Academic Journal of Science and Technology,
Aug, 2025, vol.6, No.1, p277-282

The Effect of Temperature on the Optical and Electrical Properties of ZnO Thin Films

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ABSTRACT

ZnO thin films were deposited on stainless steel substrates using the immersion method by immersing the substrates in a sol-gel coating solution at temperatures of 70 °C and 80 °C for varying durations of (1, 2, 3, and 4minutes). The results indicated that increasing the immersion time significantly influenced the film thickness. Optical measurements showed that transmittance at a wavelength of 350nm increased with higher deposition temperatures. Additionally, the Urbach tail energy increased with temperature, whereas the band gap (Eg) decreased markedly. Furthermore, the electrical conductivity of the ZnO films improved with increased temperature and immersion time.

KEYWORDS: *: ZnO, Optical Properties, Electrical properties, Thin films.*

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1. INTRODUCTION

Zinc oxide is a transparent conducting oxide with wide applications in industrial and technical fields. It is non-toxic, poorly soluble in water, has high transmittance to visible light, good reflectivity in the infrared region, and good n-type electrical conductivity. Zinc oxide (ZnO) is classified as a semiconductor with a relatively large direct bandgap of 3.37eV at room temperature, a high electron-hole bonding energy of 60meV, and n-type electrical conductivity in most cases [1,2]. ZnO films have high transmittance in the visible and infrared regions, while transmittance decreases in the ultraviolet region due to the large bandgap that allows most visible light to pass through [3,4].

These physical properties enable zinc oxide to be used in many important industrial applications, such as solar cells, lasers, photo detectors, variable resistors, gas sensors, photo catalysis [5], and blue and ultraviolet light-emitting devices. Many previous studies have been conducted on zinc oxide (ZnO) films and their potential use in many important applications. In 1982, Coporaletti [6] prepared ZnO films using the bias sputtering technique. Optical results showed that the film exhibits high transmittance and an energy gap of 3.3eV, with an absorption edge in the range (340-380nm) for films with thicknesses ranging from (0.3 to 0.56µm). In 1986, Chartier and his colleagues [7] were able to prepare ZnO films with good adhesion on glass substrates using the decomposition method in an aqueous solution. A study of the optical absorption spectrum revealed that the films had a high transmittance ranging from 90-95% in the visible region, with an absorption edge at a wavelength of 380 nm. In 2004, Norton and his colleagues [8] prepared a pure ZnO film using pulsed laser deposition on Al_2O_3 substrates at a temperature of 600°C, where a concentration of charge carriers (electrons) was obtained in the range of (7.5×10^{15}-$1.5\times10^{20}cm^{-3}$).

The properties of deposited ZnO films are influenced by several factors, most notably substrate temperature, deposition time, and other conditions that control the quality and properties of the resulting film. In this paper, we examine the effect of these factors on the optical properties of ZnO films prepared using the dip-deposition technique

2. Experimental part:

Thin films of zinc oxide (ZnO) were prepared on steel substrates using the immersion deposition technique. This technique is considered one of the simplest and easiest methods for depositing thin films. Zinc phosphate ($Zn_3(PO_4)_2$) was used as the zinc source in the solution used in the deposition process. To ensure the solution was dissolved, a magnetic stirrer was used to mix the solution for a sufficient period of time, approximately half an hour (30min), until the material was completely dissolved at room temperature. Before the deposition process, we cleaned and prepared the substrates by removing impurities and dust from their surfaces by washing them thoroughly with distilled water. They were then immersed in sulfuric acid (H_2SO_4) to activate the substrate surface and remove any possible rust. Finally, to remove excess reactants and any residual acid on the substrate surface, the substrates were thoroughly washed with distilled water again, thus preparing the substrates for the coating (film deposition) process. These substrates were immersed in a (sol-gol) solution at temperatures of 70 and 80°C for periods of (1, 2, 3, and 4minutes). After the immersion process was completed, the slides were suspended to cool slowly to complete the crystal growth process and increase the crystal grain size.

