Introduction

Chapter 1 Introduction

1.1 Motivation

Ionizing Radiation consists of electromagnetic radiations such as X-radiations, Gamma radiation ranging from (keV-MeV) and high energy particles such as neutrons, protons, alpha particles, beta particles (β+, β-), heavy ions and other charge particles, etc. These radiations can eject electron(s) from the atom/molecules of the medium through which they pass. Due to this property, these radiations are used in various applications such as Medical, Environmental, Industrial, Space and Research(Brownell, 1961). Alpha particles are doubly charged particle and are used in various applications such as nuclear transformations, associated particle imaging, gas pressure and density gauges, thickness and humidity gauges, air ionization, analytical applications, pacemaker battery, thermoelectric generator for space, radiotherapy, neutron source preparations. Beta particles are high-energy electrons and positrons and used in beta-voltaic generator, brachytherapy, ophthalmology, etc. Gamma radiation is used in radiography, tele-therapy, sterilization, non-destructive evaluations, sprout inhibitions in vegetables, waste water or sludge disinfection, radiotherapy, sterilization of medical/food products, radiation processing of polymers, degradation of cellulose and scrap material, cross linking of polymers used in insulation etc. X-ray radiations are used in computed tomography, medical diagnosis, mammography, elemental analysis, crystallography, lithography. Neutron radiation is used in neutron radiography, oil well logging and mineral resource exploration, moisture content gauges, quality control of neutron absorbing material, analysis of nuclear materials, neutron activation. Some of these applications are shown in figure 1.1.

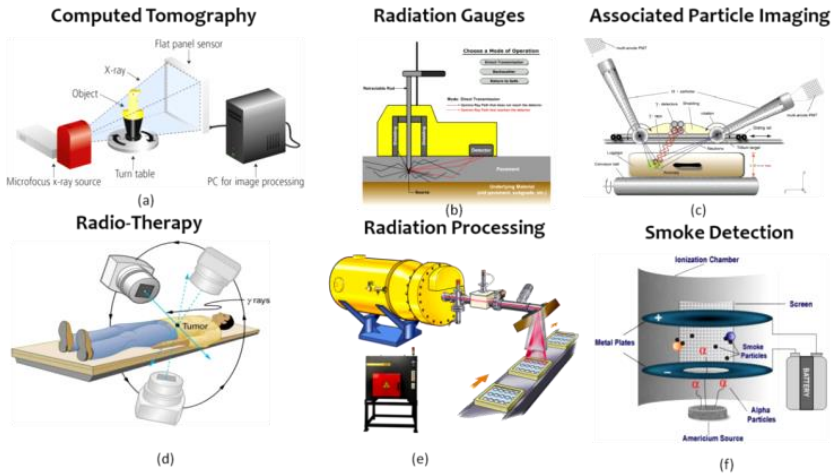

Figure 1.1: Various applications of Ionizing Radiation Sources (source: https://ndt.hamamatsu.com, https://pavementinteractive.org, https://www.fz-juelich.de, https://www.aerial-crt.com, https://www.epa.gov)

Ionizing Radiation detectors are classified into different types depending on the effects produced in a medium. A radiation detector may utilize different materials such as solid, liquid, or gases to detect various kinds of radiation. The different effects produced, such as ionization, scintillation, charge storage and radiation-induced changes in optical/physical/chemical/structural properties, are used to detect and measure ionizing radiation. The solid detector may come in different forms, such as single crystal semiconductor or scintillator, powder, composites, ceramics, etc. There are certain limitation of these detectors and thus, there is a demand to develop efficient and cost-effective detectors for detecting and measuring ionizing radiation.

