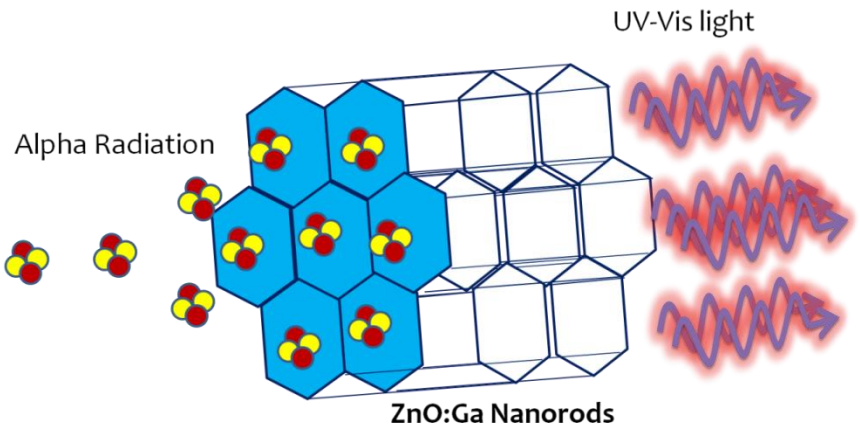


Alpha Radiation Detection using ZnO:Ga Nanorod Scintillator

Chapter 5 Alpha Radiation Detection using ZnO:Ga Nanorod Scintillator

5.1 Introduction

Zinc oxide is a wide bandgap material ~3.37 eV and suitable for optical/electronic applications such as Photocatalytic, Sensing, LEDs, UV detection application etc. The near band emission ~ 380 nm in ZnO is utilized for scintillation applications because of its fast decay times ~ ns. However, defect band emission ~ 520 nm makes it slow, which cannot be used for an imaging application. The doping of n-type of impurities suppresses the defect band emission making ZnO an ultrafast scintillator. Zinc oxide nanorods, because of their preferred orientation, have unique optical properties and are LEDs and UV lasers. ZnO:Ga doped nanorods can serve as low-cost alternative single crystals to apply fast detection of alpha particles/neutrons.

In this chapter, synthesis of ZnO:Ga nanorods over FTO glass substrate using low-temperature hydrothermal method and its application as alpha detector is presented. Photoluminescence ~393 nm shows suppression of defect band emissions due to Ga doping. These nanorods were coupled to PMT and 1k MCA to realize an alpha radiation detector. Alpha radiation measurements were performed using different alpha sources ^{241}Am and ^{239}Pu and their mixtures. Pulse height spectra show the alpha-induced scintillation is proportional to alpha source activity and thus, counts are linear. Alpha response with a variation of the source to detector distance studied and found no response at 50 mm confirming the range of alpha particles ~41 mm. Alpha sensitivity calculated as a minimum detectable activity for 95% confidence ~ 1.7 Bq. The repeatability of the device is within ±1% and the reproducibility among the samples is with ± 5% for such detectors.

5.2 Experimental Procedure

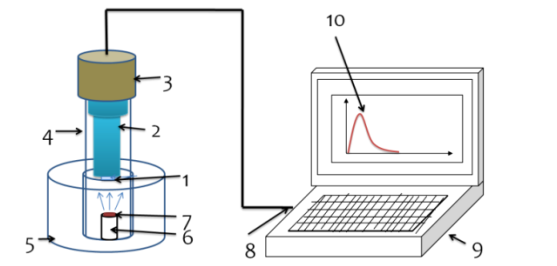
5.2.1 ZnO: Ga Synthesis on Glass substrate

