Daiichi accident brought safety to the fore by introducing the necessity to demonstrate/qualify emergency preparedness and accident- consequence- mitigation-ability during beyond design basis event (BDBE) or accident, postulated multiple external events (flood, earthquake etc.) on multi- plant site in particular, which has added emphasis to the existing Gen IV focus on core, passive cooling and sodium- water reaction [5, 11]. This is a paradigm- shift from the post- 1980s safety philosophy of FBRs which redefined core disruptive accident (CDA) from a design basis accident (DBA) or event (DBE) to BDBE (control rod scram non- mandatory) by reducing the CDA probability from $> 10^{-6}$ to $< 10^{-7}$ with a changed approach of accident- prevention rather than accident-consequence-mitigation (pre- 1980s). The post- 1980s trend, propelled by enhanced economisation drive of FBRs from slowing down of global industrial progress and falling uranium price, coupled with limitations of codes in capturing multi-physics phenomena of CDA, enabled establishing elastomeric inflatable seal (not resistant to CDA) as the primary barrier for FBR RPs (instead of the most complex, bulkiest and costliest CDA- resistant liquid metal or LM freeze

[4] Savings in seal maintenance- replacement and reactor downtime cost
[5] Lesser reactor downtime because of continued seal operation
[6] Lesser elastomer quantity because of longer life, enabled by predictive techniques of FEA (2nd and 5th element of design), Arrhenius and William Landel Ferry equations (6th element) for sizing and life assessment during synergistic ageing) which ensures utilization of maximum material potential

seal) which is the basis for unification and standardisation of this research [9- 10, 36]. ASTRID core design representing the Gen IV concepts aims at making CDA impossibility and tries to ensure by defence-in-depth that even if the impossibility occurs, energy release should be minimum. The key issue is to ensure negative void coefficient[7] of reactivity under all perceivable circumstances which necessitates avoiding gas bubble through the core by all means [11]. This outlook has profound significance for simplification- unification- standardisation in the post- Fukushima context on the following counts,

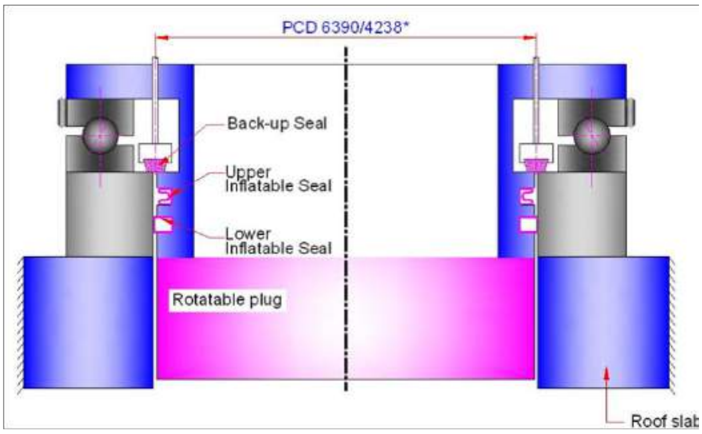


Fig.1.3. Schematic of inflatable and backup seals in support arrangement of INDIAN- SFR rotatable plugs during normal operation.

I) Elastomeric barriers are not discouraged on cover gas boundary as postulated energy release during CDA is minimum and the safety philosophy primarily focuses on fuel clad as the first boundary and RCB as the last. Majority of elastomeric static-dynamic barriers are O-rings (few V- ring, lip seal etc.) which are CDA resistant. Besides, INDIAN- SFR is designed to withstand CDA without seal at the largest leakage opening (RPs) in TS (separating cover gas and RCB air, Fig. 1.2) even though CDA- differential- pressure (2.1 MPa corresponding to 150 MJ energy release) resistance is assigned to the secondary backup seal (secondary barrier downstream of inflatable seal) made of fluorohydrocarbon rubber (FKM), installed in the INDIAN- SFR RPs for the past 6 y (Fig. 1.3). Apart from a number of inherent advantages (simplicity, economy, compactness etc.) elastomeric sealing does also offer the flexibility of access, maintainability and replacement vis-a-vis welded structures. This research is basically a continuation of the design

[7] Minimising amount of sodium coolant between fuel pins in the core by minimising spacer wire diameter is an area of cutting edge research with innovation which ensures minimum void (and hence the possibility of reactivity addition as well as power surge) in the eventuality of loss of coolant. New material technology involving advanced austenitic stainless steels and ferritic martensitic steels is a key enabler here with extensive R & D on formulating the right composition, characterisation as well as qualification.

philosophy of European Fast Reactor (EFR; 1988- 1993) where inherent advantages and flexibility of elastomeric barriers are combined with those of welded structures by minimising permeation leakage and maximising seal life [9- 10].

II) Assuring negative void coefficient of reactivity by avoiding gas bubbles through the core necessitates exclusion of liquid lubricant in cover gas barriers which enables standardisation of seal sizing by FEA with a uniform approach and paves the way for unification of antifriction coating for dynamic seals.

Simplification, unification and standardisation is carried out in pursuance of the current safety philosophy by simplifying the TS seals of INDIAN- SFR into 9 ideally representative categories, finding the categories existing in reactor application, further simplification by identifying 2 representative categories of the existing and finally identifying the most important to initiate unification in terms of standardising each of the 8 design elements.

1.2.5 **Nuclear Power:** Cover gas seals of INDIAN- SFR

Total circumferential length of elastomeric cover gas barriers[8] on TS of the **INDIAN-** SFR amounts to about 700 m [37]. Seal designs should allow the component penetration openings on TS to serve multiple purposes i.e. locating, guiding, supporting and retaining components while allowing rotary and reciprocating movements. Basic function of cover gas sealing, is to prevent outflow of higher pressure radioactive argon cover gas to RCB air above TS in order to minimize occupational hazard (INDIAN- SFR: ≤ 1 mr/ h; ≤ 30 mSv/ y/ person) and avoid diffusive ingress of RCB air into the cover gas space, driven by concentration gradient for partial pressure equalisation. TS zone spanning roof slab (RS), RPs and control plug (CP) provides thermal and biological shielding to the hot and radioactive argon cover gas (one of the reasons being personnel access above TS) and a comprehensive description of sealing functionalities here considers immediate interfacing with the TS and an overall conformity with the broader principles of FBR design [10].

[8] Millimetre to several meters in diameters, as seen from earlier FBRs and INDIAN- SFR

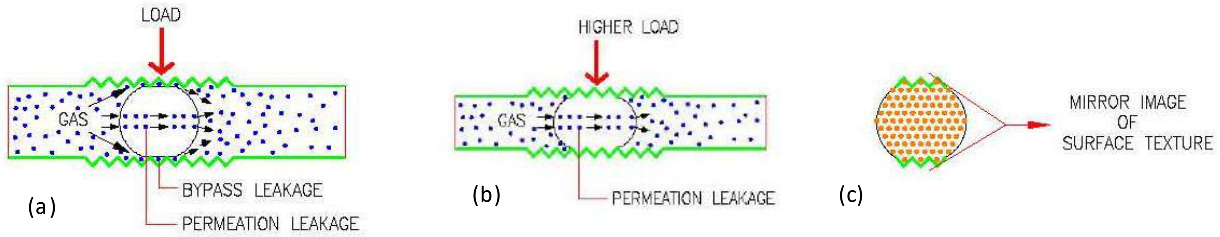


Fig.1.4. (a) Bypass and permeation leakage in elastomeric O- ring under low compression, (b) Elimination of bypass leakage with higher compression and, (c) Flow of seal elastomer (under compression) into micro- irregularities of mating surface creating mirror image of mating surface topography on sealing face.

Seals at leakage openings created by component penetrations on TS basically control the exchange of fluids (RCB air and radioactive argon cover gas), driven by various gradients (pressure, concentration, temperature, velocity) and body forces, by minimizing laminar and turbulent flow through minute unfilled passages (bypass leakage) at sealing contact and diffusion of fluids (permeation) through seal material (Fig. 1.4). Metallic seals could provide zero engineering leak-tightness (i.e. helium leak-tightness: $\leq 10^{-8}$ scc/s). Typical gas molecules of less than a nanometre in size could diffuse through the smallest engineering gap or even pores of metallic casing which is why permeation leakage through elastomers could be reduced but cannot be eliminated in engineering sense as countable diffusive passages always exist [10]. A basic thumb rule for bypass leak tightness is that contact pressure between seal and mating surfaces should be more than differential pressure across barrier, achievable by both elastomeric and metallic seals.

The argon cover- gas- blanket above liquid sodium coolant pool (immersing the UO_2–PuO_2 MOX fuel core in INDIAN- SFR MV) is laden with sodium aerosol, fission product gases (xenon and krypton) and other radioactive species (^{41}Ar, ^{24}Na etc.). Typically, the Na- Ar interface temperature of ~ 550 ^{0}C drops across the blanket to ~350–400 ^{0}C at TS bottom plate and then decreases further up exponentially to 60- 100 ^{0}C at TS top plate (Fig. 1.2). Reaction of RCB air (carrying CO_2, moisture etc.) with sodium aerosol or mist particles (NaO, Na_2O) produces NaOH, Na_2CO_3, Na_2O etc. which could corrode primary coolant circuit components and block coolant flow through core subassemblies resulting in temperature and/ or reactivity surge. Seizure of critical dynamic components (i.e. absorber rod drive mechanism or ARDM, RPs etc.) because of mist- deposition at cooler zone of leakage openings (sodium freezing temperature: 97.8 ^{0}C) had been a source of recurring operational problems in FBRs [9- 10].

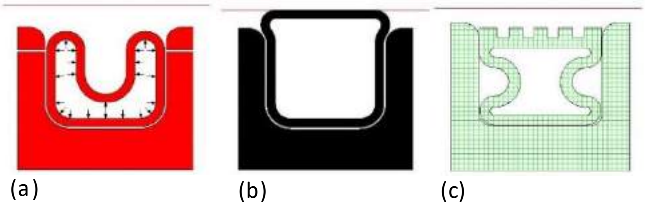


Fig.1.5. (a) INDIAN- SFR and FBTR un-beaded inflatable seal (deflated and disengaged) fitment in rectangular groove. (b) Seal inflated and engaged. (c) Beaded inflatable seal (disengaged).

Elastomeric inflatable (primary barrier: normal operation- static; fuel handling- dynamic) and backup (secondary barrier: normal operation- static, fuel handing- disengaged) seals (Fig. 1.3), used in large rotatable plug (LRP) and small rotatable plug (SRP) of INDIAN- SFR form the most important sealing combination of cover gas because of largest leakage paths (gap: 5 ± 2mm; diameter: LRP/SRP- 6.4 m/ 4.2 m) and some of the most demanding operating requirements including longest- life- demand as well as largest replacement span up to about a month of reactor downtime. The sealing system is classified as Safety Class I (Seismic Category I), safe shutdown earthquake (SSE) design and should function without failure for at least 10 y individually at design operating conditions (pressure: 25 - 25 kPa; temperature: 120 - 110 ^{0}C; γ dose rate: 23 - 23 mGy/ h; allowable leakage: 10^{-3} - 10^{-3} scc/ s/ m- seal- length) which are nearest or equal to the representative cover gas sealing conditions of INDIAN- SFR (Section1.1).

FBR inflatable seals (non-CDA-resistant) are toroidal elastomeric membranes (reinforced or non-reinforced) bonded to rectangular grooves by adhesive in general, where beaded or unbeaded surfaces are usually inflated by argon gas (supplied through inflation connector) at low inflation pressure (≤ 0.3 MPa) to engage with the mating surface (Fig. 1.5). FBTR experience (first criticality: October 18, 1985) shaped conceptualization and design of INDIAN- SFR significantly [36] where operating experience of FBTR elastomeric sealing provided key impetus for simplification, unification and standardization. Figure 1.6 shows axial fitment of the low pressure secondary barrier (inflating parallel to plug axis at pressure of 45 kPa/ 25 kPa during normal- operation/ fuel- handling) in FBTR RPs, downstream to the primary liquid metal (LM) seal, intended to add reliability (redundancy + diversity + independence) to the sealing scheme and protect the primary barrier from oxidation by RCB air, apart from avoiding reverse-leakage of radioactive argon cover gas to RCB air.

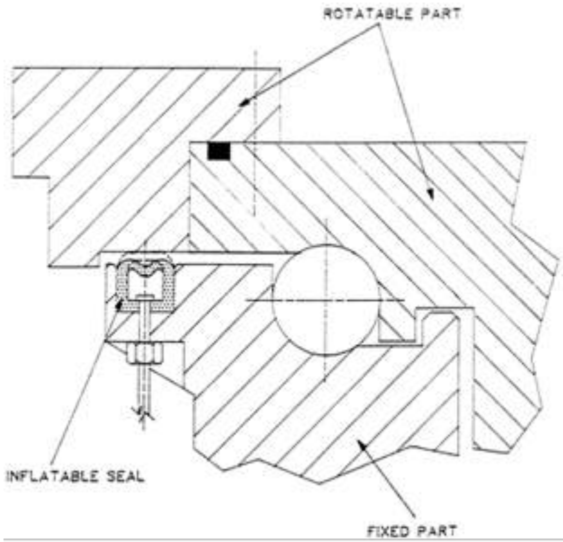


Fig.1. 6. Inflatable seal arrangement in FBTR rotatable plug.

Simplification of NDIAN- SFR cover- gas-sealing as model with classification by size/ manufacture (≤ 0.5 m diameter, moulding; > 0.5 m diameter, extrusion) and operation (static, dynamic), and further by geometry and design (O-ring and V-ring/ lip seals) for the initial 8 categories[9] is a process of consolidating- streamlining the existing design resources and knowhow (pursued in this treatise) towards unification by standardisation through an evolutionary route which is accompanied by parallel simplification of the material module (1st element of design) into 1 generic (i.e. FKM, silicone rubber or MQ, fluorosilicone rubber or FMQ, ethylene propylene diene methylene or EPDM etc.) non- reinforced elastomer representative of all categories.

1.2.6 Potential impact of research in Indian context

Nuclear power which is poised for major growth in China, India, Middle East, Russian Federation, South Africa, Republic of Korea, Turkey, Poland and UK. Contribution of nuclear in the electricity mix of China, India and U.S.A. are expected to be 19%, 18% (from ~ 3% presently) and 17% respectively by 2050. China is expected to be the largest producer of nuclear electricity (250 GWe, 27% of 930 GWe) whereas India is poised for growth to at least 100 GWe by 2050 [5] from the current installed capacity of 5.78 GWe through the 63 GWe by 2032 (Section 1.2.2) as a shorter term goal.

[9] Inflatable seal is the 9th special category because of the absence of compression set (CS) based failure

Presently identified 0.2 Mt of uranium reserve indicates an Indian potential of PHWRs worth 20 GWe from the currently installed 4.46 GWe [21]. With about 10 GWe of space for FBRs corresponding to the shorter term goal of 63 GWe by 2032 there are plans for a number of MOX and metallic fuelled FBRs i.e. 600 MWe, pool type MOX fuelled FBR 1 & 2- from 2023; 115 MWe Metallic Fast Test Reactor- from 2025; 600 MWe Metallic Demonstration Fast Reactor- from 2028; series construction of MOX and metallic fuelled Commercial Fast Breeder Reactors- 2030 onwards [18- 19].

Classification and categorization are done till they influence and bring changes in seal elastomeric formulations. A representative seal corresponds to the most demanding operating requirements of that category where the representative compound is applicable to all other seals with lower operating demand [36]. As would be seen eventually, temperature becomes the representative operating requirement which facilitates choice of 1 common generic elastomer.

1.3 Research Methodology

The FKM formulation of INDIAN- SFR RP inflatable seal (S- 1) obtained from past research is taken as the initiator of unification by standardisation of the following 7 elements of design in the most effective way for reasons explained in Chapter 3.

i) FEA based seal sizing, optimisation and design, ii) Seal manufacture by cold feed extrusion (> 0.5 m diameter) and injection moulding (≤ 0.5 m diameter), iii) Teflon- like antifriction coating by Plasma Enhanced Chemical Vapour Deposition (PECVD), iv) Synergistic ageing and stress- strain based life assessment by FEA, v) Synergistic ageing and stress- strain as well as CS based life assessment by Arrhenius and William Landel Ferry (WLF) equations and, vi) Quality control.

The 8^{th} element (standardised R & D framework) evolves from the first 7 for effective and efficient future implementation of unification with minimum effort and time while ensuring safety, reliability, economy with maximum predictability and reproducibility of performance.

Figure 1.7 depicts the entire outline of research methodology based on the above to be elaborated Chapter 3 onwards.

1.4 General

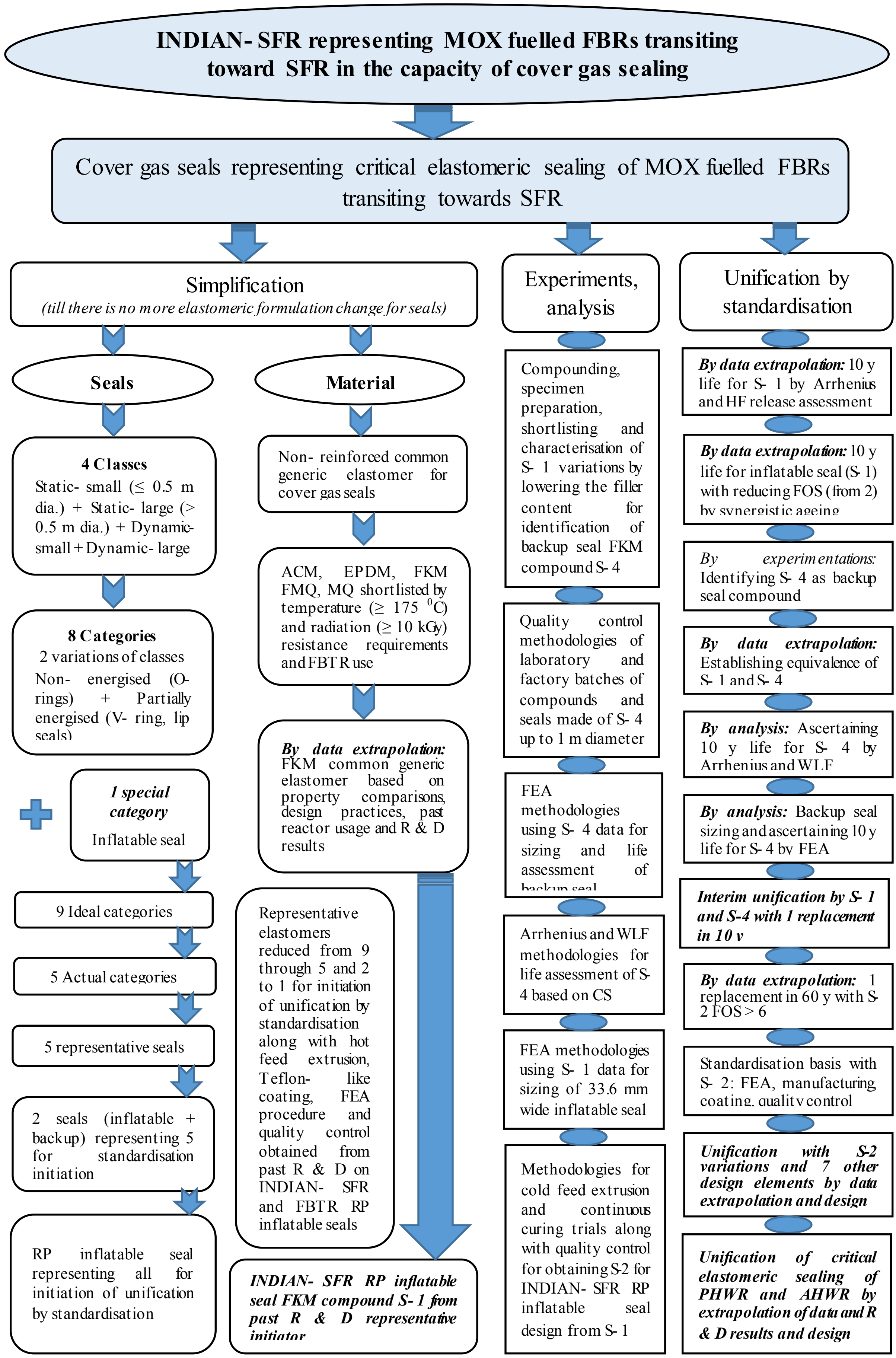
This research belongs to Pasteur's Quadrant (application of science rather than applied science) where use had been the prime motivator. Producing useful research often requires advancement of knowledge with generation of new information [38]. The subsequent chapters are all about straying from the goal of immediate use only to return back at the end, as the developed knowledge shapes understanding sufficiently towards progress of science and its useful applications.

Generic names of elastomers have been abbreviated as per the designations ASTM standard, ASTM D1418. Expansions for abbreviations, valid across tables/ figures and not used in the text, are indicated in the tables/ figures at appropriate places. Code names of elastomeric formulations used during development[10] have been retained in parallel with the corresponding *S- Series,* used for unification. The treatment of elasticity is largely based on continuum mechanics, supported by Appendix (B). Two more Appendix (A and C) are provided on generic elastomer selection and standards accessed respectively. Index notations have been used mostly in defining scaler (z, tensor of rank 0), vector (z_i, tensor of rank 1) and tensors of rank 2 to 4 (z_{ij} to z_{ijkl}). Boldface letters (**z** or **Z**) have been used for vectors and tensors sparingly, when necessary.

List of publications arising from the thesis are referred at the beginning of a *Section* whereas others are mentioned in the body of text. Local *Conclusion Sections* added Chapter 3 onwards (till Chapter 7) in order to consolidate outcome in each, supported by shaded versions of Figure 1.7 for highlighting the progress.

[10] Such as the laboratory DMF fluoroelastomer compound series for inflatable seals and the laboratory IG fluoroelastomer compound series for backup seal; production fluoroelastomer compounds, Comp #1- #9, extending DMF3 or the INDIAN- SFR- RP- FKM- inflatable- seal- formulation to new production system as well as compound; production batch variations of IG3, 10a-c, during extrusion.

Fig.1. 7. An outline of research methodology used in thesis.



Chapter 2

Motivation and Objectives

2.1 Statement of Motivation

To enhance safety, reliability, economy and sustainability of SFRs in Indian context and extend the benefits to reactors of other genres with minimization of elastomeric compounds for cover gas seals and maximization of seal life towards 1 replacement during a reactor life of 60 y while keeping the related (seal and reactor) R & D as well as capital and operating costs of reactor minimum in longer term through evolutionary route, taking safety as the highest priority.

2.2 The Canvass of Motivation [9- 10, 36-37, 41- 43, 45]

2.2.1 Uniqueness of elastomers

Accident of space shuttle Challenger (January 1986) due to failure of a small FKM O-ring highlighted the danger of treating elastomer as a mere replacement of metal [39]. Exclusive properties, fabrication techniques and design paradigms of elastomer is to be appreciated for proper exploitation of its unique benefits [40] i.e. simplicity, economy, compactness, resilience, low contact pressure, low or zero assembly load, surface conforming capacity, tolerance absorbing ability etc.

Nonlinear hyperelasticity and geometric nonlinearity, accompanied by near incompressibility under mechanical load and considerable sensitivity of product properties to geometry, material composition, processing parameters, environmental stresses and operating time are some of the unique characteristics of elastomers. Material formulations and processing parameters are mostly owned by manufacturers/ developers as per rubber- industry- practice and usable design data are rare in handbooks. This is more so for nuclear applications involving ionizing radiation, uncommon fluids and long service span because of inaccessibility, maintenance hazard and cost of reactor downtime. Uniqueness of elastomers and specialty of nuclear applications therefore often tend to present a bizarre array of innumerable proprietary compounds, processes and properties for the elastomeric sealing applications with random service spans as demonstrated by studies of Canadian PHWRs and FBTR. Complexity is compounded by the fact that elastomeric failure parameters (Section 1.1) degrade at different rates under the synergistic ageing influence (air + ozone + sodium aerosol + temperature + radiation) of same operating environment where diffusion limited oxidation (DLO) effects add additional variability.

Complete characterization of elastomeric compounds to the standards of nuclear needs is an arduous task which often tends to seal/material-specification-oversimplification resulting in spurious/ unqualified proprietary compounds. The solution lies in founding the sealing applications of reactor on a minimum number of well- characterized and qualified compounds for enhancing safety, reliability, economy and sustainability.

2.2.2 INDIAN- SFR representative of FBRs in terms of critical elastomeric sealing

INDIAN- SFR representing FBRs (transiting towards SFRs, Table 2.1) in the capacity of cover gas sealing (Section 1.1) suggests multiplicative gains from standardisation of compound minimisation within and across reactor genres. Benefits could be maximised by maximising elastomeric seal life from the existing specification of 10 y.

2.2.3 Specialty of nuclear applications and international experience

Limitations of using off- the- shelf commercial elastomeric items in nuclear applications was highlighted by the failure of successful, Fermi- RP- lip- seal-design (MQ rubber) in Rapsodie during late- 1960s which was subsequently confirmed by FFTF- CRBRP (Table 2.1) cover gas seal development (mid- 1970s). For similar reasons, leakage beyond limit and frequent unpredictable replacements of elastomer seals started to surface in Canadian PHWRs from 1960s which necessitated minimizing the number of compounds, specifying the elastomers in more detail, correlating sealing failures and mechanics of sealing in a better way and replacement/ refurbishment with a better validated design. Large leakage (1000 cc/ s) of KNK I (Table 2.1) cover gas seals during mid- 1970s suggested similar improvements which continued to determine the R & D scheme even

Table 2.1. Design features of FBRs along with types of rotatable plug seals and their materials

Reactor: circuit configuration/ No. of primary loops	First-criticality/ Final-shutdown	Power (MWt/MWe)	No. of RPs [a]	MV dia. [b] (m)	RP seals and materials [c]
Experimental Fast Reactors					
[@] DFR: loop/24 (UK)	1959/1977	60/15	2	3.2	O-ring (MQ) + LM double-leaf dip (Hg)
[#] EBR-II: pool/2 (USA)	1961/1998	62.5/20	2	7.9	LM freeze (Sn-Bi)
[#] Fermi: loop/3 (USA)	1963/1975	200/61	1	4.8	Ar pressurized lip (MQ) + LM dip (NaK) [d]
[~] Rapsodie: loop/2 (France)	1967/1983	40/0	2	2.4	LM freeze (Sn-Bi) + Lip (Fermi type) [e]
BOR-60: loop/2 (Russian Federation)	1968/----	55/12	2	1.4	LM freeze (Sn-Bi)
KNK-I and II: loop/2 (FRG)	1972/1991	58/20	2	1.9	**Inflatable (IF)**, HP- 50 with perbunabase (CSM)
JOYO: loop/2 (Japan)	1977/----	140/0	2	3.6	LM freeze
FFTF: loop/3 (USA)	1980/1996	400/0	3	6.2	Mechanical + IF (NBR) + LM dip
FBTR: loop/2 (India)	1985/----	40/13	2	2.4	LM freeze (Sn-Bi) + IF (CR)
PEC: loop/2 (Italy)	Project cancelled	120/0	1	3.1	Double gasket + LM dip (Hg)
Demonstration or Prototype Fast Reactors					
BN-350: loop/6 (Kazakhstan)	1972/1999	750/130	2	6.0	LM freeze (Sn – Bi) + Rubber ring
Phenix: pool/3 (France)	1973/2010	563/255	1	11.8	LM freeze (Sn – Bi) + IF (NBR)
PFR: pool/3 (UK)	1974/1994	650/250	1	12.2	O – ring (MQ) + LM double-leaf dip (Hg)
BN-600: pool/3 (Russian Federation)	1980/----	1470/600	2	12.9	LM freeze (Sn-Bi) + Rubber ring
MONJU: loop/3 (Japan)	1994/----	714/280	1	7.1	LM freeze (Bi- Si- In) + IF (Viton®)
INDIAN- SFR: pool/2 (India)	----	1250/500	2	12.9	If (FKM) + Backup rubber ring (FKM)
CRBRP: loop/3 (USA)	Project cancelled	975/280	3	6.2	IF (NBR)
SNR-300: loop/3 (FRG) [f]	---	762/327	3	6.7	IF (MQ with FKM layer inside)
Commercial Size Reactors					
SPX-1: pool/4 (France)	1985/1998	2990/1242	2	21.0	LM freeze (Sn- Bi) + IF (MQ)
SPX-2: pool/4 (France)	Subsumed to EFR	3600/1440	2	20.0	IF (MQ)
SNR 2: pool/4 (FRG)	Subsumed to EFR	3420/1497	2	15.0	IF (MQ)
DFBR: loop/3 (Japan)	Not determined	1600/660	2	10.4	Lip (FKM) + IF [g](FKM)
CDFR: pool/4 (UK)	Subsumed to EFR	3800/1500	2	19.2	Spring energized elastomer seal + LM dip (Hg)
EFR: pool/3	Not determined	3600/1580	2	17.2	IF (MQ)

BN-350/BN-600: Bystrieneytrony (Fast neutrons); BOR-60: Bystrij Opytnyj Reactor (Fast Experimental Reactor); CDFR: Commercial Demonstration Fast Reactor; CRBRP: Clinch River Breeder Reactor Plant; DFBR: Demonstration Fast Breeder Reactor; DFR: Dounreay Fast Reactor; EBR-II: Experimental Breeder Reactor II; EFR: European Fast Reactor; FBTR: Fast Breeder Test Reactor; FFTF: Fast Flux Test Facility; KNK-II: Kompakte Natriumgekuhlte Kernreaktoranlage (Compact Sodium Cooled Nuclear Power Plant); PEC: Prova Elementi di Combustibile; PFR: Prototype Fast Reactor; SNR 300: Schneller Natriumgekühlte Reaktor; SPX-1: Super-Phenix 1; SPX- 2: Super-Phenix 2;

[a] Approximate RP diameters (m): BN-600: 3, 4.8; CRBRP: 6.4,4.6,1.8; EBR-II: 3.4, 2.4; EFR: 6.9, 5.2; FBTR: 3.9, 2.4; Fermi: 2.7; FFTF: 1.9, 1.9,1.9; JOYO: 4.7,2.9; KNK-II: 2.8, ---; MONJU: 4.5; INDIAN- SFR: 6.9, 4.7; PFR: 3.4; Phenix: 4.5; Rapsodie: 3.9, 2.4; SNR 300: 5.5, 3.5, 1; SPX-1: 12, 10.

[b] Inside diameters (approximate)

[c] Cover gas pressures (kPa): BN-350: 186 (abs.); BN-600: 137 (abs); BOR-60: 49; CDFR: *; CRBRP: 2.5; DFBR: 90; DFR: 25; EBR-II: ± 0.24; EFR: 105-123 (abs.); FBTR: 3 ± 1; Fermi: 100 (abs.); FFTF: 3 (gauge); JOYO: 1; KNK-II: 2; MONJU: 50; PEC: 2 (gauge); INDIAN- SFR: 10; PFR: 7; Phenix: 2.0 + 3.0/-1.5; Rapsodie: 3.0 ± 1; SNR 300: 152 (abs.); SPX-1: 7-11 kPa; SNR 2: *; SPX- 2: *; *- Slightly above/below atmospheric.

[d] Provided for maintenance; [e] Inflatable seals were tried following large leakage (20-40 l/h) in lip seal; [f] Completed in 1985 but not put into operation; [g] Planned for DFBR.

[@] NaK coolant, metallic fuel; [#] Metallic fuel; [~] Helium cover gas at a later stage. All other FBRs are MOX fuelled, sodium cooled, argon cover gas at a much later stage i.e. during conceptualization of the representative MOX fuelled EFR (1988-1993, Table 2.1) from 6- nation (Belgium, France, Germany, Italy, Netherland, UK) Western European (WE) collaboration (1984) which aimed at economy of scale (< 1.3 times of LWR), sharing of R & D, licensability across Europe, construction within 5 y and safety through standardisation.

2.2.4 Absence of finite element analysis based design and international experience

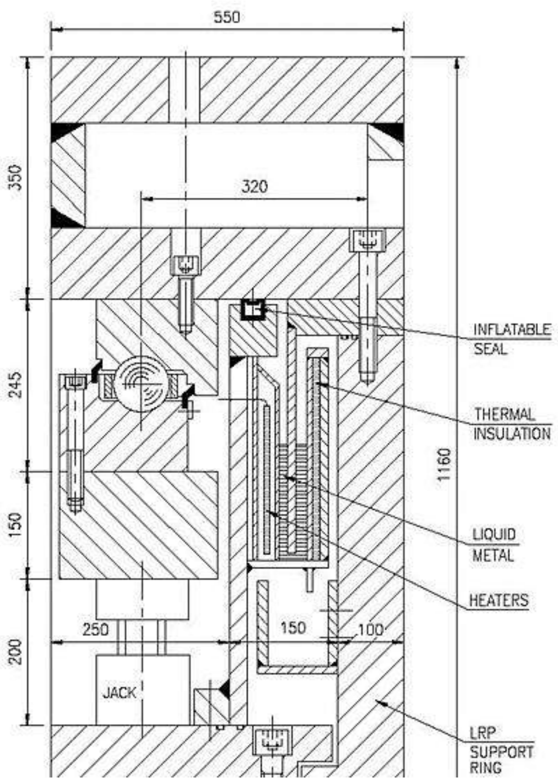


Fig.2.1. Mid- 1980s 4- loop, INDIAN- SFR large rotatable plug design with liquid metal freeze (primary)and axial inflatable (secondary) seals.

Sizing and design of elastomeric barriers by performance trials in scaled test rig was one of the main reasons for long developmental span (late 1960s to early 1990s) of FBR RP inflatable seals (Table 2.1). Non- adoption of FEA in American, European and Japanese FBRs could be attributed to already developed major test facilities and knowledge base, awaiting to be taken to their logical end. Sizing and design of critical elastomeric sealing cannot be addressed by closed form solutions even for the simplest of cases because of material, geometric and contact nonlinearity coupled with strains often lying at the non-linear zone of stress- strain curve (> 10%). Using closed- form/ exact solutions` or empirical relations for design cannot assure meeting the specification the fast time resulting in trial- error based tests and time as well as budget uncertainties. Absence of exact causative relation between load and performance necessitates repetition of the whole process again if minor design change is demanded. These could be avoided by FEA [44].

2.2.5 All- elastomer sealing and development of special elastomeric components

Failure of chloroprene rubber (CR) inflatable seals in FBTR RPs (1985) inspired the planned, collaborative initiative for indigenous development of *Special Elastomeric Components* at the dawn

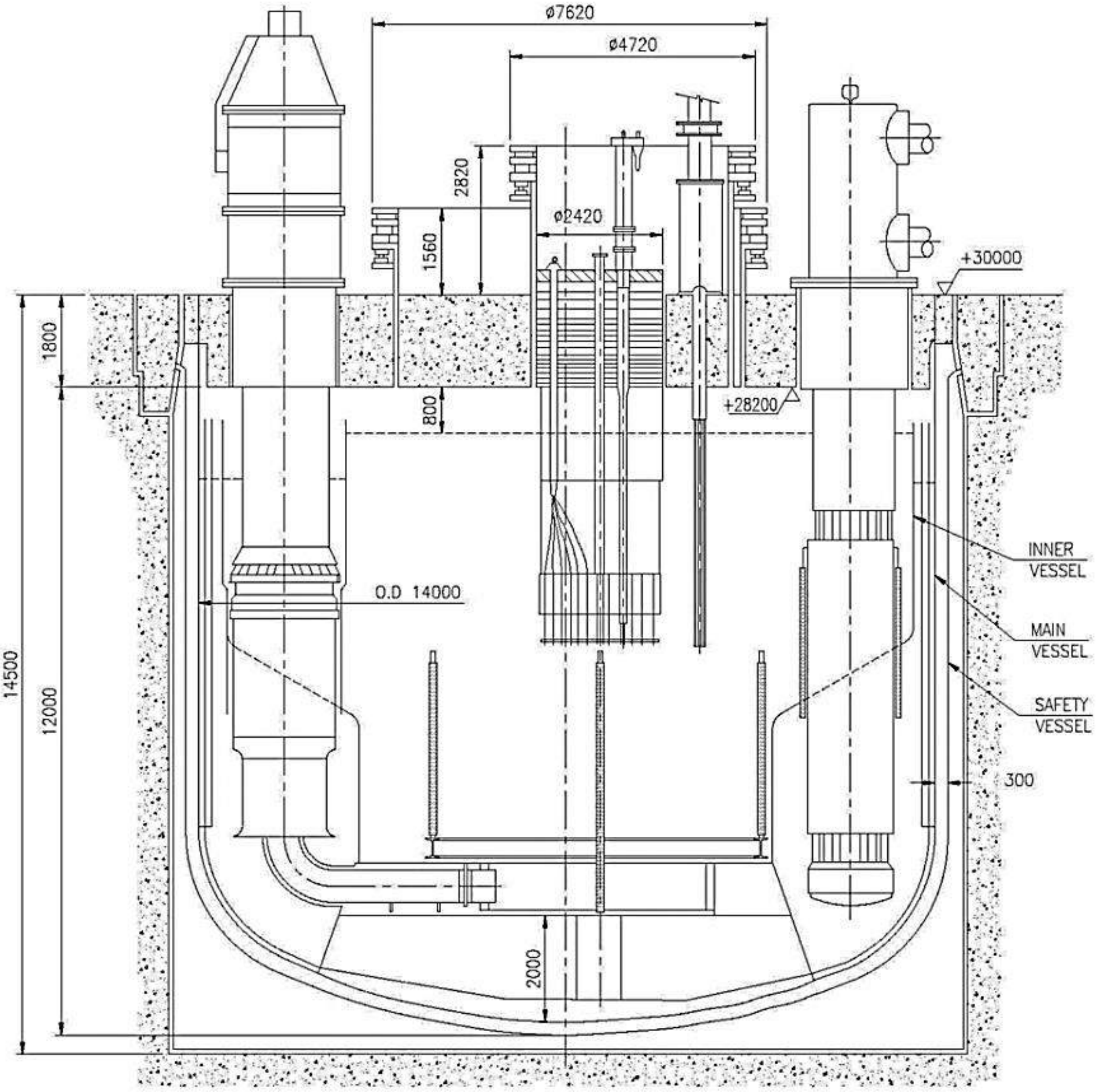


Fig.2.2. Reactor Assembly of 4- loop PFBR design.

of new millennium which was preceded by adoption of all- elastomeric sealing scheme for INDIAN-SFR RPs (early 1990s) consequent to the review of mid- 1980s, 4- Loop INDIAN- SFR design. Large width of LM freeze seal (Fig. 2.1) in the mid- 1980s design resulted in larger diameter overlapping plugs (Fig. 2.2; instead of directly supported by LRP and RS, Fig. 2.3) which increased plug heights substantially along with accompanied increase in stress, instability, cost and component (ARDM, in vessel transfer machine or IVTM etc.) alignment demands.

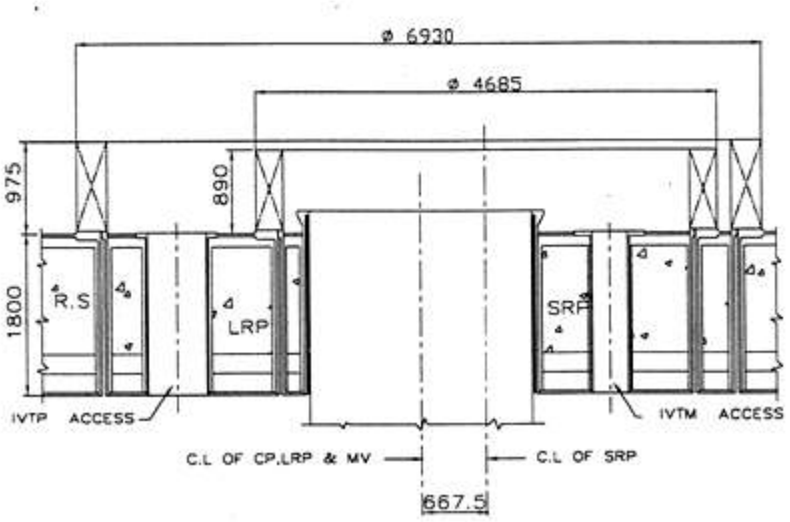


Fig.2.3. Pre- finalized, 2- loop, INDIAN- SFR rotatable plug support arrangement with small rotatable plug within large rotatable plug and supported by top shield top plate.

Significant betterment of safety (lower stress- instability, better alignment etc.), life (lower stress, slower ageing rate etc.) and economy (smaller size, longer life etc.) on these counts was brought in by RP sealing scheme (inflatable- backup, Fig. 1.3) in the present 2- loop INDIAN- SFR design (2 RPs, transfer arm, warm roof)- in line with the post-1980s global trend of enhanced FBR economization and redefined safety paradigm (Section 1.2.4). Substantial reduction (Figs. 1.2, 2.2- 2.3) of plug heights above TS (LRP/SRP, from 1.56 m/2.82 m to 0.685 m/0.65 m), MV diameter (> 1 m; Figs. 1.2, 2.2) and LRP/SRP diameters (~ 0.7m maximum) was a result of the new elastomeric sealing scheme bringing the plug bearings at about the same elevation (SRP within LRP) in pursuance of the objectives of enhanced simplicity, compactness and maintainability during the design review. Late 1990s estimate indicated a capital cost saving of > 10 crore from reduction of RP and MV diameters alone which could provide an idea about the gain from all fronts in the present context. Uniqueness of elastomers, specialty of nuclear applications[11] and absence of tailored indigenous solutions provided threshold impetus for taking up the planned initiative on *Special Elastomeric Component* development, spearheaded by development of RP inflatable seal.

2.2.6 Usage and replacement of elastomer seals in FBTR

A survey (carried out during the first decade of new millennium, Table 2.2) on usage and replacement of ~5000 elastomeric O- ring seals based on 5 generic elastomers and made of more

[11] Radiation, uncommon fluids, inaccessibility, maintenance hazards and long operation. The scarcely available compound- process specific elastomer and their design data in handbooks or commercial product literature are rarely qualified for uncommon fluids, radiation and synergistic ageing from long usage under nuclear - specific conditions.

than 20 compound formulations indicates ~ 17% seal replacement per year. Multiplicity of classes and compounds comprising of high temperature (FKM, MQ) and low temperature (Acrylonitrile-butadiene rubber or NBR, Styrene- butadiene rubber or SBR, CR) elastomers and the possible presence of spurious compounds were reflected in differing and unpredictable lifespans of seals which suggested improvements similar to those in Canadian PHWRs, German FBRs and EFR. R &

Table 2.2. Summary of data on usage of elastomeric O-ring seals in FBTR

Place of Use	Base Polymer	CR	FKM	MQ	NBR	SBR
Block Pile	Chord dia. Range (mm)	1.9 – 7	1.9 – 7	2.5– 10	1.9 – 14	1.9 – 16
	Ring dia. Range (mm)	47 - 330	9.5 - 360	47 - 515	6.4 - 2140	4.9 - 3713
	No. of different sizes	15	60	105	45	30
	Expected seal life	3 years	15 years	15 years	3 years	3 years
	Total no. of seals used	220	760	1550	537	220
Other Areas	No. of different sizes	150	150	---	100	---
	Total no. of seals used	~ 600	~ 600	---	~ 400	---
Approximate average seal replacements per year in all areas		274	61	104	314	74

D on cover gas seals indicated that there is a broader necessity of placing the seals on a common foundation apart from developing and upgrading individual items.

2.2.7 R & D model from special elastomeric component development

Results from *Special Elastomeric Component* development (with R & D prior to and part of this thesis as subsets, to be demarcated in Chapter 3) provided a model to reduce and standardise indigenous nuclear elastomeric R & D based on FKM/EPDM (INDIAN- SFR/ FBTR) inflatable and FKM backup seal (INDIAN- SFR) implementations *(withstanding pre-commissioning conditions in the INDIAN- SFR RPs for the past 6 years)*comprising of design option minimization, short term conventional and new (morphology, topography, tribology, processability from the beginning) specimen studies, fluid compatibility test as the first step for elastomer identification (new approach), FEA based sizing (new approach) to minimize seal testing for design (analogue: specimen studies) and life prediction by WLF and Arrhenius equations (with extrapolation of literature data) using data from synergistic ageing to minimise experiments and analysis (new approach) where material, profile and PECVD Teflon- like coating developments were ab initio in the absence of indigenous commercial resins, compounds, seals or coatings.

Implementation of the model enabled the basic development of inflatable (up to ~ 2m diameter) and backup seals (up to 1m diameter) to be completed within ~ 5 y (FBTR RP inflatable seal: within 3 y) and 2.5 y respectively vis-à-vis more than a decade of development (on an average) for inflatable seals internationally which provided the threshold impetus for pursuing this research[12].

2.3 Objectives

Order of objectives is more of an indication of logical continuity.

I. Taking INDIAN- SFR as the MOX fuelled SFR architype and its cover gas seals as the representative of critical elastomeric sealing.

II. Minimizing number of cover- gas- seal compounds by classification and categorization, defining specification for representative materials, choosing 1elastomer for all categories and tailoring most-demanding-category compound for other cover gas seals to standardise the material module for unification.

III. Extending the unification to sizing, manufacture, coating etc. taking material as cornerstone.

IV. Maximizing manufacturability and life for the pivotal (most-demanding-category) compound to extend the benefits to other cover gas seals.

V. Maximizing predictability of performance and life through appropriate numerical and analytical techniques such as FEA, Arrhenius and WLF methodologies.

VI. Ensuring repeatability and reproducibility of results through automation, standardisation and proper quality control scheme.

VII. Outspreading the concept to metallic fuelled FBRs, PHWRs and AHWR to encompass the Indian nuclear program.

VIII. Relevance of research in multidisciplinary domains (beyond nuclear) and global context.

[12]The comprehensive and standardised R & D scheme for future implementation of cover gas seal unification, which forms the 8th element as an outcome of this thesis, is a result of international experience, indigenous R & D and the research carried out as part of this thesis. Results of *Special Elastomeric Component* development showed that such an approach for R & D standardisation in all fronts is feasible.

Chapter 3

Literature Review and Problem Definition

3.1 Defining the Problem Through Review: *An outline* [9- 10, 43]

Evolving FBR design and scientific- technological understanding of rubber, uniqueness of elastomers, proprietary ownership, specialty of nuclear applications coupled with very limited usage of predictive techniques (FEA, Arrhenius and WLF equations) and comprehensive quality measures (spanning laboratory to reactor) were key reasons for extensive research in the area of TS sealing spanning more than four decades (1950s to early- 1990s). Development of LM (1950s to mid-1980s) and inflatable seals (late- 1960s to early- 1990s) for RPs consumed maximum effort.

Elastomer seal usage and replacement report of FBTR (Table 2.2) and R & D results from *Special Elastomeric Component* development, supported by literature review in nuclear and non-nuclear field, provided a defining outline (Sections 2.3, 3.14) for the present research. Absence of a comprehensive scheme encompassing all aspects of critical elastomeric nuclear sealing and exploiting the elastomer- sizing- manufacture- coating correlations was evident from the surveys. Developments pertaining to nuclear organic components (components and fluids made of organic materials) in general and FBR sealing in specific that contributed significantly in identifying the gap areas are listed below with representative references.

i. ***FBR Seals: USA*** – Planned initiative on cover gas seal development *(most comprehensively reported)* for USA 1000 MWe Liquid Metal Fast Breeder Reactor (LMFBR) with FFTF and CRBRP as immediate focus, carried out from 1968 to late- 1970s [46- 48]. *Representative references being too many, are distributed in subsequent text.*

ii. ***FBR Seals: other parts of the world*** – Development of inflatable seal (1967- 1993) as part of TS barriers for French, German and Japanese FBRs and EFR [49- 50]. *Representative references being too many, are distributed in subsequent text.*

iii. ***PHWR Seals:*** Upgradation of elastomeric sealing for Canadian PHWRs, initiated during 1960s with the objective of improving performance, safety margins and life-predictability and continued during 1990s [51].

iv. ***LWR Seals:*** Comprehensive R & D on elastomers and seals as part of containment structural integrity research, initiated at Sandia National Laboratories during 1982 following the Three Mile Island accident, Unit 2 (March, 1979) and continued during the first decade of 21st century [52- 54].

v. ***Seals for nuclear fuel transport flask:*** Research on elastomeric seals used in demountable lid-to-body joints of the flasks [55- 56].

vi. ***Organic components in NPP:*** Ageing and performance of components and fluids made of organic materials (elastomers and other polymers of varying molecular weight) in the form of seal, gasket, cable, hose, insulation, lubricant, coating, adhesive and seismic isolator [57- 58].

vii. ***Ageing and life cycle management:*** Comprehensive, planned, long- term studies (in parts) on ageing and life of organic components, spearheaded (1985) by the drive from International Atomic Energy Agency with accelerated ageing (and Arrhenius methodology) on specimens at the core to ensure replacement after maximum predictable interval in view of life extension of LWRs and increasing demands on safety norms [59- 66].

viii. ***Seals and materials for nuclear package casks:*** Comprehensive, long- term (more than a decade) accelerated ageing and life- prediction studies of radioactive material-package-cask-seals (particularly O- rings) including shipping packages resulting in adoption of peroxide cured APA Viton® rubber (pivotal elastomer of this research) based on extensive characterisation [67- 73].

This Chapter assumed unusual length for reasons similar to Chapter 1. Necessity to provide sufficient material in support of data extrapolation, design and logical continuity is a complimentary cause. Requirement to demarcate this research and complimentary R & D results (referred) from *Special Elastomeric Component* development is the other area.

Table 3.1. Summary of INDIAN- SFR cover gas seals and their operating conditions

Items	Location	Type	Dia. (mm)	NOS	Engaged	FIC	MIC	RSOM (mm/s)	CR (m)	MP (kPa)	NT(^{0}C)
Static O-ring Seals	RPs	Static	6300	12	NO & FH	D	P	---	---	40	110
	CP	Static	2400	2	NO & FH	D	Q	---	---	70	120
	IFTM	Static	4000	2	NO & FH	D	Q	---	---	5	150
	PI/ARDM	Static	4000	4	NO & FH	D	P	---	---	5	150
Dynamic O-ring Seals	FFIM	Rotary	76	20	NO & FH	D	Q	0.9	1300	70	120
	CSRDM	Rec.	34/69	18	NO & FH	D	S	6	3	60	100
	CSRDM	Rotary	15/50	54	NO & FH	B	R	70	5000	70	80
	DSRDM	Rotary	10/50	24	NO & FH	B	R	139	5000	70	80
Dynamic V-ring Seals	# LRP	Rotary	6300	1	NO & FH	B	P	! 75/2	*5200	30	90
	# SRP	Rotary	4200	1	NO & FH	B	P	! 75/2	*1800	30	90
	@ LRP	Rotary	6300	1	FH	B	P	! 75/2	*5200	0.5	90
	@ SRP	Rotary	4200	1	FH	B	P	! 75/2	*1800	0.5	90
	CSRDM	Rec.	110	18	NO & FH	C	Q	! 2/4000	130	60	120
Dynamic Lip Seals	DSRDM	Rec.	90	6	NO & FH	C	Q	4	100	60	120
	IFTM	Rotary	4000	2	FH	C	Q	15	*100	0.5	150
Special Lip Seals	@ LRP	Static	6300	2	NO	C	P	---	---	0.5	100
	@ SRP	Static	4200	2	NO	C	P	---	---	0.5	100
Inflatable Seals	# LRP	Rotary	6300	2	NO & FH	C	P	! 75/2	*5200	30	110
	# SRP	Rotary	4200	2	NO & FH	C	P	! 75/2	*1800	30	110
	@ LRP	Rotary	6300	1	FH	B	P	! 75/2	*5200	0.5	80
	@ SRP	Rotary	4200	1	FH	B	P	! 75/2	*1800	0.5	80

Note:
Dia. Stands for seal ring diameter
Values of dia. and CR are approximate
OC for all seals comprise of 8 months of NO followed by 20 days of FH. The OC repeats during seal life
RRO of LRP during FH: 0^0-360^0-0^0
RRO of SRP during FH: 0^0-180^0-0^0
RRO of IFTM during FH: 0^0-180^0-0^0
For all the cases cumulative γ dose at seal locations $\leq$ 2000 Gy over a period of 10 years

Symbols

A- Air
B- Argon + A;
B1- B + Xenon + Krypton
C- B + Sodium Aerosol
C1- C + Xenon + Krypton
D- Argon + Sodium aerosol
E- B + Silicone oil vapour
P- Carbon Steel (CS)
Q- Stainless Steel (SS)
R- P + Aluminium-bronze
S- Q + Aluminium-bronze
X- CS ASTM A516+ Teflon
Y- SS 304L + Teflon
@- RP Sealing Scheme 1
#- RP Sealing Scheme 2
***-** CR per FH session
! - Indicates two speeds of reciprocation or rotation

Acronyms applicable to all tables

CDD- Cumulative design (γ) dose (at seal/coating location during DL)
CDT- Cumulative design travel (between seal and mating surface during DL)
CR- Cumulative rubbing (between seal and mating surface per year or per FH session)
DCP1- Design contact pressure (on coatings)
DD- Design (frictional) drag
DL- Design life
DP- Design pressure (differential, across the seal)
DT- Design temperature (at seal/coating locations)
FH- Fuel handling
FIC- Fluids in contact (with seal)
GS- Gap (to be) sealed
MIC- Materials in contact (with seal)
MP- Maximum pressure (differential, across the seals)
NO- Normal operation
NOS- Number of seals
NT- Normal temperature (at seal location)
OC- Operating Cycle
Rec.- Reciprocating
RRO- Range of rotary oscillation
QC- Quality control
RSOM- Relative speed of motion (between seal and mating surface)
VTSR- Validatory test in scaled rig

3.2 The INDIAN- SFR Sealing System

3.2.1 Summary of cover gas seals [9-10, 36, 43]

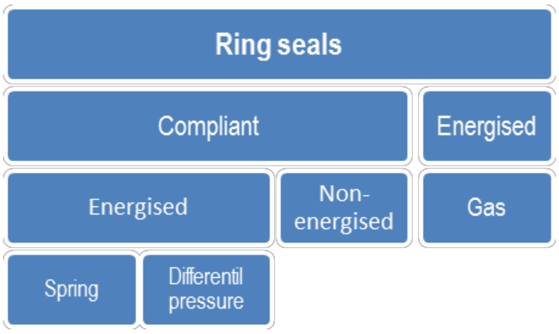


Fig.3.1. Classification of ring seals.

Table 3.1 shows a summary of cover gas seals, representative of important elastomeric sealing of INDIAN- SFR. For a category of seal corresponding to a particular location, the largest seal diameter and the most demanding combination of fluids are listed. Sealing scheme of RPs and inclined fuel transfer machine (IFTM) underwent transitions in concepts involving lip and V- ring seals before arriving at the inflatable- backup combination.

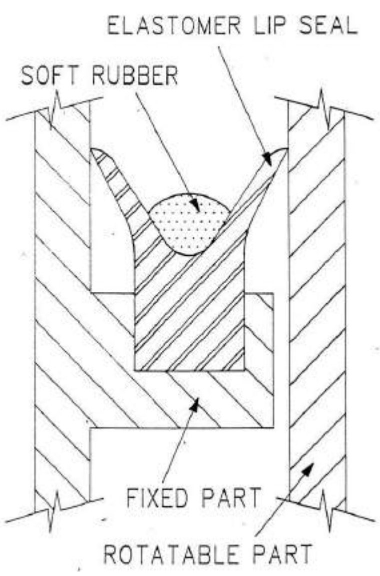


Fig.3.2. Elastomeric lip seal stiffened at base.

INDIAN- SFR TS seals are generally used in pairs, with fresh argon gas (at inter-seal space) pressurised above cover gas by ~ 10 kPa and RCB air by ~ 20 kPa. Fig. 3.1 shows classification of metallic and elastomeric ring seals of C/K/V and O/Q/rectangular/T/quad/lip/V cross-sections respectively. Cover gas elastomeric barriers in FBRs (Table 2.1) have traditionally been mostly non- reinforced (by spring etc.) in order to exploit the advantages of simplicity, economy and low assembly load- friction-damage while reducing dimensions of load- bearing shells and flanges.

Compliant seals (Fig. 3.1), with initial interference (squeeze) at seal- counterface contact determining the engagement load (initial contact pressure) and friction, get negligible energization unless seal geometry (cantilever arm in lip or V- ring seal; Fig 3.2) allows the low pressure cover gas to add to the initial contact pressure.

Compounds of moulded and extruded seals, even if made of the same base (generic) polymer (elastomer), could differ in composition. Difference in hardness of static (typical value: 50±5 ^{0}Shore

A) and dynamic (typical value: 75 ± 5 ^{0}Shore A) elastomeric seals also indicate different compositions. Inflatable seal is grouped separately as CS is not a failure criterion. CS (Fig. 3.3) is the time dependent decay of initial compression (with load, stress and contact pressure), induced by relaxation (loss of stress under constant strain) and creep (deformation under constant load) in elastomers during ageing. CS could be compensated by gas pressure from external supply in inflatable seal (Figs. 1.5- 1.6) in order to maintain the contact pressure above differential pressure for bypass leak-tightness (Fig. 1.4).

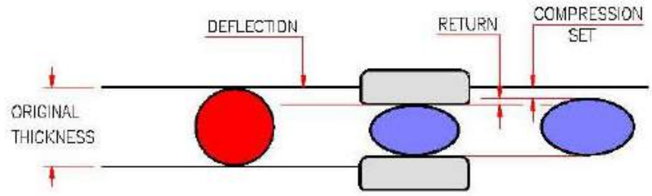


Fig.3.3. Compression set in elastomeric O- ring.

Table 3.2. A preliminary categorization of cover gas elastomeric sealing of INDIAN- SFR

Groups	Production process	Development starts with	Choice of base polymer from	Major areas of development
Large dia. static O-ring seals	Extrusion	Identification of proper base polymer and elastomer compound formulation for each group	Ethylene-propylene-diene- methylene rubber, Fluorocarbon rubber, Silicone rubber, Fluorosilicone rubber	Material, sizing by FEA, end-joining, VTSR, QC
Small dia. dynamic O-ring seals	Moulding			Material, lubricant/coating, VTSR, QC
Large dia. V-ring, Lip seals	Extrusion			Material, sizing by FEA, lubricant/coating, production, end-joining, VTSR, QC
Small dia. V-ring, Lip seals	Moulding			Material, sizing by FEA, lubricant/coating VTSR, QC
Inflatable seals	Extrusion			Material, sizing by FEA, lubricant/coating, production, end-joining, adhesive, inflation connector, seal installation/removal, VTSR, QC

CS and compression stress relaxation (CSR) of 100 % are often used interchangeably as the threshold for inception of failure in bypass leakage mode (Fig. 1.4) in non- energized, compliant, static seals (Fig. 3.1). Non- reinforced elastomeric O-rings require compounds with highest CS resistance as there is negligible energisation from low- differential- pressure cover gas (INDIAN- SFR maximum: 70 kPa). Static and dynamic V-ring and lip seals (Table 3.1) fall in between the above two (inflatable and non- reinforced O- rings, Table 3.2) as sealing is both by compliance and energisation and shrinkage as well as Joule-Gough effect have damaging influences on dynamic O-rings (class/compound specific). Table 3.2 shows an initial categorization of elastomeric sealing for INDIAN- SFR cover gas (Table 3.1).

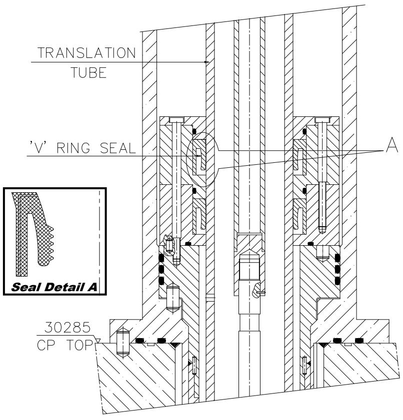


Fig. 3.4. Reciprocating elastomer V-ring seals of control and safety rod drive mechanism.

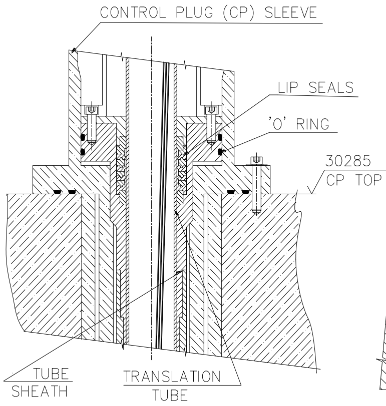


Fig. 3.5. Reciprocating elastomer lip seals of diverse safety rod drive mechanism.

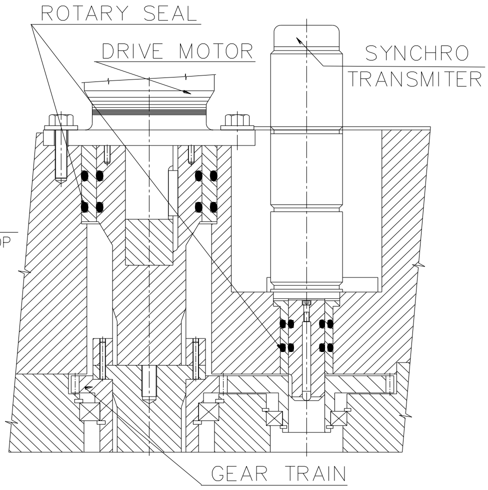


Fig. 3.6. Rotary elastomeric O-rings of control and safety rod drive mechanism.

3.2.2 Outline of categories [36, 74]

The present categorization of INDIAN- SFR cover gas seals is based on two changes vis-à-vis Table 3.1 i.e. i) A combination of 2 inflatable and 1 backup seals as primary and secondary barriers in RPs (Fig.1.3) and, ii) 3 inflatable seals as primary and secondary barriers in IFTM.

V-ring and lip seals (Figs. 3.4- 3.5) for 9 control and safety rod drive mechanisms (CSRDMs) and 3 diverse safety rod drive mechanisms (DSRDMs) of INDIAN- SFR (Table 3.1) form a part of fourth group (Tables 3.2). V-ring seals as primary barriers (Fig. 3.4) experience small rubbing every day because of reciprocation of translation tube during reactor power control. The rubbing speed may reach a maximum of 4m/s during emergency shutdown. Lip seals as primary leak-resistant barrier (Fig. 3.5) experience rubbing only before and after a planned/ unplanned shutdown (excepting emergency shutdown) exercise because of reciprocation of translation tube**.** Figs. 3.6–3.8 show locations and arrangements of rotary and reciprocating elastomeric O-ring seals of CSRDM and rotary elastomeric O-ring seals

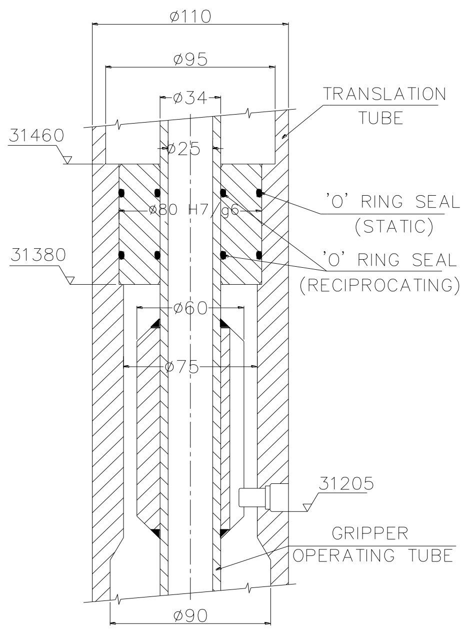


Fig. 3.7. Reciprocating elastomeric O-rings of control and safety rod drive mechanism.

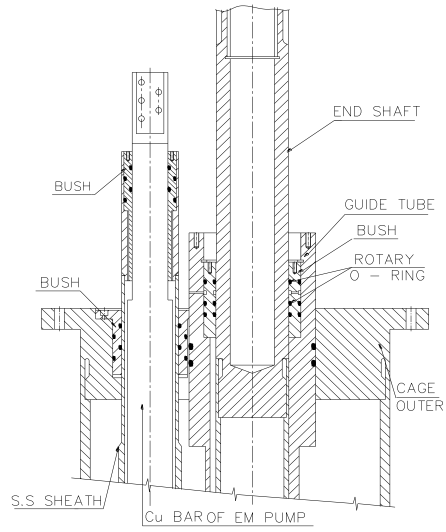


Fig. 3.8. Rotary elastomeric O-rings of failed fuel inspection module.

of failed fuel inspection module (FFIM) of INDIAN- SFR (Table 3.1) which form part of the second group (Table 3.2).

Fig. 3.9 depicts the locations and arrangement of large diameter static elastomeric O-ring seals of INDIAN- SFR SRP (Table 3.1) which form a part of the first group (Table 3.2).

Fig. 3.9 also shows the arrangement of inflatable and backup seals in INDIAN- SFR SRP which is identical with that in LRP, excepting the seal diameters. Fig. 3.10 shows the arrangement of inflatable seals in IFTM. Radially mounted inflatable seals bonded to the bases of rectangular grooves in top and middle ring (TMR, Fig. 3.9) and support table (Fig. 3.10) inflate away from RP and IFTM axes to engage with bearing support ring (BSR) and leak tight cell (LTC) respectively for preventing out leakage of radioactive cover gas to RCB air and ingress of RCB air into the cover gas space through axial openings of 5 ± 2 mm width

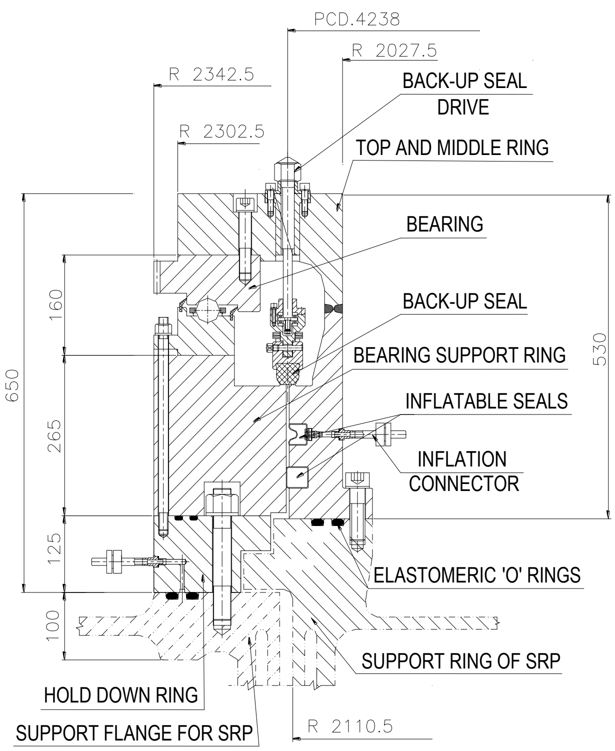


Fig. 3.9. Location and arrangement of inflatable and backup seals in INDIAN- SFR small rotatable plug.

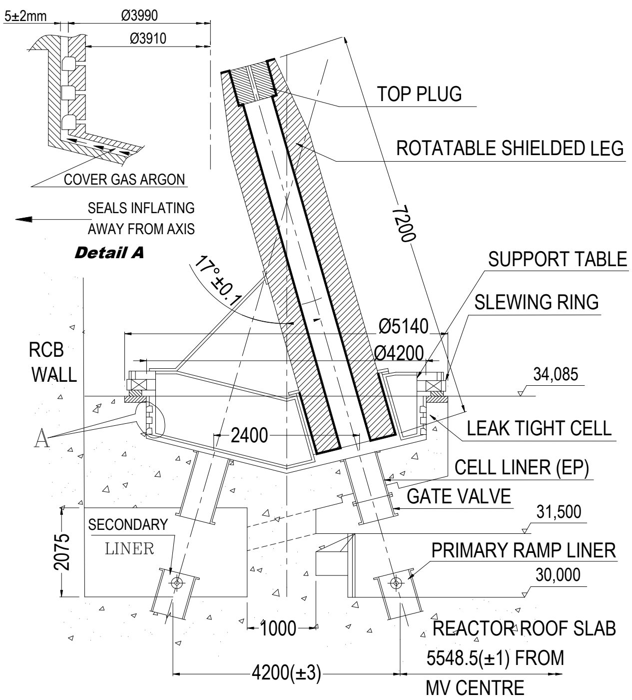


Fig. 3.10. Location, arrangement and mounting of inflatable seals in INDIAN- SFR inclined fuel transfer machine.

between shells. RPs of INDIAN- SFR hang on bearings all the time and oscillate intermittently (oscillation range- LRP: 0–360–0^0, SRP: 0–180–0^0) at a maximum speed of 75 mm/ s during fuel handling condition of reactor (after every 175 full power days of normal operation or after every 240 days, assuming a capacity factor of 75%) when the seals, treated by Teflon- like coating on rubbing surface (Fig. 3.11), rub against the BSR (Fig. 3.9), also coated with Teflon (Fig. 3.12). IFTM inflatable seals (Fig. 3.10) and the mating LTC (Fig. 3.13) are similarly coated.

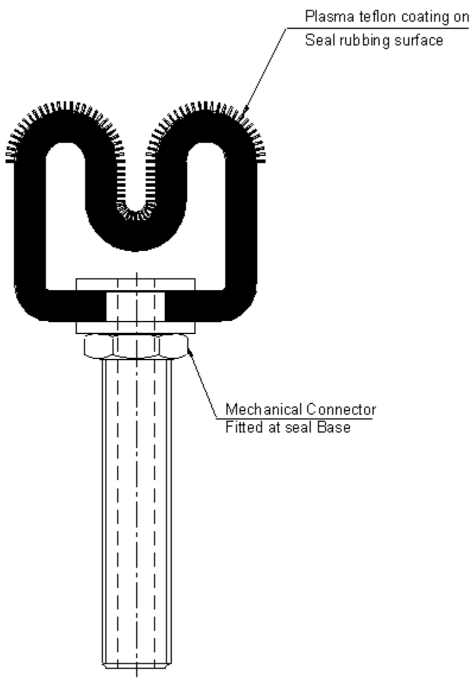


Fig. 3.11. Coating location on rubbing surface of inflatable seal.

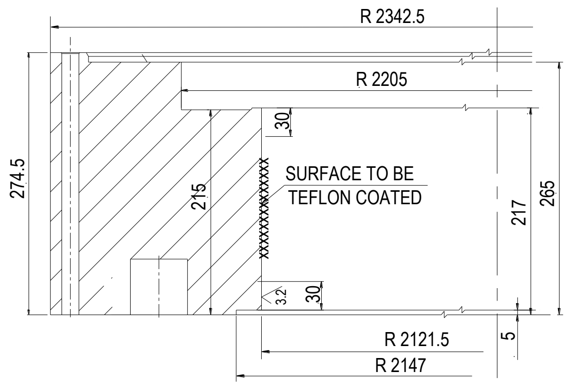


Fig. 3.12. Coating location on bearing support ring of INDIAN- SFR small rotatable plug.

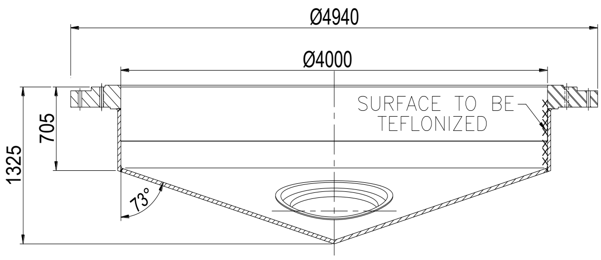


Fig. 3.13. Coating location on leak tight cell of INDIAN- SFR inclined fuel transfer machine.

Lower inflatable seal (Fig. 3.9) is always engaged as primary barrier and exposed to radioactive cover gas laden with mist and fission product gases, xenon and krypton (INDIAN- SFR is designed for 4 or 0.01% of failed fuel). The static backup seal acts as a secondary barrier during normal operation while remaining engaged with the top, horizontal, carbon steel surfaces of BSR and TMR to seal a radial gap of 5 ± 2 mm (Figs. 3.9, 3.14). The seal, held in trapezoidal groove of carbon steel seal holder, is lifted (~ 25 mm) by suitable drive prior to fuel handling to disengage while the upper inflatable seal is inflated to replace the backup seal as secondary barrier. Allowable leakage of 10 $^{-3}$ scc/ s/ m-seal-length and 10^{-2} scc/ s/ m-seal-length are specified for both the seals during normal operation and fuel handling condition respectively.

3.2.3 Specifications

3.2.3.1 Design specifications [36]

Table 3.3 summarizes the finalized design specifications of INDIAN- SFR and FBTR inflatable seals and INDIAN- SFR backup seal based on which R & D and component development were carried out.

3.2.3.2 Material specifications [36]

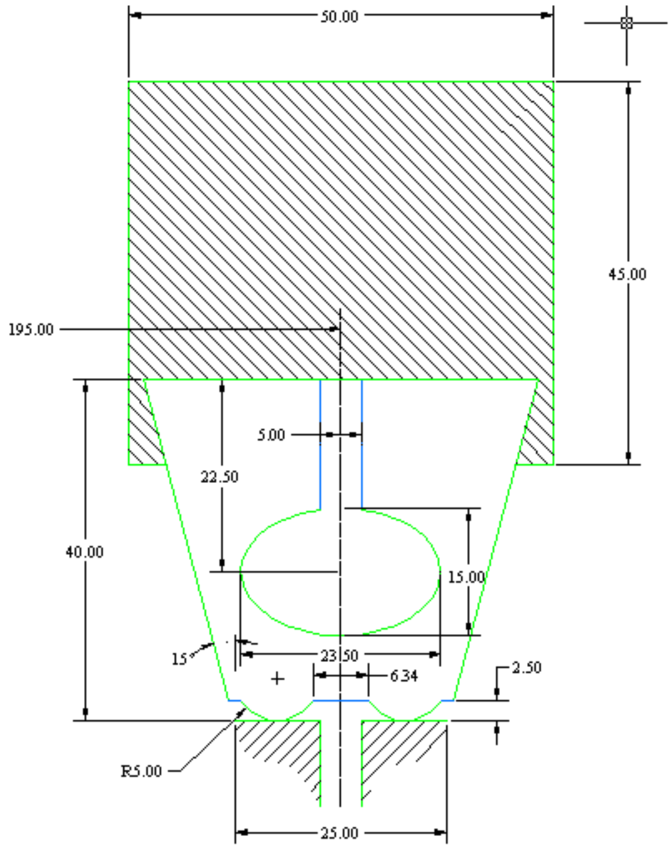


Fig. 3.14. Pre-finalized version of backup seal cross-section.

Table 3.4 shows preliminary specifications for elastomer compounds corresponding to a few important categories of seals where property measurements are done at room temperature (RT) and tests are to be carried out at RT (or

Table 3.3. Finalized design specification of INDIAN- SFR and FBTR seals used for development

Items	Location	Type	Dia. (m)	NOS	Engaged during	GS (mm)	FIC	MIC	DP (kPa)	DT (^{0}C)	CDD (Gy)	RSOM (mm/s)	CDT (km)	DD (N/m)
FBTR RP IF Seal	LRP	Rotary	3.9	1	NO & FH	3±1	E	P	10/1	80	---	14	10	1000
	SRP	Rotary	2.4	1	NO & FH	3±1	E	P	10/1	80	---	7.2	10	1000
INDIAN-SFR RP IF Seal	LRP	Rotary	6.3	2	NO & FH	5±2	C1	X	25/2.5	120	2000	! 75/2	100	1000
	SRP	Rotary	4.2	2	NO & FH	5±2	C1	X	25/2.5	120	2000	! 75/2	50	1000
INDIAN-SFR RP Backup Seal	LRP	Static	6.4	1	NO	5±2	B1	X	25	110	2000	---	---	---
	SRP	Static	4.2	1	NO	5±2	B1	X	25	110	2000	---	---	---
INDIAN-SFR IFTM IF Seal	IFTM	Rotary	3.9	3	NO & FH	5±2	C1	Y	25/6	150	2000	! 14/7	50	1000

Values of dia. are approximate
OC: 8 months of NO followed by FH repeating during seal life
RRO of INDIAN- SFR LRP, SRP, IFTM same as indicated in Table 3.1
All seals have DL of 10 years

Allowable leakage for all seals during NO and FH (excepting backup seal which is disengaged during FH) are 10^{-3} and 10^{-2} scc/s/m length of seal respectively
1st and 2nd DP (wherever mentioned) correspond to NO and FH respectively
DD between seal and mating surface correspond to per meter length of seal

Table 3.4. Preliminary material specifications

Parameters	Inflatable seal	Dynamic V/Lip seal	Dynamic O-ring seal
T_s(MPa)	13 (min.)	8.3 (min.)	8.3 (min.)
Hardness (^{0}Shore A)	65 ± 5	70 ± 5	75 ± 5
E_b (%)	150 (min.)		
Tear Strength (N/mm)	18 (min.)		
Resilience (%)	58 (Yerzley, 3.7 Hz) min.		
Hysteresis (%)	50 (Yerzley, 3.7 Hz) max.		
Splice Strength (MPa)	13 (min.)	8.3 (min.)	---
Radiation Resistance (kGy)	100		
Fluid Compatibility (swell/shrink limit in %)	≤ 20/5 under C exposure		≤ 15/3 under C exposure
Material Compatibility (shall not corrode)	P	P, Q	Q, R, S
Ozone Resistance	Shall have resistance to ozone attack		
Compression Set (%;in air at 150^{0}C for 70h)	≤ 25	≤ 20	≤ 10

Permeation leakage of inflatable seal material not to exceed 10^{-6} cc/s at RT and under differential pressure of 100 kPa when measured across thickness of an elastomeric sheet of 100 mm x 25 mm x 2 mm.

at standard laboratory temperature of 23 ± 2 ^{0}C; ASTM D1349) unless specifically mentioned. The specifications provide a framework for identification of suitable elastomeric compound (from a number of candidate compounds with compositional variations) for each category by using the upper and lower limits indicated.

3.2.3.3 Coating specifications [36]

The development of plasma based coating was initiated based on a design specification (Table 3.5), similar to the design specification for seals (Table 3.3). The cumulative distance of travel for coating durability corresponds to 10 y of

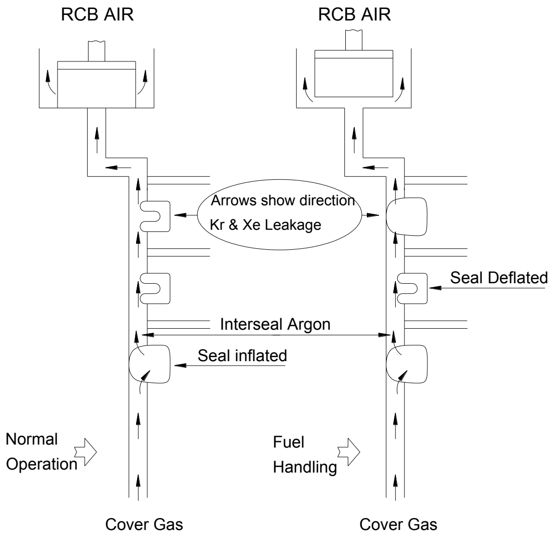


Fig. 3.15. Permeation leakage of fission product gases through inflatable and backup seal walls during normal operation and fuel handling conditions.

Table 3.5. Design specification for coating on inflatable seals and mating shells

Parameter	FBTR	INDIAN- SFR			
	RP Seal	RP Seal	RP Shell	IFTM Seal	IFTM Shell
Coating on	Seal rubbing face		BSR	Rubbing face	LTC
OC	Same as indicated in Table 3.3. The coated surfaces (seal rubbing face & mating surface) remain in static contact during NO and rubs with respect to each other during FH condition.				
RRO	As indicated in Table- 3.3				
RSOM	As indicated in Table- 3.3				
CDT (km)	10	100	400	50	200
FIC	E	C1	C1	C1	C1
MIC	P + EPDM	X + FKM	X	Y + FKM	Y
DCP1(kPa)	NO: 70; FH: 50				
DT (^{0}C)	80	120		NO: 35; FH: 150	
CDD (Gy)		2000	8000	2000	8000
DD (N/m)	1000	1000	2000	1000	3000
DL (years)	10	10	40	10	40

seal life and 40 y of shell (reactor) life which amounts to 10 km, 50 km and 100 km for the inflatable seals of FBTR RPs, INDIAN- SFR IFTM and INDIAN- SFR RPs.

