Influence of Thickness on Optical Properties of ZnO Thin Films prepared by Radio Frequency Sputtering Technique

1*Abdullahi, S., 2Momoh, M., 3Moreh, A.U., 4Isah, K.U. and 5Argungu, G.M.
1,2,3,5Department of Physics, Usman Danfodiyo University, Sokoto Nigeria.
4Department of Physics, Federal University of Technology Minna, Niger state Nigeria.

Abstract- Zinc oxide (ZnO) thin films of 75.5nm and 130.5nm were deposited at room temperature onto chemically and ultrasonically cleaned corning glass substrate by radio frequency technique and annealed at 150°C under nitrogen atmosphere for 60 minutes. The Zinc target of purity 99.99%, diameter 40mm and thickness 6.35mm was used. The optical properties of the films were ascertained by employing UV-VIS-NIR spectrophotometer (model AVASPEC 2048 MODEL). Influence of the thickness of the films on the optical properties was studied by keeping other deposition parameters constant. The optical transmittance spectra revealed an average transmittance of 81.49% and 84.26% for the 75.5nm and 130nm respectively. The band gap of the films was found to decreases with increase in thickness of the films. The band gap energy (E_g) is in the range of 3.28eV to 3.31eV respectively. Other optical parameters such as Absorbance, Reflectance and Urbach energy were also found to be thickness dependent. These effects of thickness on optical properties of ZnO thin films are suitable for solar cell applications.

Key words: ZnO, Sputtering, Film Thickness, Band Gap Energy, Urbach Energy

1.0 INTRODUCTION
ZINC OXIDE (ZnO) is an attractive material for a large variety of applications such as microelectronics, piezoelectric, optoelectronic and photovoltaic devices. It is a wide-band gap oxide semiconductor with a direct energy gap of about 3.37 eV. Zinc oxide has emerged as one of the most promising materials, due to its optical and electrical properties associated with high chemical and mechanical stability. This makes it a lower cost material when compared to the most currently used transparent conductive oxide materials such as ITO (indium tin oxide) and SnO2 (Bensmaine et al., 2007). The synthesis and characterization of polycrystalline materials have attracted much attention not only because of their exceptional properties but also due to their structure, temperature dependent properties and great potential for many technological applications. Recently there has been an increase in research and development of II-VI materials that are widely used for glazing windows, solar energy collectors, and low cost flat panel solar cells. These films offer a large number of applications in solid-state device technologies such as the target material for television cameras, microwave devices, switching devices, infrared detectors, diodes and Hall effect devices (Ziul et al., 2010). Thin films have also exhibited a wide variety of applications in environmental engineering, catalysis and gas sensor systems because they can be fabricated in small and large-scale dimensions. During the last years, several deposition techniques for thins film have been developed and studied, such as, ion-beam-assisted deposition, chemical vapor deposition CVD (Buba and Adelabu., 2010), pulsed-enhanced chemical vapor deposition (PECVD) (Tabenskaya et al., 1995), spray pyrolysis (Godbole et al., 2011), molecular beam epitaxy (MBE), sol-gel processing (Singh et al., 2009), reactive DC sputtering and magnetron sputtering technique which is one of the most widely used due to its reproductibility and efficiency (Bingyao and Weidon, 2008; Chaoyang et al., 2007; Chongmu et al., 2008; Jae et al., 2003). The objective of this paper is to study the thickness effect on ZnO thin films grown by radio frequency (Rf) magnetron sputtering at 150°C considering the fact that a lot of researchers studied such effect on films but at high annealing temperatures. Radio frequency magnetron sputtering exhibits interesting advantages such as the low substrate temperature, good adhesion of the films on the substrates, and a high deposition rate.

2.0 EXPERIMENTAL
Zinc oxide films deposited by RF magnetron sputtering are very sensitive to the deposition parameters; these should therefore be optimized in order to obtain highly orientated ZnO films. Fig. 1 shows the apparatus of RF magnetron sputtering. It consists of a cylindrical plasma chamber, sputtering gas inlet, vacuum pump etc.

