STUDY OF ORGANIC MATERIALS AND PLASMA PROCESSING FOR NUCLEAR RADIATION DETECTORS

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A thesis submitted to the
Board of Studies in Physical Sciences
In partial fulfillment of requirements
for the Degree of
DOCTOR OF PHILOSOPHY
of
HOMI BHABHA NATIONAL INSTITUTE

November, 2018

Homi Bhabha National Institute[1]

Recommendations of the Viva Voce Committee

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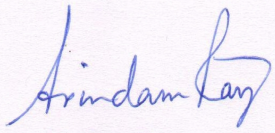

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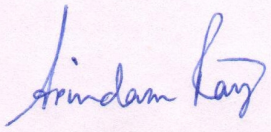

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List of Publications arising from the thesis

Journal

1. "Development of stable nuclear radiation detectors based on n-silicon/cobalt-phthalocyanine heterojunctions", A.Ray, R.Prasad, C.A.Betty, T.V.Chandrasekhar Rao, *Radiation Physics and Chemistry*, **2016**, *120*, 12-16.
2. "Development of surface barrier detectors of low leakage current employing argon cold plasma assisted silicon surface cleaning prior to detector fabrication", A.Ray, Tomi Nath Das, C.A.Betty and T.V.Chandrasekhar Rao, *Journal of Instrumentation* (*JINST*), **2018**, 13 P09019.
3. "Junction edge passivation study of silicon surface barrier detectors using organic films deposited by L-B technique", A.Ray, Sipra Choudhury, Vishal Singh, C.A.Betty and T.V.Chandrasekhar Rao, *Bulletin of Material Science (accepted).*

Not included in the thesis but very much referred

4. "Nuclear detectors based on n-silicon/copper-phthalocyanine heterojunctions", A.Ray, S.K.Gupta, J.V.Yakhmi, *Radiation Measurements*, **2009**, *44*, 47-49.

Conferences

1. "Junction edge passivation study of silicon surface barrier detectors using organic films deposited by L-B technique", A.Ray, Sipra Choudhury, C.A.Betty and T.V.Chandrasekhar Rao [Presented the paper in *M-TECS 2018 conference, September26-29,* **2018**, Organised by MRSI, Mumbai Chapter and Sponsored by Board of Research in Nuclear Science, DAE].
2. "Polymer based detectors: A preliminary study", A.Ray, *Proc. of the DAE-BRNS Symposium on Nucl. Phys.*, **2015**, *60*, 956-957. [Available online at www.sympnp.org/proceedings].
3. "Development of Rugged Silicon Surface Barrier Detector", A.Ray, Shovit Bhattacharya, Ranu Bhatt, D.K.Aswal, S.K.Gupta, *31st IARP National Conference on Advances in Radiation Measurement Systems and Techniques (IARPNC-2014)*, **2014**, p.-269.
4. "Fabrication of Ultra-thin Oxide-passivated Silicon Surface Barrier Detectors", A.Ray, D.K.Aswal, R.Prasad, S.K.Gupta, *Proc. of the DAE Symp. on Nucl. Phys.*, **2013**, *58*, 848-849. [Available online at www.sympnp.org/proceedings].
5. "Study of Aging of Nuclear Detector Based on n-Slicon/Copper-Phthalocyanine Heterojunction", A.Ray, S.K.Gupta, *AIP Conference Proc. (DAE SSPS-2012)*, **2013**, *1512*, 458-459.

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Dedicated to my parents, wife and children

ACKNOWLEDGEMENTS

To begin with, from the bottom of my heart, I am expressing my sincere gratitude to my research guide Dr. (Smt.) C.A.Betty, Scientific Officer (G), Chemistry Division, Bhabha Atomic Research Centre (BARC), Mumbai, who despite her preoccupations was kind enough to accommodate me as her research student and was always willing to make herself available for my thesis work. It is her immense support which has actually made it possible for me to complete this thesis.

It is my pleasure to express my grateful thanks to Dr.A.K.Mohanty, Director-Physics Group, BARC for his continuous encouragement and support.

I must offer my gratitude and respect to my present Section Head, Dr. T.V.Chandrasekhar Rao for his continuous support, guidance and encouragement in carrying out this work.

I'm very much indebted to Dr.S.K.Gupta, DAE – Raja Ramanna Fellow (Ex-Head, Technical Physics Division and Ex-Associate Director, Physics Group, BARC) for not only giving support but also for his constant encouragement and valuable guidance. He is also Chairman of my doctoral committee.

I'm greatly indebted to Dr.B.K.Nayak, Nuclear Physics Division and also Dean-Academic, Physical Science, HBNI, who despite his preoccupations was kind enough and attended all my presentations before my doctoral committee as an Honourable invitee. He is a constant source of encouragement to me.

I'm also very much grateful to all other doctoral committee members and invitees viz. Prof. Senthil Kumar, IIT-Bombay, Dr. D.C.Biswas, Dr. Bency V.John and Dr.(Smt.) Anita Topkar. At this juncture I must name two more personalities who

actually created my interest in the field of Nuclear Radiation Detectors and they are my Ex-Section Heads Dr.R.V.Srikantiah and Dr.D.Sahoo.

I'm highly grateful to Dr.D.K.Aswal, Director-NPL, CSIR, New Delhi for his tremendous help and support.

I'm highly thankful to my very good friends and well-wishers Dr. A.K.Chauhan, and Dr. A.K.Debnath from Technical Physics Division, BARC for motivating me all the time as well as giving their best support and valuable suggestions.

I'm very much thankful to Dr. Swapan Das, Nuclear Physics Division, for offering many valuable suggestions that has greatly helped me to complete this work.

I must express my sincere thanks to Dr. S.C.Gadkari, Dr. R.Tewari, Shri M.Gopalkrishna, Shri T.K.Saha, Dr. K.P.Muthe, Shri R.Prasad, Dr. R. Ganguly, Dr. T.N.Das, Dr. Sipra Choudhury, Shri P.G.Abichandani, Shri V.Nataraju, Shri E.Ravishankar, Dr. K.G.Bhushan, Dr. Jagannath, Shri Manish Kumar, Shri V.Parayya, Shri A.Kasbekar, Shri Sunil Sahu, Smt. Nivedita Ved, Shri Sandip Chowdhury, Smt. Deepa Bhambure, Smt. Alka Satam, Shri B.A.Narayanan and Smt. Pushpa Panchal for extending support and help at various stages of the thesis work.

I take this opportunity to record my sincere gratitude to all my teachers. Also, it gives me great pleasure to acknowledge all my friends for their invaluable support all along.

Last but not the least, I would like to acknowledge my whole-hearted indebtedness to my parents, and to my wife Suparna who have always cheered and supported me when things seemed to become burdensome. Their love has provided me energy to move forward.

Arindam Ray

To My Guide

CONTENTS

SYNOPSIS

Detection, identification and measurement of energy of ionizing radiations are very important in the research on nuclear science and technology. It also plays an important role in the development of medical diagnostics and in the study of cosmic ray effects etc. A variety of nuclear detectors have been developed over the years to fulfill this task. The charged particle detectors (e.g. Silicon Surface Barrier Detectors) find wide spread use among these applications. This very fact necessitates development of high quality (viz. low leakage current, high resolution, highly reliable in terms of their long term stability etc.) detectors and this activity has been an active as well as an important field for scientific investigations. Fabrication of high quality detectors demands stringent selection of detection material. To be very specific, the material should be of high resistivity (for larger depletion depth), small band gap (for good energy resolution), indirect band gap (high life time of carriers), high charge carrier mobility and life time (for good charge collection), availability of single crystals in large sizes (for good efficiency), should be easy to make stable electrical contact and lastly the material should have minimum trapping centers (for good charge collection). Silicon fulfills all the above requirements and is considered as one of the key detection materials.

In recent years, demand for high quality silicon detectors has greatly enhanced worldwide, especially for high-energy physics experiments (e.g. CMS experiment at CERN etc.). But the common problem always arises is that the fabricated silicon detectors breaks down at much lower reverse voltage as compared to its bulk breakdown voltage, though one chooses a very high resistivity silicon. The reason attributed to this is none other than highly active nature of silicon surface (as the

number of surface states in silicon is $2 - 4 \times 10^{15}/cm^2$) and the breakdown of silicon detectors at much lower voltage than bulk breakdown is due to surface breakdown which results in very high reverse leakage current. To prevent this surface breakdown, silicon surface needs to be passivated. Currently the leakage currents are reduced by passivating the edges either by epoxy or by silicon dioxide. Apart from these two, new passivating materials also are being explored to fabricate detectors with improved properties. Another important parameter for the good performance of the detectors is energy resolution. There are a number of factors which can affect the energy resolution of silicon based nuclear detectors viz. spectral broadening due to: (i) statistical fluctuation in number of ion pairs produced, (ii) nuclear collision, (iii) detector and amplifier noise, (iv) specifications of the radiation source and (v) detector dead layer. As a detector developer, one can improve energy resolution (a) by reducing detector capacitance (i.e. by fabricating a detector of smaller active area) which will bring down detector noise and/or (b) by reducing leakage current and/or (c) by minimizing dead layer. Bringing down the leakage current and to get longevity for good performance of fabricated detectors is a challenging task for a detector developer. **This prompted the author to take up the theme of the present thesis which is mainly aimed to explore alternate organic materials and processes to reduce leakage, since higher leakage current in silicon radiation detectors limits the detector performance as well as longevity.**

Silicon based α particle and heavy ion detectors, especially surface barrier detectors – both partially depleted as well as thin and fully depleted – play an important role in low to moderate energy physics experiments in accelerators. These detectors are having numerous advantages namely, excellent energy resolution,

particle identification, linear energy response, variety of configuration, compact and small power requirements. These detectors work on the principle of ionization in solid medium by the incident radiation which is followed by collection of electron-hole pairs (EHP). One of the major issues in many silicon detectors is the high leakage current through edges of Si/metal junction. So far the leakage currents were reduced by passivating the edges either by epoxy or by silicon dioxide. Silicon dioxide passivated Silicon detectors are known for their long term stability. However, oxidizing Silicon with an aim to get good quality thin SiO_2 layer (breakdown voltage: 7 Mega Volt/cm) requires high temperature (1000-1200ºC) oxidation furnace, high quality oxygen etc. Though organic epoxy passivated detectors are very much present in the market, epoxy has a disadvantage as it is hydrophilic in nature and reacts with moisture over a period of time to give rise to more leakage current and therefore affects long term stability. We have already developed a number of organic and oxide passivated silicon E and delta E detectors. Few rugged silicon surface barrier detectors with organic epoxy passivation were also developed. In order to have a simpler fabrication procedure for the detectors, it is essential to look for new passivating materials, which can be deposited at the edges of silicon wafer at a relatively lower temperature and slow/negligible degradation with atmospheric exposure. Growing SnO_2 film on a Silicon wafer by initially depositing Octa Decyl Amine-Stannate Complex (ODA-Stannate complex) with the help of Langmuir-Blodgett (LB) deposition technique followed by heating the sample in a furnace at a relatively low temperature (600ºC) to decompose the LB layer to form SnO_2 film on Silicon is a new as well as simpler technique which has been explored to study passivation of Silicon detectors. As tin oxide layer grown by this method consist of SnO_2 nano particles

which are not affected by humidity, the layer behaves like a depletion layer and offers very high resistance. Further, in few silicon samples hydrogenated amorphous silicon as a passivating layer has also been studied. In addition, possibilities of using organic semiconductors (CuPc, CoPc) and silicon heterojunction structures for α-detectors have been thoroughly investigated.

Semiconducting organic material can be subdivided into three categories, namely, polymers, oligomers and small molecules depending on the length of the molecular structure. This very molecular structure actually establishes the mechanical, chemical and electronic properties of the organic materials. Since, the radiation creates charge carrier inside polymer matrix by ionizing the medium, these charge carriers can be utilized for the detection of radiation. The motive behind these studies is to tap the advantages of organic based devices (the ease of fabrication, improved performance in terms of low leakage and long term stability etc.) for α detectors. Some preliminary studies were also carried out to explore new polymer viz. Bi-axially oriented Poly Ethylene Terephthalate (BoPET) based α-sensor which will help to design new polymer based flexible and easily portable α detector at low cost. Additionally, surface barrier detectors of reasonably good performance were fabricated and exhaustively studied by incorporating cold plasma assisted silicon surface cleaning (keeping aside the existing cleaning methods, e.g. cleaning of silicon surface by organic solvents and RCA cleaning of silicon surface etc. which are exhaustive and time consuming) prior to edge protection using organic epoxy followed by metallization.

The thesis has been organized into six chapters as summarized below:

Chapter 1, Introduction: This chapter includes an introductory discussion on various types of radiation, interaction of radiation with matter, radiation detection mechanism, a comprehensive literature survey on different types of radiation detectors with their associated problems and specifications. The thesis work mainly deals with role of organic materials for nuclear detectors. In nuclear detectors, organic materials can find different roles such as, (1) passivation layer in Si based alpha detectors; (2) as an active layer in organic/Si hybrid structures and (3) pure organic material as an active layer. Presence of organic impurities on Si, while fabricating Si surface barrier detectors is unwanted. Therefore the removal of organic contaminants and other impurities, by cold plasma prior to detector fabrication has also been explored. In this thesis, extensive study has been done on all these issues.

Chapter 2, Development of stable nuclear detectors based on Au/Organic thin film/n-Si structures: In earlier studies [A.Ray et al., Radiation Measurements, 44 (2009) 47-49], Copper phthalocyanine (CuPc) has been utilized by the author for fabrication of nuclear detectors based on n-silicon/copper phthalocyanine heterojunctions. Of late it has been reported that MnPc, FePc and CoPc have a stronger binding with gold than that of NiPc, CuPc and ZnPc. So in the present study CoPc has been used in lieu of CuPc. n-type silicon/cobalt-phthalocyanine (CoPc) heterojunction based nuclear detectors have been fabricated using thermally evaporated CoPc films. The detectors were characterized by measuring their current–voltage (I–V) and leakage current–time (I–t) characteristics, followed by alpha energy spectra obtained on exposure to α-particles. Variation of alpha energy resolution with applied reverse bias voltage for each of the detectors was also studied. The detectors showed very low leakage current and high breakdown voltage as compared to

conventional Au/n-Si surface barrier detectors. Lastly, the durability of the detectors was established by measuring their I–V characteristics and energy resolution for nearly 15 months

Chapter 3, Si-surface barrier detectors using Organic/Inorganic thin films as passivation layer: In conventional Silicon Surface Barrier detectors, passivation of surface is carried out at room temperature by using two-component epoxy. Right quantity of Amine-based hardener is used to properly cure epoxy resin which in turn results in very low leakage current and stable detector performance. This prompted the author to study the use of Amine-based organic thin films as well to passivate Si surface. It was carried out by depositing Octa Decyl Amine-Stannate Complex on the peripheral annular region of silicon wafer at first with the help of Langmuir-Blodgett (LB) deposition technique, followed by heating the sample in a furnace at a temperature of 600ºC to decompose the LB layer to form SnO_2 film on Silicon. Tin oxide layer so grown consists of SnO_2 nano-particles. The space charge layer between the nano-particles forms the charge carrier depletion layer and offers very high resistance, dielectric constant and breakdown strength. Growing SnO_2 film on a Silicon wafer is a new as well as simpler technique which has been explored to study passivation of Silicon detectors.

This chapter also explains the work carried out by the author towards (1) development of oxide passivated silicon delta E detectors and (2) development of organic epoxy passivated rugged silicon surface barrier detectors using DC sputtering to deposit gold film on n-silicon.

Chapter 4, Development of improved surface barrier detectors by employing argon cold plasma assisted Silicon surface cleaning: Any gas with a presence of

ions and chemically reactive species around 1% is known as cold plasma. In cold plasma electron temperature is much higher than neutral and cation (i.e. other species) temperature. That is why it is also known as non-thermal and non-equilibrium plasma. In cold plasma, moisture is also present in small quantity. This moisture, as a result of its interaction with high energy electron, produces $[H_2O]^-$ ions which decomposes afterwards into H^+ and OH^- ions. This OH^- ion is highly reactive and destroys all types of organic contaminants. This novel aspect of cold plasma has been utilized to clean silicon surface before fabricating any device. This can be a cost effective and viable replacement of the cleaning processes adopted by current industries for silicon surface cleaning as cleaning using acids (piranha solution) and organic solvents are time consuming and also may add to local environmental pollution. Surface barrier detectors of improved performance were fabricated by incorporating the cold plasma assisted silicon surface cleaning. It was a detector fabrication process step prior to edge protection using epoxy and was subsequently followed by metallization. The detectors fabricated using this new technique yielded better result compared to detectors of same active area and processed using conventional technique. The fabricated detectors were characterized by measuring their Current-voltage ($I - V$) characteristics (both forward and reverse) and alpha energy spectra obtained on exposure to α-particles. Variation of alpha energy resolution with applied reverse bias voltage for each detector was also studied. The results show that the leakage current of the new detectors treated with cold plasma formed by a mixture of Argon and Air (90% Argon and 10% Air) was substantially lower than that of surface barrier detectors fabricated by conventional methods.

Chapter 5, Purely organic radiation sensors: Two major sources of alpha emitters, because of which environment in general and mankind in particular get exposed are (1) uranium mining waste, and (2) concentrated natural radium which is generated during processing phosphate ore for fertilizer. So it is essential to detect the alpha particles in and around of such industries to prevent the environment from getting contaminated. In the present scenario, the importance of alpha detector is not only limited to environmental monitoring or carrying out some nuclear experiments but also extended to prevent nuclear proliferation. For example, plutonium, an alpha emitter, which is a classified material, can be detected by alpha detectors. For many of these applications, it will be preferable that the radiation detectors need to be inexpensive and flexible. Therefore it is expected that organic material based detectors, perhaps would be best suited, for its capability to be flexible, large scale production and that too at low cost. The working principle of organic detectors lies with the basic fact that irradiating with ionizing radiations, conductivities in polymer materials (e.g. BoPET etc.) can be changed. In present study, the changes in conductivities were investigated for α-radiation detection.

Chapter 6, Conclusion and future scope: In this chapter, a summary of all the work done is presented highlighting the main findings of this thesis. The chapter ends by indicating some future directions that have emerged from this work.

Chapter 1

INTRODUCTION

1.1 Introduction

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1.2.1 Nuclear radiations and radioactivity

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1.1 INTRODUCTION

Detection and energy measurement of radiation is very much important in nuclear science and technology, medical physics and study of cosmic rays. A variety of nuclear detectors have been developed over the years because high energy radiations cannot be measured directly [1]. Development of nuclear radiation detectors with improved features has opened the path for many important discoveries in nuclear science. For example, scientists working with data from experiments at the Large Hadron Collider (LHC), CERN (European Organization for Nuclear Research) announced the discovery of a new particle "consistent with" the Higgs boson. It is a subatomic particle also colloquially referred to as the "God particle." The CMS (Compact Muon Solinoid) is a general-purpose detector which forms a part of the LHC, is capable of studying many aspects of proton collisions at 14 trillion electronvolts (eV). Therefore, development of better nuclear radiation detectors continues to remain as an area of interest to many researchers. There are various kinds of gas detectors, scintillators and semiconductor detectors [1]. These detectors are very much useful in their respective areas of applications. Three of the oldest but at the same time very much useful detectors are the ionization chambers, the proportional counters and the Geiger-Muller (G – M) tube [2]. All these three are falling in the category of Gas Detectors. Another category of detectors, also known as scintillators, are certain materials whose atoms get ionized and/or move to excited states when ionizing particle passes through them and during their return to the normal states (de excitation) produce light emission, or scintillation. By employing

semiconductors as the basic detection medium, semiconductor radiation detectors were developed and put to use way back in 1951[3]. Both these scintillation (mainly inorganic scintillators e.g. NaI, CsI, BaF_2) as well as semiconductor detectors are falling in the category of solid state detectors and their dimensions can be kept smaller (i.e. compact size) as solid densities are ~ 1000 times larger than gas [1]. Semiconductor detectors are superior to scintillators as they offer much better energy resolution. This is achieved by reducing the statistical limit on energy resolution as a result of generation of more number of information carrier (electron-hole pair in case of semiconductors) per impinging charged particle/photon of the radiation. Semiconductor detectors are used to extract the exact information about energy, position [4] and type [5] of a nuclear radiation. Several types of semiconductor detectors have been developed to suit different experimental requirements. The various materials that can be utilized to fabricate nuclear radiation detectors in general [6] and the materials pertaining to semiconductor based radiation detectors in particular, is indeed a vast topic and will continue to remain under active consideration to a detector developer. Over 60 years ago, in terms of radiation detection, compound semiconductors were among the first direct detection media to be investigated. However, material problems caused by impurities, stoichiometric imbalances and high defect densities limited their usefulness. It is only the recent development that substantial improvement has occurred in their material growth and also in detector fabrication. Accordingly, compound semiconductor radiation detectors have become serious competitors to other established radiation detectors [7]. Among the available semiconductor materials, silicon predominates in the category of diode detectors which are mainly used for charged particle spectroscopy. Germanium also

can be used for charged particle spectroscopy but Germanium detectors need to be cooled to maintain the leakage current minimal. But for gamma ray spectroscopy, as a semiconductor material, Germanium is more advantageous because of its higher atomic number. Though silicon detectors can be operated at room temperature, the common problem always arises is that the fabricated silicon detectors breaks down at much lower reverse voltage as compared to its bulk breakdown voltage, though one chooses a very high resistivity silicon. The reason attributed to this is none other than highly active nature of silicon surface that leads to the breakdown of silicon detectors at much lower voltage than bulk breakdown. This breakdown is called surface breakdown which results in very high reverse leakage current. This leakage can be lowered by passivating silicon surface. Currently the reduction of leakage current is facilitated by employing edge passivation using either epoxy or silicon dioxide. Epoxy passivation, though very simple, won't last long as epoxy get affected by the moisture present in air. On the other hand, oxidation of silicon to prepare SiO_2 (a passivating material more stable than epoxy) is a high temperature process and also requires elaborate laboratory arrangement. So lots of new passivating materials/methods are also being explored to fabricate detectors with improved properties. Other than leakage current, another important as well as measurable parameter, what can be considered to certify the quality of a detector is energy resolution. Moreover the stability of the performance of the fabricated detectors is very much necessary. Bringing down the leakage current and to get longevity for good performance of fabricated detectors is a challenging task for a detector developer. **This prompted the author to take up the theme of the present thesis which is mainly aimed to**

explore alternate organic materials and processes to reduce leakage and also achieve better stability of the fabricated detectors.

In section 1.2, we discuss about nuclear radiation and radioactivity, various types of nuclear radiation detectors, various radiation sources and interaction of nuclear radiations with matter. In section 1.3, we describe more specifically about semiconductor detectors along with a brief definition of energy resolution and its dependence on various factors. In section 1.4, we present in a nutshell, the requisite nuclear electronics which is essential for evaluation as well as functioning of the fabricated detectors. The scope of this thesis, which mainly deals with the various fabrication methods which resulted in reduced leakage current of developed detectors, is presented in section 1.5.