3.3 Identifying Inflatable and Backup Seals as Representatives of Cover Gas Barriers

3.3.1 Identification by first principle: Intuitive mode [9, 37, 43]

3.3.1.1 Functionality and operating demands

Figure 3.15 shows permeation leakage of fission product gases through an abstraction of the inflatable- backup sealing arrangement at INDIAN- SFR RPs where an additional inflatable seal is added in between. Figure 3.16 shows the configuration and radial mounting (inflating perpendicular to plug axis) of low pressure inflatable seal at INDIAN- SFR RPs where the fold (perpendicular to seal base) in seal profile acts as a reserve of extra material length to unroll during inflation and engage with the mating surface using minimum inflation pressure and seal stress. Figure 3.17 indicates that backup seals in INDIAN- SFR RPs are to seal a maximum level

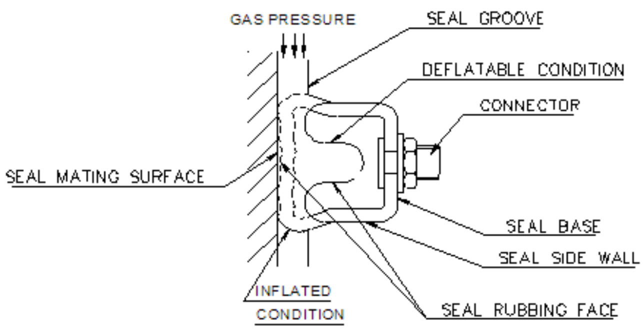


Fig. 3.16. Configuration and radial mounting of INDIAN- SFR rotatable plug inflatable seal.

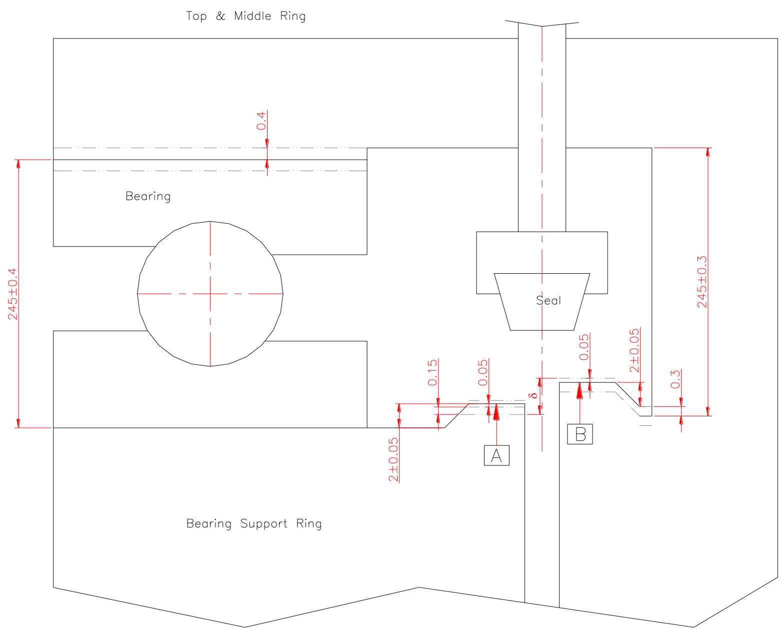


Fig. 3.17. Schematic of magnified view on fitment and arrangement of backup seal in INDIAN- SFR rotatable plugs showing level mismatch between mating surfaces.

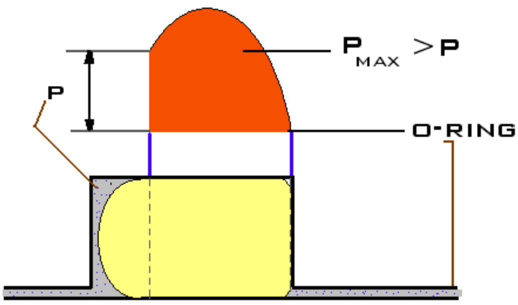


Fig.3.18. High differential pressure (P) adding with initial contact pressure (P_{MAX} – P) in the deformed configuration of elastomeric O-ring.

mismatch of 2 mm between the top mating surfaces of BSR and TMR because of fabrication and assembly tolerances.

3.3.1.2 Maximum operating demand and design make the seals representative

The representative nature of INDIAN- SFR RP inflatable and backup seals in static and dynamic categories are substantiated by maximum demands on material performance.

Tensile stress- strain zone in seals responsible for cracking failure (stress- strain more than T_s- E_b) is higher for unconstrained or nearly unconstrained sealing configuration (i.e. backup seal; Figs. 3.14 and 3.17) compared to the constrained ones i.e. O- ring seal, Figure 3.18 [75]. Backup seal requires elastomer with highest CS resistance amongst TS static seals for the same life as the design requirement (seal getting disturbed prior- to and after fuel handling for disengagement and engagement respectively) indicates that the threshold CS for leak-tightness should

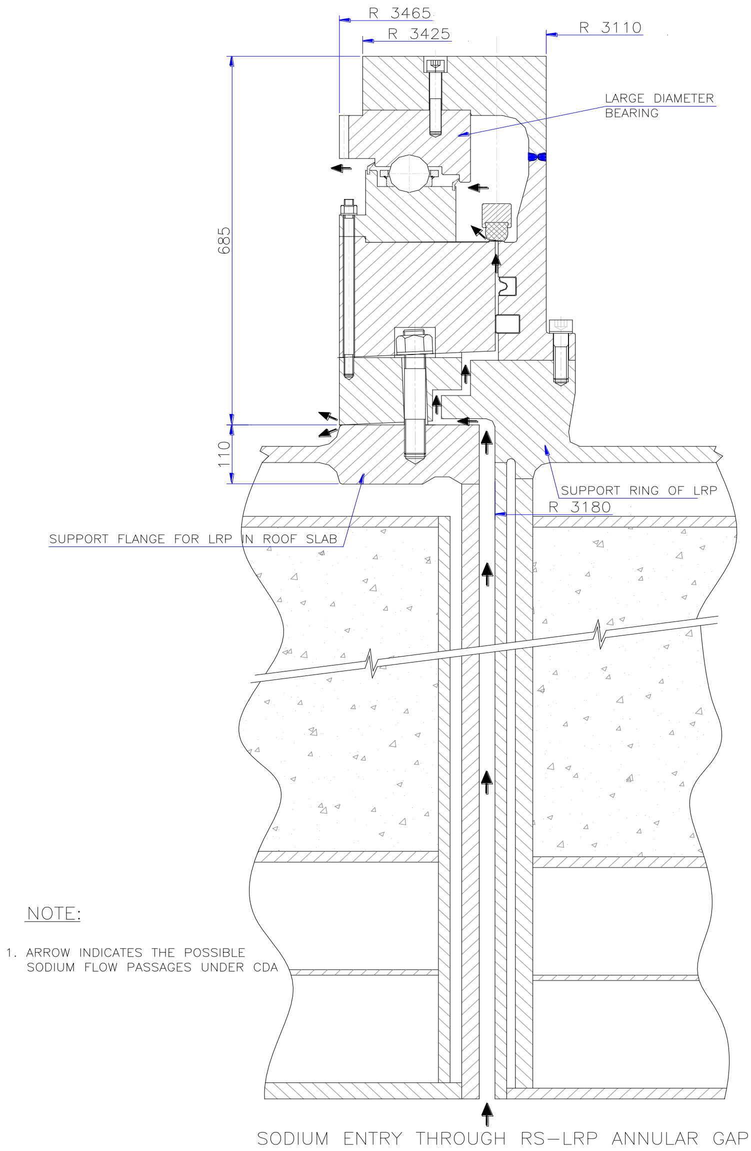


Fig. 3.19. Typical annular and radial passages between shells and flanges of INDIAN- SFR top shield and rotatable plugs providing paths for release of sodium coolant and radioactive argon cover gas to Reactor Containment Building air during core disruptive accident along with indication of flange rotations (at O- ring contact) under a differential pressure of 2.1 MPa.

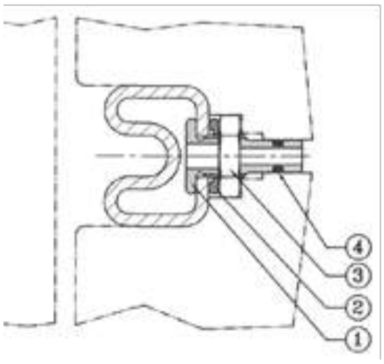


Fig.3.20. Radially mounted fluorohydrocarbon rubber inflatable seal (deflated; wall thickness: 2mm) of INDIAN-SFR fixed at groove (34 x 26 mm) base by adhesive and fitted with mechanical connector at seal base for inflation by argon gas supply. 1) Seal connector, 2) Washer, 3) Lock nut and, 4) O-ring.

be well below 100 %. In most of the compliant (Fig. 3.1), non-reinforced, cover gas static elastomeric barriers (in contact with surfaces of 0.2- 3.2 µm roughness), sealing function continues even after 100 % of CS as the contact- surface- disturbance (from thermal transient, vibration etc.) is not comparable with surface roughness and the differential thermal expansion [80, 107] of elastomer (usually ~ 10 times of steel) is not accounted for in CS, measured at RT. This is illustrated by 15- 20 y of life achieved with FKM/ MQ O- rings in FBTR block- pile and non- block- pile areas (Table 2.2).

Elastomers display liquid- like behaviour at higher pressure (0.7- 1.4 MPa, class- compound specific) and the differential pressure adds hydrostatically [51] with the initial contact pressure (from compliance, Fig. 3.18) to make the seal performance independent of CS [76- 77] i.e. during CDA (Fig. 3.19). Nearly unconstrained configuration of backup seal demands maximum exploitation of elastomer strength (by numerical procedures such as FEA) under such conditions (Fig. 3.19) as stress- strain field is essentially compressive in constrained configurations (such as O rings, Figs. 3.18- 3.19) under such condition and sealing could always be ensured by additional support from flange stiffness and bolt- pretightening to minimise flange rotation at sealing contact (Fig. 3.19).

Thin (2 mm) seal wall, largest surface area to volume ratio, inflation pressure (≥ 45 kPa) more than twice the buffer gas pressure (20 kPa), highest rubbing (100 km) during design life (10 y), presence of stress-raisers in the form of seal–connector joint (Fig. 3.20) and the role as a primary barrier (exposure to mist, static- dynamic operation etc.) to seal gap of largest diameter and width put maximum demand on INDIAN- SFR inflatable seal material vis-à-vis other seals in terms of, i) Permeation resistance, ii) Sodium aerosol compatibility and endurance under synergistic ageing, iii) Resistance to cut/crack initiation and growth and, iv) Wear and tear resistance.

3.4 Extending Intuitive Mode by Literature Substantiation: *Identifying inflatable seal elastomer as the originator of unification*

Literature substantiation on the following counts suggest that elastomer compounds for other cover gas seals should be obtained by tailoring the RP inflatable seal compound.[13].

i. Operating requirements demand maximum number of parallel functionalities from inflatable seal material, not seen by any other cover gas seal.
ii. Inflatable seal involves all material parameters of cover gas seal failure excepting CS.
iii. Choice and arrangement of inflatable seal epitomize various demands from evolving FBR design, sealing scheme, seal design and R & D - not seen in any other cover gas barrier.
iv. INDIAN- SFR and FBTR inflatable seal development imbibed experiences in design, development, operation to minimize R & D efforts, cost and time while maximizing safety.
v. Feasibility of achieving one replacement during reactor life from past experience.

3.4.1 Maximising originator life for uniform benefit and standardisation

Maximizing inflatable seal life with proper compound and then tailoring the compound formulation with marginal changes in ingredients (generic elastomer, resin unchanged) for other cover gas seals amounts to sharing the benefits of the best compound for uniform life maximisation.

*3.4.*2 Literature substantiation

3.4.2.1 Parallel functionality requirements [9, 81, 87]

Elastomer for primary inflatable seal directly exposed to mist in hanging RPs of INDIAN-SFR requires excellent chemical inertness, lowest permeability, outstanding thermal stability, low frictional coefficient (***μ***) and countable mechanical property loss initiation only at about 10 kGy of cumulative γ dose to maximize life. Sodium aerosol compatibility, excellent resistance to oxygen,

[13] The inherent vulnerability of elastomers to low cycle fatigue conditions expected during intermittent rotation of inflatable seals (fuel handling) with a varying annular gap of 5 ± 2 mm, coupled with maximum design drag of 1000 N/ m-seal-length and cumulative design distance of travel (100 km) or rubbing on thin seal wall (with maximum number of potential stress raisers on thin seal wall), makes the elastomeric compound of this seal representative amongst the cover gas barriers in terms of maximum resistance to wear, tear and cut/ crack growth.

ozone, wear, tear, abrasion, synergistic ageing and very good RT properties (T_s, E_b, tear resistance, toughness) with maximum retention at ET *(hot tensile properties)* are other requirements. Seal operating experience in nuclear field indicates requirement of high temperature elastomers with excellent thermal stability (over operating span) as the foremost criteria to maximize life. Sulphur vulcanization of rubber is avoided to prevent corrosive effects of uncombined sulphur on SS., such as the IFTM inflatable seal mating surface (Fig. 3.10).

The Westinghouse Development Test Requirement Specification (WDTRS)evolving from commercial (such as automobile) and military usages after WW- II (1939- 1945) provided R&D outline for FFTF- CRBRP cover gas seal development which suggested exclusion of halogens and restrictions on chloride content in compound recipe. These were amongst reasons for non-consideration of CR for inflatable seals of FFTF- CRBRP RP [78- 79]. Also FKM rubber which was second to EPDM rubber in terms of popularity during 1970s and 1980s. Usage of EPDM was mainly due to compatibility with water coolant and steam in thermal reactors [80] which is elaborated in Chapter 7 in relation with PHWR and AHWR applications.

Unused activator (in vulcanized or cured rubber product) in the form of strong inorganic bases, such as $Ca(OH)_2$, could dehydrofluorinate (by abstracting HF) repeating reactive sequence of vinylidene fluoride (VDF, $CH_2 == CF_2$) and hexafluoropropylene (HFP, $CF_2 == CF -- CF_3$) in the polymeric chains of bisphenol cured FKM rubber (such as Viton® A- 401C) at sufficiently high elevated- seal- operating temperature (usual or design) or ET (i.e. equivalent to curing by moulding at 160- 190 ^{0}C or autoclave at 150- 170 ^{0}C). similar to the usual curing of green elastomer. This results in release low- molecular- weight, harmful volatile (HF) along with water vapour, CO_2 etc. where the reaction between metal oxide (acid acceptor or scavenger, MgO) and HF (producing $MgF_2$and H_2O) contributes. HF has potential to induce stress corrosion cracking in PWR, BWR, PHWR or AHWR components where elastomers are in contact with coolant. Such volatile release also occurs because of polymeric chain scission by γ dose which results in weight loss of elastomers

[82]. Unused double bonds in vulcanized rubber products[14] could be further crosslinked (cured or vulcanised) by certain elements in operating environment (such as additional amines) resulting in embrittlement and failure. This is a common problem for seals in oil and gas industry [83- 84]. Unused double bonds (unsaturation) and inorganic bases (metal oxides and hydroxides) after cure make the products/specimens vulnerable to polar fluids (such as water and steam) and base attacks from corrosion inhibiters used in oil and gas industry [85]. This explains the inherent limitations of popular bisphenol cured VDF- HFP fluoroelastomers (i.e. Viton A®- 401C) for thermal reactors [86].

Viton® A was commercialised during 1957 with initial impetus from military applications and aerospace industry. Potential difficulties from volatile release (water vapour, HF, CO_2 etc.) could be avoided by peroxide cure. Peroxide curing, however, has an inherent limitation of inadequate cure in air (reflected by problems such as mould sticking and fouling) which affected its commercial growth since a successful introduction during 1976 with Viton GLT, a terpolymer of VDF, tetrafluoroethylene (TFE, CF_2==CF_2) and perfluoromethylvinyl ether (CF_2 == CF–O–CF_3 in place of HFP) with cure site monomers for peroxide curing. These limitations restricted the use of peroxide cured fluoroelastomers in thermal reactors.

3.4.2.2 Failure parameters [9- 10, 43]

Exceeding limiting leakage and frictional drag (10^{-3} scc/ s/ m-seal- length and 10^{-2} scc/ s/m-seal- length during normal operation and fuel handling respectively; 1000 N/ m- seal- length during fuel handling) could be induced by the largest number of sources in RP inflatable seals (amongst cover gas barriers) including incorrect teflonising and gluing, machining marks crossing the sealing line, uncontrolled operational variations of inflation gas pressure- temperature, deviations from specification dimensions during manufacture or smearing of mist particles on seal rubbing face during rotation which may result in friction welding and exothermic reaction under extreme conditions *(i. e. high mist deposition)* in the presence of RCB air. Design defects (wrong sizing,

[14] Originally produced during HF abstraction from uncured or green VDF- HFP dipolymer by activator and not fully utilised for crosslinking by bisphenol curative during vulcanisation.

improper compound choice, inadequate coating specification) play complimentary role. Other areas are surface and volumetric defects from production (as also unnoticed nicks and cuts on sealing surface) along with debris or oily substance in seal groove during installation. Failure- prevention requires abstraction of criteria- mechanisms to representative material parameters- limits (Table 3.4).

Generally, tensile stress and strain in elastomeric seals exceeding T_s and E_b (at ET) is taken as the initiation point of cracking failure [75, 88]. Advantages of high temperature, low permeability elastomer could be exploited for longest life only if cracking or puncture of inflatable seal wall is avoided. This implies that degradation of T_s, E_b and modification of stress-strain field in seal by synergistic ageing (air + mist+ temperature + radiation) should not take away the FOS or the margin of safety between unaged stress-strain and T_s-E_b and increase stress-strain beyond material limits in combination with stress raisers and dynamic loading. The best way is to maximize retention of unaged properties over operation time under thermal ageing (i. e. high temperature elastomer with outstanding thermal stability) so that synergistic ageing from other sources does not bring down properties below threshold. Highest feasible hot tensile properties are therefore a prerequisite for inflatable seal elastomer in specific and cover gas elastomeric seals in general towards attainment of 1 synchronised replacement in 60 y.

Stress raiser in the form of volumetric defects (voids, inclusion, porosity etc.), introduced in seal elastomer during extrusion and curing, is one of the biggest hurdles in bringing comparability between LM freeze seal (life equivalent to reactor) and inflatable seal in terms of life. Volumetric defect cannot be removed altogether as also the stress raising effects at seal connector interface (Fig. 3.20). Thin seal wall, low dose rate and continuous exposure to aerosol and radiation in a primary barrier (Fig. 3.9) signify nearly uniform ageing across material volume where surface or volumetric defects or stress concentration from other sources have comparable chances of initiating cracks. Thin seal wall provides small path length for fracture. All these aspects combine to increase probability of puncture in inflatable seal substantially. It is therefore imperative that some other material failure parameter is also considered.

Oxygen in RCB air attacks double bonds in elastomeric macromolecular chains and ozone in reactor environment accelerates chain scission to encourage propagation of crack from defects even under static loading (seal inflated, Fig. 1.5) and stress level below fatigue threshold. This is complimented by the general propensity of elastomers to low- cycle- fatigue, applicable to RP and IFTM inflatable seals (Figs. 3.9- 3.10) during dynamic operation at low speeds (75 mm/ s and 14 mm/ s) with varying annular gap (5 ± 2 mm). These aspects underline the fundamental necessity to maximise FOS (to be consumed by synergistic ageing) on the maximised hot tensile properties, supported by accurate quantification by FEA, for longest seal life.

Compatibility with oxygen and ozone and maximization of tear strength are other prime requirements for inflatable seal material. From the same count, best compatibility with sodium aerosol is to be ensured which could cause swelling in elastomer to reduce strength, hardness as well as wear resistance. Continued exposure of mist could produce bathing (immersion) effects in primary inflatable seal to demand higher level of compatibility, i. e. compatibility with liquid sodium instead of mist [89]. Different elastomers show different rates of mist accumulation and deposition, and hence the chances of friction welding which determines the generic elastomer choice. Meeting all the requirements in a single elastomeric formulation at an ideal level is very demanding. Usage of reinforcement in primary inflatable seals of many FBRs could therefore be seen as a replacement of several shortcomings in seal elastomer by a simple and single solution (with the aid of manufacturing technology) while maximizing other properties and FOS in the absence of predictive techniques such as FEA.

Irrelevance of CS based failure in inflatable seals (Table 3.3) of INDIAN- SFR RP (and IFTM) heuristically suggests that material unification by tailoring of inflatable seal compound (S-2) should dwell upon the backup seal compound (S- 4) at some point of time or other.

3.4.2.3 Epitomising reactor and seal designs, R & D [9- 10, 36, 43]

Choice, location and arrangement of FBR RP inflatable seals embody the best findings from evolution of reactor design, sealing scheme, seal design and R & D to make them suitable as originator for material tailoring towards uniform life maximisation of other TS seals. Primary dynamic barriers in FBR RPs (Fig. 3.21) show 3 trends viz. a) Sn- Bi based LM freeze seal (France, Japan, USSR), b) LM dip seal based on mercury (UK) and, c) Inflatable seal (Germany, USA).

Continuation of LM freeze seal as primary RP barrier in pre- 1980s FBR designs by France, Japan and USSR (in spite of maximum bulk, cost, complexity and fabrication challenges) could be attributed to the second lowest leakage and friction (RP rotational torque) after mercury dip seal, life equivalent to reactor and the ability to resist CDA to a large extent.

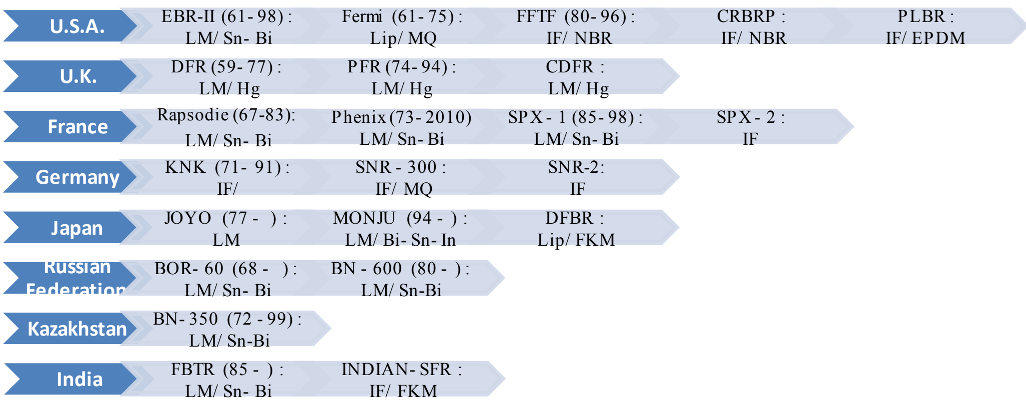


Fig. 3.21. Evolution of primary dynamic barrier in FBR rotatable plugs.

Numbers in bracket indicate first criticality and final shutdown; EBR-II first criticality corresponds to dry criticality.

CSM: Chlorosulfonated polyethylene (Hypalon); IF: Inflatable.

The pre- 1980s design and usage of inflatable seal (not resistant to CDA) as primary barrier in RPs of Germany (KNK I and II, SNR- 300) and USA (FFTF, CRBRP and ≥1000 MWe LMFBR CDs) FBRs could be attributed to ready availability of commercial compounds, seal and coating for nuclear- specific evaluation, matured production knowhow (machining and assembly with close tolerance) and existence of a rich tradition of crafts (Germany). Progress in inflatable seal technology with original development for cockpit integrity in high-tech aircrafts such as Saab Griffen and Panavia Tornado [90] and subsequent design evolution from 1940s to 1970s provided the preparedness. Inherent advantages of inflatable seals assumed complimentary role i.e. simplicity, compactness, economy, ability to seal large diameter- gap- tolerance (particularly suitable for > 0.3 m diameter), ability to engage at the required place and time (manoeuvrability) with zero assembly

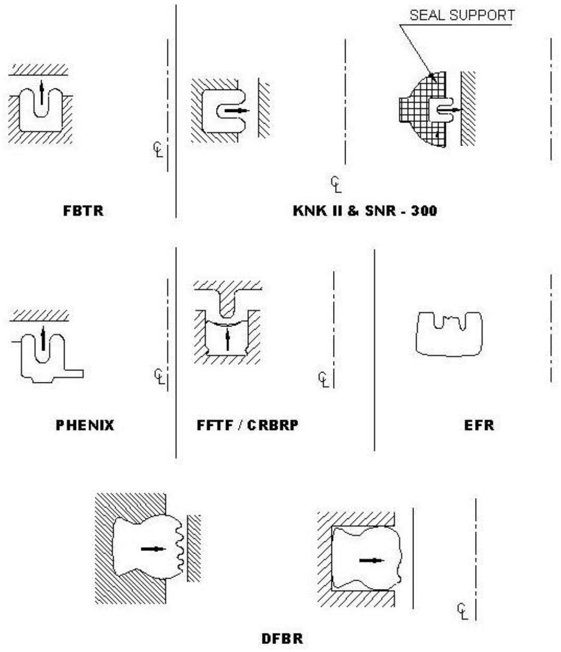


Fig. 3.22. Low pressure inflatable seal geometries in various FBRs.

load and friction, reasonably good leaktightness and third lowest rotational drag (torque) after LM dip and freeze seals for RP.

Independence of CS based failure mechanism and ability to seal with low inflation pressure (< 0.3 MPa), seal-stress as well as its variation (from gap tolerance, during rotation) with trademarked designs (such as *Membramatic*, from Le Joint Francais, France) used in French and German FBRs (similar to:

Table 3.6. Design feature of inflatable seals in rotatable plugs of various FBRs

Design features	**Reactors**										
	Rapsodie	KNK	FBTR	Phenix	FFTF	CRBRP	PLBR	SNR-300	SPX- 1	MONJU	INDIAN-SFR
[a] **Seals/ plug**	1(---)	2(NR)	1(NR)	1(NR)	2(RI)	2(RI)	2+2 (RI)	2(NR)	1(RI)	1(NR)	2(NR)
Function	S	P	S	S	P	P	P	P	S	S	P
Engaged	NO + FH	NO + FH	NO + FH	NO + FH	FH	NO + FH	NO + FH	NO + FH	NO +FH	NO + FH	NO + FH
[b] **Mounting**	[c] R/A (DP)	R(SP)	A1(SP)	A1(SP)	A1(DP1)	A1(DP1)	R+A1 (DP1)	R(SP)	A1(SP)	A1(SP)	R(DP1)
Fixing	---	G	G	C	G	G	G	G	G	G	G
[d] **Lubrication**	---	TBC	TBC	TBC	LL	LL	LL	TBC	Coating	---	TLC
Material	---	CSM	CR	NBR	NBR	NBR	EPDM	MQ	MQ	FKM	FKM
Inflating	O	I	PPA	PPA	PPA	PPA	I	I	PPA	PPA	O

[a] Seal material reinforcement: NR – Non reinforced, RI- Reinforced; [b] R - Radial, A1 - Axial, DP1 - (Seal fitted to) dynamic part (shell/bearing-race) of RP, SP - Static part (shell/bearing- race); [c] Radial and axial mounting changed several times; [d] Lubrication of seal rubbing face: LL - Liquid lubricant, TBC - Teflon based coating, TLC – Teflon like coating.

C: (Seal) clamped; FH: (During) fuel handling (condition); G: (Seal) glued (to groove); I : Inward (towards plug axis); NO : Normal operation; O : Outward (away from plug axis); P : Primary seal; S: Secondary seal; PPA: Parallel to plug axis.

Figs. 1.5a- b, 3.16 and 3.20) created the potential of competing with LM freeze or dip seals in terms of seal life. Figure 3.22 shows similar low pressure inflatable seal geometries (inflation pressure: ~ 50- 100 kPa) with variations of folds and beads on sealing face developed for and used in various

FBRs (Table 2.1). Table 3.6 shows design features of inflatable seals developed or used in various FBRs which, along with Fig. 3.21, indicates preference for the same primary barrier both during normal operation and fuel handling conditions in order to blend and optimise aspects of simplicity and reliability.

RP sealing schemes of various FBRs (Table 3.7) show that only three experimental (DFR, PFR and PEC) and one demonstration (FFTF) FBRs proposed or used different barriers. The monopoly of inflatable seal as primary barrier (excepting USSR which continued with LM freeze seal) in post-1980s, MOX fuelled, modern FBRs could be attributed to departure from zero leakage philosophy of cover gas, redefinition of CDA as BDBE and enhanced economisation of FBRs.

The near- total departure from liquid lubricants to Teflon coating of inflatable seals (Table 3.6) was induced by spillage- avoidance and aided by advent of Teflon coating technology [91] as well as its inherent advantages i.e. very low coefficient of friction, absence of sticking and stick-slip, continuous operation at higher temperature, independence of friction with respect to temperature and velocity of rubbing etc. Possible reactivity surge because of gas- bubbles (H_2 and CH_4), releasing from oil- spill into sodium coolant pool and passing through the core (as suggested by pump seal and bearing oil spillage in PFR during 1974 and 1991), was a major reason for spillage avoidance which has assumed paramount importance during post- Fukushima period for total evading of positive reactivity coefficient of voids [5, 11].

Enhanced economisation drive of FBRs during post- 1980s necessitated minimisation of high cost steels in RA by minimising MV diameter [92] where minimisation of RP diameter by proper sealing and fuel handling scheme, reduction of number of loops (simplicity, improved constructability, lower failure probability, higher availability) and minimisation of primary sodium pump (PSP) and intermediate heat exchanger (IHX) diameters were complimentary [93].

One of the common measures in pre- 1980s FBR RP sealing scheme (Tables 2.1, 3.6; Fig. 3.21) was to keep the inflatable seal furthest from TS top plate at usual operating temperature

Table 3.7. Rotatable plug sealing schemes for various FBRs

FBRs	RP sealing combinations				
	Normal operation	Fuel handling	[b] Protection seal	Maintenance seal	Mist deposition control
Experimental					
DFR	EOR	Na- Hg + Hg	----	----	----
EBR-II	Sn- Bi	Sn- Bi	----	----	None
Fermi	NaK + Lip	NaK + Lip	----	NaK	NaK
Rapsodie	Sn- Bi + Lip	Sn- Bi + Lip	Lip	----	P1 + CV
BOR-60	Sn- Bi	Sn- Bi	----	----	----
KNK	IFS + ESS	IFS + ESS	ESS	----	CV
JOYO	LMS freeze	LMS freeze	----	----	----
FFTF	SS1	IFS	----	NaK	P1+ CV
FBTR	Sn- Bi + IF	Sn- Bi + IF	IFS	----	P1 + CV
PEC	DG	Hg	----	----	----
Demonstration					
BN-350	Sn- Bi + RR	Sn- Bi	RR	----	----
Phenix	Sn- Bi + IFS	Sn- Bi + IFS	----	----	CV
PFR	EOR	Hg	----	----	P1 + NaK
BN-600	Sn- Bi + RR	Sn- Bi	RR	----	----
MONJU	Sn- Bi- In + IFS	Sn- Bi- In + IFS	IFS	----	P1 + CV
INDIAN- SFR	IFS + BES	IFS	BES	----	Na
CRBRP	IFS + Lip	IFS + Lip	Lip	----	Na
SNR-300	IFS + ESS	IFS + ESS	ESS	Yes	P1 + CV
Commercial					
SPX-1	LM + IFS	LM + IFS	IFS	----	----
SPX-2	IFS	IFS	----	----	----
SNR 2	IFS	IFS	----	----	----
DFBR	Lip + IFS	Lip + IFS	----	----	----
CDFR:	SEES	Hg + SEES	-----	-----	-----
EFR	IFS	IFS	----	Yes	Yes

EOR: Elastomeric O ring; P1: Purge; CV: Convection barrier; DG: Double gasket; IFS: Inflatable seal; ESS: Elastomeric static seal; LMS: Liquid metal seal; SS1: Static seal; RR: Rubber ring; BES: Backup elastomeric seal; SEES: Spring energized elastomeric seal. [a] Based on available data. Liquid metal freeze and dip seals are indicated by metals and alloys; [b] RCB air side protector seal mentioned

of 40- 60 ^{0}C (SNR- 300: 80 ^{0}C) in order to minimise the synergistic ageing effect and permeation leakage while maximising access, maintainability and availability. The post- 1980s, > 1000 MWe, modern FBR designs (German SNR- 2, French SPX- 2, British CDFR and EFR, Table 2.1) introduced warm roof (TS top plate temperature: 110- 120 ^{0}C) for economisation by minimising

Fig. 3.23. Design and development areas of inflatable seal.

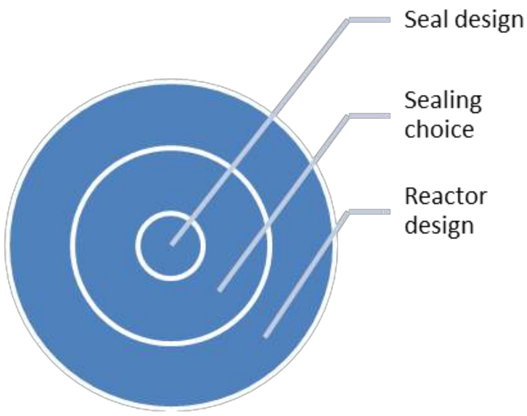


Fig. 3.24. Progressive influences for seal life maximization.

insulation. Placing SRP within LRP (bearings at same elevation, Fig. 2.3) was parallel necessity for minimisation of RP- MV size (diameter and height), cost and stress along with improvement of accuracy and ease of alignment of safety- critical components (IVTM, IFTM). Locating the primary inflatable seals closest to TS top plate for minimisation of plug- height, stress and mist- deposition maximised cover- gas- loads on primary inflatable seals operating at about 100 ^{0}C. This was one of the main reasons for continued search of low permeability, high temperature elastomers with longest life even at the end (1993) of EFR RP inflatable seal development.

Post- 1980sdesign philosophy indicates maximisation of life as the main challenge. Upscaling effects (test seal: from < 0. 5m diameter; test reactor: from ~ 2 m diameter; commercial reactor: up to 10 m diameter), transition from secondary to primary barrier (Tables 2.1. 3.6- 3.7; Fig. 3.21) and then from farthest to closest of TS top plate kept extensive R & D continuing in various areas (Fig. 3.23).

Progressive roles of reactor design, sealing scheme, seal design and R & D (Fig. 3.24) in maximising seal life could also be found in minimising non- mechanical (cover gas, Fig. 3.25) load (temperature + radiation + mist) by dilution at source with argon purification and capturing of long- lived fission product isotopes by cold trap before reducing further by ledge/ ring/ convection- barrier (CV) at TS annulus and then by argon- helium purge (towards cover gas). Alternatively, sodium dip seal (instead of CV and purge) at the bottom of TS annulus replaces the large coolant pool in MV with a tiny LM pool in seal and a much smaller surface area (spanning annular gap) to minimise the

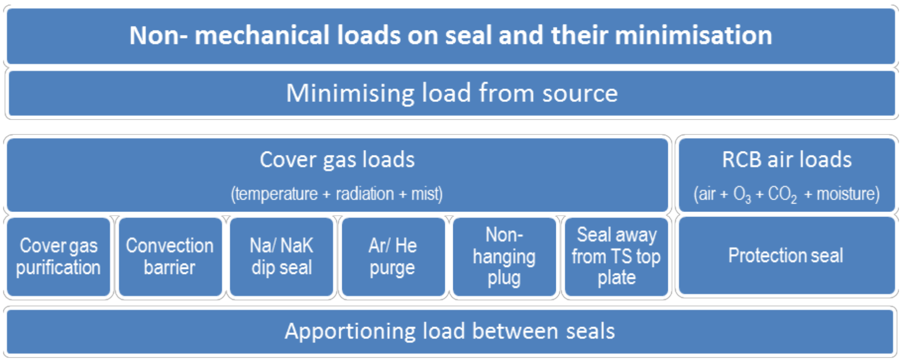


Fig.3.25. Sequential approach (top- down) for minimisation of non- mechanical loads on rotatable plug seals.

heat transfer (radiation + convection + diffusion) and the convective mass (mist) transfer through annulus to the above- TS area of seals in a hanging plug (Figs. 3.9, 3.19). Cover gas loads are subsequently reduced by distribution amongst RP sealing scheme comprising of primary and secondary barriers etc., also designed for reliability. RCB air load (oxygen, ozone, CO_2, moisture etc.) are similarly taken by the protector seal (Table 3.7), which could also be the secondary barrier (backup seal in INDIAN- SFR RPs, Figs.3.9) for design simplicity.

Subsequent distribution- minimisation by seal design could be dividing the static (inflation) and dynamic (friction) loads between membrane and beads (Fig. 1.5) or lip (membrane: reinforced, lip: non- reinforced, Fig. 3.26), for example. The EFR inflatable seal design (Fig. 3.26) was an epitome of collective wisdom i.e. non- reinforced bonded chord (on non- reinforced membrane) of SNR-300 and reinforcement of SPX-1 inflatable seals amalgamating with the German concept (of inflatable seal as primary barrier without liquid lubricant, Tables 3.6- 3.7) within the framework of sodium cooled, pool type, MOX fuelled FBRs based on French philosophy under the aegis of WE collaboration (Fig. 3.21, Tables 2.1), founded on sharing of developmental cost. SPX 1 was the model for the common European design. Net effects of these on RP inflatable seals was convergence to MQ and FKM rubber based designs in Europe (EFR) and Japan (DFBR).

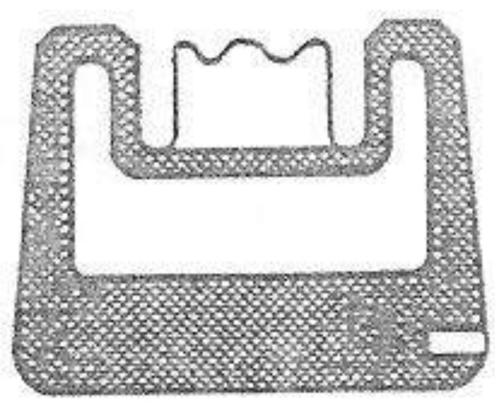


Fig. 3.26. EFR inflatable seal design

Antifriction coating is an example of minimising mechanical load by seal design (Fig. 3.11). Table 3.7 shows the evolving sealing scheme in FBR RPs towards attaining the objectives discussed.

Use of maintenance seals in Fermi (NaK dip seal), FFTF (NaK), SNR-300 (auxiliary seal inserted), CRBRP (argon gas purge above sodium dip seal) and Prototype Large Breeder Reactor or PLBR (inflatable) RP designs and emphasis on fast maintenance during German and Japanese developments indicate importance attached to plant availability during pre- 1980 period which relates to plant operating cost i.e. ~ 15% of reactor cost (Section1.2.2). Developing provisions for ready dripping of condensing sodium during EFR conceptualisation (1988- 1993) implied minimizing sodium bathing of seals and making the elastomer choice independent of elastomer-specific, differing aerosol deposition rates (FFTF-CRBRP results).

Minimising non- mechanical, synergistic ageing load and subsequent minimisation as well as apportionment of mechanical loads by conceptual design indicate longer life for the same seal elastomer. Subsequent R & D is about utilisation of the full potential of the material by using predictive techniques (FEA, Arrhenius and WLF equations) and quality control. Improving material further is one of the final steps for life maximisation (when mechanical design potentials, i.e. reinforcement, bead, membrane etc., have been utilised) which is central to this dissertation based on non- reinforced and unbeaded inflatable seal design of INDIAN- SFR.

3.4.2.4 Imbibing past experience: Finalising and shortlisting of design option s [9-10, 36, 43]

<u>Arriving at a validatory testing scheme for inflatable seal in scaled down test rig:</u>

FBR RP inflatable seal development (late 1960s to early 1990s) employed trial and error based tests in scaled down test rigs under simulated service conditions (without radiation, sometimes in the presence of mist) as a tool for design rather than arriving at a seal size closest to the actual by FEA to minimise the tests and confine them to validation only. Evaluation of 1 m diameter test inflatable seals for KNK- I RPs (involving various profiles, elastomeric formulations and coatings) began during later part of 1960s in the 5 MW KNK test loop (commissioning: 1965) under temperature, pressure and rotational simulations in the presence of sodium aerosol. The USA 1000 MWe LMFBR cover gas seal development programme during early 1970s assigned compatibility, chemical degradation and material evaluation tests (mist, temperature, radiation, permeation, CS etc.) to specimens at the beginning itself (FFTF development) in order to minimise the number of seals to be manufactured and tested. Issues pertaining to upscaling (manufacturability, retention seal in groove, more detailed specification and quality control) from 1 m diameter test seals to reactor scale (KNK-I; RSB test facility, equivalent to SNR- 300 with ~ 5 m seal diameter, Table 2.1) and the necessity for maximising seal life introduced similar specimen based material studies (1977) during the German development which evolved to encompass the approach of synergistic ageing and common materials for all seals by the end (1993) of the unfinished endeavours (1993) for EFR based on the WE collaboration (1984).

Failure of Fermi lip seal (*Klosure*) in Rapsodie delayed introduction of inflatable seal as secondary barrier in Rapsodie RPs and LM freeze seal was continued as the primary (Tables 2.1, 3.7; Fig. 3.21) in Phenix and SPX-1 [94]. The French and Japanese inflatable seal development, limited in specimen studies because of secondary functions, involved testing in reactor (Rapsodie and Phenix for inflatable seal mounting), 0.5 m diameter, 1 m diameter and 1.5 m diameter scale with full scale testing carried out for MONJU (along with LM freeze seal) and Phenix (Table 3.6). The unfinished research and testing program at later part of 1970s (PEC project cancelled; Table 2.1) for inflatable as part of Italian- French collaboration indicates maturity and standardization of specimen- rig based R& D scheme in continuation with the USA and German developments [95-98].

German full scale testing could be seen in the RSB test facility (1970 onwards) with ~ 5 m diameter inflatable seal simulating the SNR- 300 diameter scale where seal leakage was brought down from 200 scc/ s to 10^{-3} scc/ s prior to 1975 by changing the higher inflation pressure (normal-operation/fuel-handling: ~ 300 kPa/ ~ 100 kPa)KNK- I inflatable seal design in dovetail groove to the rolling- in, Membramatic type in rectangular groove (Fig. 3.22) at an inflation pressure of 70 kPa [99- 101]. A bilayer Membramatic inflatable seal design made of MQ shell and FKM (Viton®) inner layer (hose) was envisaged at around the same time to minimise permeation (by ~ 100 times) which

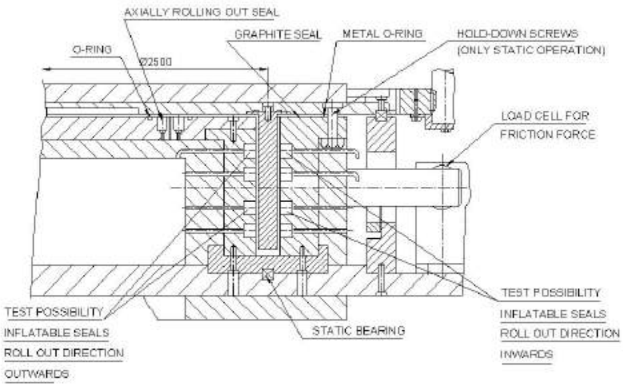


Fig. 3.27. Siemens 2.5 m rig with testing provisions for axial and radial inflatable seals.

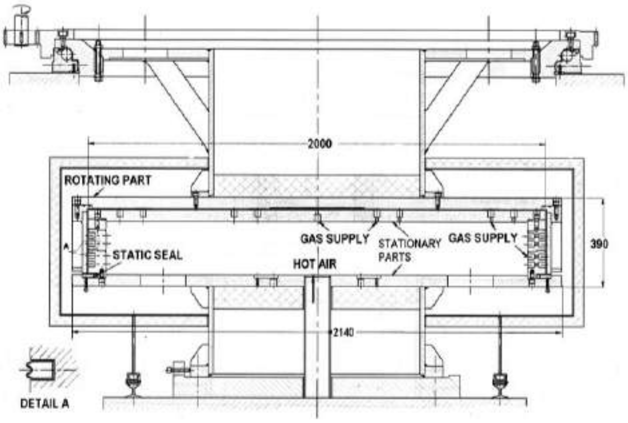


Fig. 3.28. Validatory test facility for INDIAN-SFR and FBTR inflatable seals.

initiated specimen- studies on both the elastomers from 1977.The specimen based systematic studies on MQ and FKM rubbers (1977 onwards) resulted in finalisation of the MQ- FKM (Viton®) bilayer, non- reinforced inflatable seal design by 1979.The American endeavour covered test seal diameter range from < 0.5 m to ~2 m (for CRBRP; Fig. 3.21, Table 3.6-3.7).

By the time EFR inflatable seal testing were being carried out in the 2.5 m diameter Siemens test rig (1992- 93, Fig. 3.27), a two- stage pattern for validatory testing (without sodium and radiation) evolved i.e. i) Testing in 0.5 - 1 m diameter range for arriving at a seal profile and, ii) Testing in 1.5- 2.5 m diameter range (diametric scaling- 1: 2.5 – 1:3) for upscaling effects (including reactor gap tolerance), parametric studies or seal characterisation (leakage and friction with pressure, temperature and rotation variations) and life assessment (endurance tests) by continuous rubbing (or rotation) of seal equivalent to or more than seal design life. Pressure, temperature, rotational speed, seal cross- sections were simulated 1:1. These were reflected in the validatory scheme of inflatable seals (Fig. 3.28) for INDIAN- SFR and FBTR RPs in 0.5m- 0.5 m and 1.5 m- 2 m diameter scales.

<u>Choice of non- reinforced elastomer and extrusion for inflatable seal</u>

Minimisation of design options (INDIAN-SFR and FBTR inflatable seals) by literature survey resulted in conceptualisation of low pressure, axially- radially mounted (Fig. 3.29), coated (on seal rubbing and mating metallic faces) inflatable seal (bonded to rectangular groove base) with one (mechanical) inflation connector and end joint per seal (Table 3.6). Simplicity of design, manufacture and capability of Indian industry were deciding factors for going ahead with the choice of non- reinforced elastomer and production by extrusion instead of moulding. Reinforced elastomers are composite materials involving more demanding design and manufacture (co-extrusion) where bonding interface (elastomer- reinforcement) adds to design- weak- link (leakage path)- indicated by operating experience of airlock door seals in thermal reactors and results from FFTF- CRBRP inflatable seal development [102- 104]. Seal geometry, material and coating however had more than one options, to be finalised by R & D.

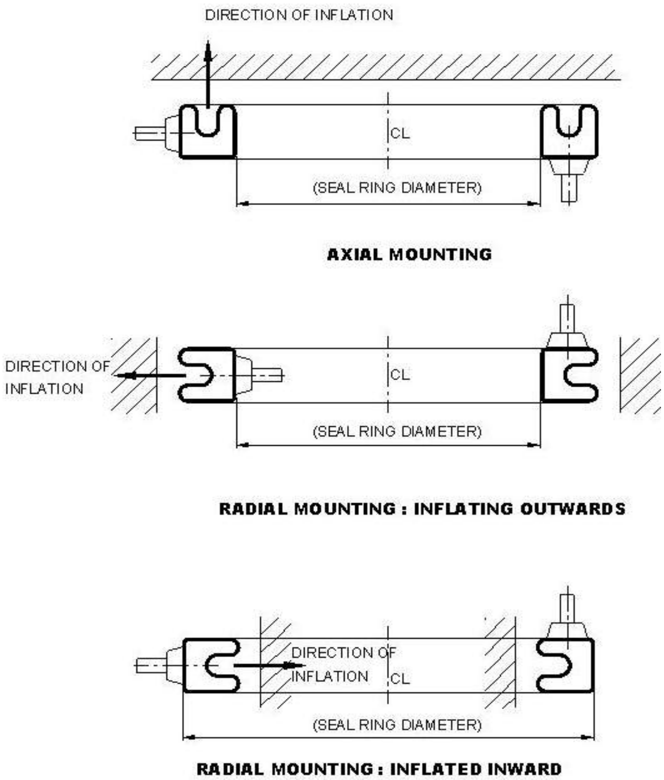


Fig. 3.29. Axial and radial mounting of low pressure inflatable seal, deflated and disengaged.

<u>Minimising design options</u>: *Coating*

Coating deposition by brushing, spraying and plasma processes were kept as options with preference for plasma processes to ensure minimum elastomer substrate temperature during deposition and dense, homogeneous, lubricious, thin coating of substantial adhesion strength for long life (Table 3.5).

<u>Minimising design options</u>: *Generic elastomer*

Seal operating experience in nuclear field indicates requirement of high temperature elastomers with excellent thermal stability (over operating span) as the foremost criterion to maximize life (Fig. 3.21; Tables 2.1, 3.6). This brought down the choice (Table 3.8) to polyacrylate rubber (ACM), CR, EPDM, FKM, FMQ and MQ rubbers taking ~150 ^{0}C (initial high temperature threshold used) and 10 kGy as thresholds. CR was not considered further because of failure in FBTR

Table 3.8. Ranges of operating temperature, radiation resistance and physico- mechanical properties of seal elastomers

[a] Property	[b] Generic Elastomers											
	ACM (1940s)	AU, EU	CO, ECO (1965)	CR (1932)	CSM	EPDM (1961)	FKM (1957)	FMQ	SBR (1936)	IIR (1937)	MQ (1942)	NBR (1935)
WTR (^{0} C)	- 18/ 177	- 54/ 93	-54/ 135	- 54/ 149	- 54/ 121	- 54/ 149	- 29/ 204	- 73/ 177	- 51/ 93	- 54/ 107	- 114/ 232	- 54/ 121
[c]RR-1 (kGy)	$> 10^2$	$> 10^2$	---	$< 10^2$	$< 10^2$	$> 10^3$	$< 10^2$	---	$> 10^2$	$> 10^2$	$\sim 10^2$	10^2
[c]RR-2 (kGy)	$< 10^2$	---	---	~ 10	---	---	---	---	$< 10^2$	~ 10	10	10^2
TSR (MPa)	16.1/ ---	28/ 55	13.2/ ---	3.4/ 21	---	14/ 21	10.3/ 21	---	3.4/ 21	14 +/ ---	4.1/ 10.3	6.9/ 24
ELR (%)	272/ ---	250/ 800	630/ ---	650/ 850	---	500/ ---	100/ 450	---	450/ ---	300/ 800	90/ 900	400/ 600
HDR (^{0}Shore A)	40/ 90	55/ 100	30/ 40	20/ 95	---	30/ 90	50/ 95	---	40/ 100	30/ 100	25/ 90	20/ 90

[a] Properties measured at RT in air; [b] ASTM designation (D 1418); [c] Radiation exposure at RT in air. ~: Approximately equal; $> 10^x$: Less than 5 x 10^x; $< 10^x$: Greater than 5 x 10^{x-1}.

* AU, EU: Polyurethane elastomers; CO, ECO: Epichlorohydrin elastomers; IIR: Butyl rubber; * Common names (from trade names) indicated in parentheses.

WTR: Working temperature range; RR- 1: Radiation range (for ≥ 80 % retention of T_s, E_b and hardness); RR- 2: Radiation range (for ≤ 80 % CS); TSR: Typical T_s range; ELR: Typical E_b range; HDR: Typical hardness range.

and non- consideration as well as usage in any other FBR either (Fig. 3.21; Tables 2.1, 3.6). ACM was also not considered because of similar reasons. EPDM with lower thermal resistance was retained for further shortlisting because of lower operating demands of FBTR secondary inflatable seal (10 kPa/ 80 ^{0}C/ 2 kGy (10 y)/ 10 km (10 y); Table 3.3).

Fluoroelastomer is perhaps the best choice if limitations of anticipated halogenated volatile release and lower hot tensile properties (because of loss of ionic interactions at elevated temperature) are discounted. Inherently lower resilience in fluoroelastomers is not a problem because of slow rotary oscillation (INDIAN- SFR: 75 mm/s; FBTR: 14 mm/s, Table 3.3) and external actuation by inflating gas. These were demonstrated by experimentations on FKM inflatable seals for MONJU and DBFR RPs (1970s to early 1990s) without any reported effect from harmful volatile release. Relook at the real necessity of non-halogenated recipe during release of seal design guide (1975), evolving from FFTF cover gas seal development provided an earlier indication on lesser importance attached to this aspect, in particular when seals are not in contact with the coolant. Lower hot tensile properties are addressable by accurate quantification of deformation and stress-strain using FEA.

Overcoming larger limitations of (lowest) strength and (highest) permeability of MQ (compared to FKM) in many FBRs (Fig. 3.21; Tables 2.1, 3.6) does also provide precedence for going ahead with fluoroelastomers, as benefits outweigh limitations. FKM rubber was therefore retained for final shortlisting. Primary reason for retaining silicone rubber was its development and wide usage in many FBRs (Fig. 3.21; Tables 2.1, 3.6). FMQ was taken out of candidature because of potential difficulties in bonding with metallic parts by glue. The final list therefore contained EPDM, FKM and MQ as candidates for compounding, moulding, extrusion and curing trials as well as for other detailed studies to identify one elastomeric formulation.

Fig. 3.30. Inflatable seal geometric options.

Minimising design options: *Seal geometry*

EFR inflatable seal design (Fig. 3.26) was partially emulated during shortlisting of four non-reinforced, low pressure geometric options (Fig. 3.30) to be finalised by FEA and validatory tests. Unbeaded, rolling-in geometry with fold perpendicular to seal base (Figs. 1.5, 3.30) was the first choice because of simplicity and adoption for inflatable seals of French and German FBR RPs (Fig. 3.22). Single lip on vertical fold (similar to EFR) was the second option. Reduction of single lip height brings in multiple serrations on the vertical fold which becomes the rubbing face on inflation. This was the third option. The fourth option was a variation of proven Japanese design (Fig. 3.22) with horizontal fold and multiple beads on the rubbing face.

3.4.2.5 Feasibility of achieving one replacement during reactor life [9- 10]

Increasing envisaged life of RP primary inflatable seals from FFTF (5 y; design initiation: 1963) to the 1000 MWe LMFBRs (40 y; end 1970s; with sodium dip seal) and similar indications from the operating experience of Phenix (4 y), FFTF (7 y) and KNK- II (at least 10 y, 15 y by unconfirmed data) suggested improving comparability with LM freeze seal in terms of one of the main limitations of elastomeric inflatable seals i. e. life (Fig. 3.21; Tables 2.1, 3.6-3.7). Increasing fuel burn-up (normal operating span) from 50000 MWd/t for >1000 MWe FBRs during the days of

Phenix construction (first criticality: 1973; Table 2.1) through 1,70, 000 MWd/t in EFR to 1,50,000 MWd/t for the latest Japanese loop concept (normal operation or static sealing span: 26 months) imply further improvement of seal life because of reduced plug rotation and wear which created the confidence that a maximum of 1 replacement per reactor life of 60 y is feasible with high temperature elastomers (maximum temperature rating ≥ 175 ^{0}C) operating at around 100 ^{0}C in warm roof [15]. It is noted that all the lifespans of inflatable seals indicated earlier involved low temperature elastomers (NBR and Chlorosulfonated polyethylene elastomer or CSM with highest continuous operating temperature of 121 ^{0}C; Tables 3.6, 3.8) operating at a temperature range of 40- 60 ^{0}C with seals located furthest from TS top plate.

Continuing search for better diffusion proof silicone rubber at the end (1993) of the unfinished EFR endeavour had similarity with renewed R & D on identifying new (high temperature, low permeability) elastomer during the closing phase (end 1970s) of USA LMFBR cover gas seal development. For EFR, in view of the increasing burnup and normal operating span, the other objective could have been to achieve a joint equivalent to the welded one (metal fuelled FBRs) in terms of leaktightness while retaining the flexibility of rotation as well as opening/closing of joint (seal engagement/disengagement) at will for maintenance and replacement.

3.5 Identification of Common Generic Elastomer Across Categories: *First assessment* [9-10, 43]

The initial USA and European pursuit for commercialisation of FBRs (≥ 1000 MWe scale) by 1980s was supported by joint purchase

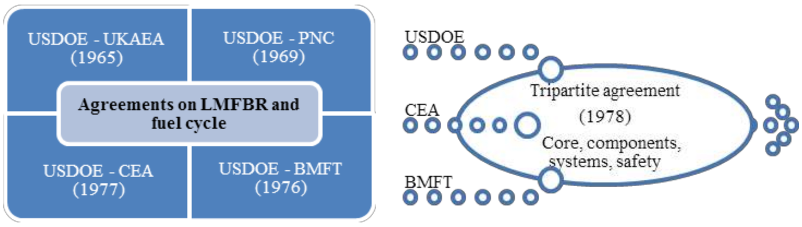


Fig. 3.31. Some of the agreements between research centers of USA., Europe and Japan during 1960s and 1970s.

BMFT : Federal Ministry for Research and Technology of the Federal Republic of Germany; CEA: Commissariat a l Energy Atomique, France; PNC: Power Reactor and Nuclear Fuel Development Corporation, Japan; UKAEA: United Kingdom Atomic Energy Authority; USDOE: United States Department of Energy.

[15] Normal operating span increased from a reference of 3 months in SNR-300 (6–12 months more likely as per early-1970s assessment) to 24 months in EFR.

initiative (France, Germany and Italy) of large breeders (1970) which reduced design differences. The German-French agreement (1976) departed from earlier collaborations (Fig. 3.31) based on exchange and sharing [105] by introducing joint ownership which shaped into broader WE collaboration (1984). Departure to the outlook of tolerable cover gas leakage during late- 1970s (after FFTF design based on zero cover gas leakage, Table 2.1) was an outcome of this backdrop and increasing understanding on mechanisms as well as outcome of cover gas leakage. The later was influenced by licensing issues in SNR- 300 and renewed LMFBR conceptualization drive in USA because of drastic reduction in funding from changed policy. The USA conceptual design (CD) drive on LMFBRs eventually gave way to another departure by mid- 1980s i.e. sodium cooled, pool type, metallic fuelled, modular FBR designs represented by Power Reactor Innovative Small Module (PRISM). WDTRS 1.25 conservatively specified allowable xenon and krypton permeation leakage of 80 x 10^{-8} scc/s/m for elastomeric cover gas seals of FFTF [78- 79]. Applying this limit to INDIAN-SFR TS sealing length of ~700m yields total allowable leakage of ~ 5.6 x 10^{-4} cc as against the design limit of 10 cc/ s (1cc/s actually applied conservatively) which shows the quantitative order of departure from the zero leakage approach.

The changing outlook was also accompanied by a key finding (end- 1970s) that radiation dose at TS is insignificant in influencing elastomer seal performance (in isolation, because of small γ dose rate[16]) which indicated that thermal ageing has greater influence on the overall synergistic (temperature + radiation + mist + air) ageing degradation [47]. Mid- 1990s reporting from Canadian PHWRs (since 1960s) substantiated this by observing that thermal ageing induces failures in elastomeric seals even in an environment (PHWRs) where operating radiation dose (10 kGy) and temperature (200 ^{0}C) are much higher [51]. These findings were established by end- 1990s when survey of organic components in nuclear field identified thermal ageing as the root cause of failure

[16] Assessment based on above- TS dose rate of 0.25 mr/ h; PFR design specified shielding to limit radiation level < 0.75 mr/ h; Corresponding limit for INDIAN- SFR- FBTR: ≤ 1 mr/ h (4 nos. or 0.01% failed fuel pin assumed in INDIAN- SFR core); USA 1000 MWe LMFBR specified ≤ 2 mr/ h during end- 1970s.

Table 3.9. Generic elastomers and normal operating temperature combination of barriers in FBR rotatable plugs

FBR	DFR (P- S1)	Fermi (P- S1+ D)	KNK (P- S1+ D)	Phenix (B- S1+ D)	PFR (P- S1)	FFTF (P- D)	CRBRP (P- S1+ D)	SNR 300 (P- S1+ D)	SPX- 1 (S- S1+ D)	MONJU (S- S1+ D)
Seal	O- ring	Lip	Inflatable	Inflatable	O- ring	Inflatable	Inflatable	Inflatable	Inflatable	Inflatable
Material	MQ	MQ	CSM	NBR	MQ	NBR	NBR	MQ	MQ	FKM
Temperature (^{0}C)	< 60	57	< 60	40- 50	< 60	51	51	80	50- 60 [a]	< 60
FBR	FBTR (S- S1+ D)	SPX- 2 (P- S1+ D)	SNR- 2 (P- S1+ D)	EFR (P- S1+ D)	DFBR (S- S1+ D)	INDIAN- SFR (P- S1+ D)				
Seal	Inflatable	Inflatable	Inflatable	Inflatable	Inflatable	Inflatable				
Material	CR	MQ	MQ	MQ	FKM	FKM				
Temperature (^{0}C)	< 60	115 [b]	120 [b]	~ 100	60	100				

P- Primary seal; S- Secondary seal; S1- Static (normal operation); D- Dynamic (fuel handling)

[a] Reactor deck temperature taken

[b] TS top plate temperature taken

with failure rates maximum in cables[17] followed by those in elastomer seals [57]. Subsequent review (2001) of polymeric ageing in nuclear power industry showed beyond doubt that thermal ageing is the decisive factor [58] where operating temperature propels (accelerating or decelerating) other ageing (oxidation, radiation and aerosol) mechanisms and rates. Identifying thermal ageing as pivotal highlights the necessity of high temperature (≥ 175 ^{0}C) generic elastomer for MOX fuelled modern FBRs (represented by INDIAN- SFR) in a warm roof maintained at ~ 110^{0}C[18]. Table 3.9 gives a very clear indication about the intuitive preference for a generic elastomer (MQ) with highest temperature (232 ^{0}C) rating (Table 3.8) to maximise the life of low temperature (40- 60 ^{0}C) RP elastomeric sealing starting from the very beginning (DFR, 1959, Fig. 3.21, Table 2.1).

Non- usage of high temperature elastomers (NBR, CSM, CR; 121- 149 ^{0}C, Tables 3.8- 3.9) for RP inflatable seals, in spite of the necessity to compete with LM freeze seals in terms of life[19]*, could be attributed to greater influences from other rubber characteristics, evolving rubber technology and FBR designs. These are given closer look.*

[17] Such as instrumentation and control or I & C cables which serve as vital links between the transducers and instruments for effective monitoring and control of plants.

[18] The temperature range for the TS top plate in warm or lukewarm roof (German parlance), modern FBRs by post-1980s definition is 110- 120 ^{0}C. SPX- 2 (115 ^{0}C) and SNR- 2 (120 ^{0}C) subsuming to EFR design (similar) are indicators.

[19] RP LM freeze seals were categorized as non- replaceable item (along with the RPs) during reactor life at an early stage of FBR development (Table 2.1) i.e. EBR II [106].

Generic elastomers used for inflatable seals of FBR RPs (Table 3.9) have a commonness in excellent resistance to oxygen, ozone, weathering and UV radiation. NBR is an exception [39, 54, 80, 107- 109], primarily because of the presence of double bonds in the carbon- hydrogen structure (CH_2—CH==CH_2—CH) of butadiene (the reason why RCB air side protector seals were required, Table 3.7). Continued use and recommendations of this rubber in USA designs (FFTF, CRBRP, 1000 MWe LMFBR CDs) are attributable to property- improvements by suitable compounding and familiarity with the elastomer because of initial development in Germany (1935) and independent production in USA during WW II along with other elastomers such as CR, Butyl rubber (IIR) and SBR [39, 80, 107, 109]. Of equal importance were safety concerns[20] and zero leakage philosophy of FBRs (1950s - 1970s) which gave maximum importance to radiation and permeation resistance. NBR, chosen as the first elastomer for FFTF cover gas seals (Fig. 3.21, Tables 2.1 and 3.9), displayed most balanced resistance to radiation (from various property aspects, Table 3.8)and the second lowest permeability (to various gases, Table 3.10) during specimen tests, as also indicated by seal-design- guide published (1975) after completion of the FFTF cover gas seal development program [110]. Lower operating temperature of Phenix RP inflatable seal and its secondary function, and compatibility with oil (liquid lubricant) in FFTF and CRBRP (Tables 3.6, 3.9) justified the choice of NBR from lifespan considerations. Phenix construction and testing report (1972) indicated potential difficulties in RP inflatable seal replacement from depositions (sodium and Na_2O) at cooler areas which could also result in friction- welding and seal failure during rotation. The choice of NBR was justified from another very important count, demonstrated by sodium- aerosol- exposure- studies (mid- 1970s) for FFTF- CRBRP cover gas seals which showed heavy deposition on EPDM and Polyurethane elastomeric (AU EU) specimens vis-à-vis very small and negligible on MQ and NBR respectively. FFTF- CRBRP cover gas seal development suggested 5y life for NBR in reactor at design (66 ^{0}C) and operation (51 ^{0}C) temperatures, extendable to 10 y for EPDM qualified for 95 ^{0}C

[20] On higher probability of positive coefficient of reactivity with upscaling to ≥ 1000 MWe rating and melting of metallic fuels in EBR 1 and Fermi.

Table 3.10. Comparison of permeability for seal elastomers

[a] **Gases**	[b] **Material comparison**
[c] Ar, H_2, Kr, Xe permeation	MQ > EPDM > ACM ~ SBR > IIR > NBR > AU, EU
[d] Ar permeation	MQ (x ~ 50) > EPDM (x ~ 11) > ACM (x ~ 4) > FKM ~ SBR (x ~ 3) > AU, EU (x ~ 2) > IIR (x ~ 1.5) > CR ~ NBR
[d] H_2 permeation	MQ (x ~ 250) > EPDM ~ SBR (x ~ 3) > ACM ~ AU, EU ~ FKM (x ~ 2) > NBR > IIR
[d] Kr permeation	MQ (x ~ 60) > EPDM ~ ACM (x ~ 7) > SBR (x ~ 3) > AU (x ~ 2.5) > FKM (x ~ 2) > NBR ~ IIR
[e] O_2 permeation	MQ (x ~ 400) > FMQ (x ~ 90) > SBR (x ~ 15) > CR (x ~ 3.5) > FKM (x ~ 2) > AU, EU ~ IIR ~ NBR
[d] Xe permeation	MQ (x ~ 200) > EPDM (x ~ 16) > ACM (x ~ 10) > SBR (x ~ 6) > AU (x ~ 4) > NBR (x ~ 1.5) > FKM ~ IIR

[a] Permeation studies carried out mostly on 2 mm thick specimens up to 149 ^{0}C – numbers in parentheses indicate permeation multiplication factors on the minimum; [b] Permeation equivalence of materials assumed within a permeability band of ± 15%; [c] Atomics International (AI) study: RT data comparison of Green Tweed, Parker Seal and Minnesota Rubber compounds of various hardness (or filler content); [d] AI study: Comparison taking FKM permeability at 93 ^{0}C as reference on specimens mostly of 70/75 ^{0}Shore A hardness; [e] Aerospace study: Comparison taking FKM permeability at 26 ^{0}C as reference on specimens mostly of 70/75 ^{0}Shore A hardness.

(5 y). Designers' paradox at the end of FFTF development was trade-off between higher temperature capability of EPDM and MQ and lower permeation of AU CU as well as NBR [111]. The FFTF RP inflatable seals made of NBR operated for at least 7 y without failure.

The straight chain macromolecular structure of rubber hydrocarbon, a polymer of isoprene(C_5H_8), is obtained by replacing the hydrogen atom in (CH of) butadiene with methyl pendant group (CH_3) which is again replaced by a chlorine atom (acting as shield to polymeric backbone of 2- chloro-1,3- butadiene) to enhance resistance to oxidation, ozone attack and ageing in CR (indicated by higher temperature rating, Table 3.8), used in the failed inflatable seals of FBTR RPs because of better twisting and dimensional stability compared to NBR in addition. The use of CSM rubber (HP 50 with perbunabase) in RP inflatable seals of KNK- I during early 1970s was a departure from the earlier and subsequent trends of MQ rubber and NBR respectively (Table 3.9). This elastomer has all-round ability with thermal and radiation resistance equivalent to NBR, outstanding resistance to oxygen, ozone, fatigue, cracking, chemicals, oil, grease and good hot tensile properties. The change to silicone rubber in SNR- 300 was probably motivated by chlorine and sulphur (potential of reduced resistance to ageing and corrosion) in CSM rubber and changed location of inflatable seals in SNR- 300 below bearing (vis- a- vis above bearing in KNK- I) with closer proximity to sodium coolant.

Introduction of MQ rubber in DFR (RP O- ring) and Fermi (RP lip seal) was aided by prior knowledge (1958 and before) about its good radiation resistance [112] and usage in critical application of aircraft engine even much earlier (1942). Profound involvement of UK in evolution of science and technology of rubber was a contributing factor initially (DFR, 1959; Fig. 3.21, Table 2.1) and also later during experimentations with uncommon elastomeric sealing configurations and designs for at CDFR (1980s). The innovative use of rubber blocks/ rings in Russian reactors (BN-350, BN- 600, Table 3.7) could perhaps be linked to factors such as early establishment of diene preparation method (as also in England), making of butadiene in 1910 (important role in formation of USSR synthetic rubber industry) and development of rubber similar to NBR as reported in 1942. Lower temperature operation of Fermi (which reduced probability of aerosol deposition), practically zero wear with liquid lubrication and oil compatibility aided the choice of MQ rubber (Table 3.9) which is discouraged for dynamic applications in general because of one of the lowest RT strength. Machined shells and small RP diameter (Table 2.1) supported compact elastomeric lip seal with closer dimensional tolerances (compared to LM seals) in Fermi RP which has similarity with introduction of primary inflatable seals (instead of LM freeze) in larger SPX- 2 RPs based on fabrication and testing experience in SPX- 1 [113]. The American involvement in evolution of rubber from the earliest days (wild rubber as early as in sixth century) through WW- II [109] is certainly amongst the reasons for earliest venturing with an elastomeric (MQ lip) dynamic seal (fuel handling condition) in Fermi RP, also engaged during static operation (Table 3.7)[21].

Success of Fermi lip seal made MQ the recommended rubber (along with the lip seal design) for Rapsodie, FFTF and demonstration as well as commercial FBRs envisaged based on 1000 MWe LMFBR CD follow on studies at the later part of 1960s[22]. Demonstration of sodium/ mist

---

[21]The earliest use of inflatable seal as primary barrier (static and dynamic) in the RPs of German FBR (KNK- I, 1971) is attributed to a rich tradition and development of science and technology of rubber as also to its engineering in machining shells with close tolerance coupled with a tradition of crafts. Interestingly, the first ever introduction of inflatable seal in the RP of an FBR was in France (Rapsodie, as secondary barrier) which did not attain its logical end in Phenix (as primary barrier) because of the delay caused by usage of lip seal and its failure.

[22] The USA LMFBR CD initiation (early 1960s) and follow- on study completion (1969) could be looked at as a part of the initial European and American endeavour to commercialise 1000 MWe LMFBRs during 1980s which resulted in a

compatibility for silicone rubber up to 260 ^{0}C during 1960s and early 1970s with parallel commercialization [114- 115] of aerosol- resistant compound (French product literature indications) were amongst reasons for continuation of this generic elastomer. Subsequent change to NBR (FFTF-CRBRP) was primarily induced by the highest permeability of MQ (~ 100 times of NBR, Table 3.10). The resurgence of MQ rubber as European favourite (Fig. 3.21, Table 3.9) was supported by earlier studies, chemical inertness and key advantages such as one of the best retention of RT T_s and E_b at elevated seal operating temperature or ET (*hot tensile properties*). Limitations of silicone rubber were addressed by design such as bead/ lip on sealing face (SNR 300, EFR; Fig. 3.26), additional Viton® layer beneath silicone tube (SNR 300), material reinforcement (SPX 1, EFR; Fig. 3.26) and thickening of seal wall (EFR, Fig. 3.26) to reduce wear, tear and permeation in inflatable seals where Teflon based antifriction coating was used (Tables 2.1, 3.6).

The Japanese endeavour (1970s- early 1990s, Tables 2.1 and 3.9) on RP inflatable seal development introduced FKM rubber (Viton®) as high temperature elastomer of significance with very strong all round positive aspects positive aspects such as continued high temperature performance (> 200 ^{0}C, Table 3.8),outstanding thermal stability, ageing resistivity in the presence of oxygen and ozone, one of the lowest permeability (Table 3.10), radiation resistance more than sufficient for cover gas seals (Table 3.8) and compatibility with wide range of fluids. The Japanese approach is supported by more than decade long recent studies (till now) on long- term accelerated ageing and life prediction for elastomeric seals in contact with SS (operating conditions similar to INDIAN- SFR RP inflatable- backup seals with specification temperature, radiation and life of 149 ^{0}C, ~ 2 kGy and 10 y) of radioactive material package casks (including those in shipping packages) involving peroxide cured Viton® based on APA [67- 73].

---

report during 1970, capturing the initiatives for safety, economy and sustainability. European designers' organisation observed during mid- 1980s that FBR per- kWh power generation cost could be brought down to 1.1 times that of an LWR. R & D activities in USSR during mid- 1980s were similarly fashioned with capital cost of BN- 600 1.65 times that of an LWR. The construction cost of 660 MWe DFBR- 1 was estimated as < 1.5 times that of an LWR during the year (1994) of its specification finalisation.

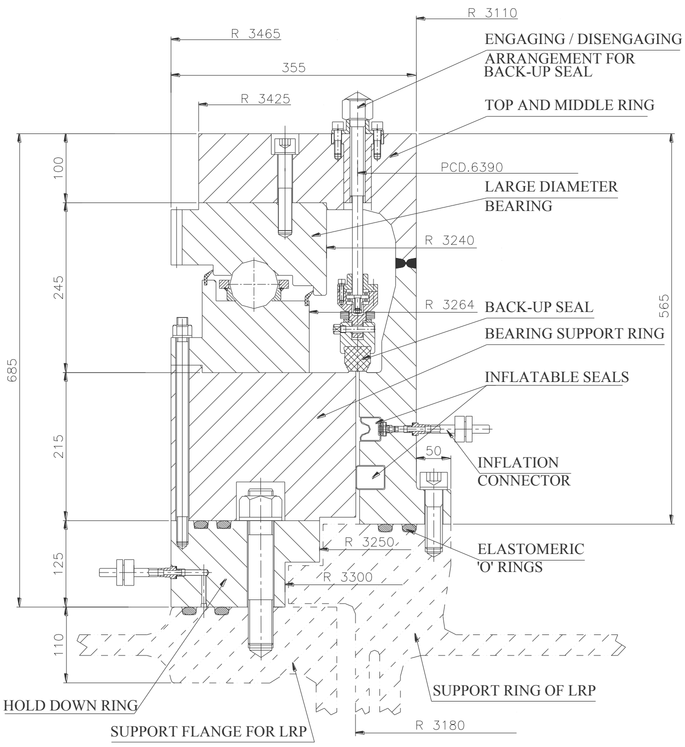


Fig. 3.32. Inflatable and backup seals in INDIAN- SFR large rotatable plug during normal operation.

These assessments provide sufficiently clear indications about the suitability of a non- reinforced, high temperature elastomer as the common generic elastomer across categories of cover gas seals (of MOX fuelled FBRs) for simplification, unification and standardisation taking material as the cornerstone, FEA as facilitator (replacing manufacturing technology) and design as an all-encompassing base.

3.6 Summary of Special Elastomeric Component R & D Results for Reference: *I*

These are material development, FEA based sizing, manufacture and quality control results of FKM and EPDM inflatable seals for INDIAN- SFR (Fig. 3.32) and FBTR (Fig. 3.33) RPs respectively, referred throughout the thesis for data extrapolation and design.

3.6.1 Background [36, 45]

Special Elastomeric Component development arrived at an extruded, non- reinforced and un-beaded inflatable seal design (mechanical inflation connector and end joint 1 each per seal, *Figs. 1.6, 3.22, 3.30*) for the RPs of INDIAN- SFR and FBTR which was unique in international context of reinforced and beaded inflatable seals (FFTF, CRBRP, SNR- 300, SPX- 1, EFR; Table 3.6, Fig. 3.26), with production technology as facilitator. Absence of production support (co- extrusion

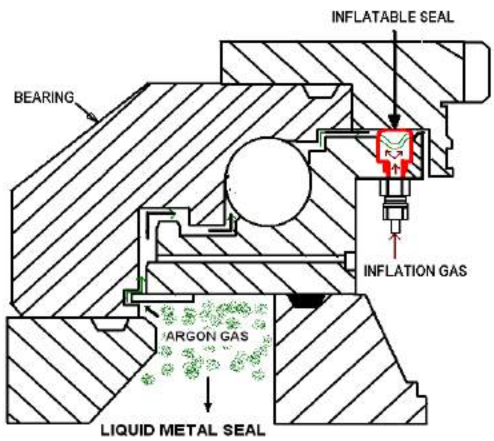


Fig. 3.33. Location and mounting of inflatable seal in FBTR rotatable plugs.

Table 3.11. Inflatable seal compound formulations for hot feed extrusion and autoclave cure

Compounds	**Ingredients** *(Parts per hundred parts of rubber or phr)*								
	Viton® A-401C	Nordel® IP 4520	FEF black (N550)	MT black (N990)	MgO	$Ca(OH)_2$	TMQ	DCP-40	Paraffin oil
S1 (FKM)	100	---	---	20	3	6	---	---	---
S3 (EPDM)	---	100	50	---	---	---	1	7	10

etc.) in Indian context for compensation of material shortcomings (by beads, reinforcement etc. in NBR and MQ) in primary inflatable seal (operating at ≥ 100 ^{0}C in warm roof) made development of non- reinforced elastomer much more challenging compared to international endeavours as readymade commercial compounds, seals and coating technology were not available for nuclear-specific evaluations. Material development was always at the centre stage in terms of R & D efforts internationally (with particular emphasis on permeability minimisation and life maximisation during the later stage of LMFBR cover gas seal development, German development and those for EFR during 1993) even with the aid of production technology and commercially available items. In this context, the research addressed in this thesis contemplating for 1 synchronised replacement during 60 y of reactor life is entirely new in terms of concept, efforts and implementation till date.

Table 3.11 shows the FKM/ EPDM formulations (S- 1/S- 3) developed for INDIAN- SFR/ FBTR RP inflatable seals (suitable for hot feed extrusion and pressurised autoclave cure in steam at typical temperature/ pressure of 150- 170 ^{0}C/ 0.55- 0.7 MPa) assuming continuous operation at 120/ 80 ^{0}C under differential pressures of 25/ 10 kPa during design life (10 y) in the presence of γ dose, contacting materials and fluids indicated in Table 3.3. Figure 3.34 shows cross-sectional view of the first radial, non-reinforced, unbeaded INDIAN- SFR inflatable seal design (groove size: 36.4 mm x 27 mm) made of bisphenol cured Viton® A-401C based S-1 and sized by FEA to operate at inflation pressures of 50- 30 kPa (maximum contact pressure at sealing surface: 70- 50 kPa; minimum contact pressure for leaktightness: 35- 7 kPa) during normal-operation- fuel-handling condition.

3.6.2 Demarcation of referred and thesis research

This thesis refers R & D results from the development of inflatable seals in the domains of material (S-1, S- 3), FEA based sizing and design (S- 1, S-3 for 36 mm/ INDIAN- SFR and 23. 6 mm/ FBTR seal- width respectively), life assessment by Arrhenius methodology (S- 1, S- 3), PECVD Teflon- like coating (S- 1, S-3; S- 2 or 50:50 blend of peroxide cured Viton® GBL 200S: 600S), manufacture (S- 1, S- 3), validation (S- 1, S- 3) and quality control (S- 1, S- 3).

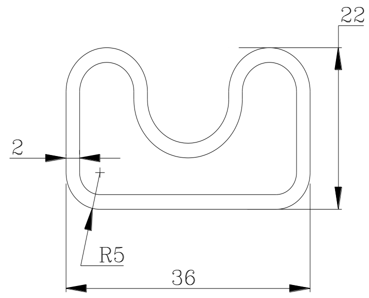


Fig. 3.34. Unbeaded design of non-reinforced inflatable seals for INDIAN- SFR rotatable plugs made of S-1.

