Irradiation Effects on The Sensitivity of ZnO Thin Films Synthesized on Glass Substrate by Sol-gel Method

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Abstract
This work investigates the structural, optical, and surface properties of ZnO thin films prepared by sol-gel method. The effect on waveguide sensor was examined at different irradiation durations of alpha particles. The X-ray diffraction (XRD) measurements revealed that the crystalline phase of ZnO thin films does not change after irradiation and showed a hexagonal structure of wurtzite type with an orientation toward (002). Moreover, ZnO thin films absorbance was increased with increasing irradiation time, whereas the transmittance was decreased. Additionally, increasing the irradiation time of alpha particles caused an increase in the extinction coefficient and the imaginary part, while the optical energy gap of the ZnO samples was decreased. Finally, the maximum value of sensitivity was 42%, found at 6 min of irradiation duration.

Keywords: Alpha particles, ZnO thin films, Sol-gel.

Introduction
Zinc oxide (ZnO) thin films have been extensively studied due to the high optical and electrical properties, which can be employed in different applications, such as solar cells and sensors [1,2]. Moreover, ZnO has attracted considerable attention as a promising candidate for inexpensive, light weighted, and light-absorbing mediums [3,4]. ZnO thin films can be deposited in several methods such as the evaporation method [5,6], metal organic chemical vapor deposition (MOCVD) [7],...
sputtering [8] and spray pyrolysis [9]. Furthermore, ZnO thin films have been used as sensing materials of reactions of chemicals and gases [10]. The researchers attempted to improve the sensitivity by different methods, such as the alloying, doping, annealing, and irradiation, which increase the sensitivity of ZnO for detecting [11,12]. On the other hand, the irradiation technique has been recently used to enhance the properties of ZnO thin films [13,14,15]. The interaction between the radiation and matter, especially alpha particles, changes the optical and electrical characteristics of the materials [16,17,18]. Alpha irradiation plays a key role to enhance the sensitivity of the ZnO thin film sensor, deposited on glass substrate by sol-gel method, to detect glucose solution.

**Experimental part**

Sol-gel method has attracted the attention of many researchers and used in the deposition of various types of oxides due to essential for the growth along the preferred orientation and give uniform surface. The ZnO thin films were prepared using sol-gel method. Zinc hydroxide acetate (Zn(CH3COO)2HO with a weight of 0.2195 g was dissolved in a mixture of 10 ml ethanol with 90 ml deionized water in room temperature. The mixture was kept under constant stirring for 20 min using magnetic stirrer to completely dissolve. To enhance the homogeneity, 0.5 g polyvinylpyrrolidone (PVP) was added to the mixture which was then kept under constant stirring for 24 hours. Afterwards, the glass substrates were washed with distilled water and methanol and cleaned with ultrasonic waves, respectively. Finally, the ZnO thin films were deposited on 320nm-thick glass substrates and exposed to alpha particles emitted from Am-241 source with energy of 3MeV to increase its sensitivity during the glucose detection process. The energy of alpha particles was calculated based on the following equation:

\[
E_a = E_0 \left[1 - \frac{x}{R}\right]^{2/3}
\]

where \(x\) represents the distance between sample and irradiated source, \(E_0\) is the maximum energy of alpha particles at \(x= 0\) (equal to 5.485 MeV), and \(R\) is the range of alpha particles in the space and can be obtained by SRIM program [19].

**Setup of glucose sensing test**

Figure-1 illustrates the experimental setup of the glucose sensing test applied in the present study. The laser source emitted at 650nm wavelength and a high sensitive avalanche photo detector (EPPT-2000) with a range of 600nm to 1200nm was used. The waveguide, coated with different ZnO thin films, was used as a sensor and placed between the source and the detector of glucose solution. The irradiated waveguide sensor coated with ZnO thin films was exposed to different concentration of glucose solution. The light emitted from the laser source was passing through the surface of the irradiated waveguide during glucose detection. Then, the absorption of the power of light detected by the photo detector was increased, leading finally to increase the sensitivity of the sensors.

