Investigation on the characteristics of ZnO and ZnO-Pb structure for gamma radiation detection

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Abstract. Zinc oxide (ZnO) thin films of multilayers structure were fabricated by chemical bath deposition (CBD) method on a glass substrate. After growing, the optical, structural and morphological characterizations of the samples were studied. Moreover, the crystallite size and energy band gap were calculated by using X-ray diffractometer (XRD) and UV-visible spectrometer, respectively. XRD showed that the crystalline size of the samples decreased with increasing the layers up to four layers, while the energy band gap increased with increasing the layers. Field emission scanning electron microscope (FESEM) used to study the morphological properties of the samples. Based on the effective atomic number, electron density and light yield properties of the samples make it suitable to be used as an ionizing radiation detector. Eventually, the samples will be used as a scintillator material to detect ionizing radiation.

1. Introduction
ZnO belongs to the II-VI group of semiconductor materials with a broad wide bandgap of 3.37 eV and high binding energy of 60 meV. Moreover, due to its nature-friendly, low cost and its high melting point ZnO used in both industrial and medical fields concerning their branches. On the other hand, ZnO can be used as a suitable radiation detector, regardless of its difficulty in the producing of high-quality crystals [1]. The material density of ZnO is 5.61 g/cm³ which is larger than the density of plastic scintillator, but lower than that of other heavy metal oxide scintillators [2]. Recently, number of studies successfully fabricated a scintillator material from undoped and doped ZnO thin film to detect ionization radiation [3-5]. Lead (Pb) has a low melting point, small potential difference, one of the densest materials with density around 11.34 g/cm³ and has atomic number 82. Because of these unique properties, some studies studied the scintillation properties of lead in different forms, such as lead carbonate [6], lead tungstate [7] and lead sulfate [8].

CBD is one of the most excellent methods to prepare nanorods thin films because it can be coated in several substrates, the process is using low temperature and its low cost [9]. The ionizing radiation detection system consists of a radiation source which emits the ionizing radiation, a photomultiplier tube (PMT) to convert radiation photos into electrical signals, power supply and then connected to the computer. Multi-channel analyzer (MCA) and MAESTRO used for data analysis.

In this present study, undoped ZnO and Pb doped ZnO thin films were prepared by CBD. Then the structural, morphological and optical properties of the films were studied by different techniques.
Finally, undoped ZnO and Pb doped ZnO thin films were tested as scintillator detectors. Cs-137 was used as a gamma radiation source to study the scintillation properties of the films.

2. Methodology

2.1. Preparation of seed layer

The thickness of ZnO thin films was 200 nm, this layer done by Radio frequency sputtering (CESAR RF Power Generator) on glass substrates. The power of this process carried out at 150 W, a pressure at \( \sim 5 \times 10^{-5} \text{ mbar} \) and the sputtering rate was at 0.8 Å/s. In addition, Ar flow gas was calibrated at 14 sccm and the sputtering operation has settled at room temperature.

Pb was deposited on the glass substrate using thermal evaporation method. 0.07g of lead added for deposition at pressure 3.4\( \times 10^{-5} \) Torr. The film then annealed at 300°C for 2 hours and a half.

2.1.1. Chemical bath deposition (CBD).

CBD of ZnO nanorods has done from mixtures containing 2.615 g zinc nitrate (Zn(NO\(_3\))\(_2\)) and 1.4 g hexamethylenetetramine (C\(_6\)H\(_{12}\)N\(_4\)) in 200 ml deionized water with continuous stirring for 1 hour. After that the ZnO seed layer of 200 nm immersed reflection symmetry in the solution to prevent the deposition of unwanted ZnO. The same previous steps were done for Pb thin film. Then the solution kept in chemical bath deposition at temperature 90°C for 7 hours. The samples then rinsed with deionized water and left to dry in air.

2.1.2. Study the characterizations and scintillation properties of the samples.

Firstly, the structural properties of ZnO and Pb doped ZnO thin films was studied by using XRD to investigate the structure of the crystalline phase and then to calculate the grain size for samples by using the Scherer's formula [10] in (equation (1)).

\[
D = \frac{(0.89 \lambda)}{(B \cos \theta_B)} \tag{1}
\]

where B expressed the full width at half maximum (FWHM) of the XRD peaks, \( \theta_B \) referred to the Bragg diffraction angle and \( \lambda \) sign of the X-ray wavelength (0.154nm).