FEA based sizing and design of inflatable seal (33.6 mm width, 34 mm x 26 mm groove) withstanding precommissioning conditions in INDIAN- SFR RPs for the past 6 y is purely a part of this thesis. Testing, inflatable seal production trials and optimisation of material and production to arrive at S- 2, testing to arrive at S- 4 (Viton® A-401C based, developed for backup seal) and sizing, design, optimisation by FEA to produce and validate 1 m diameter test seal is part of the thesis along with quality control. Production of reactor seal made of S-4 and related quality assessments are referred to complete the circle. Life assessment by application of S-4 data (accelerated aged) in Arrhenius and WLF equations and FEA are purely a part of this thesis as also extending the results by literature data extrapolation (in combination with principles of design, R & D results and an approach of material engineering) towards 1 synchronised seal replacement in

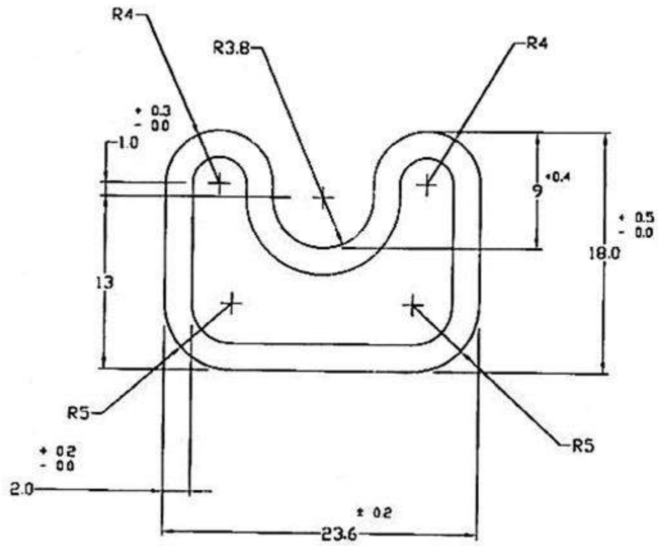


Fig. 3. 35. Unbeaded design of non-reinforced inflatable seals for FBTR rotatable plugs made of S- 3

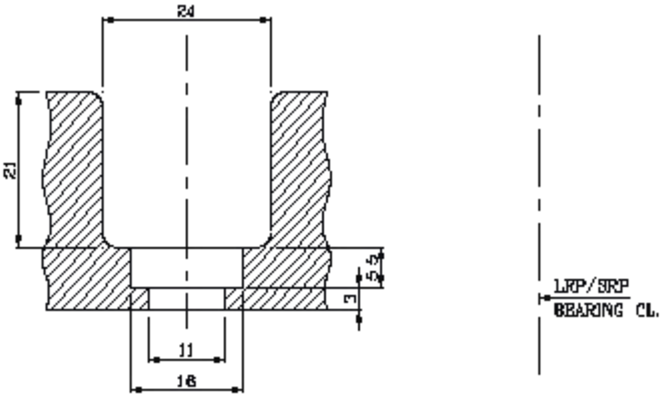


Fig. 3.36. Detail of existing rectangular groove in FBTR rotatable plug for inflatable seal

60 y and unification by simplification as well as standarsation. Standardisation of the hybrid module of R & D (8^{th} element of design) for future implementation in SFRs and other nuclear genres is made of complimentary testing and analysis ingredients from development of *Special Elastomeric Components* (as already indicated) and this research, while amalgamating the results for standardisation fall purely within the purview of this thesis.

This work is basically a combination of three domains (i.e. research based on already generated data, i and ii; research based on data generated as part of this thesis, iii; research combining iv and v belonging to design- namely, i) Literature review and survey (INDIAN- SFR, FBTR seal studies etc.) for problem definition, ii) Literature review for data extrapolation, iii) Experiments (specimen testing + production trials), analysis (FEA + Arrhenius + WLF) and conclusions obtained from them, iv) Design (includes representative seal and generic elastomer identification, problem definition, material- geometry- groove- production- coating- validation- quality choices, life assessment from all aspects, overall benefit assessment from simplification-unification-standardisation etc.) and, v) Concepts and processes of simplification-unification- standardisation including R & D standardisation.

3.6.3 Material development and sizing by finite element analysis [36, 45]

Axial, EPDM inflatable seals of FBTR (Fig. 3.33) were sized (Fig. 3.35) for existing 24 x 21 mm groove (Fig. 3.36; inflation pressure: 45/25 kPa). Design drag of 1000 N/m was applied to arrive at the INDIAN- SFR (FBTR) inflatable seal designs by FEA for the ~0.5- 2 m (~ 0.5- 1.5 m) diameter test seals and ~6.3- 4.2 m (3.9- 2.4 m) diameter LRP- SRP seals made of non-reinforced FKM (EPDM).The limiting principal and von Mises stress- strain in S-1 and S-3 used for design were 1.63 MPa- 40% and 3.35- 105% respectively using a FOS of 2 on T_s/E_b of the seal materials determined at 120^{0}C and 80^{0}C (or ET).S- 1 displayed T_s, E_b, hardness and tear resistance of 12.28 MPa, 217%, 68 ^{0}Shore A and 31 N/mm, measured at RT. Drop of 70–75% in T_s at 120 ^{0}C is in tune with the fact that fluorocarbon rubbers derives a great deal of RT strength from ionic attraction which

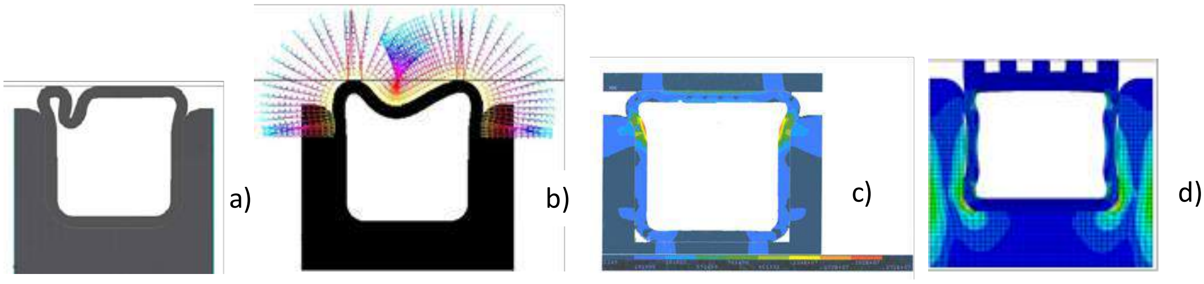


Fig. 3.37.a) Inflatable seal buckling at main sealing face, b) Contact pressure profile, c) von Mises stress distribution with uniform inflation and contact at seal rubbing face and, d) Inflatable seal with beads on seal rubbing face.

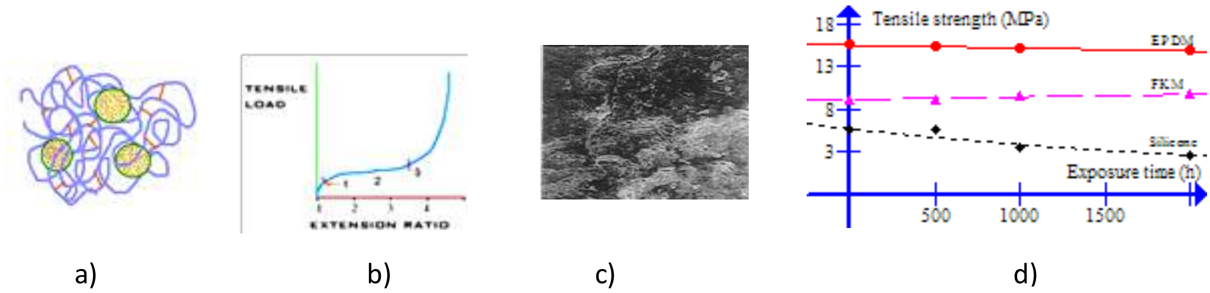


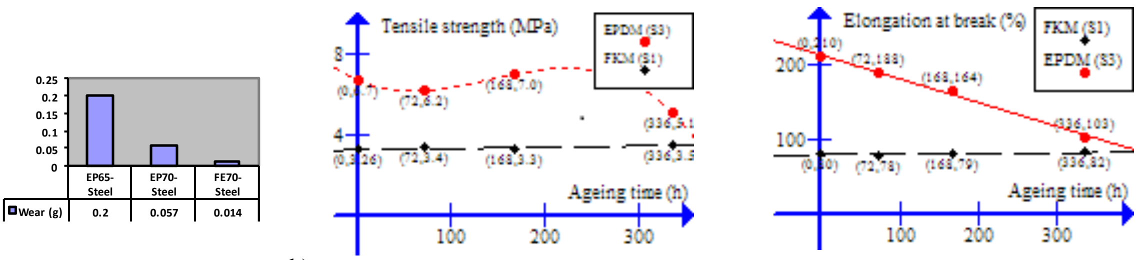
Fig. 3.38.a) Elastomer compound structure with filler aggregates and cross-links, b) Non-linear load-deflection behaviour of elastomers: I-near-Hookean, II- softening, III- stiffening, c) Surface morphology of silicone rubber after sodium aerosol exposure at 120^{0}C for 2000 h and, d) T_s variations (measured at RT) of EPDM, FKM and MQ compound specimens during sodium aerosol exposure at 120^{0}C.

reduces drastically at elevated temperature [54].

Figures 3.37 a-d show FEA implementation of INDIAN- SFR and FBTR RP inflatable seals in ABAQUS and ANSYS using Mooney- Rivlin material model as constitutive relation, plane strain condition for first approximation and axisymmetric condition for finer and final evaluation of load, stress, strain and failure. Differential pressure exceeding contact pressure (leakage) and principal stress- strain in seal exceeding T_s- E_b (divided by FOS; cracking) were applied as the basic criteria of failure by leakage and cracking of seal material respectively. Viton® A-401C (S-1, Table3.11) is a dipolymer of HFP and VDF with nominal fluorine content of 66% by weight (wt. %)and specific gravity of 1.82. This is supplied as a precompound to assure better dispersion of accelerator (phosphonium or amino- phosphonium salt) and curative (bisphenol AF) which ensures enhanced reproducibility of cure and properties between different compound batches. Peroxide cured EPDM grade Nordel IP 4520 (S-3, Table 3.11; specific gravity: 0.86; Mooney viscosity: 20 ML 1 + 4 @ 125 ^{0}C) contains 50 wt.% ethylene and 45 wt.% propylene. Figures 3.38 a-b show structure and load-deflection behaviour of elastomer. Slab samples of MQ exposed to sodium aerosol showed discolouration, pitting and crazing during visual examination from 500 h onwards which were subsequently substantiated by scanning electron micrograph (Jeol JSM-35 G at 480 × magnification; Fig. 3.38c) and large T_s-E_b drops of 54-112% (Fig. 3.38d).

Further studies with EPDM and FKM (unaffected by aerosol exposure) specimens of hardness 65 ^{0}Shore A (EP 65) and ~70 ^{0}Shore A (EP70, FE70) indicated much better wear resistance (measured at RT) of FKM (Fig. 3.39a). Fig. 3.39b shows exceptional thermo-oxidative ageing stability of S-1 (FKM, Table 3.11) compared to S- 3 (EPDM, Table 3.11) with accelerated ageing in air carried out at 150 ^{0}Cand properties measured at 120^{0}C (FKM) and 80^{0}C (EPDM) respectively. Predictions using accelerated thermo- oxidative ageing up to 200 ^{0}C and Arrhenius methodology indicated 24 y and 10 y of life for S-1 (FKM, at 120 ^{0}C) and S-3 (EPDM, at 80 ^{0}C) respectively taking 50% drop in T_s and E_b (measured at 120 ^{0}C and 80^{0}C) as the common criterion of failure [116].S- 1 (FKM, Table 3.11) and S- 3 (EPDM, Table 3.11) were assigned as the compounds (as well as generic elastomers) for INDIAN- SFR(primary) and FBTR (secondary) inflatable seals respectively based on these results and other studies on processability, permeability etc.

All the 4 beaded and un-beaded geometries (Fig. 3.30) were designed by FEA in ABAQUS and ANSYS (with S- 1 and S-3 stress- strain data) using plane strain, axisymmetric conditions and 30–60^0 sector models of seal torus which involved mixed and hybrid formulations and a number of constitutive relations (including the Mooney-Rivlin material model). A FOS of 2 was applied to account for seal inflation pressure > 50 kPa during operational variations, batch-to-batch variation of T_s and E_b in elastomeric compounds [107], laboratory to laboratory (inter-lab) variation in measurements of T_s and E_b [117], synergistic ageing (temperature + radiation + mist + air) etc.



a) b)

Fig. 3.39. a) Comparative wear of EPDM and FKM and, b) T_s and E_b variations of S-1 and S-3 with ageing at 150^{0}C in air.

Mooney-Rivlin material model was used to finalise seal dimensions (including Figs. 3.34- 3.35) as maximum tensile strain in seal during normal operation was always below 40% (Table 3.3). The Mooney-Rivlin model correlates well up to ~225% of tensile strain when results from plane- stress FEA model of ASTM D412, uniaxial (1- D) tensile specimens are compared with experimental results from rubber with low filler content (static seals), such as medium thermal or MT black [108]. Use of Mooney-Rivlin for strains of 100–200% has been reported [77]. However, engineering elastomers have higher filler (for higher strength/hardness of dynamic seal, S- 1, Table 3.11) which influences stress–strain curve. In such cases the model works well up to about 50% strain [118- 119].

3.6.4 **Manufacture** [36]

Successful deliveries of test inflatable seals of ~0.5mdiameter were based on hot feed screw extrusion and autoclave cure. Seal end-joints were obtained by holding the under-cured open ends of extruded seal profile under heat and pressure in a mould.

3.6.5 **Coating** [36, 45, 124]

The development of plasma based coating procedure was initiated based on a design specification (Table 3.5), similar to the design specification for seals (Table 3.3).

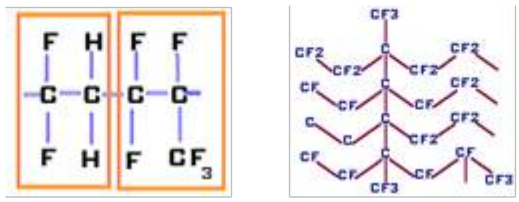


Fig. 3.40.a) Chemical structure of fluorohydrocarbon rubber and, b) Typical structure of Teflon-like coating deposited by plasma enhanced chemical vapour deposition.

PECVD technology at the initial stage employed radiofrequency power source (13.56 MHz) and capacitive type electrode coupling (parallel electrode configuration) to grow smooth, defect free, lubricious fluorocarbon films on the substrate under vacuum. In this process fluorocarbon precursor molecules or monomers ($CF_2{=\!=}CF_2$ and CF_3–$CF{=\!=}CF_2$), obtained from pyrolysed Teflon, are fed into the vacuum of reaction chamber by high purity carrier gas, nitrogen. The monomers are subsequently decomposed (fragmented) by radiofrequency plasma into various radicals (CF, CF_2, CF_3, etc.) and are made to react with the substrates, kept at various temperatures (ambient to150^{0}C) for the deposition of films [120- 122].

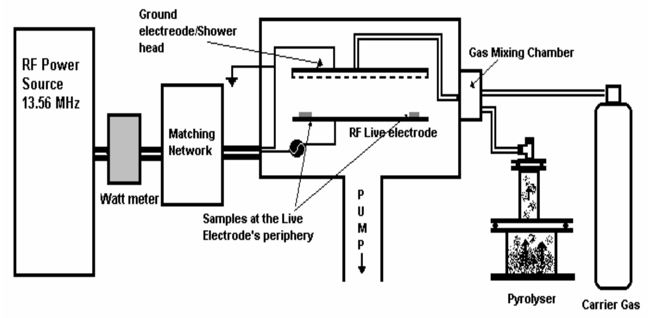


Fig. 3.41. Schematic of Teflon- like coating facility in 0.5 m diameter scale by plasma enhanced chemical vapour deposition.

Fig. 3.42. Reactor for Teflon- like coating of 1-7m diameter seals by plasma enhanced chemical vapour deposition.

Fig. 3.40b shows typical chemical structure of PECVD based Teflon-like coating along with the chemical structure of typical bisphenol cured FKM rubber based on VDF and HFP (Fig. 3.40a). A schematic of the PECVD coating setup is shown in Fig. 3.41.

The basic PECVD technology was augmented by low substrate temperature technique [122] and pulsed direct current discharge to deposit coatings of required thickness (elastomer: 5- 8 µm; steel: 10- 15 µm) and adhesion strength (≥ 4.5 MPa) on elastomer (EPDM, FKM) and steel (carbon steel, SS) surfaces, respectively. The initial specifications on adhesion strength (≥ 7MPa) and porosity (≤ 25%) of coating was modified to ≥ 4.5MPa (minimum) and 10% (maximum) respectively based on the study. An interesting study carried out during this period used atomic force microscope in friction force mode to detect the onset of chain scission and cross-linking in the macromolecules of the γ- ray irradiated EPDM and FKM samples by measuring the local frictional properties [123].

Sample level studies on elastomers were successfully extended to coat ~0.5m diameter EPDM test inflatable seals. Subsequently a PECVD reactor (length: 8m; diameter: 0.8m) was designed and commissioned based on this concept which drives large diameter (1- 7m) inflatable seals by motor-pulley arrangement under vacuum to carry out coating continuously (Fig. 3.42). A few trials were carried out on ~1.5/2mdiameterEPDM/FKM test inflatable seals.

Results of PECVD coatings on steel were mixed. Coating on carbon steel showed peeling because of corrosion of the substrate. Coating on 2 m diameter test SS shell (emulating the coated mating surface for IFTM inflatable seals on LTC; Fig. 3.13) was carried out successfully [124].

3.6.6 **Quality control** [126- 127]

Quality ensures consistency across multidisciplinary domains of laboratory compound development, FEA based seal sizing and test as well as prototype seal manufacture by using a set of representative parameters and their limits (Table 3.12). The set (evolving through development) does also attempt to provide specifications of material, manufacture and quality control for regular production of reactor seals and minimisation of future R & D [125] with better unification by standardisation and reproducibility. The set has potential use in PHWRs and AHWR. The evolution summarised here for reference and extended to complete the quality control scheme (with this thesis work) was instrumental in arriving at S- 1, S- 2 and MQ compounds and the seals manufactured by them, define stress–strain limit of the seals for sizing and ensure consistency across stages.

Table 3.12 shows the major standards used for determination of properties. Conceptual Phase was completed with sodium aerosol compatibility tests which did also mark the beginning of Developmental Phase I with a revised specification (Tables 3.4, 3.12). Developmental Phase I ended with identification of S- 1 and S- 3 including production of seals which initiated Phase II with a revised specification involving S- 2, to be addressed in Chapters 4 and 5.

During the Conceptual Phase and Developmental Phase I compounds were mixed in laboratory Banbury and Brabender Plasticorder (as per ASTM D 3182), with the later used for assessment of extrudability of uncured compound stock along with Monsanto processability tester (MPT 2000, USA). Rubber process analyser, Mooney viscometer and moving die rheometer were used for measurement of curing and rheological characteristics in accordance with ASTM D5289 and ASTM D 1646. Dumbbell specimens for tensile stress–strain measurements were punched from test slabs moulded as per ASTM D3182.

Table 3.12. Property limits of INDIAN- SFR rotatable plug inflatable seal material during different phases of development

Properties [a]	Property limits [a]		
	Applicable standards	Specimens from laboratory batches	Specimens from laboratory batches
		Conceptual phase	Development phase I
T_s(MPa)	ASTM D412 Die C, ISO 37 Type 2 Die (struin rates: 5 mm/min, 50 mm/min, 500 mm/min)	≥ 10.5	≥ 13
E_b (%)		≥ 300	≥ 150
Hardness (^{0}Shore A)	ASTM D 2240, Durometer A	70 ± 5	65 ± 5
Tear Strength (N/mm)	ASTM D 624	18	
Resilience (%)	ASTM D 945	≥ 58 (Yerzley, 3.7 Hz)	Not used
Hysteresis (%)		≤ 50 (Yerzley, 3.7 Hz)	Not used
Abrasion resistance (%)		70	Not used
Fluid compatibility, swell/shrink (%)		≤ 20/5	Not used
CS (%)	ASTM D395, Method B (25% deflection)	≤ 25 [b]	≤ 25 [b]
Dry Heat Resistance	ASTM D 573	[c] Change in hardness, T_s and E_b not to exceed 10%, 20% and 25% respectively.	[c] Change in hardness T_s and E_b not to exceed 10%, 20% and 30% respectively.
Specific gravity	ASTM D 297, ISO 2781(Method A)	Difference between two compound batches: ± 0.01 – 0.05	

[a] Limits of properties measured at RT. [b] After 70 h at 150 ^{0}C. [c] After ageing in air at 150 ^{0}C for 70 h.

Properties were specified during the conceptualization phase (Table 3.12) considering the seal operating requirements, design practices and development as well as operating experiences of inflatable seals of FBTR and other FBRs. About 20 nos. of EPDM, FKM and MQ compounds (60/70/80 ^{0}Shore A hardness) were prepared by varying the proportion of ingredients and mixing parameters. Optimum curing conditions using bisphenol (FKM) and dicumyl peroxide (EPDM and MQ) and extrusion parameters were established. Compounds were chosen for sodium aerosol tests mainly from specification hardness (which also defines the Young's modulus) requirements.

Reduction/ increase of hardness/ T_s in the revised specification during Development Phase I (Tables 3.4, 3.12) were aimed at reducing the modulus (stress in the seal material for a particular strain) and stress in the seal for the same inflation pressure which increases the FOS at ET. The presences of Teflon/Teflon-like antifriction coatings on seal and mating surfaces which minimizes friction and wear and confines the losses to the coated layer, allowed lowering of compound hardness

Table 3.13. Composition of EPDM candidate compounds

Ingredients (phr)	Candidate compounds			
	EPDM- I	EPDM- II	EPDM- III	EPDM- IV
EPDM rubber- Nordel® P 4520	100	100	100	100
Filler- Fast extrusion furnace (FEF) black N550	50	50	50	50
ZnO	0	5	5	0
Antioxidant- TMQ (phr)	1	1	0	1
Curing agent- Dicumyl peroxide (DCP- 40 % active)	7	8	7	7
Processing aid- Paraffin oil	10	10	10	10
Accelerator- Triallyl Isocynurate (TAIC)	0	1	1	1
Batch weight	168	175	173	169

to 65 ±5^{0}Shore A. The elimination of MQ rubber from candidature similarly allowed raising of minimum T_s from 10.5 MPa to 13 MPa. Minimum E_b of seal material was reduced to 150% to encompass the lower ultimate stretch of FKM grades and to address the possible variability of composition and properties during development. The only difference in FBTR seal material specification is a lower minimum T_s of 10.5 MPa. This was feasible because of higher T_s (6.7 MPa) of EPDM-I (S- 3) at 80 ^{0}C.

Better hot tensile properties of EPDM-I (S- 3) at 80 ^{0}C compensates for the drop of T_s and E_b with thermo-oxidative ageing (Fig. 3.39b). The higher E_b of EPDM-I (S- 3) at 80 ^{0}C (210%) compared to that of FKM-I (S- 1) at 120 ^{0}C (80%) similarly provides higher margin to absorb elongation drops during ageing (Fig. 3.39b). The raising of T_s for INDIAN- SFR inflatable seal material (Table 3.12, Development Phase I) was intended to ensure that the seal material retains sufficient T_s and E_b (at seal operating temperature) under synergistic ageing at the end of design life so as to avoid localized cracking failures because of stress and strains in seals exceeding the material limits, in view of inherently low hot tensile properties of FKM.

Table 3.14. Composition of FKM candidate compounds

Ingredients (phr)	Candidate compounds			
	FKM- I	**FKM- II**	**FKM- III**	**FKM- IV**
FKM rubber- Viton® A-401C	100	100	100	100
Filler- Medium thermal (MT) carbon black N990	20	30	30	15
Acid acceptor- High activity MgO	3	3	3	3
Activator- Rubber grade $Ca(OH)_2$	6	6	0	6
Batch weight	129	139	133	124

Manufacture (by hot feed extrusion and autoclave cure) and validation of test seals for INDIAN- SFR and FBTR designs in scaled rig (Fig. 3.28) authenticated the effectiveness of revised property limits in compound development, manufacture and quality control.

Fig. 3.43 a–c shows deformation, failure and Mises stress distribution in 2- D (static sealing) and 3- D (dynamic sealing) models of FBTR inflatable seal created in ANSYS based on Mooney-Rivlin material model using hyperelastic elements HYPER 74 (2-D) and HYPER 84 (3-D). Fig. 3.43a shows buckling failure (functional rather than material) of seal in 2-D model because of oversized fold (Figs. 3.34- 3.35).

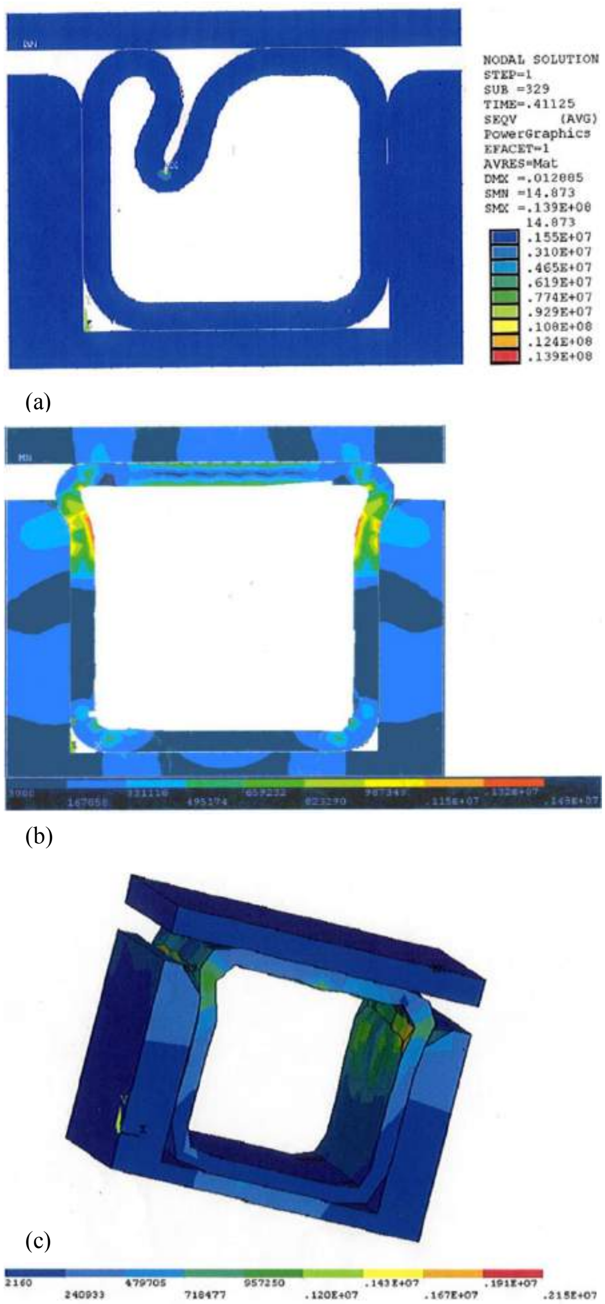


Fig. 3.43. Deformation, failure and Mises stress field in FBTR inflatable seal models. (a) Buckling failure at seal rubbing face under inflation pressure of 82 kPa. (b) Deformation and Mises stress field under inflation pressure of 200 kPa. (c) Deformation and Mises stress field under inflation pressure of 50 kPa with frictional drag acting perpendicular to seal cross-section (μ= 0.2).

The FEA results qualify the assumed limits of T_s and E_b in the revised specification (Table 3.12, Development Phase I) for seal design considering the effects of synergistic ageing.

3.6.7 Capturing special elastomeric component development [9- 10]

Adoption of all-elastomeric sealing scheme for INDIAN- SFR RPs to place SRP within LRP (Fig. 2.3) and two decades of R&D on elastomer, seal and coating involving Defence Materials Stores Research and Development Establishment (DMSRDE; Kanpur, India) as the first partner from 1999, Institute for Plasma Research (IPR; Gandhinagar, India), Hari Shankar Singhania Elastomer and Tyre Research Institute (HASETRI; Kankroli, India), 4 Indian Institute Technologies (IITs; Delhi, Kanpur, Kharagpur, Madras) and about equal number of other Indian agencies have been captured in a number of publications, out of which this thesis forms a part [9-10, 36- 37, 41- 43, 45, 74, 81, 87, 91, 116, 120- 124, 126- 145].

3.7 Harmful Volatile Release from Fluoroelastomer by Operating Temperature [81]

This assessment is carried out by literature data extrapolation.

Given the fact that the possible emission of volatiles (HF, CO_2, water vapour etc.) is from unused inorganic bases in cured product (Section 3.4.2.1), the same cannot exceed the release of volatiles during original curing of green elastomer.

Product literatures report physical properties of Viton® based on tests carried out on specimens that are extracted from standard test slabs prepared by press-curing (compression moulding) at 177 ^{0}C followed by post-curing at 232 ^{0}C [146- 149]. Early product literature [150] indicates that toxic vapours which may include HF may be liberated from Viton® products at service temperatures above 200 ^{0}C. This is demonstrated by a later information bulletin [151] based on studies carried out under simulated conditions of press-cure (193 ^{0}C/10 min) and post-cure (232 ^{0}C/24 h) on specimens made of bisphenol cured Viton® E-60C, a copolymer of VDF and HFP, which contained 3 parts per hundred parts of rubber (phr)of MgO, 6 phr of CaO and 30 phr of MT carbon black in its recipe. Tests showed that HF constitutes about 1 wt. % out of the total 1.5 wt. % of volatiles evolved during press and post cure of specimens [151]. The INDIAN- SFR RP inflatable seal made of S- 1 weighs about 0.5 kg/m length of seal profile (Fig. 3.34). Estimations indicate that the INDIAN- SFR LRP inflatable seal weighing about 10 kg would generate ~150 g of volatiles during cure out of which HF takes only ~1.5 g.

As the inflatable seals of INDIAN- SFR RPs usually operate at ~ 100 ^{0}Cand the threshold temperature for HF release is 160 ^{0}C, it is concluded that HF release from S-1 due to operating temperature is going to be nil for all practical purposes, irrespective of the operating span.

3.8 Harmful Volatile Release from Fluoroelastomer by Operating Radiation [81]

This estimation is also by literature data extrapolation.

Nuclear and space radiation studies on materials [152] indicated that 10 kGy of cumulative dose imparted on fluoroelastomer at RT results in 25% drop in T_s accompanied by gas evolution. Product literature indicated that a proprietary sealing compound based on Viton® B displays scission dominated phenomenon to produce fluorohydric acid vapour at a dose of 0.1 kGy imparted in air at RT at a high dose rate of 1.4 kGy/h [153] which could be explained by radiation-induced oxidation, supported by dissolved oxygen in test specimens [154]. Another seminal research showed that for Viton® B specimens vulcanized by diamine, weight loss from release of low molecular weight volatile products (such as HF) increases with cumulative dose and the temperature at which such dose is imparted in air [82]. Reports from earlier works [154] indicate a linear relationship between the increasing dose and weight loss in VDF–HFP fluoroelastomers. It was shown that VDF–HFP fluoroelastomer specimens loose 0.4% weight under cumulative dose of 650 kGy at 150 ^{0}C.

Simple extrapolation of this data indicates that the INDIAN- SFR LRP inflatable seal would release 0.123 g of HF at the most under cumulative design dose of2 kGy at 150 ^{0}C. Considering the facts that such release would occur over a period of 10 y and operating temperature is ~ 100 ^{0}C, it is observed that the effect of HF release because of γ dose should be insignificant.

3.9 Finalising the Common Generic Elastomer for Unification [9- 10, 36,41-43, 45, 81, 145]

Non- reinforced fluoroelastomer is chosen as the common generic elastomer for unification across categories of INDIAN- SFR cover gas seals, aiming at 1 synchronised replacement in 60 y. Two high temperature elastomers (ACM and FMQ), not considered for inflatable seal (Section 3.4.2.4), are given a relook from the consideration of common generic elastomers. ACM has the lowest heat resistance amongst all the 4 high temperature generic elastomers (ACM, FKM, FMQ, MQ) and the high temperature rating (177- 204^{0}C) provided in Table A.1 (comparison of complementary properties not included in main text, Appendix A) is for short term only. The elastomer has limitations in tear resistance and CS (Table A.1) is one of the lowest amongst elastomers which reinforce its exclusion from candidature for cover gas seals. Above all, ACM rubber experiences embrittlement (loss of flexibility, elongation, and tensile strength) on extended elevated temperature use which it out of any consideration. Apart from difficulties in adhesion to metallic parts, FMQ has the other strong shortcoming in one of the lowest CS resistance which leaves EPDM, FKM and MQ for comparisons [54].

3.9.1 Eliminating EPDM rubber

EPDM was never used in any inflatable seal of FBR RP and there were only 2 recommendations from later part of 1960s to early 1990s. 5 y of life (at 95 0C) indicated by results from the≥ 1000 MWe, USA LMFBR cover gas seal development is supported by 10 y of projected life obtained for S- 3 at 80 ^{0}C (FBTR, Section 3.6.3). Elimination of EPDM is further substantiated by non- usage of this generic elastomer for FBTR (Table 2.2) where replacement data of ~ 5000 elastomeric O- rings indicates that about 80% (of the ~ 17% replacements per year, driven mostly by CS based failure) of the replaced seals were made of low to moderate temperature (maximum 149 ^{0}C i.e. CR, Table 3.8) generic elastomers (CR, NBR, SBR, Table 2.2; average life: 3- 5 y) with maximum temperature rating similar to EPDM (149 ^{0}C, Table 3.8). FKM and MQ O- rings with high temperature rating (> 200 ^{0}C, Table 3.8) occupied about 12% of total yearly replacements with average life of 15- 20 y which tilted the choice towards these generic elastomers subsequently. One of the reasons for such non- usages could be attributed to one of the poorest resistance to thermal ageing in air (in terms of retention of tensile properties with time or thermal stability; Fig. 3.39) which more than nullifies better hot tensile properties initially (unaged condition) with synergistic ageing to take away the margin of safety or FOS, optimum resistance to tear and flex cracking notwithstanding (Table A.1). This inherent weakness of EPDM rubber in air is non- existent in hot water or steam resulting in popularity in thermal reactor domains. With double elastomeric sealing contact in all TS sealing usages, about 50% of barriers on average are in contact with air which suggests exclusion of EPDM from the candidature of common generic elastomer for cover gas seals.

The most important cause for non- usage of EPDM rubber as FBR inflatable seal material in specific and cover gas seal elastomer in general could perhaps be attributed to maximum mist deposition potential amongst the candidate elastomers (Section 3.5) with propensities of replacement difficulties and friction welding based failures- the later demonstrated by specimen studies as part of LMFBR cover gas seal development. About 50% of cover gas seals or the primary elastomeric barriers are continuously exposed to sodium aerosol. Recommendation of EPDM for the secondary inflatable seal in FBTR (not used ultimately) could also be justified by the fact that chances of exposure to sodium aerosol is negligible.

EPDM is therefore eliminated from the candidature of INDIAN- SFR RPs in specific and cover gas elastomeric seals in general.

3.9.2 Eliminating silicone rubber

Reasons for emergence of MQ compounds as European favourite for RP inflatable seals has been indicated. Tables 3.15- 3.16 compiled from two groups of sources, spanning from early 1980s

Table 3.15. Elevated temperature tensile properties of generic elastomers

Properties	Generic Elastomers									
	ACM	AU, EU	CO, ECO	CR	EPDM	FKM	SBR	IIR	MQ	NBR
$^{a}T_{s}$ (MPa)	16.1	28-55	13.2	3.4-21	14-21	10.3-21	3.4-21	14+	4.1-10.3	6.9-24
$^{b}T_{s}$(MPa)	15.6	12	8.5	10.3	14	2.1-5.5	8.3	5.5-6.9	3.7	4.8
$^{a}E_{b}$(%)	272	250-800	630	650-850	500	100-450	450	300-800	90-900	400-600
$^{b}E_{b}$(%)	278	300	600	350	300-500	100-350	250	200	200	120

[a] Measured at RT. [b] Measured at 121 ^{0}C excepting ACM (measured at 150 ^{0}C); Source: EPRI, 2007.

[80, 107- 108] to the first decade of new millennium [54], suggest that the earlier preferences (early 1990s) for FKM and MQ as the potential, common high temperature generic elastomer remain unchanged in the present context. A large part of global R & D efforts on RP inflatable seal material from the later part of 1970s could be attributed to permeability and strength limitations of MQ rubber which, along with increased performance (and life) demand from primary inflatable seal located near TS top plate in warm roof, kept the search for better diffusion- proof MQ compound continuing even at the concluding stage (1993) of EFR conceptualisation. Using a rubber as common generic elastomer for all the cover gas seals adds additional demand on material performance such as

Table 3.16. Operating temperature range (T_R) and glass transition (T_g) comparisons across generic elastomers and three decades

Properties	Generic Elastomers											
	ACM (1940s)	**AU, EU**	**CO, ECO**[f] (1965)	**CR**[f] (1931)	**CSM**	**EPDM**[f] (1963)	**FKM** (1957)	**FMQ**	**SBR** (1936)	**IIR**[f] (1942)	**MQ** (1942)	**NBR** (1935)
T_R[a]	-18, 177	-54, 93	-54, 135	-54, 149	-54, 121	-54, 149	-29, 204	-73, 177	-51, 93	-54, 107	-114, 232	-54, 135
T_R[b]	-34, 204	-54, 104	-51, 135	-62, 121	----	-51, 149	-34, 260	----	-51, 121	-51, 149	-112, 288	-40, 121
T_g[c]	~ -13	----	----	~ -50	----	-60	----	----	~ -55	~ - 70	- 127	----
T_g[b]	-46 [d]	-30	-22, -52 [e]	-43	----	-60 [d]	-20 [d]	----	-64 [d]	-71	-127	-37 [d]

[a] Source: Lyons, 1982 [80]; Parker Seals, 1992 [107]. [b] Source: EPRI, 2007 [54]; [c] Source: Gent, 1992 [108]; [d] Lowest of a range indicated. [e] Values correspond to CO, ECO.
[f] Year of first commercial introduction (EPRI, 2007); other years from [9].

resistance to tear and flex cracking considering wide diversity of dynamic usage (Table 3.1) and inherent vulnerability of elastomer under low cycle fatigue at slow operation speed. A comparison of all the major generic elastomers (Table A.1) on the same footing in this context suggests exclusion of silicone rubber because of the lowest resistance to tearing and flex cracking.

With non- reinforced elastomer as the choice for INDIAN- SFR RP inflatable seals[23] and the theoretical mist compatibility studies (FFTF- CRBRP R & D) bringing out inherent limitations of this elastomer, supported by INDIAN- SFR- FBTR inflatable seal material development, silicone rubber is eliminated from the consideration of common generic elastomer.

3.9.3 Adoption of fluorocarbon rubber as the common generic elastomer

Fluoroelastomers went through various transitions(since commercial introduction as Viton® A) including exclusion suggestions by WDTRS specification(Section 3.4.2.1), acceptability status at the end of FFTF cover gas seal development (1975, Section 3.4.2.4), second most popular generic

[23] Reinforcements in SPX- 1 and EFR MQ inflatable seals compensated high permeability and low strength; Viton® inner layer in MQ inflatable seal of SNR- 300 reduced permeability by ~ 100 times.

elastomer during this period and 1980s (Section 3.4.2.1) and inclusion in the finalised specification of DFBR (as part of the qualified incident seal structure; 1994) without any reported problem from release of harmful volatile (such as HF) during testing (0.5 m and 1.5 m seal diameters) in the most demanding envisaged conditions of DFBR i.e. contact with sodium at system pressure up to 10 MPa emulating CDA conditions. This is further substantiated by the estimation of negligible HF release (Sections 3.7- 3.8) from INDIAN- SFR inflatable seal (made of S- 1) during operation using data extrapolation. Table 3.10 indicates that FKM rubber is amongst elastomers of lowest permeability as also demonstrated during the INDIAN- SFR and FBTR inflatable seal development program as per specification requirement (Table 3.4).

Accurate quantification of stress- strain by FEA could address the low hot tensile properties of FKM rubber (comparable with MQ, Table 3.15), as demonstrated by INDIAN- SFR inflatable seal development with a minimum life of 24 y for S-1 (at 120 ^{0}C, Table 3.11). Besides, there are scopes to improve upon the hot tensile properties and FOS (of 2) of S- 1 to maximise life by better grade of fluoroelastomers (S- 2) with different curing process which forms a part of this thesis. Comparison of short term high temperature range of generic elastomers in Table A.1 obtained from nuclear usage provides a very clear indication about the superiority of FKM rubber over others in terms of continuous operation at elevated temperature. High temperature ratings (Table A.1) of FKM (232- 500 ^{0}C) is way ahead of even the closest competitor, i.e. MQ (204- 288 ^{0}C), which provides a very strong indication about achievability of 1 synchronised replacement of cover gas seals in 60 y if fluoroelastomer is adopted as the common generic elastomer.

All- round advantages of fluoroelastomers is given a closer look as development and FBR operating data are relatively scarce compared to the much-used MQ rubber.

High temperature rating (> 200 ^{0}C) for continuous operation, outstanding fluid resistance at high temperature and thermal stability, ability to withstand synergistic ageing for longer span, chemical inertness and purity coupled with enhanced reliability, safety and environmental friendliness have made fluoroelastomers the first choice in critical sealing applications of automotive, air transportation, chemical processing, petroleum, power generation, semiconductor fabrication, food packaging and medical applications where other elastomers cannot perform. Gaskets, packing, hose, diaphragm, couplers, lining, coating, power and instrumentation cables are some of the many usages of these rubbers where they act mainly as barrier layers to prevent harmful release of fluids, protect substrate from corrosive environment, maintain integrity of insulations, avoid contamination of packaging content or minimize fuel wastage and environmental pollution by restricting those releases which contribute to the cause [85, 154- 155].

VDF/ HFP dipolymer with mole ratio of ~80/20 is readily curable by bisphenol and account for the largest fluoroelastomer consumption (~ 80%) out of which Viton® grades, categorized under three types or families (Viton A, B and F) based on nominal polymer fluorine content, occupy a sizeable segment. Viton® A has the minimum fluorine content (66 wt.% F) amongst the families and hence the highest CS resistance. The precompound (VDF/HFP mole ratio: ~78/22), containing high level of curative for optimum CS and medium Mooney viscosity (42 ML 1 + 10 at 121 ^{0}C), is basically designed for compression moulding of seals. One Mooney unit corresponds to a torque of 0.083 Nm (0.735 lbf.in). Viton® A-401C is suitable for conventional extrusion and autoclave cure of hose, tube and profiles because of its favourable rheology at higher shear rates, required for extrusion compared to moulding [85, 146, 156- 157].

Intuitively, S- 1 (developed for 10 y of life) could be chosen as the pilot formulation to initiate the development of large diameter backup seal material based on compound modification (for minimum developmental effort) as production process for both is extrusion and the operating requirements in the later is lower (no mist exposure, no rubbing, slightly lower temperature, Table 3.3) along with higher seal wall thickness (> 5 mm vis-à-vis 2 mm). Adapting to static usage (and lower usual hardness of 50- 60 ^{0}Shore A) requires tailoring of S-1 by reducing the filler content as the first step. The stiffer polymeric chains of FKM rubber compared to other elastomers (such as hydrocarbons) and their slower recovery on removal of load make them a natural choice for static sealing applications. Lower resilience does not pose any hindrance for dynamic use either as the rotation/ reciprocation speeds of cover gas seals are mostly very low (Table 3.1). For higher speeds (inflatable seal; ARDM seal scram at 4 m/s), either the diameters (and tolerances) are small or external actuation by inflation gas (and resilience addition by metal inserts in seals) compensates for the inadequacy in recovery at rubbing contacts due to changing tolerances (Table 3.1).

Simplification and unification by standardisation of individual modules therefore takes non-reinforced fluorocarbon rubber as the common generic elastomer across categories of cover gas seals as the binding approach.

3.9.4 **The way ahead:** Data extrapolation and minimising experiments and analysis further

Tailoring S- 1 for backup seal (S- 4) marks the beginning of experimentation in Chapter 4.

Use of bisphenol cured Viton® A-401C based compounds however has limitations in, i) Stringent demands on FEA based design because of low hot tensile properties (25-30 % of RT), ii) Possible loss of elastomeric functions and failure (> 10 y) as the initial E_b of 80% for S- 1 at 120 ^{0}C could degrade much below 100 % (typical threshold for elastomeric behaviour) with synergistic ageing, iii) FOS> 2is required to absorb additional envisaged property- dimensional deviations during manufacture in large diameter (Tables 3.1 and 3.3) and by a different process i.e. extrusion, iv) 3-5 end-joints in the largest, extruded RP (inflatable, backup, O-rings) seals because of hot feed extrusion and autoclave cure, v) Profile collapse, curing difficulties and dimensional deviations experienced during pressurised autoclave cure (typical temperature: 150–170 °C; typical pressure: 0.55–0.7 MPa) of FKM inflatable seal of INDIAN- SFR design (Fig. 3.34), vi) Taking moulding as the common (unifying) manufacturing process for the > 0.5 m (to ~ 6.3 m) diameter cover gas seals multiplies infrastructure cost and development- production- quality control- synchronisation efforts because of a large number of moulds corresponding to a large range of seal diameters (Tables 2.2, 3.1); extrusion could replaces these moulds with a range of much smaller extrusion dies corresponding to the seal cross- sections (50 mm x 50 mm, maximum) in a single extruder, and vii)

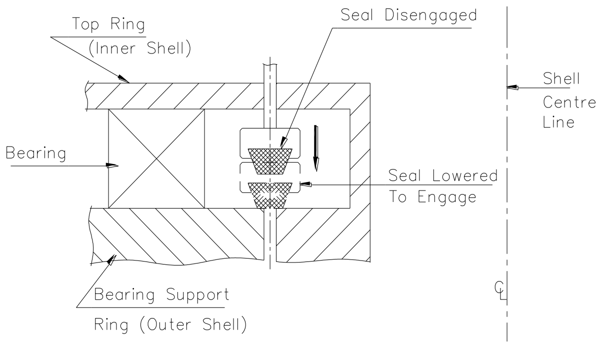


Fig. 3.44. Schematic of backup seal being lowered by suitable drive to engage with bearing support ring and top and middle ring prior to normal operating condition.

Difficulty in ensuring circumferential uniformity of temperature- cure- property in high-cost moulding of large diameter seals.

The above limitations suggesting necessity for better extrusion process and Viton® grade for better quality seals with single end- joint are addressed through other domain of this thesis based on tailoring of S-2, Chapter 4 onwards.

The entire description of Chapter 3 till now has provided most of the necessary foundations to define the problem at the end of this Chapter and proceed further which completes by including motivation, conceptualisation and R & D scheme for backup seal, depicted next.

3.10 Summary of Special Elastomeric Component R & D Results for Reference-*II* [36, 139-140]

Choice of a static, rectangular, solid rubber ring as secondary RP barrier (backup seal) to inflatable seal (RCB air side; Figs. 3.9, 3.32, 3.44) was inspired by similar arrangements in KNK and SNR-300 and rubber blocks downstream to LM freeze seals in BN-350 and BN-600 RPs, disengaged during fuel handling. The solid rectangular cross-section evolved into the present hollow, trapezoidal shape (Fig. 3.14) by FEA-based sizing of 14 geometries with the basic objective to bring down seal engagement (or assembly load, Fig. 3.44) to ≤ 10 t vis-à-vis the conceptually arrived ~ 100 t while maximising life. Figure 3.45 shows an interim solid trapezoidal cross-section arrived during conceptualisation.

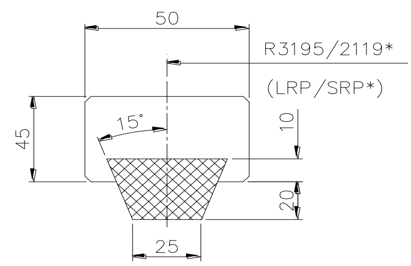


Fig. 3.45. Backup seal design arrived during conceptualization.

The assumption of bonded rubber block for assembly load estimation by stiffness calculation approach during conceptualisation emulated higher frictional contact (i.e. higher side of usual frictional coefficient range, 0.1- 1) between uncoated rubber (of backup seal) and conventionally machined (roughness: 3.2 μm) top surfaces of BSR and TMR (radial gap: 5 ± 2 mm) without radial slip or hoop stress in the sealing ring, loaded under plane strain condition. Eq. 3.1 was used to simplify calculation based on handbook where load- deflection is linearized to obtain an equivalent spring constant for the block, to be multiplied with maximum compression displacement of 6 mm (for assembly load) corresponding to a squeeze of 20 % [108].

$$K_c = A_L E_c / t \tag{3.1}$$

Where, K_c is the compression spring rate in N/ m, A_L is the load area in m^2, E_c is the effective compression modulus in N/ m^2 and t is the elastomer block thickness i.e. 0.03 m.

E_c is a function of material properties and component geometry which is calculated by Eq. 3.2 for a flat, bonded elastomeric block.

$$E_c = 1.33E_0(1 + \varphi \mathcal{B}^2) \text{ for 1- D strain} \tag{3.2}$$

Where, E_0 is Young's modulus in N/ m^2, φ is elastomer compression coefficient and $\mathcal{B}$ is the shape factor i.e. Load area (A_L)/ Bulge area (A_B).

Figure 3.46 shows ideal hyperelastic behaviour of rubber with nonlinear stress- strain relation (material non- linearity). Eqs. 3.1-3.2 overestimated the INDIAN- SFR LRP seal compressive load grossly (963 kN) by assuming linear stress- strain behaviour under large strain (35 % by FEA computations)and applying a typical Young's modulus of 4 MPa for typical fluoroelastomer (of 55 ± 5 ^{0}Shore A hardness).Simplifying and downsizing seal engagement/ disengagement drives (along with plugs) and ensuring smooth as well as synchronised engagement/ disengagement (along with seal life maximisation) were motivations for minimising the assembly load by FEA apart from eliminating trial- error based design by testing in scaled (1 m diameter) rig. The backup seal drive mechanism lowers/ lifts the seal (through parallel 36/ 30 load application points on the LRP/ SRP seal holder) by about 25 mm prior- to/ after normal operation for engagement/ disengagement. The

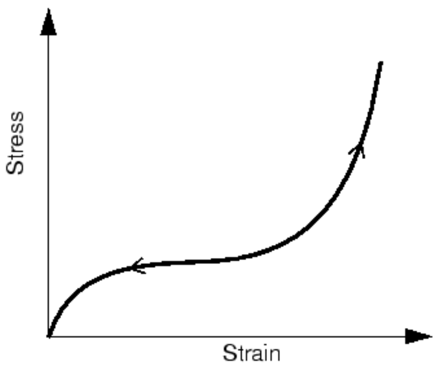


Fig. 3.46. Typical stress- strain behaviour of rubber.

drive could also compensate for compression loss (due to CS) during operation by increasing seal squeeze to enhance seal life.

3.11 Expanding Unification Beyond Fast Breeder Reactors [36]

Survey of Indian PHWRs, supported by literatures [158- 159], indicated that critical elastomeric sealing applications of FBR and PHWR could be based on the same classification scheme and unified by 8 variations of S- 1 and S- 3 (Table 3.11), some special compounds and a uniform FEA approach involving plane strain and axisymmetric conditions with Mooney-Rivlin material model. PHWR system was divided into two broad categories for the purpose viz. i) Reactor vault (including control rod drive system) and, ii) Primary heat transport system (including fuel handling machines and systems).

3.12 The First Take on Unification Scheme [36]

S- 1, S-3 (Table 3.11) and the FKM compound envisaged for backup seal (by tailoring S- 1 i.e. S- 4) cover 3 out of 8 categories. S-1 could be extended to small diameter, extruded dynamic O-ring/V-ring/lip seals (Tables 3.1-3.2) as CS of these compounds is lowest (amongst the Viton® grades) and possibility of Joule-Gough effect in the rotary O-rings does not exist because of low speed and intermittent motion.

Considerations to avoid compounds containing halogen and free sulphur in contact with polar fluids (PHWR) and SS (INDIAN- SFR IFTM inflatable seal) to avoid corrosion etc. and low hot tensile properties of fluoroelastomers suggest the necessity for other high temperature generic elastomer. High temperature (maximum:300 ^{0}C), high pressure (maximum:8.5 MPa) static sealing applications for the coolant channel seal plugs of AHWR are to operate at cumulative radiation dose of ~20 kGy/ y while in contact with water, steam, air and SS. This brings in MQ rubber and perfluoroelastomers into consideration.

The pre-conceptualised version of a unified foundation for the critical elastomeric sealing of INDIAN- SFR, FBTR, PHWR and AHWR therefore comprise of a variety of 10–12 well characterized compounds, including the special compounds. The PECVD based Teflon- like coating technique could be extended to other seals designs with minimum modification.

3.13 Defining the Problem

The problem is primarily defined as simplification of critical elastomeric sealing of MOX fuelled FBRs (represented by INDIAN- SFR) by categorising them into 9 initial representative groups (categories)and unification by taking the inflatable- backup combination as representative (of the 9 categories) and non- reinforced fluoroelastomer as the common generic elastomer where standardisation of 8 constituting elements of design completes unification with an aim of 1 synchronised replacement of the cover gas elastomeric seals in 60 y. The 8 constituting modules are, i) Material, ii) Sizing, optimisation and design by FEA, iii) Manufacture, iv) Teflon- like antifriction coating by PECVD, v) Stress- strain based life assessment by FEA, vi) CS and stress- strain based life assessment by Arrhenius and WLF methodologies, vii) Quality control, and viii) R & D.

Standardising R & D and quality control are aimed at effective future implementation of the scheme for Indian FBRs (transiting towards SFRs) in evolutionary route as the first step with minimum effort, cost and time while ensuring safety, reliability and sustainability by maximising the predictability and reproducibility of results within and across reactors.

Tailoring S- 1 to arrive at S- 4 and improvement of S- 1 with better manufacturing technology, along with FEA of inflatable and backup seals as facilitator, are pivotal in complete unification where the improved version of S-1 (i.e. S- 2) acts as the cornerstone to expand unification with the support of S- 4 and life assessment towards the objective of 1 synchronised replacement in

60 y. Usage of data extrapolation and design has been maximised to minimise experimentations as well as analysis as part of this thesis and future implementations.

3.14 Innovations, Advancement and Newness of Methodologies and Results

This research could claim for a number of innovations, advancements and newness in methodologies as well as inferences in context of results published so far, internationally.

The theme of simplification- unification- standardisation of critical elastomeric sealing, aimed at 1 synchronised replacement in 60 y for MOX fuelled Indian FBRs (and extendable to other reactor genres nationally and globally) is an innovative approach. Bringing all the critical elastomeric seals on a common foundation by maximising the exploitation of elastomer- sizing- manufacturing- coating correlations (unique for elastomers) systematically using standardisation of 8 constituting elements of design towards total unification could be identified as significant advancement.

The methodologies of data extrapolation and design adopted in this thesis for identification of representative FBR and its critical sealing, representatives of cover gas seals, common generic elastomer and estimations of HF release without any experimentation has been a departure from the conventional. Similar observations could be made on life estimation of inflatable and backup seals (made of FKM S- 1 and S-4) with minimum experimentation and analysis. Using non- reinforced elastomer as common basis is new. Standardising material module by taking the improved FKM formulation of inflatable seal (S- 2) as cornerstone and conceptual tailoring of the same, purely by data extrapolation to the entire nuclear domain (Chapters 6 and 7) and without any experimentation or analysis, has no parallel in international context. This is applicable to the standardisation and unification approaches for other 7 modules of design as well. Systematic combination of predictive tools (FEA, Arrhenius and WLF equations) for reproducible life maximisation, aided by standardised quality control, has again been a departure from the conventional as also the coating and R &D.

The FOS based methodology (on T_s and E_b) as a common thread for life assessment under synergistic ageing and mechanical loads, supported by CS, HF release and tear strength as complementary failure parameters, is unique - as also the approach of maximising life for the entire gamut of elastomeric sealing applications by choosing the cornerstone FKM elastomeric compound (S-2) of INDIAN- SFR RP inflatable seal with maximum FOS. Of particular mention should be ascertaining 10 y of life for the inflatable seal elastomer (made of S- 1) without experimentation-analysis by progressive estimation of reducing FOS with addition of synergistic ageing and mechanical load elements, which has been a common approach of design adopted in this thesis in combination with all the envisaged variabilities of reactor conditions. This novel combination of data extrapolation and design has also been used as a tool for cross- verifications of experimental and analysis findings which is in consonance with the Gen IV philosophy.

Identifying high temperature elastomer as the common thread by combining available R& D results has been the key to the whole process, as also the identification of peroxide cured, 50:50 blend of Viton® GBL 200S: 600S made of APA (S- 2) as the originating inflatable seal elastomeric formulation for unification through production trials in a commercial extruder and data extrapolation.

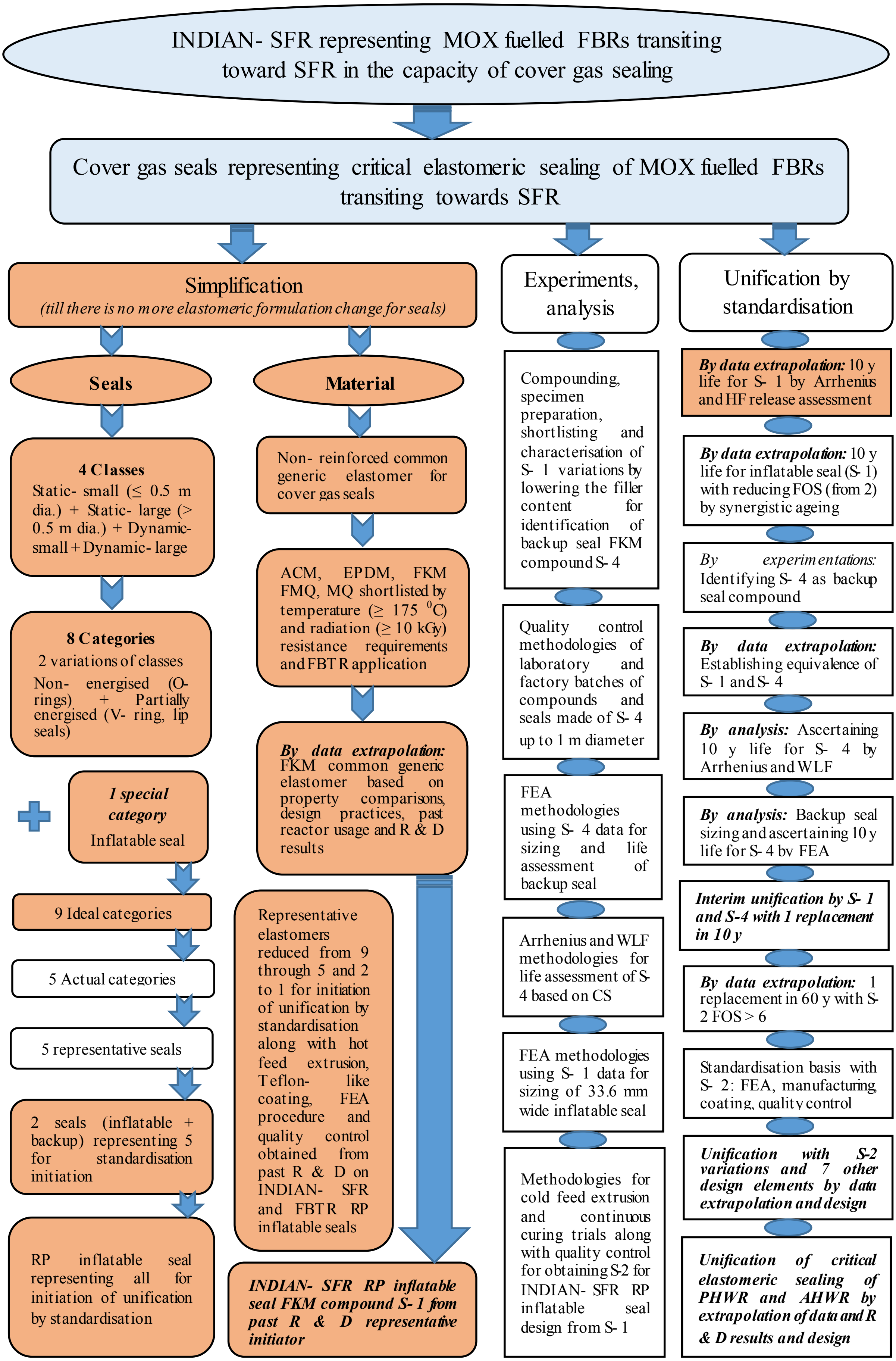
The rest of this thesis is about establishing simplification, unification and standardisation by standardising the 8 modules using experiments, analysis, data extrapolation and design.

3.16 Chapter Conclusion

Cover gas seals of INDIAN- SFR were simplified into 9 ideal categories (*towards implementation of 1 synchronised replacement in 60 y by future R & D in the most safe, reliable, economic and sustainable way*) based on operation, size, energisation level and independence of CS based failure (*inflatable seal as special category*) for unification by standardisation. RP inflatable and backup seals, installed in INDIAN- SFR for the past 6 y and withstanding precommissioning tests, are taken as representative of static and dynamic categories due to most demanding operating requirements. This eventually resulted in choice of inflatable seal FKM formulation (S- 1) as the initiator of unification by standardisation of each of the 8 design elements based on heuristic and literature substantiation approach because of the closest proximity of the seal's design requirements to the representative conditions of TS elastomeric sealing in INDIAN- SFR. The process was parallely supported by material simplification including various elastomer- failure- criteria (T_s and E_b at ET divided by FOS of 2, CS, HF release etc.) for reactor seals which identified FKM rubber as the common generic elastomer for cover gas seals, first by shortlisting the generic elastomers based on high temperature (≥ 175 ^{0}C) and minimum radiation resistance (≥ 10 kGy) and then by taking into considerations past reactor usages as well as R & D along with closer view on property comparisons. R & D results on FKM/EPDM inflatable seals of INDIAN- SFR/ FBTR, carried out as part of *Special Elastomeric Component* development at IGCAR involving more than 15 Indian agencies prior to this research, was summarised which provided an outline of standardisation modes for various design elements i.e. material (S- 1/ S- 3), sizing by FEA using common conditions (plane-strain and axisymmetric), inflatable seal production by hot feed extrusion and autoclave cure, Teflon-like coating by PECVD, quality control and a preliminary R & D framework for effective and efficient future implementations of unification scheme, supported by completion of basic inflatable seal developments within 5y/ 3 y vis-a- vis international span of more than a decade on average. The process of material simplification was identified by ascertaining 10 y life for S- 1 based on Arrhenius methodology (T_s, E_B based cracking failure) and estimation of HF release under operating temperature of seal in reactor. The first take on unification scheme involving S- 1, S- 2 and their variations highlighted feasibility of approach which indicated than PHWR and AHWR critical elastomeric sealing could be similarly unified along with those of INDIAN- SFR (i.e. covering entire spectrum of Indian nuclear genres) based on 10- 12 well characterised compounds. Innovations, advancements, newness of methodologies as well as results were indicated and the problem thus defined indicated that unification should initiate with identification of FKM compound for the INDIAN- SFR RP backup seal by tailoring S- 1 for static usage as the first step.

Figure 3.47 shows the progress of research carried out as part of this dissertation till end of Chapter 3 which encompasses the entire simplification process comprising of seals and material.

Fig.3.47. Outline of research methodologies: Progress in Chapter 3



Chapter 4

Experiment and Analysis Methodologies

4.1 Representative Materials [81, 87, 126]

Bisphenol cured, Viton® A-401C based FKM compound (S- 4) used for static backup seals (~ 6.4/ 4.2 m diameter, withstanding pre- commissioning conditions in INDIAN- SFR LRP/SRP for past 6 years) and 50: 50 blend composition of peroxide cured Viton® GBL 200S:600S (S- 2) used in ~ 2 m diameter, test dynamic inflatable seals of INDIAN- SFR RP design are chosen as representatives.

S- 4 is used as representative for quality control demonstration as this completed full circle from laboratory to reactor scale. S- 2 is chosen as pivotal to material unification and also a representative for manufacture as this is amenable to cold feed extrusion and continuous cure. S- 4, in addition, supports material unification where its behaviour is extrapolated to the pivotal blend compound with minimum experimentation. Compositions and characteristics of peroxide cured, Nordel IP 4520 based EPDM for FBTR inflatable seals (S- 3, Table 3.11) and Bisphenol cured Viton® A-401C based FKM for INDIAN- SFR inflatable seals (S- 1, Table 3.11) are referred excepting the cold feed extrusion and continuous curing trials with S- 1 which forms an important part of experimental methodologies. The transition from S- 1 to S-2 provides a complimentary quality control base (for large diameter, dynamic extruded seals) to that of S- 4, used for large diameter static extruded seal.

4.2 Complementary Roles of Experiment and Analysis

Experimental methodologies are applicable to the following modules.

I) Representative materials as cornerstone.

II) FEA based sizing, optimisation and design as facilitator.

III) Manufacture of > 0.5 m diameter seals by cold feed extrusion and continuous cure.

IV) PECVD Teflon- like coating for dynamic seals.

V) Quality control from laboratory specimen development to reactor installation stage.

VI) FEA based life assessment as facilitator.

VII) Arrhenius and WLF equation based life assessment.

Analysis methodologies are applicable to FEA and Arrhenius as well as WLF relation- based-predictions using unaged and long- term accelerated aged material properties.

4.3 Common Experimental Methodologies

4.3.1 FKM: Viton® A-401C based compounds [81, 87, 140, 145]

4.3.1.1 Overview

Tests are classified as,

I) Shortlisting of backup seal laboratory candidate FKM compounds (IG1- IG4) by processability studies (on uncured or unvulcanized compound stock, Table 4.1) and short term mechanical tests (on cured compound or vulcanizate; Table 4.1) to assess curing characteristics and rheological (or viscoelastic flow) behaviour (for manufacturability) as well as conformance to target properties (Table 4.2) respectively.

Table 4.1. Testing scheme for compound shortlisting

Candidate Compounds: IG1- IG4		
Processability characterization of stock	Mechanical characterization of vulcanizate	
	ASTM Die C *(Unaged specimen)*	**ASTM Die C** *(Aged: 150 °C/70 h)*
M_L	T_s	T_s
M_H	E_b	E_b
$t_s 2$	Hardness	Hardness
$t_s 10$		CS
t' 50		
t' 90		

Note:

- ASTM dies are applicable to specimens for T_s and E_b only, determined at crosshead speed of 500 mm/ min
- Vulcanizate properties determined at RT

II) Detailed evaluation (E- I) of shortlisted laboratory compound (IG3 or S- 4) for quality and its improvement by blending of IG3 batches (B1- B8) to a representative laboratory batch, IG3-A (Table 4.3). Ascertaining that processability and mechanical properties (determined at RT) have been translated effectively from the initially shortlisted laboratory IG3 batch to laboratory IG3- A.E-1 was aimed at producing benchmark material data for subsequent factory productions of test and reactor seals (Table 4.3) by emulating capacity of industrial scale large mixing equipment (i.e. two

Table 4.2. Target mechanical properties of INDIAN- SFR rotatable plug backup seal and its compound

Properties (Measured at RT)	**Vulcanizate** (Specimens from moulded test slabs; ASTM D3182-89)	**Extruded seal** (Specimens from seal; ASTM D3183- 02)	**Applicable Standards**
Measured on unaged specimens (ASTM Die C) at RT		Measured at RT	
T_s(MPa)	6 (minimum)	Within ±10% of the corresponding values of unaged vulcanizate	ASTM D412 – 98a (Reapproved 2002) e1
E_b (%)	150 (minimum)		
Hardness (^{0}Shore A)	50 ± 5		ASTM D 2240 – 02b
Splice Strength (MPa)	Not applicable	6 (minimum)	ASTM D2527 - 83 (2001)
Limit of % change of properties determined on heat aged specimens (150^{0}C/ 70h)at RT			
CS	10% (maximum)	Not applicable	ASTM D395 - 03
With respect to corresponding values of unaged specimens: T_s-15% (max.); E_b-15% (max.); Hardness- 10% (max.)		Not applicable	ASTM D573 - 99

Table 4.3. Quality control testing scheme for laboratory and factory batches of fluorohydrocarbon rubber compound (IG3 or S- 4) and test as well as INDIAN- SFR backup seals made of them

Laboratory Batch		**Factory Batches**							
STOCK		VULCANIZATE							
IG3- A (E- I)		**10a**				**10b**		**10c**	
Specimens from uncured blended compound	Specimens from cured blend compound	Specimens from cured compound, 200 mm- 250 mm long test seal piece				Specimens from 1 m dia. test seal		Specimens from cured compound, INDIAN- SFR seals	
		Compound			**Seal**	**Seal**		**Compound**	**Seal**
	ASTM Die C [a] Dumb- bell	ASTM Die C [a]	ASTM Die C *(150 ^{0}C/ 70h)*	ISO Die 2[a] Dumb- bell	ISO Die 2[a] Dumb- bell	ISO Die 2[a]	ISO Die 2 *(150 ^{0}C/ 70h)*	ISO Die 2[a]	ISO Die 2[a]
M_L	T_s	T_s	T_s	T_s	T_s	T_s	T_s	T_s	T_s
M_H	E_b	E_b	E_b	E_b	E_b	E_b	E_b	E_b	E_b
t_s 2	Hardness	Hardness	Hardness	Hardness	Hardness	Hardness	Hardness	Hardness	Hardness
t_s 10	CS	Relative density			Relative density	Relative density			
t' 50	TGA				TGA				TGA
t' 90	FT- IR					FT- IR			

- [a] Corresponds to unaged specimens (T_s, E_b, hardness)
- ASTM and ISO dies are applicable to specimens for T_s and E_b only, determined at crosshead speed of 500 mm/ min
- 150 ^{0}C/ 70h thermo- oxidative ageing applicable to T_s, E_b and hardness only
- CS corresponds to 150 ^{0}C/ 72h and 150 ^{0}C/ 168h thermo- oxidative ageing
- All measurements carried out at RT excepting TGA

roll mill) with blending of several small laboratory batches (B1- B8). Consistency was also required in properties and behaviours across small lots/batches (B1- B8) of stock specimens (used for

Table 4.4. Testing scheme for extrudability

Batch: IG3- A (E- II)	
STOCK SPECIMENS	
Experiments	Extrusion rate
	Die Swell
	Garvey Die rating

Table 4.5. Testing scheme for backup seal sizing and design by finite element analysis

Batch: IG3- A (E- III)			
VULCANISED SPECIMENS, UNAGED			
All properties determined at 110 ^{0}C and cross- head speed of 50 mm/ min			
Compression properties		**Tensile properties**	
Cylindrical moulded specimens		ASTM (Die C) Dumb-bell	
With stress softening [a]	Without stress softening	With stress softening [c]	Without stress softening
Stress- strain [b]	Stress- strain [b]	Stress- strain [d]	Stress- strain [d]
---	---	T_s	T_s
---	---	E_b	E_b

[a] Stress cycling of specimen in strain range of 0-20 %
[b] Stress determined at 1 % strain interval up to 40 % strain
[c] Stress cycling of specimen in strain range of 0- 50 %
[d] Stress determined at 1 % and 5 % strain intervals till failure

extrusion trials) and a large number of vulcanized specimens (used for long term accelerated ageing i.e. 32 weeks) to maximize prediction reliability.

III) Detailed evaluation (E- II) of IG3- A for extrudability (Table 4.4).

IV) Detailed evaluation (E- III) of IG3- A for sizing by FEA (Table 4.5).

V) Detailed evaluation (E- IV) with 32 weeks accelerated thermooxidative ageing of tensile and CS specimens made of IG3- A at three elevated temperatures (140 ^{0}C, 170 ^{0}C, 200 ^{0}C) for life prediction of INDIAN- SFR backup seal by FEA and Arrhenius as well as WLF methodologies (Table 4.6).

VI) Additional detailed evaluation (E- V) of unaged and long- term (32 weeks) aged tensile and compression specimens for comprehensive mechanical characterization of IG3- A (Table 4.7).

VII) Quality assessment of extra lengths of extruded backup seal profile (250- 300 mm length and 1 m dia. test seal) made of

Table 4.6. Testing scheme for backup seal life assessment and design

Batch: IG3- A (E- IV)		
VULCANIZED SPECIMENS, AGED		
Tests carried out on specimens aged for 32 weeks at 140 ^{0}C, 170 ^{0}C and 200 ^{0}C		
Specimen withdrawal intervals	**CS**	**Tensile properties** [a]
	Moulded disc specimens	ASTM Die C Dumb- bell
1 week	CS with respect to 25 % initial deflection measured at RT	Stress- strain, T_s and E_b measured at 110 ^{0}C without stress softening at crosshead speed of 500 mm/ min
2 weeks		
4 weeks		
8 weeks		
16 weeks		
32 weeks		

[a] Stress determined at 1 % and 5 % strain intervals till failure

Table 4.7. Additional testing scheme for comprehensive mechanical characterization of backup seal material

<table>
<tr><th colspan="7">Batch: IG3- A (E- V)</th></tr>
<tr><th colspan="4">AGED SPECIMENS</th><th colspan="3">UNAGED SPECIMENS</th></tr>
<tr><td colspan="4">Ageing for 32 weeks at 140 ^{0}C, 170 ^{0}C and 200 ^{0}C</td><td colspan="2">Tensile properties [a]</td><td>Compression properties [b]</td></tr>
<tr><th rowspan="2">Specimen withdrawal intervals</th><th colspan="2">Tensile properties [a]</th><th rowspan="2">Hardness</th><td colspan="2">ASTM Die C</td><td>Cylindrical moulded specimens</td></tr>
<tr><td colspan="2">ASTM Die C Dumb- bell</td><td rowspan="7">Stress- strain, T_s, E_b measured at RT with and without stress softening at crosshead speed of 50 mm/ min</td><td rowspan="7">Stress- strain, T_s, E_b measured at 110 ^{0}C without stress softening at crosshead speed of 500 mm/ min</td><td rowspan="7">Stress- strain measured at RT with and without stress softening at crosshead speed of 50 mm/ min</td></tr>
<tr><td>1 week</td><td rowspan="6">Stress- strain, T_s, E_b measured at RT without stress softening at crosshead speed of 500 mm/ min</td><td rowspan="6">M_{50}, M_{100} measured at RT, 110 ^{0}C without stress softening at crosshead speed of 500 mm/ min</td><td rowspan="6">Hardness measured on periodically withdrawn aged specimens at RT</td></tr>
<tr><td>2 weeks</td></tr>
<tr><td>4 weeks</td></tr>
<tr><td>8 weeks</td></tr>
<tr><td>16 weeks</td></tr>
<tr><td>32 weeks</td></tr>
</table>

[a] Stress determined at 1 % and 5 % strain intervals till failure; Stress cycling of specimen in strain range of 0 - 50 %

[b] Stress determined at 1 % strain interval up to 40 % strain. Stress cycling of specimen in strain range of 0 - 20 %

two different factory batches (10a and 10b) of IG3 by short- term mechanical tests and material fingerprinting (Table 4.3).

VIII) Quality assessment of INDIAN- SFR LRP and SRP inflatable seals made of another factory batch (10c) of IG3 or S- 4 (Table 4.3).

4.3.1.2 Candidate compounds

Four modified versions (Table 4.8) of the initial INDIAN- SFR RP inflatable seal compound (DMF3 or S- 1) with lower MT black content and varying level of rubber grade $Ca(OH)_2$ were chosen for studies. MgO was kept unchanged (3 phr) in the candidates to ensure maximum CS resistance. Proportion of MT black was varied to obtain minimum CS and meet the strength as well as hardness requirements of static backup seal (Table 4.2).

Table 4.8. Compositions of candidate compounds

Ingredients	Candidate compounds			
	IG1	IG2	IG3	IG4
Viton® A-401C(phr)	100	100	100	100
MT black N990(phr)	3	2	2	1
MgO(phr)	3	3	3	3
$Ca(OH)_2$(phr)	6	6	3	3
Batch weight	112	111	108	107

4.3.1.3 Compound mixing

Viton® A- 401C, obtained from DuPont Dow Elastomers (USA), was mixed with MT black, high activity MgO of small particle size (or larger surface area per unit volume) and rubber grade $Ca(OH)_2$ with small particle size (activator) in a standard internal mixer (laboratory Banbury;

Stewart Bolling) of 1.5 x 10^{-3} m^3 (1.5 L) capacity to produce the compounds in accordance with ASTM D3182 and as per specific processing requirements of Viton®. Rotor speed, ram pressure and mixing duration of 30 rpm, 0.7 MPa and 3.5 min were employed to produce green compound stock.

Table 4.9. Processability results of candidate compounds

Properties	IG1	IG2	IG3	IG4
Stock properties				
M_L (lbf.in/ N.m)	0.80/0.09	0.78/ 0.09	0.69// 0.08	0.69/ 0.08
M_H (lbf.in/ N.m)	11.67/ 1.32	11.35/ 1.28	10.75/ 1.21	10.53/ 1.19
t_s **2** (min)	1.56	1.67	2.18	2.40
t_s **10** (min)	1.48	1.59	2.03	2.23
t' 50 (min)	1.69	1.81	2.40	2.62
t' 90 (min)	2.13	2.25	3.10	3.31

4.3.1.4 Curing mechanism

Details of curing mechanism not provided so far, are given here. Bisphenol curing system requires activation by water. Ca $(OH)_2$ as activator releases water at curing temperature (moulding: 160- 190 ^{0}C; autoclave: 150- 170 ^{0}C). The nucleophilic curing system, involving a cross-linker or curative and an accelerator, is still used in about three- fourth of all FKM applications. Activator determines the rate of formation and extent of unsaturation/ double- bonds/ cure- sites in the green or unvulcanised compound stick. Accelerator level decides the rate at which cure sites are cross-linked. Proportion of curative largely determines the number of cross- links or the state of cure.

4.3.1.5 Processability: Characterization of viscoelastic flow and curing

Processability results of IG1- IG4 are given in Table 4.9.

Eight different batches (B1- B8) of IG3 were mixed in order to have sufficient compound for long-term heat ageing and extrudability tests which were cross-blended subsequently. Improved consistency of curing characteristics and viscoelastic flow behaviour of the blend (batch identification: IG3- A) are reflected by lower standard deviations after blending (Table 4.10).

Measurement of curing characteristics and rheology (viscoelastic flow behaviour) were carried out on circular, unvulcanized specimens (3- 5 cm^3 volume) from IG3- A in a rotorless, oscillating die cure meter (rotorless cure meter or moving die rheometer; MDR 2000E, Alpha Technologies) as per ASTM D5289. Reference test temperature, test time, die oscillation amplitude and die oscillation frequency of 177 ^{0}C, 10 min, ± 5 ^{0} arc and 100 cycle per minute (cpm) were used in all the measurements. Maximum torque (M_H – corresponding to maximum state of cure), minimum torque (M_L – corresponding to minimum state of cure), scorch time (t_s2 – time for increase of 2 lbf.in of torque above M_L or time-lag for curing inception) and t'$_x$ (time to x% of torque increase) were determined.

4.3.1.6 Specimen preparation: Relative density

Smooth test pieces (arbitrary shape) without crevices weighing ~ 3 g each were prepared for relative density measurements as per International Organisation for Standardisation (ISO) 2781 Method A.