The distance between the cathode and the substrate holder was 100 mm. The deposition chamber was pumped down to a base pressure of 5x10-7mbar by a turbo molecular pump prior to the introduction of the argon/oxygen gas mixture for ZnO thin film production. The pressure was fixed at 4.6x10-3mbar and the time of
deposition was 60 minutes. The RF power was set at 60 watts.

The substrate holder temperature was kept at room temperature and the oxygen percentage in Ar/O2 gas mixture was kept at a ratio of ZnO films were deposited by RF magnetron system on corning glass substrates. The Zinc target (purity 99.99%) diameter was 40 mm and 6.35 mm thick. of 1 to 1. The details of the deposition conditions were summarized in the table 1. After the deposition, the two samples were annealed under nitrogen atmosphere at 150°C for 60 minutes by the use of carbolite horizontal furnace. To determine the optical characteristics of the films, UV-VIS-NIR spectrophotometer with a wavelength range of 180-1200nm was used. The transmittance of the samples was calculated using equation (1)

\[ T = \frac{1}{10^A} \]  

(1)

Where A is absorbance.

In order to determine the optical band gap of the films, equation (2) was employed

\[ a(h\nu) = B(h\nu - E_g)^m \]  

(2)

Where B is a constant, m value is respectively 1/2 and 2 for direct and indirect transitions. The variation of (ah\nu)² with photon energy h\nu of ZnO thin films. The intercepts (extrapolations) of these plots (straight lines) on the energy axis give the energy band gaps.

To calculate Urbach energy, it is assumed that the absorption coefficient near the band edge shows an exponential dependence on photon energy and this dependence is given as:

\[ \alpha = \alpha_0 \exp\left(\frac{h\nu}{E_u}\right) \]  

(3)

where \(\alpha_0\) is a constant and \(E_u\) is Urbach energy interpreted as the width of the tails of localized states, associated with the amorphous state, in the forbidden gap. The Urbach energy was obtained from the plot of photon energy vs. ln (\(\alpha\)).

### Table 1: Details of Deposition Parameters Used in This Study

<table>
<thead>
<tr>
<th>S/N</th>
<th>Parameter</th>
<th>Deposition Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Substrate</td>
<td>Corning Glass 7059</td>
</tr>
<tr>
<td>2</td>
<td>Target/Target</td>
<td>ZnO Ceramic target</td>
</tr>
<tr>
<td>3</td>
<td>Diameter</td>
<td>4N/4cm</td>
</tr>
<tr>
<td>4</td>
<td>Substrate/Target</td>
<td>7 cm</td>
</tr>
<tr>
<td>5</td>
<td>Distance</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Annealing Environment</td>
<td>Nitrogen/Air</td>
</tr>
<tr>
<td>7</td>
<td>Annealing set points</td>
<td>150°C</td>
</tr>
<tr>
<td>8</td>
<td>Annealing Ramp rate</td>
<td>10°C/min.</td>
</tr>
<tr>
<td>9</td>
<td>Annealing period</td>
<td>60 minutes</td>
</tr>
<tr>
<td>10</td>
<td>Film Thickness</td>
<td>75.5nm and 130.5nm</td>
</tr>
<tr>
<td>11</td>
<td>Deposition pressure</td>
<td>4.6 x10⁻³ mbar</td>
</tr>
<tr>
<td>12</td>
<td>Argon/Oxygen flow rate</td>
<td>1 standard cubic centimeter</td>
</tr>
<tr>
<td>13</td>
<td>Substrate temp</td>
<td>Room Temp.</td>
</tr>
<tr>
<td>14</td>
<td>Rf power</td>
<td>60 W.</td>
</tr>
<tr>
<td>15</td>
<td>Deposition time/period</td>
<td>1 Hour</td>
</tr>
</tbody>
</table>