A 10 mM of zinc acetate dihydrate ($Zn(CH_3COO)_2.(2H_2O)$) and 10 mM of monoethanolamine are dissolved in 10 ml of isopropyl alcohol (IPA) for the preparation of zinc oxide nanorod scintillator material of desired thickness on a 1"x 1" FTO glass substrate. The solution is stirred at room temperature for 4 hours and is kept for aging at room temperature for 24 hours. The aged solution is spin-coated at 3000 rpm for 30 s and then preheated at 300 °C for 5 minutes. The process is repeated five times to get the desired thickness of the ZnO seed layer. The substrate is finally heated at 450 °C to get a uniformly distributed ZnO seed layer. To get Ga doped ZnO nanorod, 25 mM zinc acetate dehydrate ($Zn(CH_3COO)_2.(2H_2O)$), 25 mM of Hexamethylenetetramine (HMTA) and 1.25 mM of gallium nitrate hydrate are dissolved in 300 ml of DI water. The solution is stirred at room temperature for 1 hour. The seed layer coated substrate is placed in the solution with the seeded surface facing down. The solution is heated at 93 °C for 6 hours in an electric oven. The substrate is allowed to cool down to room temperature and then it is washed with DI water several times to remove any residual impurities. The substrate is finally heated at 450 °C for 4 hours in the air to facilitate highly c-axis oriented ZnO nanorods.

5.2.2 Development of Alpha Radiation Detector

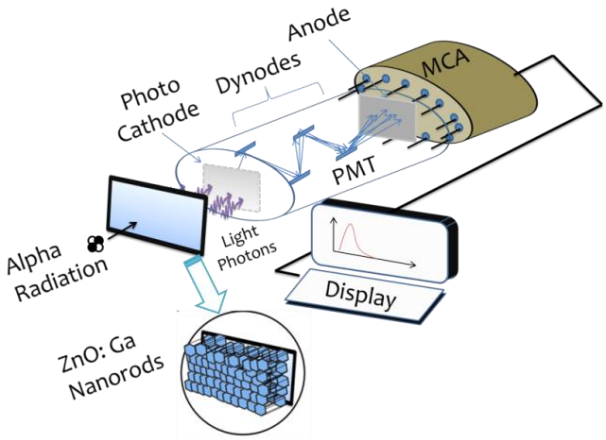
The scintillator material obtained by this process was coupled to a photomultiplier tube using silicon optical grease to match the refractive index with glass substrate to a glass of PMT to realize an alpha radiation detector. It allows the collection of maximum light generated by the PMT. Photomultiplier uses cathode made of photosensitive material that acts as the converter of a photon in electron and an anode, kept at the sufficiently high potential of kV order. Multiple dynodes are kept at different potentials between photocathode and anode. Light photon falling on photocathode generates electrons from the photocathode. These generated electron feels electric force due to the first dynode kept at a higher potential than photocathode and thus got accelerated towards the dynode and generates additional electrons on falling on the dynode. The process repeated from the next dynode to the last dynode and finally a large number of electrons generated fall on the anode of PMT, generating a current pulse (figure 5.1b).

Any optical measurement requires a dark condition so that background can be minimized or ideally can be brought to zero. A chamber is designed in such a way that it functions in two ways; 1) Act as the base of the detector and 2) Facilitates a sufficiently dark environment to detector probe so that entrance of external light in PMT can be minimized. A cylindrical vessel is used that severs such purpose. A cylindrical column is used to cover the PMT so that the entrance of light from the sides can be prevented. The commercially available MCA with an in-built high voltage divider, preamplifier and spectroscopic amplifier is used to develop an alpha radiation detector. Detector probe can be plugged in the MCA, connected with a USB cable to control and display system for acquisition and analysis of pulse height spectrum. Control and Display system is a laptop/PC equipped with spectrum acquisition and analysis software. It powers the MCA and controls the parameter such as PMT high voltage, Gain, LLD and Counting time for radiation measurement. Pulse height spectrum is acquired using the software and analyzed to get pulse height spectrum, integrated counts, count per sec (cps), etc. Figure 5.1(a) shows the schematic diagram of the developed alpha radiation detector. Alpha particle source kept in the chamber at a distance of 5 mm interacts with scintillator generate UV-Vis photon that the PMT senses in electrical voltage pulse of different height, which is displayed on the control and display system for analysis (figure 5.1 b). The actual components of the alpha radiation detector are shown in figure 5.1 (c & d).