4.3.1.7 Specimen preparation: Stress- strain and hardness

Table 4.10.[a] Improved consistency of processability of stock by cross- blending of IG3 (S-4) batches

Processability Parameters	Characteristics of IG3 prior to cross- blending across batches									
	B1	B2	B3	B4	B5	B6	B7	B8	Average	Std. Deviation
M_L (lbf.in/ N.m)	0.80/ 0/09	0.80/ 0.09	0.83/ 0.09	0.81/0.0 9	0.83/ 0.09	0.83/ 0.09	0.82/ 0.09	0.81/ 0.09	0.82/ 0/09	0.01
M_H (lbf.in/ N.m)	10.88/ 1.22	11.09/ 1.25	11.34/ 1.28	11.16/ 1.26	11.44/ 1.29	11.30/ 1.28	11.27/ 1.27	11.15/ 1.26	11.20/ 1.27	0.17
t_s 2 (min)	2.47	2.52	2.40	2.36	2.24	2.32	2.15	2.49	2.37	0.13
t_s 10 (min)	2.23	2.29	2.19	2.14	2.06	2.10	1.96	2.26	2.15	0.11
t' 50 (min)	2.76	2.82	2.70	2.64	2.51	2.60	2.43	2.78	2.66	0.14
t' 90 (min)	3.57	3.60	3.43	3.32	3.21	3.33	3.12	3.51	3.39	0.17
Characteristics of blended batch (IG3- A) stock across randomly chosen samples										
M_L (lbf.in/ N.m)	0.82/ 0.09	0.80/ 0.09	0.80/ 0.09	0.80/ 0.09	0.81/ 0.09	0.80/ 0.09	0.81/ 0.09	0.81/ 0.09	0.81/ 0.09	0.01
M_H (lbf.in/ N.m)	11.25/ 1.27	11.06/ 1.25	11.12/ 1.26	11.12/ 1.26	11.14/ 1.26	11.07/ 1.25	11.08/ 1.25	10.88/ 1.23	11.09/ 1.25	0.10
t_s 2 (min)	2.29	2.23	2.26	2.25	2.24	2.29	2.22	2.22	2.25	0.03
t_s 10 (min)	2.09	2.05	2.06	2.04	2.06	2.09	2.03	2.02	2.06	0.03
t' 50 (min)	2.56	2.50	2.52	2.49	2.51	2.55	2.48	2.47	2.51	0.03
t' 90 (min)	3.22	3.17	3.18	3.15	3.16	3.24	3.30	3.07	3.19	0.07

[a] Torque measured by test apparatus in lbf.in has been retained (while including conversions in SI unit) for the sake of proper comparisons as the t_s 2 values were also determined based on 2 lbf.in rise in torque above M_L. 1.00 lbf.in = 1.13 dN.m

Dumb- bell tensile specimens were stamped from 150 mm x 150 mm x 2 mm compression moulded test sheets (slabs) by ASTM Die C and ISO Type 2 Die. Specimens were prepared from seals as per ASTM D3183 by cutting samples with a splitting machine, careful buffing of sample surfaces and stamping dumb-bells from the samples by ISO Type 2 Die. Usage of smaller ISO Type 2 Die was necessitated by smaller width of backup seal which did not allow the wider ASTM Die C.

Smooth test sheets were compression moulded in an electrically heated press for 5 min at 177 ^{0}C and 15 MPa pressure (press-cure) as per ASTM D3182 and were oven post- cured subsequently for 24 h at 232 ^{0}C in accordance with Viton® processing guidelines. Typical press-curing conditions (177 ^{0}C/ 10 min) were modified to obtain better E_b at RT (without affecting T_s) so that elastomeric behaviour at ET is ensured by keeping E_b as close to the threshold value of 100 % as possible.

Cylindrical moulded specimens for compression stress- strain were prepared in accordance with ISO 7743.

Test pieces for ^{0}Shore A hardness measurements were prepared from compounds and extracted from seals as per ASTM D2240 and ISO 7619-1.

4.3.1.8 Specimen preparation: Compression set

Disc specimens of 29 ± 0.5 mm diameter and 12.5 ± 0.5 mm height were moulded in an electrically heated press and step post- cured (starting from about 100 ^{0}C) in air oven for 24 h using a maximum temperature of 232 ^{0}C because of higher cross-sectional thickness (> 5 mm).

Moulding (or press-cure) duration was changed from 20 min (at 150 ^{0}C) for IG1 test pieces to 35 min (at 150 ^{0}C) for those made of IG2- IG4 following development of crack in the former during short term CS tests (70 h at 150 ^{0}C) for compound shortlisting. CS specimens made of IG3-A were moulded for 35 min at 150 ^{0}C [45].

4.3.1.9 Specimen preparation: Accelerated heat ageing

Tensile, hardness and CS test pieces (IG3- A) for long term accelerated ageing were prepared as per Sections 4.3.1.7 and 4.3.1.8.

4.3.1.10 Specimen preparation: Extrudability

Stock specimens of ~ 1 kg each were randomly selected from IG3- A to carry out trials for extrusion and die- swell in a Brabender Plasticorder.

4.3.1.11 Specimen preparation: Thermal analysis

Specimens of ~ 6 mg each, weighed by vertical balance, were taken from 150 mm x 150 mm x 2 mm test sheets (IG3- A) and extra length of backup seal extrudate (10a, 10c) to generate TG thermograms for material fingerprinting.

4.3.1.12 Specimen preparation: Infrared spectroscopy

~ 20 g of sample, extracted from test sheets (IG3- A) and extra length of 1 m diameter test seal extrudate (10b) and weighed in analytical balance, was pyrolysed at 550 ^{0}C. Pyrolysed fume was condensed and collected in a vial, mixed with solvent and vacuum dried (2- 3 drops) on a salt plate as thin film for material fingerprinting by Fourier Transform Infrared Spectroscopy (FT- IR) spectra [45].

4.3.2 **Fluorohydrocarbon rubber:** Blends of Viton® GBL 200S and Viton® GBL 600S [81]

4.3.2.1 Overview

Unlike IG3- A, Comp # 9 (S- 2) made of 50: 50 blend of Viton® GBL 200S and Viton® GBL 600S was obtained by factory compounding and production trials of INDIAN- SFR inflatable seal (in cold feed extruder with continuous cure), beginning with a factory batch of DMF3 (S- 1) i.e. Comp #1. One candidate compound was used at a time and only on finding its limitations in extruding the seal profile (extrudate), the next one was chosen till Comp #9 produced conformances to seal cross-sectional dimensions (Fig 4.1) and material properties shown in Table 4.11.

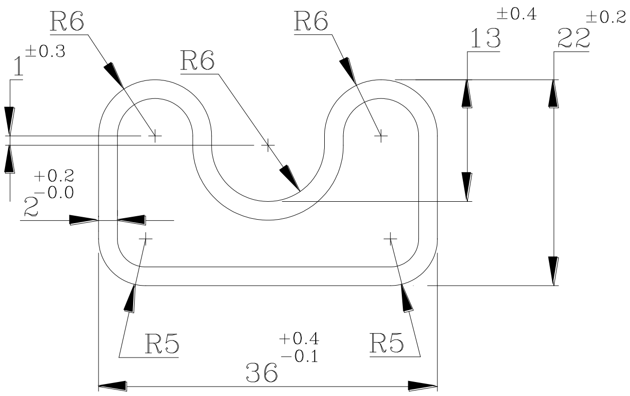


Fig. 4.1. Earlier cross- sectional dimension of INDIAN- SFR rotatable plug inflatable seal.

4.3.2.2 Candidate compounds

Table 4.11. Target properties of INDIAN- SFR rotatable plug inflatable seal and compound

Properties (Measured at RT)	**Targets: Rubber Compound** (Specimens from test slabs)	**Targets: Extruded Seal** (Specimens from seal; ASTM D3183-02)	**Applicable Standards**
Determined on unaged specimens			
T_s(MPa)	11.5 (min.)	Within ±10% of the values determined on rubber compound	ASTM D412 – 98a (Reapproved 2002) e1
E_b (%)	150 (min.)		
Hardness (^{0}Shore A)	70 ± 5		ASTM D 2240 - 05
Splice Strength (MPa)	---	10.5 (min.)	ASTM D2527 - 83 (2001)
Tear Resistance(N/mm)	18 (min.)	Within ±10%	ASTM D624 - 00e1
Determined on aged specimens			
CS 150^{0}C/168 h in air	25% (max.)	---	ASTM D395 - 03
Dry Heat Resistance 150^{0}C/336 h in air	Ts, Hardness and Eb within ±10% of the values determined on unaged specimens	---	ASTM D573 - 04

Factory batch of RP inflatable seal compound, DMF3 (Comp #1 or S- 1, Table 4.12), and its 5 modified versions (Comp #2- #6), 1 formulation based on Viton® GBL 600S (Comp #7) and 2 more based on 50: 50 blend of Viton® GBL 600S and Viton® GBL 200S (Comp. #8- #9) were studied during compounding- extrusion trials. In addition, 5 test compounds (Comp TC #1- #5) made of typical standard commercial formulations were used (Table 4.13) as benchmarks at the beginning of trials (TC # 1) and before every paradigm- shift in composition and curing (Comp #5, #7 and #8) to attune and set up factory equipment and procedures for subsequent trials. Table 4.12 shows formulations and mechanical properties of laboratory batch of DMF3 (S- 1or Comp# 1) obtained from earlier R & D and used as reference.

Reduction of filler in comp. #1 (20 phr of MT black) compared to TC #1 is reflected in decrease of modulus, T_s and hardness and an increase in E_b compared to the typical formulation (Tables 4.12 and 4.13). Filler content was reduced to attain the hardness value specified in Table 4.11 and also to increase the E_b, determined at 120 ^{0}C, nearest to 100 %. Comp. #1 was refined to Comp. #2 with addition of 1 phr of processing aid (carnauba wax) to reduce compound viscosity and improve extrusion flow as well as extrudate smoothness. Comp. #2 was further refined to comp. #3 with addition of 3 phr of CaO in order to improve strength. The content of MT black (N990) was reduced to 12 phr in the next refinement (Comp. #4) for reduced compound viscosity and improved flow. Comp #5 and Comp #6 were based on Viton® A- 201C to arrive at optimized viscosity and strength in order to meet specification (Table 4.11), while retaining amenability to production by cold feed extrusion and continuous cure.

The paradigm- shift to Comp #7 based on peroxide cured Viton® GBL 600S made of APA was intended to remove porosities, dimensional deviations, lower strength and coarseness of profiles primarily from release of volatiles during extrusion and inability of pressureless cure to address the same. The shift to 50:50 blend formulations of Viton® GBL-600S:200S (Comp #8) was aimed at reducing backpressure and die swell (by reducing the compound viscosity) in order to improve dimensional deviations and lack of finish along with complete removal of stickiness in profile. Comp. #8 was modified to comp. #9 (S- 2) by replacing 2 phr curing agent (Varox DBPH-50) with 1 phr of Luperco® 101-XL to ensure total removal of stickiness in extrudate.

Table 4.12. Formulation and properties of DMF3 (S- 1)

Proportion of ingredients
Viton® A-401C: 100 phr; MT Black N990: 20 phr; High Activity MgO: 3 phr; $Ca(OH)_2$: 6 phr; Batch Weight: 129.
Stock Properties (ASTM D5289-95)
MDR 2000 at 177⁰C, 0.5⁰ arc, 100cpm, 15 min clock
M_L: 1.29 dN.m; M_N: 22.77 dN.m; t_s2: 1.59 min; t' 90: 2.62 min; Mooney viscosity: 108 ML (1+4)100⁰C; MS@135⁰C : 39.39 dN.m
Vulcanizate Properties (ASTM D3182-89; ASTM D412-98a; ASTM D2240-02b; ASTM D 624-00e1; ASTM D573-99; ASTM D395-02)
Determined on unaged specimens at RT (press cure at 177⁰C, 10 min; post cure at 232⁰C, 24 h)
M_{100}: 4.8 MPa; T_b: 12.28 MPa; E_b: 217%; Tear strength: 31 N/mm; Hardness: 68 ⁰Shore A.
Determined on unaged specimens at 100⁰C and 120⁰C
T_b: 3.82 MPa, E_b: 98% (at 100⁰C); T_b: 3.26 MPa, E_b: 80% (at 120⁰C);
Determined on aged (336h at 150⁰C in air) specimens at RT
M_{100}: 4.8 MPa; T_b: 12.6 MPa; E_b: 209%; Hardness: 70 ⁰Shore A.
Determined on aged (336h at 150⁰C in air) specimens at 100⁰C and 120⁰C
T_b: 4.2 MPa, E_b: 97% (at 100⁰C); T_b: 3.5 MPa, E_b: 82% (at 120⁰C).
Determined on aged (336h at 200⁰C in air) specimens at RT
M_{100}: 4.9 MPa; T_b: 11.6 MPa; E_b: 191%; Hardness: 72 ⁰Shore A.
Determined on aged (336h at 200⁰C in air) specimens at 100⁰C and 120⁰C
T_b: 4.2 MPa, E_b: 93% (at 100⁰C); T_b: 3.1 MPa, E_b: 72% (at 120⁰C).
Determined on aged specimens at RT (Press cure, 30min at 150⁰C; post cure 24h at 180⁰C)
CS, 72h at 150⁰C: 9%; CS, 168h at 150⁰C: 14%

4.3.2.3 Compound mixing [81]

Viton® A- 401C, Viton® A- 201C, Viton® GBL 200S, Viton® GBL 600S ('G' for peroxide cure; 'L' for improved low temperature performance; 'S' for APA) were obtained from DuPont Dow

Table 4.13. Outline of progressive modifications of Viton® A-401C based formulation (Comp. #1 or S- 1) to 50: 50 tailored blend of Viton® GBL-200S and 600S (Comp. #9 or S- 2)

Compound ingredients	Typical Viton® A-401C (TC # I)	Comp. #1	Comp. #2	Comp. #3	Comp. #4	Typical Viton® A-201C (TC # II)	Comp. #5	Comp. #6	Typical Viton® GBL-600S (TC # III)	Comp. #7	Typical Viton® GBL-200S (TC # IV)	Typical 50/50 blend (TC # V)	Comp. #8	Comp. #9
Viton® A-401C(phr)	100	100	100	100	100	-	-	-	-	-	-	-	-	-
Viton® A-201C(phr)	-	-	-	-	-	100	100	100	-	-	-	-	-	-
Viton® GBL-200S(phr)	-	-	-	-	-	-	-	-	-	-	100	50	50	50
Viton® GBL-600S(phr)	-	-	-	-	-	-	-	-	100	100	-	50	50	50
MT black N 990(phr)	30	20	20	20	12	30	20	20	30	-	30	30	-	-
SRF black N 762(phr)	-	-	-	-	-	-	-	-	-	25	-	-	25	25
High activity MgO(phr)	3	3	3	3	3	3	3	3	-	-	-	-	-	-
Rubber grade $Ca(OH)_2$(phr)	6	6	6	6	6	6	6	6	-	-	-	-	-	-
CaO(phr)	-	-	-	3	3	-	-	3	-	-	-	-	-	-
High purity ZnO(phr)	-	-	-	-	-	-	-	-	3	2	3	3	2	2
Varox® DBPH-50(phr)	-	-	-	-	-	-	-	-	2	2	2	2	2	-
Luperco® 101-XL(phr)	-	-	-	-	-	-	-	-	-	-	-	-	-	1
Diak™ No. 7(phr)	-	-	-	-	-	-	-	-	3	2.5	3	3	2.5	2.5
Struktol® WS280(phr)	-	-	-	-	-	-	-	-	-	0.75	-	-	0.75	0.75
Carnauba wax(phr)	-	-	1	1	1	-	1.25	1.25	-	-	-	-	-	-
Vpa No. 2(phr)	-	-	-	-	-	-	-	-	-	0.75	-	-	0.75	0.75

Elastomers (USA). Viton® A- 201C is a dipolymer of VDF and HFP with nominal fluorine content of 66 wt.%, Mooney viscosity about half of that of Viton A- 401C and physical properties similar to Viton A- 401C. Viton GBL-600S (68 wt. % F; Mooney viscosity: 65 ML 1 + 10 @ 121^0 C) and GBL 200S (68 wt. % F; Mooney viscosity: 25 ML 1 + 10 @ 121^0 C) are copolymers of VDF, HFP and TFE ($CF_2 == CF_2$) with Iodine based cure site monomers in polymer chains to allow peroxide curing.

Unlike the Bisphenol cured grades, Viton® GBL 600S and 200S are not supplied as precompound along with accelerator and curative. This provides laboratory or on- site flexibility for better tailoring of compound to the parametric requirements of production system and extrudate property demands from the end use. At the same time, reproducibility of curing response and hence the dimensions as well as properties of extrudate is less assured which is compounded by about double the number of compounding ingredients compared to the Viton® A-401C or Viton® A-201C based formulations (Tables 4.8 and 4.13).

Laboratory scale batches of fluoroelastomer compounds based on Viton® A-401C (Comp #1- #4) and Viton® A-201C (Comp #5- #6) were mixed using standard laboratory equipment (internal mixer, two roll mill) as per ASTM D3182- 89 (Reapproved 2001) and adopting procedures similar to Section 4.3.1.3. Mixing steps used for comp. #9 (as given next) during the production of ~ 2m diameter test inflatable seals of INDIAN- SFR design are representative of those for Comp #7- # 8, i.e. peroxide cured fluoroelastomers made of APA. i) Premix 1: carbon black (33%) with DIAK™ 7, ii) Premix 2: rest of carbon black and other chemicals, iii) Banding of rubber in mill, iv) Adding Premix 2, proper cross-cutting and blending, v) Adding Premix 1, proper blending, vi) Cutting at 10–15mm thickness, vii) Mixing time: 30 min, viii) Bowl temperature: 12–16 ^{0}C to be maintained, ix) Stock temperature at the end of mixing: 85–90 ^{0}C, x) Keeping the stock for 16 h (maturing), xi) Mixing of stock through nip gap (2–3 mm), xii) Strip cutting at desired thickness, xiii) Maturation for another 16 h and, xiv) Use for extrusion.

4.3.2.4 Curing mechanism

For Comp #1- #6 (Table 4.13), this is same as those given in Section 4.3.1.4 and Chapter 3.

Peroxide cross-linking works through the principle of free radical mechanism where the reaction is activated by an organic peroxide that decomposes (homolytic cleavage) thermally to form alkoxy radicals during cure. The alkoxy radicals operate on special cure sites, incorporated in the form of cure site monomers having Bromine and/or Iodine substitutions during polymerization of fluoroelastomer, to displace Bromine and/or Iodine in adjacent polymeric chains which combine to form carbon–carbon bonds. Peroxide or by-products generated during vulcanization are not part of cross- links and hence do not contribute towards reducing either the strength or the inherent stability of cured rubber. Number of potential cross- linking sites is finite and is limited by the quantity of cure site monomers incorporated during polymerization. Excess curatives remain unused after the available cure site monomers are consumed. As a result, vulcanized product from peroxide curable fluoroelastomer contains much lower level of double bonds or unsaturation.

Peroxide cross- linking of FKM requires a curing agent such as Triallyl Isocynurate(TAIC) known by the trade name DiakTM 7. TAIC is considered to be the most effective curing agent as judged by the cure state, thermal stability and CS resistance of vulcanizate. TAIC is also a cross- linking promoter (prorad) which enhances cross-linking in FKM under ionizing radiation. In contrast, contents of metal oxides and hydroxides used in typical bisphenol cured FKM formulations such as TC#I- #II (Table 4.13) and their variations (i.e. Comp #1- #6; Table 4.13) are usually more than what is needed to carry out the basic functions as acid acceptor and activator respectively. As a result of excess Ca $(OH)_2$, the number of double bonds generated during cure is in excess of what is required for cross-linking. Cured parts carry these unsaturation and extra inorganic bases (metal oxides and hydroxides) during service.

Table 4.14. Processability characteristics of Comp. #9 (S- 2)

Stock Properties (ASTM D5289-95; ASTM D1646-03a)
MDR 2000 at 177^{0}C, 0.5^0 arc, 100cpm, 10 min clock
M_L: 1.1 dN.m; M_H: 30.4 dN.m; t_s2: 0.6 min; t' 90: 1.6 min; Mooney viscosity: 46 ML 1+10 @ 121^{0}C

4.3.2.5 Processability: Characterization of viscoelastic flow and curing

Rheological properties (Mooney Viscosity, torque) of uncured stocks made of TC#1 - #5 and Comp #1- #9 (Table 4.13) were measured per ASTM D1646-03a and ASTM D5289-95 (Reapproved

2001) using Mooney viscometer (MV 2000) and moving die rheometer (MDR 2000) with specimens similar to those indicated in Section 4.3.1.5. Processability characteristics of Comp #9 (S- 2) are indicated in Table 4.14.

4.3.2.6 Specimen preparation

Specimens for relative density, stress- strain, hardness, CS and short term accelerated heat ageing were prepared using equipment and procedures similar to those indicated in Sections 4.3.1.6-4.3.1.9 and conforming to standards indicated in Table 4.11.

4.4 Experimental Methodologies: *Compound shortlisting* [87]

4.4.1 Fluorohydrocarbon rubber: Viton® A-401C based formulations

All tests pertain to specimens made of IG1- IG4, carried out at HASETRI, Rajasthan.

4.4.1.1 Short term accelerated heat ageing

Short term heat ageing studies of tensile and hardness specimens in air (thermo- oxidative ageing) were carried out (Table 4.1) in rack type ageing ovens (for 70 h at 150 ^{0}C ±3 ^{0}C) equipped with thermocouples in accordance with ASTM D573- 99 and ISO 188: 1998.

4.4.1.2 Stress- strain and hardness

Uniaxial tensile stress–strain measurements were made on ASTM dumbbell specimens (unaged and short- term heat aged without stress- softening) at RT with crosshead speed (strain rate) of 500 mm/ min (Table 4.1) using universal testing machine (universal testing machine or UTM; Zwick-1445) equipped with optical extensometer and oven. Tests were conducted in accordance with ASTM D 412and ISO 37 to determine T_s, E_b and M_x (x% modulus or stress at x% strain).

^{0} Shore A hardness was measured at RT on unaged and short term aged specimens (Table 4.1) as per ASTM D2240and ISO 7619-1.

4.4.1.3 Compression set

CS was measured at RT on moulded disc specimens (exposed to short term accelerated ageing for 70 h at 150 ^{0}C; Table 4.1) in a quick clamping apparatus (Prolific Engineers, India) as per

ASTM D395 Method B (constant deflection in air). The apparatus comprises of four rigid metal plates with 9.5 mm thick spacer bars placed in between them. The plates are tightened centrally by bolts to impart the necessary compression on the specimens.

Original thickness of specimens was measured at RT to the nearest of 0.02 mm as per standard. The specimens were inserted into apparatus and compressed to the required strain at RT. Subsequently the assembled apparatus was placed in air-oven (within 2 h of completion of assembly) for thermo- oxidative ageing. Test specimens were removed from apparatus (for cooling) immediately after its withdrawal from oven following the specified test periods. The specimens were kept on surfaces with poor thermal conductivity and cooled for at least 30 min (at laboratory temperature of 23 ± 2 ^{0}C) before measuring the final specimen thickness at ambient temperature.

CS expressed as percentage of original deflection (under a compression of 25%) was calculated using the following equation from ASTM D395.

$$C_B = [(t_0 - t_i)/(t_0 - t_n)] \times 100 \tag{4.1}$$

Where, C_B is the CS (%), t_0 is the initial specimen thickness, t_i is the final thickness and t_n is the spacer bar thickness.

4.5 Experimental Methodologies: *Detailed characterization for finite element analysis and life assessment* [87, 145]

4.5.1 Fluorohydrocarbon rubber: Viton® A-401C based formulations

All measurements pertain to specimens made of IG3-A, carried out at HASETRI, Rajasthan.

4.5.1.1 Long term accelerated ageing: Tensile stress- strain and hardness

Long term accelerated heat ageing of tensile and hardness specimens in air (thermooxidative ageing) were carried out in rack type ageing ovens (± 3 ^{0}C) equipped with thermocouples. Ageing spanned 32 weeks at three elevated temperatures (140 ^{0}C, 170 ^{0}C and 200 ^{0}C) with specimens withdrawn at intervals of 1 week, 2 weeks, 4 weeks, 8 weeks, 16 weeks and 32 weeks. Dry air was circulated in the 45 x 10 $^{-3}$ m^3 (45 L) capacity ovens at a rate of 50 x 10 $^{-3}$ m^3/h (50 L/h) with 8- 10 complete air changes per hour during the entire ageing span. Ageing was carried out in accordance with the applicable provisions of ASTM D573- 99, ISO 188: 1998and ISO 11346: 2004.

4.5.1.2 Long term accelerated ageing: Compression set

Compressed disc specimens in quick clamping apparatus (Section 4.4.1.3) were aged in air ovens at three temperatures up to 32 weeks with withdrawals as indicated in Section 4.5.1.1.

4.5.1.3 Stress- strain for seal sizing by finite element analysis

Lower crosshead speed (50 mm/ min) and ET (110 ^{0}C) measurements with and without deformation cycling (stress softening) at 50% strain were applied on unaged ASTM tensile specimens to extract stress- strain data and determine T_s and E_b for seal sizing by FEA using equipment and standards indicated in Section 4.4.1.2.

Mechanical conditioning and lower testing speed were adopted for FEA vis-à-vis those for compound shortlisting (Section 4.4.1.2) in order to ensure isotropy and homogeneity in specimens by allowing relaxation of macromolecular chains with lower speed and breaking of temporary, secondary, polymer- filler bonds with stress cycling- also called Mullins effect, Fig. 4.2 [160]. This does also emulate a few cycles of slow compression- decompression and inflation- deflation of backup and inflatable seals in reactor for the same purpose which imparts repeatability and predictability in performance behaviour apart from making the specimen stress- strain data amenable to modelling by strain energy density function (W) or elastomer constitutive relation for FEA to ensure realistic performance prediction.

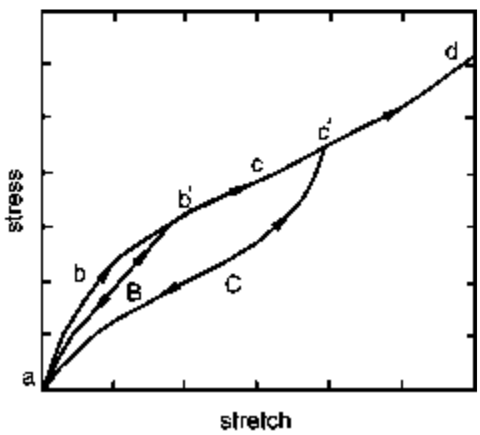


Fig. 4.2. Reduction in stiffness of stress-strain curve of rubber towards a stabilised response due to Mullins effect on repeated loading and unloading or stress cycling.

Compression stress–strain tests were carried out with and without stress softening in UTM, Zwick-1445, at 110 ^{0}C on unaged cylindrical (diameter: 29 ± 0.5 mm; thickness: 12.5 ± 0.5 mm) moulded specimens at a strain rate of 50 mm/ min in accordance with ISO 7743.

Mechanical conditioning of tensile specimens was carried out at RT by cyclic deformation of the specimens for 8 cycles in the range of 0– 50% strain at a strain rate of 50 mm/ min. A pause of 2 min was given after each cycle. The specimens were kept for 30 min after completion of 8 cycles and before proceeding for stress-strain measurements at 110 ^{0}C. Uniaxial tensile stress was determined at elongation intervals of 1 % and 5 % in the ranges of 1–12 % and 15 %- E_b respectively.

Conditioning of compression specimens differs in terms of elongation range (0- 20 %) and relaxation time (15 min) prior to final measurements. Stresses were measured at 1 % strain interval throughout. Compression strain range was increased to 40% later because of the absence of frictional effects between cylindrical test piece and lubricated platen in the UTM.

The combination of uniaxial tensile and uniaxial compressive stress- strain data input to W for determination of material or calibration constants (and to construct stiffness matrix for FEA) in effect represents triaxial state of stress in seals during operation as equibiaxial tension in an incompressible or isochoric continuum (such as rubber) is equivalent to uniaxial compression with addition of suitable hydrostatic stress imparting zero volumetric strain, Fig. 4.3 [161- 162]. For a nearly incompressible elastomer (such as IG3- A) this becomes an engineering approximation using FEA which transforms 1- D specimen behaviour with material non- linearity to 3- D response of product involving geometric and contact non- linearity with changing load and boundary conditions.

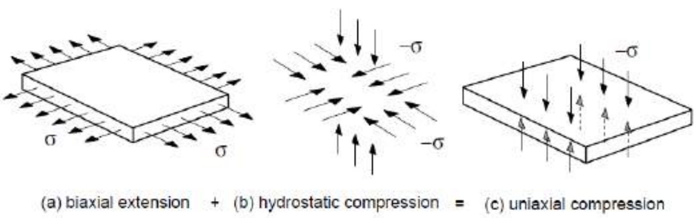


Fig. 4.3. Equivalence of uniaxial compression and biaxial tension.

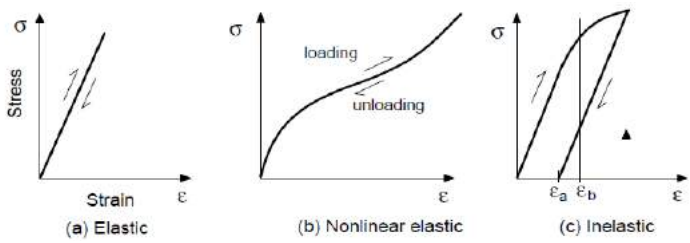


Fig. 4.4. Various types of stress-strain responses of materials.

Figure 4.4b shows material nonlinearity or nonlinear stress-strain behaviour [161].

Sizing of seal by FEA is based on comparing the principal and von Mises stress- strain in unaged seal with unaged specimen T_s and E_b (both determined at 110 ^{0}C) divided by a FOS of 2, where the later defines limiting values to avoid cracking failure. At the same time, contact pressure between seal and mating surface should be more than the differential fluid pressure across the seal as first criterion to prevent leakage.

4.5.1.4 Stress- strain for life assessment by finite element analysis

Uniaxial tensile stress- strain was measured at 110 ^{0}C at crosshead speed of 500 mm/ min on ASTM specimens (without stress- softening) exposed to thermo- oxidative ageing at three temperatures for 32 weeks using same equipment, procedures and standards as in Sections 4.4.1.2 and 4.5.1.3. Elongation interval for stress extraction was identical with those of Section 4.5.1.3.

The utility of test data for realistic prediction of seal performance and failure without using mechanical conditioning and with higher crosshead speed were enabled by negligible effects of deformation cycling and test speed on stress- strain behaviour because of increased mobility of polymeric chains at elevated temperature and low reinforcing filler (MT Black N 990) content (2 phr) in IG3- A which minimized effects of relaxation and roles of additional secondary physical (reversible) forces between macromolecules from polymer- filler interactions respectively.

4.5.1.5 Stress- strain for life assessment by Arrhenius and William Landel Ferry equations

Stress- strain data from aged specimens determined in accordance with Section 4.5.1.4.has dual utility of life prediction by FEA and Arrhenius as well as WLF equations in complementary fashion. The later maps actual time span to arrive at a specific level of degradation at ET (i.e. 110 ^{0}C for backup seal) with those corresponding to accelerated heat ageing temperatures (i.e. 140 ^{0}C, 170 ^{0}C and 200 ^{0}C). In essence, while FEA quantifies stress- strain and contact pressure in seal with accelerated aged specimen data, the Arrhenius and WLF equations find out equivalent time span in reactor corresponding to the accelerated ageing period.

The Arrhenius methodology captures degradation by chemical (or irreversible) mechanism alone, represented by activation energy of degradation ($\bar{E}$). The WLF relation encompasses physical or reversible changes in addition, such as those from strain which enhances polymer- filler- ingredient interactions to create additional secondary (temporary) bonds between macromolecules which could disappear (or break) with withdrawal of load, increased temperature or stress cycling.

4.5.1.6 Compression set for life assessment by Arrhenius and William Landel Ferry equations

CS was measured at RT (with equipment, procedures and standards outlined in Section 4.4.1.3) on disc specimens prepared as per Section 4.3.1.8 and accelerated aged (for 32 weeks) as well as withdrawn from air oven (for measurements) at intervals of 1 week, 2 weeks, 4 weeks, 8 weeks and 32 weeks as per Sections 4.4.1.3 and 4.5.1.2.

4.5.1.7 Additional tests for comprehensive characterization

Predicting backup seal life is a competing process of principal stress (and strain), contact pressure and CS reaching limiting values (CS: 80- 100 %) where life is determined by the shortest time span which defines inception of failure (Chapter 5). The necessity of integrating life prediction results from FEA, Arrhenius and WLF methodologies to the effectiveness of seal design under reactor conditions require support of comprehensive characterisation by additional tests on S- 4.

Uniaxial tensile stress- strain (with and without stress softening) was measured at RT on unaged specimens at crosshead speed of 50 mm/ min as per equipment, procedures and standards depicted in Section 4.5.1.3. These measurements were repeated at RT on periodically withdrawn, long- term (32 weeks) accelerated aged specimens (without stress softening) at crosshead speed of 500 mm/ min as per Section 4.5.1.4. In addition, unaged specimens were tested at 110 ^{0}C and crosshead speed of 500 mm/ min (without stress softening) as per Section 4.5.1.4 to provide reference for ageing degradation and bring in comparability with test results from crosshead speed of 50 mm/ min. M_{50} and M_{100} were determined at RT and 110 ^{0}C at crosshead speed of 500 mm/ min on unaged and periodically withdrawn long- term (32 weeks) aged specimens during measurements of uniaxial tensile stress- strain as per Section 4.5.1.4 to complete the picture.

Compression stress- strain tests were carried out at RT (crosshead speed: 50 mm/ min) as per 4.5.1.3 with and without stress softening of specimens prepared as per Section 4.3.1.7.

^{0} Shore A hardness was measured at RT (as per Section 4.4.1.2) on unaged and periodically withdrawn long- term (32 weeks) accelerated aged specimens (Section 4.5.1.1) prepared in accordance with Section 4.3.1.7 from IG3- A.

One of the prime objectives of additional tests for comprehensive mechanical characterization of IG3- A was to identify the usable tensile strength range of the bisphenol cured Viton® A- 401C based formulation for design at elevated seal operating temperature, attributed to covalent bonds between polymeric chains and restoring forces from transverse thermal vibrations of elastomeric macromolecules.

4.6 Experiments: *Manufacture*

4.6.1 Fluorohydrocarbon rubber: *Viton® A-401C Based Formulations* [87]

All measurements pertain to specimens made of IG3- *A,* carried out at HASETRI, Rajasthan.

4.6.1.1 Extrudability

Tests for extrudability (die swell, extrusion rate and Garvey die rating; Table 4.4) were carried out in a Brabender Plasticorder (PL 2000) equipped with a temperature control unit, extrusion head, 20 mm diameter screw and take-away conveyor using randomly selected unvulcanized blended stock. Garvey die rating was determined as per ASTM D2230.

Brabender Plasticorder is similar to commercial extruders in many ways and simulates the shear rates of factory extrusion (100– 1000 s^{-1}) more practically compared to the Mooney viscometer or rotorless cure meter (typical shear rate: 1 s^{-1}). Extrusion trials on stock specimen for Garvey die rating were carried out in the Brabender Plasticorder using die, barrel and feed-zone temperatures of 130 ^{0}C, 75 ^{0}C and 50 ^{0}C respectively with screw speed of 5 rpm during measurements. Extrusion die swell of the compound was measured through another set of trials which employed extrusion rate of 31.11 g/ min at die, barrel and feed-zone temperatures of 120 ^{0}C, 75 ^{0}C and 50 ^{0}C respectively (screw speed: 15 rpm) during measurements.

4.6.2 Fluorohydrocarbon rubber: Blends of Viton® GBL 200S and Viton® GBL 600S [81]

All tests were carried out at M/s ASP Sealing Products Ltd., Gajraula, UP, India.

4.6.2.1 Compounding trials

Tensile specimens for short- term mechanical properties without and with short- term thermo-oxidative ageing (without stress softening), indicated in the first column of Table 4.11, were die-cut (ASTM Die C) from moulded slabs (150 mm x 150 mm x 2 mm). Parameters such as proportion of compounding ingredients, order of mixing, temperature and duration of mixing and maturation were varied. Standard moulded slabs (ASTM D3182) were prepared from mixed batches with various levels of press and post cure. Temperatures and durations used for press-cure were 177 ^{0}C, 190 ^{0}C and 204 ^{0}C at 3 min, 5 min, 7 min, 10 min and 12 min (Table 4.15). Slabs were post-cured at 232 ^{0}C for 2 h, 4 h and 24 h. Measured properties were compared with targets (Table 4.11) to finalize proportion of ingredients and mixing procedure for extrusion trials. Standards used for measurement of mechanical properties, ageing characteristics and CS during the trials are indicated in Table 4.11.

Equipment and procedures adopted for property determination were similar to Sections 4.4.1.1 to 4.4.1.3 with stress- strain, T_s, E_b measured at RT and crosshead speed of 500 mm/ min at the beginning of trials (Comp #1) or during paradigm shift (Comp # 5, Comp #7 and Comp #8; Table 4.13). Stock and vulcanizate specimens for TC#1- #5 were prepared as per Viton® product literature guidelines and rheological as well as mechanical properties were compared with those reported in literature (corresponding to laboratory batch) to attune factory equipment and procedures. Subsequent departures to Comp #1- #9 (as applicable) involved variations of all parameters till the target properties for rubber compound (Table 4.11, second column) were achieved.

Table 4.15. Mechanical properties of Comp. #8 under various curing conditions

Curing Temp. (^{0}C)	Sample Set	Curing Duration (min)	Properties Measured at RT		
			T_s (MPa)	Hardness (^{0}Shore A)	E_b(%)
177	Set-I	3	12.1	72	643
		5	14.3	72	599
		7	14.4	73	557
		10	12.7	73	490
190	Set-II	3	11.9	70	468
		5	12.5	71	442
		7	13.5	72	477
		10	13.8	73	463

4.6.2.2 Extrusion trials

<u>*Equipment:*</u>

Continuous, pressure-less (atmospheric pressure) vulcanization was made possible (excepting fluoroelastomers) by vacuum extruder technology which minimises porosity in rubber vulcanizate (or extrudate) by extensive plasticisation of the green elastomer and evacuation of moisture as well as plasticizers. Steadily improving functionality of ordinary cold feed extruders during 1980s and1990s, coupled with advancements in compounding, however made it feasible to process many elastomeric formulations by pressure-less continuous cure without the use of vented extruder by the first decade of new millennium. Plasticizers are rare in fluoroelastomers. Minimisation of water vapour release by peroxide curing indicates that fluoroelastomers could be processed by ordinary cold feed extruders with improved compounding and appropriate process control [163- 166].

An ordinary cold feed extruder with continuous curing system (without vacuum degassing, gear pump, real- time feedback), equipped with basic features and good temperature control, was adopted for extrusion trials of INDIAN- SFR RP inflatable seals based on available spare capacity in Indian industry. Screw diameter, screw rotational speed, extrusion capacity, motor power and screw length to diameter ratio (L/D) were 90mm, 0–45 rpm, ~ 300 kg/h, ~35 kW and 12:1. The MW heating oven and HAT were capable of operating at 2.45 GHz and 300 ^{0}C continuously. Total line length of the system, comprising of the extruder, oven and tunnel was about 35 m.

Feasibility of processing fluoroelastomers in an ordinary cold feed extruder was however accompanied by additional demands from thin seal wall, large surface area to volume ratio and close tolerance requirements. Contradictory requirements of slow screw speed (for low shear heating, ensuring dimensional conformance in slower relaxing fluoroelastomers) resulting in higher viscosity and backpressure (compounded by additional backpressure from thin seal profile) had to be balanced for production of inflatable seal to the required dimensions (Fig. 4.1) and properties (Table 4.11).

Experimentations:

Standard moulded slabs with different levels of cure, prepared using the finalized compound and mixing procedure (conforming to Table 4.11), were run through the MW oven and HAT at different line speeds and under different HAT temperatures. Mechanical properties were measured to narrow down choice and range of process parameters to be tried for seal production. Line (conveyer) speed and HAT temperature ranges used during specimen and seal production trials were 0.5–7 m/min and 180–270 ^{0}C respectively.

Table 4.16. Design features of cold feed and vented extruders

Screw Diameter(mm)	**60**	**90**	**120**	**150**	**200**	**250**
Cold Feed Extruder						
Screw Speed (rpm; max.)	60/ 96	45/ 70	37/ 60	40	35	30
L/D Ratio	10: 1	12: 1	14: 1	16: 1	18: 1	18: 1
Motor Power (kW)	13.5 /19	32/ 44	55/ 75	130	270	425
Output (kg/h; max.)	75-150	335-500	550-870	930-1400	1500-2300	2250-3400
Vented Extruder						
Screw Speed (rpm, max.)	95	70	59	51	34	--
L/D Ratio	16: 1	16: 1	18: 1	20: 1	22: 1	--
Motor Power (kW)	19	44	75	140	250	--
Output (kg/h; max.)	50-90	150-240	265-450	440-700	800-1250	--

Dimensional checks of seal profiles were carried out as per ASTM D3767-03. The usually lower screw speeds (10- 25 rpm, Table 4.16) adopted for Viton® had to be reduced further (1- 15 rpm) in order to provide adequate time for the green exrudate profile (coming out of the extruder die) to cure sufficiently for the required properties without post-cure.

4.7 Experiments: *Quality control*

4.7.1 Fluorohydrocarbon rubber: Viton®A-401C based formulations [87]

All measurements (carried out at HASETRI, Rajasthan) pertain to laboratory blended batch IG3- A and factory batched 10a- c.

4.7.1.1 Relative density

Relative density of compound batches IG3- A, 10a and 10b were measured at RT with the same equipment and using specimens, standards and procedures as per Section 4.3.1.6.

4.7.1.2 Stress- strain and hardness

Stress- strain along with T_s, E_b and M_x were measured at RT with crosshead speed of 500 mm/ min on specimens stamped or extracted by ASTM Die C and ISO Die 2 using same equipment and as per standards as well as procedures described in Sections 4.3.1.7 and 4.4.1.2.

ASTM Die C specimens were stamped from test slabs (Section 4.3.1.7) made of compound batches IG3- A and 10a. ISO Die 2 specimens were stamped from test slabs (Section 4.3.1.7) made of IG3- A, 10 a and 10c. ISO Die 2 specimens were extracted (Section 4.3.1.7) from 200 mm – 250 mm long test pieces (10a) and extra length of extrudate for 1 m diameter test seal (10 b) as well as LRP and SRP backup seals (10c) of INDIAN- SFR.

4.7.1.3 Thermal analysis

Thermogravimetric analysis (TGA)was carried out with specimens (Section 4.3.1.11) taken from test slabs (IG3- A), 200 mm- 250 mm long test seal piece (10a) and extra length of LRP and SRP backup seal extrudate (10c).

TG thermograms were generated by a thermo gravimetric analyser (Perkin-Elmer, TGA7) equipped with automatic gas switcher in the temperature range of 80- 850^0 C with a constant heating rate of 40^0 C min^{-1} according to ASTM D6370. Sample of ~ 6 mg weight was heated in N_2 (80^0 C to ~ 600^0 C) and O_2 (~ 600^0 C to 850^0 C) with constant gas purge rate of 20 $mLmin^{-1}$ to determine the amount of fluoroelastomer, carbon black and ash.

4.7.1.4 Infrared spectroscopy

Infrared spectra were generated on specimens (Section 4.3.1.12) taken from test sheets made of IG3- A and extra length of 1 m diameter test backup seal extrudate (10b).

FT- IR spectra of samples prepared by pyrolysis from compounds and seals were taken at RT using a spectrophotometer (Perkin-Elmer, Systems 2000). Specimens were scanned from 4000 to 400 cm^{-1} with resolution of 4 cm^{-1}.

4.7.2 **Fluorohydrocarbon rubber:** Blends of Viton® GBL 200S and Viton® GBL 600S [81]

Usage of same set of equipment, procedures and standards (Table 4.11) for testing of specimens (moulded piece, stamping of slabs and extraction from seal) for short- term mechanical properties without and with short- term thermo-oxidative ageing (Sections 4.6.2.1 and 4.6.2.2) are indicative of intra- laboratory quality control framework in factory production context.

4.8 **Analysis:** Finite element analysis of inflatable and backup seals for sizing, optimisation, design and life assessment [87, 144]

The relevance of maximising isotropy and homogeneity in elastomeric products during extrusion (for performance predictability with reproducibility) by ensuring better uniformity of processing conditions (pressure, temperature, shear rate etc.) across sealing dimensions and minimum defects are of similar relevance to the sizing of seals by FEA using a strain energy density function or W where isothermal deformation[24]under constant pressure generates stress [108, 118- 119, 167- 168]. The amenability of all the elastomeric ring seals of cover gas (*without liquid lubrication for safety*) to analysis by using plane strain and axisymmetric conditions are common threads of standardisation *(similar to taking S- 2 as the standardising cornerstone for material which extrapolates to other modules)* as also the constitutive relation which relates stress and strain. The depiction of methodologies for FEA of inflatable and backup seals in ABAQUS takes a route through infinitesimal- finite- hyperelasticity.

Descriptions with and without tensors has been used in parallel for continuity through the transitions from 1- D specimens to the product. One of the prime objectives for the methodologies is to minimise specimen testing for FEA by choosing a proper constitutive relation. FEA of inflatable and backup seals (made of S- 1 and S- 4) as part of this thesis required specimen stress- strain data (determined at ET) as inputs to ABAQUS as against usage of any 2 out of 4 material constants

[24] Slow deformation such as compression or inflation of backup or inflatable seals and slower strain rate of 50 mm/ min or 5 mm/ min during FEA data generation on uniaxial dumb- bell or biaxial elastomeric specimens.

(Young's modulus, shear modulus, Poisson's ratio and bulk modulus) for common structural materials (such as steel), readily available in handbooks [44].

4.8.1 A note about nomenclature

Same symbol (S) has been used for engineering stress, stress under infinitesimal strain and the corresponding tensor components of stress (S_{ij}) for elastic, linear, isotropic, Hookean solid (infinitesimal deformation or strain). The tensor shear strains (ε_{ij}, $i \neq j$) are half of the corresponding engineering shear strains (ϵ_{xy}, ϵ_{yz}, ϵ_{zx} or in terms of γ corresponding to shear stress of τ), with the former equivalent to the Lagrangian (E_{ij}) and Eulerian (e_{ij}) measures of strain because of infinitesimal strain or displacement. The usage of E_{ij} (*deformation described with respect to a system of coordinates in the undeformed state, Appendix B*) and e_{ij} (*similarly with respect to the deformed state, Appendix B*) are therefore confined to finite elasticity and hyperelasticity respectively where the original cross-sections under finite strain vary significantly to bring in notable difference between true (or Cauchy) stress (σ) and engineering stress (S). The tensor components of Lagrangian (E_{ij}) and Eulerian (e_{ij}) strains in shear ($i \neq j$) are similarly one half of the corresponding components of finite strains expressed in terms of É and é [168- 170].

4.8.2 Infinitesimal, finite and hyperelasticity

Stresses for the hyperelastic elastomers are derived from the principle of virtual work using potential stored (or strain) energy density function instead of calculating directly from strain which is the approach for small strain, linear elastic materials [168].

Engineering materials such as crystalline metals are typically classified as linear, elastic, Hookean solid (Fig. 4.4a) under infinitesimal strain. The non- linear relation of stress and strain (material nonlinearity) shown in Figure 3.47 (or 4.4b) is typical of rubber with the ability to stretch large (geometric nonlinearity from changing load application points and boundary conditions) while remaining elastic [161].

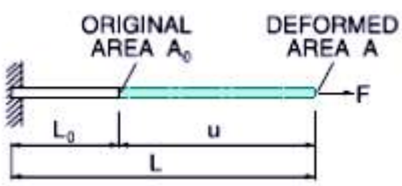


Fig. 4.5. Stretching of a rod.

Small strains in metals, adequately characterised by engineering strain, is generally defined in uniaxial loading as the percentage change in original length [161, 171- 172].

$$\text{Є} = \varepsilon = \frac{\Delta L}{L_0} \times 100 \tag{4.2}$$

Where Є, ε, L_0 and ΔL are engineering strain, corresponding tensor counterpart (as a zero order tensor or scaler), original length and change in length respectively (Fig. 4.5).

Infinitesimal strain theory is a limiting case of finite strain where true strain is one of the conjugate energy measures, employed for capturing large strains in rubber [168- 170, 173].

$$\text{é} = e = \ln\frac{L}{L_0} \tag{4.3}$$

Where é, e, L (L + ΔL or L + u) and L_0 are logarithmic (or true) strain, corresponding Eulerian tensor counterpart, deformed length and original length respectively.

True strain (Eq. 4.3) takes into account changes in original length (Eq. 4.2, Fig. 4.6). The familiar engineering (Biot) stress and the true (Cauchy) stress (as energetically conjugate measure for true strain) for infinitesimal and finite strain in 1- D could be written as [171],

$$\text{S} = \frac{\text{Ƒ}}{A_0} \tag{4.4}$$

$$\sigma = \frac{\text{Ƒ}}{\text{A}} \tag{4.5}$$

Where S, σ, Ƒ, A_0 and A are Biot or engineering stress, Cauchy or true stress as tensor of zero rank, force, original area and deformed area respectively (Fig. 4.5).

Stress- strain pairs in engineering or true domains are complimentary. Transition to finite strain or elasticity is often marked by the introduction of a deformation gradient [161, 170].

$$F_{i,A} = \frac{\partial x_i}{\partial X_A} \tag{4.6}$$

Where $F_{i,A}$ is the deformation gradient tensor and x_i as well as X_A refer to deformed and original coordinates of the body being deformed (Appendix B).

It is customary to analyse elastomers using a stretch ratio (λ)for convenience due to the magnitude of deformation [161, 171].

$$\lambda = \frac{L}{L_0} = \frac{(L_0+u)}{L_0} \tag{4.7}$$

Where λ, L_0, L and u are stretch ratio, initial gauge length, deformed gauge length and deformation respectively (Fig. 4.5).

In large deformation analysis of nonlinear materials as a 3- D solid in *Cartesian Coordinate (X- Y- Z)*, stretch ratios could be used to define strain invariants (I_j), used in many strain energy density functions. A particular point P (Fig. B.1, Appendix B) could be widely separated from its initial location during large deformation which necessitates definition of strains and stresses in two different ways i.e. referencing them to the initially undeformed and presently deformed configurations by Lagrangian and Eulerian descriptions respectively which merge for infinitesimal strains. The strain tensor for a 3- D Hookean solid could be defined as [169- 170],

$$\varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \tag{4.8}$$

Where ε_{ij} is strains and u_i and u_j are displacements, replaced by the Lagrangian finite strain tensor (E_{AB}) and the Eulerian finite strain tensor (e_{ij}) in finite elasticity theory (Appendix B).

Cauchy formalised the definitions of stress and strain in a continuum at mechanical equilibrium under body and surface forces.

"The assertion that there is defined upon any imagined closed surface ʃ in the interior of a continuum a stress vector field whose action on the material occupying the space interior to ʃ is equipollent to the action of the exterior material upon it, is the stress principle of Euler and Cauchy" [169]

$$\boldsymbol{T}^{\eta} = \frac{d\mathfrak{f}}{d\mathfrak{ʃ}} \tag{4.9}$$

$\Delta\boldsymbol{f}$ is a part of total force (exerted by the material exterior to ʃ and belonging to the continuum) on part of the total closed surface (i.e. Δʃ) and the ratio of the two tends to a definite

limit $d\boldsymbol{f}/d\int$ (i.e. $\boldsymbol{T}^{\eta}$ or traction or the stress vector on a limiting surface with normal $\boldsymbol{\eta}$) as $\Delta\int$ approaches zero (with the assumption that the external moment or couple stress vanishes to zero in the limiting process) at zero body moment because of equilibrium [172].

The stretch ratio is defined in terms of original and current relative lengths.

$$\frac{1}{\lambda} = \frac{dX}{dx} \tag{4.10}$$

Where dX and dx are original and current relative lengths (Appendix B).

At zero deformation, λ is 1 whereas E and e are zero. Stress measures in finite elasticity centres around Cauchy or true stress, defined in deformed configuration.

The usual design procedure is to measure strain or stretch ratio under load and correlate them to stress using appropriate constitutive relation- one of the simplest being Hooke's law which relates stress- strain under uniaxial deformation as,

$$\mathrm{S} = \sigma = E_0 \epsilon = E_0 \varepsilon \tag{4.11}$$

Where σ, S, E_0, ϵ and ε are Cauchy or true stress, engineering stress, Young's modulus, engineering strain and the corresponding tensor (of rank zero) notation respectively.

For elastomers, the constitutive relation could be derived from strain energy density function where the stresses are expressed as [161, 167- 168, 173],

$$\sigma_{ij} = \frac{dW}{de_{ij}} \tag{4.12}$$

Where σ_{ij}, W and e_{ij} are true (Cauchy) stress, strain energy density function (strain energy per unit volume) and strain measures such as finite Eulerian strain tensor or stretches.

Convecting the coordinates with deformation in Eulerian measure in effect forms a link between finite strain elasticity and the simple idea of stretch or extension ratio which is used in the molecular theory of rubber elasticity. The derivation of stress is similar to the derivation of force on a particle in a potential field from a differentiable potential function because of the analogy between path independent work done on the particle and the reversibility of deformation (linear or non- linear elastic) as well as independence of stress to deformation history (linear elastic). Any material (both linear and nonlinear elastic) for which such a potential strain energy density function exists is called Green- elastic or hyperelastic [161, 168]. Taking series expansion of Eq. 4.12 and considering linear terms only result in Hooke's law (Eq. 4.11) which takes the following 3- D form for an isotropic, linear elastic sold with infinitesimal deformation and strain [169].

$$S_{ij} = \sigma_{ij} = \lambda\delta_{ij}\varepsilon_{kk} + 2G\varepsilon_{ij} \tag{4.13}$$

Where, δ_{ij} is Kroenecker delta (1 for $i = j$, 0 for $i \neq j$), λ and G (shear modulus) are Lamé constants and ε_{kk} as well as ε_{ij} are tensor components of infinitesimal normal and shear strains respectively, with the tensor shear multiplied by 2 to obtain engineering shear strain for the engineering stress (S_{ij}) which is equal to the Cauchy or true stress (σ_{ij}).

This is a derivation from the following generalised Hookean form of mechanical constitutive relation (which states that stress tensor is linearly proportional to the strain tensor) where zero body-moment, material symmetry and isotropy have not been assumed [168- 169].

$$S_{ij} = \sigma_{ij} = C_{ijkl}\varepsilon_{kl} \tag{4.14}$$

Where, C_{ijkl} is a tensor of rank 4 of elastic constants or moduli, comprising of 81 elements which comes down to 2 independent Lamé constants with progressive imposition of the assumptions.

Similar to the symmetry of the stress tensor ($S_{ij} = S_{ji}$, $\sigma_{ij} = \sigma_{ji}$) because of zero body moment under equilibrium which brings down the independent stress components to 6 (from 9) and the tensor for elastic constants to rank 2 (C_{ij}, 36 constants), the material symmetry indicates symmetry of C_{ij} which reduces the constants further to 21. This is a condition for real materials i.e. the existence of strain energy function with the elastic energy a single valued function of strain [169, 172].

For finite strain solutions, a suitable strain energy density function is to be constructed. It is imperative that the reliability and effectiveness (*or safety in larger context*) of FEA predictions on seal performance is largely dependent on reproduction of the actual material behaviour by a suitably chosen strain energy density function which, at the same time, should involve minimum material constants and specimen- level (tension- compression stress- strain on unaged and aged specimens) tests for economy [160, 171].

Knowing the 6 components of stress (3 normal and 3 shear) at a given point in a body under equilibrium ($S_{ij} = S_{ji}$, as body moment is 0, reduces independent stress components from 9 to 6) determines the state of stress at that point from which normal and shear stresses on any plane passing through the point could be determined. The Cauchy's formula of stress vector on a plane in terms of the stress tensor is given as [169, 172],

$$T_i^{\eta} = \eta_j S_{ji} \tag{4.15}$$

When, $\boldsymbol{\eta}$ is the normal to the plane on which stress is being defined.

The traction or stress ($S_{(n)}$) normal to the surface (or along $\boldsymbol{\eta}$) could be written as,

$$S_{(n)} = T_i^{\eta} \eta_i = S_{ij} \eta_i \eta_j \tag{4.16}$$

The engineering shear stress (τ) or stress tangential to the surface then becomes,

$$S_{(t)}^2 = \tau^2 = \left|T_i^{\eta}\right|^2 - S_{(n)}{}^2 \tag{4.17}$$ /

The set of 3 governing equations of static equilibrium for a 3- D solid under surface traction (aerodynamic pressure, friction, mechanical contact etc.) and body (gravitational, electromagnetic, inertia) forces in rectangular Cartesian coordinate system could be written as [169],

$$\frac{\partial S_{ji}}{\partial x_j} + \dot{f}_i = 0 \tag{4.18}$$

When, $\dot{f}_i$ is the body force vector along the 3 axes, 1(x)- 2(y)- 3(z).

A plane is under pure strain if the shear stresses on that plane is zero. In rectangular Cartesian coordinate system (defined by three independent variables) there are 3 such representative and orthogonal planes and the normal to these planes form the principal axes of strain i.e. where the strains (pure tensile or compressive) are maximum. The normal stresses (principal stress) as well as the extension ratios (principal extension ratios i.e. $\lambda_1, \lambda_2, \lambda_3$) along the principal axes are maximum at the same time for pure, homogeneous strain which determines material failures (such as cracking) to a large extent. The condition is feasible (i.e. same principal axes for stress and strain) if the material is isotropic, which signify that the constitutive relation given in Eq. 4.14 is isotropic (does not change with orthogonal transformation comprising of translation, rotation and reflection) and the tensor C_{ijkl} is isotropic i.e. the array of material constants do not change value with orthogonal transformation (i.e. direction) to any rectangular Cartesian system in Euclidian metric space [168- 169, 173].

Isotropy also means same lateral contraction in every direction perpendicular to pulling. Symmetric stress tensor ($S_{ij} = S_{ji}$, $\sigma_{ij} = \sigma_{ji}$) also means that the three principal stresses are all real and the three principal planes are all orthogonal. If S_1, $S_2$and S_3 are the 3 principal stresses along 3 principal directions or η_1, η_2 and η_3, which are components of the unit normal vector $\boldsymbol{\eta}$, then the solution could be obtained from the following set of three equations expressed in tensor index notation which is basically an eigenvalue problem [169- 170].

$$(S_{ji} - S\delta_{ji})\eta_j = 0 \tag{4.19}$$

The algebra of tensors for infinitesimal and finite strains are similar and analogous. Thus, for a homogenous pure strain where λ_1, λ_2 and λ_3(principal stretch or extension ratios along principal axes) are the lengths in deformed state of linear elements (parallel to the x, y, and z axes of a rectangular Cartesian coordinate system respectively), originally (undeformed, X-Y-Z) of unit length, the following set of equations relating the principal stretches and Lagrangian measure of finite strain ($É_{XX}$, $É_{YY}$, $É_{ZZ}$, $É_{XY}$, $É_{YZ}$, $É_{ZX}$; Appendix B) result [167- 168, 170].

$$\lambda_1^2 = 1 + 2É_{XX}, \lambda_2^2 = 1 + 2É_{YY}, \lambda_3^2 = 1 + 2É_{ZZ}, É_{XY} = É_{YZ} = É_{ZX} = 0 \tag{4.20}$$

This is a well-known eigenvalue/ eigenvector problem where λ_1, λ_2 and λ_3 are found by taking roots of the determinant,

$$\begin{vmatrix} 1 + 2É_{XX} - \lambda^2 & É_{YX} & É_{ZX} \\ É_{XY} & 1 + 2É_{YY} - \lambda^2 & É_{ZY} \\ É_{XZ} & É_{YZ} & 1 + 2É_{ZZ} - \lambda^2 \end{vmatrix} \tag{4.21}$$

The condition of incompressibility in elastomers (as given below) is a constraint of large significance (unlike steel where bulk or K and shear or G moduli are of the same order of magnitude) in regular implementation by displacement based FEA as bulk modulus is very sensitive to change in Poisson's ratio[25] (in particular when elastomer is constrained such as O- ring in groove; Fig. 3.18) and the much larger bulk modulus (incompressibility) compared to the shear modulus[26] (deformability) takes away the isotropy [44].

$$\lambda_1 \lambda_2 \lambda_3 = 1 \tag{4.22}$$

The indeterminacy of stress for a particular strain to the extent of a hydrostatic pressure (or mean stress) due to incompressibility was addressed by Hermann's modified variational principle (1965) for incompressible and nearly incompressible isotropic solids with introduction of an additional degree of freedom (mean pressure function) which resulted in mixed and hybrid formulations as well as elements in FEA codes.

$$H = \frac{\sum_{k=1}^{3} S_{kk}}{2G\ (1+\nu)} = \frac{\theta}{E_0} \tag{4.23}$$

Where H, θ/3, S_{kk}, G and ν are mean pressure function, mean stress, normal stresses, shear modulus and Poisson's ratio respectively.

The state of stress, ideally described by the stress tensor with 9 components (3 normal + 6 shear), is written for infinitesimal strain elasticity (could be expressed in terms of σ also) as,

$$S_{ij} = \begin{bmatrix} S_{11} & S_{12} & S_{13} \\ S_{21} & S_{22} & S_{23} \\ S_{31} & S_{32} & S_{33} \end{bmatrix} \tag{4.24}$$

Where the first subscript indicates direction of normal to the plane on which stress acts and the second one defines direction of stress where S_{11}, S_{22} and S_{33} are normal stresses.

[25] Typically, 4 times increase in shear modulus (0.34 MPa to 1.36MPa) corresponding to a change in Poisson's ratio by only 0.000375.
[26] Typically, a bulk modulus of 1360 MPa corresponding to a shear modulus range of 0.4 - 1.36 MPa.

The stress tensor could be looked at as comprising of two complimentary halves i.e. dilatational (hydrostatic) and deviatoric (pure shear) components responsible for change in volume and shape respectively. Hydrostatic component (p) of the stress tensor is expressed as [168],

$$p = \frac{1}{3}(\sigma_{11} + \sigma_{22} + \sigma_{33}) \tag{4.25}$$

Deviatoric stress tensor (σ'_{ij}) is obtained by subtracting the hydrostatic part from the overall stress tensor.

$$\sigma'_{ij} = \begin{bmatrix} (\sigma_{11} - p) & \sigma_{12} & \sigma_{13} \\ \sigma_{21} & (\sigma_{22} - p) & \sigma_{23} \\ \sigma_{31} & \sigma_{32} & (\sigma_{33} - p) \end{bmatrix} \tag{4.26}$$

The deviatoric stress tensor in tensor notation reads as,

$$\sigma'_{ij} = \sigma_{ij} - \delta_{ij}\frac{\sigma_{kk}}{3} \tag{4.27}$$

Where, δ_{ij} is Kroenecker delta.

Addressing the issue of incompressibility and assuring a unique stretch measure by ensuring unchanged orientations of unit vectors (due to deformation) associated with the original ($\underline{N}$) and deformed ($\underline{n}$) configurations necessitate the assumption of homogeneous deformation where the deformation gradient ($F_{i,A}$) does not depend on the original configuration X (Appendix B). Such deformations can be completely characterised by 3 stretch ratios along orthogonal directions. Generally, for isotropic hyperelastic solids, the strain energy density function is a symmetric function of principal extension ratios (λ_i; indicial notation indicating 3 normals on 3 orthogonal planes) which could be defined by three invariants of Cauchy- Green deformation tensor expressed in terms of the principal extension ratios [167].

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$$

$$I_2 = \lambda_1^2\lambda_2^2 + \lambda_2^2\lambda_3^2 + \lambda_3^2\lambda_1^2$$

$$I_3 = \lambda_1^2\lambda_2^2\lambda_3^2 \tag{4.28}$$

Where I_1, I_2 and I_3 are the three strain invariants.

A general form of W, expressed in terms of the power series of invariants and in conformance with the principles of continuum mechanics (i.e. zero stress at zero deformation) is given as [161],

$$W\ (I_1, I_2, I_3) = \sum_{i=0,j=0,k=0}^{\infty} C_{ijk}\ (I_1 - 3)^i (I_2 - 3)^j (I_3 - 3)^k \tag{4.29}$$

Where C_{ijk} are material constants.

The function is zero at zero deformation ($\lambda_1 = \lambda_2 = \lambda_3 = 1$) if $C_{000} = 0$, as also the stress predicted by Eq. 4.12. With incompressibility ($I_3 = 1$) introduced, Eq. 4.29 reads,

$$W\ (I_1, I_2) = \sum_{i=0,j=0}^{\infty} C_{ij} (I_1 - 3)^i\ (I_2 - 3)^j \tag{4.30}$$

Zero deformation and stress similarly follows. As the first few terms of series generally dominate, taking a two-term linear approximation (i = 1, j = 0 and i =0, j = 1) results in the popular Mooney- Rivlin strain energy density function, first proposed by Mooney during 1940.

$$W\ (I_1, I_2) = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) \tag{4.31}$$

Recasting the equation for determination of material constants (C_{ij}) using Eq. 4.12expresses the stresses as [161],

$$\sigma_{ij}\delta_{ij} = \lambda_i \frac{\partial W(I_1, I_2)}{\partial \lambda_i} + þ \tag{4.32}$$

Where δ_{ij} and þ are Kroenecker delta and hydrostatic pressure respectively (corresponding to infinitesimal strain, Eqs. 4.23 and 4.25).

The additional term þ could be considered as a Lagrange multiplier for compliance with incompressibility and accounting for any hydrostatic traction. The general expression of Cauchy or true stress in Eq. 4.32 is transformed into the expression for principal Cauchy or true stress for an initially isotropic, homogeneous and incompressible elastomeric solid (undergoing finite deformation)using strain energy density per unit volume [174] as function of principal stretch ratios i.e. W ($\lambda_1, \lambda_2, \lambda_3$).

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i} - þ i \in \{1,2,3\} \tag{4.33}$$

By regarding two of the stretches as independent and treating the strain energy as a function of these through the definition $\hat{W}(\lambda_1, \lambda_2) = W(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1})$ one obtains,

$$\sigma_1 - \sigma_3 = \lambda_1 \frac{\partial \hat{W}}{\partial \lambda_1} \sigma_2 - \sigma_3 = \lambda_2 \frac{\partial \hat{W}}{\partial \lambda_2} \qquad (4.34)$$

4.8.3 Sizing and optimisation of backup seal by finite element analysis [144]

FEA was carried out at HASETRI, Rajasthan.

4.8.3.1 Overall approach

Similar to the methodologies for obtaining S- 4, the FEA procedures followed a route of shortlisting and finalization of constitutive relations (P1) as well as design (P1-P4) through 4 Phases.

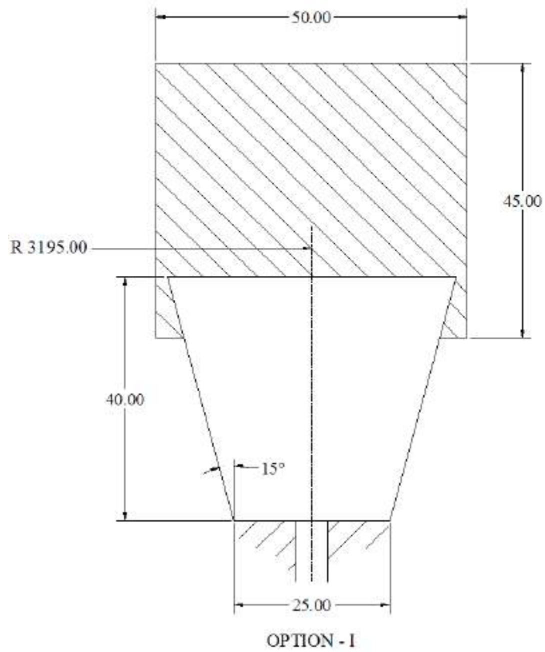


Fig. 4.6. Backup seal design: bounds of cross-sectional dimensions for optimisation.

Analysis was carried out in ABAQUS. Initial seal model was established (Phase identification: P1) using DMF3 or S-1. IG3 (S- 4) properties, LRP/SRP radius as indicated in Figure 3.46 and cross-sectional bounds of seal and holder as shown in Figure 4.6 were used during all subsequent phases. Seal compression was imparted by displacement of seal holder. Optimization was done based on simultaneous conformance of the identified design choice parameters to specified limits viz. i) Maximum true principal stress in seal cross-section at ET (110 ^{0}C) $\leq$ 0.7 MPa, ii) Maximum seal compression load at ET $\leq$ 5 kN/m-seal-length and, iii) Minimum contact pressure (threshold pressure) at ET $\geq$ 0.1 MPa ($\geq$ 0.025 MPa during initial phases).

Maximum principal stress denotes peak tensile stress in sealing configurations at various annular gaps, level mismatches and coefficients of friction ($\boldsymbol{\mu}$). Minimum principal stress or the peak compressive stress is defined similarly. Threshold pressure is the maximum contact pressure between seal and the counterfaces where seal compression is less because of level mismatch (maximum: 2 mm, Fig. 3.17).

7 variations (Fig. 4.7) of the solid trapezoidal cross-section (Fig. 4.6) were analysed for design shortlisting (Phase identification: P2). The effects of variation of geometric attributes in the shortlisted designs were studied in 6 more options (P3) from which the final design was chosen with further modifications (P4). Unaged, conditioned/unconditioned IG3 (S- 4) properties (SR: 50 mm/min) determined at ET were used to arrive at the optimized design.

4.8.3.2 Material description

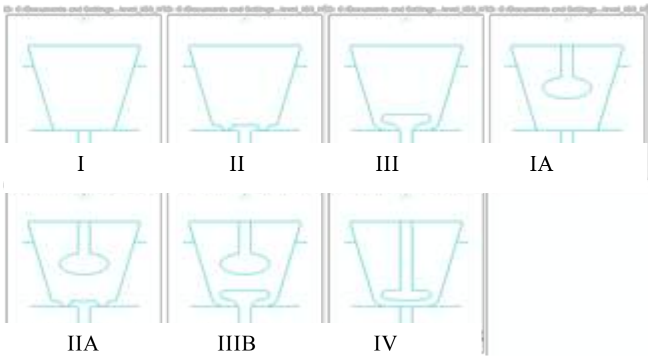


Fig. 4.7. Backup seal design: basic options.

Material constants for the chosen material (constitutive) model were determined by simultaneous inputs of tension and compression properties. As the backup seal is not constrained substantially (Fig. 4.7), pressures are low and compression/deflections are expected to be moderate, a justifiable amount of material compressibility (ν= 0.493) was used.

4.8.3.3 Geometry and mesh generation

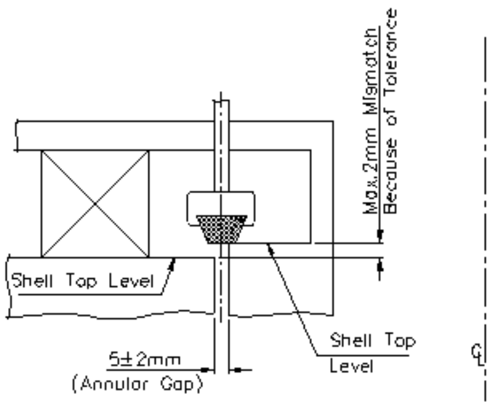


Fig. 4.8. Variable annular gap and level mismatch between backup seal mating surfaces.

The seal geometries shown in Figure 4.7 have axial and radial symmetry along with symmetry in loading as well as boundary conditions. Hence, they were analysed as axisymmetric cases. Full cross-sections were modelled considering level mismatch between the mating surfaces (Figs. 3.17, 4.8). The machined, unlubricated seal counterfaces coupled with limited constraints and moderate compression suggest utility of plane strain analysis. Axisymmetric modelling, however, was preferred because of its applicability to other INDIAN- SFR seals and to include possible compression increase, slip, change in mounting, variation of constraints and accidental pressure surge in reactor.

The models were generated using automatic mesh generation technique and 4- noded, continuum, axisymmetric elements of first order. Bending expected near the contact areas calls for employment of second order elements, which are bad in contact pressure prediction. This problem was addressed by increasing the mesh density. A uniform element length of 1mm was found to be suitable taking into account the edges in seal geometry. Hybrid type of element along with regular integration scheme was employed to address the issues of incompressibility. The seal holder and other metallic counter-surfaces of the seal were modelled as rigid body. Coefficients of friction of 0.5 and 1 were assigned for rubber-metal and rubber rubber-contact initially.

4.8.3.4 Boundary conditions and displacements

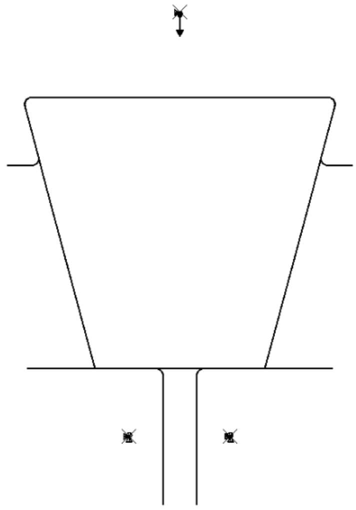


Fig. 4.9. Boundary conditions.

Figure 4.9 shows the applied displacements and boundary conditions in Option-I (Figs. 4.6- 4.7) used during analyses, which are applicable to other options as well. The contact definition is of penalty type with a slip tolerance of 0.2. The glued bonding between seal and holder is simulated by 'TIED' contact with adjustment of 0.1 mm for additional iterations. Since the sealing operation is essentially static, a 'STATIC' procedure with implicit iteration scheme was chosen. Axisymmetric (continuum) procedure/element is maintained during initial contact and deformed state of seal for all geometries.

The seal holder was given displacement till the threshold pressure between seal and mating surfaces reached (0.025/0.1 MPa). The displacement was subsequently increased to assess the differential pressure effect and absorb possible other compression variations in reactor. Annular gap variations and level mismatch were imparted by radial/axial movement of shell.

4.8.3.5 Shortlisting and finalisation of constitutive relations for minimum testing and numerical efforts and maximum reliability

FEA of backup seal in ABAQUS was preceded by shortlisting and finalisation of material model to be used for S- 4 based on the criteria of minimum testing effort, reliability of prediction in different deformation modes, stability of the model and applicability across cover gas seals of various mountings, compression, constrains, friction (slip) etc. Constitutive relations from representative categories based on statistical thermodynamics (Neo- Hookean, molecular domain), strain invariants (Mooney- Rivlin, Yeoh; phenomenological, continuum domain) and stretch ratios (Ogden; phenomenological, continuum domain) were considered for arriving at the final choice.

An outline of the phenomenological, strain- invariant based constitutive model by Rivlin incorporating near incompressibility of elastomer (shown below in its most general form) and its implementation in ABAQUS for fitment to material data (by determination of material or calibration constants) and determination of load, principal stress- strain is provided [162, 175].

$$W = \sum_{i+j=1}^{N} C_{ij}\,(I_1^c - 3)^i (I_2^c - 3)^j + \sum_i^N \frac{1}{D_i}\left(J^c - 1 - \acute{R}\right)^{2i} \quad (4.35)$$

Where N is positive, determining the number of terms in the strain energy density functions (N = 1,2,3), C_{ij} are Rivlin coefficients (describes shear behaviour of material), D_i define material compressibility, $\acute{R}$ is volumetric expansion with temperature change, I_1 and I_2 are strain invariants (superscript for compressibility; also called the first and second invariant of the deviatoric Cauchy-Green tensor) and J^c (also called elastic volume ratio) is $\lambda_1^c\lambda_2^c\lambda_3^c$ where λ_i^c is $(J^c)^{-1/3}\lambda_i$.

The first part of Eq. 4.35 is the deviatoric component (contribution from shear deformation) and the second part represents the volumetric component of stored energy for rubber. Material constants C_{ij} are found by regression to experimental data of stress strain. D_i and $\acute{R}$ are also material constants, determined from test data. The Cauchy (true) stress (or force per current area) is determined from [162],

$$\sigma = (A_1 + A_2 I_1^c)B^c - A_2 B^c B^c + PI \quad (4.36)$$

Where,

$$A_1 = \left(\frac{2}{J^c}\right)\frac{dW}{dI_1^c}; \qquad A_2 = \left(\frac{2}{J^c}\right)\frac{dW}{dI_2^c}; \qquad B^c = F^c(F^c)^T; \qquad F^c = (J^c)^{-1/3}F;$$

$$P = -\frac{dW}{dJ^c} \tag{4.37}$$

I is the identity matrix, F represents the deformation gradient matrix with information about current deformed coordinate (x_i) relative to the original, undeformed coordinate (X_i), Appendix B.

Assuming incompressibility, the true uniaxial tensile stress is derived as,

$$\sigma_{ut} = \frac{2}{J^{1/3}}\left(\lambda_1^2 - \frac{1}{\lambda_1}\right)\left(C_{10} + C_{01}\frac{1}{\lambda_1}\right) \tag{4.38}$$

On assuming incompressibility which maintains constant volume ($J^c = \lambda_1^c\lambda_2^c\lambda_3^c = J = 1$ and $\acute{R}(T) = 0$, hence: $\lambda_i^c = \lambda_i$, $I_i^c = I_i$ and volumetric component of Eq. 4.35 becomes null) Eq. 4.35 assumes a form very similar to Eq. 4.30 with the infinite series of hyperelasticity getting converted to finite (N instead of ∞) indicating limited number of material constants to be determined depending on the order of expansion. The two- term (N= 1) Mooney- Rivlin strain energy density function (Eq. 4.31) obtained this way (also from Eq. 4.30) could be further simplified through the phenomenological or continuum mechanics route to the single- term Neo-Hookean form, also derived with the approach of statistical kinetic theory of rubber elasticity [162].

$$W = C_{10}(I_1 - 3) \tag{4.39}$$

The uniaxial stress- strain derived from Mooney- Rivlin leads to the following relationship with the classical small strain Young's (E_0) and shear (G) moduli.

$$E = 6(C_{10} + C_{01}) \cong 3G \tag{4.40}$$

The ABAQUS FEA code uses a general polynomial form of the strain energy potential for hyperelastic materials without the term R in Eq. 4.35 [176].

The derivation of stresses and material constants could be looked at from another angle based on Eq. 4.32, obtained by assuming total incompressibility and then taking two terms (either through Eqs. 4.29 and 4.30 or through Eq. 4.35 and the ABAQUS representation), as below which provides an idea about the quantum of effort involved in determining the constants in uniaxial and biaxial modes with experimental methodologies (1% and 5% strain, at RT and ET, 50 mm/ min and 500 mm/ min, with and without stress softening) as already outlined in experimental methodologies.

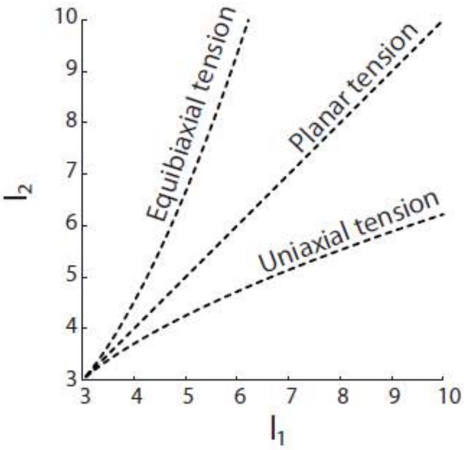


Fig. 4.10. Correlations between I_1 and I_2 for various deformation modes in incompressible solid.

A uniaxial loading along first axis could be described by stretch ratios ($\lambda_1 = \lambda$, $\lambda_2 = \lambda_2 = 1/\sqrt{\lambda}$) with λ_1 imposed and λ_2 as well as λ_3 are equal and obtained from the condition of incompressibility ($\lambda_1\lambda_2\lambda_3 = 1$) and isotropy. The stress from Eq. 4.32, assuming zero hydrostatic traction and Mooney-Rivlin relation, is-

$$\sigma_{11} = \left(\frac{\partial W \partial I_1}{\partial I_1 \partial \lambda} + \frac{\partial W \partial I_2}{\partial I_2 \partial \lambda}\right) \tag{4.41}$$

Expressing Eq. 4.41 in terms of material constants and stretch ratio takes identical form of Eq. 4.38 as J is 1 because of incompressibility.

Similar expressions could be obtained for equibiaxial loading on specimens which has been replaced by the equivalent uniaxial compression tests on specimens (Fig. 4.3) in the present study for minimising experimental efforts [161].

$$\lambda_1 = \lambda_2 = \lambda, \quad \lambda_3 = \frac{1}{\lambda^2} \tag{4.42}$$

$$\sigma_{11} = \sigma_{22} = 2\left(\lambda^2 - \frac{1}{\lambda^4}\right)(C_{10} + \lambda^2 C_{01}) \tag{4.43}$$

Figure 4.10 indicates that all deformation modes (i.e. tension, compression, shear) fall between uniaxial and biaxial tension (Fig. 4.11) with uniaxial compression being equivalent to biaxial tension [177- 178]. With experimental efforts minimised by choosing uniaxial tension and compression for specimen testing to cover the product stress field in all generality, it's the reliability of material constants in fitting the experimental data points which comes next. Usually, least square method is employed to assume a convenient interpolating polynomial and calculate the material coefficients (C_{ij}) or calibration constants which minimise errors in fitting the strain energy density function experimental data points of stress and strain. The following example of determining the Mooney- Rivlin constants for equibiaxial loading by using the analytical expression of principal stresses (Eqs. 4.43) as the interpolating polynomial is indicative of the methodology [161].

$$f(\lambda_j) = 2\left(\lambda^2 - \frac{1}{\lambda^4}\right)(C_{10} + \lambda^2 C_{01}) = \sigma_j \tag{4.44}$$

Where, $f(\lambda_j)$, σ_j and λ_j are interpolating function, j^{th} principal true stress from experimental data (Eq. 4.43) and corresponding principal stretch ratio respectively.