1.1.1 Requirement of Detector for Radiation Protection

Because of ionizing capabilities, the radiation, when passing through the human body, results in ionization, leading to several health hazards that vary with the type and quantity of radiation absorbed. This radiation reaches humans in two ways: (i) direct external exposure and (ii) internal radioactive contamination. A person working at radiation facilities may get exposed to small amounts of ionizing radiation daily, depending upon the nature of work. Another way of getting high exposure to these radiations is radiological accidents in nuclear/radiation facilities. Either shielding is damaged, causing high radiation levels in working areas, or radioactive materials being released in the environment, contaminating the air surfaces, water bodies, plants/ food. Nuclear accidents such as nuclear reactor damage due to core melting or any other natural calamities and the explosion of nuclear weapons release vast amounts of radioactive material in the environment. The contaminants are **Fission Fragments** viz; Beta and Gamma emitters and nuclear fuel/material that have not undergone fission such as uranium and plutonium isotope are alpha emitters. A large number of fission fragments produced decays over time in few seconds to days; however, some fission fragments have a significantly large half-life, such as ^{137}Cs (30.2y), ^{90}Sr- ^{90}Y (28y), etc. Alpha emitter such ^{239}Pu, ^{240}Pu has very high half-life ~24,100y and 6561y respectively. These contaminants present in the air directly contaminate the human organ if consumed by inhalation or ingestion. Also, the contaminated food such as vegetables, milk, dairy products, meat, fish, etc. produced due to absorption of contaminated water by plant or consumption by the animal can contaminate human organ if consumed (figure 1.2)

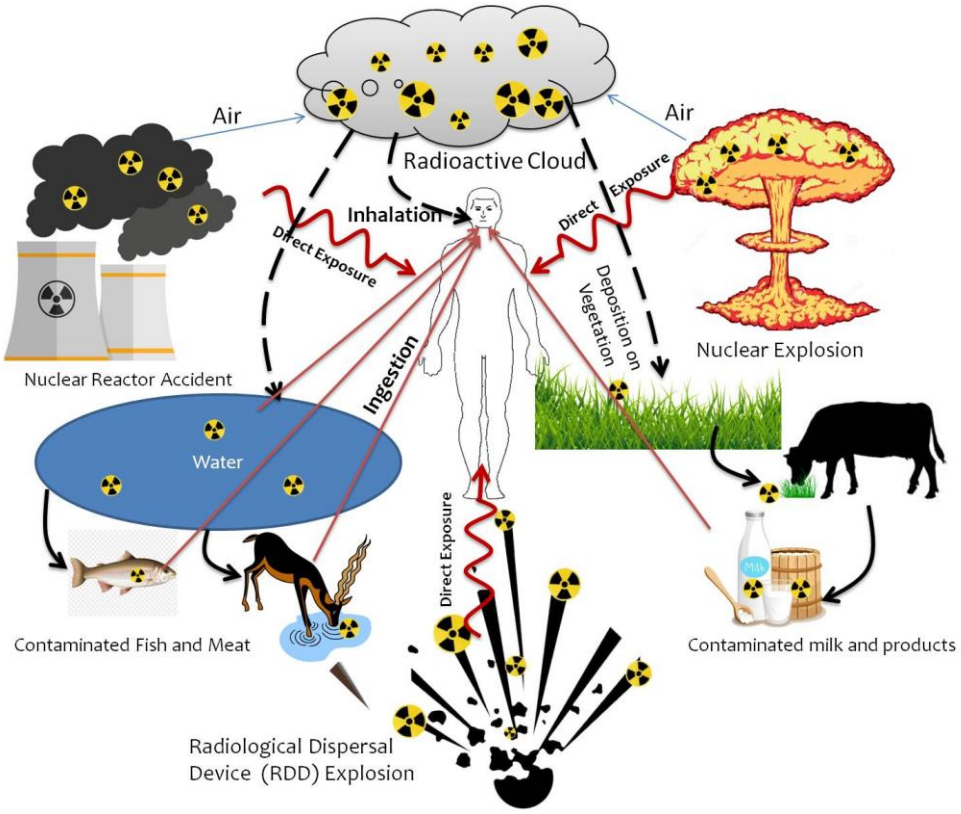

Figure 1.2: Possible ways of radiation exposure due to Direct External Exposure and Radioactive Contamination

