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Preliminary study on the properties of zinc oxide (ZnO) for alpha particles detection

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ABSTRACT

Background*: :** Due to the difficulties of locally obtaining ZnS(Ag), preliminary investigation of the radioluminescence characterization of Zinc oxide (ZnO) for alpha particle detection was performed. ***Materials and Methods*:** The scintillation properties of ZnO films were tested using alpha sources (Am-241). The correlations between ZnO scintillation responses and irradiation time and source activity were also verified and compared with the response of a ZnS(Ag) detector. ***Results: The obtained results showed that the response of ZnO was linear with the exposure time, and the uncertainty of the repeatability was less than 1%. In addition, ZnO was found to have good radiation resistance over a wide range of doses. ***Conclusion:*** The preliminary results may be indicative that the proposed ZnO detector could be considered as a promising detector for alpha particles.

Keywords: *ZnO, ZnS (Ag), Scintillation, Alpha particles detection.*

INTRODUCTION

Silver-activated zinc sulphide, ZnS(Ag), is one of the oldest inorganic scintillators, with a high scintillation efficiency for alpha particles detection[1,2]. It has been widely used for environmental radon measurements. Due to the opacity of ZnS(Ag) to its own luminescence, thicknesses greater than about 25 mg/cm^2 become unusable[3]. Due to the difficulties in obtaining or developing Zn(Ag) and its relatively high price, it was necessary to look for an alternative material with similar properties, and though to be ZnO.

Zinc oxide (ZnO) is an interesting n-type semiconductor material with a wide band gap of 3.2 eV (~ 387 nm) and a large excitation binding energy of 60 meV at 300K[4,5]. ZnO has displayed sub-nanosecond luminescence at room temperature when doped with the n-type dopants such as gallium or indium [6].

Recently, ZnO has attracted great attention due to its promising optoelectronic applications such as ultraviolet light-emitting diodes and laser diodes [7]. ZnO has a good stability under ultraviolet light and relative high electric conductivity when compared with the conventional sulphide phosphors. Single crystalline ZnO is of interest also as a substrate for the growth of GaN films [8].

ZnO is normally formed in the hexagonal crystal structure. Its luminescent property is principally related to electronic and crystalline structures. It is strongly influenced by both the preparation method and dopant [9]. Moreover, ZnO has high scintillation properties comparable to that of GaN [10]. While, ZnO/Al_2O_3 has approximately equivalent x-ray diffraction (XRD) and photoluminescence (PL) line widths, and even lower dislocation densities as GaN/Al_2O_3 [10].

Banaee *et al.* evaluated the effect of ZnO NPs doped with Gadolinium (Gd) on dose enhancement factor by 6MV photon beam irradiation [11]. The results of this study showed that ZnO NPs doped with Gd could be considered as new compound for increasing the absorbed dose.

Cooper *et al.* evaluated Ga-doped ZnO as alpha particles transducers, within a neutron

generator, between 3.2 and 5.4 MeV [12]. An average light output of up to 61 photoelectrons per event was measured. Bourret-Courchesne et al. reported that the intensity and decay time are strongly dependent on the method used for dopant incorporation [13]. In addition, the effect of introducing copper by doping on the photocatalytic activity of ZnO nanoparticles was also studied [14]. It was found that the best photocatalytic properties are for pure ZnO samples.

In this preliminary work, characterization of the scintillation response of undoped ZnO to alpha particles exposure was performed and compared with the response of ZnS (Ag) detector.

MATERIALS AND METHODS

ZnO film was prepared by using the spray coating method. An appropriate amount of ZnO (99.5 % from Honeywell Riedel-de Haën®) was dispersed in super pure water and the aqueous solution was agitated using ultra sonic bath. The ZnO solution was then sprayed on transparent plastic sheet using N2 as a holder gas to optimize the homogeneity of the oxide deposition. The prepared scintillation film dried at 80°C for 12h and kept in plastic bottle. The thickness of the ZnO layer was about 20 µm measured by a micrometer with an accuracy of ± 0.5µm (the range of 5 MeV alpha particles in ZnO is about 14 µm calculated by SRIM software). The scintillation response of ZnO due to alpha particles exposure was measured using the nuclear counting system (AB-5) manufactured by Pylon Electronics Inc. AB-5 is a portable monitor and data acquisition unit, which has a wide range of optional accessories for measuring different types of radioactive decay. AB-5 contains a rechargeable battery, an air pump, a photomultiplier tube (PMT), and associated electronics. The experimental set-up is shown in figure 1.