The principal logarithmic, Hencky or true strain (Eq. 4.3) could be obtained-

$$e_i = \ln \lambda_i \tag{4.45}$$

Where (Eqs. 4.2, 4.7),

$$\lambda_i = 1 + \epsilon_i \tag{4.46}$$

A linear least square technique (in contrast with nonlinear for Ogden) is required for determination of the unknown Mooney- Rivlin constants (Eq. 4.43) by minimising the following function (Eq. 4.47) as the constants are linearly related [161].

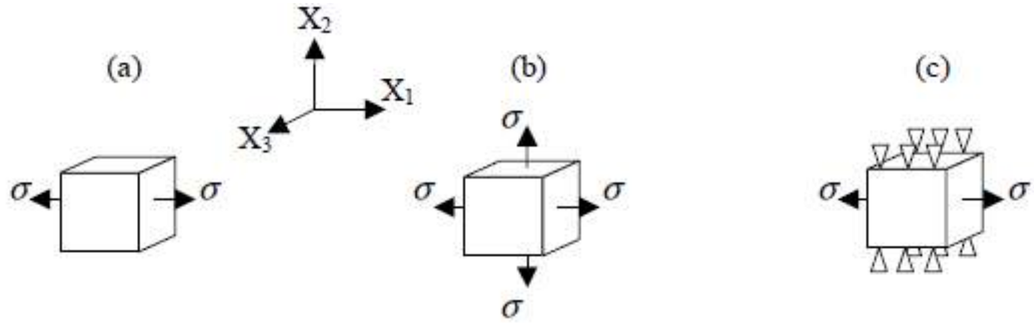


Fig. 4.11. (a) Uniaxial tension ($\lambda_1 = \lambda$, $\lambda_2 = \lambda_3 = \lambda^{-1/2}$, $\sigma_1 = \sigma$, $\sigma_2 = \sigma_3 = 0$), (b) Equibiaxial extension ($\lambda_1 = \lambda_2 = \lambda$, $\lambda_3 = \lambda^{-2}$, $\sigma_1 = \sigma_2 = \sigma$, $\sigma_3 = 0$) and, (c) Planar tension ($\lambda_1 = \lambda$, $\lambda_2 = 1$, $\lambda_3 = \lambda^{-1}$, $\sigma_1 = \sigma$, $\sigma_2 \neq 0$, $\sigma_3 = 0$). Fixed supports (triangles) do not allow free retraction in X_2 direction (on stretching in X_1 direction) and hence stress is not zero.

$$F(C_{10},C_{01}) = \sum_{j=1}^{n}\left[C_{10}\varphi_1(\lambda_j) + C_{01}\varphi_2(\lambda_j) - \sigma_j\right]^2 \tag{4.47}$$

Where, n is the number of experimental data points and φ_1 as well as φ_2 could be obtained by comparing the associativity of C_{10} and C_{01} in Eqs. 4.43-4.44 and 4.47.

The points λ_i that comply with Eq. 4.47 should meet the following conditions.

$$\frac{\partial F(C_{10},C_{01})}{\partial C_{10}} = 0, \qquad \frac{\partial F(C_{10},C_{01})}{\partial C_{01}} = 0 \tag{4.48}$$

4.8.4. Sizing and optimisation of inflatable seal by finite element analysis [36]

FEA was carried out at IGCAR.

Cross-sectional dimension of radially mounted unbeaded inflatable seal, withstanding pre-commissioning conditions in the INDIAN- SFR RPs for 6 y, was sized by FEA in ABAQUS for a groove dimension of 34 mm x 26 mm in TMR (Fig. 3.32) by optimising various seal design parameters (Fig. 4.12) with inflation pressure and principal true stress corresponding to a maximum design frictional drag of 1000 N/ m-seal-length, design differential pressure of 25 kPa, minimum contact pressure of 35 kPa between seal and mating BSR surface, radial gap

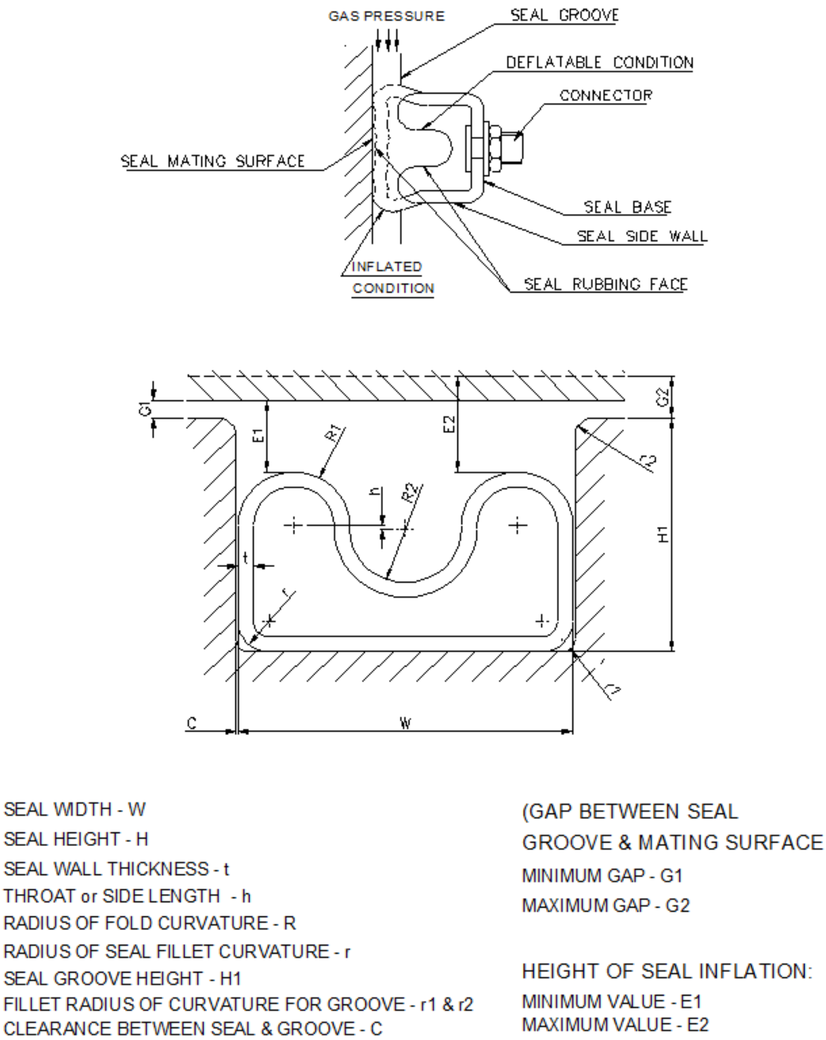


Fig. 4.12. Components and governing parameters of inflatable seal sizing and design by finite element analysis.

Table 4.17. Stress- strain data of inflatable seal fluoroelastomer compound (S- 1) determined at 120^{0}C.

Strain (%)	5	10	15	20	25	30	35	40	45	50	55	60	65	70	75	80	85	90
Stress (MPa)	0.37	0.58	0.80	0.99	1.17	1.34	1.50	1.67	1.84	2.0	2.20	2.40	2.59	2.81	3.03	3.26 (3)	3.33 (2)	3.38 (1)

of 5 ± 2 mm(between seal and mating surface, Fig. 4.12) and the limiting T_s of seal material (S- 1, 1.63 MPa) determined at 120 ^{0}C corresponding to a FOS of 2. A plane strain model was used with CPE4H hybrid quadrilateral element. Contact modelling was done using R2D2 and IRS21 elements and a FEA formulation with seal and groove acting as slave and master surfaces respectively. The basic boundary condition was to fix the seal at groove- base with groove boundaries (Fig. 4.12) and the mating surfaces (adjusted for a gap range of 3- 7 mm between BSR and TMR, Fig. 3.32) acting as limits to seal movement during inflation. Stress- strain data, determined on DMF3 (S- 1) at 120 ^{0}C (at a speed of 500 mm/ min) on ASTM Die C dumb- bell specimens, was used in Mooney- Rivlin constitutive relation for determination of true principal stress and strain.

Table 4.17 shows the stress–strain data of S-1 (determined in a Zwick UTS 1445 machine equipped with optical extensometer) used in ABAQUS. Test sheets were press-cured for 10 min at 177 ^{0}C followed by post curing for 24 h at 232 ^{0}C before stamping of the ASTM specimens. Test data (Table 4.17) were averaged over results from five specimens with figures in bracket indicating the number of specimens (when less than five) which survived a particular strain. A true principal stress limit of 1.63 MPa was applied corresponding to the T_s of S-1 at 120 ^{0}C.

Sensitivity of stress and inflation pressure were assessed for each of the governing design parameter (Fig. 4.12) by taking 3- 5 variations of each parameter and assessing the effects by simulation of inflation up to 50 kPa of inflation pressure. Optimisation was carried out taking the seal- groove boundary as well as the true principal tensile stress of 1.63 MPa as limit. A qualitative criterion was uniform contact between seal and mating surface without buckling on seal rubbing face (Fig. 3.43). Zero leakage criteria was ensured by assuming a minimum contact pressure of 35 kPa vis-à-vis the design differential pressure of 25 kPa.

Modified RIKS procedure was adopted (where load is considered as an unknown) to go through the phases of buckling of seal wall (which is not a failure criterion for rubber unlike steel) during inflation as the unbeaded seal unrolls towards its first contact (Fig. 4.12). Separate step based on restart file had to be introduced to extract the inflation pressure at first contact and during subsequent increments of inflation pressure up to 50 kPa. The inflation pressure could be extracted because of linear mapping between time and load (inflation pressure) by step functions. Sensitivity of stress and inflation pressure with respect to the element mesh size was studied and a stable mesh size of 0.5 mm was chosen. Master and slave surfaces used mesh sizes of 1 mm and 0.5 mm respectively for contact modelling.

4.8.5 **Life assessment and design of backup seal by finite element analysis** [144]

This involved determination of Principal stress- strain in backup seal by using uniaxial stress-strain data from accelerated aged specimens (determined at 110 ^{0}C) in ABAQUS and finding the time when they exceed the uniaxial, aged T_s and E_b (determined at 110 ^{0}C) including combined effects from synergistic ageing, batch- to- batch property variability, intra-laboratory measurement deviations, CDA pressure, strain etc., absorbed by a FOS of 2 applied on the ET T_s and E_b.

Prediction of life through a parallel failure mechanism (i.e. bypass leaktightness, Fig. 1.4) required finding of time in ABAQUS (using accelerated aged material data) when contact pressure (between backup seal and mating surface) is less than 0.1 MPa considering extreme effects from other variables such as tolerances from fabrication and assembly, coefficient of friction ($\boldsymbol{\mu}$= 0.1- 1) etc. Life assessment and design by FEA (including Arrhenius and WLF methodologies) were completed with validation *(results referred in this thesis for life assessment by design)* of the analysis results from ABAQUS by evaluation of 1 m diameter test seal for squeeze, assembly load and leakage in a scaled down test rig at RT and ET and ascertaining a life of 10 y from all considerations.

4.9 **Analysis:** Life assessment and design by Arrhenius and William Landel Ferry equations [87]

The following Arrhenius and WLF equations were used for life estimation from ISO 11346: 2004.

$$\ln K(T) = B - \frac{\bar{E}}{RT} \tag{4.49}$$

Where, K(T) is the reaction rate (min^{-1}), B is a constant, $\bar{E}$ is the activation energy (J/mol), R is the universal gas constant (8.314 J/molK) and T is the absolute temperature.

$$\log \alpha_T = -\frac{a(T-T_0)}{\{b+(T-T_0)\}} \tag{4.50}$$

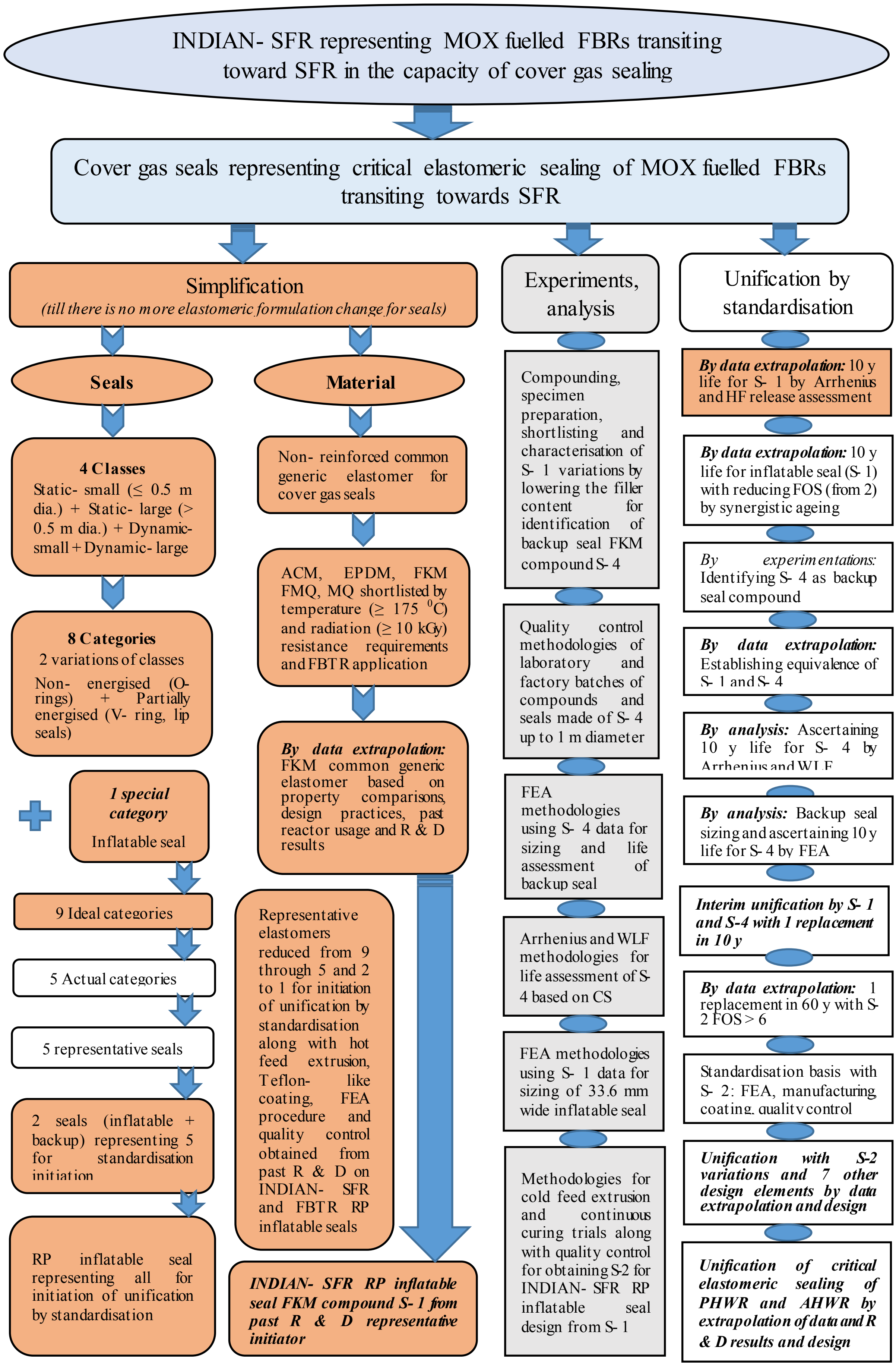
Where, α_T is shift factor, a and b are constants which depend only on material and T_0 is the reference temperature used to create the shift values.

Design was a competing process between life assessment by FEA and Arrhenius as well as WLF methodologies with the requirement that the minimum life obtained from all three should be ≥ 10 y. Life assessment by Arrhenius and WLF methodologies were based on limiting drop of 50% in unaged ET- T_s/ E_b (1.4 MPa/ 85%) of IG3 (S-4) and RT- CS of 80%. The WLF methodology is complimentary to Arrhenius in identifying contribution from physical degradation.

4.10 Chapter Conclusions

Experimental and analysis methodologies carried out as part of this thesis has been depicted (Fig. 4.13) which includes, i) Compounding, specimen preparation, shortlisting and identification of S- 4 (by tailoring S- 1) along with complete characterisation, ii) FEA for sizing, life assessment and design of backup seal made of S- 4, iii) Arrhenius and WLF methodologies for life assessment of backup seal based on CS, iv) Quality control tests of laboratory and factory batches of S-4 up to 1 m diameter test seal size, v) FEA for sizing and design of INDIAN- SFR RP inflatable seal of 33.6 mm width (*installed in INDIAN- SFR for past 6 y withstanding pre- commissioning*) made of S- 1 and, vi) Cold feed extrusion and continuous cure of ~ 2 m diameter test inflatable seals of INDIAN- SFR RP design for improved seal compound (S- 1 to S- 2) and seal towards 1 replacement in 60 y.

Fig. 4.13. Outline of research methodologies: Progress in Chapter 4



Chapter 5

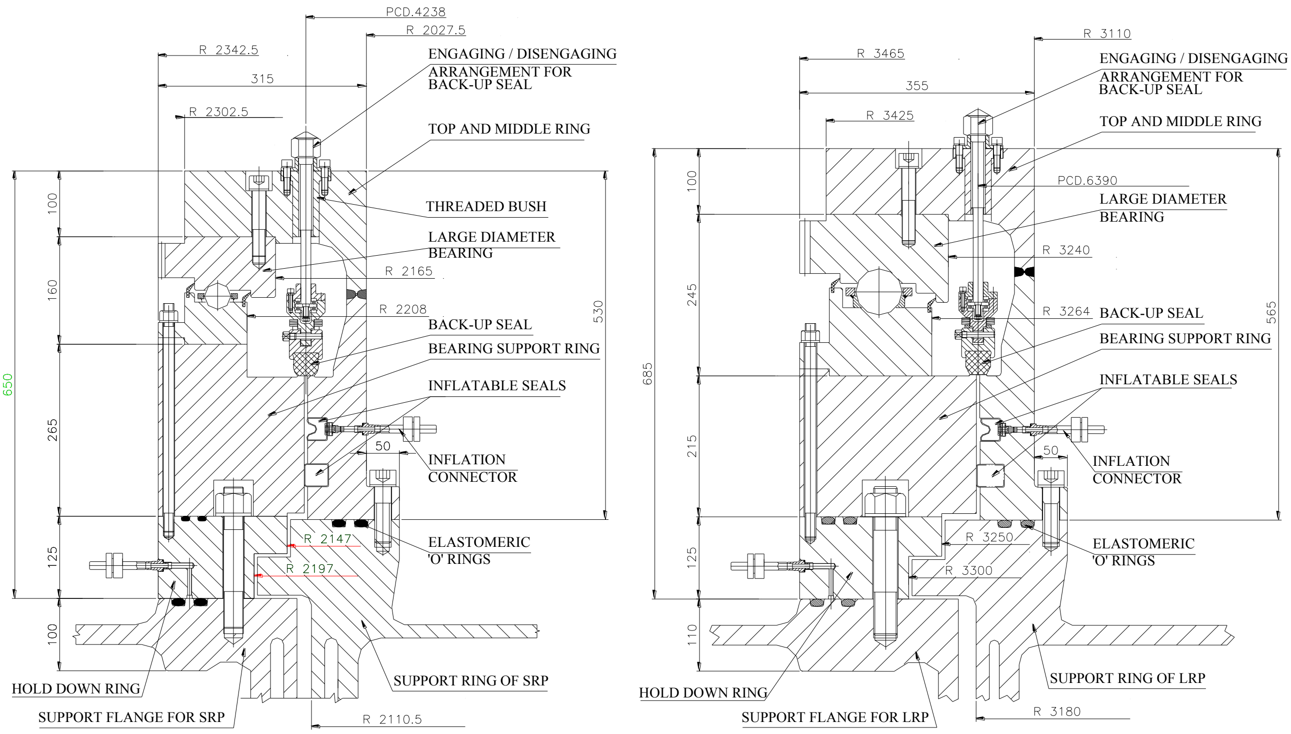
Results and Discussion

Preliminary unification and life maximisation of inflatable seal compound

5.1 Consolidating Ten Year Life for Inflatable Seal Using S- 1 by Data Extrapolation [81]

An earlier assessment (Sections 3.6.3, 3.7- 3.8) ascertained 10 y life for DMF3/ S- 1 (Table 3.11) based on 2 failure modes i.e. 50 % drop in T_s - E_b by Arrhenius methodology and estimating HF release by data extrapolation. Data extrapolation ascertains 10 y of life for S-1 here through a different route by evaluating the progressive loss of initial FOS (2 on unaged ET- T_s of 3.26 MPa and ET- E_b of 80%) with synergistic ageing from continuous exposure to temperature (120 ^{0}C), radiation (2 kGy in 10 y), mist and air (+ rubbing) at inflatable seal location in INDIAN- SFR RP.

Thresholds (5 kGy) for countable damage (modulus, T_s, E_b, hardness etc.) in elastomers generally [69] and Viton® in specific (1–10 kGy, irrespective of Viton® grade or filler content) [150] determined under high dose rate in air at RT suggests 10 y of life for S- 1without any other synergistic effect. Seminal research, carried out on 1.91mm thick Viton® specimens based on VDF and HFP (or Viton® A type) demonstrated that from a higher limiting dose rates of 5.5 kGy/h (radiation induced oxidation depth: 0.33mm;heterogeneous degradation) to the lowest of 0.13 kGy/h (complete oxygen diffusion; homogeneous degradation), imparted in air at 70 ^{0}C up to about1MGy cumulative γ dose, the specimens show progressive transition from a hard and brittle state (cross-linking dominated ageing)to a soft and stretchable state (scission dominated ageing) as higher dose rate consumes more oxygen in air to restrict its diffusion through elastomer and subsequent chain scission by radiation induced oxidation [59]. The single most important reason for equivalence between high- dose- rate- RT - damage- thresholds (elastomer: 5 kGy; Viton®:1–10 kGy) and low- dose- rate- RT γ exposure (2 kGy at the rate of 23 mGy/h) to inflatable seals in INDIAN- SFR RPs is radiation induced oxidation for the first few kGy of γ dose because of the consumption of dissolved oxygen in elastomeric parts. Similar thickness of inflatable seal wall (2mm), Viton® specimens (1.91 mm) and



SUPPORT ARRANGEMENT FOR SRP SUPPORT ARRANGEMENT FOR LRP

Fig. 5.1. Inflatable and backup seals in the support arrangement of large rotatable plug and small rotatable plug of INDIAN- SFR.

other specimens (2 mm) used for determining the thresholds signifies (by extrapolation) 10 y of life for DMF3 or S- 1 (Tables 3.11, 4.12) at RT without any loss of FOS. Negligible degradation in seal material under such condition will be driven by slow rate of scission in Viton® macromolecules at slow γ dose rate (23 mGy/h).

Results from another important work, carried out on specimens based on Viton® B (a terpolymer of VDF, HFP and TFE with 68 wt.% of fluorine) in air oven under a γ dose rate of 2.5 kGy/h and at a temperature range of 90–200 ^{0}C up to a cumulative dose of ~1.25MGy (homogeneous oxidation), indicated that S- 1 could sustain a cumulative dose of 2 kGy imparted at a temperature of 120 ^{0}Cwithout notable change in physical properties [82]. The extrapolation of results to S- 1 is based on the equivalence of Viton® B and Viton® A in this respect as shown by a review of significance in the area of fluoroelastomers [67]. Adding other attributes of seal functioning (i.e. cumulative rubbing of100km during seal design life 10 y) to correlated S- 1 and inflatable seal performance is unlikely to add further damage because of Teflon-like protective coating on seal rubbing face (Fig. 3.11). The possible synergistic influence of aerosol combined with radiation and temperature, in the absence of experimental data, has been factored in the FOS of 2.

The degradation mechanism of INDIAN- SFR RP inflatable seals (S- 1) under synergistic ageing (in the absence of mist) up to10 y comprise of competing phenomena of slow scission and cross- linking (thermo- oxidative ageing) of polymeric chains with negligible effects, as indicated by data extrapolation. Research on O-ring seals for radioactive material packages (material: Viton® GLT made of APA) which work under conditions similar to that of the inflatable seals (excepting a higher design temperature of 149 ^{0}C) also predict a similar ageing mechanism [67, 69- 71].

Results from another important work on Viton® B specimens indicate that at 10 kGy of cumulative γ dose imparted at 120 ^{0}C in air, T_s decreases by ~25% and E_b falls to about two-third of its initial value [82]. Inferior thermal resistance and physical properties of diamine cured Viton® B compared to bisphenol cure (S- 1) indicates that changes in T_s could be taken care by the initial FOS of 2. The ET- E_b of S- 1 (80%) at 120 ^{0}C (Table 4.12) however suggests that the compound may show vulnerability from localized strain exceeding limits (40%) at a cumulative dose of 10 kGy (*50 y of life at 120 ^{0}C*) apart from possibilities of seal-malfunction due to fall of E_b much below the usual threshold of elastomeric behaviour i.e. 100%. FEA results of inflatable seals indicate that the allowable principal true strain of 40 % for S- 1 (Fig. 4.1) is completely used up at 50 kPa of inflation pressure and there could be further reduction of E_b from synergistic ageing by sodium aerosol and possible hardening (cross- linking) in the inflatable seals as the dissolved oxygen is used up after 2 kGy of γ dose (Lower inflatable seal, Fig. 5.1). Data from product literature also indicate higher sensitivity (drop) of E_b to increasing radiation dose compared to that of T_s.

Consolidation of 10 y life for inflatable seals (made of S- 1) of INDIAN- SFR RPs from all the envisaged modes of failure indicates that life beyond 10 y is primarily limited by drop in E_b which suggests the necessity to increase FOS towards 1 synchronised TS seal replacement on 60 y.

5.2 Sizing and Optimisation of Inflatable Seal Used in INDIAN- SFR Rotatable Plugs [9-10, 36,43, 126]

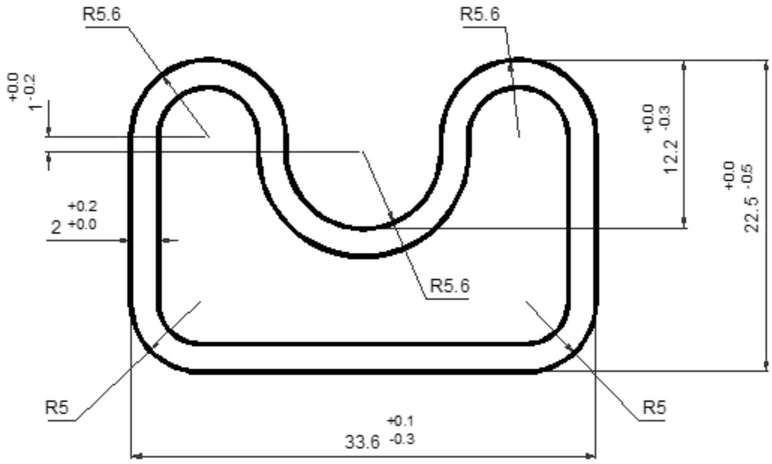


Fig. 5.2. Cross-section of inflatable seals used in INDIAN- SFR rotatable plugs.

Figures 5.2- 5.3 show the cross-sectional dimension of radially fitted inflatable seal arrived by FEA in ABAQUS (as per procedures in Section 4.8.3) using stress- strain data of bisphenol cured S- 1 and withstanding pre- commissioning conditions in INDIAN- SFR RPs for the past 6 y. The seal is designed to operate at inflation pressures of 50 kPa/ 30 kPa during normal-operation/ fuel handling conditions (Figs. 1.3, 3.20, 5.1, 5.3). Sizing for minimum stress-strain and maximum life initially suggested seal width and wall thickness of 54 mm and 3 mm respectively (Figs. 5.4- 5.5) which was modified subsequently to the present design (Figs. 5.2- 5.3) with smaller width (33. 6 mm in 34 mm x 36 mm groove) because of larger propensity of profile collapse under pressurised autoclave cure (typical pressure: 0.55- 0.7 MPa; typical temperature: 150-170 ^{0}C) following hot- feed extrusion (of inflatable seals made of S- 1) and the available space in TMR of INDIAN- SFRRPs (Fig. 5.1).

Parametric studies involving sensitivity of governing design parameters (Fig. 4.12) with respect to inflation pressure and stress indicated that maximum inflation pressure and principal stress for the first contact reduce with increasing seal width and

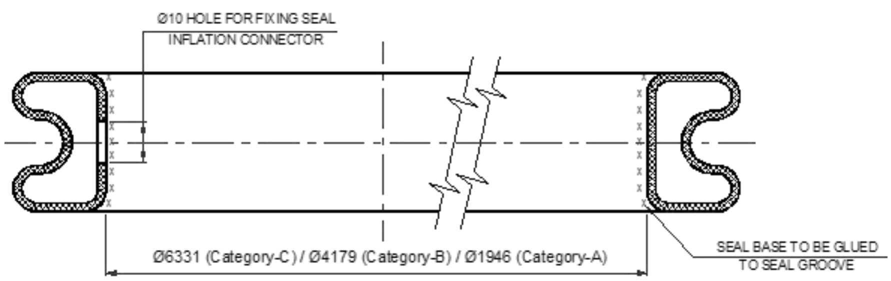


Fig. 5.3. Ring diameters of inflatable seals used in INDIAN- SFR rotatable plugs with details of hole for fitment of seal inflation connector.

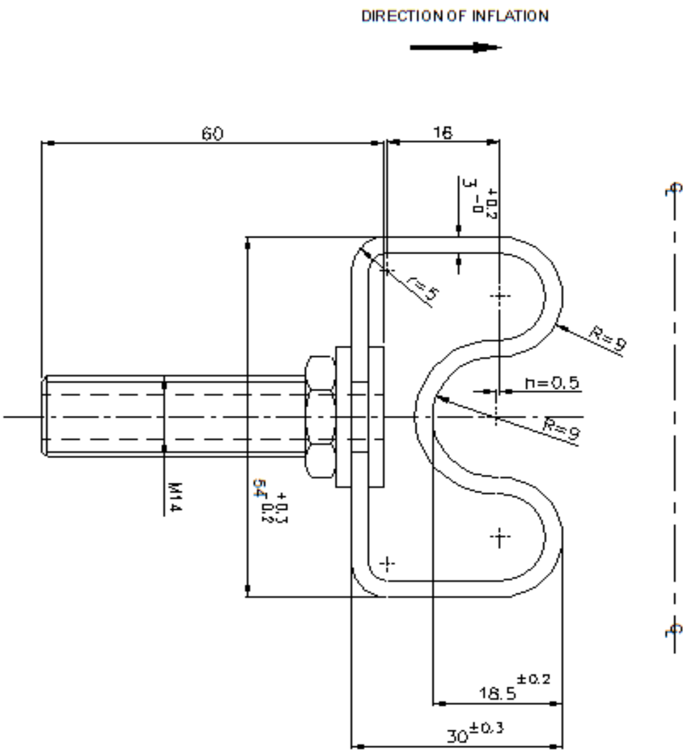


Fig. 5.4. Earlier design of INDIAN- SFR rotatable plug inflatable seal with width of 54 mm for minimum stress-strain and maximum life.

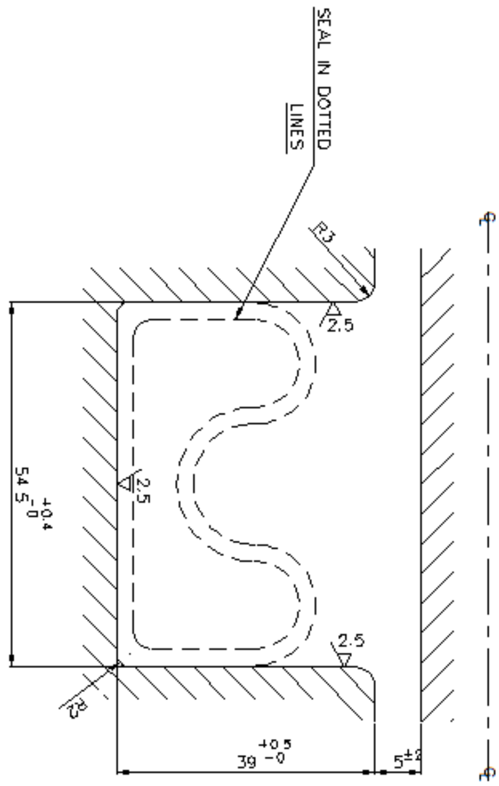


Fig. 5.5. Groove dimension of radially mounted inflatable seal corresponding to a width of 54 mm.

increase with seal wall thickness as well as radial gap from 3 mm to 7 mm. The maximum principal, true tensile stress corresponding to the seal width of 36 mm, 42 mm and 48 mm (seal wall thickness for all: 3 mm) were found to be 2.47 MPa, 2.02 MPa and 1.64 MPa using plane- strain condition, Mooney- Rivlin material model, hybrid quadrilateral element (CPE4H) and S- 1 as seal material in ABAQUS (Section 4.8.3) with 1.63 MPa as the limiting stress (T_s of S- 1 determined at 120 ^{0}C divided by the FOS, 2). Maximum principal, true tensile stress was lower (1.59 MPa) for seal width of 36 mm for a wall thickness of 2 mm (Figs. 3.34, 4.1).

Parametric study for optimisation was carried out by increasing the seal width in steps of 6 mm for simplicity in die design and manufacturability in Indian context. The folded, unbeaded inflatable seal design made of non- reinforced rubber comprises of 3 folds. Radius of fold was taken as odd or even integer and was increased by 1mm in each step (Figs. 3.35, 4.1, 5.4) where the present inflatable seal design (33.6 mm wide) has been a departure (Fig. 5.2). The inflatable seal design of

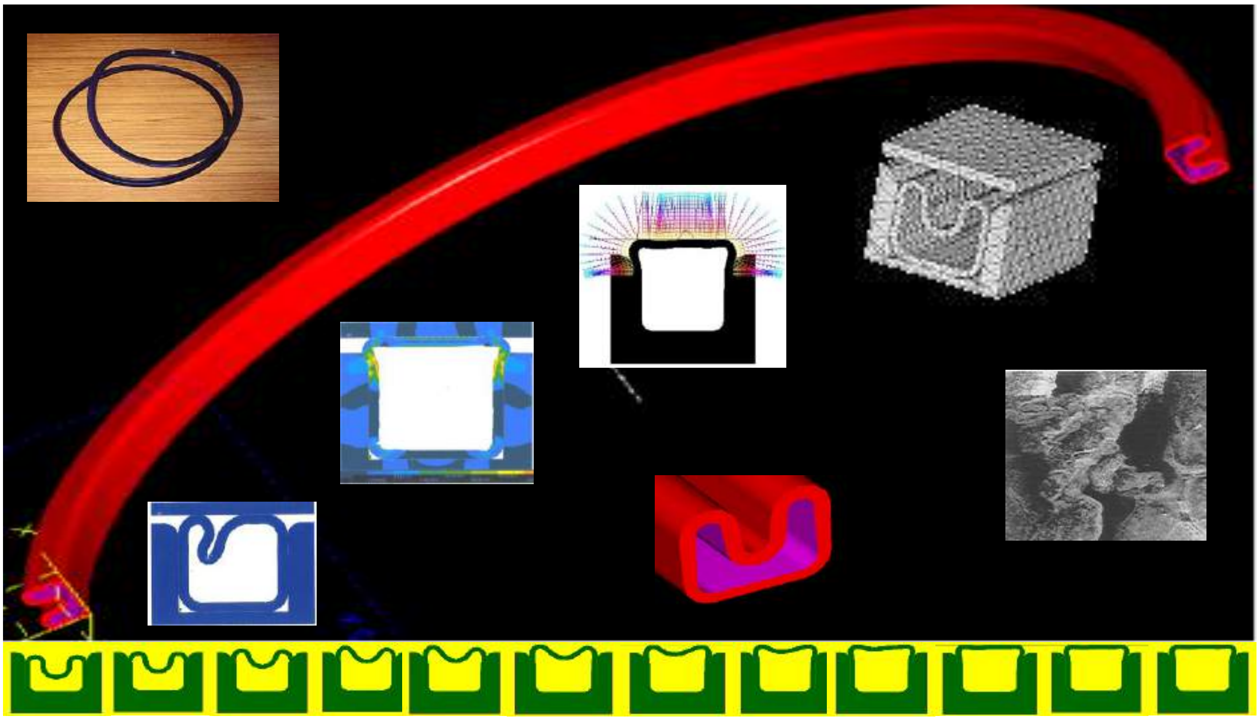


Fig. 5.6. Panorama of inflatable seal development: 3-D solid model of inflatable seal. *Insert:* ~ 0.5 m diameter test seals *(top left corner)*. *Clockwise from bottom left:* Finite element models of, a) Seal failure, b) Stress-field in inflated seal, c) Contact pressure profile and, d) 3-D seal-groove mesh. Scanning electron microscope image of surface degradation of silicone after 2000 hours of exposure to sodium aerosol at 120^{0}C. *Bottom:* Finite element simulation on progressive seal inflation.

33.6 mm x 22.5 mm used in INDIAN- SFR RPs was arrived based on these conditions and a minimum contact pressure of 35 kPa and design frictional drag of 1000 N/ m- seal- length.

Fig. 5.6 presents a panorama of INDIAN- SFR and FBTR inflatable seal development carried out as part of *Special Elastomeric Component* development where a solid model of inflatable seal is shown (Courtesy: IIT Kharagpur). Figs. 5.6a–d show FEA models of failure mode, stress field, contact stress profile and a 3D seal-groove mesh of inflatable seal as inserts (courtesy: IIT Madras, IIT Kanpur) along with a scanning electron microscope image of MQ rubber degradation on exposure to sodium aerosol (courtesy: DMSRDE, Kanpur). The progressive inflation of inflatable seal is shown as a finite element simulation in ABAQUS at the bottom of the figure. The inset does also show a 0.5 m diameter EPDM inflatable test seal of FBTR design (made of S- 3, Table 3.11; seal courtesy: DMSRDE; manufacture: M/s Ailga Rubber, Kanpur) which was qualified in uncoated and coated (Teflon- like, PECVD) conditions in scaled down test rig at IGCAR (Fig. 3.28).

5.3 Ascertaining Ten Year of Life for Fluoroelastomer Backup Seal Made of S- 4

This is carried out by combining 3 competing failure modes (i.e. drop of T_s and E_b to 50%, maximum CS of 80% and HF release) along with validation of the 1 m diameter test seal in scaled rig at IGCAR (Fig. 5.7) which is referred. The basic approach is that of loss of FOS with operating time because of synergistic ageing.

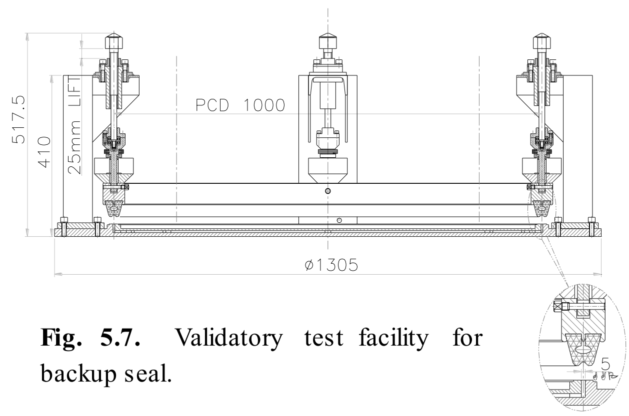


Fig. 5.7. Validatory test facility for backup seal.

5.3.1 Shortlisting IG3 (S- 4) as potential backup seal elastomeric formulation [87]

Table 5.1, in continuation of Table 4.9, summarizes the results of studies carried out as part of compound shortlisting as per Table 4.1. The abrupt increase of crosslinking (or cure), M_H, M_{100}, T_s and reduction of t_s2 as well as curing span (t'90 - t_s2) in IG2 compared to those of IG3 could be attributed to an increased level of $Ca(OH)_2$ (6 phr) in the former (Table 4.8). Influence of comparatively lower increase of MT black is reflected in the marginal increase of M_H, M_{100}, T_s and hardness (Table 5.1) coupled with decrease of t_s2 and E_b in IG1 (compared to IG2) and IG3 (compared to IG4), which are consistent with the trends reported in literature.

Results of mechanical tests on unaged specimens of IG2 – IG4 (Table 5.1) meet the target values of properties (Table 4.2). IG1 shows a marginal overstepping of the hardness limit. The improved T_s and E_b, coupled with a lower percentage increase of M_{100} compared to that of T_s in IG2 (with respect to IG3) gives the best combination of properties at RT. IG1 and IG2, however, were not chosen for further evaluation because of their sensitivity to ageing conditions (Table 5.1) even though the allowable variations (Table 4.2) of hardness (± 10%), Ts (± 15%) and E_b (±15%) under such conditions were conformed to. IG3 (S- 4) was selected for detailed investigation because of its higher T_s compared to IG4 and trends of property change during ageing which suggested better elevated temperature behaviour in terms of availability of safety margin or FOS.

5.3.2 Ascertaining IG3 (S- 4) as the backup seal elastomer through detailed studies [87]

Detailed studies were carried out as per testing schemes in Tables 4.3- 4.4 and 4.6 using the blended batch of IG3 i.e. IG3- A.

5.3.2.1 Properties of the blended batch

M_{50}, M_{100}, T_s, E_b and hardness of randomly selected blended batch, when measured at RT, were 0.9 MPa, 1.5 MPa, 6.2 MPa, 252% and 54 ^{0}Shore A, respectively. When measured at 110 ^{0}C, the M_{50}, T_s and E_b values changed to 1.1 MPa, 1.4 MPa and 85%, respectively. CS measured for 150 ^{0}C/72h and 150 ^{0}C/168 h were 7% and 9%, respectively. When compared with the earlier batch of IG3 (Table 5.1) the variations of properties (at RT) are insignificant considering the acceptable band of batch-to-batch property variability (T_s: 15%; E_b: 20%) and variations in reproducibility of test results (T_s: ~6%; E_b: ~9%; Hardness: ~3%) during intra- laboratory measurements.

Table 5.1.[27]Evaluation results of candidate compounds

Properties	IG1	IG2	IG3	IG4
Stock properties				
M_L (lbf.in/ N.m)	0.80/ 0.09	0.78/0.09	0.69/ 0.08	0.69/ 0.08
M_H (lbf.in/ N.m)	11.67/ 1.32	11.35/ 1.28	10.75/ 1.21	10.53/ 1.19
t_s 2 (min)	1.56	1.67	2.18	2.40
t_s 10 (min)	1.48	1.59	2.03	2.23
t' 50 (min)	1.69	1.81	2.40	2.62
t' 90 (min)	2.13	2.25	3.10	3.31
Vulcanizate properties *(Determined on unaged specimens at RT)*				
M_{100}(MPa)	1.8	1.7	1.5	1.4
T_s(MPa)	8.7	8.6	6.5	6.1
E_b(%)	263	280	252	257
Hardness (^{0}Shore A)	56	55	53	52
Determined on heat aged (150^{0}C/70 h in air) *specimens at RT*				
M_{100}(MPa) ^{a}	2 (11)	1.7 (0)	1.4 (-7)	1.4 (0)
T_b(MPa) ^{a}	7.5 (-14)	7.5 (-13)	6.5 (0)	6.3 (3)
E_b (%) ^{a}	230 (-13)	252 (-10)	262 (4)	258(0)
Hardness (^{0}Shore A)^b	56 (0)	55 (0)	53 (0)	53 (1)
CS: 150^{0}C/70h in air	150^{0}C/20 min ^{c}		150^{0}C/35 min ^{c}	
CS (%)	Cracked	7	7	7

[a] Numbers in brackets indicate percentage change of properties due to ageing; [b] Numbers in brackets indicate points (^{0}Shore A) of change in hardness due to ageing; [c] Specimen moulding conditions.

5.3.2.2 Extrusion characteristics

Extrusion trials carried out in Brabender Plasticorder using randomly selected blended batch produced excellent Garvey die rating of A8 at extruder die/barrel/feed-zone temperatures of 130 ^{0}C/75 ^{0}C/50 ^{0}C employing a screw speed of 5 rpm. The letter ratings ranging from A (excellent) to

[27]Torques measured by test apparatus in lbf.in has been retained (while including conversions in SI units) for the sake of proper comparisons as the t_s 2 values were also determined based on 2 lbf.in rise in torque above M_L. 1.00 lbf.in = 1.13 dN.m.

E (poor) indicate smoothness of extrudate. The digit ratings ranging from 1 (poor) to 10 (excellent) signify sharpness and continuity of extrudate edge produced through the 30 ^{0} edge of Garvey die. Extrusion die swell of the compound was measured through another set of trials which produced a die swell index of 1.36 at an extrusion rate of 31.11 g/min using die/barrel/feed-zone temperatures of 120 ^{0}C/75 ^{0}C/50 ^{0}C (screw speed: 15 rpm). These laboratory studies on manufacturability of IG3 (batch: IG3- A) by hot feed extrusion indicated that a combination of screw-speed/die-temperature of 10- 15 rpm/120 ^{0}C coupled with optimum backpressure could be suitable for production of backup seals with smooth surface and porosity-free cross-sections.

5.3.2.3 Long term heat ageing

Figs. 5.8a- h show the ageing behaviour of IG3 (batch: IG3- A) in air at 140 ^{0}C, 170 ^{0}C and 200 ^{0}C over a span of 32 weeks drawn as smooth curves through experimental data points. All the tensile tests shown in the Figs. 5.8a- f were carried out at a crosshead speed of 500 mm/min.

It is observed that variations of T_s and E_b (Figs. 5.8a-d) with ageing follow similar cyclic patterns. E_b measured at RT (Fig. 5.8c) tends to remain close to the unaged value of 252% (~ +4% to -5%) which are within experimental errors. E_b measured at 110 ^{0}C (Fig. 5.8d) remains above the unaged value (85%) by ≥ 10% most of the time which suggests unlocking of physical entanglement of polymeric chains at ET because of increased molecular mobility and the kinematic reconfigurations therefrom. Higher E_b and increased M_{50} or stiffness (unaged- RT: 0.9 MPa; unaged- ET: 1.1 MPa) because of Joule-Gough effect are the major contributors for the aged- ET- T_s dwelling at a level ~ 20% higher than the unaged- ET- T_s of 1.4 MPa most of the time (Fig. 5.8b).

The general cyclic variations of T_s and E_b with ageing at RT and ET (Figs. 5.8 a-d) comprise of two parts i.e. i) Covalent, chemical crosslink (bond) exchange in the macromolecular polymeric chains of specimens and kinematic reconfigurations therefrom, and ii) Presence of ionic groups at the end of polymer chains of IG3, which enhances the RT modulus, strength and viscosity by forming ionic clusters that act as chain extenders or temporary secondary network of weaker physical bonds.

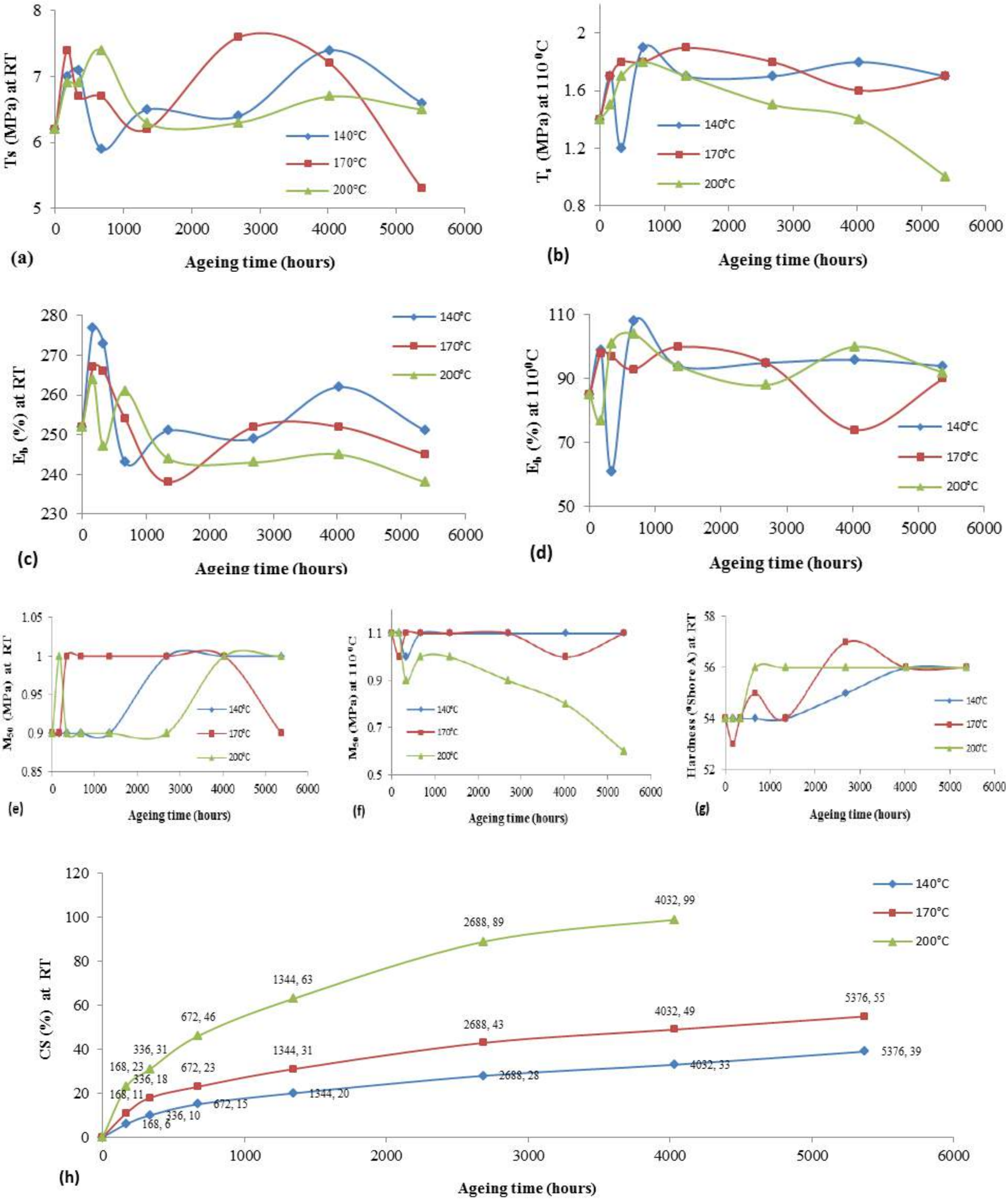


Fig. 5.8.(a) Tensile strength of IG3 measured at RT as functions of ageing time and temperature; (b) Tensile strength of IG3 measured at 110 ^{0}C as functions of ageing time and temperature; (c) Elongation at break of IG3 measured at RT as functions of ageing time and temperature; (d) Elongation at break of IG3 measured at 110 ^{0}C as functions of ageing time and temperature; (e) 50% modulus of IG3 measured at RT as functions of ageing time and temperature; (f) 50% modulus of IG3 measured at 110 ^{0}C as functions of ageing time and temperature; (g) Hardness of IG3 measured at RT as functions of ageing time and temperature and; (h) Compression set of IG3 measured at RT as functions of ageing time and temperature.

The second effect reduces in a reversible way during elevated temperature measurements because of the detachment of weaker physical bonds (courtesy: increased thermal energy and chain mobility), manifested by much lower variations of T_s with ageing at ET (Fig. 5.8b) and large reduction of unaged T_s (> 75%) and E_b (>65%) for IG3- A (similar to the unblended batch of IG3). These general characteristics of fluoroelastomers could also be found in DMF3 or S- 1 where the RT T_s (12.28 MPa, Table 4.12) and E_b (217%, Table 4.12) reduces by ~ 75% (to 3.26 MPa, Table 4.12) and ~65%(to 80%, Table 4.12) during ET (120 ^{0}C) measurements respectively. The strength at ET is primarily dependent on the restoring forces from the transverse thermal motion of chain segments between crosslinks (increased from RT because of Joule- Gough effect[28]) and hence do not reflect the unpredictability associated with ionic attractions which are built upon the probabilities of interactions between the end groups, rubber and its additives- manifested by much larger variation of T_s (measured at RT) with ageing compared to the ET T_s. This is substantiated further by the variability of RT and ET M_{50} (deformation resisted by thermal vibration of polymeric chains and kinematically limited by the chemical crosslinks or covalent bonds) within experimental errors with ageing (Figs. 5.8 e-f) and the invariability of RT M_{100} with ageing at 1.5 MPa.

The exceptional thermal stability of fluoroelastomers in general and IG3 (S- 4) in specific is evident from the invariabilities of modulus (M_x) which is also a measure of crosslink density. Absence of any perceivable increase (crosslinking) or decrease (scission of chains) of modulus clearly indicates absence of any chemical degradation effect from the thermooxidative ageing, supported by a virtually unchanged RT hardness (within experimental errors) with ageing (Fig. 5.8g) and absence of DLO effects, generally observed in elastomers with lower thermal stability.

Potential increase of E_b-FOS and T_s-FOS by ~10% and ~20% with ageing of S- 4 suggests backup seal life equivalent to reactor at least, under thermooxidative ageing alone and at unstrained state (specimens aged at unstrained state),in keeping with the general observation that

---

[28]Joule- Gough effect allowed by low MT carbon black content of 2 phr, Table 4.8, which leaves enough free volume in the macromolecular conglomeration, for the lateral thermal motion of chain segments between covalent crosslinks to intensify with temperature resulting in greater resistance to stretch or uncoiling of chains.

fluoroelastomers could work continuously at 200 ^{0}C. Monotonic increase (with ageing time) of another failure parameter (CS) under strain (Fig. 5.8h) indicates applicability of Arrhenius methodology and a life of at least 10y under synergistic ageing and strain by first assessment. which ascertained the choice of IG3 (S-4), supported by the extrudability results.

5.3.3 Equivalence of S-1 and S-4 in radiation resistance by data extrapolation [87]

Different fluids in contact with inflatable (argon gas, S- 1) and backup (air, S- 4) seals and thickness (2 mm vis-a-vis > 5 mm respectively) do not bring any performance difference up to cumulative γ dose of 2 kGy (or 10 y life) as the dissolved oxygen in S- 1 and S- 4 are consumed. It is known that incorporation of carbon black improves the radiation resistance of elastomer vulcanizate. Variation of the level of carbon black (S-1, 20 phr; S- 4, 2 phr), however, does not influence radiation resistance of Viton® A compounds significantly up to about 10 kGy, as has been reported by several sources. The level of $Ca(OH)_2$ influences water vapour and volatile generation (apart from controlling vulcanisation) rather than altering the radiation resistance.

S-4 could be assumed to be equivalent to S-1 in terms of radiation resistance.

5.3.4 Release of volatile from backup seal during operation in reactor [81, 87]

HF release induced by the usual operating temperature (~ 90 ^{0}C) of backup seal (S- 4) would be nil for all practical purposes irrespective of operating duration (Section 3.7) which is also applicable to all other cover gas seals of INDIAN- SFR because of much lower representative temperature of 120 ^{0}C vis- a- vis the threshold of release i.e. 160 ^{0}C. Release of HF from backup seal, induced by operating γ dose (23 mGy/ h), is expected to be ~7 times to that of inflatable seal (0.123 g) in 10 y because of the proportionality of release with weight (i.e. ~ 3.5 kg/m-seal-length). This is inconsequential considering the fact that the secondary RP barrier is located away from TS top plate and protected from direct mist- contact by primary inflatable seal.

About a third of TS seals (total circumferential length: ~ 700 m; total weight: 250 kg maximum) are in direct contact with mist. This suggests necessity for experimental verifications of possible effects of cumulative HF release from γ dose for the sake of unification by fluoroelastomer.

5.3.5 Design approach for life estimation by reducing factor of safety and increasing compression set [87]

This takes into consideration determination of stress- strain field in backup seal by FEA (using aged stress-strain data) in advance for the sake of illustration. Fig. 5.9 shows Mises stress distribution in the backup seal cross-section used in reactor.

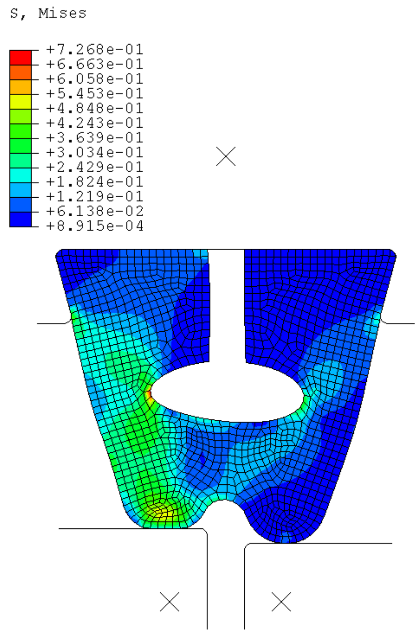


Fig.5.9. Deformation and stress field in backup seal cross-section used in INDIAN- SFR.

Results of ageing and FEA give a very clear indication that backup seal made of IG3 (S- 4) can work without failure in reactor for 10 y without any loss in the FOS of 2 on the ET- T_s of 1.4 MPa. FOS additions above 2 because of 20 - 10 % increase in T_s- E_b during thermooxidative ageing and maximum true strain of 35 % during normal operation (vis-à-vis ET E_b of 85%) take care of the variations in IG3 properties from batch-to-batch (T_s/E_b: 15/20% maximum) and intra-laboratory (T_s/E_b: 6/9 % maximum) deviations leaving the FOS of 2 for CDA pressure (2.1 MPa) resistance at any point of time during seal life. Possible synergistic effects of radiation and strain (+ variations from moulded specimens to extruded seals, 10%, Table 4.2) are to be combined with this prediction in order to assess the design life of IG3 (S- 4) and the seal for comparison with CS based prediction and choosing the minimum

5.3.6 Life prediction based on compression set [87]

The CS data of 39/55/99% after 32/32/24 weeks of air ageing under temperatures of 140^0/170^0/200^0C (Fig. 5.8h) are in tune with earlier findings. FKM compound (V 747-75) based on

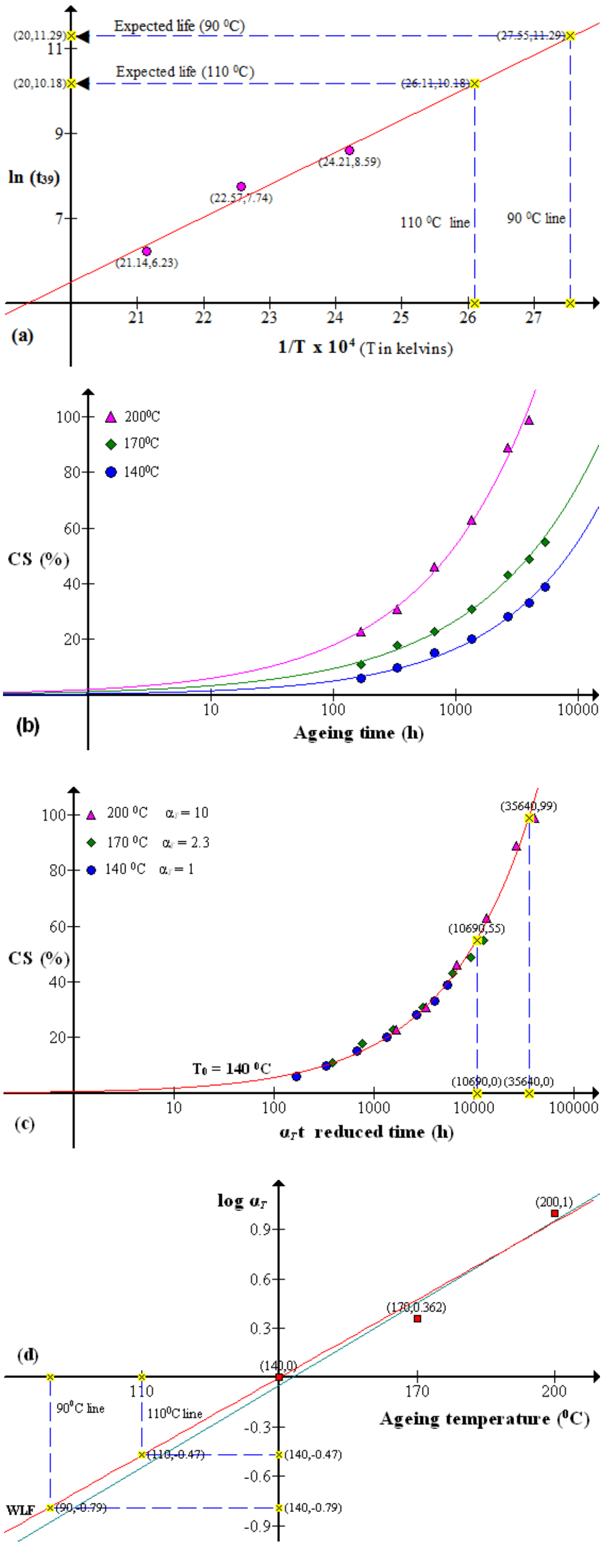


Fig. 5.10. (a) Service life prediction of IG3 assuming threshold compression set of 39% based on Arrhenius plot. (b) Compression set of IG3 plotted against log (time) x-axis at three ageing temperatures for construction of master curve. (c) Extraction of extrapolated data from compression set master curve of IG3 referenced to 140 ^{0}C. (d) Service life prediction from log α_T vs. ageing temperature plot based on William Landel Ferry equation.

Viton E-60C (VDF-HFP dipolymer precompounded with bisphenol curative and accelerator) with ~15 phr of carbon black produced a CS of 66% at 204 ^{0}C/1000 h in air. The lower CS (55%) in the current study (Fig. 5.8h) under comparable condition (200^{0}C/ 1000h) could be attributed to a better bisphenol curing system incorporated in the later generation Viton®A-401C.

The Arrhenius relation (Eq. 4.49) could be rewritten as below for CS.

$$\ln t_x = A + \frac{\bar{E}}{RT} \qquad (5.1)$$

Where, $\bar{E}$ is the activation energy (J/mol), R is the universal gas constant (8.314 J/molK), A is a constant and t_x is the threshold time required to reach a specific stage (x) of CS determined by the ageing temperature T.

The maximum CS common to all ageing temperatures is 39% (Fig. 5.8h). This is used as stage ‘x’ to plot Eq. 5.1 and construct a best fit straight line through the points as shown in Fig. 5.10a. It is observed that the points deviate from a straight line marginally. The threshold times required to reach 39% of CS (t_{39}) at 140 ^{0}C (5376h), 170 ^{0}C (2292 h) and

200 ^{0}C (508 h) are obtained from Fig. 5.8h. Activation energy (Ē) for the ageing process, calculated from the slope of the best fit straight line, is 63.49 kJmol^{-1} which predicts that attainment of 39% CS at 110 ^{0}C and 90 ^{0}C should take 3.01 y and 9.13 y respectively.

Eq. 5.1 is written as below to utilize data points beyond 39% of CS (Fig. 5.8h) for extrapolation.

$$\ln\left(\frac{t_2}{t_1}\right)_x = \frac{\bar{E}}{R}\left(\frac{1}{T_1} - \frac{1}{T_2}\right) \quad (5.2)$$

Where, t_1 and t_2 are times corresponding to the ageing temperatures T_1 and T_2 to reach a particular stage of CS, x.

The calculated periods for attainment of 55% and 99% of CS at 110 ^{0}C and 90 ^{0}C, using the known value of activation energy and extrapolating the experimentally determined data points at 170 ^{0}C and 200 ^{0}C in Eq. 5.2, are listed in Table 5.2. The tabulated results show some mismatch such as those between 55% CS extrapolation results (t_{55}) from 170 ^{0}C and 200 ^{0}C. The WLF method (Eq. 4.50) is therefore used for predictions.

Fig 5.10b shows Fig. 5.8h redrawn with ageing time (x- axis) represented in a log(time) scale. The curves are superimposed to produce a master curve for thermal ageing (Fig. 5.10c), referenced to 140 ^{0}C, by giving horizontal shifts (log α_T) to the non-reference temperature (200 ^{0}C and 170 ^{0}C) lines on the log(time) axis towards the positive direction. The shift factors (α_T) shown in Fig. 5.10c indicates that the ageing times corresponding to 170 ^{0}C and 200 ^{0}C are to be multiplied by 2.3 (log α_T: 0.362) and 10 (log α_T: 1) to obtain the same level of CS at 140 ^{0}C. Near-

Table 5.2. Backup seal life predictions based on compression set

Predicted times	Arrhenius			WLF
	Eq. 5.1, Fig. 5.10a	Eq. 5.2*(from 170 ^{0}C)*	Eq. 5.2*(from 200 ^{0}C)*	Eq. 4.50, Figs. 5.10c & 5.10d
t_{39} at 90 ^{0}C (y)	9.13	---	---	3.79
t_{39} at 110 ^{0}C (y)	3.01	---	---	1.81
t_{55} at 90 ^{0}C (y)	---	27.41	15.21	7.53
t_{55} at 110 ^{0}C (y)	---	9.14	5.07	3.6
t_{80} at 90 ^{0}C (y)	---	---	---	15.94
t_{80} at 110 ^{0}C (y)	---	---	---	7.62
T_{99} at 90 ^{0}C (y)	---	---	61.35	25.1
T_{99} at 110 ^{0}C (y)	---	---	20.38	12

perfect superimposition indicates realistic extrapolation of elevated temperature results to backup seal service condition.

Fig. 5.10d shows the logα_T vs. ageing temperature plot where the best fit straight line and the best fit for the WLF equation are constructed through the points using standard curve fitting technique involving linear regression. The times for attainment of 39%/55%/80%/99 % CS at 90 ^{0}C and 110 ^{0}C are computed by multiplying the corresponding values at 140 ^{0}C (from the master curve, Fig. 5.10c) by α_T (obtained from Fig. 5.10d) and are listed in Table 5.2. Only the modulus (or positive values) of logα_T are used for calculation of α_T as the data on the master curve at 140 ^{0}C (Fig. 5.10d) is effectively shifted in the same direction as those of 170 ^{0}C/200 ^{0}C lines in order to make the predictions. The best fit straight line in Fig. 5.10d does not pass through the 140 ^{0}C point (a requirement of the WLF methodology for computations of the material constant as per Eq. 4.50) and gives more optimistic predictions based on higher multiplication factors. These results are therefore not considered.

The temperature dependence of shift factor often follows an Arrhenius relation. Plotting the logarithm of α_T against the reciprocal of T (as in Fig. 5.10a) using the three points indicated in Fig. 5.10c produces a best fit straight line which gives an activation energy ($\bar{E}$) of about 60 kJmol^{-1} which is similar to the earlier energy calculated by Fig. 5.10a. The similarity indicates that a single chemical reaction determines the degradation at all the 3 ageing temperatures. Non- inclusion of physical changes in Arrhenius approach results in optimistic predictions and mismatch (Table 5.2). The near-perfect superimposition of IG3 (IG3-A batch) CS data obtained from accelerated thermo-oxidative ageing and the value of activation energy calculated are consistent with results from other short and long term ageing studies carried out on Viton® E-60C and Viton® GLT-S O-rings [72].

5.3.7 Ascertaining ten year of life for backup seal made of S-4 based on compression set by overall approach of design [87]

5.3.7.1 Assessment based on thermo-oxidative ageing alone

The CS results from standard specimens (Table 5.2) are applicable to backup seal because of similarities in the thicknesses, air exposure, long-term oxygen diffusion effects and compression of specimen as well as seal wall. Use of threshold CS values of 75-100% as elastomer seal failure criteria is common. The backup seal is disturbed during every fuel handling operation because of disengagements and engagements. The large seal diameter is expected to see squeeze variations along its circumference because of tolerance stack-ups from various sources. Considering these aspects, the threshold CS is reduced to 80%. As the CS results reported here are based on RT measurements, additional compression loss (~15%) because of differential thermal expansion between seal and metallic surfaces are included in the CS values measured. This margin of ~35% on the maximum seal squeeze of 6 mm accounts for the above uncertainties.

The backup seal life of 7.62 y and 15.64 y at 110 ^{0}C and 90 ^{0}C based on attainment of 80% of CS (Table 5.2) is conservative. It does not consider the disengaged condition of the seal during fuel handling condition which constitute ~8% of seal life. The reversible physical component of CS is also expected to be recovered during seal disengagement. A rough idea of the reversible component could be obtained by drawing a tangent to the master curve (from origin of coordinate) in Fig. 5.10c which indicates that the contribution of physical changes should be more than 10% of the total CS accrued. The master curve predicts shorter life compared to the experimentally determined points. As an example, the time for attainment of 99% CS at 200 ^{0}C are 4032 h and 3564 h as per experiments and master curve predictions respectively.

Going by engineering judgment it could be said that IG3 could last for more than 10 y and 20 y (based on CS) under thermo-oxidative ageing conditions of reactor considering backup seal design and usual operating temperatures of 110 ^{0}C and 90 ^{0}C respectively. These findings are consistent with the predictions from long-term accelerated ageing (~16000 h) studies of Viton® GLT-S O-rings carried out at a maximum temperature of ~175 ^{0}C considering the fact that the later contains more carbon black and hence is less resistant to CS [72].

5.3.7.2 Adding radiation: Assessment of dose rate effects by data extrapolation

The equivalence of S- 1 and S-4 in terms of radiation resistance and the role of radiation in synergistic ageing for S- 1, as depicted in Section 5.1, gives a very clear indication that adding radiation (to thermooxidative ageing) is going to produce negligible additional effects of degradation in synergism till 10 y of operation in reactor. Such observations are substantiated by decade long studies on O-ring seals of radioactive material package casks made of similar material and operating under comparable conditions [67, 72].

5.3.7.3 Considering complimentary strain effect on factor of safety by data extrapolation

Literature accounts indicate additional degradation of T_s and E_b when synergistic ageing is added with strain (seal compression in reactor), not evaluated by accelerated ageing of unstrained IG3 tensile specimens. Investigations on V 747-70, a proprietary compound based on Viton® E-60C, predicted 60% retention of stress under a tensile strain of 10% after ten years of ageing at ~150 ^{0}C. The study also revealed that below a temperature of 110 ^{0}C the changes or relaxation are entirely caused by physical or reversible effects [67].

5.3.7.4 Ascertaining ten-year life based on compression set under synergistic ageing by data extrapolation and engineering judgement

Studies on Viton 747-75 indicate 66.7% CS after 100 kGy of cumulative γ dose in air at RT. It is also stated that the effects of 10 kGy of dose under the same condition is minor [107]. Recent results from ageing studies of Viton GLT O-rings (cross-section: ~3.5 mm) of shipping package cask indicate a life of ~3.5 years at temperature and cumulative dose of 149 ^{0}C and 2 kGy [73]. Another study on fluoroelastomer indicates that 80% CS would be attained in ~1.14 years of time under temperature and cumulative γ dose (dose rate: 13 Gy/h). of 80 ^{0}C and 130 kGy [67].

These results give an indication that backup seal made of IG3 (S- 4) will require more than 10 y to attain 80% of CS considering synergistic influences of 90 ^{0}C temperature, 23 mGy/h dose rate and RCB air in reactor with the FOS of 2 on ET T_s and E_b kept to absorb CDA pressure surge (2.1 MPa) at any point of time during 10 y life and additional degradations expected from strain along with the variabilities (10 % maximum) in properties from moulded specimen to extruded seals

(Table 4.2). The latent additional FOS unaccounted so far is the reduction of contact pressure and stress with CS and provision of CS compensation by increasing squeeze by seal holder (Figs. 3.17, 3.45, 3.48, 5.1) if so required. Maximization of life (> 10 y) with quantification *of these aspects is* to be qualified for a temperature of 110 ^{0}C through experimentations involving assessment of long term synergistic ageing and dose rate.

5.3.8 Ascertaining ten year of life for backup seal made of S- 4 based on factor of safety by finite element analysis and design [144]

5.3.8.1 Shortlisting of constitutive laws

The uniaxial tensile stress-strain plot of IG3 (S- 4) determined at 110 ^{0}C (Fig. 5.11) illustrates minimal effect of stress softening due to low filler (MT black) content (2 phr). Low differential pressure (≤ 0.1 MPa) in FBR cover gas seals and their moderate strains (< 50% in most of the cases) during normal operation and fuel handling conditions, as demonstrated by studies on INDIAN- SFR and surveys on FEA results of rubber sealing applications [179- 186], suggest that a simple constitutive law should serve the purpose. Commercial codes such as ABAQUS and ANSYS use the same form of general constitutive relation for rubber with the term R (volumetric expansion corresponding to temperature change) omitted from the most general form of Rivlin equation (Eq. 4.35) for nearly incompressible, hyperelastic materials [160, 177]. Applicability could be expanded to dynamic seals if the chosen material model is able to simulate behaviour of rubber compounds with higher filler (up to 30 phr, at least) content [108, 118-119]. The possibility of standardizing the analysis and design procedures of FBR elastomeric seals are further enhanced by the fact that the low pressure, low

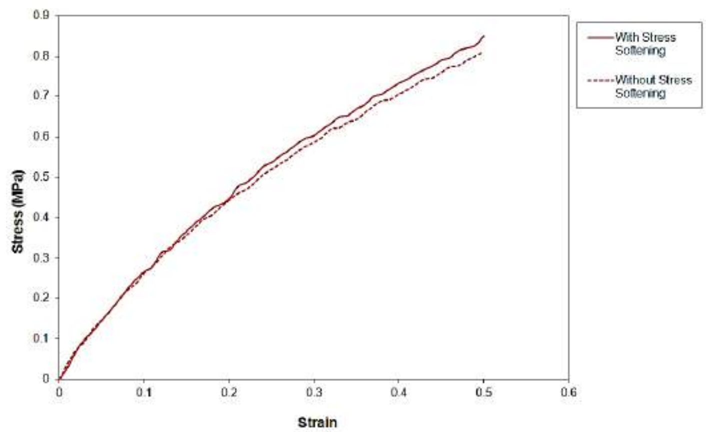


Fig. 5.11. Tensile stress-strain plot of IG3 (S- 4) determined at 110 ^{0}C with and without stress softening (strain rate: 50 mm/ min).

squeeze, static, elastomeric ring seal applications of reactor (having different cross-sectional geometries) could be represented by plane- strain condition under unlubricated contact, with axisymmetric analysis applied to account for other mountings, higher compression, larger constraints (imposed by seal groove) and possible slip between seal and mating surfaces [187- 188]. The shortlisting of constitutive model for IG3 (S- 4) was aimed at addressing these issues concurrently.

The slow rate of engagement of backup seal prior to normal operation implies isothermal deformation [168]. The behaviour of a hyperelastic, nearly incompressible, initially isotropic solid deforming under isothermal condition could be approximated by strain energy per unit volume as function of three strain invariants in continuation of Eq. 4.29 [167, 173]. The most generalised form of Rivlin constitutive model (Eq. 4.35) segregates Eq. 4.29 in two parts with the first part containing only the deviatoric components (without I_3or compressibility) while the second accounting for compressibility with addition term for volumetric expansion (R) with temperature [162, 175]. In ABAQUS and ANSYS the second term involving D_i (material compressibility) and J^c (elastic volume ratio) require additional experimentally determined pressure versus volume data at least (apart from uniaxial stress- strain) or introducing a Poisson's ratio (ν) to minimize experiments.

$$W = \sum_{i+j+k=1}^{\infty} C_{ijk}(I_1 - 3)^i(I_2 - 3)^j(I_3 - 3)^k \quad (5.3)$$

Where, W is the strain energy density and I_1, I_2 and I_3 are the three invariants of the Cauchy-Green deformation tensor expressed in terms of the principal extension ratios, λ_1, λ_2 and λ_3 ($\lambda_i = 1 + \epsilon_i$ where ϵ_i are the engineering strains along the principal axes) as below.

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$$

$$I_2 = \lambda_1^2\lambda_2^2 + \lambda_2^2\lambda_3^2 + \lambda_3^2\lambda_1^2$$

$$I_3 = \lambda_1^2\lambda_2^2\lambda_3^2 \quad (5.4)$$

Eq. (5.3) takes the following first order forms to present rubber-like behaviour on assuming incompressibility ($\lambda_1\lambda_2\lambda_3$=1) and taking the first (neo-Hookean model) and first two terms (Mooney-Rivlin model) of the power series, similar to Eq. 4.31 from Eq. 4.29 or Eq. 4.39 from Eq. 4.35.

$$W = C_{10}(I_1 - 3) \tag{5.5}$$

$$W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) \tag{5.6}$$

Where C_{10} and C_{01} are material constants, also called material parameters or calibration constants to be determined by regression of test data (4.8.2.5).

Combining Eq. 5.3 and isochoric behaviour with homogeneous deformation, the general stress-strain relations for uniaxial tension/compression (dumbbell specimen tests) and simple shear are derived as (uniaxial tension: $\lambda_1 = \lambda$ and $\lambda_2 = \lambda_3 = \lambda^{-1/2}$, Fig. 4.10),

$$\frac{S}{(\lambda - \lambda^{-2})} = 2\left[\frac{\partial W}{\partial I_1} + \frac{1}{\lambda}\frac{\partial W}{\partial I_2}\right] \tag{5.7}$$

$$\frac{\tau}{\gamma} = 2\left[\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\right] \tag{5.8}$$

Where, S, τ and γ are engineering or nominal stress, shear stress and shear strain respectively ($S_i = \sigma_i/\lambda_i$, when σ is the true stress).

Eq. 5.7 could be a direct derivation from Eq. 4.38 assuming J (elastic volume ratio) = 1 and using the engineering- true stress conversion in uniaxial mode along principal axis. The higher order Yeoh model expresses W in terms of the first strain invariant or I_1 only which minimises specimen tests for FEA data generation by confining to one uniaxial test only (*present study used uniaxial compression to cover biaxial behaviour, Fig. 4.3)*. This is obtained by ignoring $\partial W/\partial I_2$, as it is much smaller than $\partial W/\partial I_1$ generally.

$$W = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3 \tag{5.9}$$

Where, C_{10}, C_{20} and C_{30} are material constants.

Eqs. 5.7- 5.8 takes the following forms using neo-Hookean model.

$$\frac{S}{(\lambda - \lambda^{-2})} = 2C_{10} \tag{5.10}$$

$$\frac{\tau}{\gamma} = 2C_{10} \tag{5.11}$$

The interchangeability of Eqs. 5.10 and 5.11 indicate that results from a single uniaxial tensile test can represent other deformation modes, which minimizes testing requirements. Good correlations with test results from gum rubber are obtained up to strains of 0.4-0.9 in simple shear and uniaxial extension of 40-50%. Eq. 5.5, which could also be obtained from kinetic theory, has limitations in characterization of filled rubber. The shear modulus ($G = \tau/\gamma$) is constant and is related to the molecular parameters of statistical theory, which predicts linear behaviour in simple shear and is suitable for unfilled or lightly filled rubbers. The neo-Hookean model, therefore, is not considered.

The uniaxial tension/compression and simple shear stress-strains using Mooney-Rivlin model are related as,

$$\frac{S}{(\lambda-\lambda^{-2})} = 2C_{10} + 2C_{01}\left(\frac{1}{\lambda}\right) \tag{5.12}$$

$$\frac{\tau}{\gamma} = 2(C_{10} + C_{01}) \tag{5.13}$$

The shear modulus is constant like the neo-Hookean relation. Non-interchangeability of Eqs. 5.12 and 5.13 implies necessity of FEA data inputs from more than one deformation mode. The constants C_{10} and C_{01}, however, could be thought of symbolizing stiffness and change of stiffness in uniaxial tests, which gives good agreement with test data up to strains of 150% and 50% for unfilled/lightly-filled and filled compounds respectively. In spite of the known inaccuracies and inadequacies in describing the compression mode of deformation, the Mooney-Rivlin form finds wide use in commercial codes because of its smooth behaviour outside the measured strain range and monotonically increasing W with deformation. As previous studies demonstrate, the two-parameter Mooney-Rivlin model gives equivalent prediction of uniaxial compression in typical elastomer up to ~50% strain using data from uniaxial tension and another set of inputs from uniaxial, pure shear and equibiaxial tests. This brings in the relevance of this model in the present context in economizing the testing efforts.

The three-term Mooney-Rivlin model given below accommodates variable shear modulus and predicts tensile behaviour of highly filled elastomers quite accurately beyond strains of 100%.

$$W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) + C_{11}(I_3 - 3) \quad (5.14)$$

Where, C_{10}, C_{01} and C_{11} are material constants.