1.2 NUCLEAR RADIATION DETECTORS

1.2.1. Radioactivity and Nuclear Radiations:

Radioactivity is a nuclear phenomenon and it is connected with the instability of the nucleus. Since most of the elements are not radioactive, the ratio of neutron to proton of the unstable, radioactive nucleus is the factor responsible for radioactivity. Scientists have studied this problem exhaustively and arrived at the conclusion that the stability or instability of a nucleus is related with the pairing of nuclear spins. It is available in literature that electrons spin around their own axis and electron-spin pairing leads to stable chemical bonds. Likewise inside a nucleus, protons and neutrons too spin around their own axes and pairing of spins of protons among protons as well as pairing of spins of neutrons among neutrons leads to nuclear stability. For all non – radioactive i.e. stable isotopes, it is found that nuclei with even number of protons and with even number neutrons are most abundant. The even numbers result

in spin pairing and odd numbers result in unpaired spins. So nuclei having either of the proton number or the neutron number odd are slightly less stable than the even numbered ones. The nuclei which have odd numbers of protons and odd numbers of neutrons are the least stable ones.

In addition to the odd or even number of protons and neutrons, the nuclear stability is also very much influenced by the neutron/proton ratio *(n/p)*. As the number of protons increases inside a nucleus, more and more neutrons are required to minimize the proton-proton repulsion and eventually add to nuclear stability. As a matter of fact, neutrons are serving as binding material inside the nucleus.

The activity of a radio isotope source is defined as its rate of decay and as per the fundamental law of radioactive decay

$$(dN/dt)_{decay} = - \lambda N$$

where N is the number of radioactive nuclei and λ is called radioactive decay constant. One of the units of activity is Curie (Ci) which is actually the activity of one gram of pure Radium (^{226}Ra) and its value is 3.7×10^{10} disintegrations per second. The SI unit of activity is Becquerel (Bq) and is defined as one disintegration per second.

1.2.2. A Glimpse of Various Types of Radiation Detectors:

There are different kinds of detectors; one can come across in literature. A majority of them are listed below:

- Fluorescent Screen (Roentgen – discovery of X-ray)
- Photographic film and emulsion
- Cloud and bubble chamber

- Gas detector
- Scintillation detector
- Cerenkov detector
- Semiconductor detector
- Crystal spectrometer
- Thermoluminescence dosimeter
- Self powered neutron detector
- Solid state nuclear track detector
- Multi wire proportional counter
- Electron multipliers
- Photodiodes
- Scintillating fibres
- Microstrip detectors
- Drift detectors
- Charge Coupled Devices
- Calorimeters etc.

1.2.3 Radiation Sources:

The radiations of our concern get generated in nuclear and atomic processes. In a broader perspective, radiations are of three kinds:

i) Charged particles (e.g. fast electrons, alpha particles and fission fragments etc.)

ii) Uncharged particles (e.g. neutrons)

& iii) Photons (e.g. gamma and X-rays)

Radiation sources are also broadly considered to be of two kinds:

i) Natural (i.e. radioactive substance)

& ii) Artificial (i.e. produced due to operation of reactor and accelerator)

A. Fast electron sources

There are three different types of fast electron sources:

i) Negatively charged electrons are arising out of **beta-minus decay** and the process is written schematically as:

$$^{A}X_{z} \rightarrow {}^{A}Y_{Z+1} + \beta^{-} + \bar{\nu}$$

where X and Y are called the parent and daughter nuclei, and $\bar{\nu}$ is antineutrino. The recoil nucleus Y comes out with a negligibly small recoil energy (< ionization threshold). So the beta decay energy is essentially shared by neutrino/anti-neutrino and beta particle. Beta minus emission occurs for nuclei with too many neutrons.

ii) Positively charged electrons (also known as positrons) are arising out of **beta-plus decay** and the process can be written schematically as:

$$^{A}X_{z} \rightarrow {}^{A}Y_{Z-1} + \beta^{+} + \nu$$

where X and Y are called the parent and daughter nuclei, and ν is neutrino. Beta plus emission occurs for nuclei with too many protons. Any beta source (i.e. β^{+} or β^{-}) produces a continuum of energy.

Example: ^{22}Na (β^{+}) and ^{36}Cl (β^{-})

iii) The nuclear process of **internal conversion** is the source of conversion electrons and in certain cases they are mono-energetic. Sometimes beta decay leaves the

daughter nucleus in excited state and the excited nucleus cannot de-excite through emission of γ-ray. In such cases, the nuclear excitation energy is transferred to one of the orbital electrons and the electron comes out.

Example: ^{113}In, ^{207}Bi etc.

iv) Auger electrons are somewhat analogous to internal conversion electrons and produce a discrete energy spectrum. The process of electron capture leaves an atom with a vacancy in a normally complete electron shell. This vacancy needs to be filled by electrons from outer shell along with emission of an X-ray. But many a times, the excitation energy of the atom is transferred to one of the orbital electrons. As a result of which, the electron comes out. This ejected electron is known as Auger electron.

B. Heavy charged particle sources

i) Alpha decay:

Alpha decay is schematically written as

$$^{A}X_{Z} \rightarrow {}^{A-4}Y_{Z-2} + {}^{4}He_{2}$$

Some heavy nuclei are spontaneous emitter of alpha particles as they are energetically unstable against the spontaneous emission of alpha. The phenomenon called *barrier penetration mechanism* is governing the probability of alpha decay. The energy of the emitted alpha particles lies in the range between 4 – 6 MeV. It is also observed that the alpha particle emitters emitting alpha particles of energy beyond 6 MeV are having very short half life and those emitting alphas with energy lower than 4 MeV are having exceedingly long half life.

Example: ^{239}Pu, ^{241}Am etc.

ii) Spontaneous fission:

All heavy nuclei are, in principle, having a tendency to undergo spontaneous fission into two lighter fragments. But for that they need to overcome a large potential barrier. For this reason, the spontaneous fission process is limited to few extremely heavy nuclei.

Example: ^{252}Cf .

C. Electromagnetic radiations

i) Gamma rays following beta decay:

Apart from emission of beta particles, beta decay ends up with an excited nucleus which subsequently de-excites (either partly or completely) by emitting gamma rays. But the emitted gamma rays are limited to energies below about 2.8 MeV.

Example: ^{22}Na (1.274 MeV γ), ^{60}Co (1.173 MeV & 1.332 MeV γ) and

^{137}Cs (0.662 MeV γ) etc.

ii) Gamma rays following nuclear reactions:

For gamma rays of higher energies, one has to get it from nuclear reactions:

$$^{4}He_2 + {}^{9}Be_4 \rightarrow {}^{12}C_6^{*} + {}^{1}n_0$$

$$\downarrow \rightarrow 4.4 \text{ MeV } \gamma$$

$$^{4}He_2 + {}^{13}C_6 \rightarrow {}^{16}O_8^{*} + {}^{1}n_0$$

$$\downarrow \rightarrow 6.13 \text{ MeV } \gamma$$

iii) Bremsstrahlung:

When fast electrons interact with matter, part of their energy comes out as electromagnetic radiation in the form of *bremsstrahlung*. The part of electron energy converted into bremsstrahlung is more for higher electron energy and for absorbing material of high atomic number, this energy conversion becomes highest.

iv) Annihilation radiation:

The positrons coming out of β^+ decay, initially looses energy in an absorbing medium. Near the end of their range, when their energy becomes very low, they combine with normal negative electrons in the absorber and disappear. Followed by this two oppositely directed γ-rays of energy 0.511 MeV comes out and this process of joining of one positron with an electron is known as annihilation.

v) Characteristics X-ray:

When the orbital electrons of an atom get disrupted from their normal configuration due to some excitation process, it causes the atom to move to an excited state for a short period of time. This is followed by a rearrangement of electrons amongst themselves which in turn bring back the atom to its ground state. All these things happen within a small time which is characteristically a nanosecond or even less for solid material. The liberated energy in the transition from the excited state to the ground state gets converted into a *characteristics X-ray* photon.

D. Neutron sources

i) Spontaneous fission: In each fission of a transuranic isotope there will be emission of few neutrons.

Example: ^{252}Cf

ii) Radioisotopes: By mixing an alpha emitting isotope with an appropriate target material a self-contained (α,n) source can be made.

Example: $^{4}He_{2} + {}^{9}Be_{4} \rightarrow {}^{12}C_{6} + {}^{1}n_{0} + 5.71$ MeV

iii) Photoneutron sources: It is based on providing requisite excitation energy to the target nucleus by way of absorption of a gamma ray photon

which results in emission of a free neutron.

Example: $^{9}Be_{4} + h\nu \rightarrow {}^{8}Be_{4} + {}^{1}n_{0} - 1.666$ MeV

$^{2}H_{1} + h\nu \rightarrow {}^{1}H_{1} + {}^{1}n_{0} - 2.226$ MeV

iv) Reaction from accelerated charged particles:

Example: (1) Deuteron – Deuteron Reaction

$^{2}H_{1} + {}^{2}H_{1} \rightarrow {}^{3}He_{2} + {}^{1}n_{0} + 3.26$ MeV

(2) Deuteron – Tritium Reaction

$^{2}H_{1} + {}^{3}H_{1} \rightarrow {}^{4}He_{2} + {}^{1}n_{0} + 17.6$ MeV

1.2.4. Interaction of Nuclear Radiation with Matter

A. Interaction of heavy charged particle

Heavy charged particles (e.g. alpha particles, fission fragments etc.) interact with matter by exerting coulomb force on orbital electrons and nuclei of the absorber atoms. In this process they lose energy while moving in the absorber medium. The *linear stopping power* for heavy charged particles in a given absorber is defined as

$$S = -(dE/dx)$$

'S' is also called *specific energy loss*.

The expression through which one can calculate the value of specific energy loss is known as *Bethe – Bloch formula* which is as written below:

$$-\ (dE/dx) = [\ (4\pi z^2 e^4) / (m_0 v^2)\ ]\ NB$$

$$\text{where } B = Z\ [\ln(2m_0 v^2 / I) - \ln(1 - v^2 c^{-2}) - v^2 c^{-2}]$$

ze and v are the charge and velocity of the primary particle, N and Z are the number density and atomic number of the absorber atoms, e is the charge of an electron and m_0 is the rest mass of an electron.

The plot of specific energy loss against the distance of penetration in the absorber is known as Bragg curve and it is shown in Fig.1 below:

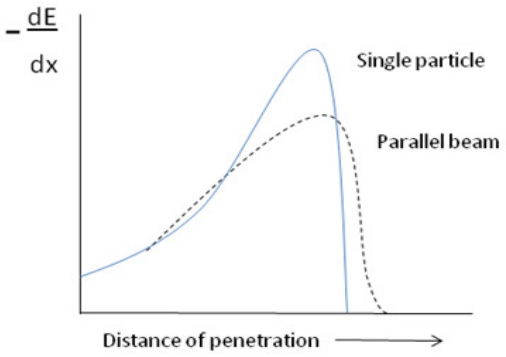

Fig.1. Bragg curve

It can be seen from the curve that there is a peak immediately before the particles come to rest. This is due to the fact that decrease in particle energy results in increase in interaction cross section [8].

B. Interaction of fast electrons

For fast electrons as well, an expression for specific energy loss (due to ionization and excitation) was derived by Bethe. Unlike heavy charged particles, electrons lose energy in an absorber both by collisional losses (i.e. ionization and excitation) and also by radiative losses.

$$(dE/dx)_{Total} = (dE/dx)_{collision} + (dE/dx)_{radiative}$$

The ratio of the specific energy losses can be written as

$$[\ (dE/dx)_{radiative} / (dE/dx)_{collision}\ ] \approx [\ EZ / 700\ ]$$

where E is expressed in MeV [1].

C. Interaction of gamma rays

There are three physical phenomenon through anyone of which gamma rays (i.e. gamma ray photons) interact with absorber atoms and lose energy. These are as follows:

i) Photoelectric effect: The energy of a gamma ray photon is transferred to an outermost orbital electron and it comes out from the absorber as photo-electron. This process is predominant for absorber of higher atomic number and/or for gamma ray photons of lower energy. Probability of photoelectric absorption per atom over all ranges of E_γ and Z can be expressed as: $\tau = \text{const.} \times (Z^n / E_\gamma^3)$ where n varies between 4 and 5 [1].

ii) Compton effect: In this process, the gamma ray photon transfers part of its energy to an electron (considered to be at rest in the beginning) of the absorber atom and then get deflected through an angle θ from its original direction. The electron which receives part of the energy of the incident photon is known as *recoil electron*.

For any given interaction, the relation between energy transfer and the scattering angle can be expressed as:

$$h\nu' = h\nu/[\ 1 + (h\nu/m_0c^2)(1 - \cos\theta)] \qquad [1]$$

The probability of Compton scattering per atom of the absorber increases linearly with Z as it depends on the number of electrons available as scattering targets.

iii) Pair Production: When the energy of the incoming gamma ray, attains twice the rest mass energy of an electron (1.02 MeV) or has even more than this value, the process of pair production becomes energetically possible and this process is predominantly confined to high energy gamma rays.

Gamma ray attenuation

If 'μ' is the probability per unit path length that a gamma ray photon is removed from a beam of gamma ray energy, then μ can be expressed as

$$(I/I_0) = e^{-\mu t}$$

where μ = linear attenuation coefficient

I = number of transmitted photons after crossing absorber of thickness 't'.

I_0 = number of photons without an absorber

The coefficient μ depends on density of absorber material (ρ) and the gamma ray attenuation expression can be written as

$(I/I_0) = e^{-\mu t} = e^{-(\mu/\rho)(\rho t)}$ where (μ / ρ) is called mass attenuation coefficient [9].

D. Interaction of neutrons

i) Slow neutron interaction: Slow neutrons interact with absorber material either by some neutron-induced reactions or by elastic scattering with absorber nuclei. Each neutron induced reaction gives rise to secondary radiations of adequate energy which can be detected directly.

Example: (n,α) reaction

$$^{6}Li_{3} + ^{1}n_{0} \rightarrow ^{4}He_{2} + ^{3}H_{1} + 4.76 \text{ MeV}$$

In this reaction alpha comes out as secondary radiation which can be detected easily.

ii) Fast neutron interaction: A fast neutron, when collides with an absorber nucleus, it transfers a significant part of its energy to the nucleus and for such interactions, the secondary radiations are the *recoil nuclei*. If the energy of the fast neutron is too high, it may take part in inelastic scattering with an absorber nucleus and put the nucleus in an excited state which subsequently de-excites by emitting a gamma ray. If the absorber is hydrogen then the neutron loses almost its entire energy in a single collision. So to detect fast neutrons one needs to opt for *proton recoil*.

1.3 SEMICONDUCTOR RADIATION DETECTORS

1.3.1 Successful working of semiconductor detector needs a junction:

Let us consider a Silicon bar of following specifications:

Resistivity (ρ) = 10^4 Ω-cm, Length (L) = 1cm and cross-sectional area (A) = 1 cm^2 ,

Then the resistance of Silicon bar (R) = ρ (L/A) = 10000 Ω

So for applied bias of 1V, current (i) = 100 μA

Now, charge generated by 1 MeV particle in Silicon by ionization

$$Q = (E/\varepsilon) \times e = (10^6 / 3) \times 1.6 \times 10^{-19}$$

$$= 0.5 \times 10^{-13}$$

where ε = minimum energy required to produce one electron-hole pair in Silicon

= 3.62 eV

If charge collection time is ~ 10^{-8} sec, then the current due to 1 MeV particle will be of the order of few micro amperes. So the signal will be completely merged in noise. Hence junction is essential as it reduces the current in the absence of radiation.

1.3.2 Types of junctions based on fabrication methods:

Based on fabrication method, junctions can be classified into four categories which are as follows:

i) Diffused junction: Junction is formed due to thermal diffusion of impurities.

ii) Schottky barrier: Junction is formed out of metal-semiconductor contact. Here junction formation is based on the semiconductor surface condition (i.e. density of surface states), work function of metal and semiconductor work function. As this barrier or junction forms very near to the surface of the semiconductor (5 – 10 ˚A), it is also called ***surface barrier***. This thesis mainly deals with *surface barrier* type of detectors. There are four types of surface barrier detectors:

a) Partially depleted (used for measuring energy of incident radiation and hence also known as *E-type detector*)

b) Fully depleted (used for measuring type of incident radiation and hence also known as Δ*E-type detector*)

c) Annular (used to measure angular correlation)

d) Position Sensitive Detector (used to get position information of the incident radiation and also called PSD)

Both annular and position sensitive detectors may be utilized either in partially-depleted mode or in fully-depleted mode. Apart from above four, surface barrier detectors also find its application for neutron energy (both slow and fast) and flux measurement.

iii) Ion-Implanted junction: Introducing impurity by impinging accelerated ions on to semiconductor substrate.

iv) LASER doping: At first, the impurity elements are to be evaporated on to semiconductor substrate. Then this surface of the substrate will be exposed to LASER beam to form a junction.

1.3.3. Important development of semiconductor detectors (Chronological order):

1) Semiconductor Counter (Point Contact) 1951 [3]
2) Germanium Surface Barrier 1955 [2]
3) Silicon Surface Barrier 1958 [10]
4) Silicon Diffused Junction 1959 [11]
5) Lithium Drifted Silicon 1960 [12-13]
6) Lithium Drifted Germanium 1962 [14]
7) Ion Implantation 1964 [15]
8) Cadmium Telluride 1967 [16]
9) High Purity Germanium 1972 [17]
10) Gallium Arsenide 1970 [18]
11) HgI_2, PbI_2 1971 [19]
12) Diamond detector 1975 [20]
13) semiconductor drift detector 1984 [2l] etc.

Existing semiconductor detectors differ from one another mainly because either the material which is adopted for fabrication or the method of treating the material is not same. Fabrication methods of various detectors, their working principles and operational characteristics are very much available in the literature [22-24] (Tsoulfanidis, 1995; Akimov, 2007).

1.3.4. Material requirement to fabricate good detector:

1) High Resistivity – For large depletion depth

2) Small Band Gap (E_g) – For good energy resolution

$$\varepsilon = 2.67E_g + 0.87, \text{ No of ion pairs}=E/\varepsilon$$

3) Indirect Band Gap – For high carrier lifetime

4) High Atomic Number – As probability of photo electric absorption is directly proportional to Z^5

5) High Mobility (μ) And Life Time (τ) – For good charge collection

6) Availability in Large Sizes – As it improves efficiency

7) Contact – Electrical contact should be easy to make as well as must be stable

1.3.5. Energy resolution of radiation detector:

Energy resolution is a parameter which is directly linked with the performance of a detector. It is measured as the width of the pulse distribution of a mono-energetic source, measured at half of the maximum of the Gaussian shaped curve. This width measured at half of the maxima is also known as FWHM (Full Width Half Maxima). The definition of energy resolution can be expressed as the capability of a detector to differentiate the incident particles with respect to energy.

There is another way of expressing energy resolution as follows:

FWHM = $2.35\sigma = 2.35\sqrt{N} = 2.35\sqrt{(E/\varepsilon)}$

Energy Resolution = $R(E_0) = [FWHM/E_0]$

$R(E_0)$ is normally expressed as a percentage.

1.3.6. Factors affecting energy resolution:

$\Delta^2 = (\Delta\eta)^2 + (\Delta\gamma)^2 + (\Delta g)^2 + (\Delta s)^2 + (\Delta w)^2$

where Δ = total spectral broadening in the energy spectrum

Now, $\Delta\eta$ = Spectral broadening due to fluctuation in no. of ion pairs

$= 2.35\sqrt{(Fe\varepsilon)}$

Again ε = minimum energy required to create one electron-hole pair

= 2.98 eV Ge

= 3.60 eV Si

F = Fano Factor = 0.1 for silicon

For 5 MeV alpha, $\Delta\eta$ = 3.0 keV

$\Delta\gamma$ = Spectral broadening due to nuclear collision

$\alpha\ Z^{1/2}\ A^{4/3}$

For alpha particles, $\Delta\gamma$ = 5 keV

Δg = Spectral broadening due to detector + amplifier noise

$(\Delta g)^2 = [\ (\Delta C)^2 + (\Delta Y)^2\ ]$

ΔC = spectral broadening due to detector capacitance

$= 2.35\ (\varepsilon/e)\ (KTC)^{1/2}$ keV

$= 3.29\ (\ C\ )^{1/2}$ keV where $C(pF) = [106\ (pF/m) \times A(mm^2)/d(\mu m)]$

and ΔY = Spectral broadening due to amplifier noise

For C = 25 pF, ΔC = 16 keV and ΔY = 6 keV

$(\Delta g)^2$ = 256 + 36 = 292 keV

ΔS = Spectral broadening due to source ≈ 4 keV

ΔW = Spectral broadening due to dead layer = $23.5\sqrt{x}$

where x = Window thickness in microns

For S.B. detectors, $\Delta W \approx 4$ keV

So, Δ^2 = 9 + 25 + 292 + 16 + 16; Hence $\Delta \approx 20$ keV

So, the detectors of 10 mm diameter, 25 pF junction capacitance and operated at room temperature can offer energy resolution of 20 keV.

To fabricate detectors of higher energy resolution, one needs to fulfill the following two major requirements:

(A) Smaller value of detector capacitance which can be achieved by fabricating detector of smaller active area.

(B) By reducing the reverse leakage current because the value of FWHM will be larger for higher value of leakage current. This is evident from the following expression:

$$(FWHM)_{I\text{-}Rev} = 34.4\ (\text{`I-Rev'} \times \Gamma_a)^{1/2}\ \text{keV}$$

Where 'I-Rev' is the reverse leakage current (in μA)

and Γ_a is the amplifier shaping time (in μS)

1.3.7 Various components of reverse leakage current:

1. Diffusion Current is due to generation of minority carriers in the undepleted region within the diffusion length of space charge region. It is very small current.

$$I_D = 16\rho_P / \tau_e^{1/2}\ \text{nA/cm}^2$$

for a typical diffused junction detector, $\rho_P = 1$ kΩ-cm & $\tau_e = 500$ μs,

$I_D = 0.73$ nA/cm^2

2. Generation current is the major source of leakage current resulting from thermal generation of e-h pairs in space charge region. The accurate knowledge of space charge current depends on recombination of carriers in the depletion region

So, $I_g = edn_i / 2\tau_e$ μA/cm^2 $= 1.2\ (\rho V)^{1/2} / \tau_e$

For $\rho = 1$ kΩ-cm, $\tau_e = 500$ μs, V = 100 V, we get $I_g = 0.72$ μA/cm^2. This is much higher than diffusion current. Here 'd' is the width of depletion region. It is desirable to have long lifetime (τ_e) of carriers, not only to achieve complete charge collection, but also to minimize generation current.

3. The third and most important contribution to current comes from **generation at surface states** or conduction through surface channels at the edge of the device. This is primarily due to manufacturing procedures.

4. The final contribution to the current comes from the contacts. In surface barrier detectors this current comes from the electron emission over the potential barrier created by metal-semiconductor contacts and is given by

$$I_e = A^* T^2 e^{q(\Phi - \Delta\Phi)} / kT$$

where A^* = Richardson's constant for thermo ionic emission, Φ = Potential barrier and ΔΦ = correction term for the barrier lowering by the image force. The contribution of this current is significant in fully depleted detectors.

5. In Si Surface barrier detector the additional source of current is due to light. Any light penetrating the depletion layer of the device contributes to increase in *dark current*. The surface barriers with thin gold films have high photosensitivity of value

0.13 μA/μW at 5460^0A wavelength is generated in case of a detector covered with 50^0A gold film.