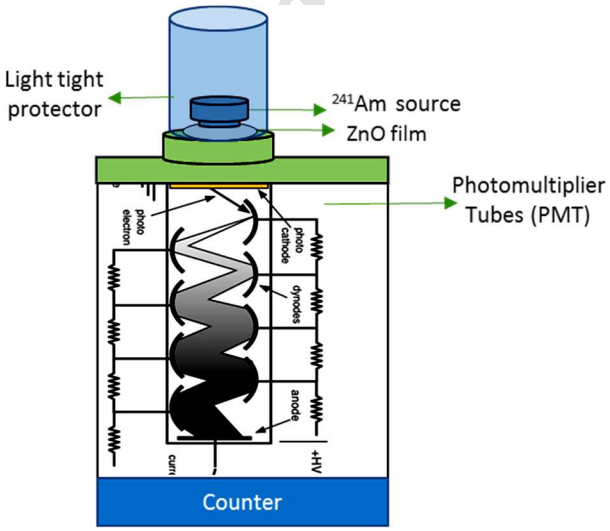

Figure 1. The experimental set-up for testing the response of ZnO.

Five tests were performed to evaluate and characterize the scintillation properties of ZnO. These tests were: characterization using photoluminescence (PL), repeatability, exposure time response, radioactivity response and radiation resistance. Similar tests were performed with scintillation discs ZnS(Ag) manufactured by Pylon Electronics Inc (a transparent plastic base coated with ZnS (Ag)) using the same exposure conditions and results were compared. Both ZnO and ZnS (Ag) detectors are circular discs with a 3 cm of diameters. In addition, the same counting geometry was used to test the two detectors (as shown in figure 1) in order to remove the effect of the geometry changing on the response of the

detectors.

PL measurements were used for the characterization of ZnO and ZnS(Ag). The experimental set-up was consisted of UV excitation using a 325 nm He/Cd laser, and spectral measurements via a grating monochromator with 1200 groves/mm equipped with a cooled Photomultiplier Tube PMT (HAMAMATSU). A long-pass filter was used to reject plasma and laser light from the helium-cadmium laser below 360 nm. Synchronous detection technique was implemented by chopping the laser beam and employing a lock-in amplifier to process the electrical output of the PMT. Via a set of optical devices, the excitation beam was guided and focused on to the tested sample.

The repeatability test was done by recording the responses of ZnO and ZnS (Ag) when repeating their exposures for 1 min to an alpha source (Am-241 source with activity around 7 kBq) fifty times.

The exposure time response was performed by exposing the two detectors, to the same alpha source used for the repeatability test, for different exposure times ranging from 1 sec to 900 sec. The correlations between their scintillation response and exposure time were examined.

To evaluate the radioactivity response, ZnO and ZnS (Ag) were exposed for a fixed time (1 min) to Am-241 alpha sources with different activities (from 0.7 kBq up to 62.0 kBq). The correlations between scintillation response of the two detectors and alpha sources activities were studied.

Finally, the radiation resistance of ZnO and ZnS (Ag) was examined by exposing the two detectors to different doses up to 71 kGy by using Am-241 alpha source with activity of 62.0 kBq. The absorbed dose rate was calculated using the following equation:

$$D^{\bullet} = \frac{A.\bar{E}}{m} \cdot \left(\frac{MeV}{kg} \right) \times 1.6 \times 10^{-13} \frac{J}{MeV}$$

Where:

$D^{\bullet}$: is the absorbed dose rate in Gy/sec

A = the activity of the alpha particles source in Bq.

m: the mass of the ZnO film

$\bar{E}$ = the average alpha particle energy, in MeV per disintegration

The responses for an exposure time of 1 min were compared before, during and after the exposures. The results were normalized to the value of its response obtained before the irradiation.

RESULTS AND DISCUSSION

PL Characterization

Figure 2 shows room temperature PL spectra of ZnO film. ZnO exhibits two emission Bands: the first is around l = 384 nm (~3.23 eV), which is attributed to band to band emission. The second is referred to broad green-band emission centered at 530 nm. It has been attributed to different intrinsic defects in ZnO, such as oxygen vacancies, zinc vacancies and interstitial zinc [10].

On the other hand, ZnS (Ag) exhibits a wide emission peak centered at 450 nm (~2.76 eV) (figure 3). This result is in agreement with those obtained by Erfurt *et al.* [15].