![Figure 1: Sensing experimental setup](image-url)
Results and Discussion
X-ray diffraction (XRD) measurement

The XRD patterns of ZnO films, with different thickness, deposited on the glass substrate before and after alpha irradiation are presented in Figure- 2. Alpha particles of 3MeV emitted from Am-241 source were irradiated on the ZnO at different time periods of 2, 4 and 6min. It can be seen from Figure- 2 that the patterns of XRD indicate a hexagonal structure and well C-axis orientation after alpha irradiation [20]. Moreover, the XRD analysis shows a strong peak of ZnO (002) at 2θ=34.24, while the other peaks are found toward the plane orientations (100), (101), (102) and (110) at 2θ =31.73°, 2θ =36.23°, 2θ =48.34° and 2θ =57.32° respectively. Furthermore, the XRD results confirmed that the peaks intensity increases with increasing irradiation time, which could be due to the breaking of bonds on the surface of ZnO thin film. The XRD analysis also revealed that the irradiated ZnO films were still in the crystalline phase and in good homogeneity. The grain size of the deposited ZnO thin films can be calculated using Scherrer equation 1 [ 21] :

\[
D = \frac{\kappa \lambda}{\beta \cos \theta}
\]

where D is the grain size, \(K\) is a constant which is equal to 0.89, \(\lambda\) is the wavelength of XRD (Cu K\(_{α}\) ) which is 1. 54178Å, and \(\beta\) is the full width at half maximum. It can be observed that the grain size increases with the increase in irradiation time, due to the defect of clusters of samples that were formed after irradiation, as well as the interface in the localization of states on the ZnO surface, as shown in Table-1.

![Figure 2-XRD patterns of ZnO thin films before and after irradiation at time periods of 2min, 4min , and 6min.](image)

<table>
<thead>
<tr>
<th>Irradiation time (min)</th>
<th>Grain Size (nm)</th>
<th>FWHM (radian)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>18.34</td>
<td>0.52</td>
</tr>
<tr>
<td>2</td>
<td>22.67</td>
<td>0.45</td>
</tr>
<tr>
<td>4</td>
<td>28.65</td>
<td>0.43</td>
</tr>
<tr>
<td>6</td>
<td>32.22</td>
<td>0.39</td>
</tr>
</tbody>
</table>

Optical properties

The optical properties before and after irradiation of ZnO thin films were studied. Figure- 3 shows the absorption spectra as a function of the wavelength. The results indicate that the absorbance of ZnO thin films was increased for all irradiation times. This might be due to the breaking of bonds on the ZnO surface after alpha irradiation, which leads to increase the absorbance mechanisms. Increasing of irradiation time leads to formation defects. These defects are considered as an interesting factor in
atomically manipulating photonic devices as they play important roles in the optical properties of a material by introducing absorption bands or color centers [22]. The ZnO thin films were still in crystalline phase and did not change in crystal structure after irradiation. Furthermore, the diffraction peak of the ZnO thin film was oriented along the C-axis (002) after irradiation. Figure- 4 shows the results of transmittance against wavelength of the samples. The transmittance of ZnO thin films was decreased for all irradiation times in comparison with the non-irradiated ZnO samples. This is related to defects on the rough surface of ZnO after alpha irradiation, leading to a noticeable decrease in the transmittance [23]. Figure-5 illustrates the absorption coefficient of non-irradiated and irradiated ZnO thin films as functions of wavelength. The absorption coefficient increases as the irradiation time increases for all the samples. This could be due to the change in the crystal structure of ZnO after irradiation [24]. Moreover, all the irradiated ZnO samples exhibited high magnitudes of the absorption coefficient, which could lead to the direct transition band gap of ZnO. These results are consistent with those reported by Muhammad [25]. Figure- 6 shows the relationship between $(\alpha h\nu)^2$ and photon energy ($h\nu$) of non-irradiated and irradiated ZnO thin films. The energy gap ($E_g$) can be calculated via Tauc formula:

$$a h\nu = B(h\nu - E_g)^r$$

where $B$ is constant, $\alpha$ is the absorption coefficient, $h\nu$ is photon energy, and $r$ takes the value of 1/2 at direct transition and 2 at indirect transition. The results show that the optical energy gap decreases with irradiation time, with values of 3.26, 3.21, 3.16, 3.1 eV for irradiation times of 2, 4 and 6 min, respectively. This behavior can be justified by generating extra energy states after irradiation as well as interface band transition at localized states in the energy gap. The values of calculated energy gap are in agreement with those reported by Khan [26]. The results of optical properties after irradiation are attributed to the defects in ZnO thin films. Most of these defects are created by the localization of oxygen vacancies and change of electronic structure [27]. Furthermore, ionized radiation through thin films causes the loss of most of the energy, due to non-elastic collision of radiation with electrons on ZnO surface or losing of energy due to elastic collision with atomic nucleation of material [28].

In addition, the extinction coefficient of ZnO thin films, which includes both the real and imaginary parts of dielectric constant, was studied in the wavelengths range of 300-800 nm. The extinction coefficient ($k_o$), real part ($\varepsilon_r$), and imaginary part ($\varepsilon_i$) was decreased with increasing wavelength for all samples. The values of extinction coefficient and imaginary part of irradiated samples were larger than those of the non-irradiated samples. Moreover, the values of the real part were smaller than those before irradiation, as shown Figure-9 [29, 30].
Figure 7- Extinction coefficient versus $\lambda u$ for different irradiation times

Figure 8- Real part $\varepsilon_r$ versus $\lambda u$ for different irradiation times.

Figure 9- Imaginary part $\varepsilon_i$ versus $\lambda u$ for different irradiation times

Scanning Electron Microscopy (SEM)

Figure-10 shows the SEM images of non-irradiated and irradiated ZnO thin films for different times (2, 4, and 6 min). The images reveal that the grain size evidently increases when the irradiation time is increased [31]. This could be due to the defects formed on the ZnO surface caused by alpha particles, which lead to the formation of large-sized nuclei, especially at the longer irradiation time. Therefore, this can open the possibility for manipulating the crystallinity, shape, and size of the grains via controlling irradiation time [32].
Waveguide sensor for glucose detection

The prepared samples were used in the detection of various concentrations of glucose solution. Figure 11 shows the normalized power of the light passing through the ZnO surface versus glucose concentration, before and after irradiation. As can be observed, the normalize power was increased as the time irradiation increased during the detection processes. This could be due to defects, such as those in color centers, on the ZnO surface after irradiation which lead to increase the absorbance process. Additionally, the response of ZnO to glucose solution, for all irradiation times, was found to be linear. The gradients of the lines were employed to give the sensitivity of ZnO. The results showed that the sensitivity of the ZnO for glucose detection increases as the irradiation time is increased. The maximum sensitivity of the ZnO is approximately 42% at 6 min irradiation time, whereas the minimum sensitivity is approximately 25% and 14% at 4 and 2 min, respectively. Figure 12 shows the linear fitting of the ZnO sensitivity with various ZnO thicknesses. The R-square value for the linear line fitting is 0.98221, which indicates a good linearity. The proposed ZnO sensor can be implemented for glucose detection due to the good line fitting characteristics.

Figure 11- Normalized power versus glucose concentration of ZnO after Alpha irradiation

Figure 12-Sensitivity percentage versus irradiation time of ZnO waveguide sensors
Conclusions

Waveguide sensor was designed and fabricated by depositing ZnO thin films on glass substrates using sol-gel method and then irradiated by alpha particles. Structural, optical, and morphological properties of the ZnO thin films were examined and analyzed. The X-ray diffraction patterns revealed a slight shift, while the crystalline size increases, with increasing irradiation time. Moreover, the band gap slightly decreases after irradiation. Furthermore, the waveguide sensor showed an improved sensitivity to detect glucose solution with increasing irradiation time. The higher value of sensitivity was 42% dB/con at irradiation time of 6min, while the value before irradiation was found to be 9.4% dB/con. The irradiation process increased the absorbance of the sensor, which in turn increased the sensitivity of waveguide sensor. This may be used to monitor liquid leakages in different fields, such as fuel stations, wells, and oil pipes.

References