Radiation exposure in either form, i.e., direct exposure or exposure due to internal radioactive contamination, causes adverse effects on health due to its biological effects of ionizing radiation. Alpha radiation has less penetration power because of high ionization and mass and is stopped in the skin, whereas beta radiation has more penetration because of lesser

mass. Other energetic radiations such as Gamma and Neutrons have high penetration to the different parts of the body depending on the energy. The penetration of these radiations inside the human body is schematically presented in figure 1.3a. Neutrons with a high radiation weighting factor than gamma radiation have a more damaging effect than gamma radiation. Hazard due to alpha radiation when exposed externally is minimum. But in the case of internal exposure, alpha radiation may be severe. Alpha emitter present in radioactively contaminated air or food/water goes inside the body of the person consuming in the process of inhalation and ingestion, as schematically shown in figure 1.3b. Also, these contaminants may go into the bloodstream if deposited on broken skin/wounds. Ionizing radiation passing through the human body interacts with the cell of different tissues. These interactions may be directly breaking the DNA strands present in the nucleus or radiation interaction with water molecule generating free-radical which further damages the DNA (figure 1.3 c and d). The damage of DNA, in either case, results in either Cell death or Cell repair. In the case of cell death, the deterministic effects of radiation are observed such as chromosomal aberration, nausea-vomiting and diarrhea syndrome (NVD), fever, death due to gastrointestinal (GI) failure, etc. as result of whole body exposure and burn, blister, epilation, sterilization due to local exposures. These effects are certain in nature and have a threshold radiation quantity (radiation dose) over which these are observed. However, in the case of Cell repair, no effects are noticed if the cells are repaired correctly, but in case of mis-repair the function of the cell is altered, leading to uncontrolled growth or mutation. These effects are not certain but stochastic. The stochastic effects such as Cancer and Genetic mutation may occur at any dose with some probability which increases with the absorbed radiation dose. Biological effects depend on the type and quantity of radiation, exposure area (local or whole-body) and tissues under gone exposure (local exposure). Radiation biological effectiveness of the different types of radiation varies with the charge, mass and energy (in case of the charged particle) and interaction with matter.