Repeatability

The recorded scintillation counts using ZnO and ZnS (Ag) were approximately unchanged for 50 runs and remained within ± 0.5% and ± 0.4%, respectively, as shown in figure (4). It is clear that both detectors have good repeatability with very low variations. However, the response (count rate) of ZnS is higher than that of ZnO. This could be due to the preparation process of ZnO, as the process used in this work was a simple uniform distribution aqueous solution of ZnO on transparent sheet. This process is currently under optimization to give the highest response.

Exposure time response

The counts measured by both ZnO and ZnS (Ag) detectors for different irradiation times between 1.0 sec and 900 sec demonstrate excellent linearity (R^2=1) (figure 5). It is noteworthy to mention that the characteristics of ZnO were comparable with those of ZnS (Ag) in terms of linearity with exposure time, except

the slightly lower efficiency (about 18 %). This, as mentioned before, may be due to the preparation process of ZnO.

Radioactivity response

Figure 6 shows the response of ZnO and ZnS (Ag) for different alpha activity sources. It is clear that both have quasi-linear response with activities and reach saturations for activity higher than 40 kBq. It should be noted that the response of ZnS(Ag) was about 20% more than ZnO at low activities, which could be due to the preparation process as mentioned before. While, at higher activities the differences become lower than 5%, which could be due to the saturation occurrences. This could be explained by the differences in scintillation response times. As the response time of ZnS(Ag) is about 1.473μs [16], while for ZnO is 0.1μs [17]. This also explains why ZnS(Ag) saturated at lower activity (~25 kBq) than ZnO (~40 kBq).

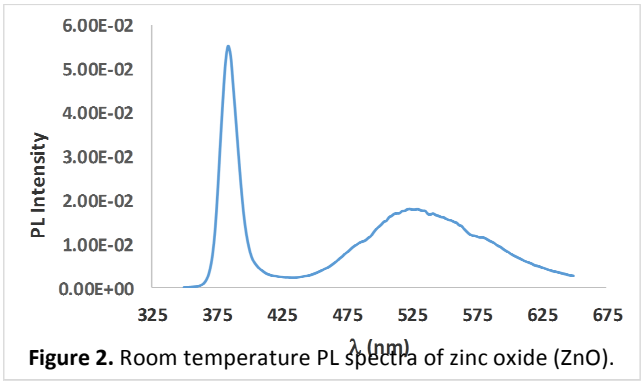

Figure 2. Room temperature PL spectra of zinc oxide (ZnO).

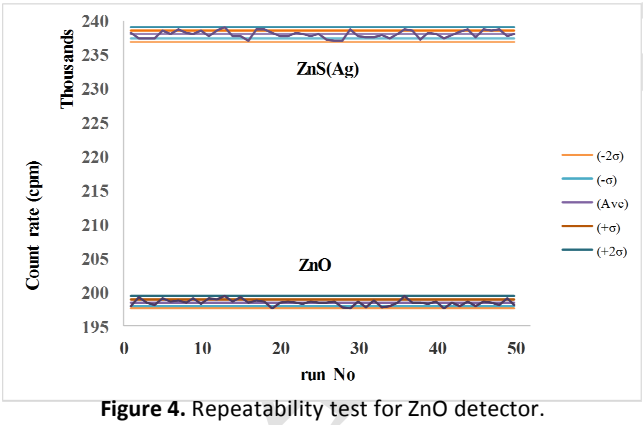

Figure 4. Repeatability test for ZnO detector.

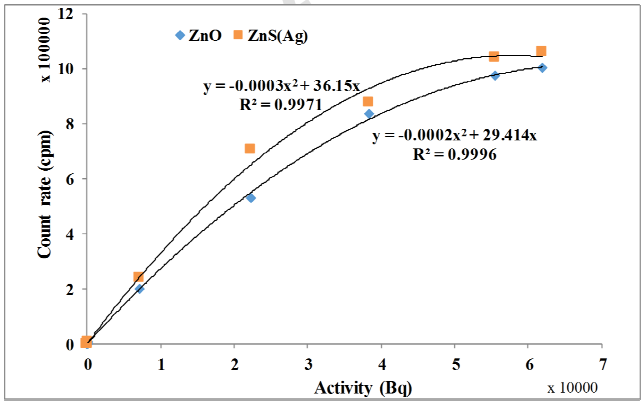

Figure 6. ZnO and ZnS(Ag) radioactivity dependence.