This resulted in thin films of varying thicknesses. A (GE DMS 2 Thickness Gauge) was used to measure the thickness. The surface morphology of the prepared films was studied using a scanning electron microscope (SEM), and the crystal structure was studied using X-ray diffraction (XRD). X-ray diffraction results showed that the prepared films have a polycrystalline structure and are of the (Hexagonal Wurtizite) type. Absorbance and transmittance measurements were recorded using (UV-visible) spectroscopy. A (Fischer Scope mms) was used to measure electrical conductivity.

3. Results and Measurements:

3.1. Thickness Measurement:

The thickness measurement was performed using a (GE DMS 2 Thickness Gauge). The obtained results showed that the thickness ranged between (9.12 µm and 19.02 µm) at a deposition temperature of 70°C over deposition times of (1-4min), as shown in Table (1). This thickness increased to (25.28 µm) when the deposition temperature increased to 80°C for a deposition time of (4min), as shown in Table (2). The reason for this increase is the increased crystal growth of the films deposited on the metal surface.

Table 1. Thickness of thin films at a temperature of 70°C.

Time (Sec)	Thickness (µm)
1	9.12
2	12.68
3	16.22
4	19.02

Table 2. Thickness of thin films at a temperature of 80°C.

Time (Sec)	Thickness (µm)
1	13.12
2	16
3	20.34
4	25.28

3.2. Transmittance:

The transmittance measurements of the prepared ZnO samples were studied over the wavelength range (300

to 900nm). Figure (1) shows the graph of the transmittance as a function of wavelength for a ZnO sample at 25°C.

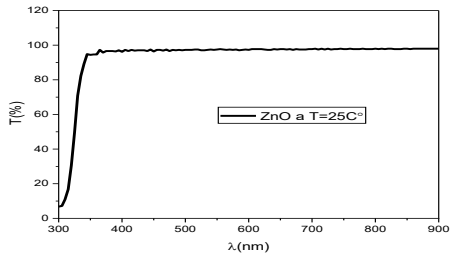


Figure 1. shows the transmittance as a function of wavelength at a 25°C.

Figure (2) shows the transmittance curve of a ZnO sample obtained at 70°C, which shows that the transmittance is at its lowest in the UV wavelength region at 300-320nm, which indicates porosity (gaps). Then, the transmittance increases sharply at 320-350 nm. This region is called the fundamental absorption edge, and this indicates the high absorption of photons incident on the film. Due to the high absorption of incident photons by the valence band electrons, the electrons move down the conduction band, which in turn indicates that the material is a large energy gap semiconductor.

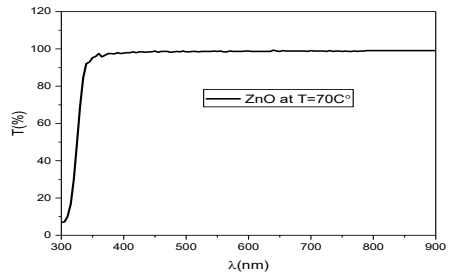


Figure 2. shows the transmittance as a function of wavelength at a of 70°C.

These results also showed that the zinc oxide films have high transmittance in the visible and near infrared region and became almost constant at (800 nm). We note from these results that the transmittance increased significantly when the deposition temperature increased from 25°C to 80°C. The average transmittance value was about 92% at 25°C and increased to about 96% at 80°C within the visible spectrum region Figure (3). The high transmittance value we obtained can be attributed to the smoothness and relative homogeneity of the prepared films' surface, which reduces the scattering of incident light. The transmittance spectrum of the studied samples shows an optical behavior similar to the transparent conductive oxide (TCO) group, which indicates that the films are suitable for solar cell applications because the spectrally active region in the cells lies in the visible region (9).