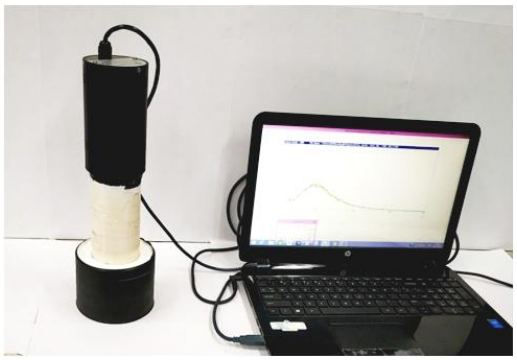
1. Nanorod Scintillator Film
2. Photomultiplier Tube
3. MCA with inbuilt HV, Preamp. and Specamp.
4. Cylindrical column for light protection
5. Cylindrical Vessel
6. Stand for Radiation Source
7. Radiation Source
8. USB Connection to Power MCA
9. Display unit
10. Spectrum

(a)



(b)

(c)



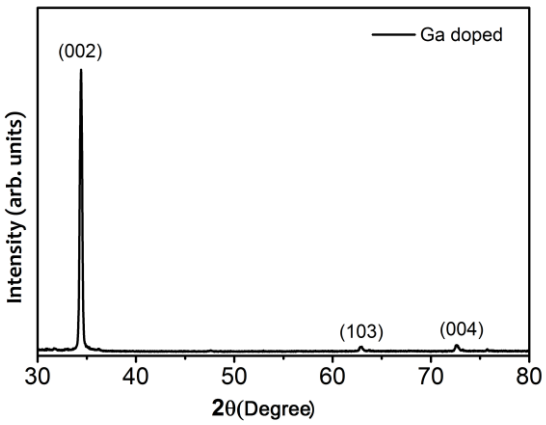
(d)

Figure 5.1: (a) Actual component of used in the development of alpha radiation detector (b) developed alpha radiation detector (c) schematic diagram of alpha detector (d) schematics of the process involved in the scintillation process

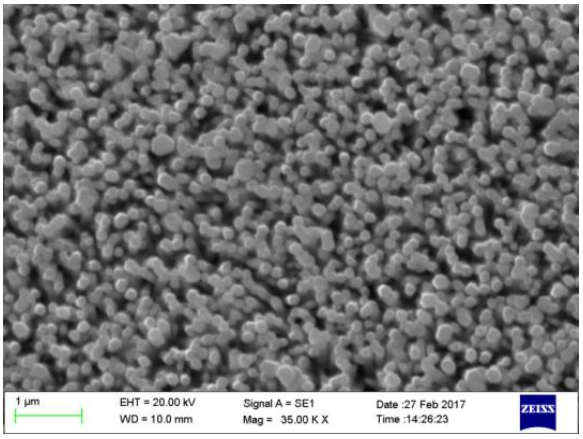
5.3 Results and Discussion

5.3.1 Structural and Microstructural Properties

XRD pattern of ZnO:Ga nanorods are shown in fig 5.2 (002) peak shows grown nanorods are highly oriented along the c-axis. Scanning electron micrograph shows ZnO:Ga Nanorods uniformly grown on the glass substrate and are vertical as the hexagonal face is clearly visible. The average diameters of these nanorods were obtained as ~150±10 nm. (figure 5.2)



(a)



(b)

Figure 5.2 : (a) X-ray diffraction pattern (b) Scanning electron micrograph of ZnO:Ga Nanorods on Glass substrate

5.3.2 Optical and Luminescence Properties

Diffused reflectance measurement carried out UV-Vis spectroscopy and Absorption coefficient calculated from Kulbeka-Munk function $\alpha = \frac{(1-R)^2}{2R}$; plotted w.r.t. energy hv in figure 5.3 a. Bandgap is obtained by drawing a line to x-axis and is ~ 3.22 eV. Photoluminescence emission of ZnO:Ga nanorods, excited with 320 nm, is shown in figure 5.3b. Sharp emission at 396 nm shows the near band edge emission in ZnO.