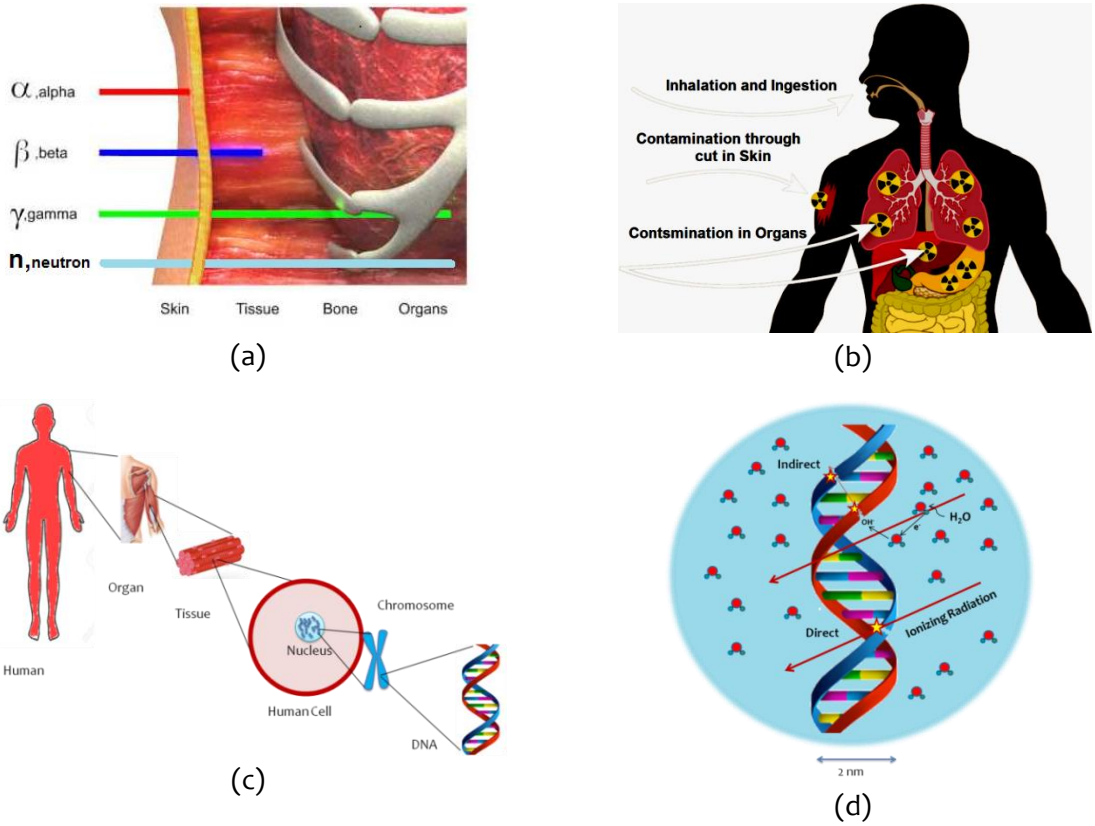

Figure 1.3: (a) Penetration of different radiation in external exposure (b) Internal radioactive Contamination in human organ (source: https://www.liquiglas.jp/gamma, https://www.cdc.gov) (c) Composition of the human body (b) Direct and Indirect damage of DNA by ionizing radiation

The regulatory authorities have decided the minimum radiation dose limits to be received by a worker/public personnel to prevent deterministic effects and minimize stochastic effects. Regular personal monitoring of radiation doses received by radiation workers is carried out by using (i) Thermo-luminescent dosimeters (TLD) and (ii) Fast Neutron Monitoring (FNM) dosimeters. In case of Radiological Accident/emergencies, radiation exposure due to direct and radioactive contamination is to be avoided by measuring radioactive contamination in air, food and water which requires radiation detectors for detection and measurement of the invisible radiation.

1.1.2 Detector Requirement for Imaging Application

Imaging is one of the novel applications of ionizing radiation to use in nondestructive testing. X-rays and gamma rays are widely used for Industrial Computed Tomography/Radiography for internal structure detection. Neutron Radiation is used for the identification of hidden explosive/nuclear material through associated particle imaging. Fast Neutrons of energy 14.1 MeV are generated due to a neutron generator employing the D-T reaction. Deuteron from ion source is allowed to fall on tritiated target, which emits neutron and alpha particle, exactly opposite to each other. The object under interrogation is irradiated with neutrons, which interact with constituents' atomic nuclei, thus generating gamma radiation. This gamma radiation is monitored and analyzed. The timing and direction associated with alpha particles gives the information of neutrons' direction and timing, which help in determining the location of hidden explosives in the container (figure 1.4 a).