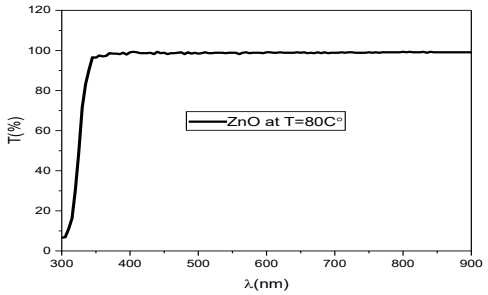
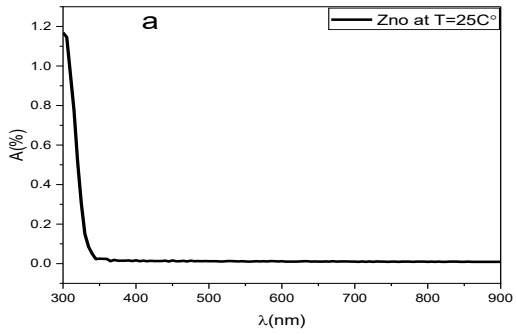


Figure 3. shows the transmittance as a function of wavelength at a of 80°C.

3.3. Absorption:

Figure (4(a,b and c)) shows the graph of the absorption as a function of wavelength from (300 to 900nm) at different temperatures. We can see from these figures that the absorption spectrum is greatest at the negative absorption edge for short wavelengths. This means that the prepared films exhibit high absorption at short wavelengths, which fall within the visible region, and then decrease with increasing wavelengths. This is explained by the fact that the incident photon was unable to excite the electron and transfer it from the valence band to the conduction band, since the energy of the incident photon is less than the energy gap value of the semiconductor. Therefore, the absorption decreases with increasing wavelengths [10]. The absorption at high energies is due to negative absorption processes arising from the transfer of electrons from the valence band to the conduction band [11]. We also note that the absorption decreases with increasing deposition temperature in the infrared and visible regions. By comparing the average absorbance values, we find that they decrease with increasing deposition temperature, as their value ranges between (0.025%, 0.016%) when the temperature rises above (°C).



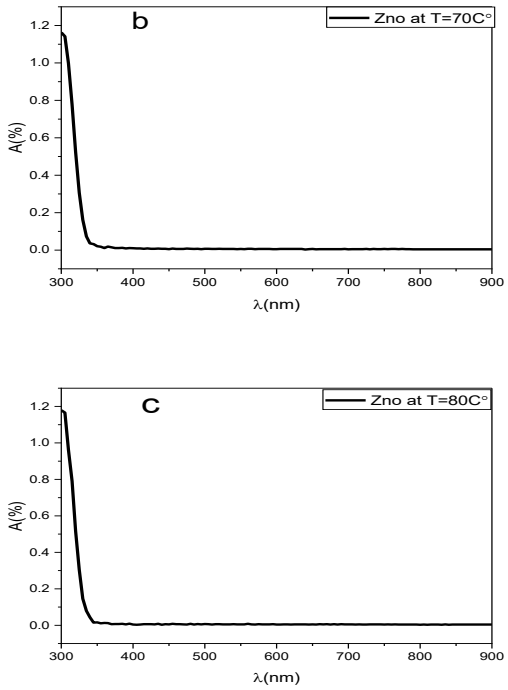


Figure 4. shows the absorption as a function of wavelength at (a. 25C̊, b. 70C̊ and c.80C̊).

3.4. Energy gap:

The energy gap was calculated according to the equation $(\alpha h\upsilon)^2 = B(h\upsilon - Eg)$, where B is a constant, Eg(ev) is the energy gap, and $h\upsilon(ev)$ is the photon energy. The energy gap values for pure ZnO films prepared at different substrate temperatures were determined by plotting the curve of $(\alpha h\upsilon)^2$ as a function of photon energy $(h\upsilon)$ and drawing a tangent to the straight part of the curve. The extension of this tangent intersects with the photon energy axis $(h\upsilon)$. This intersection point represents the value of the optical energy gap for the allowed transitions.

Figure (5) shows the optical energy gap for a ZnO sample deposited at 25°C. This curve shows that the point of intersection of the tangent with $(h\upsilon)$, which represents the energy gap, was approximately 3.7 eV.