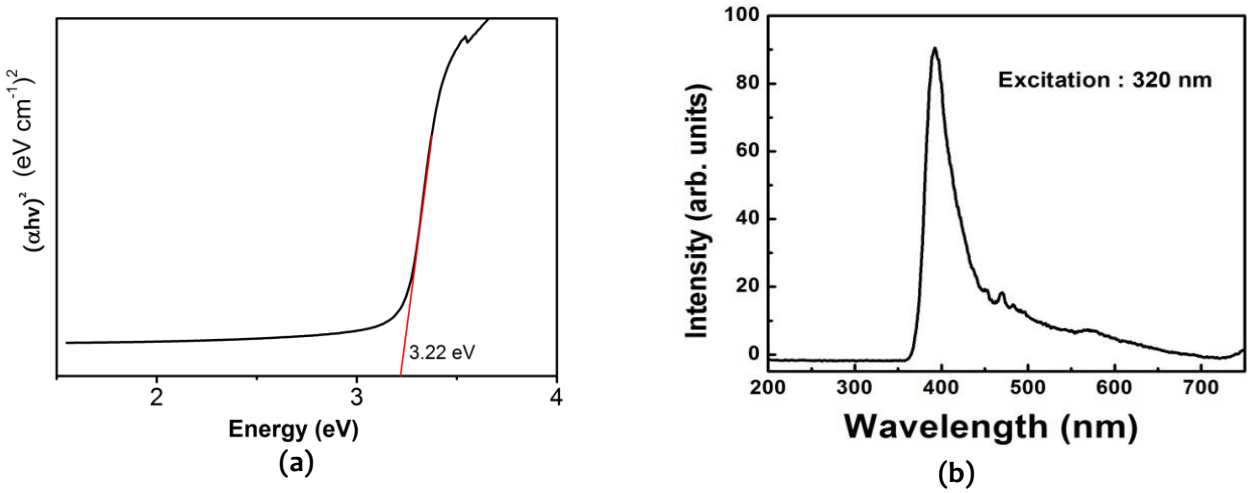


Figure 5.3: (a) Band Gap Measurement (b) Photoluminescence emission of ZnO:Ga nanorods

5.4 Alpha Radiation Response

5.4.1 Scintillation Pulse Height Spectra and Linearity

Alpha pulse height spectra from different alpha sources ^{241}Am +^{239}Pu (13 Bq), ^{241}Am (3700 Bq) and ^{239}Pu (22200 Bq) are shown in figure 5.4 a. The zoomed view of the background pulse height and pulse height from ^{241}Am+^{239} Pu source being extremely low activity source is shown in the inset of figure 5.4 a, which can be clearly seen in the contrast of the background. The intensity of pulse height spectra increases with an increase in the activity of the alpha source. Net counts obtained by subtracting the background counts from source counts are plotted vs. activity in figure 5.4 b. Net counts are found linearly increasing with activity.