The use of higher order terms and more material constants to fit the experimental data with enhanced accuracy, however, is accompanied by poorer prediction outside the measured strain range and greater testing efforts which could be justified for special reactor sealing applications involving higher strain and analysis of accidental conditions under high differential pressure (2.1 MPa). The much-used Ogden strain energy function, expressed as a separable function of principal stretch ratios, is another example which accommodates different deformation modes, variable shear modulus and slightly compressible material behaviour along with accurate fitment to tensile test data up to 700% elongation. The general expression of Ogden strain energy potential takes the following form.

$$W = \sum \frac{2\mu_i}{\alpha_i^2}(\bar{\lambda}_1^{\alpha_i} + \bar{\lambda}_2^{\alpha_i} + \bar{\lambda}_3^{\alpha_i}) + \sum_{i=1}^{N} \frac{1}{D_i}(J^{el} - 1)^{2i} \quad (5.15)$$

Where, $\bar{\lambda}_i$ are the deviatoric principal stretches $\bar{\lambda}_i = J^{-\frac{1}{3}}\lambda_i$; λ_i are the principal stretches; N is a material parameter; and μ_i, α_i, and D_i are temperature-dependent material parameters; J^{el} is the elastic volume ratio.

Simple tension, pure shear and equibiaxial specimen test data are progressively added in the Ogden strain energy density function for increasing accuracy. The limitations of prediction outside the range of fit, however, is evident from FEA studies on 50 IRHD (international rubber hardness degree) NBR O-rings where Mooney-Rivlin model predicts contact stress and seal compression load with better accuracy compared to the Ogden when uniaxial tension data are used for extrapolation to compression domains [186]. The three-term Mooney-Rivlin and Ogden relations are therefore assigned for analysis of special cases and accidental conditions.

The uniaxial tension/compression and simple shear stress-strains using the higher order Yeoh form are related as,

$$\frac{S}{(\lambda-\lambda^{-2})} = 2C_{10} + 4C_{20}(I_1 - 3) + 6C_{30}(I_1 - 3)^2 \quad (5.16)$$

$$\frac{\tau}{\gamma} = 2C_{10} + 4C_{20}(I_1 - 3) + 6C_{30}(I_1 - 3)^2 \quad (5.17)$$

Eqs. 5.16 and 5.17 are interchangeable and the shear modulus varies with strain. Deviations (in modulus) from test results at low strains are acceptable as stress-errors are small in absolute terms. The model is applicable to wider strain ranges compared to the Mooney-Rivlin.

The Yeoh and Mooney-Rivlin relations are shortlisted for further numerical evaluation based on the study involving data extrapolation and principles of design.

5.3.8.2 Material model finalization

The calibration constants (C_{ij}) of Yeoh and Mooney-Rivlin models were determined by least square curve fitting technique using test data points. The coefficients were used to interpolate/extrapolate in the range of $0.1<\lambda<10$ in uniaxial, biaxial, planar and volumetric tension/compression modes to assess the material stability by determining whether the energy density function is positive definite. This is defined by Drucker stability criterion which demands that tangent stiffness matrix (or Hessian) must be positive definite. The Hessian, a 4th order tensor, could be written in terms of strain energy density function as,

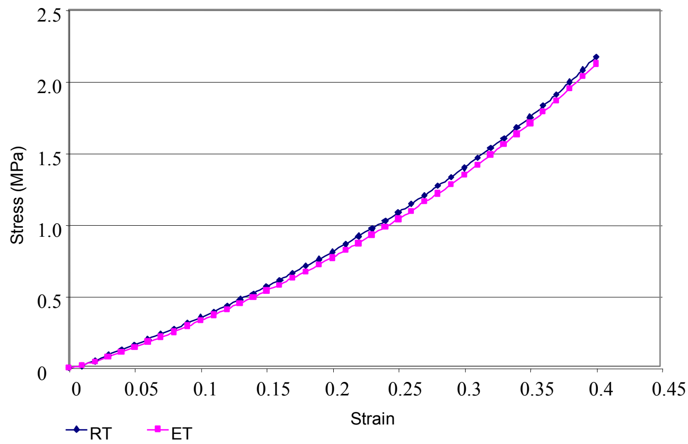


Fig. 5.12. Compression stress-strain plot of IG3 determined at RT and 110 ^{0}C(ET) without stress softening at a strain rate of 50 mm/min.

$$H_{ijkl} = \frac{\partial^2 W}{\partial E_{ij} E_{kl}} \quad (5.18)$$

Where, E_{ij} is the finite Lagrangian (Green) strain tensor.

Mooney-Rivlin model was adopted for sizing and optimization of backup seals, as it remained stable in most of the deformation modes and strain ranges while the Yeoh form showed instability using ET material data at strains well within the 50/20% (tension/compression) range.

5.3.8.3 Compression stress-strain plot

The compression stress-strain plots of IG3 (S- 4), Figure 5.12, indicate little change in behaviour because of increase in temperature of measurement.

5.3.8.4 Optimising fourteen geometries: Shortlisting of design

Figs. 5.13a-c show the variations of design choice parameters in the 7 basic seal geometric options (Fig. 4.8) obtained by FEA in ABAQUS using stress-softened specimen data. The maximum von Mises stress and contact pressure for LRP/SRP seals and maximum compression load for LRP seal (limiting engagement load for LRP/SRP seal: 100/65 kN) are presented considering the effects of possible level mismatch between the seal counterfaces (Fig. 3.17). Maximum load/stress is generated at maximum level mismatch.

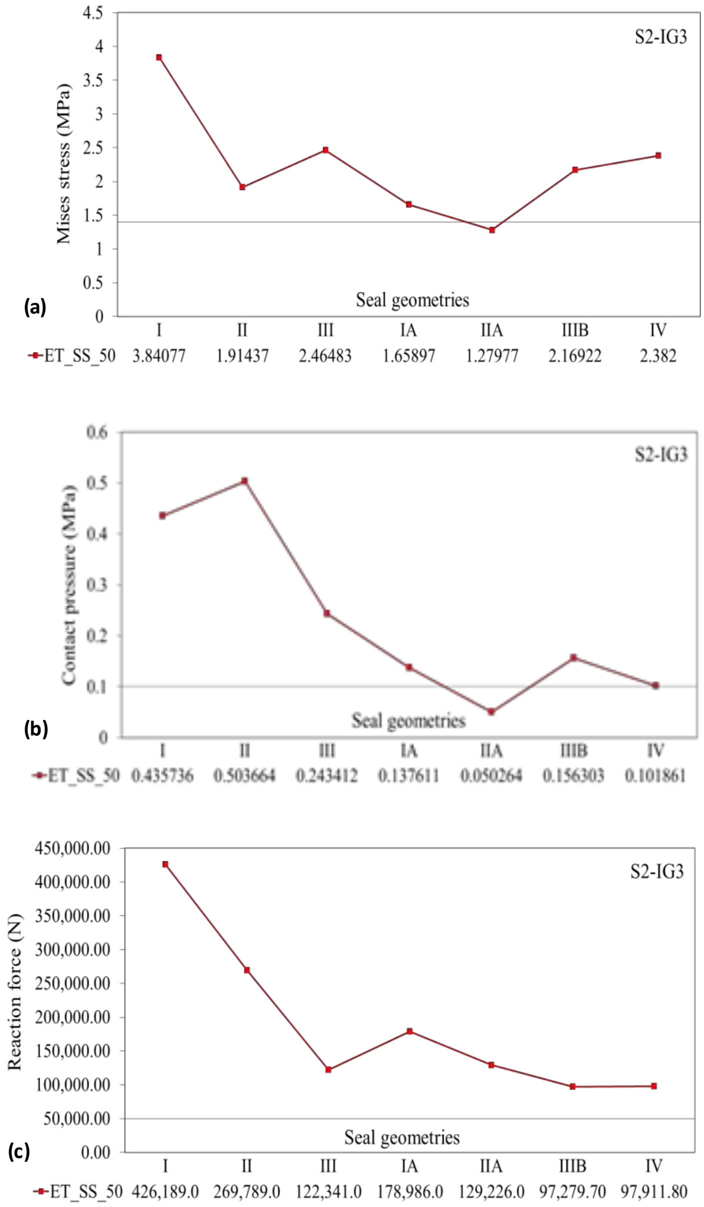


Fig. 5.13. Design choice parameters of basic geometric options obtained by using stress-softened (SS) IG3 properties determined at 110 ^{0}C (ET, strain rate: 50 mm/min). (a) Maximum Mises stress. (b) Minimum contact pressure. (c) Maximum reaction (seal compression) force.

It is observed (Fig. 5.13c) that introduction of O-ring (Option- II, Fig. 4.7) and lip profiles (Option- III, Fig. 4.7) on the main sealing face of the solid trapezoidal block (Option- I) brings down the seal compression load drastically by ~ 37% and ~71% as the solid cross-section in effect acts as a rigid body with the flexible sealing contacts allowing the deformation.

Table 5.3. Variation of design choice parameters with seal geometries (P2-IG3)

Design choice parameters[a]	Seal geometries *(Fig. 4.8)*						
	I	II	III	IA	IIA	IIIB	IV
Maximum Mises stress (MPa)	3.72581	1.87833	2.41293	1.63633	1.26435	2.12851	2.33833
Minimum contact pressure (MPa)	0.434983	0.502037	0.240186	0.13713	0.0500022	0.155777	0.101522
Maximum reaction force (N)	425,078	269,063	121,994	178,483	128,846	97,004.80	97,630.20

[a] Parameters obtained using unaged IG3 stress-strain data measured at ET without stress softening (strain rate: 50 mm/min)

Stiffness reduction (~58% drop) comparable to the addition of lip profile is seen when hollowness in seal cross-section is introduced (Option- IA, Fig. 4.7) keeping the flat sealing face intact. Recombining the O-ring (Option- IIA, Fig. 4.7) and lip (Option- IIIB, Fig. 4.7) profiles on the sealing face of the hollow cross-section leads to lesser gains in the seal assembly force (IIA/IIIB: ~28/42% drop compared to IA) as the lower stiffness of hollow seal section also shares some of the deformation, thus contributing towards the overall effective stiffness of the seal.

The contact pressure variations show interesting trends (Fig. 5.13b). Options I and II (Fig. 4.7) show much higher maximum contact pressure because of stiffening of the solid cross-section under deformation (at level mismatch) by the time the seal comes in contact with the lower counterface. This increases the sensitivity of contact pressure to deformation. The introduction of lips (Option- III, Fig. 4.7) having much higher flexibility reduces this sensitivity considerably. Hollowness provides space for the near incompressible material to absorb the deformations caused by the initial contact with the higher sealing counterface resulting in near disappearance of the sensitivity effects (Options IA, IIA, IIIB, IV; Fig. 4.7).

Figs. 5.13a-c reveal that Options IIA and IIIB (Fig. 4.7) are the best choices for further design refinement with contact pressure (~50 kPa) and von Mises stress (~1.28 MPa) minimum in the former and engagement load (~ 97 KN) minimum in the later even though the stress remains on the higher side. The design choice parameters determined by using data from specimens without stress softening (Table 5.3) substantiates the earlier observation (Fig. 5.11) that deformation cycling does

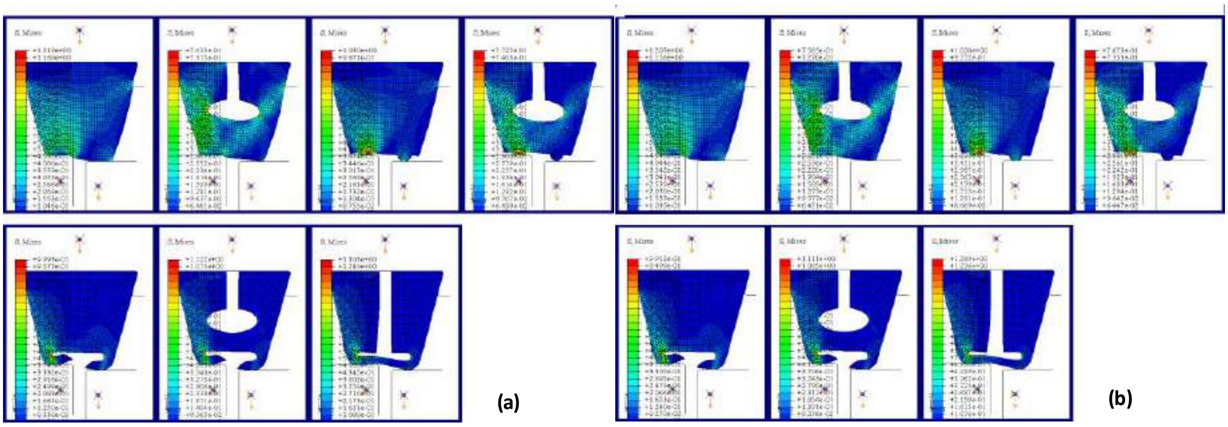


Fig. 5.14. Deformation and Mises stress distribution in basic geometric options obtained by using IG3 properties determined at 110 ^{0}C (strain rate: 50 mm/min) (a) Properties from stress softened IG3 specimens. (b) Properties from IG3 specimens without stress softening.

not influence the performance of backup seal made of IG3. This is further demonstrated by the deformation and von Mises stress fields in seal geometries (Figs. 5.14a-b).

5.3.8.5 Optimising fourteen geometries: Design refinement

Table 5.4 lists the design choice parameters of Options IIA and IIIB (Fig. 4.7) with varying μ in seal-metal interface obtained during design refinements in ABAQUS (3rd Phase, P3) while keeping the seal deformations same as those in Figs. 5.13a-c (μ= 0.5). The results indicate that μ has insignificant effects on the performance of IIA. The very large seal diameter compared to its cross-section and slip (between seal and counterfaces) makes the effects of friction negligible. IIIB shows some (~15%) reduction in compression load as the lips are allowed to bend (because of slip) when μ is reduced from 0.7 to 0.3. As a result of reduced stiffness and increased contact area the contact pressure drops rather drastically (~35%) indicating higher sensitivity, lesser control and greater

Table 5.4. Effects of coefficient of friction on design choice parameters of short-listed seal designs (P3-IG3)

Design choice parameters [a]	Seal geometries and μ					
	IIA (μ= 0.3)	IIA (μ= 0.7)	IIA (μ= 0.9)	IIIB (μ= 0.3)	IIIB (μ= 0.7)	IIIB (μ= 0.9)
Maximum Mises stress (MPa)	1.25419	1.29234	1.30028	2.08881	2.10993	2.19582
Maximum contact pressure (MPa)	0.0480116	0.0515321	0.0528081	0.116127	0.173004	0.180691
Maximum engagement load (N)	128,740	129,447	129,579	82,119.40	97,990.80	98,617.50

[a] Parameters obtained using unaged IG3 properties measured at ET with stress softening (Strain rate: 50 mm/min)

unpredictability in sealing response. This possibility, however, is unlikely under reactor conditions

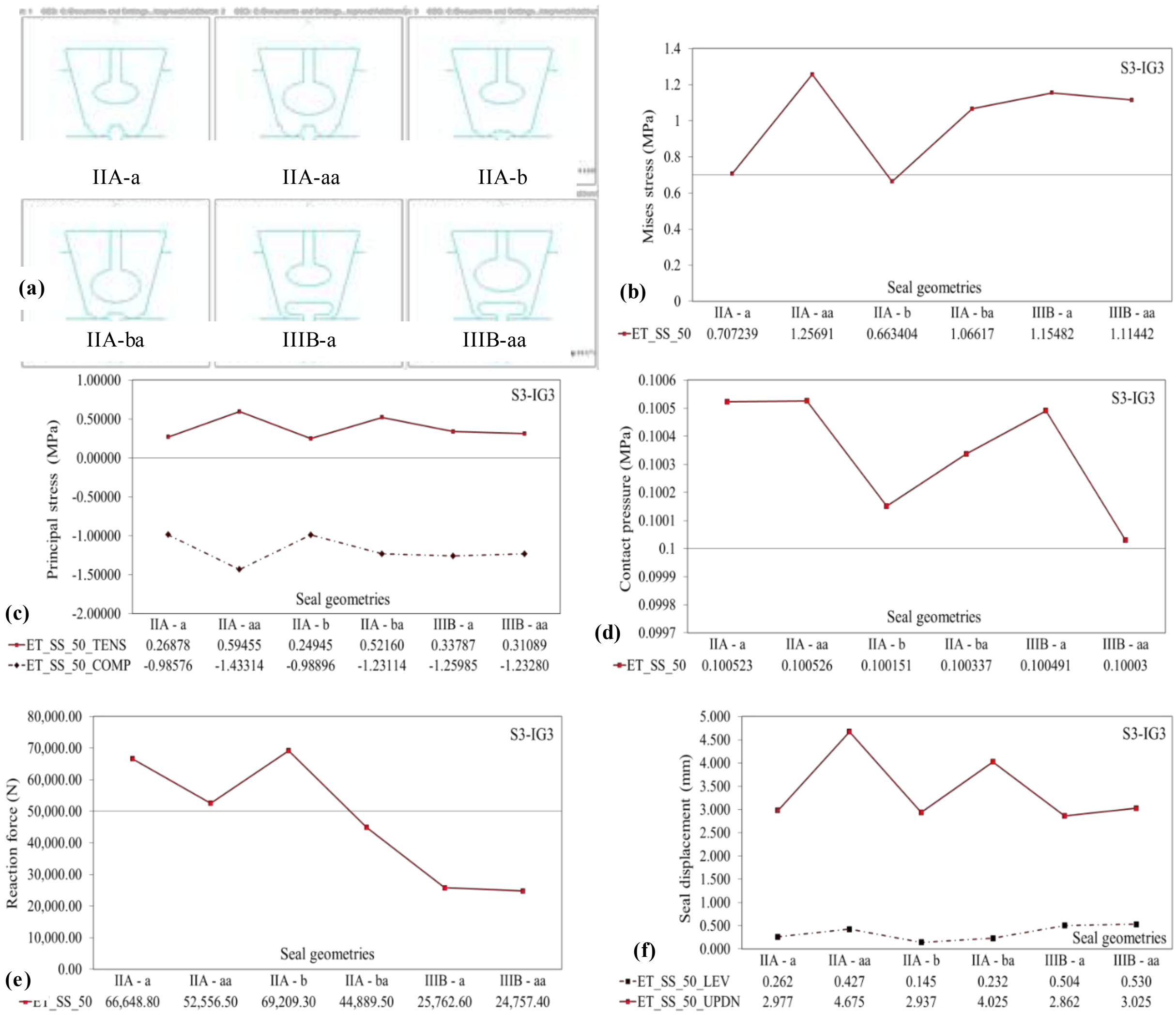


Fig. 5.15. Modifications of shortlisted designs and design choice parameters of the modifications obtained by using stress-softened (SS), unaged IG3 properties determined at 110 ^{0}C (ET, strain rate: 50 mm/min) to attain minimum contact pressure of 0.1 MPa. (a) Geometric modifications of IIA and IIIB. (b) Maximum Mises stress in modified geometries. (c) Maximum tensile principal stress (TENS) and maximum compressive principal stress (COMP) in modified geometries. (d) Maximum contact pressure in modified geometries. (e) Maximum reaction (seal compression) forces in modified geometries. (f) Maximum seal displacement (squeeze) in modified geometries with mating surfaces at level (LEV) and level mismatch of 2 mm (UPDN).

because of unlubricated contact where μ is taken close to 1. Variation of annular gap in the range of 3-7 mm does not influence performance of IIA and IIIB, as boundary conditions remain unchanged.

Figs. 5.15b-f presents the results of parametric studies on 6 geometric variations (Fig. 5.15a) of IIA and IIIB (Fig. 4.8) carried out using stress-softened specimen data in ABAQUS during the 3rd Phase (P3) to attain the threshold pressure of 0.1 MPa considering the level mismatch (2 mm maximum, Fig. 3.17) effects. The peak tensile and compressive principal stresses and seal displacements are plotted along with other parameters for options IIA-a to IIIB-aa where the attributes of the hollowness in seal section (height of annular space, size of ellipse) and lip (thickness) are altered. It is observed that thinning of lips brings down the stress, contact pressure and load considerably (IIIB-a and IIIB-aa). Mises, principal stresses and seal displacement increase with increase of hollowness in seal sections having O-ring contours, while the seal engagement load reduces because of lowering of stiffness of the seal. Options with lip profiles on the main sealing faces are however insensitive to such changes as the trapezoidal seal cross-section (with the extremities of hollowness) acts as a rigid body with respect to the thin lips.

Option IIA-b involves the lowest stress and displacement. The higher requirement of seal compression force is acceptable as the same is well within limit. Option IIIB-aa gives the lowest seal assembly load (at a low peak tensile stress) and highest displacement (with seal mating surfaces at level) amongst all the geometries. These designs from two representative categories are taken up for manufacturing trials.

Options excepting IIA-aa and IIA-ba require displacement of ~ 3mm to achieve the threshold pressure with counterfaces at maximum level mismatch (Fig. 5.15f). This provides the necessary reserve in the designs to absorb increase in seal compression from various requirements; either at the initial stage or during later part of design life. The low levels of engagement load (~69/25 kN) and peak tensile stresses (~ 0.25/0.31 MPa) in the chosen designs indicate that such increases are feasible (Figs. 5.15c and 5.15e).

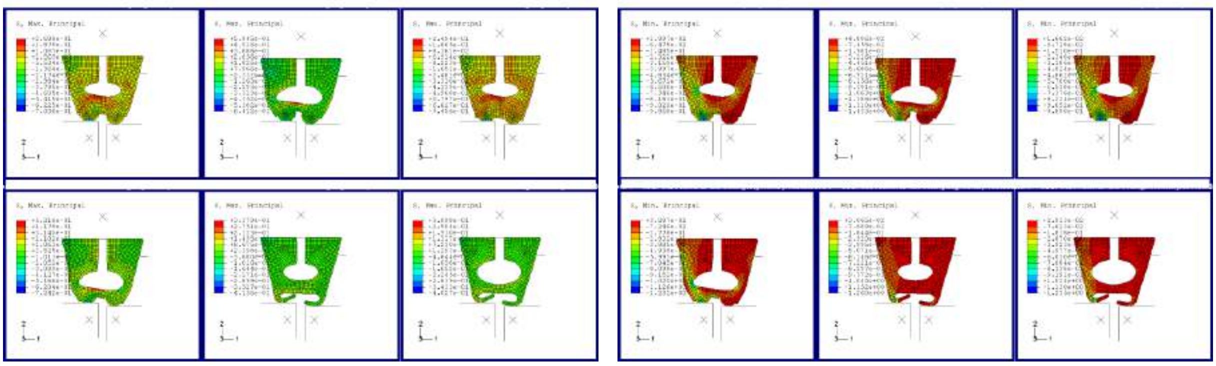


Fig. 5.16. Deformation and principal stress distribution in modifications of IIA and IIIB obtained by using stress-softened IG3 properties determined at 110 ^{0}C (strain rate: 50 mm/min) to attain minimum contact pressure ≥ 0.1 MPa. (a) Contour plots of maximum principal stress. (b) Contour plots of minimum principal stress.

Figs. 5.16a and 5.16b show the deformation and distribution of principal stresses in seal geometries corresponding to conditions indicated in Figs. 5.15a-5.15f. The reason for higher seal displacement in IIA-aa and IIA-ba is evident from the deformed profiles, which indicate tendency of buckling at the lowest seal wall thickness zone because of large elliptical hollowness. Figs. 5.17a and 5.17b show the optimized cross-sectional details of the chosen designs arrived by FEA in ABAQUS during the 3rd Phase (P3) and used for manufacture.

5.3.8.6 Optimising fourteen geometries: Finalization of design

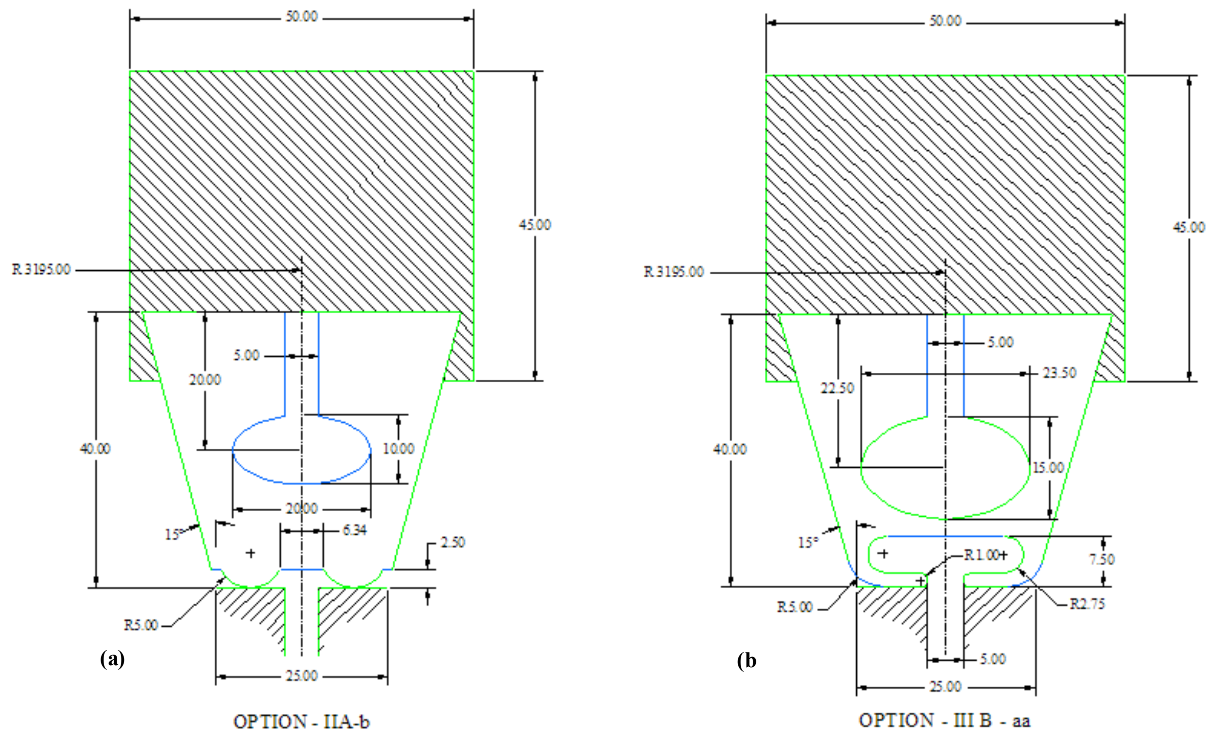


Fig. 5.17. (a) Optimised backup seal cross-section with sectioned double O-ring profiles on the main sealing face. (b) Optimised backup seal cross-section with lips on the main sealing face.

Trials in commercial, variable speed hot feed extruder with autoclave curing showed difficulties in producing the backup seal geometry with lips, shown in Figure 5.17b. The thin lips come in contact with seal during deformation (Fig. 5.16a), which limits their compression and brings in elements of unpredictability in performance prediction. The advantages of IIIB-aa is further offset by larger possibilities of ageing degradation because of higher surface area to volume ratio of the lips and their low thickness (Fig. 5.17b). It has been demonstrated that IG3 will be virtually unaffected by long term ageing in unstrained state. The highest level of stress at the root of the lips (Figs. 5.16a-b), however, suggests greater possibilities of loss of resilience and elasticity during ageing, which could multiply at the open ends of the lips to impair sealing functions.

The design with sectioned double O-ring contour on the main sealing face (Fig. 5.17a) was therefore finalized during the 4th Phase (P4) with modification (identification: IIA- b modified) to enhance the maximum admissible squeeze and eliminate the possible effects of sharp edges and flat surface on the main sealing face.

Fig. 5.18 shows the profile and basic dimensions of IIA-b modified. Table 5.5 lists the design choice parameters of the finalized design corresponding to attainment of a threshold pressure of 0.1 MPa and a seal squeeze of 5 mm, obtained during 4thPhase (P4) from ABAQUS. The peak tensile stress and strain does not exceed 0.43 MPa and 40%, which assures FOS in excess of 3/2 on T_s/E_b at ET in unaged condition. Optimization based on attainment of threshold pressure $\geq$ 0.1 MPa produces

Table 5.5. Effects of specimen test data input (to finite element analysis) on design choice parameters of IIA-b modified (P4-IG3)

Design choice parameters		**Tension/compression data at 110 ^{0}C** *(50 mm/min)*					**RT tension/compression data** *(50 mm/min)*		
		From unaged specimens		From aged (16 weeks/170^{0}C) specimens		From unaged specimen	From unaged specimens		
		Up to 50/20 % strain				Up to 50/40 % strain	Up to 50/40 % strain	Up to 50/20 % strain	Only tension (50%) data
Maximum principal stress/strain (MPa)	Level	0.03/0.03	0.43/ 0.40	0.03/0.03	0.45/0.40	0.43/0.40	0.39/0.40	0.41/0.40	0.45/0.40
	UPDN	0.24/ 0.20	0.42/0.38	0.25/0.20	0.44/0.38	0.43/0.38	0.38/0.38	0.41/0.38	0.44/0.38
Minimum principal stress/strain (MPa)	Level	-0.11/ -0.03	-1.75/ -0.41	-0.11/-0.03	-1.84/ -0.41	-1.78/-0.41	-1.63/-0.41	-1.75/-0.41	-1.91/-0.41
	UPDN	-0.92/-0.20	-1.67/ -0.39	-0.89/-0.21	-1.76/-0.39	-1.70/-0.39	-1.56/-0.39	-1.67/-0.39	-1.82/-0.39
Minimum contact pressure (MPa)	Level	0.10	1.07	0.11	1.12	1.08	0.98	1.04	1.12
	UPDN	0.10	0.67	0.16	0.70	0.68	0.61	0.65	0.70
Maximum squeeze force (kN)	Level	4.1	196.3	4.3	206.1	198.59	179.52	191.14	205.96
	UPDN	69.7	153.5	73.2	161.2	155.29	140.35	149.41	160.97
Maximum seal squeeze (mm)	Level	0.16	5.00	0.16	5.00	5.00	5.00	5.00	5.00
	UPDN	2.93	5.00	2.93	5.00	5.00	5.00	5.00	5.00

highest load/stress during maximum level mismatch between seal mating surfaces. When a fixed squeeze is given to the seal (as would happen during actual operation in reactor), these conditions are generated during contact with level surfaces without any mismatch (Table 5.5).

The LRP seal compression load of 196.3 kN, however, goes way beyond the limit of 100

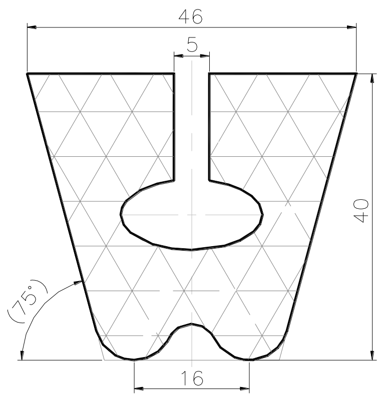


Fig. 5.18. Finalised design of backup seal used in INDIAN- SFR rotatable plug: option IIA-b modified.

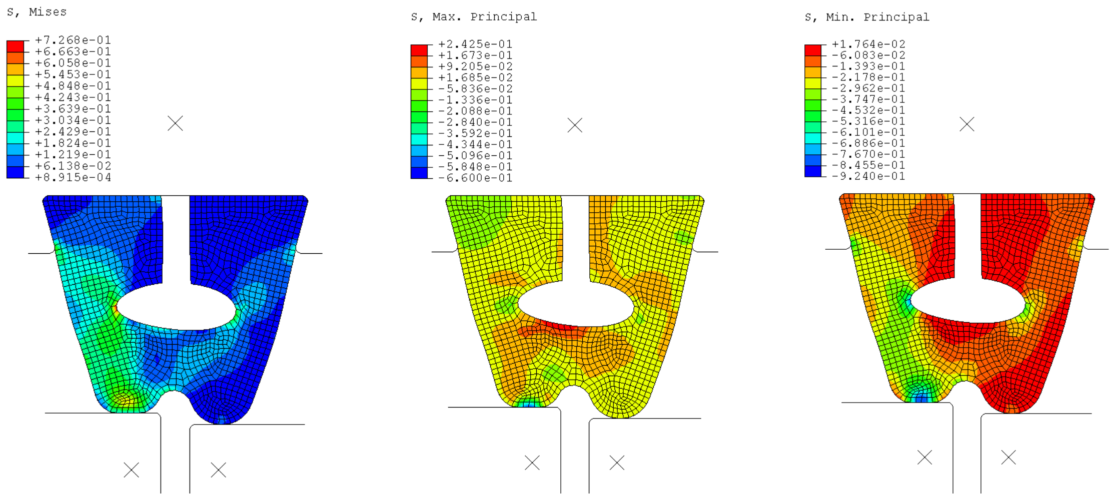


Fig. 5.19. Deformation, Mises and principal stress distribution in IIA-b modified obtained by using unaged, stress- softened, IG3 properties determined at 110 ^{0}C (strain rate: 50 mm/ min; input tension/ compression strain range: 50/20%) to attain threshold pressure ≥ 0.1 MPa.

kN. The remaining compression of ~1/0.5 mm on the upper/lower sealing counterfaces at the end of design life, considering an initial overall squeeze of 5 mm (at maximum level mismatch) and CS losses (80%), indicate substantial lowering/increase of compression-load/FOS. However, the original values are retained as a conservative measure to absorb possible effects of ageing on stress/strain/stiffness/load, tolerances and CS compensation.

Figs. 5.19 and 5.20 show deformation and stress contour plots in IIA-b modified based on attainment of threshold pressure of 0.1 MPa and a squeeze of 5 mm. Fig. 5.21 shows the contact pressure distribution at the interfaces of seal and its mating surfaces (squeeze: 5mm).

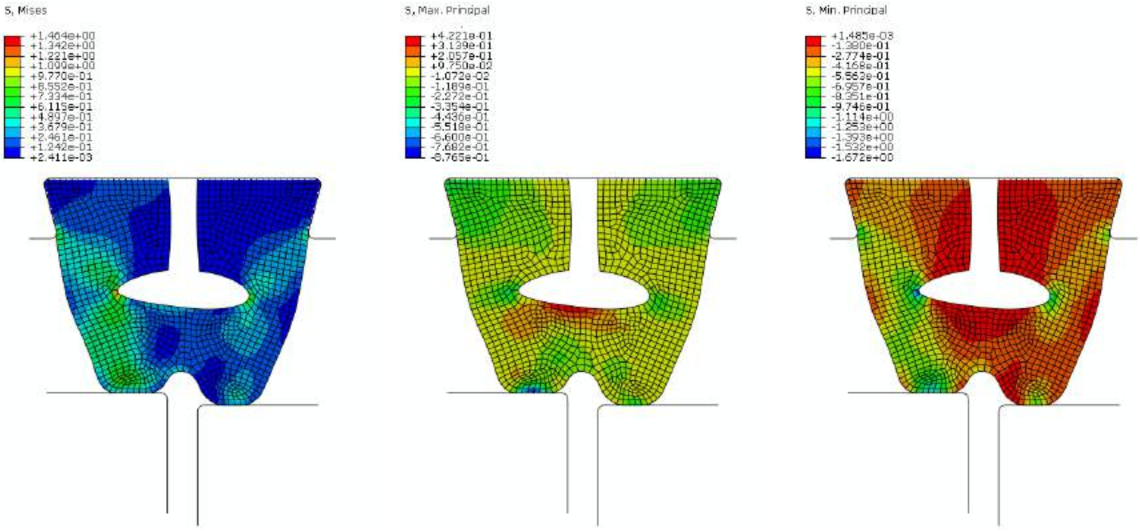


Fig. 5.20. Deformation, Mises and principal stress distribution in IIA-b modified obtained by using unaged, stress-softened, IG3 properties determined at 110 ^{0}C (strain rate: 50 mm/ min; input tension/ compression strain range: 50/20%) for 5 mm squeeze.

5.3.8.7 Ascertaining ten year of life by finite element analysis using aged material data

The negligible effects of IG3 specimen ageing (16 weeks/170 ^{0}C) on the design choice parameters of IIA-b modified are shown in Table 5.5. Use of specimen properties after ageing at other temperatures, strain rate and durations gives similar results.

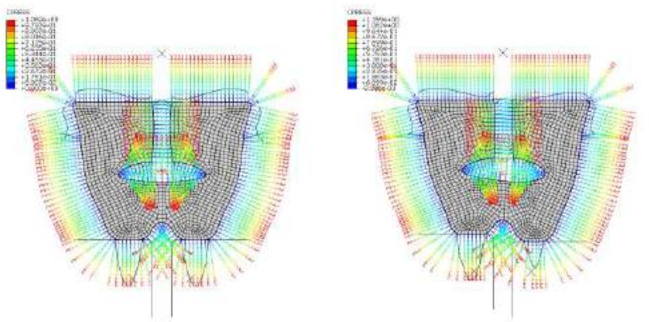


Fig.5.21. Contact pressure distribution at seal and counterfaces at 5 mm squeeze.

The variations of T_s, E_b and M_{50} with ageing at different temperatures and the near identical stress-elongation behaviour of IG3 samples (Fig. 5.22) during ageing in air at 170^{0}C substantiate these findings which signify that FOS, stiffness and compression load of the unaged seal will remain practically unaltered by ageing during design life. The close similarity of test curves (Fig. 5.22) and FEA results (Table 5.5) is also attributed to moulding of all unaged/aged specimens from one blended batch of compound (IG3- A) and testing in the same machine, which minimized batch-to-batch material property deviations and repeatability errors during intra-laboratory measurements.

5.3.8.8 Ascertaining ten year of life by safety assessment, validation and design

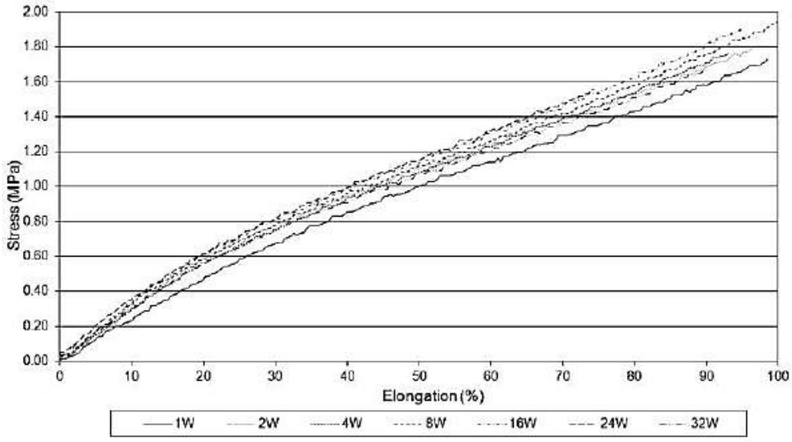


Fig. 5.22. Uniaxial tensile stress in aged (at 170 ^{0}C), unconditioned, IG3 specimens measured at 110 ^{0}C (strain rate: 500 mm/ min) as functions of elongation and ageing duration in weeks (W).

As the temperature at backup seal location is 90 ^{0}C normally, the FOS should be more than 4/3 because of higher T_s/E_b at that temperature. $T_s/E_b/M_{100}$ variations of up to ± 15/20/25% could be experienced between batches of seal compound. The additional tolerance of ± 10% given to include the extrusion effects of backup seal (Table 4.2) takes the maximum overall variations to ~ ± 25/30/35%. The most unfavourable combination of variations (T_s and E_b reduced by maximum amount accompanied by marginal increase of M_{50}) indicates reduced FOS (T_s/E_b more than 3/2) at 90 ^{0}C.

The FEA results from ABAQUS using aged material data after the 4th Phase ascertain ten year of life for backup seal made of IG3 (S-4), with the initial FOS of 2 on T_s and E_b assigned to CDA pressure surge (2.1 MPa) at any point of time during design life and additional envisaged degradations because of strain in seal (maximum: 35%) during normal operation.

Experimental verification of 1m diameter test seal with the finalized design (IIA-b modified) under simulated service conditions (Fig. 5.7) indicate compression load of ~50 kN for LRP seal (average squeeze of 2.7 mm; counterfaces at level) which is ~2.5 times lower than that (~125 kN) obtained by FEA. The initial assembly load of 11.2 kN for the test seal (average squeeze: 5.1 mm; counterfaces at maximum level mismatch) indicates squeeze load requirements of ~72/47 kN for the LRP/SRP seals which are about half of the corresponding computed values obtained by using unaged specimen properties at ET (Table 5.5). The loads were measured at RT owing to functional limitation of the load cells at ET. The design choice parameters were therefore determined using IG3 RT/ET properties with various combinations of strains (Table 5.5).

For every 20% reduction of input specimen-compression-strain-range to the FEA code, the RT seal compression load is overestimated by ~7% (Table 5.5). The overestimations could be attributed to numerical extrapolations (in compression zone) beyond the measured strain range using Mooney- Rivlin material model in ABAQUS which could be even more for higher order constitutive relation such as Ogden. The possible reason for higher estimated squeeze load with ET data (tension/compression: 50/40%) is the higher stiffness of stress-elongation curve compared to that at RT (Fig. 5.23), which increases the bending resistance of seal under deformation.

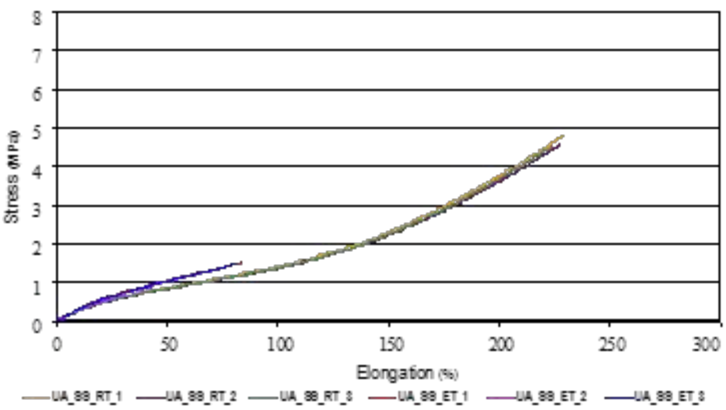


Fig. 5.23. Stress- elongation behaviour of unaged (UA), stress softened (SS), IG3 tensile specimens measured at RT and 110 ^{0}C (ET) at strain rate of 50 mm/min.

The large difference between computed (~140 kN) and tested (~72 kN) loads could be explained. Tolerances on the basic dimensions (Fig. 5.18; inflatable seal tolerances, Figs. 5.2 and 5.4 could give some idea) of IIA-b modified allow some reduction in manufactured dimensions and squeeze load. The most unfavourable variations of $T_s/E_b/M_{50}$ (between specimens used for FEA and the test seal) do not appear to induce significant change in the calculated loads. Polymeric chain scission, reduction and redistribution of molecular weight and introduction of directionalities of properties (or anisotropy, i.e. stronger along extrusion length or seal circumference) in green elastomer during extrusion, however, suggest some reduction in material stiffness compared to those of moulded specimens used for FEA (Figs. 5.11, 5.22-5.23). The overrating of compression load from these counts could be anywhere between 25-50% of the actual. The other part of deviation is attributed to FEA approximations, which include inaccuracies related to the material model, meshing/element/solution schemes and differences between the modelled and real contact/friction conditions.

The FOS increases irrespective of whether the overestimations are caused by dimensional reduction, batch-to-batch and production variations or numerical approximations. Ideally, an increase of FOS on T_s/E_b at 90 ^{0}C to more than 5/4 is envisaged (squeeze: 5 mm). Conservatively, FOS of more than 4/3 (or more than 3/2 at ET) is arrived by keeping 50% of overestimations to absorb dimensional/material variations, which are unavoidable. This is consistent with the uniform safety margin of 2 at ET arrived during IG3 development, which is preserved at the end of design life to absorb possible additional ageing effects under strain and accidental pressure surge. The gains in squeeze load and FOS (because of CS) are retained for CS compensation and additional tolerances such as positional accuracy of seal drive.

The optimization study gives a very clear indication that the safety margin could be determined more accurately and enhanced further along with improvements in numerical approximation by using properties of specimens extracted from seal. The results of FEA with unaged and aged material data coupled with validation and an overall assessment by design confirms a minimum life of 10 y for back backup seal made of S- 4 in INDIAN- SFR RPs.

5.3.8.9 Aspects of anisotropy and absorbing the possible effects

The hyperelastic material models or constitutive relations based on stretch ratios or strain invariants assume isotropy throughout the deformation process under isothermal condition. Possible effects from anisotropy or directionality of properties introduced by loading (different Mullin's effect in different directions), manufacturing (moulding, extrusion etc.) or material structure (reinforcement etc.) are not reflected in computation of stress- strain by FEA and hence included in the margin of safety or FOS instead for simplification. Structural anisotropy is not applicable for the unification by standardisation pursued in this dissertation because of the adoption of non- reinforced common generic elastomer (fluoroelastomer) across categories. Directionalities from Mullin's effect is expected to be insignificant for elastomeric formulations of static seals (such as S- 4) because of low filler content which minimises stress- softening and hence the directionalities therefrom.

Spatial orientations of macromolecular chains in unstrained and unprocessed, non- reinforced rubber could be assumed as a sphere which explains the initial isotropic behaviour. The origin of stress for unfilled or low- filled rubber is essentially based on statistical thermodynamics with integration of change in entropy over the sphere considering each and every chain. There had been efforts to simplify this process of integration by choosing finite number of representative directions (such as 3 and 8) which follows the principal directions of gradient deformation tensor in order to satisfy the conditions of material isotropy. Progressive introduction of anisotropy was an extension of this process where direction specific phenomenological constitutive models were envisaged. Addressing the issue of capturing non- linear hyperelastic behaviour with considerations of further details (such as type and intensity of loading) is evolving for more realistic representation of product behaviour. In this context, the process of design simplification and unification by standardisation calls for an evolutionary process of maximising the utilisation of existing by including the effects in FOS while allowing the quantification process to grow in maturity, similar to the design simplification and standardisation of the Gen III reactors towards the Gen IV SFR or peaking of CO_2emission by 2020 while allowing space for the infrastructure, knowhow and resources (funding, material etc.) to grow and consolidate across the globe.

Unifying the material module (or the 1st element of design) as cornerstone by standardisation with 1 fluoroelastomer formulation and its variations requires quantification of directionality (from manufacture) in static elastomeric sealing of cover gas for MOX fuelled FBRs as the first step. Processing introduces orientation (commonly referred to as ‘grain’ of rubber) in the random macromolecular structure of non- reinforced, initially isotropic elastomer represented by a sphere and then a few directions with detailing of loading etc. and manufacture as subsequent steps. Differing tensile properties along and across grains is manifestation of anisotropy from processing which could be non- existent to countable in the final product depending on the choice of manufacturing process and elastomer. An indication on the range of deviations in T_s (for 50% maximum strain) could be obtained from the range of deviations in two representative generic elastomers during calendaring i.e. natural rubber (30% for 150% stretch) and SBR (10% for 300% stretch). Absorbing this anisotropy in FOS (of 2) has been illustrated for S- 4 (Section 5.3.8.8).

Increased filler content to 20- 30 phr (usually) for dynamic sealing application (e.g. inflatable seal, S-1) introduces additional anisotropy from directional deviations of Mullin's effect because of increasing polymer- filler interactions and secondary, reversible bonds therefrom which breaks (disappears) with repeated loading (or increasing temperature) resulting in reducing stiffness etc. Quantification of this anisotropy is left for future research (different- direction specimen extraction from seal) with possible deviations absorbed in the increased FOS (> 2) envisaged using improved FKM compound and better manufacturing process.

5.3.9 Combining compression set, stress-strain and HF release results

The predictive studies involving Arrhenius, WLF and FEA methodologies ascertain 10 y of life for backup seal made of S- 4 from all counts and including CDA with a CS of 80 % as failure criterion along with T_s as well as E_b of unaged material (determined at 110 ^{0}C) divided by a FOS of 2 which is consistent with the life estimation based on HF release.

5.4 Interim Unification with Ten Years of Seal Life Towards Complete Unification

The qualification of S- 1 and S-4 from most of the perceivable aspects of nuclear use provides sound basis for evaluating an interim unification scheme for synchronized replacement of cover gas seals with minimum life of 10 y in continuation of the first take in Section 3.12 and towards complete unification (Chapters 6 and 7) for 1 synchronised replacement in 60 y as the objective of this thesis.

5.5 Interim Unification Scheme Involving S-1 and S-4[45, 139]

Identical ingredients and curing system of S- 1 (Table 3.11) and S- 4 (Table 4.8) suggests sodium aerosol compatibility of S- 4, also substantiated by sodium vapour exposure data of Viton® in nitrogen environment. Insignificance of HF release from inflatable (S- 1) and backup (S- 4) seals of INDIAN- SFR RP during 10 y life individually, is extended to other cover gas seals individually because of much lower weight of the later (Sections 3.7- 3.8, 5.3.4). Mooney-Rivlin constitutive model, plane strain condition as first approximation and axisymmetric model for finalisation are applicable to all the static and dynamic cover gas seals of INDIAN- SFR during normal operation and fuel handling conditions (strain ≤ 50 %), as they don't use liquid lubricant.

Screw extrusion and steam curing in an autoclave are followed by post-curing and end-joining. End-joining involves holding the open ends of seal (cut at a 45° angle) under heat and pressure. This method was used for the production of fluoroelastomer backup seals (S- 4), and is applicable for other large-diameter elastomeric seals of INDIAN- SFR. This puts all the large diameter static elastomeric seals of INDIAN- SFR on a single foundation of material formulation, FEA procedure and manufacturing technique developed and used for the RP backup seals. Multiple end joints result from this production process for all the large diameter static elastomeric cover gas seals, listed in Table 3.1. The bisphenol-cured, Viton® A-401C-based compound developed for backup seal (S- 4) is also appropriate for small-diameter, moulded, static elastomeric ring seals (mainly O-rings) of the reactor assembly, as their operating requirements (Table 3.1) do not exceed those of backup seal (Table 3.3) and S-4 is suitable for both extrusion and moulding (Section 3.9.3), being based on Viton®A-401C. The compound provides higher safety margin and promises longer life for the smaller diameter moulded, static seals as the seal chord diameter and squeeze are lower (because of lower fabrication and assembly tolerance stack-ups) and the uniformity of cure as well as other properties are better compared to the large diameter seals.

The suitability of Viton A®-401C based inflatable seal compound for the dynamic O-ring/V-ring/ Lip seal applications of INDIAN- SFR cover gas has been indicated in Section 3.12. The cover gas elastomeric sealing applications of MOX fuelled FBRs (transiting towards SFR), modelled by the barriers of INDIAN- SFR cover gas, could therefore be represented by a combination of 4 Viton® A-401C based FKM compound formulations as cornerstone (S- 1, S-4 and their mouldable versions), 1 FEA approach as facilitator and 2 manufacturing techniques i.e. extrusion and moulding.

Multiplicative gains are immediately evident if effort, time and cost involved in complete development, characterisation and qualification (typical of nuclear seals) of these 4 compounds are compared with the 21 compounds of FBTR (including inflatable seal but excluding the variations other than O-rings, Table 2.2). The FEA tension/compression data generation alone on five specimens (for each test) at RT and seal operating temperature (with and without stress softening at various shear rates), withdrawn periodically during long-term ageing, provides an indication about the potential savings in R & D endeavour for complete unification. Another simple evaluation shows that by using FEA (and other novel R & D approaches indicated in Chapters 2 and 3), basic developments of inflatable and backup seals (up to 2m diameter) were completed within ~5 y (FBTR: 3 y) and ~ 2.5 y whereas similar developments in other FBRs (without FEA) took a decade or more on average.

5.6 Maximising Inflatable Seal Life with S-2 by Enhanced Factor of Safety and Better Manufacturing

The aim of attaining 1 synchronised TS- seal- replacement in 60y necessitates maximising CS resistance, FOS on T_s- E_b and further reduction of collective HF emission (by γ dose) from TS barriers which suggest the need for fluoroelastomer of better grade, also supported by extrusion difficulties (S- 1) and limitations from a FOS od 2 (S- 1, S- 4) indicated in Section 3.9.4 and this Chapter. As RP inflatable seal is pivotal to unification (Section 3.4), the search for such a betterment should start from the maximisation of FOS where cold feed extrusion and continuous cure (MW, HAT) provide an avenue for better quality seals with superior cure- uniformity and properties along seal circumference, automation (better uniformity across production lots) and 1 end-joint per seal.

5.6.1 Transition from DMF3/Comp #1 (S- 1) to Comp # 9 (S- 2) through extrusion experiments [81]

Trials of cold feed extrusion (Section 4.6.2) for inflatable seal (Fig. 4.1) using S- 1 were marked by seal profiles of low T_s and hardness and high level of porosity along with dimensional deviations and coarse seal surface. This reflected typical bisphenol-cured-fluoroelastomer-behaviour vulcanised at atmospheric pressure without post-cure. Apart from driving out the volatiles, post-cure also contributes to a thermally induced bond-breaking and bond-making process, analogous to annealing, which relieves the stress points in the cross linked network and produces a more stable network while maintaining the overall network (crosslink) density essentially unchanged. These two parallel processes during post-cure result in a vulcanized product, free of porosity and displaying improved properties, finish and dimensional stability. Mould- autoclave pressure squeezes the voids and bubbles out of uncured rubber parts and ensures fast compaction of pores which are important for fluoroelastomers considering their rather slow relaxation and recovery from strain.

Comp. #1 (S- 1) specimens, like other bisphenol cured fluorocarbon rubbers, requires standard post-cure (24 h/232 ^{0}C) to achieve the target properties (Tables 4.11- 4.12). Production experience indicates that pressure-less continuous curing in HAT takes longer time (as much as 10×t'90 depending on profile thickness) to attain equivalent level of cure and strength of a press-cured specimen. A very slow line speed of 0.5 m/min, at the most could simulate the curing conditions of a thick, press-cured specimen or profile. The line speeds had to be kept (Section 4.6.2) at or above 1m/min during the trials (after initial explorations with a speed of 0.5 m/min) to control the problems related to high viscosity and die-swell because of low shear rate. The HAT temperature was raised up to 270 ^{0}C during the extrusion trials to bring in some of the post-curing effects. This, however, led to deformation and swelling of profile and could not be corrected. HAT temperatures were kept below 220 ^{0}C during subsequent trials.

Refining Comp # 1 (S- 1) to Comp #2 with the addition of 1 phr of processing aid (carnauba wax, Table 4.13) improved flow but the quality of extrusion did not show notable change because of uncompacted pores from water vapour and other volatiles such as carbon dioxide. The typical limitations of bisphenol cured fluoroelastomers comprising of the requirement of post- cure (typical: 232 ^{0}C/ 24 h) to achieve the desired physico- mechanical properties (Table 4.11) and generation of large amount of water vapour (~ 1.5 % of product weight) during cure[29] - resulting in uncompacted pores, lower strength- hardness and surface coarseness during atmospheric pressure, continuous cure (no provision for post- cure) -were manifested in all the subsequent trials based on Comps. # 3-4 (based on Viton®A-401C, Table 4.13) and Comps. #5-6 (based on Viton® A-201C) to varied extent along with higher viscosity, backpressure and dimensional deviations related to slower screw speed and thin- seal- section.

The paradigm- shift to peroxide cured, Viton® GBL-600S(Comp. # 7) made of APA was driven by the advantages of peroxide- cure (minimum generation of water vapour, minimum double-bonds in cured product) and APA which enabled faster cure and easier flow (lower viscosity, shear heating, backpressure and dimensional deviations) with lesser amount of curative to achieve the properties with minimum post- cure, while removing the traditional problems of peroxide curing in air with non- APA Viton® (Viton® GLT, since 1976)- outlined in Sections 4.3.2 and 4.6.2.

The low reinforcing filler, MT black (N990), of typical formulation was replaced by semi reinforcing furnace (SRF) black (N762) in comp. #7 to ensure better strength and dimensional stability of the end product. The metal oxide used in typical formulation (ZnO) was retained as acid acceptor considering its ability to give better thermal stability and long-term heat resistance. The quantity of high purity ZnO was reduced to 2 phr to minimize the possible generation of water vapour. Also, a combination of two processing aids namely, Struktol® WS280 (Silicone organic compound) and VPA No. 2 (Ricebran wax) were introduced in comp. #7 along with stearic acid coating on ZnO particles to ensure improved flow, lower shear heating, minimum die swell and smoothness in the extruded profile. The type and quantity of peroxide curing agent used in the typical formulation (Varox® DBPH-50, a 45% active dispersion of 2,5-dimethyl-2,5-di-(t-butylperoxy) hexane) was retained in comp. #7. The content of DiakTM 7 was slightly modified in Comp. #7. Comp.#7 showed conformance to the target properties (Table 4.11) during compounding trials.

[29] Metal oxide acting as scavenger or acid acceptor to react with HF, generated by dehydrofluorination, and producing water.

Porosities were reduced considerably and some stickiness on cured seal surface was observed. Dimensional deviations and roughness in the extruded seal profile even after temperature profile corrections in the extruder raised some fundamental questions. The slower shear rate, increased viscosity and higher back pressure from slow rotational speed of screw (1- 15 rpm; 2- 5 rpm for best results) during extrusion trials of INDIAN- SFR RP inflatable seal (Fig. 4.1) reduced the inherent advantages of peroxide cured, APA-based- Viton® considerably which is tailored to give best performance at high shear rate (1000 s^{-1} or more), appropriate for commercial mass production through extrusion and injection moulding. Higher die resistance (because of narrow annular opening in extrusion die) led to higher back pressure at extruder head, which amplified the problem and resulted in dimensional deviations as well as lack of finish because of increased dependence of extrusion rate on the rheology of compound which is non-Newtonian and less predictable.

Table 5.6. Properties of Comp. #9 (S- 2)

Stock Properties (ASTM D5289-95; ASTM D1646-03a)
MDR 2000 at 177^{0}C, 0.5^0 arc, 100cpm, 10 min clock
M_L: 1.1 dN.m; M_N: 30.4 dN.m; t_s2: 0.6 min; t' 90: 1.6 min; Mooney viscosity:46 ML 1+10 @ 121^{0}C.
Vulcanizate Properties (ASTM D3182-89 (Reapproved 2001); ASTM D412 – 98a (Reapproved 2002) e1; ASTM D 2240 - 05)
Determined on unaged specimens at RT (press cure at 177^{0}C, 7 min; no post cure)
M_{100}: 3.3 MPa; T_s: 13.7 MPa; E_b: 343%; Hardness: 69 ^{0}Shore A.

Compound #8 (Table 4.13), a 50:50 blend formulation of Viton®600S and 200S with intermediate viscosity between the two and aimed at reducing die swell as well as back pressure, showed conformance to target properties (Table 4.11) during compounding trials some of which are shown in Table 4.15. Flow and extrudability of comp. #8 was better than that of comp. #7 and stable cross-sections with low porosity could be achieved. Some stickiness, however, was still evident on extruded seal surfaces.

Comp #9 (S- 2, Table 4.13), a refinement of Comp #8 by replacing 2 phr curing agent (Varox®DBPH-50) with 1 phr of Luperco®101-XL, showed conformance to target properties (Table 4.11) in test specimens with 7 min of press cure at 177 ^{0}C and without any post-cure (Table 5.6). Porosities and stickiness were eliminated and dimensional deviations were within limit.

~2m diameter test inflatable seal was successfully produced by some marginal factory modifications on comp. #9 with a conveyor line speed of 1 m/min and HAT temperature of 190 ^{0}C. The approximate temperature profile maintained during seal production are, i) feed zone: 60 ^{0}C, ii) head zone: 90 ^{0}C, iii) screw zone: 60 ^{0}C and, iv) zone 1/2/: 70–80 ^{0}C.

5.6.2 Ascertaining one replacement of inflatable seal made of S-2 during reactor life by data extrapolation and design [10, 37, 81, 126, 145]

Comp. #9 or S- 2promises significant improvement of seal performance in reactor in terms of fluid resistance, permeation resistance, property retention at elevated temperature, radiation resistance, life and safety. Releases of traces of HF during seal operation (0.123g from γ radiation during 10 y) are comparable for comp. #1 (S-1) and comp.#9 (S- 2). Comp. #9 (S-2) is likely to be more resistant to sodium aerosol at seal operating temperature because of lower level of unsaturation from peroxide cure and higher fluorine content (~ 68 wt.% because of TFE) as well as lower level of processing aids in S- 2 compared to S- 1. The absence of activator (calcium hydroxide) and lower level of metal oxide in S- 2could also contribute toward an improvement of aerosol resistance which, however, is to be ascertained experimentally. Superior permeation resistance is expected from comp. #9 (S-2) because of established superiority of APA grades over the non-APA grades in this respect.

Product literature and other sources indicates that typical APA formulations based on Viton® GBL-200S, Viton® GBL-600S and the 50/50 blend, on an average, offer ~50% higher E_b, ~35% lower modulus (M_{100}) and slightly improved T_s (~7%) over corresponding values of comp. #1 (S-1) without any post-cure (measured at RT) which is substantiated by Table 5.6. The much lower level of ionic end groups in peroxide cured APA Viton® grades result in minimal ionic interaction in the cured compound at RT which is reflected in reduced modulus and enhanced elongation. Non-APA older grades (such as S- 1 and S- 4) loose considerable part of RT T_s (~ 75%) because of substantial reduction of the RT- ionic- interactions at ET. E_b reduces by two- third on average. It was shown that a 90^{0} Shore A hardness, Viton® GBL-S based compound retains 80% of RT modulus

(M_{50}), 42% of RT T_s and 40% of RT E_b at 200 ^{0}C as ionic interactions have minimal contributions at RT. The retentions are expected to be much higher for the representative cover gas seal temperature of MOX fuelled FBRs i.e. 120 ^{0}C.

M_{50} values for Comp. #1 (S- 1), determined at a test speed of 500 mm/min (specimen reference: ASTM D412 Die C) and at test temperatures of 50 ^{0}C (M_{50}: 1.95 MPa), 80 ^{0}C (M_{50}: 2.04 MPa), 100^{0}C (M_{50}: 2.06 MPa) and 120 ^{0}C (M_{50}: 2.02 MPa) do not show trends of major variations with respect to the RT value (M_{50}: 2.25 MPa). Absence of Joule- Gough effect is evident here because of much higher filler content (20 phr) in S- 1 compared to S- 4 (2 phr). Also, the invariability of M_{50} (stress at 50% strain) with increasing temperature substantiates the observations on cross-link driven domain of stress-strain, virtually independent of the uncertainties of ionic interactions, in an elastomer where a large part of RT strength is arrived from ionic interactions. The cross-link driven domain of stress- strain has also been addressed in another work as part of this thesis in order to identify the usable rage of stress- strain for design by S- 4 (1.4 MPa- 85%) and S-1 (3.26 MPa- 80%) at ET on which the FOS is applied to define the limits of principal true stress- strain. Similar observation could be drawn for the Viton® GBL-S grades.

The unchanged maximum tensile strain (40%) in INDIAN- SFR RP inflatable seal, irrespective of operating temperature or compound, indicates that the reduced modulus in S- 2 (compared to S- 1) would be reflected in a corresponding reduction in stress and gain in FOS for the same T_s at 120 ^{0}C. This gain is multiplied due to higher retention of T_s at ET. Rough estimations suggest a FOS of ≥ 4 from the cumulative gains for Comp. # (S- 2, Table 4.13). Similar assessment indicates that comp. #9 (S- 2) is likely to retain at least 50% of its RT E_b (343%, Table 5.6) at 120^{0}C which is more than double compared to corresponding value of Comp. #1 (80%, Table 4.12) indicating a two-fold gain in FOS in terms of E_b. Comparing these values with the results of literature data extrapolation (Section 5.1) gives a very clear indication that S- 2with its substantially higher FOS (> 4) could absorb much larger variations of modulus, T_S and E_b or a γ dose of 10 kGy at least at 120 ^{0}Cunder synergistic ageing and rubbing i.e. 50y of life without loosing elastomeric function.

This is further substantiated by the fact that peroxide cured Viton® made of APA is designed to retain at least 100 % E_b at typical moulding temperature (160- 190 ^{0}C) as a general thumb rule [189] in order to withstand the demoulding stress- strain for defect free product, which indicates a FOS of > 4 on E_b (most important failure parameter limiting life for inflatable seal) considering the design seal temperature (120 ^{0}C) and an E_b of 80% for the bisphenol cured S- 1 at that temperature.

The gains with comp. #9 (S- 2) make it a contender for the IFTM inflatable seals (~ 4m diameter) of INDIAN- SFR(Fig. 3.10), exposed to150 ^{0}C during fuel handling and in contact with stainless steel The suitability of APA based Viton® in such condition is substantiated by the development of shipping package O-rings which are in contact with stainless steel and operate at a design temperature of 149 ^{0}C with design life of 10 y. Viton® GLT-S grades have been adopted for the seals and are being evaluated for long term ageing resistance and life [69- 71].

Considering the fact that all these variations of operating requirements of INDIAN- SFR cover gas seals are normalised to a set of representative operating requirements (10 y ---- 70 kPa ---- 120 ^{0}C ---- 23 mGy/ h ---- 1000 N/ m-seal-length ---- 100 km ---- 10^{-3} scc/ s/ m- seal- length), the FOS of > 4 (and the life maximisation) is applicable to other cover gas seals also if S- 2 is tailored.

The FOS (> 4) on T_s and E_b, determined at design seal operating temperature, is apportioned between batch-to-batch variability (T_s: 15%; E_b: 20%), intra-laboratory repeatability errors (Ts: ~6%; E_b: ~9%; Hardness: ~3%), one- fourth and one- third envisaged drops in T_s and E_b following 10 kGy of γ dose in air at 120 ^{0}C without mist contact (Section 5.1), ± 10 % variations from moulded specimens to the extruded seals (Tables 4.2 and 4.11), possible effects of sodium aerosol and strain corresponding to 10 kGy of cumulative γ dose at 120 ^{0}C corresponding to 50 y life, supported by virtually unchanged T_s and E_b under thermo- oxidative ageing as demonstrated by S- 4. The assumption of equivalence of S-1, S- 4 and S- 2 here ignores the envisaged enhancement of T_s (20%), E_b (10%) and FOS (as demonstrated by 32 weeks of accelerated ageing of S- 4, Figs. 5.8a- g, Section 5.3.2.3) as a measure of conservatism. The anticipated release of HF because of thermal ageing during seal operation in reactor is negligible and the same from cumulative γ dose (10 kGy) during

50 y should not exceed 0.4 g (per seal) considering 100 ^{0}C as the usual operating temperature of inflatable seals for INDIAN- SFR RP. Specimen test results indicate that the FOS on T_s and E_b goes above 6 considering the usual operating temperatures of 90 ^{0}Cand 100 ^{0}C for backup and inflatable seals respectively. This provides a definite indication about the feasibility of attaining 50 y of life for the large diameter extruded static and dynamic categories using non- reinforced fluoroelastomers based on anticipated changes in T_s and E_b.

The feasibility of 50 y life for the cover gas seals, assuming equivalence of S-1, S-2 and S-4 (and varying only FOS) and taking S- 2 as the cornerstone, is limited by the CS for static seals, possible variabilities from the combined effects of different fillers and additives in S-2, S-1 and S- 4 (Tables 3.11, 4.12- 4.13) and the necessity for experimental evaluation of cumulative effects from HF release by γ dose from the ~ 700 m long periphery of cover gas seals during reactor operation.

Maximising the feasibility of 1 replacement in 60 y could be aided by minimising mist exposure of primary RP inflatable seals by introducing a Na dip seal upstream, as demonstrated by CRBRP studies and >1000 MW (e) LMFBR experiments suggesting40 y of life (NBR; design temperature: 66 ^{0}C; usual operating temperature: 51 ^{0}C) using a combination of 2 inflatable and 1 sodium dip seal without the necessity of purge (simplicity and economy by removing purge gas flow). Negligible release to RCB air (1.3 × 10⁻4mr/h vis-à-vis the allowable of 2 mr/h) with this combination did also indicate minimum- surveillance- necessity of the remotely- located dip seal for leaktightness. Ease of fabrication and fitment at larger gap of rolled shells with minimum stress makes this an ideal choice which could be standardised for the future FBRs transiting towards SFR.

Considering the envisaged limitations imposed by CS, collective HF release by γ dose, and the cumulative effects of various fillers and additives a uniform, conservative basis of 1 replacement is taken for all the 9 categories of cover gas elastomeric seals during reactor life of 60 y with S- 2 as pivotal and the life of inflatable seal maximised to 50 y based on improved T_s, E_b, permeability and mist resistance. Implementation of maximised life is based on the assumptions of defect- free standardised manufacture with improved uniformity of properties along sealing periphery and 1 end-joint per large extruded seal along with better reproducibility by automation of procedures. A standardised scheme of quality control maximises these potential gains.

Even though the possible additional effects from fillers and mist could be absorbed by the much larger FOS (> 6) by conceptual assessments, tailoring of S- 2 for other cover gas seals (with variations of fillers and additives) and CS limitations indicate the necessity for experimental evaluation considering the range of seals, materials and fluids in contact along with other operating requirements for implementation of 1 synchronised replacement in 60 y with safety and reliability.

5.7 Hot Feed Extrusion and Autoclave Curing Technology of Backup Seal Using S- 4[36, 87, 141]

This is a reference to the production process using factory compound batches 10a, 10b and 10c (Table 4.3) of IG3 (S- 4) for standardisation and unification of the manufacturing module.

Fig. 5.24. 1m diameter backup test seal produced by hot feed extrusion and autoclave cure.

Production of 1m diameter test seal (validated in scaled test rig, Fig. 5.7) required adjustment of hot- feed- extrusion (with autoclave cure, Fig. 5.24; cross-section: Fig. 5.18; *Photo courtesy: M/s HASETRI*) process parameters vis-à-vis those obtained from Plasticorder experiments (Section 4.6.1.1). Production of reactor seal was a continuing process of manufacture and quality control.

Fig. 5.25. Backup seal made of S- 4 for the small rotatable plug of INDIAN- SFR and its packaging.

Table 5.7. Properties of test and reactor backup seal compounds and specimens extracted from seals

Properties	Test seals						Reactor seals	
	Specimens from compound			Specimens from seal			Compound	Seal
	Unaged [d]		Aged (150 ^{0}C/70 h) [d]	Unaged [d]	Unaged [e]	Aged (150 ^{0}C/70 h) [e]	Unaged [f]	Unaged [f]
	ASTM Die C	ISO Die 2	ASTM Die C	ISO Die 2	ISO Die 2	ISO Die 2	ISO Die 2	ISO Die 2
T_s(MPa)[g]	6.5	9.4	6.5 (0)	11.7	12.5	12.6 (+1)	10.2	10.7
E_b (%) [g]	252	286	262 (+4)	341	316	307 (-3)	362	352
Hardness (^{0}Shore A) [h]	55	55	55 (0)	55	54	54 (0)	52	52
Relative density	1.900			1.890	1.914		---	---

[d] Made of batch 10a compound; [e] Made of batch 10b compound; [f] Made of batch 10c compound; [g] Numbers in brackets indicate percentage change of properties due to ageing; [h] Numbers in bracket indicate points (in ^{0}Shore A) of change in hardness due to ageing.

The LRP and SRP back-up seals (~6.4 m and ~4.2 m in diameters, Fig. 5.25) withstanding pre- commissioning conditions in the INDIAN- SFR for past 6 years, were manufactured with 5 and 3 end-joints respectively by suitable extrapolation of procedures developed for the test seals with a single end-joint. A hot feed screw extruder (length-to-diameter ratio of 10:1) was used to form the IG3compound (S- 4 with minor factory modifications) under feed, barrel and head temperatures of 50°C, 80°C and 80°C respectively to produce un-vulcanised pieces that were subjected to steam curing (155°C for 30 min) at 0.4- 0.5 MPa pressure, followed by step post-curing (230°C for 24 hours) to produce the cured profiles. The open ends of extruded profile (Fig. 5.18), cut at 45% and containing a proprietary solution of Viton®, were held under pressure and cured in air oven (150°C/ 35 min) to obtain the joints. Visual inspection, tests on specimens extracted from the seal to meet the target properties (Table 4.2) and material fingerprinting involving TGA and FT-IR were the main quality control procedures that were employed to ensure consistency of dimensions, material compositions and properties during the entire development process.

5.8 Foundation of Quality Control for Backup and Inflatable Seals Made of S-4 and S- 2

5.8.1 Quality control: Backup seal [87]

Table 5.7 compares the properties of factory batches of IG3 (S- 4) compounds (10a, 10b, 10c) used to produce test and reactor seals and those of specimens extracted from the produced seals

(Table 4.3). Batch 10a was used for production of 200–250 mm long test seal pieces. The properties of unaged and aged specimens stamped by ASTM Die C from sheets made of batch 10a are very close to those of laboratory IG3 batches. All subsequent measurements were done on specimens stamped by ISO die 2, as smaller size die is required to extract specimens from seals. The use of smaller size die increases T_s and E_b values, as evident from Table 5.7. Such values, determined on specimens from standard sheets prepared from factory compound (batch 10a) and seal samples, were used as reference for the test and reactor seals. Comparison of the properties of specimens taken from samples (batch 10a) and 1 m diameter test seals (compound batch identification: 10b) indicate negligible variations of T_s (+6.84%) and E_b (-7.33%) in the later considering the combined allowable deviations from intra-laboratory measurements and batch-to-batch variations of properties. Aged data of test seal specimens (batch 10b) indicate similar insensitivity to thermo-oxidative ageing as that of the earlier laboratory (Table 5.1) and factory batches of IG3. Comparison of properties of specimens taken from reactor seals (compound batch identification: 10c; Table 5.7) with the reference values indicates negligible variations in the later compared to the allowable deviations. These quality studies demonstrate effective translation of laboratory batch (of IG3) properties to the test and reactor seals, and provide further substantiation to the validatory test results.

Performance of backup seal in reactor depends on long-term chemical response of the seal material apart from its mechanical characteristics. The consistency of compositions in laboratory and factory batches (10a, 10b, 10c) of IG3 (S- 4) is illustrated by the material fingerprints derived from TGA and FT-IR in Figs. 5.26 and 5.27. Fig. 5.26 indicates virtually identical content of organics, carbon black and ash in the laboratory

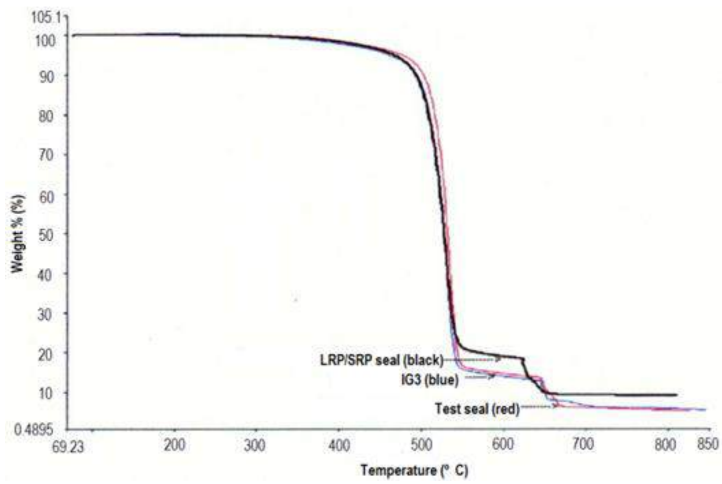


Fig. 5. 26. Comparison of thermogravimetric analysis results of laboratory batch and factory versions of IG3 used in test and reactor seals.

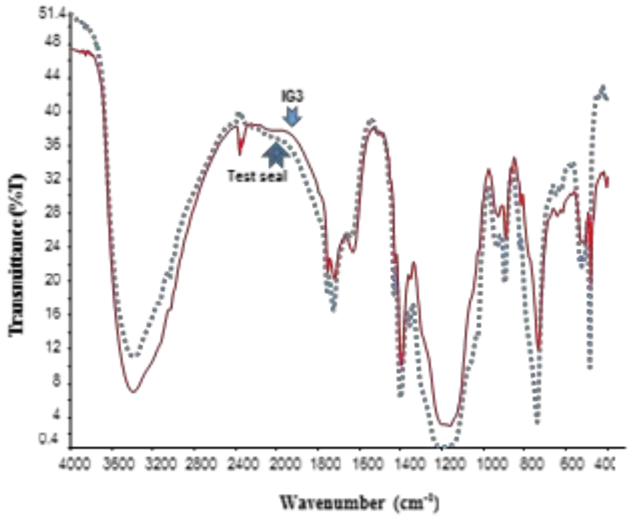


Fig. 5.27. Comparison of Fourier transform infrared spectroscopy results of laboratory batch and factory versions of IG3 used in test seals.

and factory (batch 10a) batches of IG3. The ash content of 5.2 wt.% in these batches indicates the total quantity of metal oxide and hydroxide. The MT black content of laboratory IG3 (Table 4.8) was marginally increased in the factory batch from production requirements. The reactor seal compound batch 10c displays higher ash content because of presence of talc which was used to handle ~4 m long uncured extrudate during steam curing. The same was not required for the 200–250 mm long seal samples (batch 10a). The reactor seal compound batch also required further marginal increase of MT black content from production requirements as reflected in Fig. 5.26. The addition of processing aids in the factory batch, which evaporates during the initial heating up to about 300 ^{0}C, is not prominent in the thermograms because of its low content.

The FT-IR curves of specimens taken from the batches of laboratory compound and 1 m diameter test seals (batch 10b), shown in Fig. 5.27, further emphasize the consistency of compound formulations across the batches and indicate the validity of the long term accelerated ageing data (and the FEA results derived from them) in predicting the life of the factory produced seal in reactor realistically considering the synergistic chemical influences of the operating environment.

5.8.2 **Quality control:** Inflatable seal [126]

Table 5.8 is a continuation of Table 3.12 (Section 3.6.6) to provide a set of property limits for inflatable seals, evolved through various Phases of developments and assumed a definite shape during Development Phase II i.e. cold feed extrusion and continuous curing of inflatable seal made of S-2 (Sections4.6.2 and 5.6.1). The departure from hot feed extrusion and autoclave cure to cold feed extrusion and pressureless (atmospheric pressure) cure by MW and HAT was one of the reasons to bring in changes in specification of property limits during Development Phase II (Table 5.8). The limits of T_s, hardness, dry heat resistance and CS for INDIAN- SFR inflatable seal were revised

Table 5.8. Property limits of INDIAN- SFR rotatable plug inflatable seal material across phases of development

Properties [a]	Property limits [a]				
	Applicable standards	Specimens from laboratory batches	Specimens from laboratory batches	Specimens from production batches	Specimens from seal
		Conceptual phase	Development phase I	Development phase II	
T_s *(MPa)*	ASTM D412 Die C, ISO 37 Type 2 Die (strain rates: 5 mm/min, 50 mm/min, 500 mm/min)	≥ 10.5	≥ 13	≥ 11.5	Within ±10% of that of corresponding limits of rubber compound
E_b (%)		≥ 300	≥ 150	≥ 150	
Hardness (^{0}Shore A)	ASTM D 2240, Durometer A	70 ± 5	65 ± 5	70 ± 5	
Tear Strength (N/mm)	ASTM D 624		18		
Resilience (%)	ASTM D 945	≥ 58 (Yerzley, 3.7 Hz)		Not used	
Hysteresis (%)		≤ 50 (Yerzley, 3.7 Hz)		Not used	
Abrasion resistance (%)		70	Not used		
Fluid compatibility, swell/shrink (%)			≤ 20/5		Not used
CS (%)	ASTM D395, Method B (25% deflection)	≤ 25 [b]	≤ 25 [b]	≤ 25 [c]	Not used
Dry Heat Resistance	ASTM D 573	[d] Change in hardness, T_s and E_b not to exceed 10%, 20% and 25% respectively.	[d] Change in hardness T_s and E_b not to exceed 10%, 20% and 30% respectively.	[e] Change in hardness, T_s and E_b not to exceed 10%.	Not used
Specific gravity	ASTM D 297, ISO 2781 (Method A)	Difference between two compound batches: ± 0.01 – 0.05			

[a] Limits of properties measured at RT. [b] After 70 h at 150 ^{0}C. [c] After 168 h at 150 ^{0}C. [d] After ageing in air at 150 ^{0}C for 70 h. [e] After ageing in air at 150 ^{0}C. for 336 h.

(Table 5.8) based on results of Phase-I (i.e. development of S- 1 and S-3, Table 3.11, and seals produced by them) and considering characteristics of fluoroelastomers made of APA in addition with the dual objective of using the property specification as an effective quality control tool for regular seal production and a framework for future developments.