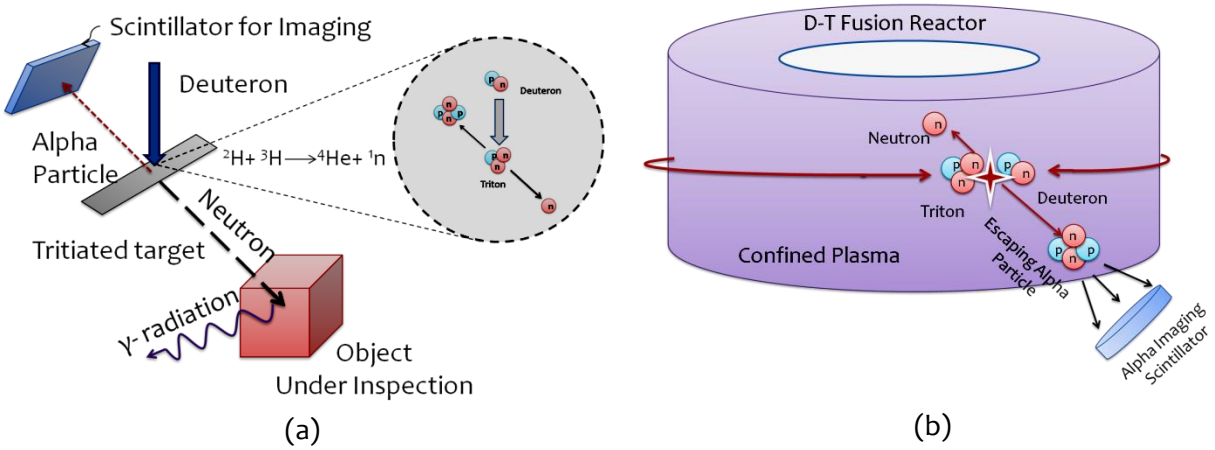

Figure 1.4 : (a) Alpha Scintillator requirement in Associated Particle Imaging (b) Alpha scintillator for escaping alpha particle flux for plasma confinement

The timing performance of the detector is very important for its applications in associated particle imaging. The decay time of the order of pico second to nano second is required for such application.

1.2 Objective of Thesis

The thesis work is on developing metal oxides, i.e., ZnO and TiO_2 based radiation detector, to detect and measure **Alpha, Neutron and Gamma** radiations. We fabricated four structures (i) ZnO:Ga/FTO Glass Nanorods as scintillator detector for alpha particles (ii) ZnO/Polystyrene composite scintillator for alpha particle monitoring (iii) ZnO-^{6}LiF/Polystyrene for thermal neutron detection (iv) TiO_2 Nanorod/FTO for measurement of Gamma radiation doses. The first structure, i.e., ZnO:Ga nanorods based scintillator, consists of vertical nanorods

synthesized by low-temperature hydrothermal method. Their structural, morphological, optical and scintillation properties are studied to realize an efficient alpha particle detector. This detector may be useful in a high-temperature environment as this can sustain temperature ~ 400 ^{0}C. Another detector for the alpha particle is prepared using a mixing process at room temperature, a composite of ZnO micro-particles and polymer polystyrene. The optical, luminescence and scintillation properties are studied for ZnO/Polystyrene composite with different % w/w of zinc oxide loaded in polystyrene. This detector shows a good response to alpha radiation and can be utilized in alpha contamination monitoring. The flexible polymer base facilitates the preparation of this detector in various shapes and sizes. It can also be coupled with other composite scintillators for making a layered scintillator for mixed radiations.

The promising response of ZnO/Polystyrene is further utilized in developing a thermal neutron-sensitive scintillator. ^{6}LiF with 95% enrichment of ^{6}Li is embedded in ZnO/PS scintillator for making a ZnO-^{6}LiF/Polystyrene scintillator to detect thermal neutrons. Thermal neutrons absorbed by ^{6}Li result in alpha particle production by (n, alpha) reaction which subsequently is detected by Zinc oxide, emitting UV-Vis emission due to alpha particle excitation. Response of ZnO-^{6}LiF/PS is tested with Am-Be neutron source by varying the composition. Neutron radiation measurements are performed with this detector and compared with a neutron dosimeter. Another structure explored for measurement of gamma radiation dose is TiO_2NR/FTO samples, prepared using the hydrothermal method. Structural, microstructure, optical properties are studied to understand the growth behavior and surface morphology. Gamma radiation-induced changes were investigated using I-V measurements. A significant change in I-V measurements was observed, which is linear with gamma dose exposed.

1.3 Brief Results and Future Scope

An extensive review of the literature was carried out to understand the mechanism of radiation absorption in a detector medium, ways for detection and measurement of ionizing radiation and current research status in the development of such detectors. We identified a low-cost alternative method for fabricating the detectors and studied the response. The research work performed during the present study resulted in the following major findings:

(i) Highly textured and vertically aligned along (002) Gallium-doped Zinc Oxide (ZnO:Ga) nanorods were obtained using the low-temperature hydrothermal method. The average diameter of these nanorods was obtained as ~150 ± 10 nm. Near band emission of 393 nm obtained photoluminescence measurement, which agrees with the bandgap value ~ 3.22 eV, measured by UV-Vis spectroscopy. Alpha radiation response of these nanorods is studied by coupling it to PMT and 1K Multichannel analyzer. Alpha detection efficiency is obtained as ~28% and minimum detectable activity ~1.7 Bq. The response of single ZnO:Ga nanorods scintillator is repeatable within ± 1%. Sample to sample reproducibility studied for four samples is found varying from ± 5 to ± 20% for high activity alpha source to low activity alpha source.