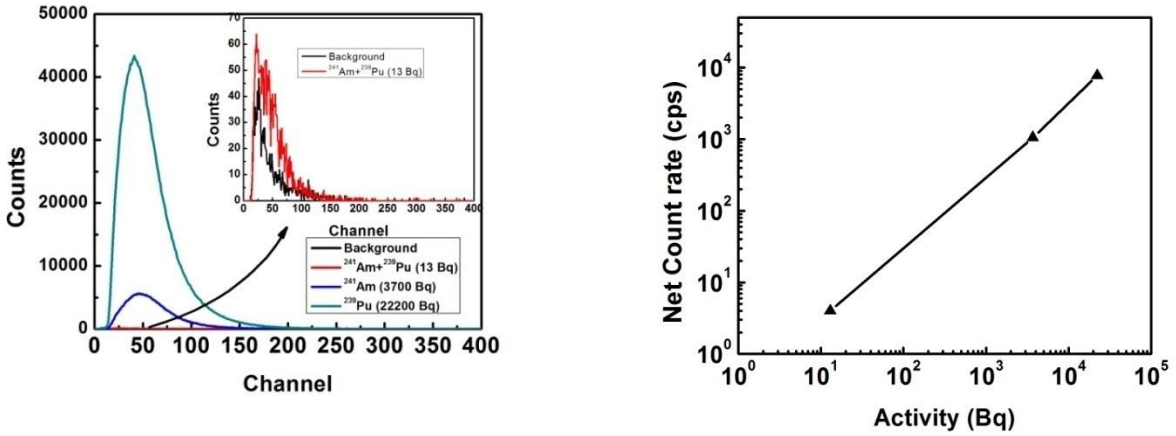


Figure 5.4: (a) Pulse height spectrum of background, ^{241}Am+^{239}Pu, ^{241}Am and ^{239}Pu obtained by alpha radiation detector utilizing ZnO:Ga nanorods (b) Count rate obtained by alpha sources of different activities

5.4.2 Alpha Response Variation of ZnO:Ga Nanorods with Variation of Source to Detector Distance

Alpha response variation of ZnO:Ga nanorod scintillator is also investigated by varying the distance of alpha particle source from 5 mm to 50 mm. The pulse height spectra obtained at these source to detector distance is plotted in figure 5.5 a. As the distance of the alpha source increses, the intensity of pulse height decreases rapidly and merges with the background beyond 40 mm. The counts are also falling with the distance, as shown in the inset of figure 5.5 a. It suggests that the alpha particle is not reaching to the detector at this distance. It agrees with the alpha particle range calculated from SRIM software ~ 41 mm. (figure 2.6 b). Absolute detection efficiency and minimum detectable activity calculated using source and background counts obtained at different distances is tabulated in table 5.1 and plotted in figure 5.5 b. Since the detection efficiency decreases with the source to detector distance, these results correspond to minimum detectable activity. Maximum efficiency is obtained at minimum source to detector distance, i.e., 5 mm, which is ~28.5% and corresponding minimum detectable activity 1.7 Bq.

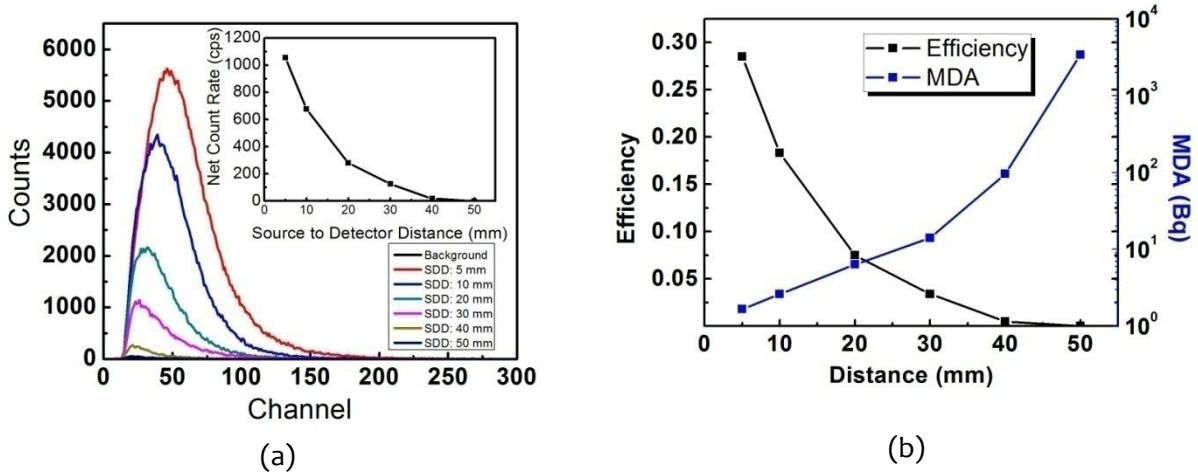


Figure 5.5: (a) Pulse height spectrum of ^{241}Am obtained at different source to detector distances (b) Detection efficiency and MDA at different source to detector distance

Table 5.1: Absolute detection efficiency calculated at different source to detector distance using ^{241}Am Source with counting time 300 sec and average background count rate 3.0 cps.