1.4 NUCLEAR ELECTRONICS TO PROCESS SIGNALS FROM DETECTORS

1.4.1. Alpha spectrometer

Alpha spectroscopy is a technique which determines the energy of the alpha particles emitted by radioactive substances. A detailed analysis of the alpha energy spectrum is used to determine the identity of the alpha emitters. The equipments used in alpha spectroscopy includes a detector, a high voltage power supply, a preamplifier, a main amplifier which is followed by a PC based Multi Channel Analyzer (MCA). The block diagram of a typical alpha spectrometer is shown in the figure (Fig.2.) below:

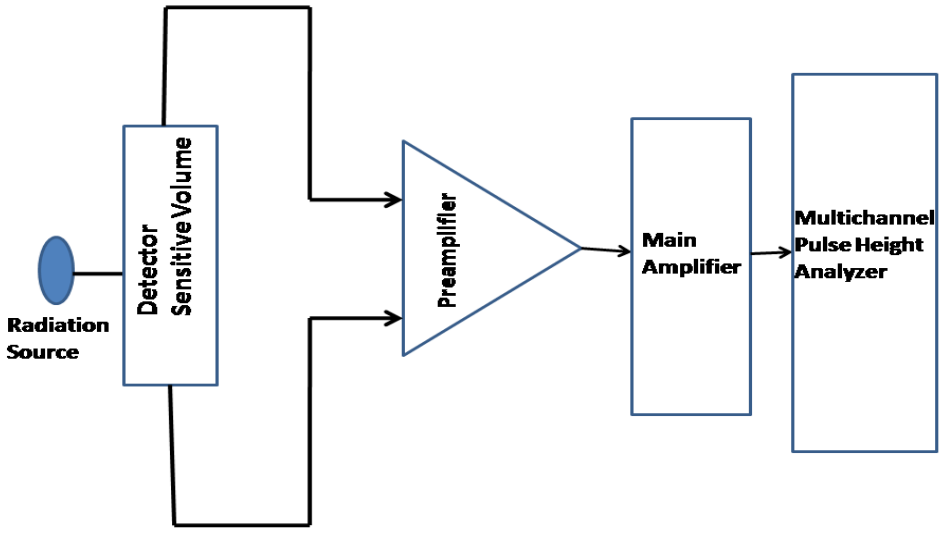

Fig.2. Block diagram of a typical alpha spectrometer

1.4.2. Preamplifier

The electrical signals at the output circuit of a detector, generated as a result of interaction of the impinging radiation, in the sensitive volume of the detector are usually small and must be amplified prior to their processing to get information about the nature of the incident radiation. The amplification of the signal is carried out with the help of a preamplifier, mounted very close to the detector. The output of the preamplifier is fed to a main amplifier which can be far away. Apart from providing some signal amplification, the preamplifier must also be able to drive several feet long coaxial cable to deliver the signal to the main amplifier.

1.4.3. Main amplifier

The amplifier plays two roles viz. *amplifying* as well as *shaping* the signal. The output signal of preamplifier is usually of a few millivolts and can't travel very far so that it can be manipulated in substantial way without losing any information it contains. These amplifiers can increase the amplitude of the input signals to as many as 2000 times in certain models [22].

1.4.4. Multi-Channel Analyzer (MCA)

The multi channel analyzer separates the pulses based on their pulse-height. Each channel corresponds to a small energy range of alpha particles. The pulse height is proportional to the quantity of energy lost by an alpha particle. By analyzing the spectrum of emitted alpha particles, the elements that caused the alpha pulse can also be identified. For our measurements, in order to obtain alpha spectrum, we use MCA

of one thousand channels for alpha particles of energy ~ 5 MeV from ^{241}Am – ^{239}Pu dual source.

1.5 Scope of the Thesis Work

Silicon based alpha particle and heavy ion detectors especially surface barrier detectors – both partially depleted as well as fully depleted (i.e. thin detectors) – play an important role in low to moderate energy physics experiments in accelerators [25]. These detectors are having numerous advantages namely, excellent energy resolution, efficient particle identification, linear energy response, variety of configuration, compact and small power requirements. These detectors work on the principle of ionization in solid medium by the incident radiation which is followed by collection of electron-hole pairs. One of the major issues in these silicon detectors is the high leakage current through edges of Si/metal junction. The usual practice to reduce the leakage current is either by passivating the edges of silicon using some passivating material (e.g. silicon di-oxide, organic epoxy etc.) or employing guard – ring structure at the edge. Silicon di oxide passivated detectors are very much known for their highly stable performance over a long period of time. However, epoxy has a disadvantage as it is hydrophilic in nature and water vapour present in the atmosphere affects its long term stability. So, in order to enhance the longevity as well as better leakage behavior of the detectors, it will be appropriate to explore some more new passivating material that do not degrade with atmospheric exposure. In this context, highly resistive tin oxide layer through organic route (i.e. by depositing initially Self Assembled Monolayer of Amine-based organic thin film with the help of Langmuir Blodgette

(L-B) deposition technique and subsequent heating of the layer causes its decomposition to tin oxide), which can be deposited easily on silicon [26-27], is ideal as the grown tin oxide is environmentally stable. It is also reported in literature [28] that hydrogenated amorphous silicon, which can be deposited easily on silicon, provides good passivation and brings down the detector leakage current. In this thesis, passivation of silicon surface by highly resistive tin oxide as well as by hydrogenated amorphous silicon was studied. With regard to Silicon dioxide passivation, one can find many reports in literature about development of Silicon dioxide passivated E-type detectors [29]. On the contrary, almost no information is available about fabrication of Silicon dioxide passivated ΔE detectors by way of thinning oxide-grown thick Silicon samples to desired thickness. Thickness and area of ΔE detectors needs to be tailor made depending on user requirements. Hence few Silicon dioxide passivated delta E detectors were fabricated. Also effort has been put to fabricate few rugged surface barrier detectors by employing better adherent gold film on silicon. Typical block diagram for the fabrication of E-type and ΔE-type detectors is shown in (Fig.3.) below:

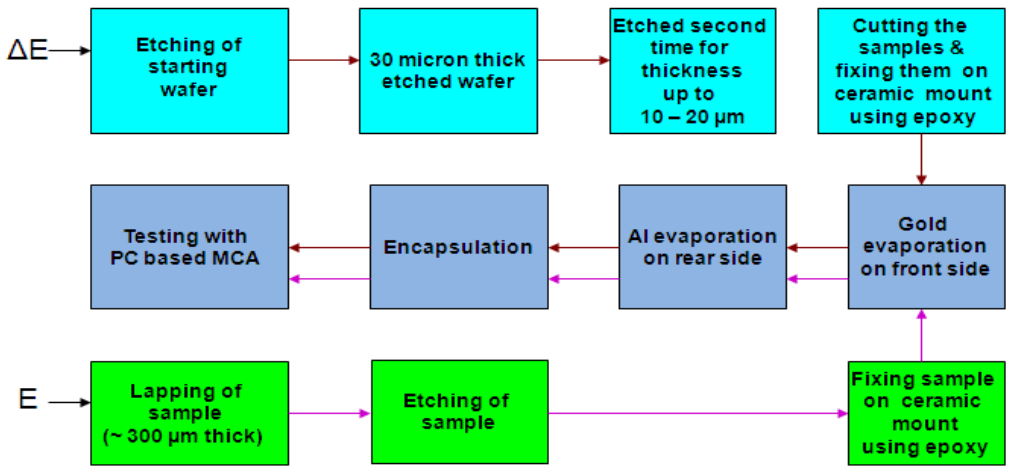

Fig.3.Typical block diagram of the steps to fabricate E-type and ΔE-type detectors

In addition, possibilities of using organic semiconductors (CuPc, CoPc etc.) and silicon hybrid structures for alpha detectors were also thoroughly investigated [30-32]. The motive behind these studies was to tap the advantages of organic based devices (the ease of fabrication and low cost) for charged particle detectors. Further, some studies on polymer based alpha detectors were also carried out. Since the radiation creates charge carrier inside polymer matrix by ionizing the medium, these charge carriers can be utilized for detection. Such studies will help to design new polymer based flexible and portable alpha detector at low cost. In short, this thesis work deals with use of organic materials for nuclear detectors. In nuclear detectors, organic materials can find different roles such as passivation layer in silicon surface barrier detectors; as an active layer in organic/Si hybrid structures and pure organic material as an active layer. In addition, a new cleaning methodology of silicon surface using cold plasma along with optimization of plasma conditions was also studied at length. From the experimental results of the above study, we can conclude that the leakage current of the detectors treated with cold plasma generated by a mixture of Argon and Air (90% Argon and 10% Air) is very much lower as compared to the surface barrier detectors fabricated by us after treating the detector surface using organic solvents.

Chapter 2

DEVELOPMENT OF STABLE NUCLEAR DETECTORS BASED ON Au/ORGANIC THIN FILM/n-Si STRUCTURES

2.1 INTRODUCTION:

It is well known that organic materials undergo irreversible chemical processes under high dose of irradiation. This phenomenon is known as radiolysis and it occurs due to rupture of C – H and C – C bonds which in turn produces new organic compounds [33-35]. In spite of all these facts, several researchers have reported in the literature [36-37] about the successful applications of organic thin films in electronic and opto-electronic devices. Among the organic compounds in general and organic semiconductors in particular, porphyrins [38], phthalocyanines [39] and conducting polymer etc. were extensively used to develop these devices. Later on, the organic compound copper-phthalocyanine (CuPc), in the form of thin films was used in the fabrication of nuclear detectors by J. Assaf and A.Al-Mohamad [40] and they reported remarkable performance and highly improved resolution of the fabricated detectors of Au/organic thin film/n-Si structure as compared to Au/n-Si (Schottky type) detectors fabricated on identical substrates and also under similar conditions. Any detector developer looks forward to adopt new fabrication methodology which yields better performance (i.e. lower leakage current and higher energy resolution etc.) and nuclear particle detectors with CuPc/n-Si heterojunctions was exhaustively studied by us before working for the scope of the present thesis [30]. It was observed that the alpha energy resolution of the fabricated Au/CuPc/n-Si detectors was much better than a similar detector reported by Assaf et.al. [40] and is nearly same as that of similarly prepared Au/n-Si surface barrier detectors. It was also observed by us that Au/CuPc/n-Si detectors show lower leakage current than the similarly fabricated Au/n-Si surface barrier detectors [30]. One of the five year old detectors (fabricated by us) of Au/CuPc/n-Si structure, which offered best performance at the time of fabrication,

was studied for its leakage behavior and alpha energy resolution to evaluate the aging effect of such detectors [31]. The results indicated very good stability of Au/CuPc/n-Si detectors for particle detection. Of late it was reported that MnPc, FePc and CoPc have stronger binding with gold than NiPc, CuPc and ZnPc as suggested by the theoretical calculations involving free standing molecules of 3d-transition metal phthalocyanines (MPc, where M stands for anyone of Mn, Fe, Co, Ni, Cu or Zn) on gold [41]. Accordingly, CoPc was utilized to fabricate semiconductor heterojunction detectors in place of CuPc and it yields some promising results in terms of lower leakage current and good stability [32].

2.2 EXPERIMENTAL

The cross-sectional diagram (side view) of the Au/CoPc (or CuPc)/n-Si detector is given in Fig.1. Detailed procedure to fabricate CuPc/n-Si & CoPc/n-Si heterojunction detectors was described at length by A.Ray et. al [30, 31]. In the case of CuPc/n-Si detectors, only the aging effect on the best performing detector was studied whereas for CoPc/n-Si detectors, CoPc films of two different thicknesses (100 nm and 200 nm)

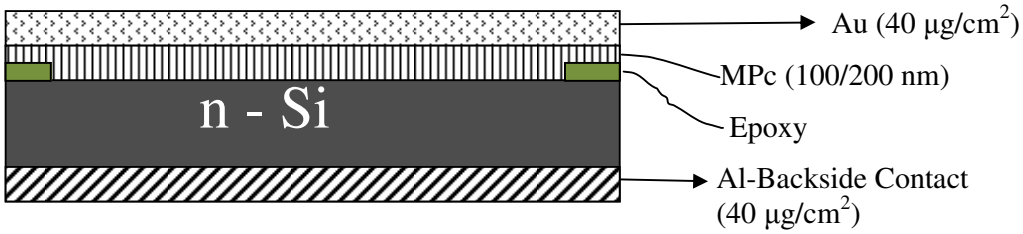

Fig.1. Cross-sectional (side view) diagram of a mounted Au/(CoPc or CuPc)/n-Si detector (in the figure 'M' stands for either Co or Cu).

were attempted to fabricate and evaluate the performances of the detectors. The detector with CoPc film of thickness of 100 nm is termed as Detector1 and for CoPc thickness of 200 nm, the detector is termed as Detector2. For preparation of MPc/n-Si heterojunction devices, silicon wafers (n-type) of 14.9 kΩ-cm resistivity and 34.3 mm diameter were taken as starting material. These wafers were lapped with various grain size carborandum/alumina powders which is followed by chemical polishing on both sides in 6:1:1 (by volume) solution of HNO_3:HF:CH_3COOH. Prior to deposition of MPc, the samples were thoroughly cleaned using trichloroethylene and dried. Then the wafers were mounted on ceramic mount using organic epoxy [a mixture of one drop epoxylite hardener No. 323 (obtained from M/s Epoxylite corporation, U.S.A.) and five drops of vacuum pumped CIBA 230 Araldite epoxy resin (Make: Hindustan CIBA-GEIGY Ltd. under license from CIBA-GEIGY AG, Basel, Switzerland)]. The thin organic film of metal(copper or cobalt)-phthalocyanine (MPc) with a thicknesses of 100 or 200 nm were thermally deposited on one side of the substrate using tantalum boat under high vacuum (~ 10^{-6} Torr). The depositions were carried out at a substrate temperature of 300°K. Subsequently, Au contacts (thickness ~ 40 μg/cm^2) were deposited on MPc films by thermal evaporation without exposing the films to atmosphere. Al Ohmic contacts (thickness ~ 40 μg/cm^2) were also deposited on rear side of the substrates. Subsequently, the edge-protection of the junctions were carried out with the help of an organic epoxy. After fabrication, the diameters of the active areas of the fabricated detectors were measured to be ~ 26 mm. The forward and reverse *I-V* characteristics were measured using Keithley make programmable electrometer. For CoPc/n-Si detectors, *I-t* characteristics were also studied. Alpha spectra for the fabricated detectors were obtained using ^{241}Am – ^{239}Pu dual α-particle

source and PC based multi-channel analyser. For alpha spectroscopic measurement, both the source and the detector were placed in a vacuum chamber (at ~ 10^{-3} Torr).

2.3 RESULTS AND DISCUSSIONS

2.3.1 Study of aging of CuPc/n-Si heterojunction detectors

The electrical (i.e. forward and reverse) characteristics and alpha energy spectra of these detectors were measured after a long gap of nearly five years and the best result obtained for the best detector is described below:

The forward characteristics of the detector, both 'As-prepared' (marked as 'A') and 'Five years later'(marked as 'B') are shown in Fig.2. It is observed that forward current is very much lower after five years as compared to its value in 'As-prepared' case. The answer lies in the fact that the backside aluminium contact turned out to be rectifying in nature over the period of time (aging of the detector) [42] which was actually ohmic in nature at the time of fabrication. The reverse *I-V* curve for both 'As-prepared' and 'Five years later' is shown in Fig.3. Though some marginal increase in leakage current is observed, which can be due to aging of the detector, the device still has good diode behaviour after a gap of five years. Degradation of organic epoxy due to aging may be the reason for this marginal increase. Both the alpha energy spectra (initial and after five years) were obtained by exposing the detector to alpha particles from ^{241}Am – ^{239}Pu dual source inside a vacuum chamber and the spectra are shown in Fig.4. The observed deterioration in energy resolution is not much significant during the span of five years.

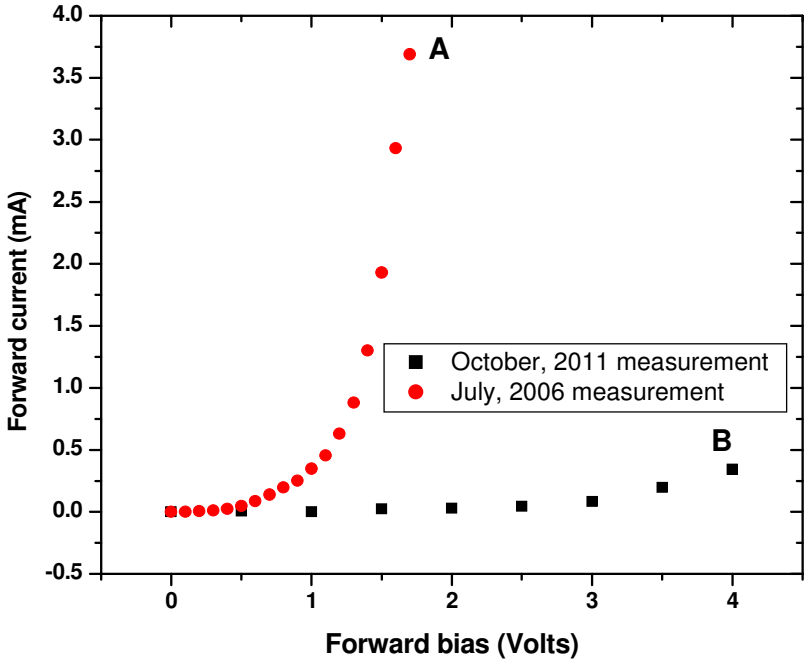

Fig.2. Forward Characteristics of Au/CuPc/n-Si heterojunction

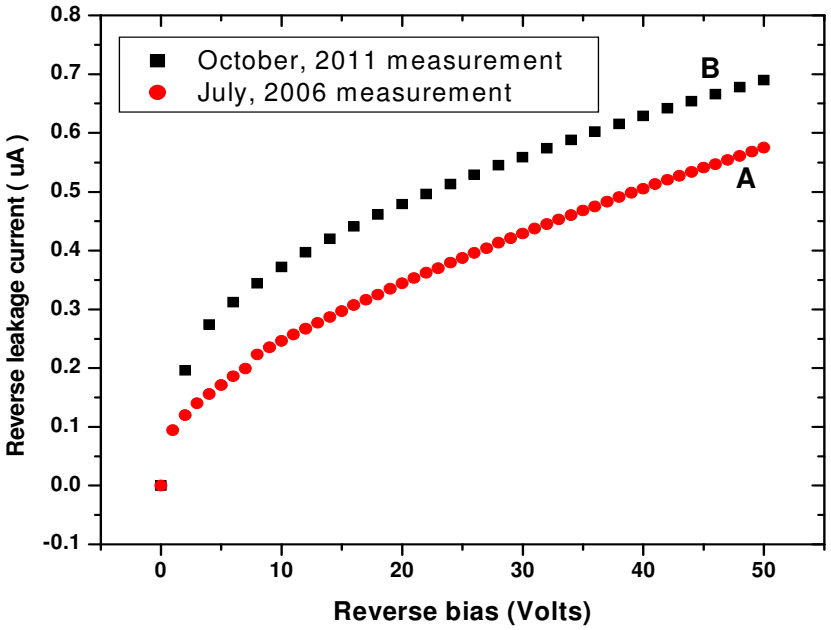

Fig.3. Reverse Characteristics of Au/CuPc/n-Si heterojunction

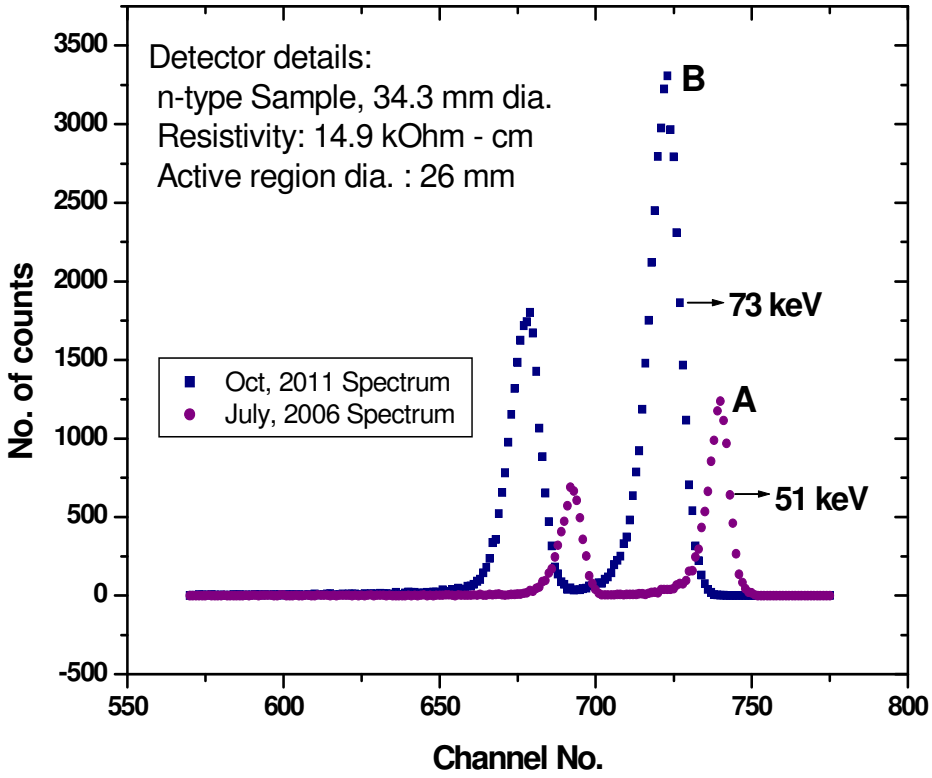

Fig.4. Alpha Spectrum of Au/CuPc/n-Si heterojunction

2.3.2. Study of CoPc/n-Si heterojunction detectors

The forward and reverse *I-V* characteristics of detectors 1 & 2, just after their fabrication are shown in Fig.5 & Fig.6 respectively. From their characteristics we can say that both the detectors showed good diode behavior. The I-V curves of Detector1 got fitted with diode equation with diode ideality factor of n = 0.536 which is shown in Fig.7. The *I-t* characteristics i.e. variation of reverse leakage current with time (elapsed) for a fixed value of applied reverse voltage (40 V) for both Detector1 & 2 are plotted in Fig.8. It is observed that the leakage behaviors of both the detectors are quite stable. Leakage current through Detector1 is almost double than that of Detector2 for a given applied bias.