The limits of T_s, hardness, dry heat resistance and CS for INDIAN- SFR inflatable seal were revised (Table 5.8) based on results of Phase-I (i.e. development of S- 1 and S-3, Table 3.11, and seals produced by them) and considering characteristics of fluoroelastomers made of APA in addition with the dual objective of using the property specification as an effective quality control tool for regular seal production and a framework for future developments.

Differences between Phase-I specification (Table 5.8) and developmental results (Chapter 3) indicate that the short term heat resistance limit could be brought down well within 10% while increasing the ageing span to 336 h (at the same ageing temperature of 150 ^{0}C) considering excellent thermooxidative ageing resistance of fluoroelastomers. The ageing period for CS was increased similarly to168 h keeping its target value and ageing temperature unchanged. The earlier specification of short term thermooxidative ageing (with wider band of degradation) and CS were maintained for EPDM-I (S- 3) considering sharp fall in properties (Fig. 3.39b). The limiting value of E_b was retained considering possible variability in the new compound because of anticipated compounding and production trials in cold feed extrusion and continuous curing system which eventually led to the transition from S- 1 (DMF3 or Comp #1) to S- 2 (Comp #9). The limiting value of T_s could be refined to 11.5 MPa (Table 5.8) based on a few trials which indicated that the lower modulus and higher hot tensile properties of the 50/50 blend of Viton® GBL-200S/600S could result in at least twofold gain in the FOS. Requirement for lowering of T_s limit (for quality control) was also indicated by a RT T_s of 12.5 MPa for S- 1 (vis-à-vis the minimum required 13 MPa, Table 5.8), made of another batch (12.28 MPa for the development batch of DMF3, Table 4.12). Lowering of T_s specification was feasible as the atmospheric pressure continuous cure in cold feed extrusion system contemplates to achieve the properties of moulded S- 2 specimens without post-cure which is sufficient for the present purpose considering the characteristics of commercial fluoroelastomer grades made of APA.

The S- 2 compound batch used for production of ~ 2 m diameter test seals had T_s, E_b and hardness (all measured at RT) of 13.7 MPa, 343% and 69^{0}Shore A (Table 5.6). The allowable hardness range of seal compound was raised to 70 ± 5 ^{0}Shore A based on the results of trials. The lower RT ionic interactions in fluoroelastomers belonging to APA grades leads to lower M_x and higher E_b for the same hardness (compared to the bisphenol cured VDF-HFP fluorohydrocarbon rubber such as Viton® A-401C based S- 1 and S- 4) which allowed raising of the hardness limits.

The higher E_b of S- 2 indicates necessity to revise the specification limit ($\geq$ 150 %, Table 5.8) in view of completeness and potential usage of the entire specification for regular production of seals and during compounding, design, manufacture and quality control for unification of FBR elastomeric sealing based on tailoring of S- 2. The conservative E_b limit is retained to be finalized during manufacture of reactor size seals. Sodium aerosol compatibility tests carried out during INDIAN-SFR and FBTR inflatable seal development (Chapter 3) and the changes in T_s (2%), E_b (−3%) and hardness (−1^0 Shore A) of Viton® fluoroelastomer reported based on sodium vapour exposure at 175 ^{0}C for 7 days in nitrogen gas indicate a limiting specification of variation for T_s/E_b/hardness (±5%/±5%/±5^{0}Shore A) of S- 2 under sodium aerosol exposure at 120 ^{0}C in argon gas for 2000 h.

5.9 Arriving at the Foundation for Overall Unification and Standardisation [37]

Continuation of inflatable and backup seals in the INDIAN- SFR RPs for the past 6 years through various phases of construction and pre-commissioning evaluations (with leakage across the secondary backup seals grossly within limit and that across inflatable seals varying beyond the limits within acceptable range to allow continuation) is a validation of the research on simplification, unification and standardisation addressed in this discourse. Validatory tests of seals in scaled rigs along with various other specimen- level tests, numerical analysis, data extrapolation and design approaches for life maximisation provide further substantiation through various parallel cross-verification routes.

Taking the foundation towards overall standardisation and unification of the 8 modules aimed at synchronised 1 replacement of cover gas seals (belonging to 9 categories) during MOX fuelled FBR life of 60 y (transiting towards SFRs) and the implementation of concept require extending the foundation through an overall framework of unification (Chapter 6). Readiness of 3 (FEA, manufacture, coating) out of 8 modules towards overall unification is indicated in the following

Sections for the sake of continuity in transition from specifics to general while taking S-2 (i.e. the 50:50 blend formulation of peroxide cured Viton® GBL 200S and 600S for INDIAN-SFR RP inflatable seals) as the cornerstone for tailoring to all cover gas seals at the end of Chapter 6 for uniform life maximisation and total unification.

5.9.1 Finite element analysis

A uniform approach of standardisation for FEA of cover gas seals based on plane strain (first approximation) and axisymmetric (finalisation) conditions, with Mooney-Rivlin constitutive model during normal operating and fuel handling conditions and 3-term Mooney-Rivlin as well as Ogden material model for special cases and accidental conditions (strains exceeding 50%), has been established based on studies carried out prior to this research and during this research as part of *Special Elastomeric Component* development.

5.9.2 Manufacture

Similar elements of standardisation for integrating the manufacturing module to the overall simplification-unification-standardisation scheme (comprising of 8 modules) could be found in extrusion of large diameter seals carried out prior to and during this research.

Fig.5.28. Progressive improvement of end joint for RP backup seal in INDIAN-SFR.

The hot feed extrusion and autoclave curing developed for inflatable-backup seals encompass extremes of static-dynamic compounds, cross-section (within 50 mm x 50 mm square) and number of seal end joints (inflatable: 1; backup: 3-5) in reactor scale. Production issues such as varying extrusion viscosity/ speed/ backpressure/ die-swell from extremes of reinforcing filler content in static-dynamic compounds, thin profile collapse under pressurized autoclave cure and scorching in thick cross-sections were addressed. The hot-splicing technique developed for backup seal and the joining procedure of inflatable seals by holding the under-cured open ends of extruded seal profile in mould under heat and pressure, similarly, includes the range of static/dynamic use, joint strength, seal wall thickness and hollow profiles seen in the reactor. The cold feed extrusion and continuous cure process is specific to the thin-walled, hollow profiles of INDIAN- SFR RP inflatable seal (Fig. 4.1) and APA based fluoroelastomers formulation used for the same. The extruded profiles are end-joined by transfer/compression moulding. Similar elements of standardisation are to be found in moulding for total unification of the manufacturing module towards the overall unification, addressed in next Chapter.

Figure 5.28 (*M/s HASETRI)* shows progressive improvement in end joint of INDIAN- SFR RP backup seal (made of S- 4) by hot splicing. The stress field perpendicular to end- joint cross-section of large diameter extruded static seals in reactor under low differential pressure and covered mostly by plane- strain condition is mostly compressive. Experiments in scaled rig on 1m diameter test backup seals indicated that seals are functional even with half of the specified splice strength (i.e. 3 MPa, Table 4.2) which suggests lower vulnerability of end- joint failure under such conditions involving long- term use. This is substantiated by backup seals made of S- 4 (and the design depicted in this thesis) withstanding the precommissioning conditions in INDIAN- SFR for the past 6 y without failure where 3-5 nos. of end joints were used in SRP- LRP seals. Further, cold feed extrusion and continuous cure proposed in this dissertation reduces the number of end joints to 1 in all large diameter elastomeric cover gas seals which minimises weak links in design and indicates feasibility of unification by standardisation based on this approach for long life and synchronised replacement.

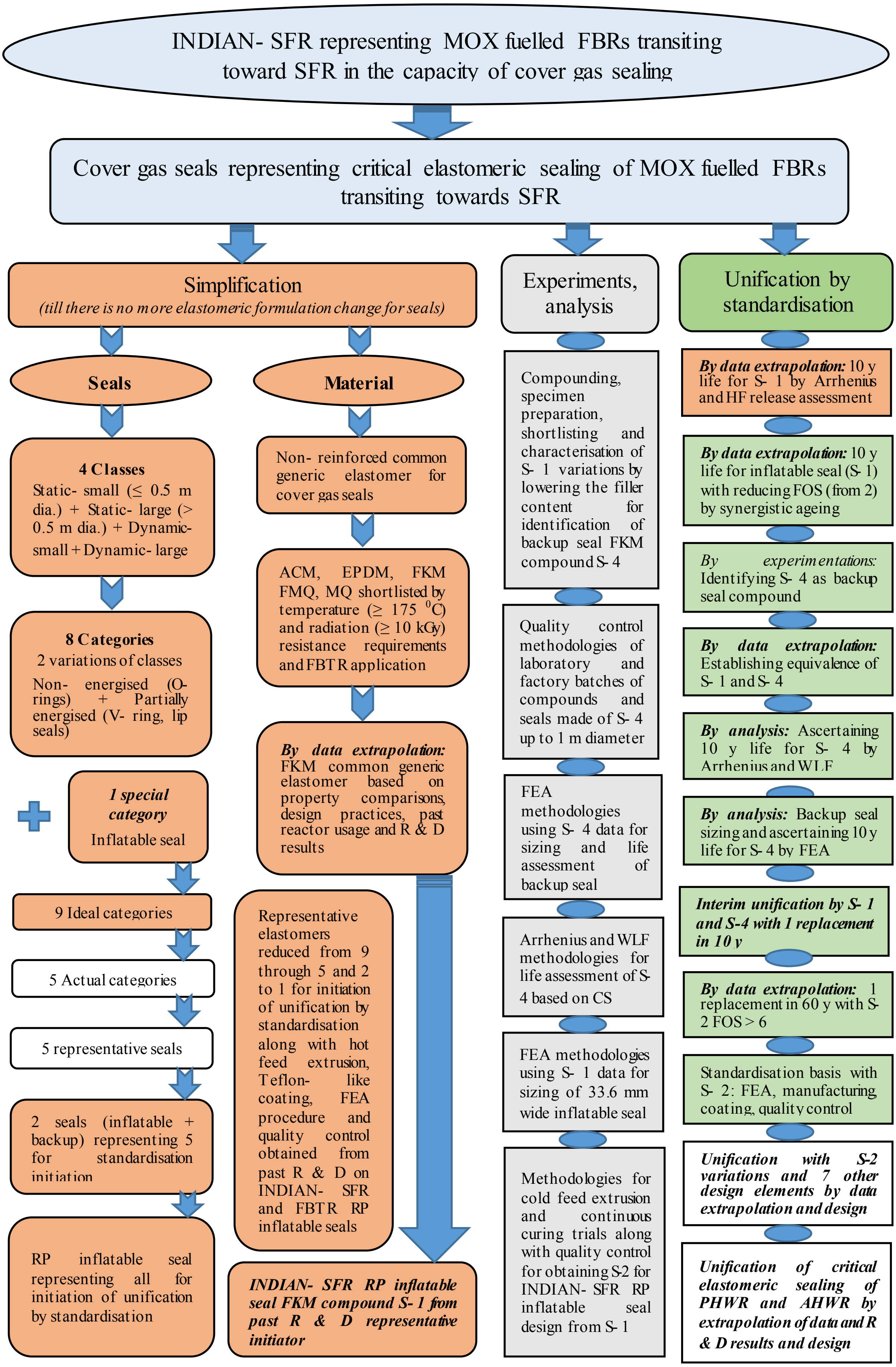
5.9.3 Coating

Procedures for the Teflon-like coating, developed for inflatable seals based on PECVD, are specific to the corresponding FKM- EPDM compounds (S- 1, S- 3; Table 3.11) and large diameter, extruded, dynamic sealing category-limited by the maximum seal diameter capability (7 m) of the coating facility. The limiting diameter encompasses other major dynamic seals (Table 3.1). Total standardisation of the coating module requires generalisation of the findings for moulded seals, taking into account the contact pressure variations (as maximum drag of 1000 N/m- seal- length has already been applied) etc., addressed in the next Chapter for overall unification.

5.10 Chapter Conclusions

10 y life for INDIAN- SFR RP inflatable seal made of S- 1 was ascertained by data extrapolation where reducing FOS (from 2 on T_s and E_b determined at ET) with synergistic ageing (temperature + radiation + mist + air) in reactor was assessed. S- 4 was identified as the elastomeric formulation for backup seal based on short- term experimentations on green and cured elastomeric specimens where 4 tailored variations of S- 1 with considerably reduced filler content and different levels of $Ca(OH)_2$ were studied. Results from short and long term experiments including detailed characterisations for extrudability and 32 week accelerated thermo-oxidative ageing at 140/170/200 ^{0}C were combined with FEA (stress- strain from unaged- aged tensile specimens), Arrhenius and WLF (CS) methodologies to ascertain 10 y life for backup seal made of S- 4 which included usage of data extrapolation in various fronts such as equivalence of S- 1 and S- 4 and progressive erosion of FOS with synergistic ageing. A preliminary unification of the cover gas seals of INDIAN- SFR (as model) for 1 replacement in 10 y was arrived by data extrapolation with standardisation of material module as cornerstone (S- 1, S- 2 and its 2 mouldable versions) considering parallel failure mechanisms comprising of FOS of 2 on tensile properties, CS of 80% and HF release. The increase of FOS to > 6 because of the change in INDIAN- SFR RP inflatable seal elastomeric compound from bisphenol cured S- 1 (based on Viton® A-401C) to peroxide cured S- 2 (based on 50:50 blend of Viton® GBL 200S:600S) through cold feed extrusion and continuous cure (by MW and HAT) experiments implies feasibility of 1 replacement in 60 y by data extrapolation (with reducing FOS estimation) which makes this compound the initiator for unification by standardisation of the 8 design elements. Foundations for other major elements (FEA as facilitator, seal production, coating and quality control based on production of reactor/test backup/inflatable seals) were obtained from experiments, analysis and data extrapolation along with past R & D results (Fig.5.29).

Fig.5.29. Outline of research methodologies: Progress in Chapter 5



Chapter 6

Results and Discussion

Overall unification, standardisation and benefits for SFRs

6.1 Final Categorisation of INDIAN- SFR Cover Gas Seals [36- 37, 87, 126]

Table 6.1 shows the final categorisation arrived through the initial categorisation (Table 3.1), subsequent refinement (Table 3.2), the finalised design specifications of these seals (Table 3.3), corresponding preliminary material specification (Table 3.4) and the finalised material specification for the categories of large diameter (extruded) static seals (backup, O- rings, Table 4.2) and large diameter dynamic seals (inflatable, Table 4.11).

The model of INDIAN- SFR TS sealing is abstracted into 4 basic categories (Table 6.1) and additional category (reinforced seals) from the initially 9 with corresponding target properties shown

Table 6.1. Operating condition, target properties of representative seal groups of INDIAN- SFR and FBTR

	Seals (manufacturing process)	Seal location(s)	Dia. min./max. *(mm)*	FIC	MIC	Speed max. *(mm/s)*	Temp. max. *(^0C)*	Pr. max. *(kPa)*
INDIAN SFR	**Static O-rings**(extrusion)[a]	RPs, CP, IFTM, PI, ARDM	~ 2400/6300	Air, Na aerosol	Carbon steel, SS	--	150	70
	Backup seals(extrusion)[a]	RPs	~ 4200/6400	Air	Carbon steel	--	110	25
	Rotary O-rings(moulding)[b]	FFIM, CSRDM, DSRDM	.10/76	Air, Na aerosol	Carbon steel, SS, Al-bronze	139	180 (120)	70
	Reciprocating O-rings(moulding)[b]	CSRDM	34/69	Air, Na aerosol	SS, Al-bronze	6	100	60
	Reciprocating v-ring/lip(moulding)[d]	CSRDM, DSRDM	90/110	Air, Na aerosol	SS	4000	120	60
	Inflatable seals(extrusion)[c]	RPs, IFTM	~4000//6300	Air, Na aerosol	Carbon steel, SS	75	150	25
FBTR	**Inflatable seals**(extrusion)[e]	RPs	~ 2400/3900	Air	Carbon steel	15	80	10

FIC: Fluids in contact; MIC: Materials in contact; Pr.: Pressure; Temp.: Temperature.

[a] **Target properties at RT:** T_s: 6 MPa (min.); E_b: 150% (min.); Hardness: 50 ± 5 ^{0}Shore A; CS (150 ^{0}C/ 70 h): 10% (max.).[b] **Target properties at RT:** T_s: 8.3 MPa (min.); E_b: 150% (min.); Hardness: 75 ± 5 ^{0}Shore A; Tear strength: 18 N/mm (min.); CS (150 ^{0}C/70 hours): 10 % (max.).[c] **Target properties at RT:** T_s: 11.5 MPa (min.); E_b: 150% (min.); Hardness: 70 ± 5 ^{0}Shore A; Tear strength: 18 N/mm (min.); CS (150 ^{0}C/168 h): 25 % (max.).[d] **Target properties at RT:** 8.3 MPa (min.); E_b: 150% (min.); Hardness: 70 ± 5 ^{0}Shore A; Tear strength: 18 N/mm (min.); CS (150 ^{0}C/ 70h): 20% (max.)[e] **Target properties at RT:** 10.5 MPa (min.); E_b: 150% (min.); Hardness: 70 ± 5 ^{0}Shore A; Tear strength: 18 N/mm (min.).

at the bottom of Table 6.1. The target properties for large diameter static and dynamic categories (extrusion) are parts of Tables 4.2 and 4.11, resulting from this thesis and earlier research (as complimentary subsets of *Special Elastomeric Component* development), which are outlined in Tables 5.7 ad 5.8. The preliminary specifications of elastomer target properties for small diameter (moulding) dynamic O- rings and V- ring/ lip seals, as indicated in Table 6.1, are for future implementation with refinements and finalisation as part of unification.

6.2 Arriving at Representative Conditions for INDIAN- SFR Cover Gas Seals [37, 45]

Maximum temperature for each group of seals are indicated (Table 6.1) considering upset conditions and incidents such as safety grade decay heat removal. The usual operating temperatures at INDIAN- SFR RP inflatable (~ 100 ^{0}C) and backup (~90 ^{0}C) seal locations are attributed to warm roof where the top surface of TS is maintained at ~110 ^{0}C using air cooling. The temperature will not be exceeded under loss of TS cooling during Category- III station blackout. TS cooling may have to be initiated (by emergency diesel generator) during Category- IV station blackout and the temperature could rise above 110 ^{0}C for a few hours influencing the temperature at seal locations. The maximum temperature of 180 ^{0}C is likely to occur at FFIM during such incident (maximum duration: 8 h), which, otherwise, should not exceed 120 ^{0}C as indicated in parenthesis (Table 6.1). The maximum differential pressures across O-ring and V-ring/lip seals take into account the possible failure of pressure regulators in argon supply lines.

The maximum speeds of relative reciprocating/rotary rubbing between seals and mating surfaces include possibilities such as absorber rod scram during emergency shutdown. The maximum rotational speed of 75 mm/s for INDIAN- SFR inflatable seals (Table6.1) is chosen from the two-speed scheme of RP (75/2 mm/s; 90% of total distance of travel at the speed of 75 mm/s; Fig. 5.1) and IFTM (~14/7 mm/s; Fig. 3.10) inflatable seals as the former corresponds to maximum distance of relative cumulative rotational travel (rubbing and wear, 100 km) of inflatable seal during 10 y. Cumulative design γ dose of 2 kGy over the design life is specified for the seals assuming negligible neutron flux and a maximum γ dose rate of 23 mGy/h at TS sealing locations. The maximum temperature of 150 ^{0}C for static O-rings (Table 6.1) correspond to loss of TS cooling and fuel handling condition. The IFTM inflatable seal design temperature of 150 ^{0}C (as against 120 ^{0}C for RP inflatable seals) is attributed to operating temperature of 35/150 ^{0}C during normal-operation/fuel-handling conditions.

The representative conditions arrived on this basis are abstracted as 10 y ---- 70 kPa ---- 120 ^{0}C ---- 23 kGy ---- 1000 N/ m-seal-length ---- 100 km ---- 10 $^{-3}$ scc/ s/ m- seal- length (Section 1.1).

Table 6.2. Composition and properties of inflatable and backup seal compounds

Compound ingredients	Inflatable Seal			Backup seal
	INDIAN- SFR RP design (FKM)		FBTR RP design (EPDM)	INDIAN- SFR RP design (FKM)
	S-1 (phr) [a]	S-2 (phr) [b]	S-3(phr) [c]	S-4 (phr) [d]
Viton® A-401C	100	--	--	100
Viton® GBL-200S	--	50	--	--
Viton® GBL-600S	--	50	--	--
Nordel® IP 4520	--	--	100	--
FEF (N550)	--	--	50	--
MT black N 990	20	--	--	2
SRF black N 762	--	25	--	--
MgO	3	--	--	3
$Ca(OH)_2$	6	--	--	3
ZnO	--	2	--	--
TMQ	--	--	1	--
Luperco® 101-XL	--	1	--	--
Diak™ No. 7	--	2.5	--	--
DCP-40	--	--	7	--
Struktol® WS280	--	0.75	--	--
Vpa No. 2	--	0.75	--	--
Paraffin oil	--	--	10	--

[a] **Properties at RT:** M_{100}: 4.8 MPa; T_s: 12.28 MPa; E_b: 217%; Tear strength: 31 N/mm; Hardness: 68 ^{0}Shore A. [b] **Properties at RT:** M_{100}: 3.3 MPa; T_s: 13.7 MPa; E_b: 343%; Hardness: 69 ^{0}Shore A . [c] **Properties at RT:** M_{100}: 2.6 MPa; T_s: 13.6 MPa; E_b: 316%; Tear strength: 40 N/mm; Hardness: 66 ^{0}Shore A. [d] **Properties at RT:** M_{100}: 1.5 MPa; T_s: 6.2 MPa; E_b: 252%; Hardness: 54 ^{0}Shore A; CS (150 ^{0}C/168 hours): 9 %.

6.3 Summarising Elastomeric Formulations for Inflatable and Backup seals [37]

EPDM and FKM compounds (S- 1 to S- 4) developed for inflatable and backup seals of INDIAN- SFR and FBTR RPs based on design (Table 3.3) and material (Tables 4.2, 4.11, 5.8) specifications, meeting target properties (Table 6.1) and described at various part of this thesis, are summarised in Table 6.2 along with compositions and RT properties for consolidation towards standardisation and overall unification. S-1/S-2/S-3/S-4 were developed for minimum 10 y

Table 6.3. Elements of design for unification as obtained from inflatable and backup seal development

Compound ingredients		Material			
		INDIAN- SFR inflatable seal		FBTR inflatable seal	**Backup seal**
		S- 1: Viton ® A-401C based (FKM)	**S. 2: Viton® GBL 200S/600S based (FKM)**	**S- 3: Nordel® IP 4520 based (EPDM)**	**S- 4: Viton® A-401C based (FKM)**
FEA	Analysis scheme	Plane-strain (first approximation) and axisymmetric			
	Constitutive relation	Two-term Mooney-Rivlin; Three-term Mooney Rivlin and Ogden (accidental conditions and special cases)			
Production	Extrusion	Hot feed	Cold feed	Hot feed	Hot feed
	Curing	Autoclave cure	Continuous cure	Autoclave cure	Autoclave cure
	End-joining	Heat and pressure at joints	Transfer/compression mould	Heat and pressure at joints	Hot-splicing
Teflon-like coating		PECVD	PECVD	PECVD	Not applied

of uninterrupted operation (S- 2 potential up to 50 y with 1 replacement in reactor life of 60 y assigned) at 120/120/80/110 ^{0}C under differential pressures of 25/25/10/25 kPa and γ dose rate of 23 kGy/h assuming continuous presence of air, sodium aerosol, argon, xenon and krypton (Tables 3.1-3.2 converging to Table 6.1).

6.4 Combining the Modules

6.4.1 General approach [37, 87]

Table 6.3 shows 4 elements of design for unification out of 8 where 4 material failure parameters (limiting Principal stress- strain based on FOS, CS, HF release and tear strength) had been the binding common thread along with the functional failure parameter of contact stress i.e. i) FEA as facilitator for sizing, optimisation and design, ii) FEA as facilitator for life assessment and design, iii) Production technology confined to extrusion for large diameter seals, and iv) Teflon- like coating based on PECVD for large diameter seals. Accelerated thermooxidative ageing studies of tensile and CS specimens spanning 32 weeks at 3 elevated temperatures in combination with life assessment by Arrhenius and WLF methodologies provide a standardised framework of parallel life assessment by the 6th element of design.

Findings from this research and long- term studies[30] on peroxide cured APA fluoroelastomers (similar to S- 2, Table 6.2) in nuclear applications indicate necessity of accelerated ageing studies (at 3 temperatures) spanning at least 100 weeks (up to 225 ^{0}C) for life assessment and nuclear grade qualification of representative elastomers and seals for future implementation of unification. These studies should include evaluation of the effects of additives, fillers and strain on synergistic ageing and life using specimen data extracted from seals. Assessment on possible consequences from collective release of HF (from 250 kg of cover gas seals) and drive for CS maximisation could provide a streamlined, efficient and comprehensive basis for future R & D. Tables 5.7 and 5.8 provide a standardised framework of quality control for large diameter, static and dynamic extruded seals across elastomer grades and laboratory- testing- reactor domains along with a set of applicable standards, validated through the results of development. Chapters 3- 5 provide the standardised testing scheme and methodologies for quality control used to arrive at this framework.

These 6 modules along with standardisation of elastomeric formulation by tailoring S- 2 (i.e. material module, 1st element of design) cover 7 modules out of the 8 elements of design to be unified by standardisation; the 8th module being future R & D for implementation. The approach and model for an efficient and reliable R & D scheme obtained through design option and compound minimisation and FEA implementation during *Special Elastomeric Component* development prior to this research, has been made all-encompassing by adding additional predictive techniques (Arrhenius and WLF) and a quality control framework for predictability and reproducibility with certainty, which could be extended to the other 2 categories of cover gas seals i.e. small diameter (moulded) dynamic O- rings and small diameter lip/ V- ring seals during future implementations. Effectiveness of framework has been demonstrated by completion of basic development- validation of EPDM- FKM inflatable and backup seals made of S-1, S-3 and S-4 within 5 y, 3 y and 2.5 y

[30]Examples: i) 32 weeks accelerated ageing studies during USA LMFBR cover gas seal development programme involving ACM, AU CU, CR, EPDM, FKM, IIR, MQ, NBR and SBR out of which AU CU, EPDM, MQ and NBR were shortlisted [110]. Ii) Accelerated ageing for ~16,000 h at a maximum temperature of 175 ^{0}C on Viton GLT- S O-rings for life assessment as part of more than decade long studies on radioactive material package casks.

respectively and continued functioning of inflatable as well as backup seals for the past 6 years in INDIAN- SFR RPs through various precommissioning phases.

The remaining works pertaining to overall unification is to find the variations of techniques and facilities for production and coating corresponding to all categories of cover gas seals along with tailoring of S- 2 for standardisation and unification of material module by data extrapolation and design, without any experimentations or numerical analysis.

6.4.2 Correlating finite element analysis and S- 2 with variations [37]

Identification of constitutive relations for seal elastomer (S- 2 and its variations) for all possible reactor conditions (Table 6.3) implies that the set of tests required to generate FEA input data (for determination of material/calibration constants of the constitutive models) are well defined. The tests are to be repeated on laboratory made standard specimens, specimens prepared from production compound batches and samples extracted from seals (including ageing effects) made of various seal compounds for accurate assessment of life and FOS at the end of life.

Effective integration of the standardized FEA procedure to the unification scheme and the universal code therefore becomes mostly compound specific where the remaining works are confined to the determination of test data on unaged and aged specimens made of modified versions of inflatable seal compound (S- 2) or new compounds tailored for other cover gas seals.

6.4.3 Correlating coating and S- 2 with variations [9- 10, 37, 43]

The PECVD coating procedure is largely independent of seal size because of evolution of the process through a 0.5 m diameter coating facility (Fig. 3.41) to the PECVD reactor (Fig. 3.42), which drives large diameter (1–7 m) seals under vacuum by motor-chain-pulley arrangement or continuous coating of seal rubbing faces. Coating of small diameter, dynamic moulded seals (Table 6.1) is likely to introduce certain alterations in PECVD coating parameters established for inflatable seals because of, i) Tailoring of extrusion specific inflatable seal compounds for moulding, ii) Introduction of new compounds for moulded seals, iii) Requirements of complete coating of the moulded seal surfaces (particularly the O-rings) as against coating of rubbing faces of bonded inflatable seals and, iv) Higher contact pressure in moulded seals (excluding CS/CSR effects) compared to the inflatable seals (normal operation/fuel handling: 70/50 kPa) which could necessitate an increase of the required coating adhesion strength to the originally specified value of ≥ 7 MPa (Chapter 3). The requirements of adhesion strengths ≥ 4.5/7 MPa (extruded/moulded seals), porosity ≤ 10%, thickness of 3–10 μm and 1/none overlap along circumference on coated surfaces of extruded/moulded seals for the PECVD based Teflon-like coatings correspond to the unified design requirements of INDIAN- SFR dynamic seals (Table 6.1). The smaller diameters, closer tolerances and better finishes of the moulded seals allow thinner coatings (3–5 μm) with higher adhesion strength while keeping the substrate (elastomer) temperature sufficiently low.

Complete standardization of coating procedure for reactor seals is a prerequisite for it to be part of unification scheme as a reproducible module and the measures to meet this objective is largely oriented to tailored and new compounds.

The life of Teflon- like coating deposited by PECVD and properties like stick slip assume importance in view of the aimed 1 synchronised replacement of seal in 60 y which suggests that coating life should be comparable with the seal for life maximisation and unification to be effective. Usage of Teflon coating in all the inflatable seals using non- liquid lubrication (Table 3.6) provides an indication about its unquestioned all- round supremacy in dry antifriction coatings, indicated in Sections 3.4.2.3 and 6.6. Low stick-slip of coating is a requirement in view of slower rotational speed of INDIAN- SFR RPs (2 mm/ s, ~ 10 % of cumulative travel) for finer and final positioning of the plug during fuel handling condition, applicable for other FBRs transiting towards SFRs as well. The life of Teflon- coated RP inflatable seals in Phenix (4 y, brushing) and KNK I-II (minimum 10 y, 15 y by unconfirmed data; spraying) using conventional deposition techniques are indicative of the feasibility of long life which was substantiated by Teflon- like coating deposited by PECVD on ~ 0.5 diameter tests seal for 10 km of travel (i.e. FBTR) without failure at IGCAR in scaled rig (Section

5.2). Unification by standardisation of Teflon- like coating for long life do find further substantiation based on the following counts.

I. Tefllonised coatings assure minimum coefficient of running or dynamic friction ($\mu = 0.037$) for Teflon- Teflon and 0.49 for Teflon- metal based on laboratory data) amongst all the types of dry polymeric coatings and impregnations [190- 191].

II. Teflon coating is virtually free from stick- slip. For all other types of coatings stick- slip are present more or less [77, 192].

III. Teflon based coating has the maximum temperature resistance and thermal stability where μ is virtually independent of either temperature or speed of relative motion between surfaces at usable temperature range [108, 190, 193].

6.4.4 **Correlating manufacture with S- 2 and variations** [37]

Moulded seals do not have end joints and curing takes place under pressure which has natural suitability for the bisphenol cured fluoroelastomer formulations of inflatable and backup seals i.e. S-1 and S- 4 based on Viton® A- 401C. For fast curing and easy flowing APA fluoroelastomer grades, moulding time reduces and injection moulding (instead of compression moulding) involving different process parameters and high shear rate (10^3- 10^4 s^{-1} compared to 10^2- 10^3 s^{-1} generally used for extrusion) could be employed to exploit the processing and property advantages of the elastomers. The combination of APA grades (S- 2 and variations) and injection moulding provides a unification basis for production of large number of small diameter moulded seals in the reactor (Tables 3.1 and 6.1) because of enhanced automation, precision with closer dimensional tolerances and reproducibility of dimensions/ properties in different cross-sectional geometries. The use of injection moulding simulation programs to determine an optimum combination of machine, mould configuration, runner design, material characteristics and process conditions for a given material can aid substantially in simplification and standardization.

It is required that one cold feed screw injection moulding machine capable of covering the viscosity range of INDIAN- SFR moulded static and dynamic seal compounds and the geometry/size variation is employed to minimize material wastage (owing to premature vulcanization in hot runner) and number of new/modified compounds because of machine specific requirements. In the same way, one cold feed extruder with MW and HAT continuous curing should be employed for all the large diameter extruded seals. Such steps could simplify process standardization considerably.

6.5 Unification by Data Extrapolation and Design, Tailoring S-2: *Static seals* [37]

The lower CS resistance of Viton® grades belonging to 'B' family (S- 2) compared to 'A' family (S- 1 and S- 4) is acceptable *(considering S- 2 as basis for standardisation of static sealing instead of S- 4)*because of several reasons namely: i) Reduction of filler level in S- 2 corresponding to static application will improve CS resistance, ii)CS resistance could be improved further by post-cure, iii) Major static sealing applications of INDIAN- SFR (excepting backup seal) are expected to retain sealing function at CS- CSR of 100% and beyond, iv) Backup seal life estimation based on CS is conservative, v) Reduction of CS resistance in Viton® GBL 200S and 600S and their blend are not substantial compared to Viton® A-401C, vi) Closer tolerances in injection moulded seals indicate that greater percentage of initial squeeze is available for sealing and, vii) CS resistance of S- 2 could be further improved by changing the type of filler. Reduction of permeation resistance for static seal compounds (because of lower filler content, obtained by tailoring of S- 2) is acceptable as permeation leakage through thicker seal walls of most of the static seals (inflatable seal wall thickness: 2 mm) under lower differential pressure (from buffer gas, 20 kPa) is less.

The distribution of ring and chord diameters of INDIAN- SFR (Table 3.1) and FBTR static elastomeric seals (Table 2.2, Chapter 2) indicate a broad division of moulded and extruded seals, viz. i) Moulded seals up to 0.5 m ring diameter and 10 mm chord diameter and, ii) Extruded seals of 10- 30 mm chord diameter with ring diameters in the range of ~0.5–6.5 m. Two basic formulations based on 50/50 blend of Viton® GBL 200S/600S tailored for cold feed extrusion with continuous cure (using MW and HAT heating) and injection moulding are envisaged to cover these applications.

The blend ratio of 50/50 is kept unchanged because of prior experience in extrusion (as depicted in Chapters 4- 5 with S- 2), maximum T_s and E_b after post- cure (duration: 4 h) compared to Viton® GBL 200S or 600S and natural suitability of intermediate viscosity compounds for injection moulding. Unchanged monomer composition is also intended to maintain the fluid compatibility of S- 2 in its modifications.

Use of mineral fillers is excluded as grades, properties, ingredient proportion, compounding procedures and quality control of furnace or thermal carbon blacks are better defined and standardized. The lower target strength, elongation and hardness of static seals (Table 4.2) do not indicate the requirements of special filler either. Most of the FKM compounds give hardness of 50-60^0 Shore A without filler because of higher viscosity of fluoroelastomers. MT and SRF blacks are therefore used mostly.

Furnace blacks with smaller particles and higher structure are used more in hydrocarbon rubbers for reinforcement rather than fluoroelastomers. Furnace blacks may interfere with peroxide curing and cause mould- sticking. The target hardness range of 50 ± 5^0 Shore A for backup and other static elastomeric seals (Table 4.2) indicate that MT blacks, often used in fluoroelastomers as non-reinforcing fillers, will be suitable. Thermal blacks with large spherical particle size (100–500 nm), almost non-existent structure and fewer reactive functional groups on particle/ aggregate/agglomerate surface give the best combination of CS, long term heat ageing and processability as also suggested by their wide usage in APA/Non-APA Viton® compounds in the form of MT N 990. This filler is therefore retained in the tailored formulations of S- 2 (Table 6.2) along with ZnO to ensure the best possible high temperature and long term heat ageing resistance. TAIC, known by the trade name Diak™ 7, is a universal choice as curing co-agent (cross-linker or radical trap) for the extruded/moulded seals as it is most effective in terms of CS resistance and cure state. The proportion of TAIC in moulded compound is likely to be different from extrusion formulation (and lower than the standard level of 3 phr) as higher hot tensile strength/elongation is necessary to overcome the resistance from deformation/sticking/fouling at de-moulding temperature to ensure extraction of parts from mould without tear. As a general thumb rule, E_b at moulding temperature should always be more than 100%.

Types and proportions of peroxide curing agent and processing aids are likely to be different in extrusion and moulding compounds. A typical low- filler formulation comprising of Viton® GBL 600S (100 phr), N 990 (5 phr), Diak™ 7 (3 phr), processing aid, Armeen® 18D (1- octadecanamine, N-octadecyl: 0.5 phr) and Varox® DBPH-50 (45% active dispersion of 2,5-dimethyl-2,5-di-(t-butylperoxy) hexane: 3 phr) give RT $M_{10}/M_{100}/T_s/E_b$/hardness/CS (22 h/200 ^{0}C) of 0.4 MPa/1.3 MPa/11 MPa/423%/52^0 Shore A/21% and 0.4 MPa/1.4 MPa/11.7 MPa/379%/55^0 Shore A/17% on standard specimens prepared without post-cure and with 2 h of post-cure at 232 ^{0}C (press cure: 5 min/177 ^{0}C). Compounds made of Viton® APA generally require 2–4 h of post- cure to optimize properties. The low filler content in static sealing formulations, however, minimize the effect of post-cure as seen from the results of typical formulations. This suggests that post- curing could be eliminated altogether in moulded and extruded seals (if CS requirements are met) for further process simplification as the hot-tensile/tear properties are required in non-post-cured compounds only. This argument is further strengthened by the large available margins in T_s/E_b (typical compound) with respect to the corresponding target values for static seals (Table 4.2) and the fact that post- curing improves T_s and CS with lowering of E_b. The role of post- cure in eliminating residual volatiles and imparting better dimensional/property stability suggests otherwise.

Processing aids serve multiple purposes during moulding and extrusion to minimize mould-fouling and improve mixing, compound flow, mould- release as well as surface smoothness of profiles because of which a combination of two to three chemically different aids are preferred traditionally. A 3- component system specified for earlier generation, peroxide cured, non- APA fluoroelastomers comprised of 0.25 phr stearic acid, 0.2- 0.5 phr fatty acid amide and 1- 1.5 phr hard wax such as carnauba or VPA No. 2 (ricebran wax). The characteristics of APA polymers indicate requirement of lower level (total: 1–1.5 phr) of aids. This minimizes the possibilities of negative impact on T_s and CS from some of the earlier generation aids (Armeen® 18D and waxes). Reduction of Armeen® 18D (widely used because of excellent mould release) content in compound formulation is also a requirement from the consideration of lowering of cure rate (and improving scorch safety) during injection moulding. A typical combination of processing aids for black filled APA grades comprise of Armeen® 18D (0.2–0.4 phr) and wax (0.3–0.5 phr) for flow and mould release and a silicone organic compound, Struktol® WS-280 (0.5–0.8 phr), for flow. Processing aids used at this level require 6- 8 h of post cure for optimum properties of compounds loaded with 30 phr of N 990 black. As per literature indications on later generation aids (which are free from the above disadvantages), mixture of fatty-acid-derivative/wax (flow and release), PAT 777 (up to 0.5 phr), and blend of fatty acid derivatives (all-round performance), Struktol® HT-290 (0.5–0.75 phr) should be sufficient for INDIAN- SFR static seals using APA formulations. Alternatively, Struktol® HT-290 used at around 1 phr could meet the functions which need to be ascertained. Carnauba wax or VPA #2 may be required at an optimum level (0.2–0.3 phr) to maximize release and smoothness in extruded seals. The required post- curing durations are expected to be 2- 4 h.

Reduced duration of post- curing is favoured during cold feed extrusion and continuous cure as the same could be carried out by multiple passes of the long extrudate through the HAT of extruder. The bonding at seal end joint also improves with reduction of post- cure duration. Luperco® 101-XL45 (45% active dispersion of 2,5-dimethyl-2-5-di-(t-butyl-peroxy) hexyne- 3) or Varox® DBPH-50 are the peroxide curing agents for extruded seals because of fast cure and low CS. For injection moulded seals, peroxides with slower cure rates such as Varox® DBPH50-HP (45% active dispersion of 2,5-dimethyl-2-5-di-(t-butyl-peroxy) hexane) or Varox® 130XL (45% active dispersion of 2,5-dimethyl-2-5-di-(t-butyl- peroxy) hexyne-3) could be the choice to increase the cure onset time and improve scorch safety. Standardization of approach demands the least feasible compositional variations between the extrusion and moulding compounds so that additional validations of the modified compounds could be minimized. In this context, use of Varox® DBPH-50 at 1.25–1.5 phr is preferable along with possible lowering of curing temperature than usual.

The number of static elastomeric seals in the RPs, CP, IFTM, ARDM, PSPs, IHXs, etc. of TS requiring extrusion is about 50 including the backup seals. Effective simultaneous preheating of green extrudate by microwave prior to its travel through the HAT is a prerequisite for uniform cure of the thick sections. The cross-sectional dimensions of backup seal (Fig. 5.18) and chord diameter range of the O-rings indicate that at least two variations of the basic extrusion compound (with different levels of peroxide curing agent) and corresponding process parameters (screw/extrudate speed, temperature distribution in extruder and HAT, etc.) will be required to ensure adequate cure rate so that the possibility of inadequate cure because of factors such as migration of TAIC to the product surface is eliminated and extrudate properties (determined on extracted specimens) consistent with those of non-post-cured standard specimens (belonging to seal production batch) could be obtained in a single pass of continuous cure.

These two variations of the basic extrusion compound along with their process parameters, joining of seal ends by hot splicing and the standardized FEA procedures form the basis of unification of extruded static elastomeric seals of INDIAN- SFR. The injection moulding compound for the static seals is likely to have two sets of process parameters corresponding to part thicknesses (O-ring chord diameter) below and above 5 mm.

6.6 Unification by Data Extrapolation and Design, Tailoring S- 2: *Dynamic seals* [37]

Dynamic O-rings with maximum chord diameter of ~5 mm, reciprocating V-rings of CSRDM (Fig. 3.4) and reciprocating lip seals of DSRDM (Fig. 3.5) could be moulded in the same injection-moulding machine used for static seals. The negligible release of volatiles from S-2 during service indicates its compatibility with other materials in contact such as stainless steel (Tables 3.1 and 6.1).

The V-ring seals of CSRDM reciprocates ~5 mm (speed: 2 mm/s) every day for continuous reactor power control and rub against the counterfaces for a distance of ~1.1 m at the same speed during reactor shutdown. The maximum rubbing speed of 4 m/s corresponds to absorber rod scram

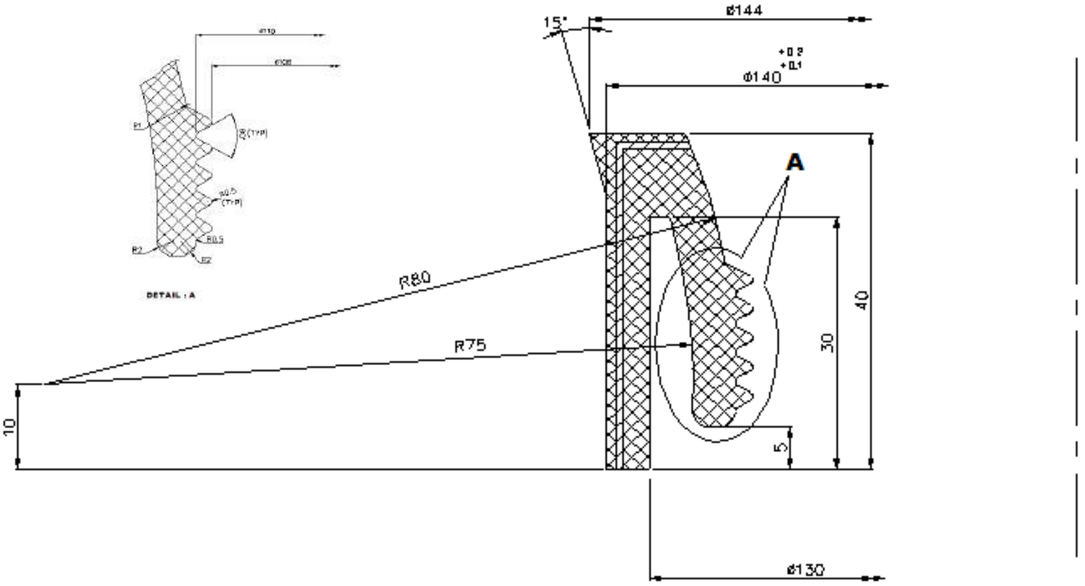


Fig. 6.1. Reciprocating V-ring seal of control and safety rod drive mechanism.

during emergency shutdown. The lip seals of DSRDM rub ~1.5 m during reactor shutdown (speed: 4 mm/s). The larger deformation of lips (compared to the O-rings), serrated rubbing faces (Fig. 6.1) and assistance from differential pressure ensure completely fail-proof and uninterrupted functioning of these seals during design life considering scram. The reciprocating and rotary O-rings operate in the squeeze range of ~10–20% (static O-rings: ~20–30%). The higher filler loading required for enhanced strength of dynamic seals increase CS. The CS resistance should therefore be maximized.

Higher hot-tensile/tear properties of APA polymers (typical value of hot tear strength for 50/50 blend of Viton® 200S/600S with 30 phr N 990 at 150 ^{0}C: 10.7 N/mm) give distinct tribological advantage in such applications over the Viton® A-401C based formulations. The advantages extend further to better radiation resistance and higher FOS as discussed earlier. These aspects, along with the overall maximum operational demands on RP inflatable seal materials vis-a-vis other dynamic seals (Tables 3.1, 3.3, 6.1), indicate that using modified versions of S- 2 should be the best solution for other dynamic seals. The blend ratio is maintained in the

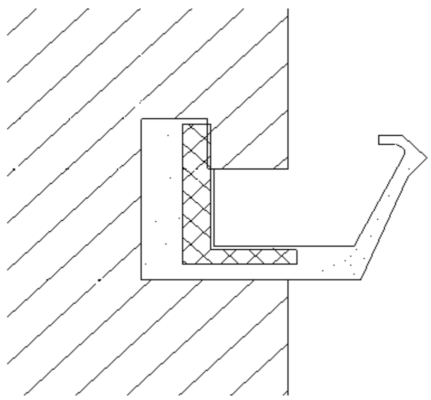


Fig. 6.2. Reciprocating lip seal of diverse safety rod drive mechanism.

modifications. Permeation resistance of the modifications should be more than static seal compound because of higher filler loading.

The design drag (starting/running: 1000/300 N/m length of seal) specified for INDIAN-SFR/FBTR inflatable seals are unsurpassed in moulded dynamic seals (in spite of higher contact pressure) as calculations are based on assumptions which are either very conservative or higher than corresponding values of moulded seals namely, i) Inflatable seal contact width of 30 mm and design contact pressure of 50 kPa during rotation corresponding to INDIAN- SFR inflatable seal design inflation pressures of 50/30 kPa during normal-operation/fuel-handling conditions, ii) Uninterrupted contact (dwell) between uncoated seal and mating surfaces for 8 month as basis for determination of starting drag and, iii) 0.6 and 0.2 as starting and running coefficients of friction ($\boldsymbol{\mu}$) as per design thumb rule even though laboratory studies indicate that amongst all types of dry contacts, Teflon–Teflon coating between surfaces give the minimum $\boldsymbol{\mu}$ (~0.04) which is practically same during starting and running. This indicates that combining the PECVD based Teflon-like antifriction coating (tailored for moulded dynamic seals) with the modified formulations could address all the design, manufacture and performance issues of the moulded dynamic seals.

Two formulations are envisaged for the moulded dynamic O-ring and V-ring/lip seals using the same injection moulding machine for static and dynamic seals. The content of N 990 filler in the composition of moulded static seals has to be increased to ~30 phr to suit the requirements of dynamic O-ring seals. All other ingredients will be unchanged even though their proportion could vary marginally to match the filler level and improve CS resistance. The general requirement of higher post- curing duration (4–8 h) for black filled APA polymer recipes of dynamic seals (to optimize wear resistance) is also expected to enhance the CS resistance. Austin black (coal fines) gives better high temperature CS resistance than the MT blacks but are poorer in T_s/E_b, reinforcement and processing. This could be blended with N 990 at low level to enhance the CS resistance further. One set of injection moulding process parameters should cover all the dynamic O-ring applications as the maximum chord diameter is limited to ~5 mm.

The dynamic V-rings and lip seals fitted to shaft diameters of 110 mm and 80 mm and produced by injection moulding should have a formulation different from that of the dynamic O-rings because of presence of metal inserts in the shaft/oil seal configurations (Figs. 6.1- 6.2) which necessitates the use of a blend of ZnO and MgO to simultaneously ensure good rubber-metal bond and heat ageing resistance. CS and CSR have lesser importance in these seals compared to the O-rings. The N 990 filler should therefore be replaced by a combination of untreated mineral fillers, Blanc Fixe® ($BaSO_4$) and Nyad 400 (fibrous $CaSiO_3$) at a level of about 40 phr. Blanc Fixe® can provide the best CS amongst the non- black fillers but has lower T_s compared to the MT blacks. Nyad 400 can provide T_s comparable to the MT blacks. A suitable blend of these could therefore provide the necessary reinforcement, hot tear resistance and tensile properties to ensure adequate de-moulding of the lip seal and V-ring geometries. The tensile properties and moulding characteristics could be improved further by using treated calcium metasilicate fillers such as Wollastocoat®. All other ingredients will be same as that of the dynamic O-ring compound even though proportions may vary. The post- curing temperature should not exceed 200 ^{0}C considering thermal stability of adhesives used for rubber- metal bond. Longer post- cure (8–16 h) will be needed because of the use of mineral fillers.

Dynamic sealing of INDIAN- SFR is therefore represented by 3 variations of Viton® GBL 200S/600S corresponding to the inflatable seals (S- 2), V-ring/lip seals of shaft/oil designs with inserts (or reinforced seals of other design) and moulded O-rings (or non-reinforced seal of other cross-section). The O-ring compound without post- cure (with minor modifications in formulation and other process parameters) will be useful for other dynamic seals with non-metallic inserts. These formulations and the FEA-processing-coating procedures form the standardised basis of unification for the dynamic elastomeric seals, taking INDIAN- SFR as model.

6.7 Benefits for SFR

The envisaged gain of ≥ 10- fold in R & D for implementation indicated in Chapter 5could be reduced further substantially (perhaps to half in an average) as studies for permeability, mist compatibility and accelerated ageing may not have to be carried out for all the compounds in view of the fact that the blend ratio (50:50) of Viton® GBL 200S and 600S are kept unchanged in the 4 variations of S- 2.

Realising1 synchronised replacement of TS seals in 60 y requires studies on cumulative effects of HF release (from γ dose) from all TS seals, assessment of the effects of strain on tensile specimens (ageing rate), more accurate predictions with specimens extracted from seals and a concerted effort to maximise CS which limits static-seal-life. Here, one representative elastomer formulation from static and dynamic classes could be chosen for evaluation as the extent of CS is largely determined by filler content. In essence a 20 times gain in R & D effort could be assumed.

The gains are expected to be enhanced further (considering minimum 100 weeks of accelerated ageing studies at 3 elevated temperatures with periodic specimen withdrawals and taking 5-specimen average for test results) as it has been demonstrated in this research that for elastomers with low filler content (i.e. static cover gas seals), only one strain rate (500 mm/ min) for tensile testing (for quality control, FEA, Arrhenius, WLF and design) is to be employed for complete nuclear- grade characterisation and qualification of compounds (i.e. variations of S- 2) instead of 3 strain rates (5- 50- 500 mm/ min) required for dynamic seals. The gains envisaged in reactor downtime, operating cost (15% out of the total cost) and avoidance of maintenance hazards are expected to be substantial which could be illustrated by comparing the 827 seal replacements (out of ~ 5000) per year in FBTR vis-à-vis 1 envisaged synchronised replacement in 60 y, which is supported by Canadian PHWR data and those from other FBRs such as KNK. Substantial reinforcement in safety and reliability is readily evident by gross simplification of design, operation and maintenance which reduces unforeseen failure probabilities and the operating synchronisations, documentations as well as record- update efforts considerably. These potential benefits are maximised by standardised predictive techniques, R &D and quality control which do also maximise reproducibility within and across reactors to minimise cost.

Economic benefits in repeat production is evident from assigning 1 extruder and injection moulding machine each for the production of seals (Section 6.4.4) which indicates time saving with ready spare availability and assured quality (with the standardised material- manufacture-quality control framework propounded in this thesis) which in turns adds to safety and reliability by ensuring 1 synchronised replacement in 60 y given the fact that elastomer part material composition, processing parameters, properties and performance are machine specific. Significant delays (one and half years) in FFTF-CRBRP test inflatable seal production, driven by non-availability of (machine specific) proprietary compounding details to changed manufacture is one of the many examples [43]. Indirectly, this approach does also signify better sustainability of the unification concept from another viewpoint apart from assured supply of material as the future R & D framework and repeat productions are based on joint- ownership of proprietary data in the form of specifications.

Quantifying overall economic benefits from elastomeric sealing and unification for SFRs has its first indication in Rs. 10 crore saving from large RP diameter and height reduction (late 1990s estimation, Chapter 2) using inflatable seal in addition to significant potential gains with reduced stress, increased stability, improved alignment and enhanced reactor life apart from savings in LM freeze seal cost or that from reduction in MV diameter etc. The list of benefits could be endless which is substantiated by large saving and improved reliability in Canadian PHWR plants (mid-1990s) from retrofitting of improved fuelling machine snout plug seals. The same report indicates three fold gains in life by improving CS resistance to heat and radiation and total envisaged savings of several hundred thousand dollars annually because of these improvements [51].

Looking at the issue of sustainability from assurance of continued seal material (S- 2 its variations) supply for SFRs in view of maximising critical elastomeric seal life (up to 1 synchronised replacement in 60 y), the availability of FKM rubber in a number of trade names globally provides the necessary basis for continuation of the concept of simplification- unification- standardisation i.e.

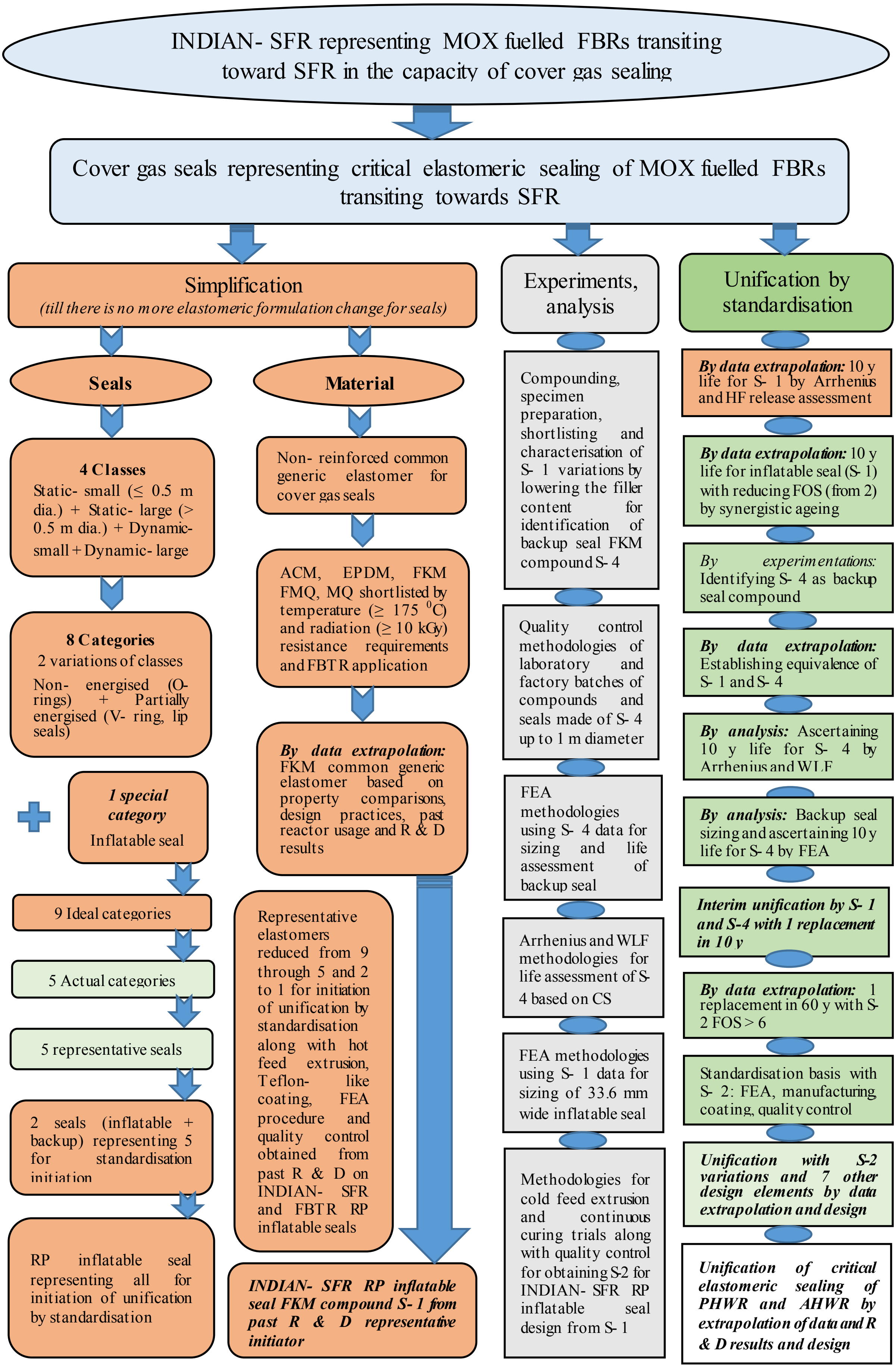
Viton (The Chemours Company, previously DuPont); Technoflon (Solvay Solexis), Dyneon fluoroelastomer (Dyneon, previously 3M), Dai-el (Daikin) etc. There is, however, a necessity to ascertain equivalent (to S- 2) elastomeric resins and compounds experimentally (along with corresponding specifications) as part of future research for assured sustainability due to specific requirements for life maximisation such as hot tensile properties, extrudability for better quality seals, amenability to injection moulding etc.

All the above aspects suggest maximisation of safety, reliability, economy and sustainability and the same could be extended to the metallic fuelled FBRs of future as the operating requirements of cover gas seals differ only marginally.

6.8 Chapter Conclusion

The 9 ideal categories of cover gas seals for INDIAN- SFR as model is brought down to 5 categories based on actual usage which includes reinforced seals as separate category and inflatable seal as special category along with large diameter extruded static seal and small diameter moulded O- rings as well as V- ring/lip seals. Past R & D from *Special Elastomeric Component* development and findings from this research coupled with the uniqueness of elastomer correlativity (other design elements mostly compound specific) is used to bring out maximum dependence of the other 7 design elements with S- 2. The INDIAN- SFR cover gas material module (or the first element of design) is standardised purely by data extrapolation to illustrate that TS sealing could be entirely covered by S- 2 and its 4 various where the blend ratio of 50:50 is kept unchanged. This, along with FEA based on plane- strain (first approximation) and axisymmetric conditions (finalisation) with Mooney-Rivlin material model, two basic variations of Teflon- like coating by PECVD, manufacture by cold feed extrusion and injection moulding (1 machine assigned for each), FEA- Arrhenius- WLF based life assessment and quality control in combination with standardised future R & D model provides a framework for unification which is extendable to metallic fuelled FBRs (Fig. 6.3). The envisaged gains in future R & D for implementation is estimated to be 20 times.

Fig. 6.3. Outline of research methodologies: Progress in Chapter 6



Chapter 7

Results and Discussion

Extending Unification to PHWR and AHWR by Material

Extrapolating literature data and R & D results

7.1 Design Requirements, Failure Modes and Desirable Properties [37]

The essential operating differences in critical elastomeric sealing applications of PHWR and AHWR are presence of water and steam at high pressure (up to ~10 MPa), temperature (up to ~315 ^{0}C) and γ dose rates. The use of various grades of SS as mating surfaces indicates necessity for exclusion/minimization of halogen and free sulphur in the seal material formulation to avoid corrosive effects on reactor components. Cumulative γ dose resistance of 10– 100 kGy is a necessity considering typical extremes of seal replacement intervals (3 months to 5 y). Snout plug and coolant channel pressure boundary seals of PHWR and AHWR are representative examples of such seals.

O-rings are widely used with chord and ring diameters up to ~10 mm and ~0.5 m respectively. CS does not influence sealing significantly in a purely static (without vibration, pressure or thermal cycling etc.) application and is not a failure criterion either (usually) as the seals act like fluid- filled bags at high differential pressure which adds hydrostatically to the assembly contact pressure at the sealing surface generated by initial squeeze. Presence of tensile zones and crack propagation probabilities are minimum usually because of totally confined sealing configurations under high differential pressure (Section3.3.1.2). Leak- tightness is less dependent on CS- CSR also because liquids are easier to seal than gases. Extrusion is a major failure mode and seals of 80–90^0 Shore A hardness are usually required. High hot tensile properties, compatibility with water and steam, sustaining the synergistic influences of temperature, radiation and fluids (including air) and minimization/elimination of release of low molecular weight volatile products or halides during operation are some of the major requirements that govern seal material selection.

Studies on oil and gas applications (temperature- pressure ≤ 225 ^{0}C- 100 MPa) and plate heat exchanger gaskets (hot water and steam at 180–200 ^{0}C) indicate that Viton® GBL-S with various blend ratios could satisfy most of these requirements.

The presence of pressure and temperature fluctuation/cycling in PHWR and AHWR critical elastomeric barriers along with possible presence of vibration as well as other forms of mechanical disturbance however introduce CS as a failure criterion which was demonstrated by late- 1970s studies on snout and magazine seals for coolant channels and fuel handling machine of PHWR belonging to CANadian Deuterium Uranium (CANDU) design. Experiments with O- rings made of 4 generic elastomers (Viton®, silicone, ethylene propylene and perfluoroelastomer) from 4 suppliers in pressurised hot water for long (up to 1y at up to 6.9 MPa and 232 ^{0}C) and short terms (about 30 min, up to 8.3 MPa and 270 ^{0}C) under weekly pressure- thermal cycling and pressure- thermal shock respectively indicated a common trend of failure by leakage beyond 75% CS [194].

Sudden generation of leakage gap at sealing face by cycling or shock and time lag on the part of seal to fill the gap by hydrostatic force (generated by differential pressure) require additional simultaneous support from remaining elasticity and resilience of the O- ring rubber sourced from significantly below- 100%- CS. This is substantiated by results from the same experiment that failure of absolutely undisturbed seals is not governed by CS [194]. The reasons for leakage and premature failure of O- rings made of Viton (diamine or bisphenol cured at that time) and silicone rubbers in the above experiments with and without splitting and cracking were mainly due to fluid incompatibility as extrusion was absent because of zero clearance in the face (flange) seal test arrangement where stress field was essentially compressive under a differential pressure up to 8.3 MPa. O- rings made of ethylene propylene rubber with much lower temperature rating performed impressively up to 52 weeks of leaktightness [194] because of inherent compatibility with hot water and steam [80, 107] and an essentially compressive stress field which camouflaged the weakness of rapid drop in tensile strength/elongation properties of the elastomer under high temperature ageing as indicated in Chapter 3. The performance was aided by much lower CS of ethylene propylene in hot water (about half) compared to that in air under identical conditions as demonstrated by a previous experiment [195]. The above observations are a designer's take based on current state of knowhow and could be easily demonstrated by FEA to confirm the compressive stress- strain field in the ethylene propylene O- ring. The inadequacy of interpretations in reporting test results as well as failures could be attributed to evolving understanding on rubber technology (late 1970s) illustrated by the Challenger Space Shuttle failure due to a fluoroelastomer O- ring [40] about a decade later (1986).

The key aspect of unification by standardisation of material module here is that unlike bisphenol cured FKM grades (i.e. S- 1 and S- 4), the peroxide cured ones made of APA (i.e. S- 2) are not vulnerable to aqueous medium (polar fluids). Also, the range of critical elastomeric sealing applications in PHWR and AHWR cannot assure purely compressive field (with very low tensile in some) everywhere (because of radial mounting, radial/diametrical clearances and propensity of extrusion in such cases) which brings in the necessity of improved hot tensile properties and thermal stability for unification and standardisation.

7.2 Unifying PHWR Elastomeric Sealing [37]

Operating temperature limit of PHWR seals (200 ^{0}C) suggests natural suitability of APA polymers for these applications. A 70/30 blend of Viton® GBL 200S/600S studied for oil and gas application contains 3 phr/5 phr $MgO/Ca(OH)_2$, 20 phr Ensako® 250 black, 10 phr Austin black®325 and is cured by a combination of Varox® DBPH50-HP (1.25 phr), Viton® curative no. 50 (2 phr) and TAIC (0.5 phr) to give RT $T_s/M_{10}/M_{25}/M_{50}/M_{100}/E_b$/hardness of 19 MPa/3.6 MPa/6.1 MPa/9.7 MPa/16 MPa/133%/91^0 Shore A. 80%/41%/40% of RT $M_{50}/T_s/E_b$ is retained at 200 ^{0}C. The hot tear strength at 200 ^{0}C is very impressive at 14 N/mm. The low E_b (53%) at 200 ^{0}C requires improvement and this could be done by use of lower reinforcing filler (such as MT N 990) which increases elongation with the reduction of modulus and strength.

Use of thermal black also reduces impurities in the form of heteroatoms (hydrogen, oxygen, sulphur, etc.) incorporated in the carbon black structure. Thermal blacks typically contain <1% heteroatoms as against 2–3% for furnace blacks. Sulphur impurities are tightly bound to black structure and does not affect vulcanization process (typical temperature range: 160- 190^{0}C) which suggests that they are unlikely to contribute towards corrosion of reactor components. Increasing the content of N 990 in Viton® GBL 600S formulation to 60 phr gives M_{10}/M_{100}/T_s/E_b/hardness of 2.1 MPa/8.6 MPa/16.4 MPa/240%/87^0 Shore A at 2 h/232 ^{0}C post-cure which appears to have suitability for PHWR applications from the requirements of extrusion resistance and demoulding considering seal geometries of varying complexities. The higher CS of 29% (22 h/200 ^{0}C, Method B, O-rings) because of higher filler content compared to the 70/30 blend should not be a problem in many cases. Low level of Austin black could be blended along with other variations of blacks. From processability requirements some blending with Viton® 200S is likely. Lowering or eliminating metal oxides is a requirement to minimize unsaturation and volatile release. At the same time, metal oxides are required to enhance thermal stability – ZnO at 3 phr could be ideal in many applications. Maximizing thermal stability is a prime necessity to preserve the M_x, T_s and E_b for countering the synergistic degradations from high radiation. Case studies on high-duty sealing applications of PHWR indicate adhesion to mating surface and tearing (driven by thermal degradation) as the main failure modes rather than radiation effects.

The above examples show utility of S- 2 variations (in terms of blend ratio and fillers) for PHWR sealing applications in terms of fluid compatibility, mechanical properties, processability and ageing which forms a basis for unification along with the FEA procedures and injection moulding based production process adopted for the INDIAN- SFR model. Extrapolation of earlier results from S- 2 and S- 4 could easily show that release of volatiles from PHWR seals at 200^{0}C and at a typically high γ dose rate of 50 Gy/h will be negligible at a cumulative dose of 100 kGy- at least if this release is assumed to happen in FBR TS. Studies carried out on Viton® B (a terpolymer of VDF, HFP and TFE with 68 wt.% F) specimens in air at the same temperature indicates drop of RT T_s//E_b by

~35/50% (maximum M_{25}: ~3 MPa) at a cumulative dose of 100 kGy which suggests that S- 2 variations tailored for PHWR seals will remain functional.

These aspects along with the effects of volatiles on PHWR system, however, should be ascertained experimentally to validate the basis of unification.

Static and dynamic O- rings are usually fitted in axial (face or flange) and radial configurations where the dynamic seals could be either reciprocating, rotating or oscillating. Surface finish (groove and mating surface) of 1.6 µm – 3.2 µm are tolerable for static seals even through the finish should be limited to 0.8 µm. For reciprocating and rotary elastomeric O- rings the surface roughness ranges are 0.2 µm – 0.4 µm and 0.4 µm – 0.8 µm respectively [196- 200]. Even though NBR carries an edge amongst oil resistant rubbers in terms of resistance to extrusion and wear, operating temperature of the seals are limited to 120 ^{0}C because of material limitations [196] which could be increased to 150 ^{0}C by using hydrogenated nitrile butadiene (HBNR) rubber with better thermal rating and increased strength [197, 199]. Lower hot tensile properties of bisphenol cured fluoroelastomers (such as Viton® A-401C) are general limitations in sealing higher differential pressure (as in PHWR and AHWR) which is why these seals are used in below- high- pressure applications (low pressure seal ≤ 0.7 MPa differential pressure transiting to high- pressure ≥ 10 MPa) with an usual higher- end hardness of 75 ^{0}Shore A [199]. Peroxide cured APA fluoroelastomers (i.e. varying the blend ratio of S- 2) remove this limitation with much higher strength retention at ET. This is indicated by utility and usage of these rubbers (up to ~ 95 ^{0}Shore A hardness) in oilfield and gas applications with high pressure (≥ 100 MPa), high temperature (≥ 150 ^{0}C) operating range including ultra- high temperature domains (≥ 200 ^{0}C or ≥ 225 ^{0}C) where extrusion resistance is of prime importance [201- 202].

Elastomeric O- rings starts transmitting the differential pressure hydrostatically to add to the contact pressure, 0.7 MPa onwards. This initiation of hydrostatic transmittance could be in the range of 0.7 MPa – 1.4 MPa which is genre- class- compound specific (Section 3.3.1.2). Extrusion is the flowing of seal's body into axial/ radial clearance gap between seal groove and mating surface [197].

A standard rectangular O- ring groove design could be used to seal a differential pressure of 10 MPa or more. This could be increased further to 100 MPa or more reliably with non- reinforced rubber if proper design precautions are taken to address the issues of seal- counterface movements, elastomer ageing and extrusion [196, 199]. Flange and face seals with zero axial clearance handles 1360 MPa using a seal compound of 70 ^{0}Shore A hardness [196]. Extrusion resistance depends on differential pressure, temperature, compound hardness and the radial clearance. As design charts of handbooks and various sources of supply indicate [196- 200], the radial clearance could be as much as 0.25 mm for a 90 ^{0}Shore A rubber involving seals of up to 8.4 mm cross- sectional diameter (metric series: 1.6 mm – 8.4 mm; inch series: 1.78 mm – 6.99 mm) and 655 mm ring diameter (3.5 mm – 250 mm; 2mm – 655 mm) covered by various international standards which should include all PHWR elastomeric O- ring sealing applications adopted so far. The clearance limit takes into account possible gap increase from pressure- expansion of cylinder/ bore, concentricity of cylinder/bore-shaft and further breathing effects which vibrations from pressure cycling could bring in. Static and dynamic seal cross-sections are generally compressed by 10% - 40% and 10% - 30% [200]. It is always better to go for the highest clearance by using rubber of highest hardness (usually 90 ^{0}Shore A) from economic considerations as precision machining costs are reduced substantially which could assume significant proportion if adopted with uniformity by unification and standardisation in PHWRs.

Ideally the seal grooves should be rectangular with straight sides. However, for the sake of ease- of- machining, convenience therefrom and economy, 5 ^{0} slant in groove- side- faces could be tolerated up to 10 MPa differential pressure [196, 198- 199]. The rectangular groove edges should have a minimum radius of 0.15 mm ideally (could be reduced to 0.125 mm) in order to optimise avoidance of nicking- cutting from sharp edges and extrusion- probability from wider gap, created by higher radius [198]. Up to 10 MPa, backup rings are not required as additional support (at low pressure side of seal) to resist extrusion for NBR or HBNR O- rings of 90 ^{0}ShoreA hardness [196-197, 199]. The possibility of introducing backup ring at <10 MPa differential pressure because of lower hot tensile properties of fluoroelastomers is reduced substantially by peroxide cured Viton® rubber made of APA. These aspects imply that the feasibility of unification of PHWR critical elastomeric sealing with variations of blend ratio in S-2 could be translated into total standardisation with the usage of backup- ring- free seals, grooves, finish and clearance as indicated.

One of the important criteria in this respect is to ensure that the direction of frictional force or drag in reciprocating dynamic sealing acts opposite to the differential pressure (by proper positioning of groove in cylinder/bore or rod/plug as applicable) failing which the maximum no-extrusion differential pressure rating of O-rings without backup ring could be reduced to 30% - 40% of 10 MPa for the same temperature- hardness – clearance combination. Design criterion by most of the standards and manufacturing catalogues indicates 70 % filling of groove by O- ring where differential thermal expansion between seal and elastomer and volume swell of rubber by the sealing fluid typically consume about 13% and 5- 10% of free space [196- 200]. Out of a number of cross-sectional dimeters available as per standards corresponding to each ring diameter, the larger ones are preferred in order to ensure effective absorbance of various tolerances (also reduces machining cost by allowing larger tolerance band) and improved resistance to ageing as well as extrusion. For critical applications such as these, tolerances on O- ring cross-sectional diameters (inch- metric maximum: 0.15 mm) may have to be considered. Choice of larger cross-section for the same ring diameter is limited by the necessity of surrounding metal works in terms of space, weight and cost [196]. Economic factor could also be a factor of consideration in terms of total cost of high- priced specialty elastomers considering standardisation of the entire PHWR domain with second largest installed capacity (11 %; LWR: 82%) and growth rate (under construction: PHWR 7%; LWR 85%) where the 0.2 Mt of Indian uranium reserve signifies expansion to 20 GWe (Chapter 1).

The inch series of O- rings is covered by British Standard (BS) BS 1806 and Society of Automotive Engineers (SAE) Aerospace Standard (AS) SAE AS568 (USA) with ISO 16032 covering the corresponding metric dimensions. The most widely used pure metric seals are based on the size ranges of BS 4518 and similar Swedish document SMS 1588. A number of alternative series of standards have been devised for specialised applications including those from the aerospace industry for high precision and quality seals which could be of relevance for the present applications. SAE AS5857 provides standardised design criteria and dimensions for grooves corresponding to SAE568 size static O-rings without backup rings (< 10.3 MPa) and with backup rings (> 10.3 MPa). SAE AS4716 and SAE AS568A is the combination for aerospace dynamic (reciprocating) O-rings. Various other O-ring groove design specifications being used in industry include Aerospace Recommended Practice (ARP) 1232, 1233, 1234 and ISO 3601/2. Other country-specific standards for O-ring manufacture are AFNOR 47501 (France), DIN 3771 (Germany), JIS B2401 (Japan) and SMS 1586 (Sweden) along with ISO 3601 [198-199].

The critical elastomeric sealing applications of PHWR could be standardised with blend-ratio variations of S-2 using the above guidelines and standards discussed as per the requirements of specific application.

7.3 Unification of AHWR Elastomeric Sealing [37]

The static sealing application of coolant channel seal plugs of AHWR works at design differential pressure and temperature of 8.5 MPa and 300 ^{0}C while remaining in contact with air, hot water, steam and SS. The γ dose rate (2 Gy/h) at full reactor power during normal operation results in a design dose of 15 kGy/y. A typical 50/50 blend of Viton® GBL 200S/600S loses 27/34% of M_{100}/T_s (with E_b gain of 18%) after 70 h/275 ^{0}C oven ageing which indicates the requirement of improved high temperature resistance. This shifts the material selection further to peroxide cured perfluoroelastomer(FFKM) with low envisaged release of volatiles and halides because of highest purity and lowest extractive contaminations from the material amongst elastomers. Commercial grades of silicone rubber of recent developments hold some promise in this respect because of compatibility with water/steam up to about 260 ^{0}C even though the issue of retention of properties at that temperature remains.

A rule of thumb indicates drop of hardness by 10 ^{0}Shore A for every 100 ^{0}C increase in temperature [199] which implies that radial clearance should be lower and tolerances are to be tighter for the AHWR seals with other guidelines and standards as indicated for the PHWR seals. FFKM, special subgroup of fluorocarbon elastomers, are essentially rubbery derivatives of polytetrafluoroethylene (PTFE) which exhibit exceptional chemical inertness and thermal stability. Kalrez and Perlast are two types of commercial FFKMs [155]. Grades of Kalrez are resistant to more than 1800 chemical substances and are capable of service at temperature up to 327 ^{0}C, outperforming FKM, PTFE and other generic elastomers. Kalrez Spectrum 6375 has the broadest chemical resistance to solvents, steam, amines, bases and acids with the lowest temperature rating (275 ^{0}C) amongst the other frontrunner Kalrez grades (4079 and 7075) from M/s DuPont Performance Elastomers. Kalrez 4079 (Industry standard for > 20y) has lowest chemical resistance amongst the three with temperature rating (316 ^{0}C) falling in between Kalrez Spectrum 6375 and 7075. Kalrez Spectrum 7075 on the other hand has the highest temperature rating (327 ^{0}C) and resistance to acids while other chemical resistance falls in between Kalrez 4079 and Kalrez Spectrum 6375 [199, 203].

AHWR seals require temperature rating in excess of 300 ^{0}C and maximum resistance to aqueous medium. Kalrez Spectrum 6375 appears to be the best choice as Kalrez Spectrum 7075 is not suggested for severe aqueous and amine applications and Kalrez 4079 has the lowest resistance to these amongst the three. Much better performance is envisaged compared to the bisphenol and peroxide cured Viton® discussed in the thesis as the high temperature stability translates to increased chemical resistance in all fronts. The temperature rating of 275 ^{0}C for Kalrez Spectrum 6375 is however life limiting. Besides, typical usage of the two Spectrum grades corresponds to a hardness of 75 ^{0} Shore A which is unlikely to be suitable for the differential pressure of 8.5 MPa in view of high operating temperature. In this context Kalrez Spectrum 7090 (latest addition to Kalrez Spectrum family) with typical hardness of 90 ^{0} Shore A, lowest CS (12 %, 70h/ 204 ^{0}C), impressive mechanical properties (RT T_s: 22.75 MPa; M_{50}: 15.5 MPa) and outstanding sealing force retention (as well as chemical compatibility) could be a potential solution for unification and standardisation of critical elastomeric sealing applications for AHWR [199]. Compatibility with materials and fluids in contact (SS, aqueous media) and life under synergistic ageing however are to be experimentally verified in terms of corrosion, volatile release, leakage etc.

Late- 1970s experimentations involving elastomeric O– rings for PHWR snout plug of Canadian deuterium uranium (CANDU) design also used perfluoroelastomer as one of the candidate materials which proved to be ineffective [194]. Even though commercial FFKM grades improved significantly since then there is a necessity of experimental validation in terms of quantification of the benefits.

7.7 Economic Aspects

Relative cost of the raw materials (Indian price) for S- 1 (FKM- Viton® A-401C based), S- 2 (FKM- 50:50 Viton® GBL 200S:600S), S- 3 (EPDM- Nordel IP 4520) were approximately in the ratio of 10:20:1. Price of Kalrez could be taken as 25- 30 times of S- 1 [204]. Given the small amount of elastomer used in critical sealing, INDIAN- SFR being an instance (250 kg maximum), the seal material cost is negligible (~ Rs. 1M and Rs. 1 crore using S- 2 and Kalrez for INDIAN- SFR; going to be lower for PHWR and AHWR) compared to the reactor as also indicated by estimates from other industries. Installation cost is independent of elastomer grade. It's the cost of unforeseen reactor shutdown and labour for regular seal replacement (reactor operating cost: ~ 15%, Section 1.2.2) from which the main economic benefits accrue [198]. Maximising seal life with high temperature elastomer (slower synergistic ageing rate) and impressive hot tensile properties (S- 2 and its variations; higher FOS and more room for ageing) is supported by virtually unchanged extrusion resistance (i.e. M_{50} and hardness or cross-link density) because of outstanding thermal stability of fluoroelastomers, demonstrated by 32 week accelerated ageing studies using S- 4. Standardising the process by adopting common generic elastomer reduces cost from both counts by ensuring synchronised replacement and minimising its number during reactor life. Extent of gain could be judged from comparative studies of S- 1, /S- 4 and S-2 used for INDIAN- SFR as tensile-hardness properties are correlated with extrusion resistance. The multiplicative gains in life (i.e. 50 y for S-2 vis-à-vis 10 y for S-1 and S-2, INDIAN- SFR context) indicates multiplicative gains in operating cost and efforts (+ maintenance hazard + additional quality control + additional specifications and records etc.) which outweighs the increased material and seal cost grossly, apart from significant safety, reliability and sustainability advantages. Klarez information bulletin indicates at 5 times gain in life vis-à-vis ordinary FKM and PTFE, similar to the expected gains using S- 2 (peroxide cured APA 50:50 blend OF Viton® GBL 200S:600S) with respect to S- 1 or S- 4 (bisphenol cured, based on Viton® A- 401C).