Source to Detector Distance (mm)	Net Source Counts (cps)	Absolute detection efficiency	Minimum detectable Activity (Bq)
5	1054.8	0.285	1.66
10	677.9	0.183	2.59
20	280.4	0.075	6.32
30	126.3	0.034	13.94
40	18.5	0.005	94.80
50	0.5	0.00014	3385.95

5.4.3 Alpha Response Repeatability of ZnO:Ga Detector

Repeatability of the alpha response of ZnO:Ga nanorod scintillator is investigated by analysing the net counts variation in multiple measurements of 60 second duration. Pulse height spectra obtained from background and from all three alpha source in five successive runs is shown in figure 5.6 a-d. The net count rate from all sources is summarized in table 5.2 and plotted in figure 5.6 e. the variation found in net count rate is within ±1%, which suggests that the performance of ZnO:Ga detector is highly replicable.

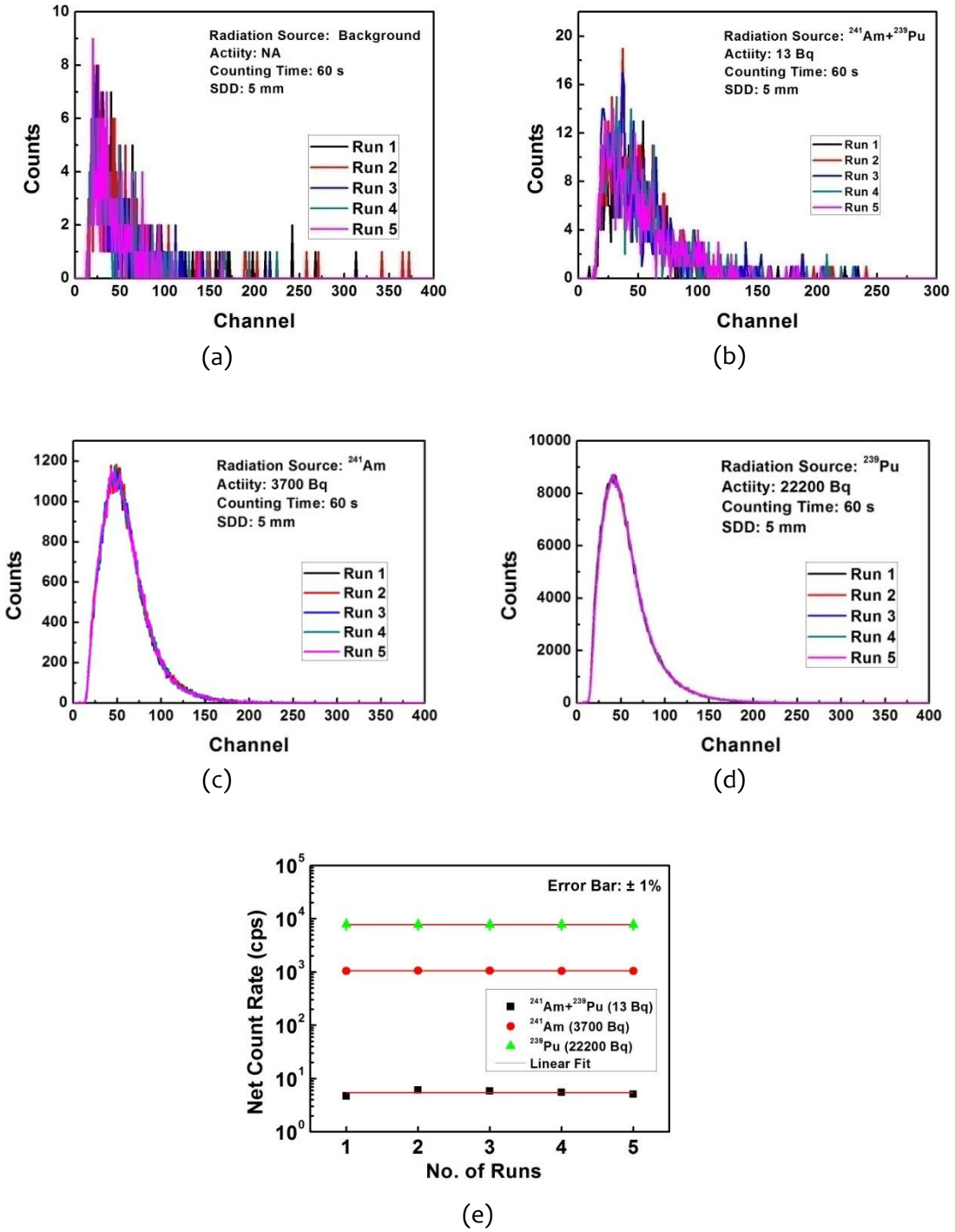


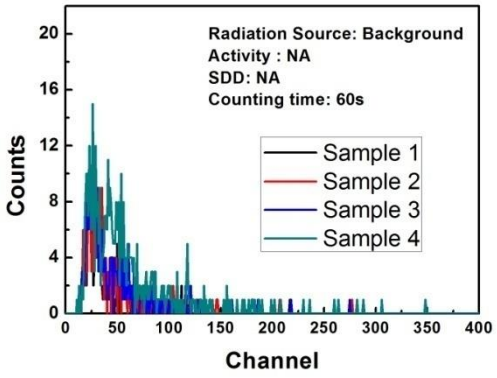
Figure 5.6: Pulse height spectrum of (a) background (b) ^{241}Am+^{239}Pu source (c) ^{241}Am source (d) ^{239}Pu source recorded five times by alpha radiation detector (e) Variation in net count rate obtained by different radiation sources in five no. of runs

Table 5.2 Count rates of different sources at a distance of 5 mm and Counting time 60 s

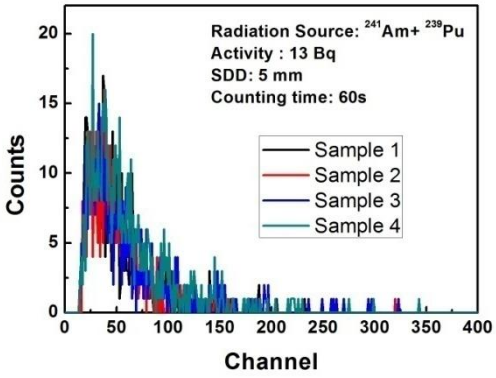
No. of Runs	Average count rate (cps)			
	Back-ground	**^{241}Am+^{239}Pu (13 Bq)**	**^{241}Am (3700 Bq)**	**^{239}Pu-239 (22200 Bq)**
1	2.8	7.5	1044.3	7747.8
2	3.2	8.9	1057.1	7736.6
3	2.7	8.6	1056.9	7735.0
4	2.7	8.3	1049.3	7739.5
5	2.6	7.9	1051.9	7720.3
Average	**2.8**	**8.2**	**1051.9**	**7735.8**