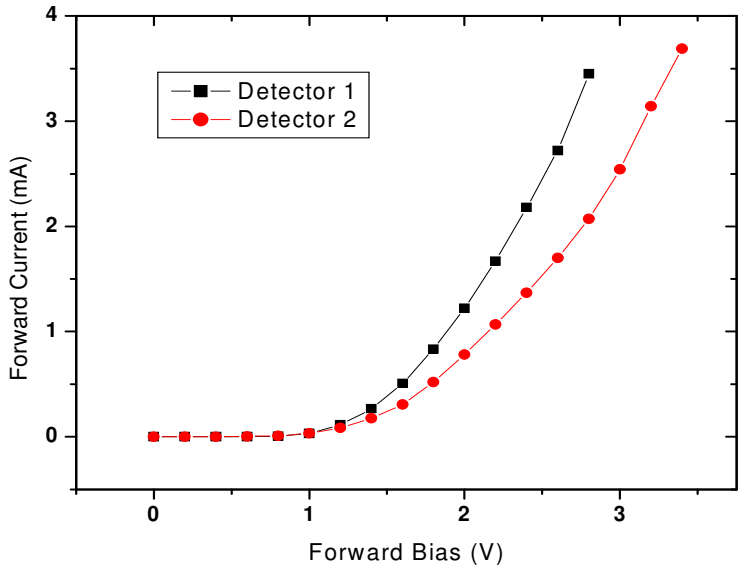

Fig.5. Forward characteristics of Au/CoPc/n-Si heterojunction

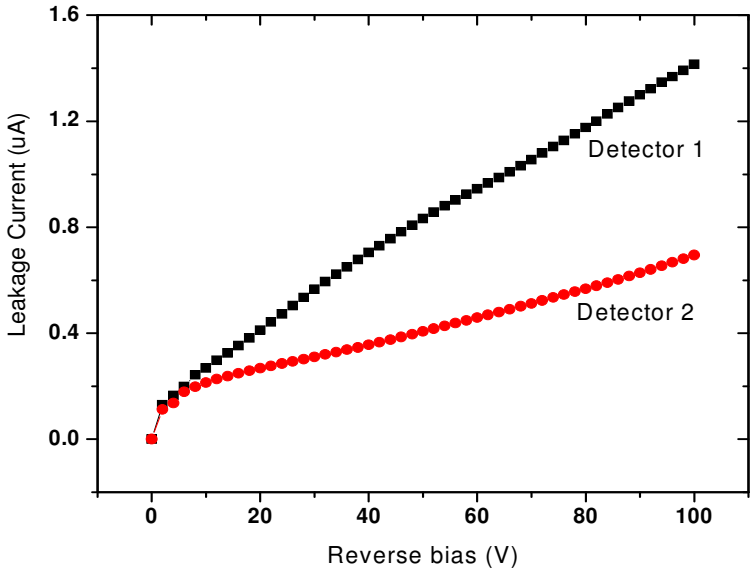

Fig.6. Reverse characteristics of Au/CoPc/n-Si heterojunction

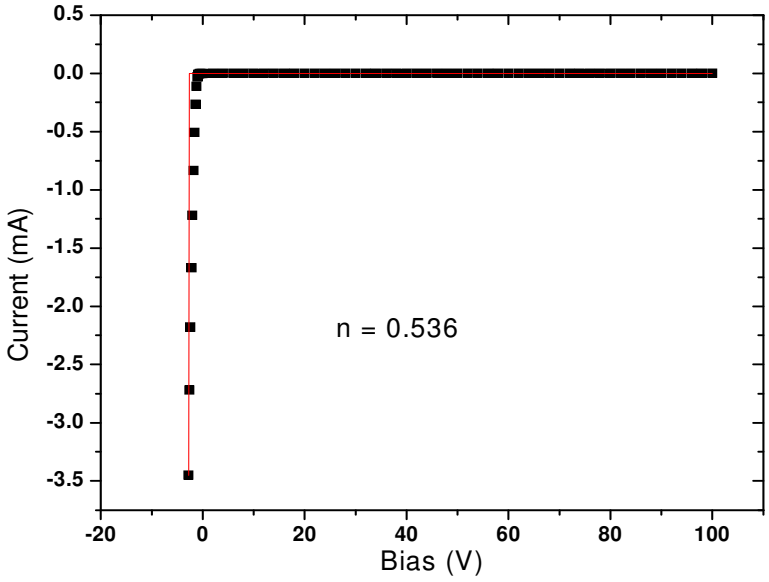

Fig.7. Determination of Diode Ideality Factor

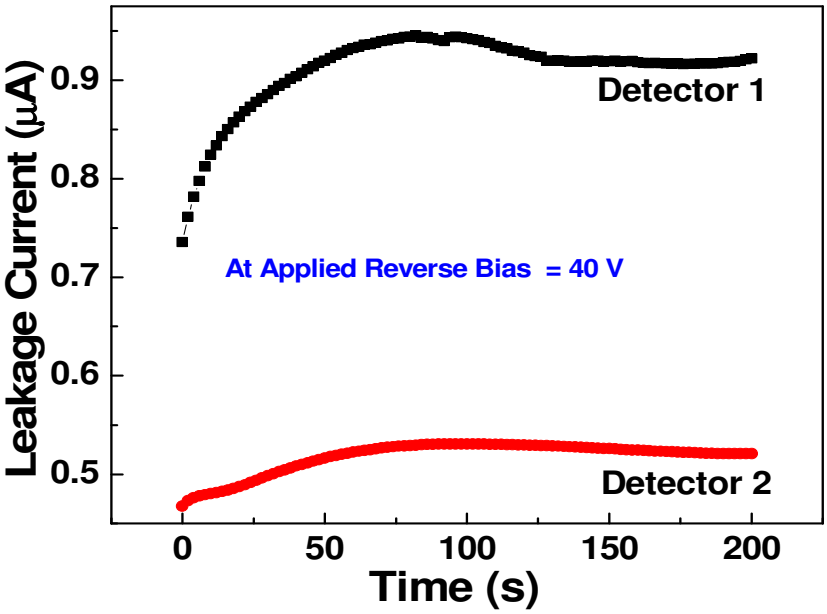

Fig.8. I-t characteristics for Au/CoPc/ n-Si heterojunction

Performances of both the detectors (i.e. Detector1 & Detector2) were studied over a longer period of time (15 Months) to get information about their 'long term performance'. Fig.9 (a) and (b) indicate the long term performance with regard to forward characteristics for Detector1 and Detector2 respectively. From the graphs marked "15 Months after fabrication" pertaining to both the detectors (i.e. Detector1 & 2), it is seen that the kink in the forward current (where it starts rising exponentially) shifts to a at a higher value of forward bias which may be due to aging of these detectors in a similar fashion to what has been already described for CuPc/n-Si heterojunction detectors above [31]. Actually there are two junctions, one at Au/CoPc which is forward biased and the other one at Al/n-Si junction. So when the detector was forward biased, though the Au/CoPc junction was forward biased, the weaker junction formed due to aging of the detector (i.e. Al/n-Si junction formed due to aging) was reverse biased. When the bias was increased, at certain higher value of voltage, the device started conducting.

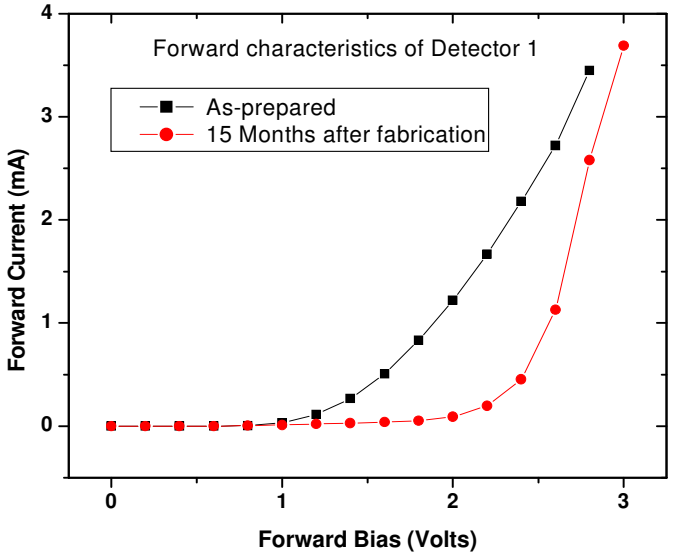

Fig.9(a). Long term performance of Detector1 for forward characteristics

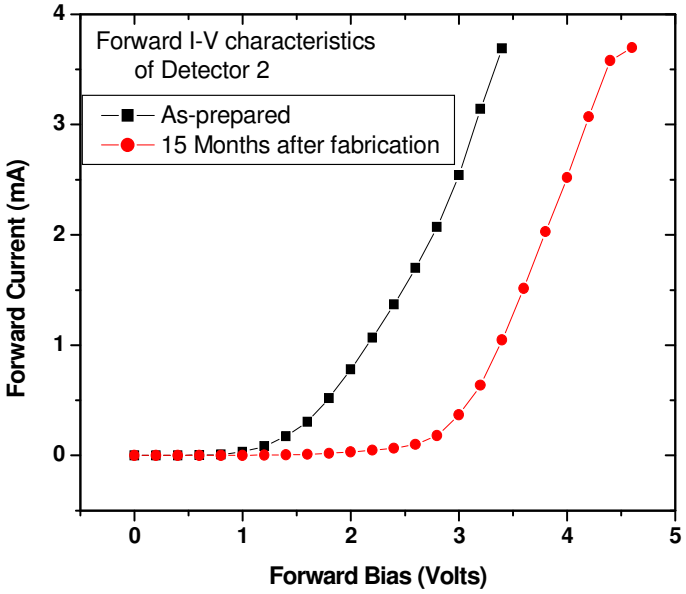

Fig.9(b). Long term performance of Detector2 for forward characteristics

Fig.10 (a) and (b) show the 'long term performance' of Detector1 & 2 with respect to their reverse characteristics. The result reveals only marginal increase in leakage current over a period of 15 months. Degradation of epoxy during the period of 15

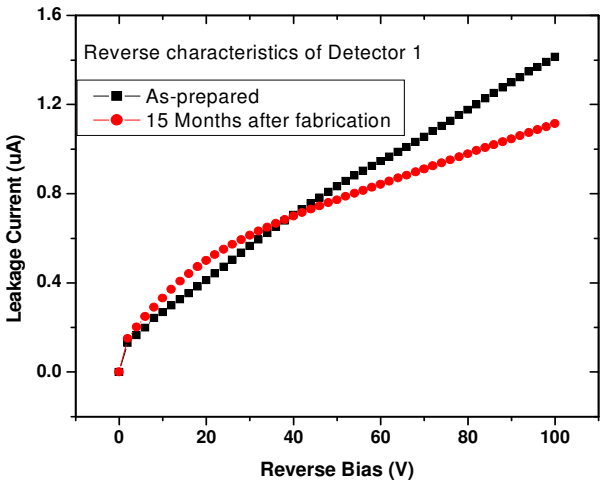

Fig.10(a). Long term performance of Detector1 with respect to reverse characteristics

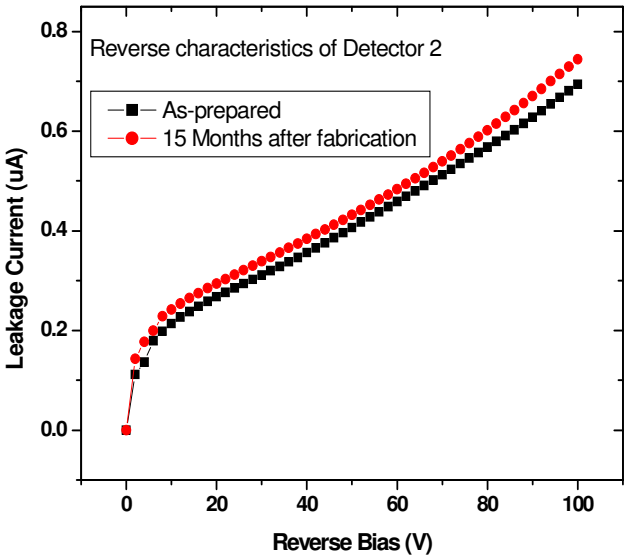

Fig.10(b). Long term performance of Detector2 with respect to reverse characteristics

months may be the primary reason for such marginal increase in leakage current. But only a marginal increase in leakage current over a time gap of 15 months indicates about good stability of the fabricated detectors. For alpha particle spectra, both the detectors (i.e. Detector1 & 2) placed under an alpha particle source (one detector at a time) of ^{241}Am – ^{239}Pu inside a vacuum chamber with vacuum of ~10^{-3} Torr. The distance between the source and the detector was maintained at 20 mm. These detectors could be operated smoothly up to a reverse bias of 500 V (applied through a load of 110 MΩ). The alpha spectroscopic measurements were carried out for both the detectors (Detector1 & 2) and the spectra corresponding to optimum resolution, studied just after fabrication as well as after a gap of 15 months for detectors 1 & 2 are shown in Figs.11 and 12 respectively. The energy resolution of Detector1 (41 keV) is

better than that of Detector2 (45 keV) though the active area of Detector2 (180 mm^2) is less than that of Detector1 (260 mm^2). This may be due to the double thickness of CoPc layer which might have created thicker dead layer for impinging alpha particles. Though the energy resolution was remained almost same for Detector1, it got decreased for Detector2 over a period of 15 months. To conclude, one can say that the detector fabricated with 100 nm CoPc, can be produced by simple fabrication method and offers stable performance over a period of 15 months. The graphs indicating the variation of energy resolution with reverse bias voltage for Detector1 & 2 is presented in Fig.13.

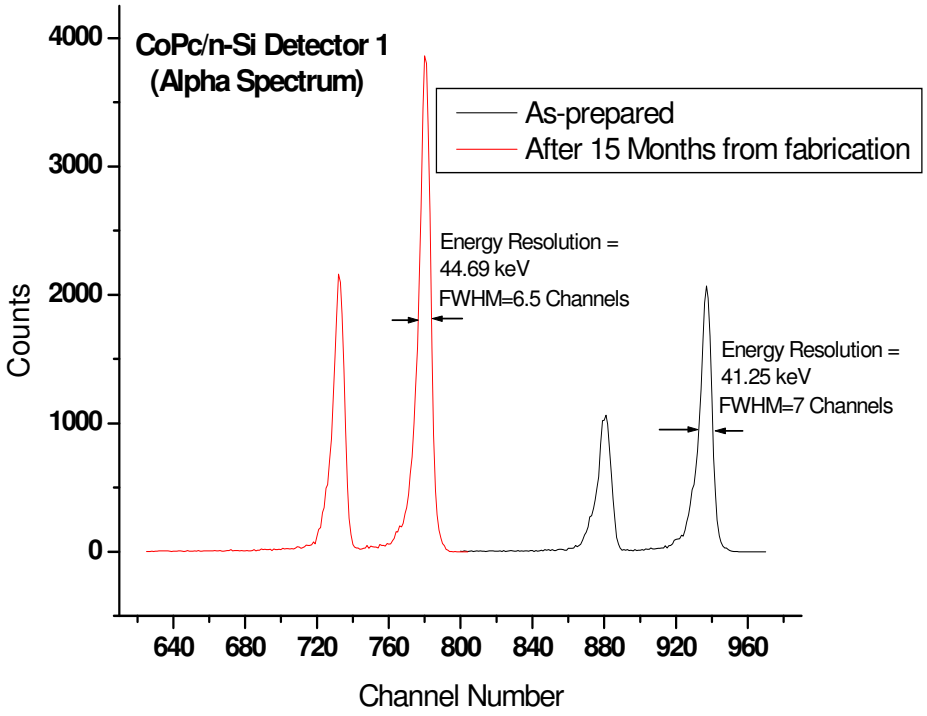

Fig.11.Alpha spectra of detector1 (energy resolution of 41.25 keV is obtained corresponding to Am-peak of energy 5.48 MeV).

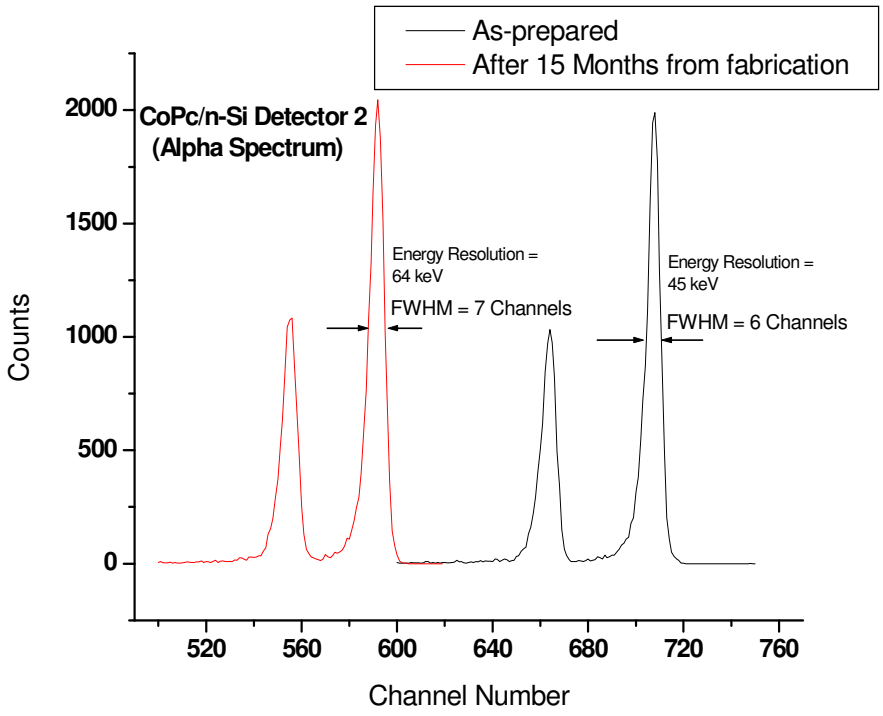

Fig.12.Alpha spectra of detector2 (energy resolution of 45 keV is obtained corresponding to Am-peak of energy 5.48 MeV)

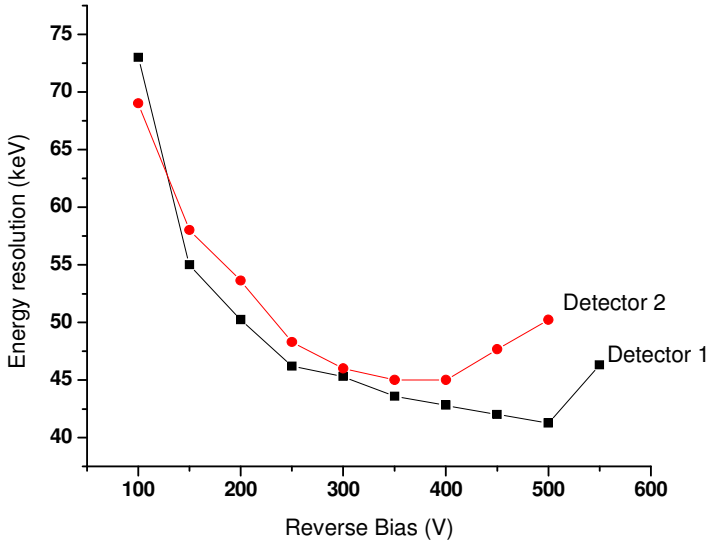

Fig.13. Variation of energy resolution with reverse bias

It is observed from these plots that the energy resolution improves with increase in reverse bias initially but after reaching the optimum value, subsequent increase in reverse bias causes deterioration of energy resolution. It can be explained as, at lower reverse voltage, the collection of the generated charge, in the active volume of the detector was incomplete. So there is improvement in energy resolution with increase in reverse voltage. But once all the generated charge got collected, further increase in reverse bias will augment the leakage current in the device which in turn deteriorates its energy resolution.

2.4 CONCLUSION

In this chapter we have discussed the modification of silicon surface barrier detectors to MPc/Si heterojunctions using CoPc (which is an organic thin film) as an active layer. Two film thicknesses have been tried to investigate the influence of organic active layer thickness in reducing the leakage current. It was observed that the leakage behavior is better than conventional surface barrier detectors with 100 nm thick CoPc layer. Long term stability studies over a period of 15 months also suggested stability and reliability of this new heterojunction device.

Chapter 3

SILICON SURFACE BARRIER DETECTORS USING ORGANIC/INORGANIC THIN FILMS AS PASSIVATION LAYER

3.1 INTRODUCTION

One of the major problem areas faced by a detector developer is the highly active nature of silicon surface. Passivation basically means the junction electrical properties are not significantly degraded at the surface compared to their bulk properties. It is now very much important to concentrate on the following data pertaining to silicon:

For silicon: No. of atoms/cc = 5×10^{22}

Hence number of atoms/cm.^{2} = $(5\times10^{22})^{2/3}$ = 1.85×10^{15}

So the no. of surface states = $1.85\times10^{15}\times$ 1-2 (No. of broken bonds)

= 2 to 4 times 10^{15}/cm^2

So the number of surface states is quite high which gives rise to larger value of leakage current. So silicon surface needs passivation to bring down surface states to ~ 10^{11} /cm^2. Various materials have been explored to passivate Silicon surface. Some of them are well established and are in regular use to fabricate silicon detectors e.g. amine-based epoxy (organic material) to fabricate silicon surface barrier detectors and silicon dioxide film (inorganic material) on silicon to fabricate diffused junction/ Passivated Ion-implanted Planar Silicon (PIPS) detectors etc. Now in the case of silicon surface barrier detectors, epoxy gets deteriorated with time and accordingly detector performance too gets deteriorated. In the case of diffused junction/(PIPS) detectors, the passivating oxide layer won't deteriorate with time but still some problem remains. PIPS detectors [43-44] are having lesser dead layers as compared to diffused junction detectors and are known for their long term stability. Unlike diffused junction detectors, the junction depth can be accurately controlled by choosing appropriate implantation energy for PIPS detectors and its fabrication involves low

temperature processes. But to fabricate PIPS, elaborate laboratory arrangement is required. Typical configuration of a PIPS detector is as given in Fig.1 and its fabrication steps are shown in Fig.2(a) &2(b) respectively. On the contrary fabrication of surface barrier detectors requires very simple set-up.