### 3.0 Results and Discussion

#### 3.1 Transmittance

Fig. 2 shows the wavelength dependence of optical transmittance spectra of ZnO thin films deposited on corning glass by reactive RF magnetron sputtering at different thicknesses (75.5nm and 130.5nm). Both films sputtered at ambient temperature shows an average transmittance of 81.49% and 84.26% respectively in the wavelength range from 350 to 750 nm. It is clear from Figure (2), that the optical transmittance increases slightly with increasing of film thickness. Similar trend was reported by Chitra et al., (2013) and Asel and Ali (2015). The sharp absorption edge was observed at the wavelength of about 400nm and shifted towards higher wavelengths. Displacement of the absorption edge is due to Fermi level moving into the conduction band with the increase in carrier concentration according to the theory of Burstein-Moss effect (Guilleen and Herrero, 2010).

#### 3.2 Reflectance

Figure 3 depicts the plot of reflectance versus wavelength for ZnO thin films of thicknesses 75.5nm and 130.5nm. The film having a thickness of 75.5nm shows an average reflectance of 38.78%, while 130.5nm
thickness exhibits an average reflectance of 30.76%. These indicate that as thickness increase thin film become less reflective.

3.4 Energy band gap

The variation of \((\alpha h\nu)^2\) with photon energy \(h\nu\) of ZnO thin films of 75.5nm and 130.5nm is shown in Fig. 5. It has been observed that the plots of \((\alpha h\nu)^2\) versus \(h\nu\) are linear over a wide range of photon energies indicating the direct type of transitions. The direct band gaps (\(E_g\)) of 75.5nm and 130.5nm thin films were determined at 3.31eV and 3.28eV, respectively. This implies that band gap energy decreases with increase in ZnO film thickness.

3.5 Urbach energy (\(E_u\))

The photon energy vs. \(\ln(\alpha)\) plots for ZnO thin films of 75.5nm and 130.5nm are shown in Figure 6. The values of \(E_u\) obtained from this figure are given in Table II. It is believed that the exponential dependence of \(\alpha\) on photon energy may arise from random fluctuations of the internal fields associated with the structural disorder in many materials. The Urbach energy (\(E_u\)) value of 130.5nm thick sample is (2.4eV) higher than that of 75.5nm thick sample which has 2.3eV. This suggests that the film thickness increases the Urbach energy referred the width of the band tail. This is probably due to the structural disorders in the samples and increase in the degree of amorphous character (Tariq and Almushtak, 2010). The change in the value of \(E_u\) is also associated with the breakdown of crystal structure of
ZnO at high annealing temperature (Ezema and Nwankwo, 2010).

![Graph showing ln α vs hv for 75.5nm and 130.5nm thick thin ZnO](image)

**Fig. 6:** ln α vs. hv for 75.5nm and 130.5nm thick thin

<table>
<thead>
<tr>
<th>Sample thickness (nm)</th>
<th>Urbach energy (E_u)</th>
</tr>
</thead>
<tbody>
<tr>
<td>75.5nm</td>
<td>2.3eV</td>
</tr>
<tr>
<td>130.5nm</td>
<td>2.4eV</td>
</tr>
</tbody>
</table>

**TABLE II: VALUES OF URBACH ENERGY (E_u)**

### 4.0 CONCLUSION

ZnO thin films of different thicknesses were prepared on glass substrate by RF sputtering technique. Changes in the optical properties of these films were studied with respect to their thickness. The band gap energy (E_g) was found to increase with thickness. Other optical parameters such as Absorbance, Reflectance and Urbach energy were also found to be thickness dependent. These effects of thickness on optical properties of RF sputtered ZnO thin films are suitable for solar cell applications.

**ACKNOWLEDGMENT**

The authors are indebted to the World bank through the Science and Technology Post Basic (STEP-B) of the Usman Danfodiyo University Sokoto.

**REFERENCES**


Guilleen, C. and Herrero, J. (2010). Optical,electrical and structural characteristics of Al:ZnO thin films with various thickness deposited by DC sputtering at room temperature and annealed in air or vacuum. Vacuum.vol. 84 pp.924-929.