(ii) ZnO-Polystyrene composite scintillator prepared using solution mixing process. ZnO microparticles loaded with different % w/w in polystyrene on a quartz substrate. Absorbance and transmittance show the exponential decrease in transmittance with ZnO% w/w, corresponding to the increase in absorbance. Bandgap measured from diffused reflectance data shows bandgap ~4.5 eV corresponding to Polystyrene and 3.22 eV for ZnO. Photoluminescence emission ~ 385-390 nm shows the near band emission of ZnO along with some visible emission due to defects. Alpha excited radio-luminescence obtained using 5.2 MeV alpha particles of ^{239}Pu shows emission ~387 nm with intensity increasing with ZnO% w/w. Similar setup as for ZnO:Ga nanorods used for alpha radiation experiments. Pulse height spectra show pulses in a

higher channel compared to bare PS for ZnO loading 5% onwards. The integrated counts are linearly increasing with ZnO % loading up to 20% w/w and deviate at 50% ZnO because of transmittance loss. Maximum detection efficiency ~23% and MDA~0.4Bq obtained for ZnO/Polystyrene with 50% ZnO loading. The response of all scintillator films was found linear with alpha activity. The alpha radiation detection performance with good sensitivity and very low MDA make ZnO/PS a good candidate for monitoring alpha radioactivity.

(iii) Neutron radiation response of ZnO-LiF/Polystyrene studied with Am-Be neutron source. The pulse height spectra of the composite film with natural LiF (^{6}Li, 7.5%) and ^{6}LiF (95% ^{6}Li enriched) showed increased pulse height in enriched LiF, confirming the **n-α** reaction of thermal neutrons with ZnO-LiF/Polystyrene films. The response with the variation of ZnO and LiF ratio affects the integrated counts. For 1:1 ratio of ZnO and ^{6}LiF, the integrated counts are maximum. Further experiments are carried out using ZnO-LiF (1:1)/Polystyrene. Neutron radiation measurement for different time duration was carried out. The integrated net counts are found linear with time, plotted against the neutron dose measured by fast neutron dosimeter, also found linear. The sensitivity of ZnO-^{6}LiF obtained is ~206 counts/μSv dose. Neutron radiation measurement with a variation of the source to detector distance showed similar variations with the distance measured by neutron dosimeter.

(iv) TiO_2/FTO glass prepared using hydrothermal method. XRD measurements show the nanorods are highly oriented along the (002) plane and in the Rutile phase. FESEM micrograph shows the nanorods' uniform distribution over the FTO substrate with high density. The diameter of these nanorods ranges from 20-80 nm. Raman spectroscopy shows three vibrational Raman modes at 237.49 cm^{-1}, 444.02 cm^{-1} and 608.59 cm^{-1} correspond to B1g, Eg and A1g modes of TiO_2 Rutile phase. Bandgap obtained from diffused reflectance data is ~3.15 eV. Photoinduced lifetime carrier measurement is performed using a photoconductive method employing a fluorescent lamp as the light source. The average lifetime of photoinduced charge carrier is ~ 78 s. For electrical measurements, silver metal contacts are employed over top of TiO_2 nanorods. I-V measurement of unexposed nanorods film shows the Schottky behavior. On exposing the film with 375-3000 mGy gamma dose, the leakage current increases linearly w.r.t. gamma dose.

The thesis work carried out may be useful to realize detector for alpha particle, thermal neutron radiation and gamma radiation. Further, the present thesis work may also provide insight to researchers and developers to explore other materials/structures for alpha particle measurements.