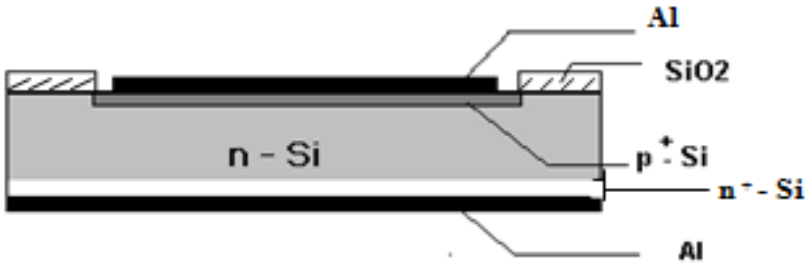

Fig.1. PIPS detector configuration

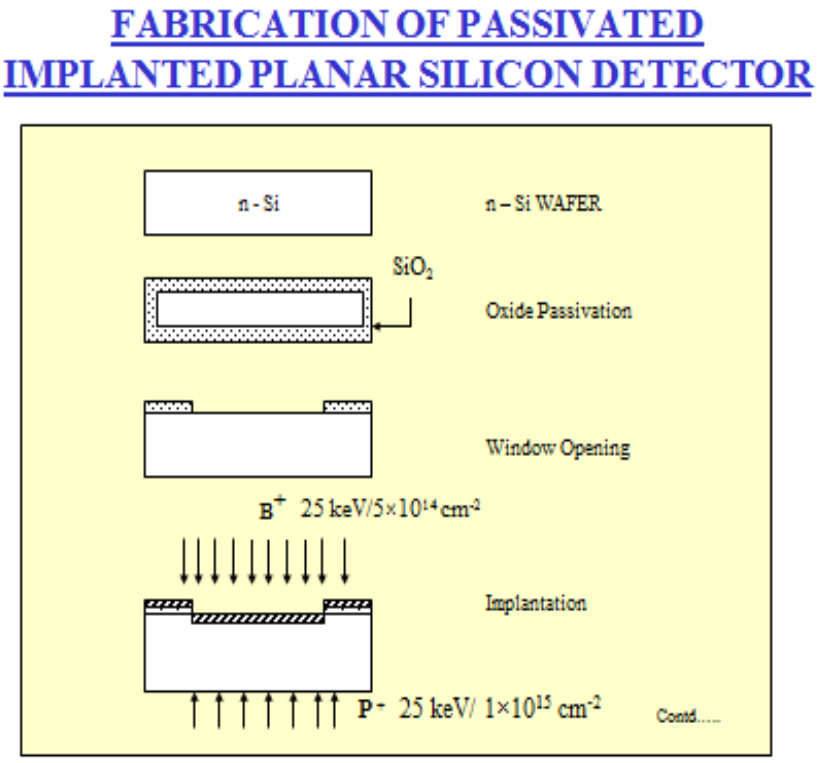

Fig.2(a) Initial fabrication steps for PIPS detector [1]

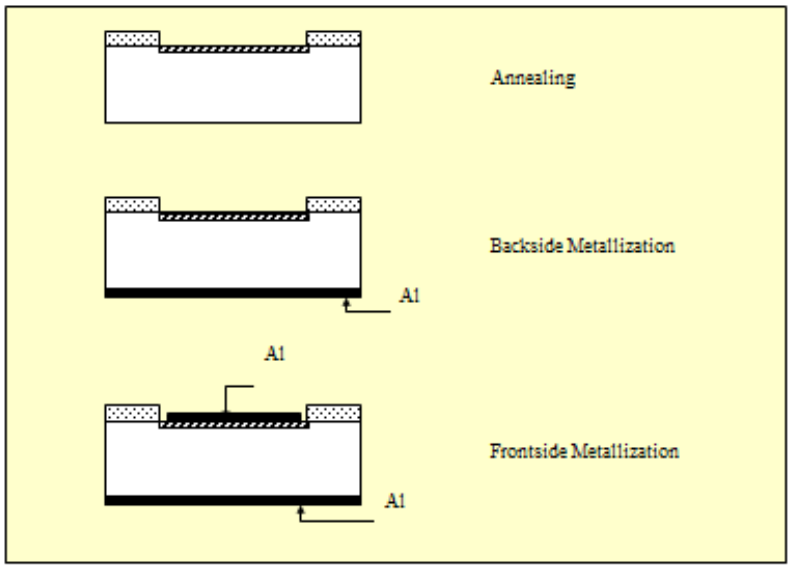

Fig.2(b) Remaining fabrication steps for PIPS detector [1]

Now in the case of surface barrier detectors, replacing epoxy by SiO_2 at the edge will improve its long term stability. In partially depleted E-type surface barrier detectors, replacing epoxy with SiO_2 is implemented well by detector manufacturers and is also reported in literature. But it is still not popular and not implemented in the cases of very thin (10 – 30 μm), standalone and fully depleted ΔE-type detectors. This is due to the fact that, heating such thin samples of silicon to 1000-1200˚C to grow SiO_2 on it is extremely difficult to achieve and these samples are more vulnerable to breakage due to thermal stress at this high temperature. But there is another method to develop oxide-passivated fully depleted ΔE-type detectors. In this method, one can start with growing SiO_2 on a thick sample and subsequently thinned down this sample to the desired thickness. This method was utilized to develop few prototype oxide passivated delta E detectors and it is described at length in the next section. Another problem

encountered in our laboratory is that the epoxy passivated surface barrier detectors are not rugged due to poor adhesion of thermally evaporated gold film on n-silicon surface. To overcome this, few detectors were fabricated using sputtered gold film on silicon in place of thermally evaporated gold film to make them rugged. This is also described in the next section.

It is well known that organic epoxy passivation of silicon surface can be done by utilizing two-component epoxy. In order to cure the epoxy resin properly, one needs to add right quantity of amine-based hardener. This results in very low leakage current and stable detector performance. This prompts one to make use of amine-based organic thin films as well either as passivation layer or it can behave as an organic route to end up with an appropriate passivation layer. With this motivation, a new material has also been explored to study edge-passivation of silicon surface barrier detectors. This new material is tin oxide film and was formed as a result of decomposition of an amine based L-B film [Octa Decyl Amine (ODA) – Stannate complex] by heating it to a moderate temperature (i.e. by organic route). Initially Octa Decyl Amine-Stannate Complex was deposited on Silicon wafer with the help of L-B deposition technique. Subsequently the silicon wafer was heated in a furnace in presence of air at a relatively low temperature (~ 600ºC). This heating causes decomposition of L-B layer to form SnO_2 film on silicon. This tin oxide contains SnO_2 nano-particles. This film of tin oxide offers very high resistance as it behaves like a depletion layer. Details of fabrication and test results of these detectors are described in the next section.

3.2 MATERIALS/ METHODS ADOPTED FOR Si-PASSIVATION

3.2.1 Silicon dioxide passivated delta E detectors

3.2.1.1 Experimental: Thin and fully depleted delta E detectors are very much useful for many applications e.g. particle identification measurements and heavy ion spectroscopy [45]. These detectors are Au/n-Si Schottky barrier detectors with their junction edges passivated by organic epoxy. Fabrication of these detectors are quite difficult because of the following two major factors viz. (a) device breakage due to brittleness of thin silicon and (b) difficulty in achieving desired thickness uniformity.

In an oxide passivated thin detector, the edge of the thin silicon wafer is covered by SiO_2 layer in place of organic epoxy (which is used in the case of epoxy passivated thin detectors). This was achieved by oxidation of thicker (greater than 500 micron) Silicon sample, followed by thinning, removal of SiO_2 layer from all sides of the sample except the radiation-entry side and finally opening a window there at the centre. To start with, an n-type Si-wafer of 1″ diameter, of nearly 800 µm thickness, 11 kΩ-cm resistivity and oxide layer on all sides of 0.7 µm thickness was taken as the starting material. The wafer was thinned to 30 µm (approx.) keeping the oxide layer only on the radiation-entry side with the help of in-house thinning technique which involves following four steps:

Step 1: Lapping of thick silicon samples was carried out in a lapping machine (Make: M/s Speedfam, Japan) to bring down the sample thickness up to 200 µm. The rotational speed of the lap plate was kept at 35 rpm. The slurry used, was made up of a mixture of 20 µm grain size powder and lapping oil.

Step 2: Preparation of samples of uniform thickness of 100 μm (with a thickness variation of ± 8 to 10 μm for samples of 1 inch diameter) has been carried out using a lapping fixture (Make: M/s Lapmaster, USA). The lapping fixture contains three adjustable screws (each screw provided with a diamond tip). Height of the screw tips from crystal mounting surface have been set at 100 μm with the help of the angular scale attached to each screw (coarse adjustment) and dial gauge (fine adjustment). Then by lapping the crystals mounted on this fixture (one crystal at a time), the thickness of the wafers was brought down to 100 μm.

Step 3: Etching/chemical polishing of the 100 μm thick samples was performed to bring the thickness down to 50 μm using 6:1:1 volumetric mixture of HNO_3:HF:CH_3COOH (etching solution of etch rate ~ 10 μm/minute).

Step 4: Final etching of ~ 50 μm samples is carried out to bring down the thickness below 25 μm using HNO_3 : HF :: 20:1 etchant (etch rate ~ 3 μm/minute).

Two 30 micron thick samples, thus prepared and window opened at the centre of the oxide layer were fixed on ceramic mounts. Then a thin gold layer of 20 nm was deposited onto the side having the oxide layer at the edge. On the backside, ohmic aluminum contact of 200 nm was deposited by vacuum evaporation at 10^{-6} Torr pressure. The ceramic mounts were held in transmission type metallic containers provided with microdot connectors. The results presented hereafter were obtained from the better performing detector. The detector is having active area of 95 (~11 mm dia.) mm^2. The photograph of the thinned sample is shown in Fig. 3 (a). Thickness variation within ± 2 μm was measured in sample area of 5-6 mm diameter.

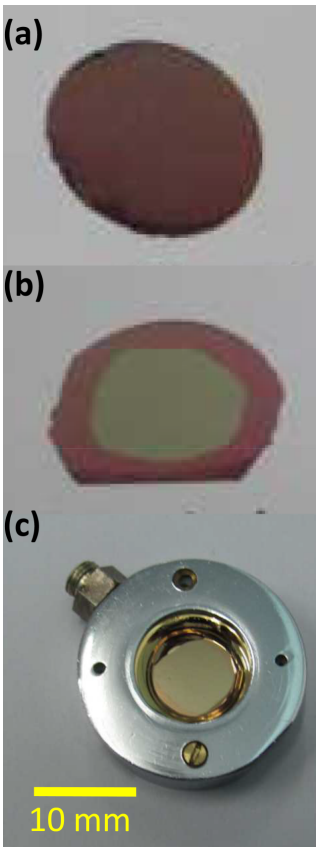

Fig.3. Photographs of different fabrication process steps of the ultrathin oxide-passivated silicon surface barrier detectors: (a) ultrathin (30µm) oxidized Si wafer; (b) oxide is etched out from the center; (c) final assembly of the detector after Au metallization

3.2.1.2 Evaluation of results & Discussion: The detector was tested for its I-V characteristics and alpha spectrum. The reverse characteristics of the detector are shown in Fig.4. For charged particle detection, the detector was exposed to α-particles from dual source (^{241}Am – ^{239}Pu). Alpha spectrum of the detector is shown in Fig.5.

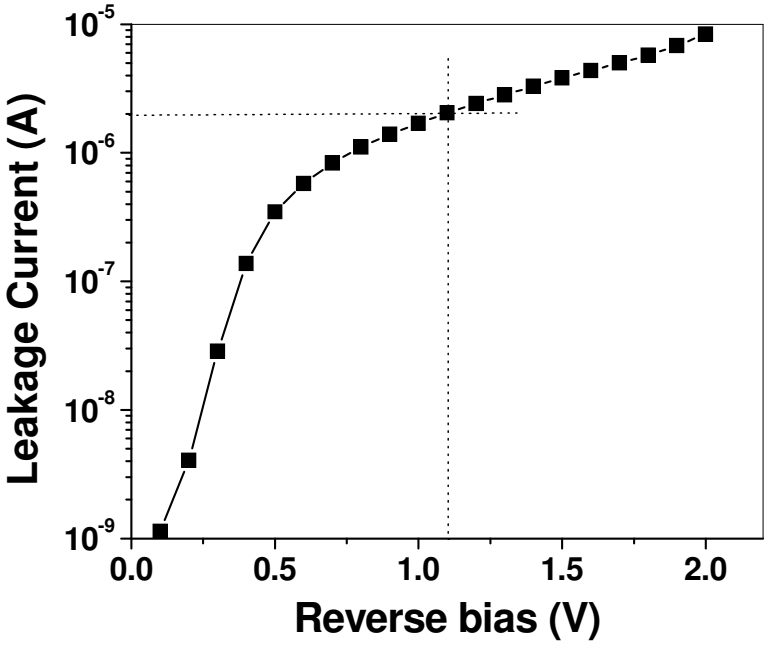

Fig. 4. Reverse characteristics of the detector (The dotted line indicates that a leakage current of >2μA occurs at >1.1 V).

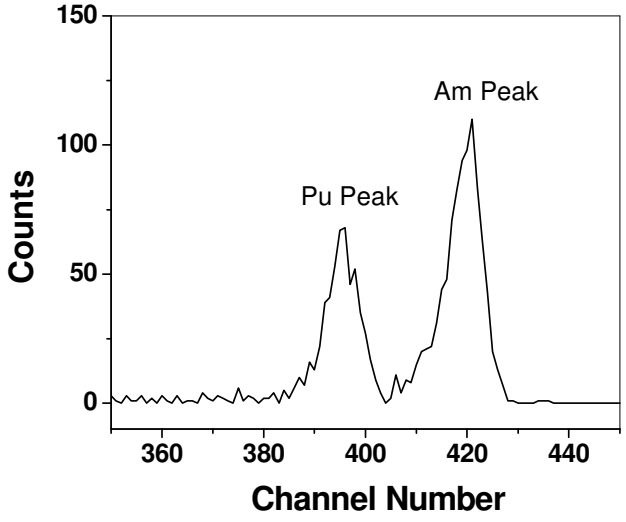

Fig.5. The Alpha spectrum obtained from the detector.

So the fabrication of thin (≤ 50 μm) of fully depleted silicon surface barrier detector with its edges passivated with thermally grown SiO_2 has been carried out successfully. The detector exhibited leakage current of 2 μA for an applied reverse bias of 1.1 V. It could also resolve Am and Pu peaks. Though the reverse characteristics and alpha spectrum of these detectors are comparable with our own standard detectors with edge passivation done by epoxy, these detectors promise durability because of silicon dioxide passivation. Better quality of oxide as a starting material may improve the leakage behavior and breakdown voltage further of these devices. This work was presented and published in DAE symposium on nuclear physics (2013) [46].

3.2.2 Development of epoxy passivated rugged silicon surface barrier detectors

3.2.2.1 Experimental: Detection and measurement of low energy nuclear particles and fission fragments are carried out mainly by surface barrier detectors. There are numerous fields where these detectors find their applications such as nuclear reaction, isotope identification, space research, biology and radiation dosimetry etc. Surface barrier detectors are actually based on the formation of Schottky barriers at metal/doped-semiconductor interface and various researchers have reported about this work [47-53]. As fabrication of these detectors, involves thermal evaporation of thin gold film (~ 20 nm) onto n-Si wafer and its adherence with silicon is poor, these detectors are highly vulnerable to damage and once damaged they automatically starts malfunctioning. In literature, there is mention about various thin film deposition techniques viz. electro- deposition, sputtering of gold etc. As requirement of wet surface is a pre-requisite for electro-deposition, it is not suitable for detector

fabrication. So fabrication of surface barrier detectors has been attempted by employing sputtering of gold on n-Si to develop prototype rugged detectors. The fabrication of these detectors can be briefly summarized as follows. Two n-Si wafers of 25 mm diameter, 600 μm thick, 1 kΩ-cm resistivity and <111> orientation were taken as starting materials. They were subsequently lapped on both sides by various grain size lapping powders (starting from 16 μm diameter to 5 μm diameter) till a smooth finished surface is obtained. Then these wafers were cleaned in deionized water ultrasonically and thoroughly dried under infrared lamp. Afterwards the wafers were etched in 6:1:1 volumetric composition of HNO_3, HF and CH_3COOH for about 5 minutes to obtain a mirror finished surface and fixed in ceramic mounts with the help of epoxy to passivate their edges. A thin gold layer of 20 nm was deposited onto the radiation-entry side by dc sputtering technique under an Ar pressure of ~5×10^{-2} torr. 200 nm thick Aluminum was thermally evaporated and deposited on the backside to form ohmic back contact of the device. Finally, the ceramic mount was hooked up in SS transmission assembly having the microdot connector. The diameters of the active area of the detectors were measured as 15 mm (active area ~ 177 mm^2).

3.2.2.2 Evaluation of results & Discussions: Typical reverse *I-V* characteristic of the fabricated silicon surface barrier detector is shown in Fig.6. It is seen from the figure that the leakage current remains lower than 5×10^{-6} A even up to an applied reverse bias of 100 V which indicates that the detector is good. In order to test the applicability of the fabricated surface barrier detectors for detection of charged particles, they were exposed to α-particles from a dual source (^{241}Am – ^{239}Pu). The typically recorded α-particle spectrum is shown in Fig.7. The two energies have clearly been resolved by the fabricated detector. The optimum energy resolution

(FWHM) of 70 keV was obtained for the detector when a reverse voltage of 400V was applied through a preamplifier of 110 MΩ load resistance and the measured leakage current was 3.34 μA. The measured energy resolution was slightly poorer as compared to our own detectors fabricated by vacuum thermal evaporation i.e. the energy resolution is compromised at the cost of better ruggedness. The higher energy of the gold atoms during sputtering could have caused little damage to the silicon surface which in turn made the resolution poorer. The energy of gold atoms can be fine tuned by varying the pressure of the Argon gas during sputtering. Making some more detectors by varying pressure of Argon, one can optimize the detector performance.

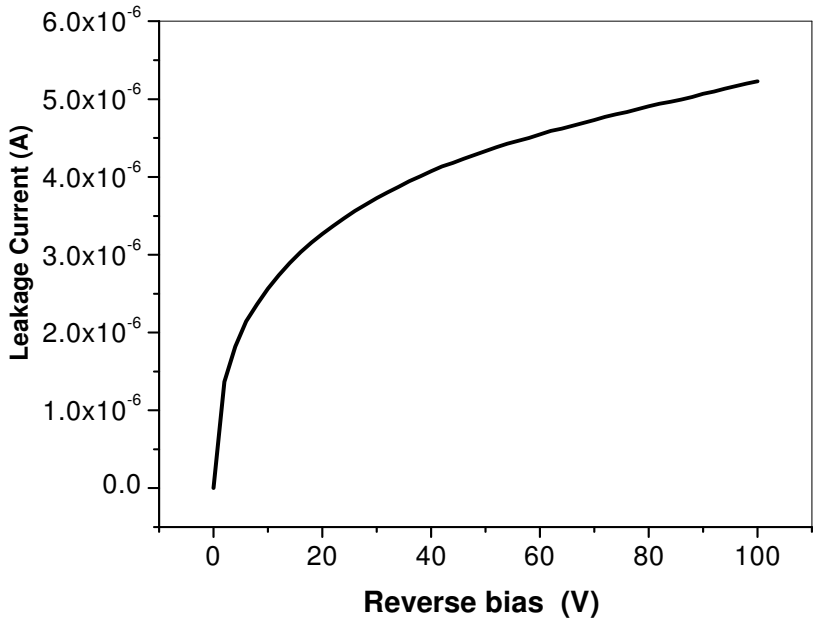

Fig.6. Reverse I-V characteristic of the gold sputtered surface barrier detector.

The adhesion of sputtered gold film on n-Si was evaluated by two methods: (a) At first the adhesion was tested as per ASTM standard by pasting special adhesive tape 3M Scotch on gold film and subsequent withdrawal of the tape witnessed no damage

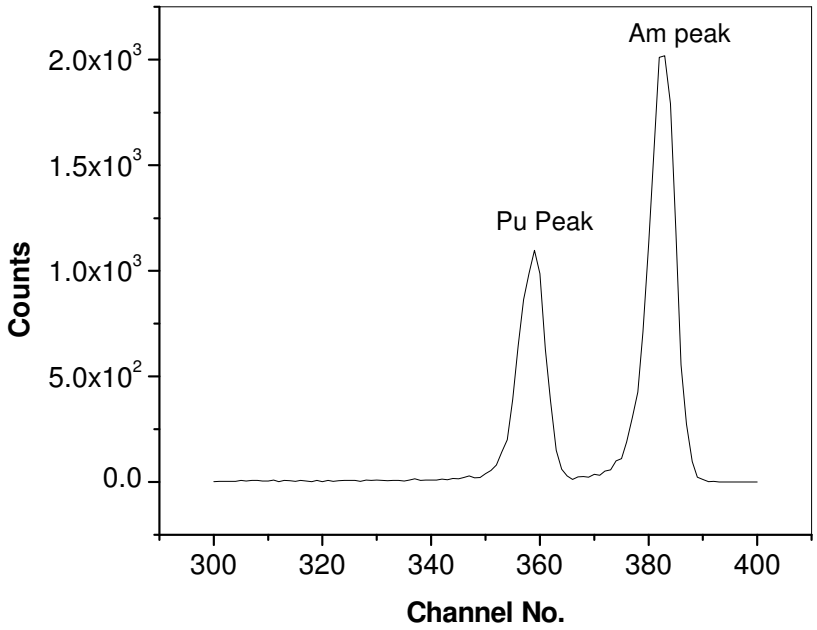

Fig.7. Alpha spectrum of the gold sputtered surface barrier detector.

to the film and (b) Secondly, the gold film was gently scrubbed using a tissue paper. It was observed that, unlike the thermally evaporated gold films, the sputtered gold films do not peel off and thus suggesting better adhesion. The higher kinetic energy of the gold atoms during deposition by sputtering may be the reason for this better adhesion. So the fabrication of rugged surface barrier detector has been demonstrated and the detector could resolve the energies of the α-particles from a dual source (^{241}Am – ^{239}Pu). This entire work has been presented and published in 31st IARP National Conference on Advances in Radiation Measurement Systems and Techniques [54].

3.2.3 Junction edge passivation of silicon surface barrier detectors by tin oxide layer formed as a result of thermal decomposition of Amine based organic films deposited by L-B technique

3.2.3.1 Experimental: To fabricate detectors using this passivation method, p-type silicon slices of 2 inches diameter, 1 mm thickness and 8 kΩ-cm resistivity were taken as starting materials. They were subsequently lapped with various grain size (starting with highest available grain size and completed with lowest available grain size) carborandom/alumina powders. Then thick layers (~ 500 nm) of gold were deposited on the backside of the lapped wafers using electro-less plating and the gold layers were masked properly by black-wax before proceeding for chemical polishing. Afterwards chemical polishing was carried out by dipping the mechanically-polished wafers (one at a time) into an etchant solution composed of 6:1:1 volumetric mixture of HNO_3:HF:CH_3COOH for 5 minutes. Removing the black wax from the back side resulted in front-side polished Si-slices with their backside covered with gold. The chemically polished front sides of the wafers were then partially covered by tape (as mask) of 38 mm diameter, covering the central part leaving the peripheral annular region (at the edge) open. Tape was also utilized to cover their entire backside gold layer. Multilayer L-B films of Octa Decyle Amine (ODA) – Stannate complex of 95 to 100 layers were deposited on the entire surfaces of both sides of these already masked silicon wafers. After deposition, the masking-tapes were removed and the covered areas under the masks were thoroughly cleaned with trichloroethylene (which actually degreased the Si-surfaces and also made it free from various other organic/inorganic contaminants). After removal of tapes, the deposited L-B film remained only at the annular area (at the edge) on the front side. In the next step the

Si-samples were kept inside a pre-cleaned quartz tube (one sample at a time) and placed the quartz tube loaded with the sample inside a furnace. Subsequently, the samples were heated with a ramping rate of ~ 5˚C/min to attain 600˚C (in air) in two hours and remain there for three hours (i.e. dwell time). This heating results in decomposition of multilayer L-B film to form high resistive SnO_2 film at the edges of the wafers. Growing SnO_2 film ($\rho \approx$ 10-20 MΩ-cm) on a Silicon wafer is a new as well as simpler technique [26 – 27] and has been explored to study passivation of Silicon detectors. Thermal evaporation of Aluminum was carried out to deposit Al-film of thickness 40 µg/cm^2, on to the central portion, at the front-side of the Si-wafer, covering partly the SnO_2 layer. Gold film of thickness 40 µg/cm^2 was evaporated on the rear side as an Ohmic contact.

3.2.3.2 Evaluation of results and Discussions: The fabricated detectors were evaluated with the help of their *I-V* characteristics measurement (both forward and reverse) and alpha spectroscopic study. The front and backside of one of the fabricated detectors prior to mounting and encapsulation are shown in Fig.8.