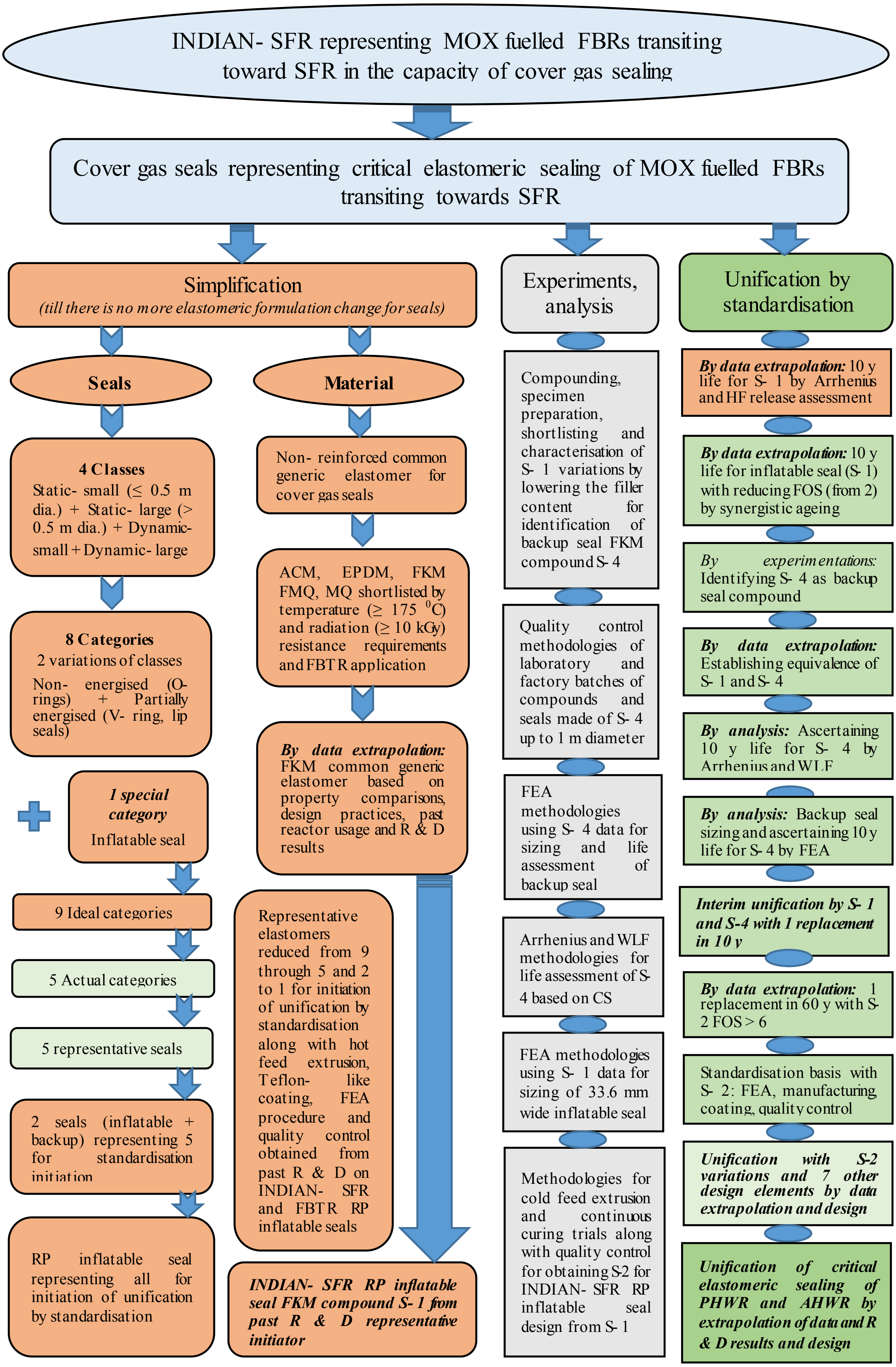
7.8 Sustainability Aspects

Perfluoroelastomers are in supply for critical elastomeric sealing applications for about four decades (the first supply was produced by M/s DuPont in the 1970s) as indicated by one of its early evaluations for Canadian PHWR and PWR in hot water [194, 198] Growing popularity of Kalrez in the most critical chemical and thermal environments could be seen from various product catalogues, handbooks and technical information bulletins [197- 200, 203- 205}. Perfluoroelastomers (FFKM) are sold in a wide range of trade names Kalrez, (DuPont), Chemraz (Greene, Tweed), Simriz, (Freudenberg Simrit), Isolast (Trelleborg Sealing Solutions), Parofluor (Parker Hannifin), Perlast, (Precision Polymer Engineering), Technoflon (Solvay Solexis) etc. [198] which provides initial assurance of continued seal- material supply for sustainability in the eventuality of non- availability of the suggested brand for unification by standardisation.

7.9 Chapter Conclusion

Applicability of peroxide fluoroelastomer grades over other generic elastomers and compounds in thermal reactors are driven by minimum number of synchronous replacements. It was shown by data extrapolation that varying the blend ratios of S- 2 could encompass PHWR sealing. For AHWR, the choice shifts to perfluoroelastomers (with silicone as a fall-back option) because of higher operating temperature. Chapter progress is indicated in Figure 7.1.

Fig.7.1. Outline of research methodologies: Progress in Chapter 7



Chapter 8

Conclusions and Future Research

8.1 Conclusions

I. The envisaged all- round growth of nuclear power across the globe (from 372 GWe presently to 930 GWe by 2050) towards decarbonisation through an evolutionary route (with SFRs likely to dominate the post- 2050 scenario) is being pursued by rapid increase in grid- capacity addition rate (from 5 GWe/ a to 20 GWe/ a) with parallel drives of NPP life extension (≥ 60 y) and ascertaining preparedness (and mitigation abilities) for BDBE, such as multiple external events (flood, earthquake etc,) on multiple- plant sites in particular. Safety is taken as the first and immediate priority during post- Fukusima age towards sustainability philosophy of GEN IV reactors with economy in NOAK plant, where design simplification and standardisation as well as harmonisation of codes and standards play defining roles.

II. The focus on ensuring negative coefficient of reactivity by innovative core design, minimising possibilities/ effects from sodium water reaction and decay heat removal (coupled with intensified studies on breach- resistant fuel clad as the first preventive barrier) creates a provision for continuing with elastomeric sealing of cover gas in FBRs (transiting towards the SFRs) to retain the flexibility of access and maintenance while bringing in equivalence with welded joints - by minimising permeation with better elastomers, maximising life with design (combining better material, predictive techniques and sound quality control) and maximising safety- reliability- economy by simplification, harmonisation and standardisation.

III. Material and its global reserve is the key enabler in decarbonisation as also in the growth of nuclear energy where classifying reactor systems in representative modules and materials, pursued by Europe and America during the past decade, finds a parallel in polymeric domain from this dissertation- the first of its kind.

IV. This thesis is about simplification, unification and standardization of critical elastomeric sealing (with material as cornerstone, FEA as facilitator and design as an all-encompassing foundation) for MOX fuelled, sodium cooled FBRs transiting towards SFR which has depicted a new research in the domain of organic materials pertaining to nuclear usage by applying novel techniques of data extrapolation (for experiment and analysis minimisation) and design towards 1 synchronised replacement of cover gas elastomeric seals during reactor life of 60 y in consonance with the global trends of energy, materials and design, sustainability philosophy and taking the Gen IV developments into perspective.

V. The research was implemented by taking cover gas seals as the epitome of critical elastomeric sealing and INDIAN- SFR as the model for MOX- fuelled sodium- cooled FBRs, simplifying the cover gas seals (categorisation and classification) into 9 representative categories and elastomeric formulations (and abstracting further into 4), identifying high- temperature non-reinforced fluoroelastomers as the common generic elastomer across categories based on 1 set of representative operating requirements for the 9 categories, defining RP inflatable and backup seals as architypes for the large diameter (extruded) static and dynamic sealing categories and identifying the inflatable seal as well as its elastomeric formulation as cornerstone for subsequent tailoring to other TS seals and complete unification by standardising 7 other elements of design i.e. i) Seal sizing, optimisation and design by FEA, ii) Seal manufacture by cold feed extrusion (continuous pressure-less cure,> 0.5 m diameter) and injection moulding (≤ 0.5 m diameter), iii) Teflon- like antifriction coating of dynamic sealing surface based on PECVD, iv) Stress- strain based life assessment by FEA, v) Stress- strain and CS based life assessment by Arrhenius and WLF methodologies, vi) Quality control and, vii) R & D.

VI. Simplification, unification and standardisation was carried out using a common set of material failure parameters (limits of Principal stress- strain with a FOS on ET- T_s and E_b, CS, HF release, tear strength) and functional- failure- parameter (contact pressure) contributing towards seal failure (leakage and drag), where a FOS of 2 was applied to ascertain 10 y life for the INDIAN- SFR RP inflatable seal by FEA along with an estimation of HF release as well as parallel assessment of life under synergistic ageing by data extrapolation using bisphenol cured, Viton® A- 401C based compound (S- 1) developed for the inflatable seals (suitable for hot feed extrusion and autoclave based production) as part of earlier (prior to this research) development pertaining to *Special Elastomeric Components* (elastomer, seal, coating) of reactors and laboratories.

VII. Inadequacy of the FOS (2, S- 1) in taking the life beyond 10 y, profile collapse of fluoroelastomer inflatable seal under pressurised cure, necessity of better quality seal with single end joint and reproducibility during repeat production, requirement of maximising FOS for uniform maximisation of TS- seal-life through tailoring of inflatable seal compound etc. resulted in adoption of a new extrusion process (cold feed extrusion and continuous cure) as part of this research which did also result in an improved compound based on peroxide cured, 50:50 blend of Viton®600S and 200S (S- 2, made of APA) with better resistance to permeation, mist, radiation and a vastly improved life from significantly improved FOS (> 6) due to much better hot tensile properties and lower modulus- attributed to much lower ionic interactions in the APA fluoroelastomer at RT.

VIII. Ascertaining 50 y of life for S- 2 (with 10 y confirmed life of S- 1 as foundation) by an innovative approach of progressive introduction of synergistic ageing elements (temperature, radiation, mist, air) in eroding the FOS and a similar methodology of FOS enhancement for the new formulation (S-2) through data extrapolation was combined with an original synthesis of reactor and seal design elements to confirm feasibility of achieving 1 replacement for INDIAN- SFR RP inflatable seal in 60 y and its subsequent tailoring for unification of the cover gas seals, taking material as the first module of standardisation.

IX. The absence of a comprehensive outlook (national and international) to bring nuclear-elastomeric- sealing on a common foundation (rather than addressing component specific problems) and a consolidated approach for predictive techniques (FEA, Arrhenius and WLF methodologies), along with an all- encompassing framework of quality control, (by exploiting the inherent elastomer-sizing- manufacture- coating- life correlations)has been addressed in this thesis with standardisation and unification of the remaining 7 modules where the development of bisphenol cured, Viton® A-401C based formulation (S- 4), its sizing- optimisation- design by FEA and ascertaining 10 y of life by predictive techniques (in combination with data extrapolation and design) contributed significantly.

X. Complete unification of cover gas sealing by simplification into 9 ideal categories and standardisation of 8 modules have produced the following results.

XI. The aim of synchronised cover gas seal replacements (1 in 60 y) for MOX fuelled Indian FBRs transiting towards SFRs (represented by INDIAN- SFR) could be realised by standardising the material module with peroxide cured, FKM compound S- 2 made of APA (INDIAN- SFR RP inflatable seal compound) and its 4 major variations (2 each, for static and dynamic) supported by FEA based sizing, life assessment and design (based on T_s and E_b with a FOS) with a uniform approach of plane- strain (initial approximation) and axisymmetric (finalisation) conditions, Mooney- Rivlin material model (normal operation) and 3- term Mooney-Rivlin as well as Ogden constitutive relations for special sealing and accidental conditions involving strain in excess of 50 %. Arrhenius and WLF methodologies completes the circle for life assessment and design involving CS and specimens under strain in a complimentary way with cross-verifications as necessary. The manufacturing module is similarly standardised with 2 processes (cold feed extrusion and continuous cure; injection moulding) for small (≤ 0.5 m) and large (> 0.5 m) diameter cover gas seals with dedicated extrusion and moulding machines (1 each), supported by 2 PECVD based Teflon- like coating procedures corresponding to large/ small diameter cover gas seals with 2 sets of adhesion strength (≥ 4.5/ 7 MPa) as well as thickness (3- 5/ 5- 10 μm) requirements. Tables 5.7- 5.8 provide the standardised quality control framework (along with the standards) for large diameter, extruded, static and dynamic seals supported by material and coating specifications (Tables 3.4-3.5) for future implementation of unification. Future R & D standardisation comprise of 7 standardised modules for implementation along with an approach of design options and R & D minimisation based on survey. The whole scheme of standardisation and unification provides a standardised design framework for the cover gas seals of Indian and global SFRs alike for sustainability which could result in a design code in smooth interfacing with commercial FEA codes i.e. ABAQUS, ANSYS and MARC.

XII. This scheme of simplification, unification and standardisation is applicable equally to metallic fuelled FBRs as the operating requirements of cover gas seals are marginally different.

XIII. The APA FKM formulation of INDIAN- SFR RP inflatable seal (S- 2) based on a blend of peroxide cured Viton® GBL 200S and 600S has natural applicability for unifying the PHWR elastomeric barriers by varying the blend ratio (from the 50:50 for FBRs), operating at a maximum temperature, pressure and γ radiation range of 200 ^{0}C, 10 MPa and 10- 100 kGy respectively with usual replacement span varying from 3 months to 5 y. Higher operating temperature (300 ^{0}C) of the critical elastomeric sealing for AHWR expands the material selection further to grades of perfluoroelastomers, with silicone rubber as an option.

XIV. A closer look at the operating requirements of INDIAN- SFR, PHWR and AHWR provides an indication that a variety of 10- 12 well characterised compounds (mostly based on peroxide cured blend of APA fluoroelastomers) could encompass the entire panorama of critical elastomeric sealing applications of Indian nuclear genre.

XV. Feasibility of implementation of the scheme for Indian FBRs hinges on CS-resistance- maximisation as a prime element of future R & D where the already- achieved life of 15-20 y in static O- ring seals of FBTR (made of fluoroelastomers and silicone rubber) provide the substantiation.

XVI. Feasibility of the scheme for Indian PHWRs depends on compatibility demonstration of S- 2 variations with coolant in terms of effects from volatile release, where purity of peroxide cure and estimated HF release (0.4 g for INDIAN- SFR RP inflatable seal in 50 y at 120 ^{0}C and 10 kGy of γ dose) provides substantiation considering smaller size and shorter life (5 y, maximum) of PHWR elastomer seals at a maximum temperature and cumulative γ dose of 200 ^{0}C and 100 kGy respectively.

XVII. A minimum gain of 10 fold in R & D effort, cost and time for future implementation of unification in FBR domain is envisaged based on minimisation of compounds, standardisation of R & D and past gains in inflatable- backup seal developments in context of international progress. Future R & D could be reduced further by choosing representative compounds (out of 5) and seals from static and dynamic domains to enhance the gains to 20 times. The benefits are expected to improve by another step*(≥ 100 weeks of accelerated ageing at 3 elevated temperatures, with periodic specimen withdrawals and 5- specimen average for test results)* as this dissertation has demonstrated that for elastomers with low filler content (i.e. static cover gas seals), only one strain rate (500 mm/ min) for tensile testing (quality control, FEA, Arrhenius, WLF, design) is sufficient for nuclear-grade characterisation/ qualification of compounds (i.e. variations of S- 2) vis-à-vis 3 rates (5- 50- 500 mm/ min) for compounds of dynamic seals. The gains envisaged in reactor downtime, operating cost (15% out of the total cost) and avoidance of maintenance hazards are expected to be substantial which could be illustrated by comparing the 827 seal replacements (out of ~ 5000) per year in FBTR vis-à-vis 1 envisaged synchronised replacement in 60 y- supported by data from Canadian PHWRs and those FBRs (KNK I and II). Substantial reinforcement in safety and reliability is readily evident by gross simplification of design, operation and maintenance which reduces unforeseen failure probabilities and the operating synchronisations, documentations as well as record- update efforts considerably. These potential benefits are maximised by standardised predictive techniques, R & D and quality control which do also maximise reproducibility within and across reactors to minimise cost and maximise safety, along with reliability.

XVIII. Cumulative potential impact of the current research in Indian context could be judged by all- round envisaged short- term (63 GWe by 2032) and longer- term (100 GWe by 2050) growth of nuclear power where FBRs occupy a space of 10 GWe in shorter term (2032) and indigenous PHWRs carry a potential of 20 GWe considering 0.2 Mt of uranium reserve.

XIX. This research has applied a phenomenological approach (rather than molecular) as a novelty in Pasteur's Quadrant (application of science rather than applied science) to provide a comprehensive scheme for simplification, unification and standardisation of critical elastomeric sealing in nuclear domain, so far unaddressed.

8.2 Future Research

Complete nuclear characterisation of S-2 and its 4 variations comprising of mist compatibility in long term, permeability, processability (curing characteristics and rheological behaviour), extrudability, tribology, determination of physico- mechanical properties (stress- strain, modulus, Ts, Eb, hardness, CS etc.at various strain rates and specimen conditioning, RT and elevated operating temperatures) on unaged, short- term aged and long- term aged (at least 100 weeks, up to 225 0C) specimens along with sizing, life assessment and design by FEA, Arrhenius and WLF methodologies form the essential gamut of future work ideally. The unchanged blend ratio (50:50) in S-2 and its 4 various for the cover gas seals of FBRs transiting towards SFRs indicate similar FOS, fluid compatibility and processability which signify that choosing 2-3 representative compounds out of the 5 from static and dynamic domains (including S-2) for complete characterisation may be sufficient. Similarly, representative seals could be chosen from 5 categories (extruded large diameter static O- rings, moulded small diameter dynamic O- rings, moulded small diameter dynamic V-ring/lip seals, large diameter extruded dynamic inflatable seal and large diameter extruded static backup seal) derived from the initial 9 categories for sizing, design and life assessment implementations by FEA, Arrhenius and WLF methodologies using the qualified and designated compounds. Performance validation of the representative seals in scaled down test rig with provisions for accelerated ageing (up to 100 weeks and 225 0C) of test seals is an essential part of the whole process. Assessment of the potential effects of cumulative release of HF (induced by γ dose) from the cover gas seals on reactor performance in long term form an important part of future research, as also the determination of strain effects on ageing and that of transition from moulded specimens to moulded/ extruded products by using specimens extracted from seals for accurate quantification of FOS erosion and safe life. Attaining an uniform aim of 1 synchronised replacement of cover gas seals during reactor life of 60 y is influenced significantly by another key failure parameter i.e. CS. This research has shown that attaining 1 replacement during 60 y of life based on FOS maximisation could be a trivial issue (with the proposed scheme in this thesis) if the cumulative effects of HF release on long- term reactor performance is ascertained. Demonstration of 10 y of life based on a CS threshold of 80% and several improvement suggestions on this count in tailoring of S- 2 notwithstanding, there is significant scope in future research to maximise CS resistance further (by finer compositional variations with fillers and additives in the 50:50 blend) towards attainment of uniformity of maximum life i.e. 1 replacement in 60 y.

Production and installation of inflatable seals with Teflon- like coating by PECVD in INDIAN- SFR RPs made of S-2, completion of development of PECVD Teflon- like coating for the inflatable seal mating steel shells and development of adhesion-less end joining as well as automated glue dispensation technique are some of the component- oriented future research in Pasteur's coordinate which could complete the circle of simplification, unification and standardisation for Indian FBRs transiting towards SFRs.

Demonstration of the possible effects of release of low molecular weight volatiles (such as HF) induced by γ dose on long- term reactor performance is key to the implementation of unification in the PHWR domain along with validation of extrusion resistance of the seals. For AHWR the future research could be maximising the temperature rating of peroxide cured perfluoroelastomers for life maximisation and unification along with explorations with silicone rubber.

References

1. President, CoP21, 2015. Adoption of the Paris Agreement, proposal by the President. Draft decision -/CP.21. In: FCCC/CP/2015/L.9/Rev.1. Conference of the Parties, twenty-first session, United Nations Framework Convention on Climate Change, Paris, 30 Nov. – 11 Dec. 2015.
2. The Hindu, 2015. India to press for equity at climate talk despite pressure. Report. 25 November 2015.
3. Narain, S., 2015. Path breaking work on climate justice. Article. Times of India (TOI), 30 November 2015.
4. OECD/IEA, 2015. World energy outlook 2015: Executive summary. International Energy Agency (IEA), Organisation for Economic Co-operation and Development (OECD), Paris.
5. OECD/IEA and OECD/NEA, 2015. Technology roadmap: Nuclear energy, 2015 edition. IEA and Nuclear Energy Agency (NEA), OECD, France.
6. Kelly, E. J., 2015. International perspective on the future of nuclear power. Presentation. 3rd Gen IV International Forum (GIF) Symposium presentations, Makuhari Messe, Japan, May 2015.
7. Garg, P., 2012. Energy scenario and vision 2020 in India. Journal of Sustainable Energy and Environment 3, 7-17.
8. IAEA, 2015. Nuclear technology review. International Atomic Energy Agency (IAEA), Vienna, August.
9. Sinha, N. K., Raj, Baldev, 2013. Development of inflatable seals for the rotatable plugs of sodium cooled fast breeder reactors: a review, Part I—Key areas. Kerntechnik78/5, 380–399.
10. Sinha, N. K., Raj, Baldev, 2015. Choice of rotatable plug seals for Prototype Fast Breeder Reactor: Review of historical perspectives. Nucl. Eng. Des. 291, 109- 132.
11. CEA, 2012. 4th- Generation Sodium- Cooled Fast Reactors: The Astrid technology demonstrator. Nuclear Energy Division, cedex- France, December.
12. Bhoje, S. B., Chellapandi, P., 1996. Prototype fast breeder reactor main options. IAEA-TECDOC-907, Vienna, pp. 71-82.
13. Jain, S. K., 2010. Inevitability of nuclear power. Presentation. CMD, NPCIL & BHAVINI.
14. Chetal, S. C., Chellapandi, P, 2013. Indian fast reactor technology: Current status and future programme: Sadhana 38 (5), 795- 815.
15. TOI, 2015. Japan gives India its second most important nuclear deal. Report. 12 December 2015.
16. TOI, 2015. India closing in on Westinghouse deal to build 6 nuclear reactors. Report. Reuters, 24 December 2015.
17. *Data Courtesy: Shri Sandip Singharoy, FormerPD Jaitapur, NPCIL.*
18. Chellapandi, P., 2015: Status of PFBR. Presentation. Joint IAEA- GIF Technical Meeting Workshop on Safety of Sodium Cooled Fast Reactors, Vienna, 23- 24 June 2015.
19. Chellapandi, P., 2015: Status of FBR 1 & 2. Presentation. Joint IAEA- GIF Technical Meeting Workshop on Safety of Sodium Cooled Fast Reactors, Vienna, 23- 24 June 2015.
20. Stanculescu, A., 2012. Liquid metal cooled fast reactors: Worldwide overview. Presentation. International Workshop on Innovative Nuclear Reactors Cooled by Heavy Liquid Metals: Status and Perspectives, Pisa, Italy, 17-20 April 2012.

21. *Data Courtesy: Shri G. Srinivasan, Former Director RDG, IGCAR.*
22. OECD/ NEA, 2014. Technology roadmap update for Generation IV nuclear energy systems. OECD/ NEA on behalf of GIF, January.
23. Sinha, R. K., Kakodkar, A, 2006. Design and development of the AHWR- the Indian thorium fuelled innovative nuclear reactor. Nucl. Eng. Des. 236, 683- 700.
24. BARC. AHWR300- LEU: Advanced Heavy Water Reactor with LEU- Th MOX Fuel. Bhabha Atomic Research Centre (BARC), DAE, Mumbai.
25. OECD/IEA, 2015. World energy outlook 2015 factsheet. IEA, OECD, Paris.
26. Arpe, H. J., 2010. Industrial Organic Chemistry. 5th ed., WILEY- VCH Verlag GmbH & Co. KGaA, Wienheim.
27. Yvon, P., Carre, F, 2009. Structural materials challenges for advanced reactor systems. J. Nucl. Matl. 385, 217–222.
28. Busby, J., 2010. Advanced Structural Materials Development: Accomplishments and Plans. Presentation. NEUP Workshop, U. S. Department of Energy (DOE), 28 July 2010.
29. Fazio, C., 2012. Nuclear materials research in EU: Present status and future perspectives. Presentation. Seminar on Generation IV Nuclear Energy Systems, Rise, Denmark, 29- 31 October 2012.
30. Busby, J. T., 2012. Challenges for reactor materials. Presentation. Nanonuclear Workshop, U. S. DOE, Rice University, 28 February 2012.
31. Natesan, K., Meimei, L., 2013. Materials performance in sodium- cooled fast reactors: Past, present and future. Presentation. Int. Conf. Fast Reactors and Related Fuel Cycles: Safe Technologies and Sustainable Scenario, U. S. DOE, Rice Paris, France, 4- 7 March 2013.
32. Sham, S., 2013. Advanced reactor concepts program: ARC materials development – accomplishments and plans. Presentation. DOE- NE Materials Crosscut Coordination Meeting, U. S. DOE, 30 July 2013.
33. Malerba, L., 2014. The Joint Program on Nuclear Materials of the European Energy Research Alliance (EERA JPNM). Presentation. EERA, 2014.
34. EERA, 2015. Vision paper for the EERA Joint Programme for Nuclear Materials. EERA, February 2015.
35. U. S. DOE, 2011. Critical materials strategy. Report.
36. Sinha, N. K., Raj, Baldev, 2010. Development of polymeric applications for sodium cooled fast breeder reactors: chronicles of inception, progress and achievements. Nucl. Eng. Des. 240, 2925–2947.
37. Sinha, N. K., Raj, Baldev, 2012. Unification of reactor elastomeric sealing based on material. Nucl. Eng. Des. 243, 404–418.
38. Ravishankara, A. R., 2015. Why do we do the science we do. Current Science 109 (4), 659-660.
39. Crawford, R. J., 1985. Plastics and Rubbers: Engineering Design and Applications. Mechanical Engineering Publications Ltd., London.
40. Metcalfe, R., Baset, S. B., Lesco, R., Seleander, W. N., 1989. Modelling of space shuttle rocket O-rings. Atomic Energy of Canada Limited (AECL) Report. AECL-11167, Chalk River Laboratories, Ontario, Canada.
41. Sinha, N. K., 2009. Design, development, production, validation and performance of organic components to ensure safety of nuclear applications: a finite element analysis perspective. In: Proc. Int. Conf. Peaceful Uses of Atomic Energy (Peacefulatom), Vol. I, New Delhi, 29 Sept. – 1 Oct. 2009, pp. 186- 187.

42. Sinha, N. K., 2011. Design, development, production, validation and performance of organic components to ensure safety of nuclear applications: a finite element analysis perspective. In: Dharne, S.P., Hajela, S., Sahaya, R.R. (Eds.), Reactor Systems & Applications: Thematic Volume evolved from Peacefulatom 2009. BARC, DAE, Mumbai, pp. 131–141.
43. Sinha, N. K., Raj, Baldev, 2013. Development of inflatable seals for the rotatable plugs of sodium cooled fast breeder reactors: a review, Part II—R & D necessities and development across the world. Kerntechnik78/6, 463–476.
44. Finney, R. H., 1986. Springy finite elements model elastomers. Mach. Des. May, 87–92.
45. Sinha, N. K., Chetal, S. C., Raj, Baldev, 2012. Simplification and unification of elastomeric sealing for fast breeder reactors based on Fluorohydrocarbon rubber. In: Mendez-Vilas, A. (Ed.), Fuelling the Future: Advances in Science & Technology for Energy Generation, Transmission and Storage. Brown Walker Press, Boca Raton, Florida, pp. 445–451.
46. Berg, G. E., 1977. CRBRP design and test results for fuel handling systems, plugs and seals. Conf-771217-1.
47. Sharbaugh, J. E., 1979. A recommended design approach for breeder reactor cover gas seals. J. Eng. Power 101, 647-650.
48. Horton, P. H., 1980. LMFBR inflatable elastomeric seals: Development and test history. DOE Research and Development Report. Rockwell International Corp., Canoga Park, CA, USA.
49. Mausbeck, H., 1976. Materials experience with large sodium loops and components. In: Cooper, M. H. (Ed.), Proc. Int. Conf. on Liquid Metal Technology in Energy Production. Pennsylvania, pp. 67–74.
50. Marth, W., 1994. A review of the collaborative programme on the European Fast Reactor (EFR). In: Proc. 26th Meeting of the International Working Group on Fast Reactors. (IWGFR). IAEA-TECDOC-741, IAEA, Vienna, pp. 103–137.
51. Wittich, K. C., Wensel, R., Larose, R., Kuran, S., 1995. Upgrading elastomer seals for nuclear service. Atomic Energy of Canada Limited (AECL) Report. AECL-11385, Chalk River Laboratories (CRNL), Ontario, Canada.
52. Parks, M. B., Clauss, D. B., 1992. Performance of containment penetrations under severe accident loadings, Nucl. Eng. Des. 134, 177-197.
53. Hessheimer, M. F., Dameron, R. A., 2006. Containment integrity research at Sandia National Laboratories: An overview. SNL Report. NUREG/CR-6906, SAND2006-2274P.
54. EPRI, 2007. Plant support engineering: Elastomer handbook for Nuclear Power Plants. Electric Power Research Institute (EPRI) Report. 1014800, Palo Alto, CA, USA.
55. Chivers, T. C., George, A.F., 1984. Factors influencing the design and assessment of elastomer seals for nuclear fuel transport flasks. Central Electricity Generating Board (CEGB) Report. TPRD/B/0442/N84, Berkley Nuclear Laboratories.
56. Chivers, T. C., George, A. F., 2004. EPDM and fluorocarbon seal materials: A comparison of performance for nuclear fuel transport flasks. 14th International Symposium of Packaging and Transport of Radioactive materials (PATRAM), Berlin, Germany, 20- 24 September 2004.
57. NEA, 1999. Survey on organic components in NPPs. OECD/ NEA Report. NEA/CSNI/R (98)7, Paris.
58. Burnay, S.G., 2001. An overview of polymer ageing studies in nuclear power industry. Nucl. Inst. Meth. Phys. B 185, 4–7.
59. Clough, R.L., Gillen, K.T., Quintana, C.A., 1984. Heterogeneous oxidative degradation in irradiated polymers. SNL Report, NUREG/CR-3643, SAND83-2493. Sandia National Laboratories (SNL), Albuquerque, NM.

60. Burnay, S.G., Hitchon, J.W., 1985. Prediction of service lifetimes of elastomeric seals during radiation ageing. J. Nucl. Mater. 131, 197–207.

61. Clough, R.L., Gillen, K.T., 1990. Stabilizer additives in ionizing radiation environments under oxidizing conditions. Polym. Degrad. Stabil. 30, 309–317.

62. Achenbach, M., 2000. Service life of seals: numerical simulation in sealing technology enhances progress. Comput. Mater. Sci. 19, 213–222.

63. IAEA, 2000. Assessment and management of ageing of major Nuclear Power Plant components important to safety, volume 1 and 2. IAEA Report. IAEA-TECDOC- 1188, Vienna.

64. Celina, M., Gillen, K.T., Assink, R.A., 2005. Accelerated ageing and lifetime prediction: review of non-Arrhenius behavior due to two competing process. Polym. Degrad. Stabil. 90, 395–404.

65. IAEA, 2008. Nuclear Technology Review 2008, Vienna.

66. Placek, V., Kohout, T., Hnat, V., Bartonicek, B., 2009. Assessment of EPDM seal lifetime in Nuclear Power Plants. Polym. Test. 28, 209–214.

67. Skidmore, T. E., 1998. Performance evaluation of o- ring seals in model 9975 packaging assemblies (U). WSRC Report. WSRC-TR-98-00439 (Rev. 0), Westinghouse Savannah River Company (WSRC), Aiken, SC; December.

68. Skidmore, T. E., 2003. Performance evaluation of o- ring seals in the SAFKEG 3940A package in KAMS (U). WSRC Report. WSRC-TR-2003-00198 (Rev. 0), WSRC, Aiken, SC; May.

69. Skidmore, T. E., Counts, K. M., Fox, E. B., Hoffman, E. N., Dunn, K. A., 2007. Aging performance of Viton® GLT O-rings in radioactive material packages. SRNL Report. SRNL-STI-2009-00420, Savannah River National Laboratory (SRNL), Aiken, SC.

70. Skidmore, T.E., 2008. Seal materials in shipping containers. WRSC-STI-2008-00282- S. Presentation at Material Science & Technology 2008 CTMA Workshop.

71. Skidmore, T. E., Hoffman, E. N., Daugherty, E. L., Dunn, K. L., 2009. Aging and surveillance of Viton® GLT O-rings in model 9975 shipping packages. SRNL Report. SRNL-STI-2009-00420. SRNL, Aiken, SC.

72. Hoffman, E. N., Skidmore, T. E., 2009. Lifetime prediction for model 9975 o- rings in KAMS. SRNL Report. SRNL-TR-2009-00259 (Rev. 0), SRNL, Aiken, SC; November.

73. Hoffman, E. N., Skidmore, T. E., Daugherty, W. L., 2010. Long-term leak tightness of o-ring seals in the 9975 shipping package. In: Proc. PVP 2010, 2010 ASME pressure vessels and piping (PVP) division conference (SRNL-STI-2010 037, PVPm2010-78086), Bellevue, WA, 18- 22 July 2010.

74. Sinha, N.K., Raj, Baldev, 2014. Leakage limits for inflatable seals of sodium cooled fast breeder reactor. Nucl. Eng. Des. 266, 194–198.

75. Medri, G., Strozzi, A., 1986. Stress–strain fields in compressed elastomeric seals and their extension to fracture mechanics. Rubber Chem. Technol. 59, 709–721.

76. Chivers, T. C., Hunt, R. P., 1978. The achievement of minimum leakage from elastomeric seals. In: Proc. 8th Int. Conf., BHRA, Durham, UK. Vol. 1, pp. F3–F31.

77. Muller, H. K., Nau, B. S., 1998. Fluid Sealing Technology Principles and Applications. 1st ed. Marcel Dekker, New York.

78. Kurzeka, W. J.; Welch, F. H., 1975. Permeation leakage and compression set testing of elastomeric seals for LMFBR use. Report. AI-AEC-13146, Rockwell Inc., CA, U.S.A., April.

79. Rockwell Inc., 1973. Quarterly technical progress report: Sodium technology and cover gas seal development programs, Jul.-Sept. 1973. Report. AI-AEC- 13113, Rockwell Inc., CA, U.S.A., November.

80. Lyons, J. L., 1982. Lyon's valve designers' handbook. Van Nostrand Reinhold Company, pp. 405–450.
81. Sinha, N. K., Raj, Baldev, 2011. Optimization of material and production to develop fluoroelastomer inflatable seals for sodium cooled fast breeder reactor. Nucl. Eng. Des. 241, 739–751.
82. Ito, M., 1996. Radiation induced degradation of fluorine containing elastomers at various temperatures. Radiation Physics and Chemistry 47 (4), 607–610.
83. Arcella, V., Albano, M., Barchiesi, E., Brinati, G., Chiodini, G., 1993. Development of new nucleophile resistant vinylidene fluoride fluorocarbon elastomers. Rubber World.
84. Bowers, S., Eric, T., 2001. New FKM class provides low-temperature service. Rubber and Plastics News 30(15), p. 14-6.
85. Moore, A. L., 2006. Fluoroelastomers Handbook: The Definitive User's Guide and Databook. 1st ed. William Andrew Publishing, New York.
86. Horton, P.H., Oliva, R., Shimazaki, T., 1975. Cover Gas Seals: FFTF-LMFBR Seal Test Program. Progress Report: Jan.–Mar. 1975. DOE/SF/71011-T2. Rockwell Inc., WA, USA.
87. Sinha, N.K., Mukhopadhyay, R., Dhupia, D., Das Gupta, S., Raj, Baldev, 2011. Development of fluorocarbon rubber for backup seals of sodium cooled fast breeder reactor. J. Mater. Des. 32, 5141–5153.
88. Stevens, C. A., 1988. Prediction of fracture in unconstrained elastomeric O- ring seals. CEGB Report, RD/M-1782/RR89.
89. Eude, J. C. (Translation-Ellyn, G.), 1976. Use of elastomers in nuclear reactors. Report. (Revue Generale Du Caoutchoic Et Des Plastiques, 49/5, 1972, p. 395) ANL-TRANS-1003, ANL, Illinois, U.S.A.
90. Sealing Technology, 1996. Inflatable seals for gas tight cleaning unit. Sealing Technology 26, 1.
91. Sinha, N. K., 2006. Teflon coating for nuclear applications. In: Alphonsa, J. (Ed.), Plasma Processing Update, Golden Jubilee Issue. FCIPT, IPR, Gandhinagar, pp.36–40.
92. Crette, J. P., 1971. Industrial aspects of fast breeder reactor construction. Kerntechnik 13, 491-495.
93. Chikazawa, Y., Kotake, S., Sawada, S., 2011. Comparison of advanced fast reactor pool and loop configurations from the viewpoint of construction cost. Nucl. Eng. Des. 241, 378–385.
94. Lagrange, M.; Chevallier, R.; Perona, M. (Translation-Ellyn, G.), 1975. Problems raised by the design and manufacture of loading plugs for sodium cooled fast reactors. (Nuclex 1972, Paper 16) ANL-TRANS- 999, Illinois, U.S.A.
95. Pierantoni, F., 1976. Review of fast reactor activities in Italy March, 1976. IWGFR 9th Annual Meeting, Summary Report Part II & III. IWGFR/12, IAEA, Vienna.
96. Pierantoni, F., 1978. Review of fast reactor activities in Italy, April 1978. IWGFR 11th Annual Meeting, Summary Report Part II. IWGFR/24-2, IAEA, Vienna.
97. Pierantoni, F., 1979. A review of the Italian fast reactor programme, March 1979. IWGFR 12th Annual Meeting, Summary Report Part II. IWGFR/30- 2, IAEA, Vienna.
98. Pierantoni, F., 1980. A review of the Italian fast reactor programme. IWGFR/34-2, IAEA, Vienna.
99. Althaus, S., Jansing, W., Kesseler, R., Kirchener, G., Menck, J., 1974. Experience with handling core subassemblies in sodium cooled reactor KNK and test rigs. In: Proc. Int. Conf. Fast Reactor Power Stations, BNES, London, March 1974, p. 545.

100. Babilas, C., Hoeft, E., Kenkmann, W., Kirchner, G., Menck, J., 1975. Tests carried out for the SNR-300 handling and the influence of test results in view to design of handling equipment. In: Proc. European Nuclear Conference, Vol. 2, Paris, 21- 25 April 1975, p. 445.
101. Le Joint Francais (LJF): Membramatic sealing. Catalogue, LJF, Bezons, France, 1988.
102. Welch, F. H., 1978. Inflatable seal gamma radiation study: A Test Report. DOE/SF/70027-T5, Rockwell Inc., CA, U.S.A.
103. Parks, M. B., 1989. Evaluation of leakage behaviour of inflatable seals subjected to severe accidental conditions. Report. NUREG/CR-5394.
104. Parks, M. B., 1989. Performance of containment penetration under severe accidental loading. Report. SAND-89-1631C, SNL, NM, U.S.A.
105. Horton, K. E., 1986. Status of the fast reactor development in United States of America- April, 1985. IWGFR/56, p. 122.
106. Sackett, J. I., 1997. Operating and test experience with EBR- II, the IFR prototype. Prog. Nucl. Energy 31, 111- 129.
107. Parker Seals, 1992. Parker O-ring handbook. ORD 5700, Parker Seal Group, KY, U.S.A.
108. Gent, A. N. (Ed.), 1992. Engineering with rubber: how to design engineering components. Carl Hanser Verlag.
109. Stern, H. J., 1987. History. In: Blow, C. M., Hepburn, C. (Eds.), Rubber technology and manufacture. 2nd ed., Butterworths.
110. Kuruzeka, W. J., Welch, F. H., 1975. Design guide for reactor cover gas elastomeric seals. AI-AEC-13145, Atomics International Div., Rockwell Inc., CA, U.S.A.
111. Horton, P. H., Steele III, O. P., Asquith, J. G., 1975. Design considerations for dynamic elastomeric seals for LMFBR application. Trans. ANS 22, 224- 225.
112. Harrington, R., 1958. Elastomers for use in radiation fields, Part IV. Rubber Age, June.
113. Lions, N.; De Fremont, R.; Lemercier, G., 1986. R & D in support of the 1500 MWe fast neutron reactor in the light of the experience gained on Super Phenix. 14eme Symposium Sur La Fusion Controlee, Avignon, 8- 12 September 1986.
114. LJF, 1983. Seals for nuclear power. Product catalogue.
115. LJF, 1972. Elastomer seals for nuclear energy. Product catalogue, 10-72-207-an.
116. Shaktawat, P. R., Dhupia, D., Adhikary, A., Singh, R. K., Sinha, N. K., 2002. Performance evaluation of elastomeric inflatable seals based on FKM. Presented at Int. Conf. and Exhibition of Rubber and Allied Materials, New Delhi, 28- 30 November 2002.
117. Del Vecchio, R. J., 1992. Tensile testing of elastomers. In: Patricial, H. (Ed.), Tensile Testing. ASM International, pp. 135–146.
118. Brown, R. P., 1996. Physical Testing of Rubber. Chapman & Hall, London.
119. Brown, R. P., 2006. Physical Testing of Rubber. 4th ed. Springer, New York.
120. Nema, S. K., Kikani, P., Raole, P. M., Ganeshprasad, K. S., Sinha, N. K., Raj, B., 2004. Study of Teflon-like films grown on elastomer and steel surfaces in radiofrequency plasma. Presented at DAE Workshop on Plasma Surface Engineering, BARC, Mumbai, 23- 24 September 2004.
121. Nema, S. K., Kikani, P., Raole, P. M., Ganeshprasad, K. S., Sinha, N.K., 2005. Study of Teflon like films grown on steel and elastomer surfaces using RF plasma. Presented at National Symposium (IVS-NS)-2005, Indian Vacuum Society, Gandhinagar, 16- 18 November 2005.
122. John, P. I., 2006. Commercial spin-off of fusion technologies. Nuclear India 39 (11–12), 24–29.

123. Banerjee, S., Sinha, N. K., Gayathri, N., Ponraju, D., Raj, B., 2007. Detecting onset of chain scission and crosslinking of γ-ray irradiated surfaces using frictional force microscopy. J. Phys. D: Appl. Phys. 40, 834–839.

124. Satyaprasad, A., Nema, S. K., Sinha, N. K., Raj, Baldev, 2010. Deposition of thick and adherent Teflon-like coating on industrial scale stainless steel shell using pulsed DC and RF PECVD. J. Appl. Surf. Sci. 256, 4334–4338.

125. Chellapandi, P., Chetal, S. C., Raj, B., 2009. From 500 MWe prototype fast breeder reactor to 500 MWe commercial fast breeder reactors. In: Reddy, A.V.R., Venugopal, V., Sinha, R.K., Banerjee, S. (Eds.), Proc. Int. Conf. on Peaceful Uses of Atomic Energy–2009, vol. II. BARC, DAE, Trombay, Mumbai, pp. 490–491.

126. Sinha, N. K., Raj, Baldev, 2012. Basis of property limits for inflatable seal fluoroelastomers. Nucl. Eng. Des. 244, 100–108.

127. Sinha, N. K., Suresh Kumar, S., Babu, A., Raj, Baldev, 2013. Quality control strategy for critical elastomeric seals of sodium cooled fast breeder reactor. Book of abstracts. In: International Conference on Rubber and Rubber-like Materials (ICRRM 2013b), 6- 9 March 2013, IIT Kharagpur, West Bengal, pp. 169–170.

128. DAE, 2006. Towards reliable and indigenous polymer materials for application in Nuclear Power Plants (NPP). In: Raj, Baldev (Ed.), Atoms with Mission: A Golden Jubilee Commemorative Volume (1954–2004), Vol. II. Department of Atomic Energy (DAE), Government of India, Mumbai, pp. 211–212.

129. IGCAR, 2003. Development of inflatable seals. In: IGC Annual Report 2002. IGCAR, DAE, Kalpakkam, pp. 58.

130. IGCAR, 2005. Experience with replacement of inflatable seals of both rotatable plugs. In: IGC Annual Report 2004. IGCAR, DAE, Kalpakkam, p. 14.

131. IGCAR, 2008. Qualification of backup seals. In: IGC Annual Report 2007. IGCAR, DAE, Kalpakkam, p. 22.

132. IGCAR, 2009. Testing of backup seal. In: IGCAR Annual Report 2008. IGCAR, DAE, Kalpakkam, p. 23.

133. Raj, B., 2006. Plasma applications in nuclear industry. In: Alphonsa, J. (Ed.), Plasma Processing Update, Golden Jubilee Issue. FCIPT, IPR, Gandhinagar, pp. 13–16.

134. Sinha, N. K., 1995. Finite element analysis of elastomer ring seals. In: Project Report. Machine Elements Laboratory, IIT Madras, Madras.

135. Sinha, N.K., 1999. Inflatable seal development in fast breeder and thermal reactors: a review. Internal Note. TFDOE/TECHDOC/05. IGCAR (1999).

136. Sinha, N.K., 2000. Elastomers for Indian fast reactors: IGCAR and defence join hands to realize indigenization. IGC News Lett. 44, 9.

137. Sinha, N. K., Kasiviswanathan, K. V., Chander, S., Vasudeva Rao, P.R., Veerasamy, R., 2000. Development of inflatable seal for prototype fast breeder reactor: methodology and input. In: Proc. Conf. Recent Advances in Polymers and Composites, Kanpur, India, p. 727 (2000).

138. Sinha, N.K., 2001. Conceptual design of inflatable seals for PFBR rotatable plugs. Internal Note. TFDOE/TECHDOC/10/R-0. IGCAR.

139. Sinha, N. K., Raj, Baldev, 2011. PFBR backup seal development. Nucl. Eng. Int. 56 (March), 36–38.

140. Sinha, N. K., Raj, Baldev, Mukhopadhyay, R., 2011. Development of fluoroelastomer backup seal: snapshots of a novel effort in rubber engineering for a critical application (Part I). Sealing Technol. 4, 10–13.

141. Sinha, N. K., Raj, Baldev, Mukhopadhyay, R., 2011. Development of fluoroelastomer backup seal: snapshots of a novel effort in rubber engineering for a critical application (Part II). Sealing Technol. 5, 9–12.
142. Sinha, N. K., Chetal, S. C., Raj, Baldev, 2012. Simplification of elastomeric sealing of fast breeder reactors based on a few material formulations, analysis procedures, coating technique and manufacturing processes. In: Book of Abstracts, The Energy Materials Research Conference—EMR 2012, June 20–22, 2012, Torremolinos, Spain, p. 303.
143. Sinha, N. K., Chetal, S. C., Raj, Baldev, 2012. Unification of elastomeric sealing applications of fast breeder reactors based on peroxide cured fluorohydrocarbon rubbers. Book of Abstracts, EMR 2012. Formatex Research Centre, Spain, pp.306.
144. Sinha, N. K., Ghosh, P., Saha, A., Mukhopadhyay, R., Raj, Baldev, Chetal, S. C., 2012. Design optimization of backup seal for sodium cooled fast breeder reactor. J. Mater. Des. 36, 396–407.
145. Sinha, N. K., Mukhopadhyay, R., Raj, Baldev, 2013. Mechanical behavior of fluoroelastomer considering long term ageing. Nucl. Eng. Des. 254, 89–96.
146. DuPont, 2003. Viton® A-401C. Technical Information Bulletin Rev. 2. VTE-H71110-00-C0203. DuPont Performance Elastomers, USA.
147. DuPont, 2003. Viton® A-200. Technical Information Bulletin. Rev. 2. VTE-H71121-00-C0203, DuPont Performance Elastomers, USA.
148. DuPont, 2005. Viton® GBL-200S. Technical Information Bulletin. VTE-A10165-00-B0604, DuPont Performance Elastomers, USA.
149. DuPont, 2005. Viton® GBL-600S. Technical Information Bulletin. VTE-A10086-00-C0604, DuPont Performance Elastomers, USA.
150. DuPont, 1978. Radiation resistance of Viton®. DuPont Data Bulletin. V-A-4-303. DuPont Dow Elastomers, USA.
151. DuPont, 2005. Handling precautions for Viton® and related chemicals. Technical Information Bulletin. VTE-H71129-00-E1105, DuPont Performance Elastomers, USA.
152. National Aeronautics and Space Administration (NASA), 1970. NASA Space Vehicle Design Criteria (Structures): Nuclear and Space Radiation Effects on Materials, Monograph, NASA SP-8053, USA.
153. LJF, 1972. Elastomer Seals for Nuclear Energy: Recommendations for the Selection of Elastomer Compounds for the Principal Applications in Nuclear Engineering. 10-72-207-an, Nuclear Department, LJF, Bezons, France.
154. Scheirs, J. (Ed.), 1997. Modern Fluoropolymers: High Performance Polymers for Diverse Applications. 1st ed. John Wily & Sons Ltd., New York.
155. Drobny, J. G., 2009. Technology of Fluoropolymers. 2nd ed. CRC Press, Boca Raton.
156. DuPont, 2003. Viton processing guide. Technical information bulletin (Rev. 2), VTEH90171-00-A0703, DuPont Dow Elastomers, USA.
157. DuPont, 2005. Viton selection guide. Technical information bulletin (Rev. 6), VTEH68134-00-F1105, DuPont Performance Elastomers, USA.
158. Finn, P.A., 1991. Guidelines to achieve seals with minimal leak rates for HWR-NPR coolant system components. Argonne National Laboratory (ANL) Report, ANL- 90/29.
159. Finn, P.A., 1992. Sealing systems for a heavy water reactor. Fusion Technol. 21, 377–382.
160. Dassault Systems, 2013. Abaqus 6.13 Documentation, Dassault Systemes Simulia Corp., RI, USA.

161. Makino, A., Hamburgen, W. R., Fitch, J. S., 1993. Fluoroelastomer pressure pad design for microelectronic applications. Western Research Laboratory (WRL) Research Report, 93/7, CA, USA, November.

162. Charlton, D. J., Yang, J., The, K. K., 1994. A review of methods to characterise rubber elastic behavior for use in finite element analysis. Rubber Chem. Technol. 67/3, 481- 503.

163. Indian Rubber Industries (IRI), 2000. Rubber Engineering. McGraw- Hill, NY, pp. 646- 716.

164. Folie, G., 1996. Continuous production of rubber profiles- state of extrusion line technology. Rubber World, 1 December, p. 594.

165. Capelle, G., 2000. Extrusion, continuous vulcanisation of profiles. Rubber World, 1 May.

166. Klein, H., 2000. Continuous production of rubber profiles. Rubber World, 1 September, p. 594.http://www.thefree1ibrary.com/Rubber+World/2000/September/1-p594.

167. Treloar, L. R. G., 1975. The Physics of Rubber Elasticity. 3rd ed. Clarendon Press, Oxford.

168. Ward, I. M., 1983. Mechanical Properties of Solid Polymers. 2nd ed. John Wiley & Sons.

169. Fung, Y. C., 1969. A First Course in Continuum Mechanics. Prentice-Hall, Inc., Englewood Cliffs, New Jersey.

170. Malvern, L. E., 1969. Introduction to the Mechanics of a Continuous Medium. Prentice- Hall, Inc., Englewood Cliffs, New Jersey.

171. MSC Software, 2014. Nonlinear finite element analysis of elastomers. White paper.

172. Love, A. E. H., 1926. A Treatise on Mathematical Theory of Elasticity. 4th ed. Dover Publications, New York.

173. Green, A. E., Adkins, J. E., 1970. Large Elastic Deformation. 2nd ed. Clarendon Press, Oxford.

174. Ogden, R. W., 2001. Elastomer models. In: Lemaitre, J. (Ed.), Handbook of Materials Behavior Models Vol. 1. Academic Press, CA, USA, pp. 91–94.

175. Ali, A., Hosseini, M., Sahari, B. B., 2010. A review of constitutive models for rubber-like materials. American J. of Engineering and Applied Sciences. 3 (1), 232- 239.

176. Wagner, D.A., Morman, K. N. (Jr.), Gur, Y., Koka, M. R., 1997. Nonlinear analysis of automotive door weatherstrip seals. Finite Elem. Anal. Des. 28, 33-50.

177. Wadham-Gagnon, M., Hubert, P., Semler, C., Paidoussis, M. P., Vezina, M., Lavoie, D., 2006. Hyperelastic modelling of rubber in commercial finite element software (ANSYS™).

178. Oswald, J. J., Daniels, C. C., Steinetz, B. M., Dunlap, P. H. (Jr.), 2007: Simulating elastomer seal mechanics for a low impact docking system. AIAA SPACE- 2007 Conference & Exposition, Long Beach, California, 18- 20 September 2007.

179. Medri, G., R. G. Molari, Strozzi, A., 1978. Numerical and experimental stress- strain analysis on rubberlike seals in large elastic deformation under unilateral contact. In: 8th Int. Conf. Fluid Sealing, Cranefield, UK, BHRA Fluid Engineering, pp. F2 19- 30.

180. Medri, G., Strozzi, A., 1984. Mechanical analysis of elastomeric seals by numerical method. Ind Eng Chem Prod Res Dev, 23, 596- 600.

181. Strozzi, A., 1986. Experimental stress- strain field in elastomeric O- rings. In: Weiringa, H. (Ed.), Experimental Stress Analysis, Martinus Nijhof Publication.

182. George, A. F., Strozzi, A., Rich, J. I., 1987. Stress field in a compressed unconstrained O-ring seal and a comparison of computer predictions and experimental results. Trib Int, 20(5), 237- 247.

183. Dragoni, E., Medri, G., Strozzi, A., 1987. Analysis of an elastomeric X- ring seal. In: Nau, B. S. (Ed.), 11th Int Conf Fluid Sealing, London and New York, Elsevvier Applied Science Publisher, pp. 160- 174.

184. Dragoni, E., Strozzi, A., 1988. Analysis of an unpressurised laterally restrained, elastomeric O- ring seal. Trans ASME J Trib, 110, 193- 199.
185. Strozzi, A., 1986. Static stresses in an unpressurised, rounded, rectangular, elastomeric seal. ASLE Trans, 29(4), 558- 564.
186. Boast, D., Coveney, V. A., (Eds.) 1999. Finite element analysis of elastomers. 1st ed. London and Bury St Edmunds: Professional Engineering Publishing.
187. Staken, Borg, M. J. L., 1988. On the sealing mechanism of radial lip seals. Tribol Int, 21(6), 335- 340.
188. Green, I., English, C., 1992. Analysis of elastomeric O- rings in compression using the finite element analysis method. Tribol Trans, 35(1), 83- 88.
189. Ferrandez, P., 2008. Fluoroelastomers, FKM, and FEPM. In: Klingender, R.C. (Ed.), Handbook of Specialty Elastomers. 1st ed. CRC Press, Boca Raton, pp. 133–153.
190. Progelhof, R. C.; Thorne, J. L., 1993. Polymer engineering principles: properties, processes, tests for design. 1st ed. Hanser Publisher, pp. 666-679.
191. Bartenev, G. M.; Lavrentev, V. V., 1981. Friction and wear of polymers. Elsevier Scientific Publishing Company, Amsterdam, pp. 81-100.
192. Brown, M. W., 1990. Seals and Sealing Handbook. 3rd ed. Elsevier Science Publication Ltd., pp. 470- 473, 495- 525.
193. Glaeser, W. A., 1992. Materials for tribology. 1st ed. Elsevier Science, Amsterdam, pp. 177-210.
194. Wensel, R. G., Cotnam, B. M., 1978. Evaluation of O-rings of various elastomer compounds for service in hot water. AECL Report. AECL- 6214, CRNL, Ontario, Canada.
195. Wensel, R. G., 1977. Resistance of O-ring seal compounds to heat and ionising radiation in air and in water. AECL Report. AECL- 5697 Report, CRNL, Ontario, Canada.
196. Parker Seals, 2007. Parker O-ring Handbook. ORD 5700, Parker Hannifin Corporation, KY, U.S.A.
197. Jackson, E. (Ed.), 2004. The Seal Man's O-ring Handbook. 1st ed. EPM, Inc. – The Seal Man, Stockbridge, USA.
198. Flitney, R., 2007. Seals and Sealing Handbook. 5th ed. Butterworth- Heinemann, Elsevier Ltd., Oxford, UK.
199. ERIKS, 2013. Sealing Elements: Technical Handbook O- rings.
200. Apple Rubber, 2016. Seal Design Guide. Apple Rubber Products, NY, USA.
201. Fuller, R. E., 2006. Advanced polymer architecture sealing solutions for oil and gas applications. Sealing Technol. 9, 6- 11.
202. Fuller, R. E., 2006. Fluoroelastomers made with advanced polymer architecture for oil and gas applications. Oilfield Engineering with Polymers, UK, London, March 29-30.
203. DuPont, 2015. DuPont Kalrez perfluoroelastomer parts: Reliable, long-term sealing in extreme chemical and thermal environments. Reference Number KZS-A10105-00-A071, DuPont Performance Elastomers, USA.
204. DuPont, 2015. DuPont Kalrez perfluoroelastomer parts: Optimised total system cost for use in pumps and processing equipment. Reference Number KZS-D10651-00-A0710, DuPont Performance Elastomers, USA.
205. DuPont, 2012. DuPont Kalrez perfluoroelastomer parts: Physical properties and product comparisons. Technical Information Bulletin Rev. 11, Reference No. KZE-H68254-00-K0912, DuPont Performance Elastomers, USA.

Appendix A

Property- spectrum- comparison of Generic Elastomers for Selection [54]

Table A.1. Property comparisons for generic elastomers

Properties	Generic elastomers						
	ACM	AU, EU	CO, ECO	CR	EPDM	FKM	IIR
Density (g/cm^3)	1.03- 1.12	1.04- 1.10	1.36, 1.27	1.23	0.86	1.4- 1.95	0.92
HTR (^0C)	177- 204	82- 104	121- 135	98- 121	104- 149	232- 500	121- 149
OR	EX	EX	GE	GE	GE	EX	EX
OXR	EX	GE	GE	GE	EX	EX	EX
STR	PF	PR	FG	FG	EX	GE	GE
SUR	EX	GE	GD	GE	EX	GE	EX
WER	EX	EX	GD	FG	EX	EX	EX
WAR	EX	PG	GD	FG	EX	EX	GE
FR	PR	PG	PG	FG	PR	----	PR
AR	GE	EX	FG	GE	GD	FG	FG
CS	PG	PG	GE	PG	PE	GE	FG
FCR	GD	FG	GD	GD	GD	FG	GE
IMR	GE	GE	FE	GE	GE	GD	GD
TR	GE	GE	FE	GE	FG	FG	GD
VD	GE	FG	GD	GE	FG	FG	EX

Properties	Generic elastomers				
	IR	MQ	NBR	NR	SBR
Density	0.93	0.95	1.00	0.91	0.94
HTR	82- 104	204- 288	99- 121	82- 104	99- 121
OR	PR	EX	FG	PR	PR
OXR	GD	EX	GD	GD	FE
STR	GD	FG	FG	GD	FG
SUR	PF	EX	PG	PF	PR
WER	PF	EX	FG	PF	FG
WAR	EX	EX	GE	EX	GE
FR	FG	FE	PR	FG	PR
AR	GE	PG	GE	GE	EX
CS	EX	GE	GE	EX	GE
FCR	EX	PG	FG	EX	GD
IMR	GE	PG	FG	GE	EX
TR	GE	PG	GE	GE	FE
VD	GE	FG	FG	GE	FG

AR: Abrasion resistance; FCR: Flex cracking resistance; FR: Flame resistance; HTR: High Temperature range; IMR: Impact resistance; OR: Ozone resistance; OXR: Oxidation resistance; STR: Steam resistance; SUR: Sunlight resistance; TR: Tear resistance; VD: Vibration damping. WAR: Water resistance; WER: Weather resistance;

EX: Excellent; FE: Fair- to excellent; FG: Fair- to good; GD: Good; GE: Good- to excellent; PF: Poor- to fair; PG: Poor- to good; PR: Poor;

Appendix B

Deformation and Strain in Finite Elasticity [161, 168, 170]

An initially unstressed configuration (C_0) assumes configuration C_1 after finite deformation (Fig. B.1) with points in the two configurations referenced to different coordinate systems X_i and x_i respectively. The deformed configuration could be imagined as translation of axis d**X** along with its stretching and compression where rotation of the body abut axis completes the definition of final configuration. In essence, it's about knowing the change in length between two arbitrary points and the location of the body. Lagrangian and Eulerian descriptions emphasise happening at points P and p (Fig. B.1) i.e. undeformed and deformed states respectively. The initial and deformed configurations could be related by mapping the corresponding undeformed and deformed position vectors, **X** and $\boldsymbol{x}$ respectively.

$$\boldsymbol{x} = \boldsymbol{\Xi}(\boldsymbol{X}) \tag{B.1}$$

$$d\boldsymbol{X} = dX_A \boldsymbol{I}_A \tag{B.2}$$

Where, $\boldsymbol{I}_A$ is the base vector for undeformed configuration.

By convention, quantities associated with undeformed configuration are capitalised. Relative length between p and q could be expressed taking the deformed configuration as basis.

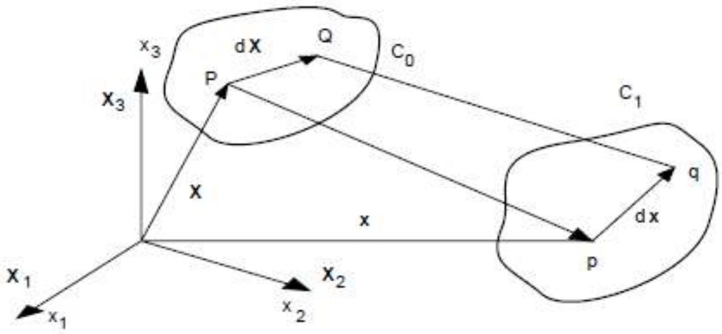


Fig. B.1. Initial and deformed configuration of a body

$$d\boldsymbol{x} = dx_i \boldsymbol{i}_i \tag{B.3}$$

Where $\boldsymbol{i}_i$ is the base vector in deformed configuration.

Differential quantities dx_i and dX_A are described as,

$$dx_i = \frac{\partial x_i}{\partial X_A} dX_A = x_{i,A} dX_A, \qquad dX_A = \frac{\partial X_A}{\partial x_i} dx_i = X_{A,i} dx_i \qquad \text{(B.4)}$$

Where $x_{i,A}$ ($x_{i,A} = F_{i,A}$) is deformation gradient tensor.

A measure of deformation could be expressed in terms of the differential quantities-

$$(dx_i)^2 - (dX_A)^2 = dx_i dx_i - dX_A dX_A \qquad \text{(B.5)}$$

Expressing the deformation in terms of undeformed configuration (Lagrangian description) results in,

$$(dx_i)^2 - (dX_A)^2 = \left((x_{i,A} dX_A)(x_{i,B} dX_B) - \delta_{AB} dX_A dX_B = (x_{i,A} x_{i,B} - \delta_{AB}) dX_A dX_B = \right.$$

$$\left(\delta_{\alpha\beta} \frac{\partial x_\alpha}{\partial X_A} \frac{\partial x_\beta}{\partial X_B} - \delta_{AB}\right) dX_A dX_B = (C_{AB} - \delta_{AB}) dX_A dX_B \qquad \text{(A.6)}$$

Where, δ_{AB} (1 at A=B and 0 AT $A \neq B$), $\delta_{\alpha\beta}$ and C_{AB} $\left(\frac{\partial x_k}{\partial X_A} \frac{\partial x_k}{\partial X_B}\right)$ are Kroenecker delta and Green deformation tensor respectively.

The strain tensor E_{AB} (also known as Lagrange deformation or Lagrangian finite strain tensor) was introduced by Green and St. Venant and is called Green's strain tensor.

$$2E_{AB} = C_{AB} - \delta_{AB} \qquad \text{(B.7)}$$

Expressing the same deformation measure (Eq. B.6) in terms of deformed configuration (Eulerian description) leads to,

$$(dx_i)^2 - (dX_A)^2 = (\delta_{ij} - c_{ij}) dx_i dx_j \qquad \text{(B.8)}$$

$$c_{ij} = \frac{\partial X_K}{\partial x_i} \frac{\partial X_K}{\partial x_j} = X_{A,i} X_{A,j} \qquad \text{(B.9)}$$

Where, c_{ij} is Cauchy's strain (or Cauchy deformation tensor) tensor.

The strain tensor e_{ij} introduced by Cauchy for infinitesimal strain is known as Almansi's strain tensor for finite strain.

$$2e_{ij} = \delta_{ij} - c_{ij} \qquad \text{(B.10)}$$

In analogy with terminology in hydrodynamics, E_{AB} is often referred to as Lagrangian and e_{ij} as Eulerian i.e. Eulerian finite deformation or finite Eulerian strain tensor. E_{AB} and e_{ij} are symmetric tensors ($E_{AB} = E_{BA}$, $e_{ij} = e_{ja}$) defined in the coordinate systems X_A (instead of X_i for expression convenience such as deformation gradient) and x_i respectively which are zero for zero deformation (Eqs. B.6 and B.8).

Introducing the displacement vector **u** with components,

$$u_\alpha = x_\alpha - X_\alpha \qquad \text{(B.11)}$$

$$\frac{\partial x_\alpha}{\partial X_A} = \frac{\partial u_\alpha}{\partial X_A} + \delta_{\alpha,A}, \qquad \frac{\partial X_\alpha}{\partial x_i} = \delta_{\alpha,i} - \frac{\partial u_\alpha}{\partial x_i} \qquad \text{(B.12)}$$

The Lagrangian and Eulerian reduce to the following simple forms.

$$E_{AB} = \frac{1}{2}\left[\frac{\partial u_B}{\partial X_A} + \frac{\partial u_A}{\partial X_B} + \frac{\partial u_\alpha}{\partial X_A}\frac{\partial u_\alpha}{\partial X_B}\right] \qquad \text{(B.13)}$$

$$e_{ij} = \frac{1}{2}\left[\frac{\partial u_j}{\partial x_i} + \frac{\partial u_i}{\partial x_j} - \frac{\partial u_\alpha}{\partial x_i}\frac{\partial u_\alpha}{\partial x_j}\right] \qquad \text{(B.14)}$$

From the tensor index to unabridged notations (x-y-z for x_1-x_2-x_3 or x_i-x_j-x_k ; X-Y-Z for X_1-X_2-X_3 or X_A-X_B-X_C; u-v-w for u_1-u_2-u_3), the typical terms are obtained.

$$E_{XX} = \frac{\partial u}{\partial X} + \frac{1}{2}\left[\left(\frac{\partial u}{\partial X}\right)^2 + \left(\frac{\partial v}{\partial X}\right)^2 + \left(\frac{\partial w}{\partial X}\right)^2\right]$$

$$e_{xx} = \frac{\partial u}{\partial x} - \frac{1}{2}\left[\left(\frac{\partial u}{\partial x}\right)^2 + \left(\frac{\partial v}{\partial x}\right)^2 + \left(\frac{\partial w}{\partial x}\right)^2\right]$$

$$E_{XY} = \frac{1}{2}\left[\frac{\partial u}{\partial Y} + \frac{\partial v}{\partial X} + \left(\frac{\partial u}{\partial X}\frac{\partial u}{\partial Y} + \frac{\partial v}{\partial X}\frac{\partial v}{\partial Y} + \frac{\partial w}{\partial X}\frac{\partial w}{\partial Y}\right)\right]$$

$$e_{xy} = \frac{1}{2}\left[\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} - \left(\frac{\partial u}{\partial x}\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}\frac{\partial v}{\partial y} + \frac{\partial w}{\partial x}\frac{\partial w}{\partial y}\right)\right] \qquad \text{(B.15)}$$

Above equations indicate that for Lagrangian and Eulerian evaluations, u-v-w are considered as functions of unstrained (X-Y-Z) and strained (x-y-z) configurations respectively. The Lagrangian measure of finite strain components, when defined as the components of strain tensor (E_{AB}, as in Eq. B.15), becomes half in shear as also the case for infinitesimal strains where Eulerian and Lagrangian measures and the engineering as well as the true definitions of stress- strain do not have any difference. The set of 6 strain- displacement equations in Lagrangian finite strain measure (which provide a basis for the governing equations of compatibility) is given below in conventional form.

$$\acute{E}_{XX} = \frac{\partial u}{dX} + \frac{1}{2}\left[\left(\frac{du}{dX}\right)^2 + \left(\frac{dv}{dX}\right)^2 + \left(\frac{dw}{dX}\right)^2\right]$$

$$\acute{E}_{YY} = \frac{\partial v}{dY} + \frac{1}{2}\left[\left(\frac{du}{dY}\right)^2 + \left(\frac{dv}{dY}\right)^2 + \left(\frac{dw}{dY}\right)^2\right]$$

$$\acute{E}_{ZZ} = \frac{\partial w}{dZ} + \frac{1}{2}\left[\left(\frac{du}{dZ}\right)^2 + \left(\frac{dv}{dZ}\right)^2 + \left(\frac{dw}{dZ}\right)^2\right]$$

$$\acute{E}_{XY} = \frac{\partial u}{dY} + \frac{\partial v}{dX} + \frac{\partial u}{dX}\frac{\partial u}{dY} + \frac{\partial v}{dX}\frac{\partial v}{dY} + \frac{\partial w}{dX}\frac{\partial w}{dY}$$

$$\acute{E}_{YZ} = \frac{\partial v}{dZ} + \frac{\partial w}{dY} + \frac{\partial u}{dY}\frac{\partial u}{dZ} + \frac{\partial v}{dY}\frac{\partial v}{dZ} + \frac{\partial w}{dy}\frac{\partial w}{dZ}$$

$$\acute{E}_{ZX} = \frac{\partial w}{dX} + \frac{\partial u}{dZ} + \frac{\partial u}{dZ}\frac{\partial u}{dX} + \frac{\partial v}{dZ}\frac{\partial v}{dX} + \frac{\partial w}{dZ}\frac{\partial w_z}{dX} \qquad \text{(B.16)}$$

For small deformations and strain the squares and partial derivatives of u_i are negligible which reduces the Eulerian finite deformation (or finite Eulerian strain) tensor e_{ij} to Cauchy's infinitesimal strain tensor (ε_{ij}), taking away the differences between Lagrangian and Eulerian definitions (Eqs. B.15- B.16).

$$\varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \qquad \text{(B.17)}$$

e_{ij} and c_{ij} (Eqs. B.8- B.10, B.14) are completely described by the subscripts of the current (deformed) configuration. In keeping with the necessity to define the stress and strains (true) with respect to the current configuration (*because of significant difference in area as well as orientation of the undeformed planes resulting in geometric nonlinearity involving changing load application points and boundary conditions*) for quantitative determination of performance and failure, basic constitutive relations for depiction of mechanical behaviour involving load, finite deformation and incompressibility could be defined by Eulerian measures involving e_{ij}. However, additional complexities such as nonlinear material behaviour of rubber, thermodynamics implication of stress-strain in terms of change in configurational entropy on deformation etc. demand more comprehensive and advanced theory for realistic depiction of behaviour. Stored energy or Strain energy density provides a better starting point in this regard- in terms of providing flexibility in solution routes by allowing various energetically conjugate stress and strain measures (for simplifying complexities without compromising essence) and also by defining material isotropy in terms of energy density, as function of strain invariants. This allows determination of Cauchy or true stress by differentiating the strain energy density function with respect to the stretch ratio of rubber instead of e_{ij}.

$$\sigma_{ij} = \frac{dW}{de_{ij}} \tag{B.18}$$

Where σ_{ij}, W and e are true (Cauchy) stress, strain energy density function (strain energy per unit volume) and strain measures such as finite Eulerian strain tensor or stretches.

Alternative measure of deformation by stretch ratios, defined in current configuration (Fig. B.1), is expressed as,

$$\frac{1}{\lambda_{\underline{n}}} = \frac{dX}{dx} \tag{B.19}$$

Where, $\lambda_{\underline{n}}$, dX and dx are stretch ratio along $\underline{\mathbf{n}}$, *original relative length and current relative length respectively.*

The subscript n signifies alignment of the stretch along a particular direction or unit vector ($\underline{\mathbf{n}}$ = d**x**/dx), referenced to the current configuration. Defining the stretch in original configuration ($\underline{\mathbf{N}}$= d**X**/dx) is expressed as,

$$\boldsymbol{\Lambda}_{\underline{N}} = \frac{dx}{dX} \tag{B.20}$$

Where, $\boldsymbol{\Lambda}_{\underline{N}}$ is the stretch ratio along $\underline{\mathbf{N}}$.

In particular cases of deformations ($\underline{\mathbf{N}}$ and $\underline{\mathbf{n}}$ are pointing towards same direction) both stretches (Eqs. B.19- B.20) become same and are treated as engineering strain. Stretch ratios were not defined as tensor quantities originally. However, they could be considered as such because of relationships with Green and Cauchy deformation tensors.

$$\frac{1}{\lambda_{\underline{\mathrm{N}}}^2} = \underline{\mathrm{n}} \cdot \boldsymbol{c} \cdot \underline{\mathrm{n}} \qquad \boldsymbol{\Lambda}_{\underline{\mathrm{N}}}^2 = \underline{\mathrm{N}} \cdot \boldsymbol{C} \cdot \underline{\mathrm{N}} \tag{B.21}$$

Where, $\boldsymbol{C}$ and $\boldsymbol{c}$ are green and Cauchy deformation tensors respectively.

This implies that the invariants of the Green and Cauchy deformation tensors are also the invariants of stretches. Assuming homogeneous deformation where orientation of the unit vectors (**$\underline{\mathrm{N}}$** and **$\underline{\mathrm{n}}$**) do not change (i.e. pure strain), both stretch ratios are same and could be stated as,

$$\bar{\wedge} = \boldsymbol{\lambda}\boldsymbol{\lambda} = \begin{bmatrix} \lambda_1^2 & 0 & 0 \\ 0 & \lambda_2^2 & 0 \\ 0 & 0 & \lambda_3^2 \end{bmatrix} \tag{B.22}$$

The invariants are expressed as,

$$I_1 = tr[\bar{\wedge}] = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$$

$$I_2 = (tr[\bar{\wedge}])^2 - tr[\bar{\wedge}\bar{\wedge}] = \lambda_1^2\lambda_2^2 + \lambda_2^2\lambda_3^2 + \lambda_3^2\lambda_1^2$$

$$I_3 = det[\bar{\wedge}] = \lambda_1^2\lambda_2^2\lambda_3^2 \tag{B.23}$$

Regardless of external tractions, $\lambda_1^2\lambda_2^2\lambda_3^2 = 1$ when a material is incompressible. Choosing λ_1 and λ_2 are the independent stretches,

$$\lambda_3 = \frac{1}{\lambda_1\lambda_2} \tag{B.24}$$

And the invariants are-

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$$

$$I_2 = \frac{1}{\lambda_1^2} + \frac{1}{\lambda_2^2} + \frac{1}{\lambda_3^2}$$

$$I_3 = det[\bar{\wedge}] = \lambda_1^2\lambda_2^2\lambda_3^2 \tag{B.25}$$

Appendix C

List of Standards Accessed

Table C.1. Stock properties

Operations and Properties	Seals					
	Inflatable seal				Backup seal	
	Compounds				Compounds	
	S- 1 (Laboratory batches: DMF3) *Data reference for Thesis*		S- 2 (Seal production batches: DMF3 or Comp # 1- Comp # 9) *Experiment as part of Thesis*		S- 4 (Laboratory batches: IG3, IG3- A) *Experiment as part of Thesis*	
	Standards					
	ASTM	ISO	ASTM	ISO	ASTM	ISO
Mixing and molding	ASTM D3182-89	----	ASTM D3182-89 (reapproved 2001)	----	ASTM D3182-89 (reapproved 2001)	-----
M_L, M_H, t_s2, t_s10, t'50, t'90(RT; ibf.in, min)	ASTM D5289-95	----	ASTM D5289-95 (reapproved 2001)	----	ASTM D5289-95 (reapproved 2001)	----
Mooney viscosity[31]	----	----	ASTM D1646- 03a	----	----	----
Garvey die rating	----	----	----	----	ASTM D2230-96 (reapproved 2002)	----

Table C.2. Vulcanizate properties- I

Properties	Seals							
	Inflatable seal				Backup seal			
	Compounds							
	S- 1 (Laboratory batches of DMF3) *Data reference for Thesis*		S- 2 (Seal production batch of Comp # 9) *Experiment as part of Thesis*		S- 4 (Laboratory batches of IG3, IG3- A) *Experiment as part of Thesis*		S- 4 (Seal production batches of IG3, 10a- c) *Data reference for Thesis*	
	Standards							
	ASTM	ISO	ASTM	ISO	ASTM	ISO	ASTM	ISO
Specific gravity	ASTM D297	ISO 2781 (Method A)	ISO 2781: 1996	----	----	ISO 2781: 1996		ISO 2781: 1996
T_s- E_b -M_x(RT, ET, MPa- %- MPa)	ASTM D412 98a	----	ASTM D412- 98a (reapproved 2002)e1	----	ASTM D412- 98a (reapproved 2002)e1	----	ASTM D412- 98a (reapproved 2002)e1	ISO 37: 2005
Hardness(RT, ^{0}Shore A)	ASTM D2240- 02b	----	ASTM D2240-05	----	ASTM D2240- 03	----	ASTM D2240- 03	ISO 7619- 1:2004

[31] 1 Mooney unit (expressed as viscosity number) is equivalent to a torque of 0.083 N-m (0.733 ibf.in)

Table C.3. Vulcanizate properties- II

Operations, Properties, Life Estimation, Nomenclature	**Seals**							
	Inflatable seal				Backup seal			
	Compounds							
	S- 1 (Laboratory batches of DMF3) *Data reference for Thesis*		S- 2 (Seal production batch of Comp # 9) *Experiment as part of Thesis*		S- 4 (Laboratory batches of IG3, IG3- A) *Experiment as part of Thesis*		S- 4 (Seal production batches of IG3) *Data reference for Thesis*	
	Standards							
	ASTM	ISO	ASTM	ISO	ASTM	ISO	ASTM	ISO
Specimens from slab	ASTM D3182-89	----	ASTM D3182-89 (reapproved 2001)	----	ASTM D3182-89 (reapproved 2001)	----	ASTM D3182-89 (reapproved 2001)	----
Specimens from seal	-----	----	ASTM D3183- 02	----	ASTM D3183- 02	----	ASTM D3183- 02	----
Compression stress- strain (RT, ET; MPa- %)	----	----	----	----	----	ISO 7743: 2004	----	----
Splice strength (RT, MPa)	----	----	ASTM D2527- 83 (2001)	----	----	----	ASTM D2527- 83 (2001)	----
Tear Resistance (RT, N/ mm)	ASTM D624- 00e1	----	ASTM D624- 00e1	----	----	----	----	----
CS (RT, up to 150^{0}C/168 h in air)	ASTM D395- 02	----	ASTM D395- 03	----	ASTM D395- 03	---	ASTM D395- 03	----
Dry heat resistance (RT, up to 150^{0}C/33h in air)	ASTM D573- 99	----	ASTM D573- 04	----	ASTM D573- 99	----	ASTM D573- 99	----
Dimensional checks	----	----	----	----	----	----	ASTM D3767- 03	----
Long term heat ageing (up to 32 weeks in air at 140-170-200 ^{0}C)	----	----	----	----	ASTM D573- 99	ISO-188:1998; ISO 11346: 2004	----	----
Life estimation by Arrhenius and WLF equations	----	----	----	----	----	ISO 11346: 2004	----	----
Material fingerprinting: TGA	----	----	----	----	ASTM D6370- 99 (reapproved 2003)	----	ASTM D6370- 99 (reapproved 2003)	----
Rubber nomenclature	ASTM D1418- 01							