5.4.4 Alpha Response Reproducibility of ZnO:Ga Detectors

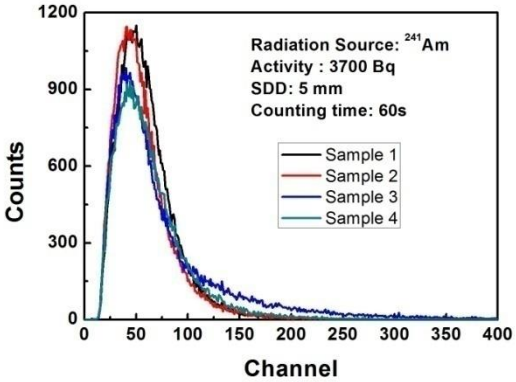
Alpha response variation in diffrent samples of ZnO:Ga nanorods preprapred in simalr conditions. Pulse height spectra recorded in five runs using all three sources along with bacground for 60 s are shown in figure 5.7 a-d. Count rate variations are shown in table 5.3 and plotted in figure 5.7e. The variation observed in net count rate of four ZnO:Ga detector is within ± 20% for extremely low activity source ^{241}Am+^{239}Pu source; ± 10% for ^{241}Am source and ± 5% for ^{239}Pu.



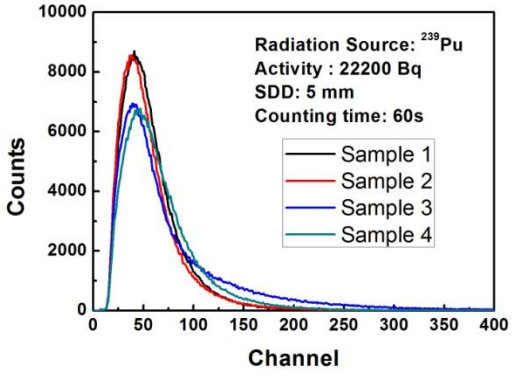
(a)



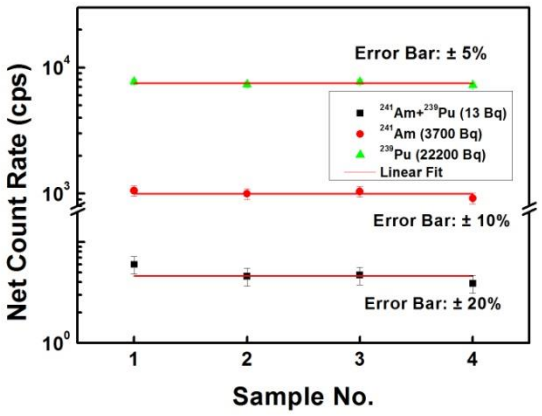
(b)



(c)



(d)



(e)

Figure 5.7: Pulse height spectrum of (a) background (b) ^{241}Am+^{239}Pu source (c) ^{241}Am source (d) ^{239}Pu source recorded with different ZnO:Ga nanorod detectors five times by alpha radiation detector (e) Variation in net count rate obtained by different radiation sources

Table 5.3: Count rates obtained by using four samples of ZnO:Ga detector and different radiation sources

Sample No.	Av. Counts (cps)			
	Back-ground	^{241}Am+^{239}Pu (13 Bq)	^{241}Am (3700 Bq)	^{239}Pu (22200 Bq)
1	2.6	8.6	1056.9	7735.0
2	3.2	7.8	997.9	7359.8
3	4.0	8.7	1042.6	7734.9
4	7.2	11.1	928.6	7272.2

5.5 Conclusion

Zinc Oxide doped with Gallium nanorods was synthesized using a low-temperature hydrothermal method. Material properties such as phase, microstructure, bandgap and luminescence properties have been investigated. An alpha radiation detector is made by coupling these nanorods to PMT, MCA and control and display system. Performance evaluation with alpha sources shows the linear performance of the detector with the activity of the radiation source used in the study. Detection efficiency at 5 mm detector distance is 28.5% and MDA obtained is 1.7 Bq. The repeatability of the alpha radiation detector is within ±1%. Four ZnO:Ga detector response varies from ± 20% for extremely low activity alpha sources to ± 5% for high activity alpha sources. High sensitivity to alpha radiation with good repeatability shows that the detector can be used for alpha radiation detection and measurement purpose. The work has been considered for patent and a patent has been filed.