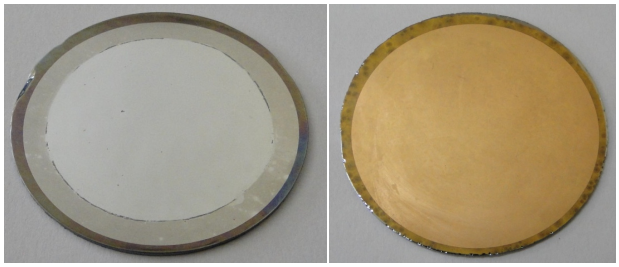

Fig.8. Photographs of front and backside of an un-mounted detector.

Forward and reverse *I-V* characteristics of the best performing detector are shown in Fig.9 and 10 respectively. Larger active area of the detector (diameter of the active area ~ 38 mm) gives rise to larger value of leakage current. Both “As-prepared” and

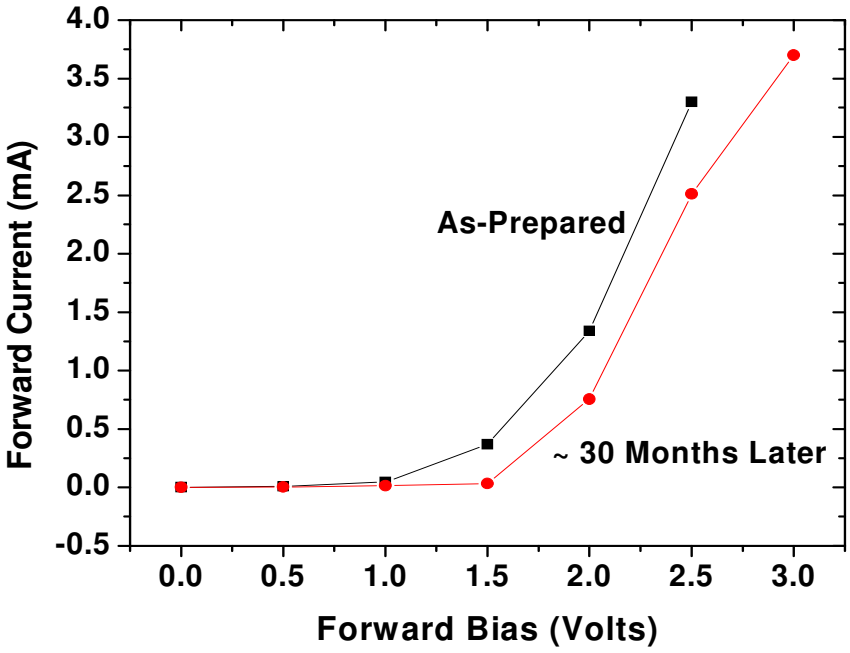

Fig.9. Forward characteristics of the detector.

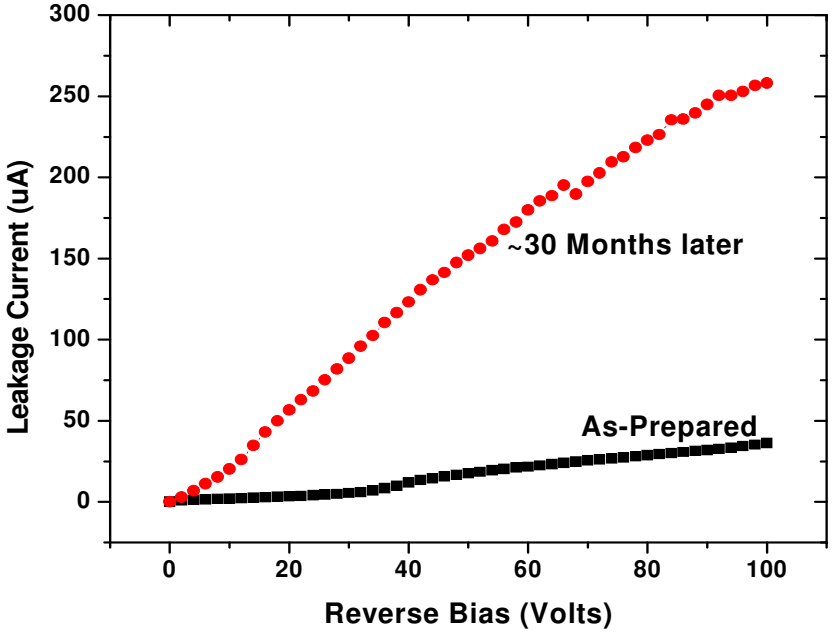

Fig.10. Reverse characteristics of the detector.

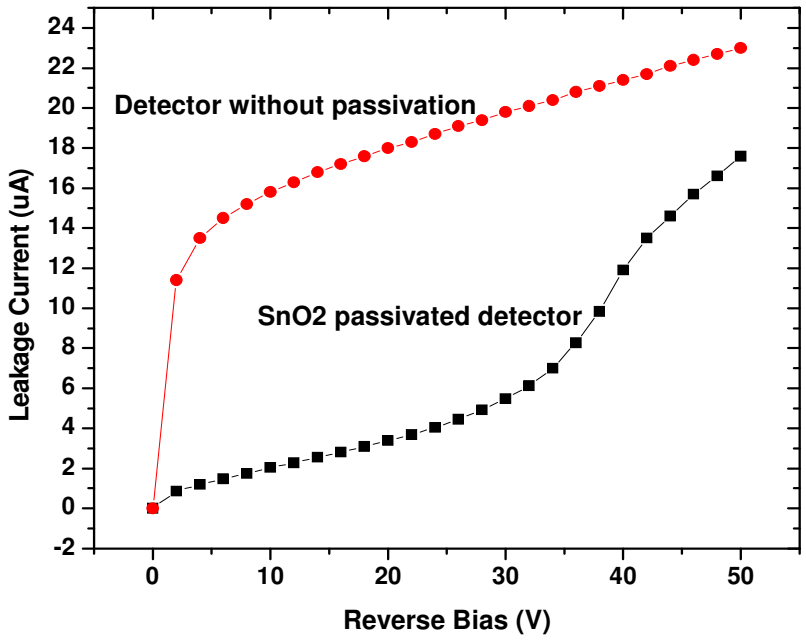

Fig.11. Comparative reverse *I-V* characteristics of detector with/without passivation.

"30 Months later" characteristics were measured and incorporated in forward as well as reverse characteristics in order to have an idea about long term performance. The effectiveness of the tin oxide passivating layer was verified by comparing the reverse I-V characteristics of the detector having tin oxide passivating layer with the reverse characteristics of another detector having no passivating layer [55]. This comparison is shown in Fig.11 which indicates that the SnO_2 passivation layer helps in reducing the leakage current. The alpha spectrum of the detector, obtained using ^{241}Am-^{239}Pu alpha source, by keeping the source and the detector 5 cm apart and under rotary vacuum (~10^{-3} Torr), is shown in Fig.12.

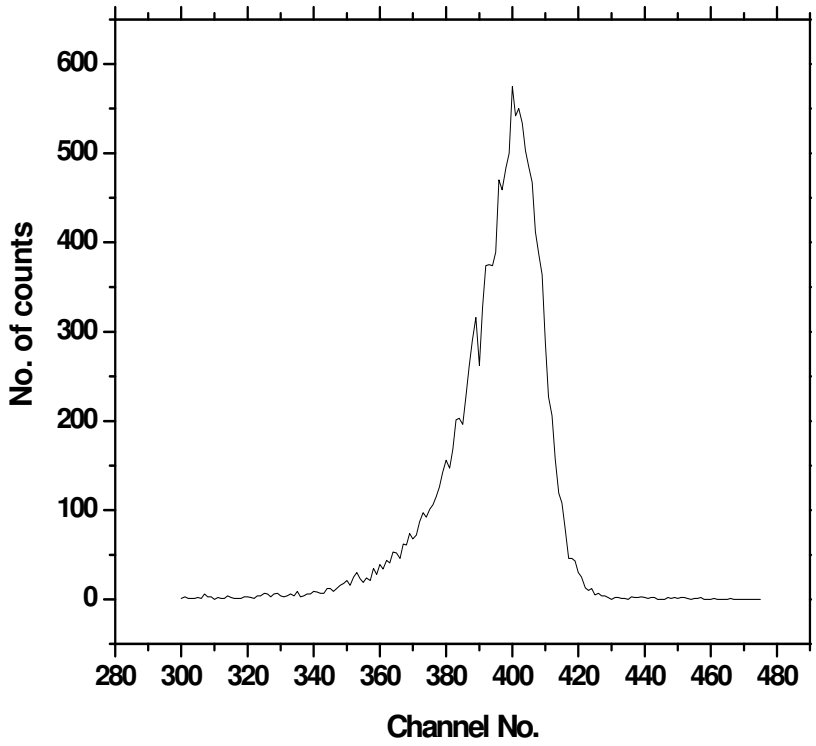

Fig.12. Alpha spectrum of the detector.

As the active area of the detector is very large, its capacitance is also very high. Higher the value of capacitance, higher will be the noise generation in the detector. This in turn affected the energy resolution thereby not resolving Am and Pu-peaks. But the detector can still work as a solid state counter. In order to have a better idea about the surface morphology of the newly grown tin oxide layer, SEM images of its surface were taken for two different scales and are shown in Fig.13. The SEM image corresponding to 100 nm scale is presented in the inset and the main image was acquired by keeping the scale set at 200 nm. SEM Micrograph reveals close packed distribution of SnO_2 nano-crystals of size about 50 nm.

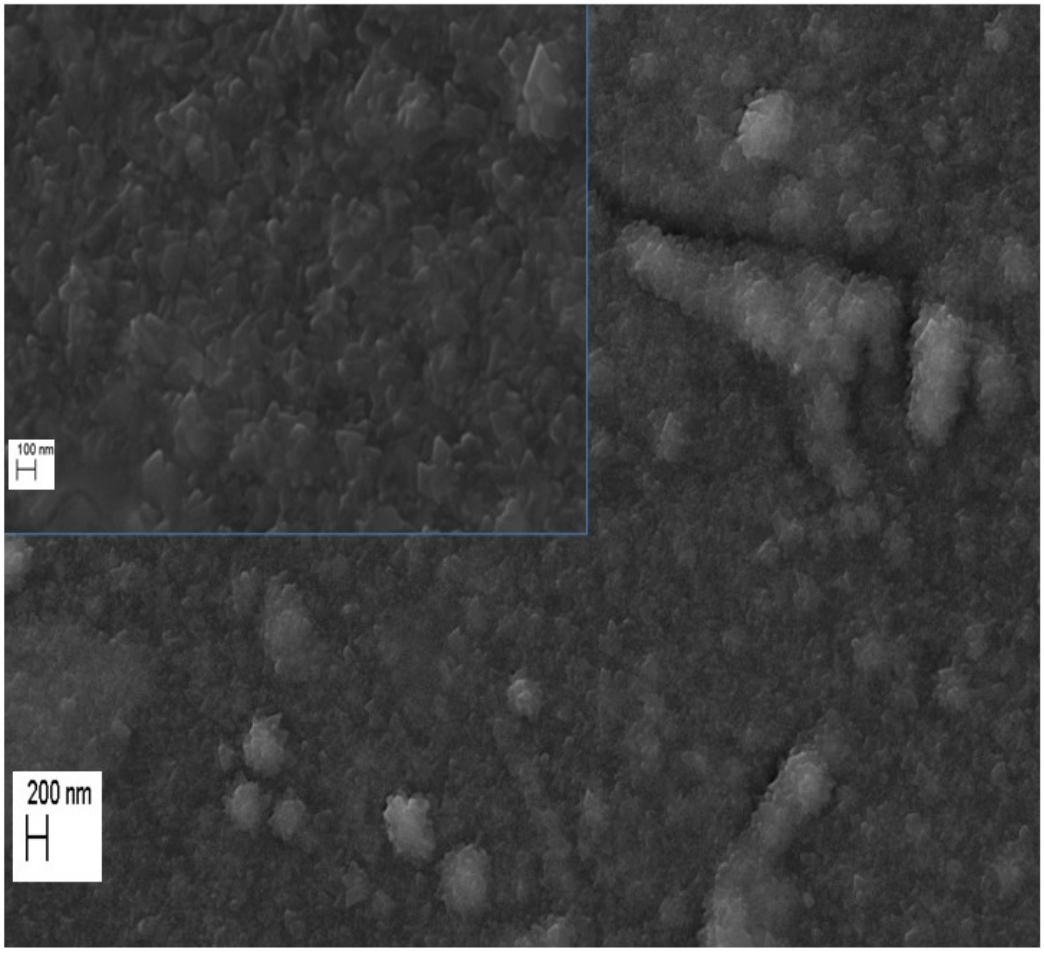

Fig.13. SEM Image of SnO_2 surface.

The EDS spectrum is measured in three different locations on the surface of the SnO_2 layer and most detailed one is presented in Fig.14. EDS spectrum shows predominantly the peaks corresponding to Sn and O in the newly formed SnO_2 layer. Also there is feeble presence of zinc as metallic impurity which might be responsible for higher reverse leakage current.

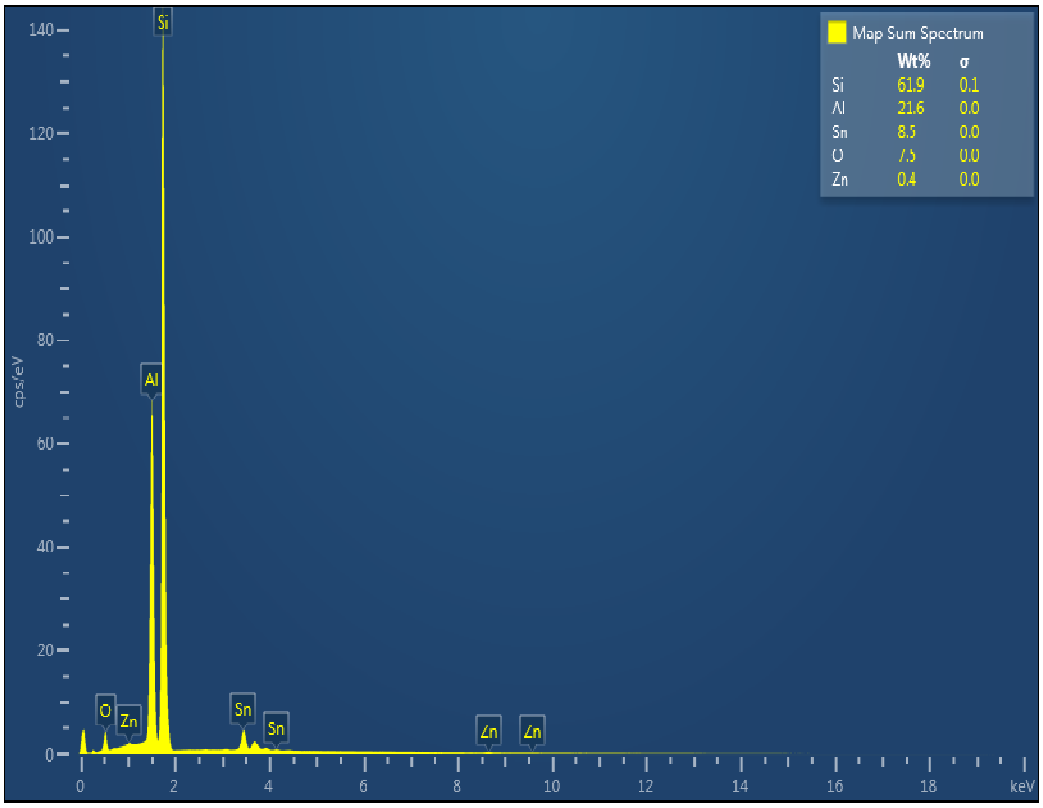

Fig.14. EDS spectra of the SnO_2 thin film prepared by LB method.

This entire work has been reported for publication in the journal "Bulletin of Material Science" (Springer Publications), which subsequently got accepted.

3.2.4 Junction edge passivation of Silicon surface barrier detectors by hydrogenated amorphous silicon layer

It is reported in the literature that Hydrogenated amorphous silicon (a-Si:H), can be used as an excellent passivant for crystalline silicon (c-Si) p-n junctions [28]. Researchers have reported a two orders of magnitude reduction in reverse leakage current from that of a typical thermal oxide passivated junction. This material has been attempted to passivate the edge of silicon surface to fabricate silicon surface barrier detector. Two n-type silicon wafers of 1 inch diameter, 4.5 kΩ-cm resistivity

and 1 mm thickness were taken as starting material. The front side (top view) of the polished, edge-passivated and mounted silicon wafers, prior to taking gold evaporation on it, is shown in Fig.15. Two surface barrier detectors were fabricated with their edges passivated by hydrogenated amorphous silicon and tested subsequently for *I-V* Characteristics. But their *I-V* didn't show diode behavior as there was hardly any difference between forward and reverse currents. Optimization in the process to make hydrogenated amorphous silicon layer need to be done to get proper results.

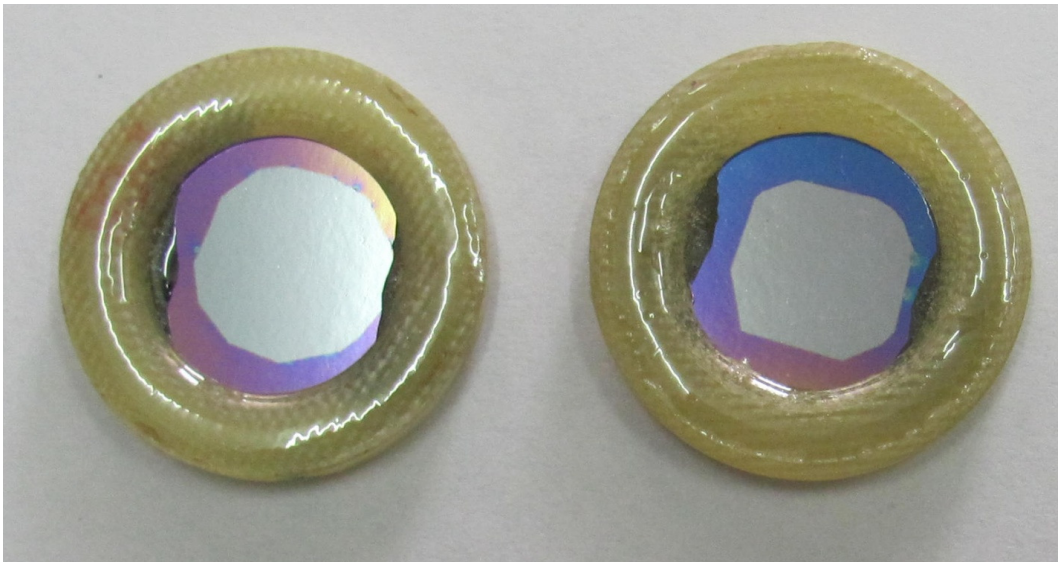

Fig.15. Photograph of the front side (Top view) of **a-Si:H** passivated silicon surface.

3.3 CONCLUSION

This chapter describes the fabrication, characterization of SSB detectors for alpha particle detection using two different edge passivations; (a) thermally grown SiO_2 and (b) SnO_2 thin films prepared by LB method. Edge passivation by SiO_2 is reliable in terms of leakage current and energy resolution for thicker silicon substrates, however,

for thin silicon detectors chances of breaking of the substrate is higher. Passivation by SnO_2 thin films is perhaps more appropriate for thinner silicon substrates, however, further studies are required to improve the energy resolution by using smaller active area of the detectors as in the case of detectors with SiO_2 edge passivation. The ruggedness of the gold contact of Au/n-Si junction has been studied by depositing gold contacts using sputtering. It was observed that sputtered gold films gave better ruggedness for the SSB detectors.

Chapter 4

DEVELOPMENT OF IMPROVED SURFACE BARRIER DETECTORS BY EMPLOYING ARGON COLD PLASMA ASSISTED SILICON SURFACE CLEANING

4.1 INTRODUCTION

Silicon is an important material whose properties have been exhaustively studied [56]. It continues to remain a material of choice for large scale use in device fabrication, which includes integrated circuits, micro-electro-mechanical systems (MEMS), solar cells, nuclear radiation detectors etc. The silicon surface plays an important as well as a crucial role in many process steps used to fabricate these devices, especially silicon surface barrier detectors. The silicon surface needs to be exhaustively cleaned prior to fabrication of these detectors. The conventional way of cleaning silicon surface by using specific organic and inorganic solvents [57-58] is time consuming and tedious, while the solvents used are toxic and are also responsible for environmental pollution. Besides, for solar cells, produced from single crystal, polycrystalline, or amorphous silicon, the respective surface must be carefully cleaned either before an oxide mask layer is allowed to grow, or prior to deposition of an anti-reflection coating [59]. The usage of atmospheric pressure oxygen plasma provides a convenient, in-line treatment process for cleaning as well as activating silicon surfaces [59-61]. Though surface modification of silicon using plasma has been reported by many researchers, no information is available in the literature about how this surface cleaning/modification can influence the performance of silicon based nuclear radiation detectors. It is observed in semiconductor industry that the properties of surface/interface of silicon based layers play a key role in fabrication of devices. Different applications require different properties of such surface/interface layers. It is also mentioned in literature that the modification of surface properties of silicon based layers involves either toxic processes or is highly energy consuming [62-64]. However the use of plasma technology provides an environmentally acceptable and cheaper

alternative. Though the low pressure plasma technology is highly efficient and widely used in the industry, efforts are on to replace it by atmospheric pressure plasma [62]. Plasma activation of silicon surface for self-assembly, in an attempt to optimize and simplify the formation of hydrophilic silicon surface, is a relatively safer and simpler route which replaces the conventional means to form hydrophilic silicon surface with the help of corrosive and harmful piranha solution [65]. Researchers have also separately studied plasma oxidation of silicon exhaustively [66-67]. However, very little information exists in the literature on the possibility of technical use of cold plasma for nuclear detector fabrications. The cold plasma facility used in the present study is a compact and low cost table-top unit and does not require any elaborate laboratory arrangements. The design, fabrication, and usage details of this low cost, table-top equipment have been published recently [68]. Moreover, by employing this equipment, usage of solvents could be avoided. So this method of cleaning silicon surface has simplified the fabrication of surface barrier detectors, over and above the inherent benefit of choosing simple surface barrier fabrication technique as compared to elaborate arrangements of ion-implantation/diffusion techniques.