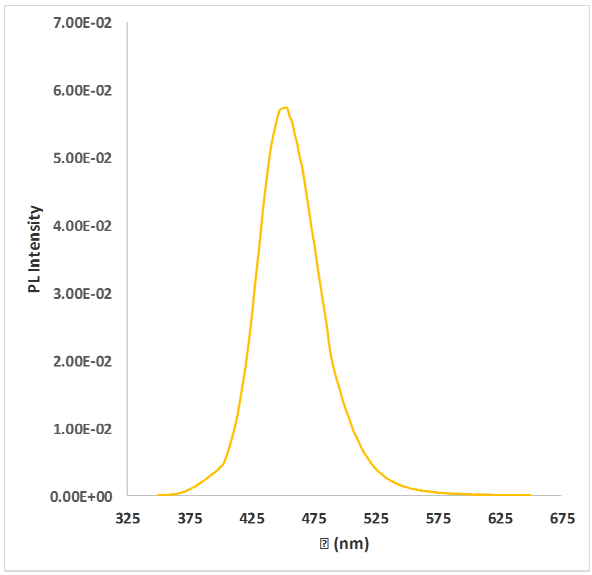

Figure 3. Room temperature PL spectra of Ag-doped ZnS.

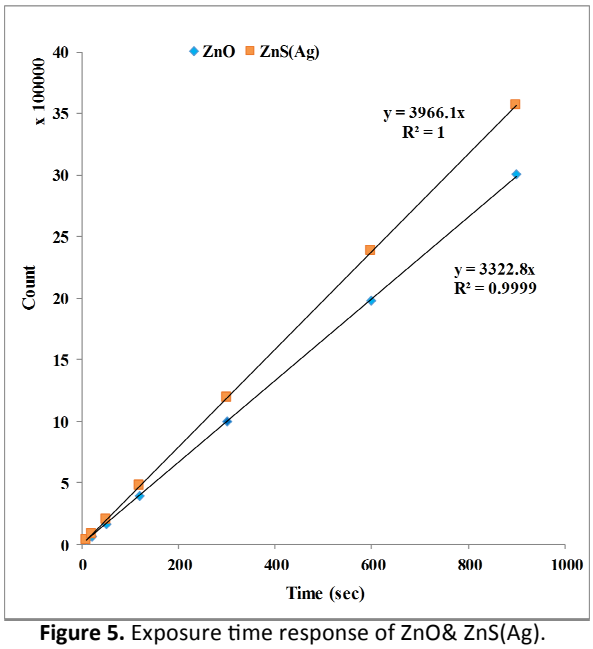

Figure 5. Exposure time response of ZnO& ZnS(Ag).

Radiation resistance

The measured counts obtained by the ZnO and ZnS (Ag) detectors showed good stability and radiation resistance (figure (7)). Variation in the response was around ± 2% after an irradiation with alpha particles source for dose about 71 kGy.

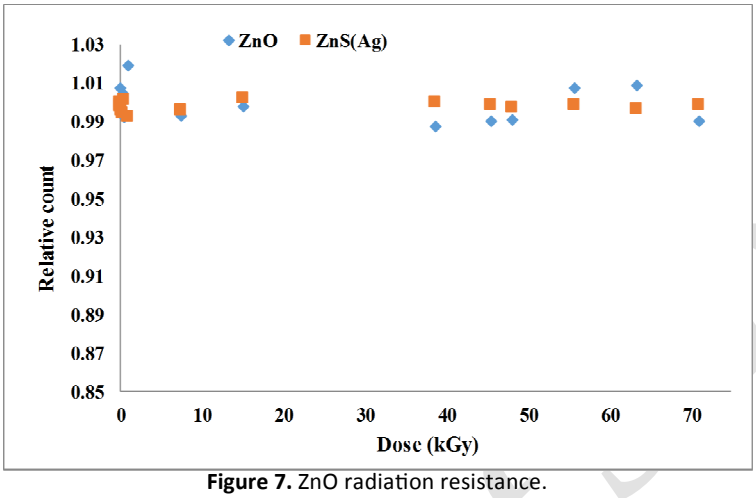

Figure 7. ZnO radiation resistance.

CONCLUSION

ZnO film was prepared by using the spray coating method and its scintillation response to alpha particles exposure was tested. The uncertainty of repeatability found to be within ±0.5% for 50 runs. Its response was linear with irradiation time and it had a very good radiation resistance. In addition, ZnS (Ag) is more sensitive than ZnO due to the preparation process of ZnO which is currently under investigation. However, ZnO is much more available and its preparation is much easier and cheaper. Therefore, the proposed ZnO detector could be considered as a promising detector for alpha particles.

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***Conflicts of interest*: Declared none.**

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