FESEM was used to study the morphology of the films. UV-visible spectrometer (UV-vis) was carried out to investigate transmission, absorption, and then to calculate the energy band gap of both samples. The energy band gap estimated by using Tauc's equation (equation (2)).

\[
\alpha h\nu = A(h\nu-E_g)^n \tag{2}
\]

where \( \alpha \) is the absorption coefficient, \( h\nu \) is incident energy; \( E_g \) presents the energy band gap and \( n \) depends on the transition type, in this work \( n=2 \) for the direct transition. Absorption coefficient can be measured by using (equation (3)).

\[
\alpha = 2.303(A/d) \tag{3}
\]

where A is the absorption and \( d \) is the thickness of the film. This calculation process discussed elsewhere [11].

Eventually, based on some scintillators properties such as atomic number, energy resolution, electron density and light yield properties of the samples, the samples were used as scintillator materials to detect gamma rays emitted from Cs-137 for ten minutes, which is enough time for photons to accumulate in the detector and then collected in PMT to convert the light into signals. The data then analyzed by the MCA and MAESTRO. Here in this work, NaI(Ti) crystal detector was the standard to compare its peak with the peak appeared from the samples prepared in this study.
3. Results and discussion

Based on the results we got from Figure 1 for XRD graph, the crystalline peaks of ZnO-Pb were at 34°, 64°, and 72° correspond to (002), (103) and (004), while the peaks of ZnO were at 34° and 72° which correspond to (002) and (004). Although the intensity of ZnO thin film is higher than the intensity of ZnO-Pb, by calculating the grain size of both samples, we found out that the particle size of ZnO is 1.68nm while doping of Pb with ZnO was increased the grain size to 2.05nm.

Figure 1. XRD for ZnO and ZnO-Pb.

Figure 2 shows that FESEM results for both samples, we can notice that the diameter of the nanofiber is larger for ZnO-Pb sample with average size around 470nm, while for ZnO thin film the average diameter size is ~280nm. These results agreed with the results of the grain size from XRD. Moreover, there are some thin fibers in the sample of ZnO-Pb which not found in ZnO thin film.

Figure 2. FESEM for ZnO and ZnO-Pb with the diameter size of nanofibre.
Optical properties of the samples showed that the absorption of ZnO-Pb thin film is higher than that of ZnO thin film, both films are saturated at 400nm and below, Figure 3. Increasing the absorption with doped Pb can be attributed to the created of the hole carrier after doping with Pb, which depends on the density of the material [12, 13]. On the other hand, the ZnO energy band gap is 3.21 eV and slightly increased after doping with Pb to be 3.23 eV, Figure 4. This small change in the energy band gap can also attribute to the increase in the carrier concentration [14].

![Absorption spectrum for ZnO and ZnO-Pb.](image)

**Figure 3.** Absorption spectrum for ZnO and ZnO-Pb.

![$(\alpha h\nu)^2$-hv graphs for the band gap energy (E<sub>g</sub>) values: (a) ZnO films and (b) Pb doped ZnO film.](image)

**Figure 4.** $(\alpha h\nu)^2$-hv graphs for the band gap energy (E<sub>g</sub>) values: (a) ZnO films and (b) Pb doped ZnO film.

Cs-137 used as a gamma source to test the scintillation properties of the films. The samples were used as a scintillation detector for gamma rays, while NaI(Ti) scintillator was used as a reference detector, Figure 5. The results indicated that, undoped ZnO thin film detected the background rays only, while ZnO-Pb could detect the background rays plus to the low energy emitted from the source. The ability of ZnO-Pb to detect better than undoped ZnO can be attributed to the high atomic number and high density of Pb. The background peaks and the small peak from Cs-137 which detected by ZnO-Pb sample and the background peak detected by ZnO sample are shown in Figure 6.
4. Conclusion
ZnO and Pb doped ZnO thin films were prepared by CBD. After studying the characterizations for both films, the results showed that Pb doped ZnO thin films has a better gamma detection property than ZnO thin film. Moreover, ZnO-Pb has larger grain size, more crystallinity, higher absorption peak and higher energy gap. Those properties increased the ability of the film to detect gamma radiation.
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