4.2 EXPERIMENTAL

Fabrication of surface barrier detectors using silicon slices cleaned by cold plasma was carried out as follows:

At first, single crystal silicon slices (n-type, ~14.9 kΩ-cm resistivity, 34.3 mm diameter and 1 mm thickness) were cut from ingot with the help of diamond wheel in a wafer cutting machine. Then these slices were mechanically lapped using carborandum/alumina powders and then chemically polished on both sides in 6:1:1

volumetric composition of HNO_3:HF:CH_3COOH. Before carrying out cold plasma assisted cleaning of the etched silicon surface, the etched wafers were cleaned by dipping them in de-ionized water in a beaker and vibrating the beaker ultrasonically. Subsequently the slices were dried by keeping them under an infrared lamp. Now the slices are ready for exposure to cold plasma. Four wafers were cleaned using cold plasma under four different conditions. First wafer was exposed to the cold plasma formed by Argon gas (Argon gas purity 99.999% and flow rate maintained at 2.8 lit/min) for 3 minutes, while the second wafer was exposed to cold plasma formed by a mixture of Argon and Ambient Laboratory Air (90% Argon and 10% Air, Argon flow rate was 2.8 lit/min and Air flow rate was ~300 ml/min), also for 3 minutes. The third wafer was first exposed for 3 minutes to cold plasma of Argon and Air (90% Argon and 10% Air, Argon flow rate was 2.8 lit/min and Air flow rate was ~300 ml/min) similar to the second wafer, followed by an additional exposure for 2 minutes to cold plasma formed by a gas mixture of Ar and moist Air [Air was passed through de-ionized water at room temperature (300 K) of 18 MΩ-cm resistivity using a bubbler] (90 % Ar and 10% moist Air). Hence total time of exposure of this wafer to plasma is 5 minutes. The fourth wafer was exposed to cold plasma of Ar and moist Air (90% Ar and 10% moist Air) for 3 minutes. Hereafter the detectors fabricated out of first, second, third and fourth wafers will be termed as D1, D2, D3 and D4 respectively. The plasma coming out of the flexible brush had a spot diameter of approximately 3 mm [68]. It was fixed at a distance of 5mm over the wafer surface in a Z-axis clamp, and the silicon wafer was made to glide underneath this tip in X-Y directions by hand, mimicking a 2D plotter movement (or raster scanning), so that the

entire silicon surface could be scanned uniformly by the plasma tip within the preset time duration. The four plasma exposure conditions are given in Table-I below:

TABLE - I

Detector	Plasma gas composition Ar flow rate 2.8 lpm, Air flow rate 300mlpm	Duration of exposure (min)	Optimum energy resolution (keV)
D1	Argon	3	α-peaks are not well resolved
D2	90% Ar + 10% Air	3	53
D3	90% Ar + 10% Air and followed by 90% Ar + 10% moist Air	3 2 (total 5)	97.77
D4	90% Ar + 10% moist Air	3	79.41

(This table is taken from our published paper: A. Ray *et al* 2018 *JINST* **13** P09019)

Then the samples were mounted on ceramic mount using organic epoxy [a mixture of one drop epoxylite hardener No. 323 (obtained from M/s Epoxylite corporation, U.S.A.) and five drops of vacuum pumped CIBA 230 Araldite epoxy resin (Make: Hindustan CIBA-GEIGY Ltd. under license from CIBA-GEIGY AG, Basel, Switzerland)]. Au of thickness ~ 40 μg/cm^2 was deposited on radiation-entry side of silicon wafer by thermal evaporation under high vacuum (~ 10^{-6} Torr). Ohmic Al contacts of thickness ~ 40 μg/cm^2 were also deposited on backside of the wafers. The depositions were carried out at a substrate temperature of 300 K. In case of conventional Au/n-Si surface barrier detectors, prior to depositing Au of 20 nm thickness (~ 40 μg/cm^2) on to the radiation-entry side of silicon wafer, the silicon surface is cleaned by organic solvents and not by cold plasma [30]. So actually it is

the silicon surface cleaning process which differs. The evaluation of the fabricated detectors was carried out by measuring current-voltage (*I-V*) characteristics (both forward as well as reverse characteristics), measurement of detector response to α-particles (i.e. measuring optimum alpha energy resolution) and also measuring the variation of alpha energy resolution with reverse bias voltage. Measurement of *I-V* characteristics under forward as well as reverse bias were carried out using Keithley make programmable electrometer. Alpha spectra for the detectors were obtained using ^{241}Am – ^{239}Pu dual α-particle source and PC based multi-channel analyzer. The energy resolutions of the detectors were measured as a function of reverse bias voltage (n-silicon positive with respect to Au). For alpha particle spectroscopic measurements, the source and the detector were placed in a vacuum chamber (at ~ 10^{-3} Torr) with a separation of the order of the diameter of the detector.

4.3 RESULTS AND DISCUSSIONS

The schematic of the Au/n-Si surface barrier detector is shown in Fig.1 and the photograph of the cold plasma facility which was utilized to clean silicon samples' surfaces is given in Fig.2. The forward *I-V* curves of the four detectors are shown in Fig.3 and the reverse *I-V* curves are shown in Fig.4. From the forward and reverse characteristics we can say that all the detectors are showing very good diode behaviour. The reverse leakage current obtained for D2 is lowest among all the four detectors and it is also lower than that for best conventional Silicon Surface Barrier (SSB) detector prepared in our laboratory previously [30]. The forward and reverse *I-V* curves of D2 and the best SSB detector are shown in Fig.5 and Fig.6 respectively purely for comparison purpose. The presence of reactive free radicals, *e.g.* H-atom,

$^{\bullet}OH$ radical and $HO_2^{\bullet}$ radical (usually referred to as reactive oxygen species, or ROS) and reactive nitrogen oxides, $NO^{\bullet}$ and $NO_2^{\bullet}$ (generally referred to as reactive nitrogen

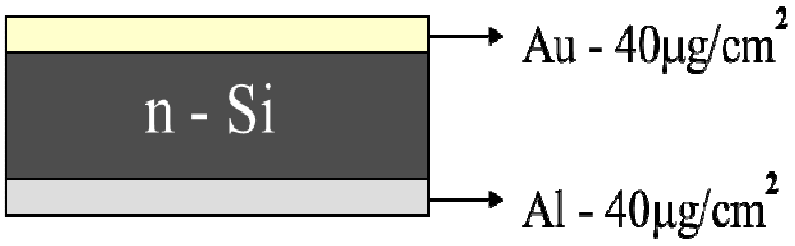

Fig.1. Schematic of Au/n-Si un-mounted surface barrier detector.

Fig.2. Cold Plasma Flexible Brush plume of Ar gas ~10 mm over aqueous sample in a beaker, with 12.8 kV_{p-p} applied voltage at Ar flow rate 10 ml $.s^{-1}$ [68].

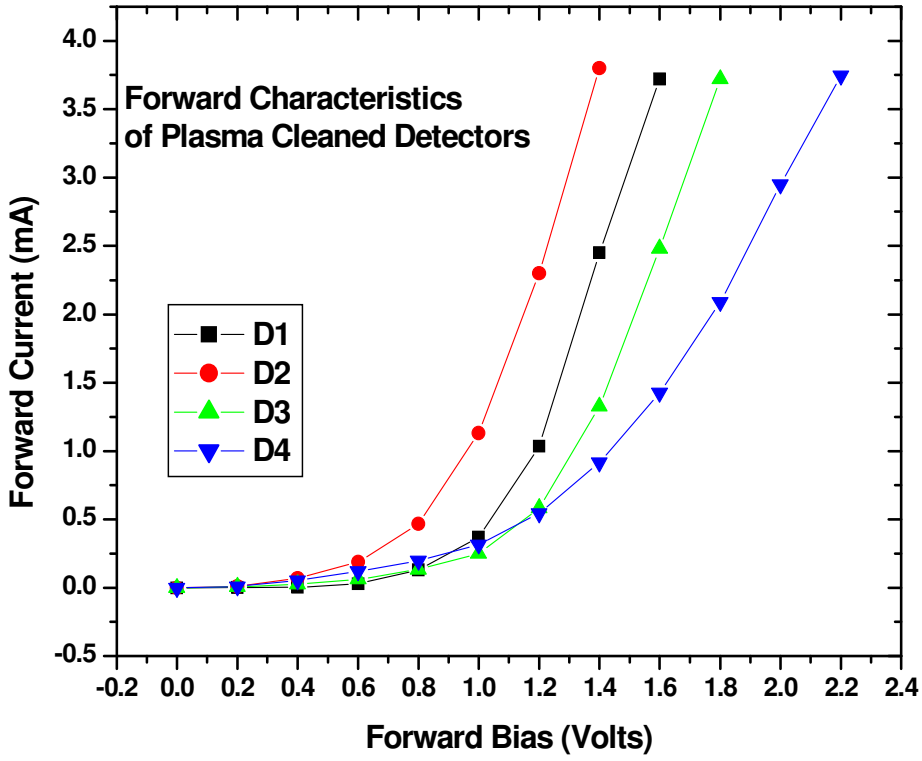

Fig.3. Forward Characteristics of the four detectors D1, D2, D3 and D4.

Species, or RNS) in the plasma as reported in details in the literature [68], is responsible for such behavior, as it quickly reacts with all sorts of organic surface materials and oxidize them into carbon dioxide and water [68]. When plasma is prepared from the gas mixture of Argon and moist air (as in the cases of D4 and D3), the small amount of water molecules present in the moist air will react with $NO^{\bullet}$ and $NO_2^{\bullet}$ and form HNO3 in presence of O_2. HNO_3 is a strong acid as well as oxidizing agent. Thus, it is expected to modify and etch the Si surface to some extent. As a result of this, the leakage current of D4 is slightly higher than D2. Between D4 and D3, D3 showed more leakage current. As a qualitative observation, we can say that prior to fabricating D3, the Si- surface was under cold plasma for a longer time (5

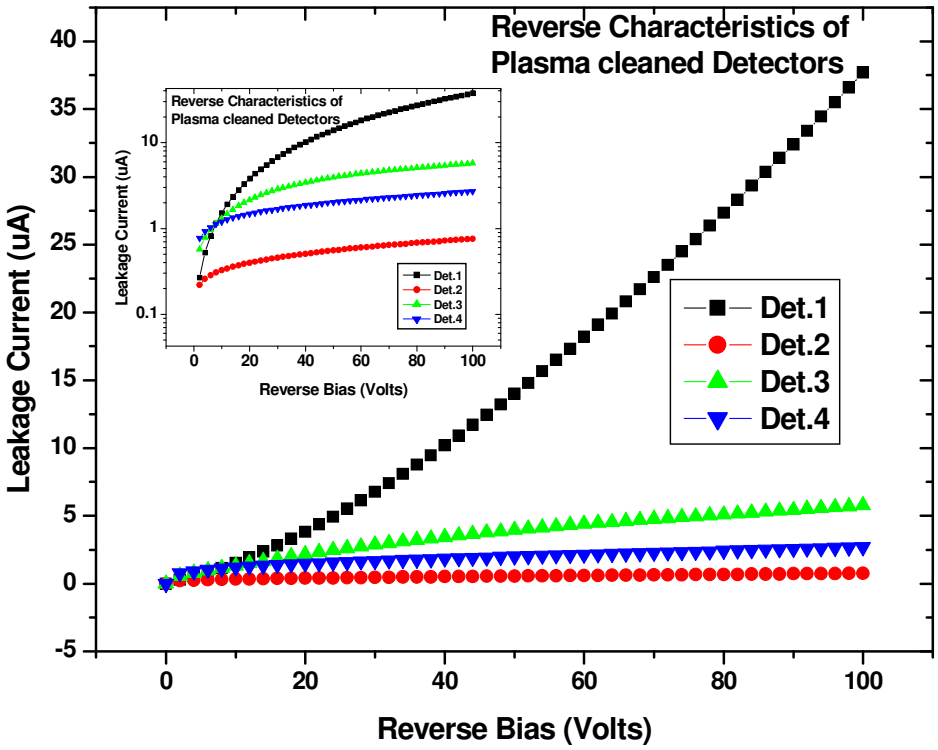

Fig.4. Reverse Characteristics of the four detectors D1, D2, D3 and D4.

minutes) than that for D4 (3 minutes). From the reverse characteristics, it is observed that the leakage behavior of D2, D3 and D4 (Argon + Air cases) are closely spaced lines which are wide apart from D1 (only Argon case). The reason for the higher leakage current of D1 can be due to the presence of energetic ions such as $Ar_2^{\bullet+}$, $Ar_3^{\bullet+}$ etc. in the plasma (when only argon cold plasma is used) which due to their heavy mass as compared to Si, possibly damage the latter surface irreversibly [1]. But in the cases of D2, D3 and D4, even though Argon is present in the plasma, due to the presence of air/moist air, the possibility of creation of $Ar_2^{\bullet+}$, $Ar_3^{\bullet+}$ ions becomes very feeble.

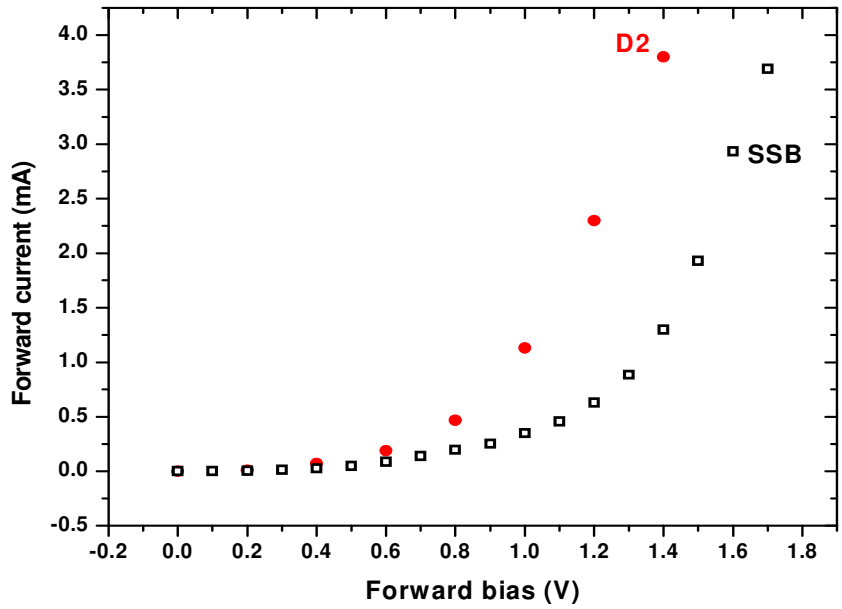

Fig.5. Comparison of forward characteristics of D2 with SSB detector.

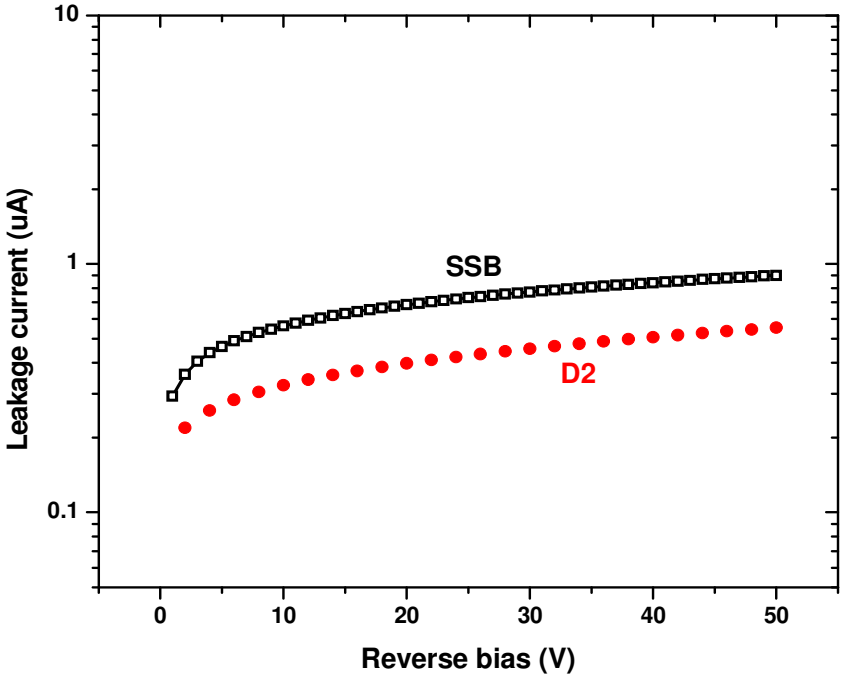

Fig.6. Comparison of reverse characteristics of D2 with SSB detector.

For charge particle detection, these detectors were exposed to α-particles from dual source (^{241}Am–^{239}Pu), irradiating particles with energies of 5.48 and 5.15 MeV. The detectors used for this measurement had an active area of ~250 mm^2. During measurements, both detector and source were placed in vacuum. Because of the higher leakage current (as discussed above), D1 offered very poor resolution and it is almost of no practical use as far as spectroscopic measurements are concerned. The best alpha spectra obtained from D2, D3, D4 and the best prepared SSB in our laboratory are shown in Fig.7, 8, 9 and 10 respectively. The variation of energy resolution for the three detectors i.e. D2, D3 and D4 with reverse bias voltage i.e. alpha energy resolution as a function of reverse bias voltage is plotted in Fig.11. In this figure, the voltage shown is the total applied voltage to the device in series with a load resistance of 110 MΩ.

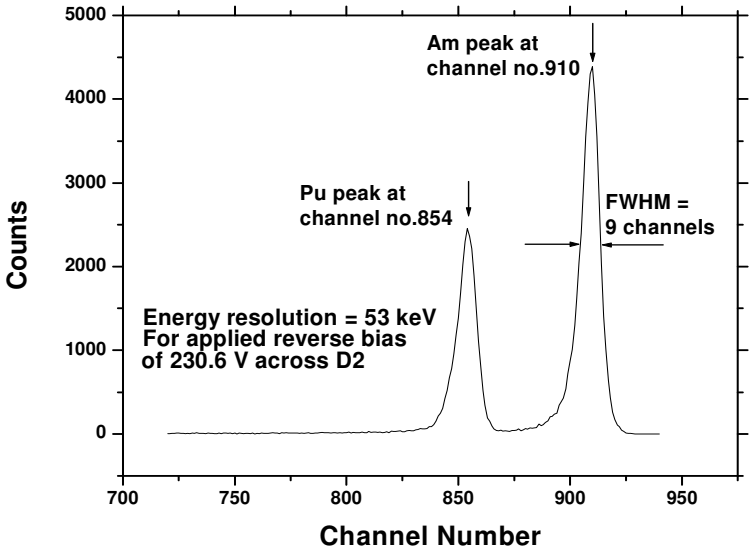

Fig.7. Best alpha spectrum obtained from D2 (energy resolution of 53 keV is obtained corresponding to Am-peak of energy 5.48 MeV).

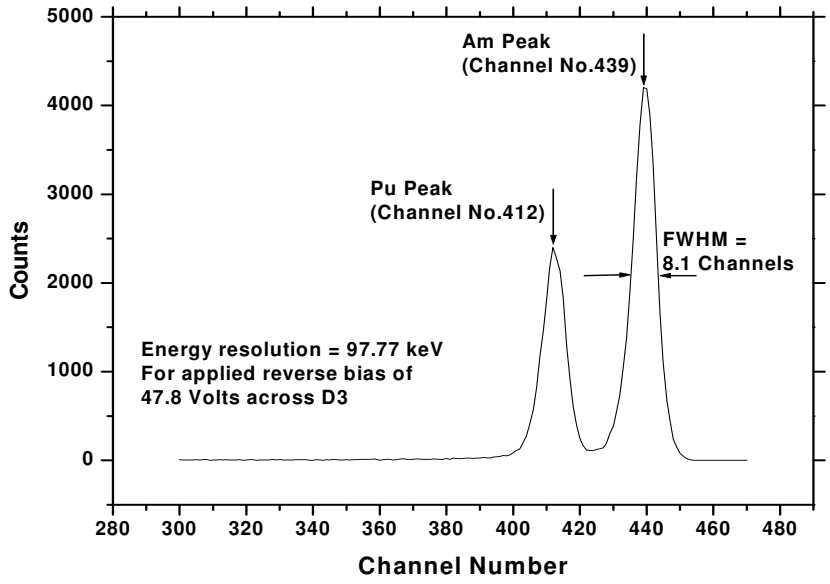

Fig.8. Best alpha spectrum obtained from D3 (energy resolution of 97.77 keV is obtained corresponding to Am-peak of energy 5.48 MeV).

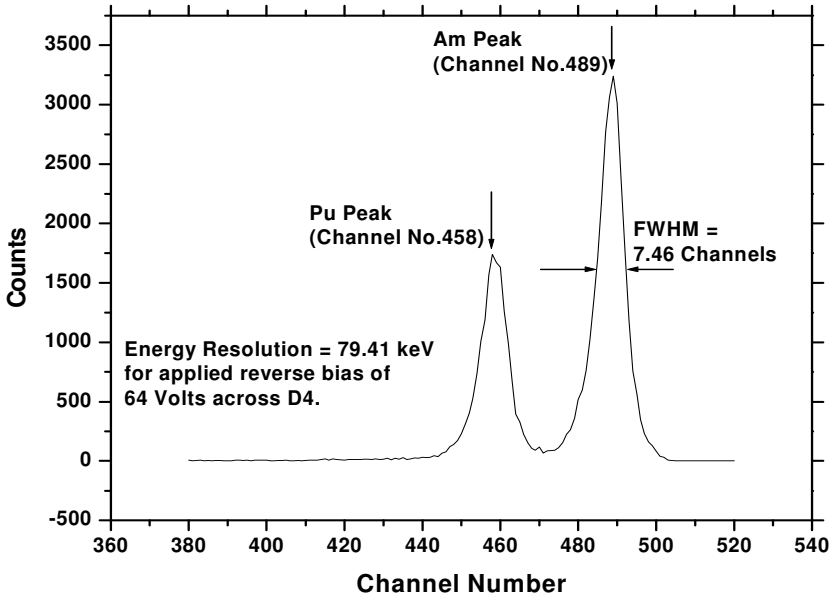

Fig.9. Best alpha spectrum obtained from D4 (energy resolution of 79.41 keV is obtained corresponding to Am-peak of energy 5.48 MeV).

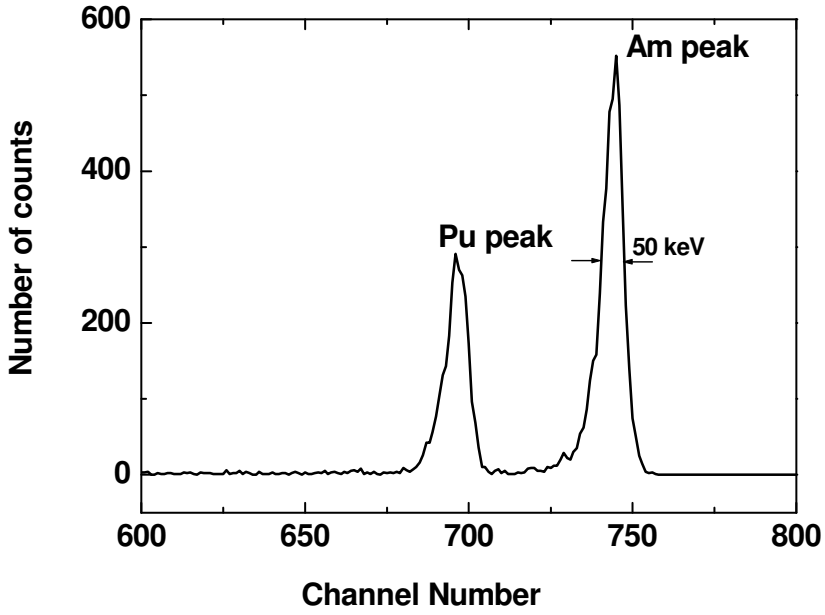

Fig.10. Best alpha spectrum obtained from SSB (Energy Resolution=50 keV).

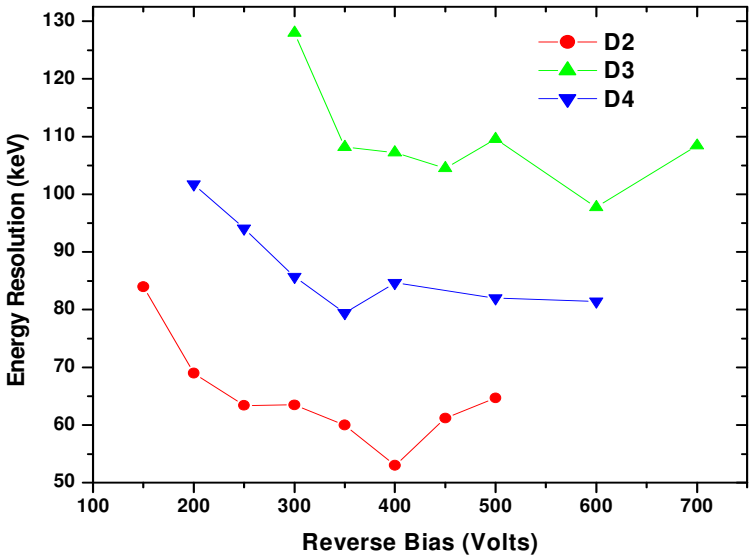

Fig.11. Variation of energy resolution with reverse bias voltage (as the leakage current through D1 is quite high, it offered very poor resolution and hence omitted in this figure).

This entire work is published in Journal of Instrumentation (JINST), IOP Publishing (U.K.) **[69].**

4.4 CONCLUSION

Environment friendly and simple method of cleaning for silicon wafers using cold plasma in place of conventional chemical cleaning has been tried. Four types of cold plasma compositions (Argon, air, moisture) and plasma exposure durations have been tried to evaluate the dependence of leakage current and energy resolution of the detectors on plasma cleaning. From the study we can conclude that cleaning with only Argon cold plasma is not very effective in terms of leakage current and energy resolution. However, the leakage current of D2 is lowest among all the four detectors fabricated and also lower compared to best conventional SSB detector fabricated in our laboratory earlier. In the present study we have optimized the gas composition of the cold plasma for best detector performance amongst "pure Argon (0% humidity), Argon with pure Air (trace humidity) and Argon with moist Air (~10% humidity)". In order to get a qualitative idea, D3 was fabricated using longer exposure time. In this case, the mixed exposure for D3 was conducted to check the effect of a sequential protocol. Since the plasma plume formed out of pure Argon is exposed to open air, there is a possibility that a trace amount of air and/or moisture may get added with that Argon and their effects will be unavoidable. However continuous gas flow in all the cases ensured that such effects remains minimal. Lastly, conducting experiment using a single detector, fabricated out of each plasma condition, has produced results which

are indicative of the effect and more number of detectors from each category i.e. more statistics are needed to make the results more conclusive.

Chapter 5

PURELY ORGANIC RADIATION SENSORS

5.1 INTRODUCTION

Detection of alpha particles, heavy ions etc. is an essential feature in various nuclear physics experiments. For this purpose, solid state detectors and especially silicon detectors are playing a leading role. Let us now switch our focus for the time being from Nuclear Physics to Health Physics. We all know that the naturally occurring radioactive materials (e.g. uranium, thorium and radon) and artificial radioactive elements (e.g. plutonium and americium) are the emitters of alpha particles and therefore some of them are very much in use to prepare alpha emitting sources. These alpha particles may cause threat in case of being inhaled or ingested by someone or even more dangerous in case they get absorbed in someone's blood. In all such cases, it may cause damage to body tissues and hence enhances the risk of being affected by cancer. To be more specific, firstly the uranium mining waste and secondly the concentrated natural radium generated while processing phosphate ore to produce fertilizer, are the two major alpha emitters through which mankind is directly getting exposed to alpha. So in order to prevent the environment from getting contaminated, it is very much essential to detect the alpha particles in and around such industries. To suit this stringent requirement, one must look for a sizable number of flexible as well as portable alpha sensors. In this context, perhaps the development of organic polymer based detectors would be best suited ones because of its capability to be flexible, as well as portable. Also they can be prepared in large scale and that too at a very low cost. There are certain properties of organic semiconductors which look alike to those of inorganic semiconductors, like Silicon and Germanium and for this very reason, one can think of treating them as a substitute for Silicon and Germanium in many devices. Researchers are taking interest in this new as well as challenging

field primarily due to the relative simplicity of the technological implementation promising substantial economical benefits and also side by side opening the scope for getting new semiconductor products e.g. Organic FET, Organic LED, Organic Photovoltaic devices etc. Owing to the differences in physical and chemical properties, organic semiconductors could serve as a starting material for various new and improved sensors and detectors [34]. Semiconducting organic material may be subdivided into three categories viz. polymers, oligomers and small molecules depending on the length of the molecular structure that sets mechanical, chemical and electronic properties [70].

This chapter deals with the interaction of alpha particles with polymer material (BoPET). The outcomes of such interactions were analyzed in detail as these outcomes can be considered as the stepping stones towards the development of polymer based detectors. Crystalline organic substance and non-crystalline materials, e.g., plastics, suffer radiation effects through other mechanisms, chiefly the breaking of chemical (covalent) bonds. The threshold energy for such breakage is also of the order of 25 eV [71]. As exposure to ionizing radiation will break many bonds in polymer material (also called radiation damage) it will cause generation of charge carriers (e-h pairs). So by irradiating with ionizing radiations, it is expected that the conductivities in polymer materials [e.g. Polyanilene, Biaxially Oriented Poly Ethylene Terephthalate (BOPET) etc.] will be changed. This change in conductivity was studied for alpha particles to carry out their detection. The results obtained for the change in conductivity of BoPET film, due to its exposure to alpha particles, are being reported in the subsequent section.

5.2 EXPERIMENTAL

BoPET film was developed in the mid-1950s, originally by DuPont, Imperial Chemical Industries (ICI) and Hoechst. It is also called Bi-axially Oriented PET where PET is called Poly Ethylene Terephthalate. The chemical structure of PET is shown in Fig.1. A number of samples were made using thin sheet of BoPET and

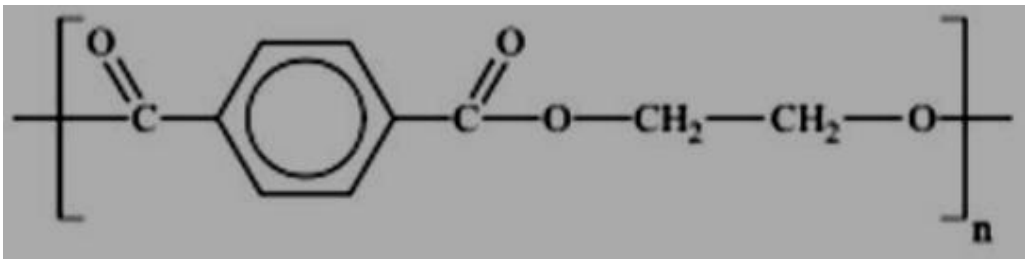

Fig.1. Chemical structure of PET.

studied their current conducting behavior before and during exposure of the samples to alpha particles. On each of these samples (i.e. on each small piece of thin BoPET sheet of size 6mm × 2mm (approx.) which was mounted on a glass slide with the help of epoxy) there are two closely separated (12 μm) and 30 nm thick gold pads (deposited by physical vapour deposition). Electrical connection to the sample was established with the help of two single strand wires, held by silver paste on the two gold pads deposited on BoPET samples. In order to see the current vs. time behavior (i.e. *I-t* characteristics), a fixed voltage in the range of 40V to 90V was applied between the two pads and the current was measured over a period of 200-600 seconds. Afterwards, an increase in this current was observed for the same applied bias across the pads of the sample when it was exposed to 5 MeV alpha particles from ^{241}Am –

^{239}Pu source. Identical measurements were carried out with the help of samples prepared out of BoPET sheet coated with gold nanoparticles. But no change in the device current was witnessed before and during exposure of 5 MeV alpha particles. Also some samples, comprising of larger separation (~ 330 μm) between the gold pads on a bigger size (15 mm × 6 mm) of BoPET sheet, were prepared and similarly tested. Extremely low value of current was observed and there was no change in the value of current before and during exposure of 5 MeV alpha particles. The reason could be: the charge carriers generated by the applied voltage and/or by radiation may get recombined before reaching the respective contacts. An experiment was conducted by exposing two BoPET samples (Size: 6 mm × 2 mm, Separation between two gold pad: 12 μm) periodically to an alpha source of higher activity (0.5 mCi), by placing the source on the sample for some time, then withdrawing the source for some duration, again placing the source on the sample for some time and so on. It was found that the current also got increased and decreased periodically in the same fashion as the source was placed and withdrawn. Keeping view of the results obtained from two samples, the better result corresponding to a particular sample (Sample I), is presented in Fig.2 which shows the variation of current through the BOPET sample with time when it was periodically exposed to 5 MeV alpha in air from a source of activity 0.5 mCi. An increase in current of ~ 60 pA during alpha exposure as compared to its value before exposure was observed. Typical values of current observed were 320 pA and 380 pA with respect to 'before exposure' and 'during exposure' cases respectively for an applied bias of 40 V. Subsequently, for testing the detector performance under vacuum conditions (2nd experiment), one vacuum measurement set-up was developed where an alpha source and another BoPET sample (Sample II) [size: 6 mm × 2 mm,

Separation between two gold pad: 12 μm] can be placed inside a vacuum chamber and the chamber can be kept under rotary vacuum (~ 10^{-3} Torr). At first the device current was measured in air for an applied bias of 90 V across the sample without exposing it to alpha particles (by placing the sample inside the chamber which was kept at atmospheric pressure by not doing any evacuation.

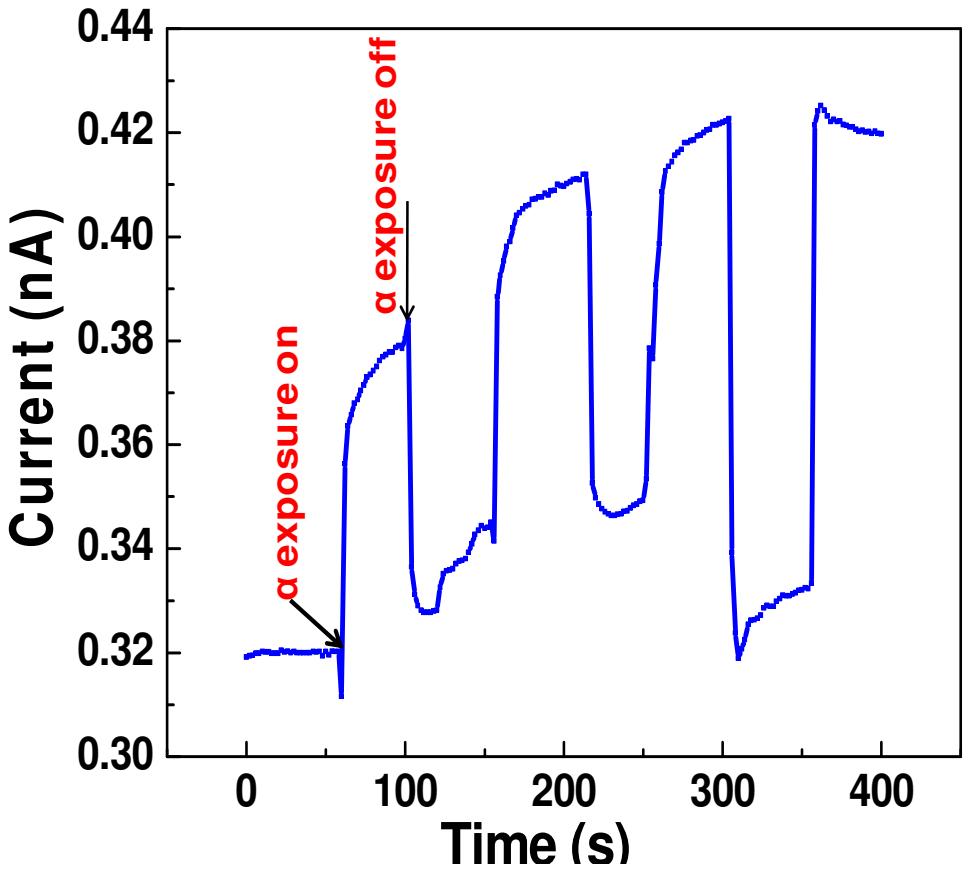

Fig.2. BOPET sample was periodically exposed to 5 MeV alpha in air from a source of activity 0.5 mCi and the device current vs. time was recorded for an applied fixed bias of 40V across the sample (Sample I).

Fig.3 shows the variation of device current, which is plotted against time-elapsed to obtain the steady-state value of the device current for a particular value of the bias voltage (90 V). Then the sample and the alpha source were kept under vacuum and a clear jump of device current was observed (from 3.44 pA to 25.1 pA by using an alpha source of lower activity ~ micro-Curie (12108.19±32.79 Bq) and from 2.68 pA to 21.5 pA by using an alpha source of still lower activity of ~ nano-Curie (36.63 Bq) as shown in Fig.4 for an applied bias of 90 V across the sample.

5.3 RESULTS AND DISCUSSIONS

It was observed that when the experiment was conducted in air, the base current (i.e. current measured in absence of alpha exposure) was more for the experiment corresponding to Fig.2 than that for the experiment corresponding to Fig.3. The reason can be attributed as follows:

(a) In the case of Fig.2, there was a surface adsorbed layer (from the ambient) on the surface of the BOPET sample which was present when the experiment was conducted in air and was absent when the experiment was conducted under vacuum. The surface adsorbed layer was getting ionised due to bombardment of incoming energetic alpha particles during periodic exposure of the sample to alpha source. As the sample was kept under a constant bias, the ions generated in the surface adsorbed layer were very much under the influence of this bias and thus augment the base current through the sample.

(b) In the case of Fig.3, the current shown in the figure was the current measured for the sample in air under a constant bias, prior to exposing it to alpha particles. So in this case, even though there was a surface adsorbed layer from ambient air, present on the sample surface, it wasn't got ionized because of the absence of alpha exposure.

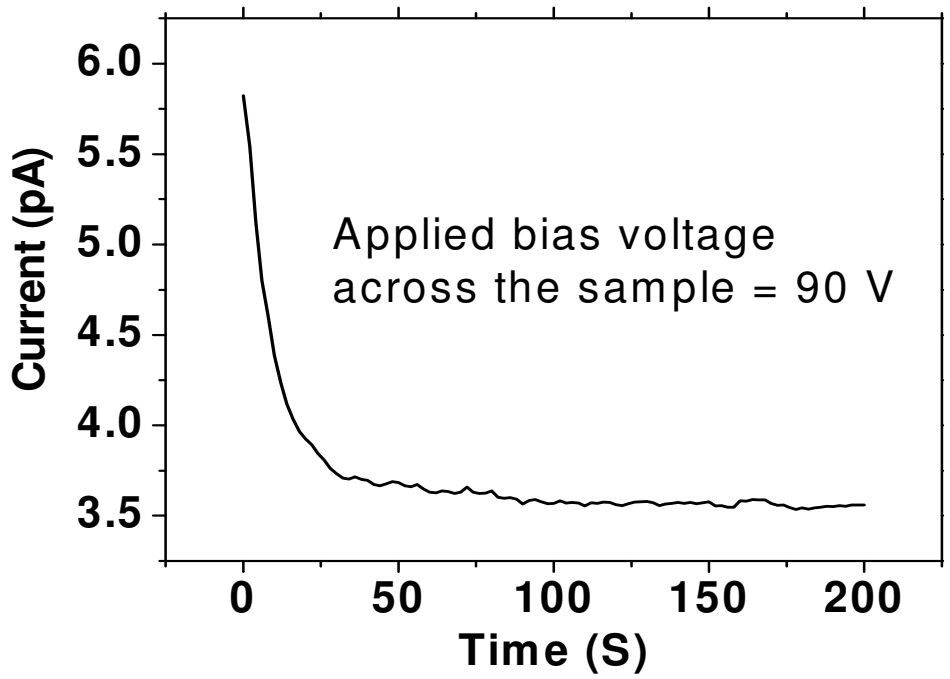

Fig.3. *I-t* characteristics of a BOPET sample (Sample II), measured in air, prior to its exposure to alpha particles.

So whatever current was flowing through the sample, it was purely due to flow of carriers present in the sample. From Fig.2 one can very well draw the inference that a sample of BoPET can act as an alpha sensor and gives incident radiation flux related information of alpha particles. From Fig.4 one can say that for higher value of the alpha source activity, greater was the current output from the BoPET sample. This is obvious because, for a source of higher activity, more number of alpha particles will come out from the source and also will interact with the BoPET sample. As a result of this, more radiation energy would be absorbed

and will give rise to more number of charge carriers in the material. Under the influence of applied bias it will cause enhanced sample current. This entire work is presented and published in DAE-BRNS Symposium on Nuclear Physics – 2015 [72].

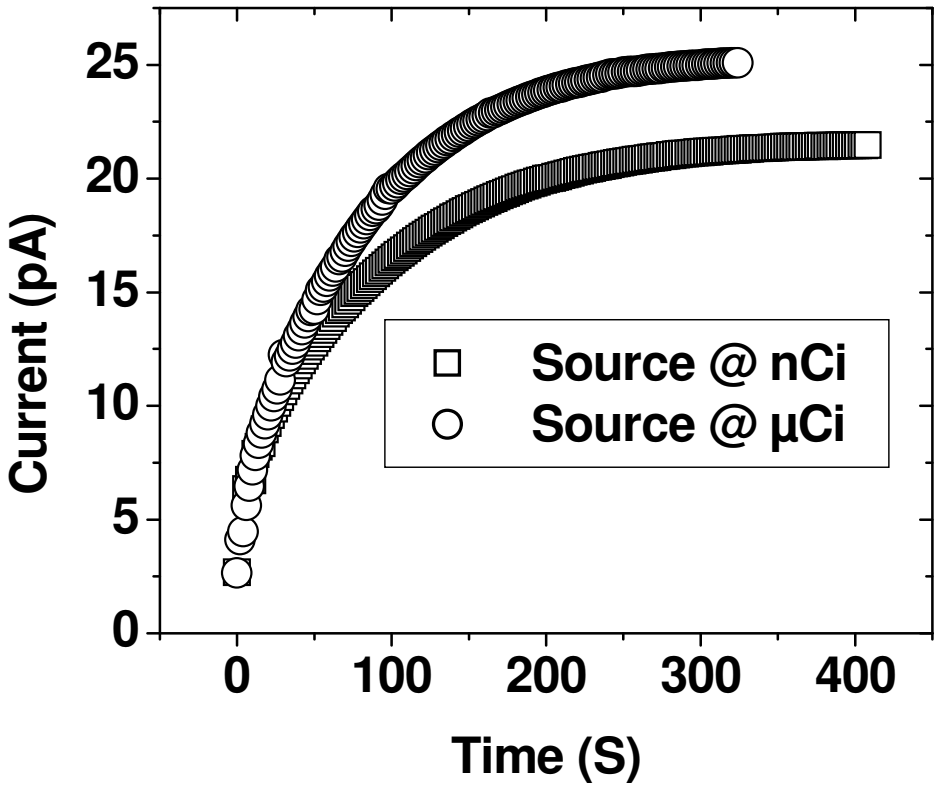

Fig.4. Same BOPET sample (Sample II) was exposed to 5 MeV α from two alpha emitting sources of different activities under vacuum and the *I-t* curve was recorded.

5.4 CONCLUSION

If some radiation is incident on a polymer, then for polymer like BoPET, the immediate change will be that it becomes opaque. It is well known that every polymer is having its own 'glass transition temperature'. If the temperature of a polymer is raised above its glass transition temperature, then whatever changes happened in the polymer during its exposure to radiation will be lost. So whatever opaqueness took place in BoPET due to its exposure to radiation will disappear (i.e. will heal up), in case its temperature is raised above its glass transition temperature.

Chapter 6

CONCLUSION AND FUTURE SCOPE

High quality nuclear radiation detectors will always remain as an important tool to conduct various nuclear physics experiments and experiments are the only available routes to unfold many mysteries pertaining to nuclear phenomenon and radiations. The topic 'Nuclear Radiation Detectors' is vast and the scope of this thesis limits the topics to only silicon and polymer based charged particles detectors. However, the major areas of concern for silicon based detectors are (a) higher leakage current and (b) stable long term behaviour. Various avenues have been explored and presented in this thesis to address these two major issues but still there is scope to do some more work. The summary of this thesis is given below:

An introductory discussion on radiation detection, origin of radioactivity, various radiation sources, interaction of radiation with matter, various types of radiation detectors, criterion to select right material for radiation detectors, definition of energy resolution, factors affecting energy resolution, various components of leakage current and nuclear electronics etc. has been given in the first chapter. This thesis work has dealt mainly with use of organic materials for nuclear detectors. In nuclear detectors organic materials can find different roles such as, active layer in organic/Si hybrid structures (as detailed in Chapter 2), as a passivation layer in Si-based alpha detectors (described in details in Chapter 3) and pure organic material as an active layer (as presented in Chapter 5). This thesis also dealt with a novel surface cleaning methodology using cold plasma flexible brush which has been adopted successfully to clean silicon surface and subsequently used to fabricate silicon surface barrier detectors. These detectors offered better performance than the conventionally prepared (i.e. silicon surface cleaned by inorganic/organic chemicals) silicon surface barrier detectors. This work has been described at length in Chapter 4.

Though lots of effort has been put and basic studies (as proof of concept) in each aspect have been carried out, there is still further scope for improvement. All these works can be considered as future scope of this thesis and are listed as given below:

(a) Study of aging of nuclear detectors based on n-silicon/copper-phthalocyanine heterojunction was carried out where the electrical and alpha particle characteristics of these detectors were studied after a long gap of 3 – 5 years and the best result obtained from one detector (five year old) was reported. Similar studies can be conducted for n-silicon/cobalt-phthalocyanine heterojunction detectors whose 'As prepared' and 'Fifteen months after fabrication' performances were already carried out. Further study over a longer duration will confirm more affirmatively the very long term performance of n-silicon/Metal-phthalocyanine heterojunction detectors.

(b) In passivation studies of silicon using tin oxide, 95-100 layers of ODA-Stannate complex was initially deposited at the edges of silicon which after thermal decomposition gave rise to a thick tin oxide layer. Fresh samples with 60 and 30 number of ODA-Stannate layers may be attempted to get thinner tin oxide layers. Thinner the tin oxide layer, larger will be its resistance and perhaps lesser will be leakage current through it. This aspect of optimizing the tin oxide layer thickness may be explored. Secondly, passivation of silicon surface using hydrogenated amorphous silicon layer can be explored exhaustively on some more silicon samples in order to give this problem a logical end.

(c) With regard to plasma cleaning of silicon surface, some more plasma conditions pertaining to mixed exposure may be explored by varying time of plasma exposure for each of its two components viz. 'Argon with laboratory Air' and 'Argon with moist Air' for further optimization of device performance.

(d) Lastly for all the fabricated detectors, apart from the off-line tests with radioactive emitters, one can perform measurements using heavy ions from accelerator facilities e.g. at FOTIA (BARC) or PELLETRON (TIFR). This will give better idea about longevity of the detectors and how leakage current changes with fluence of heavy and light ions.

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