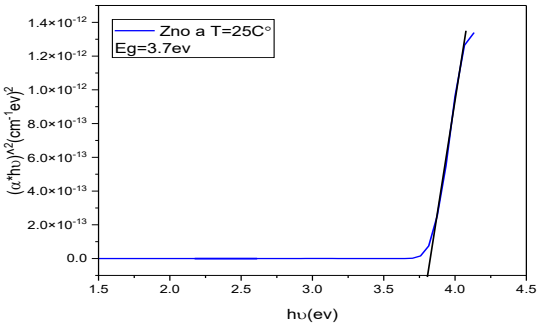


Figure 5. The optical energy gap curve of a ZnO sample deposited at a 25°C̊

We note that this value of the energy gap decreases with increasing temperature, as its values ranged from approximately 3.48eV at a temperature of 70°C to 3.25eV at a deposition temperature of 80°C, as shown in the figures (5 and 6 respectively). The decrease in the energy gap with increasing temperature is explained by the increase in the number of electrons moving to the conduction band, thus forming donor levels within the energy gap near the conduction band, which contributes to the absorption of photons with low energies [12], and also the sample resistance decreases with increasing temperature.

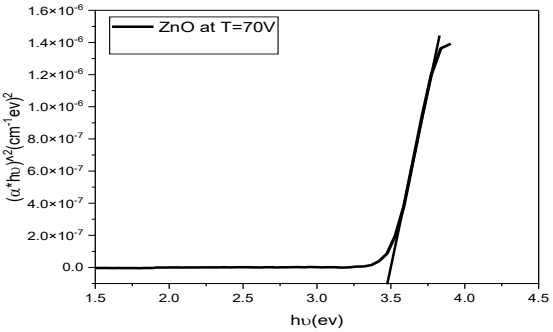


Figure 6. The optical energy gap curve of a ZnO sample deposited at a 70°C̊.

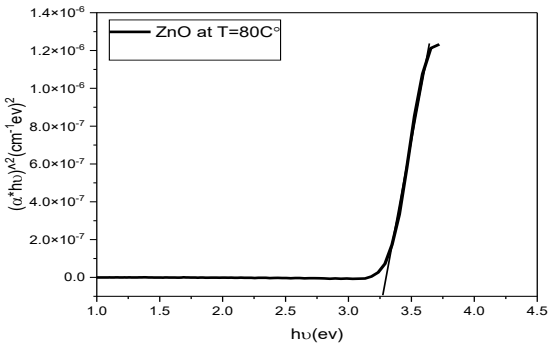


Figure 7. The optical energy gap curve of a ZnO sample deposited at a 80°C̊

3.5. Urbach tail energy:

Urbach tails are defined as the number of levels within the optical energy gap, which is equal to the reciprocal of the slope of the relationship between $(lin(\alpha))$ and $(h\upsilon)$. The Urbach energy for zinc oxide films was calculated using the mathematical equation:

$$\alpha = \alpha_{\circ} exp \frac{(h\upsilon)}{E_U}$$

This was done by taking the reciprocal of the slope of the linear region in the plot of $lin(\alpha)$ versus photon energy $(h\upsilon)$, as shown in Figures (8), (9), and (10). This linear region corresponds to the absorption edge, and its slope reflects the degree of disorder in the material.

We note that, the Urbach energy value increases with increasing temperature, as its value ranges between (0.142eV at 25°Cand 0.157eV at 80°C). The increase in the Urbach energy value indicates that, the width of the local levels or the number of local energy levels in the optical energy gap has increased with increasing temperature, and thus the number of Urbach tails increases, which leads to a decrease in the optical energy gap, as it was found that the optical behavior of the Urbach tail energy value is opposite to the optical behavior of the optical energy gap value for all films [9].

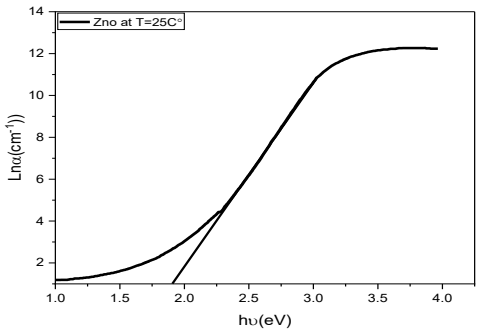


Figure 8. shows $(lin(\alpha))$ versus $(h\upsilon)$ at 25°C.

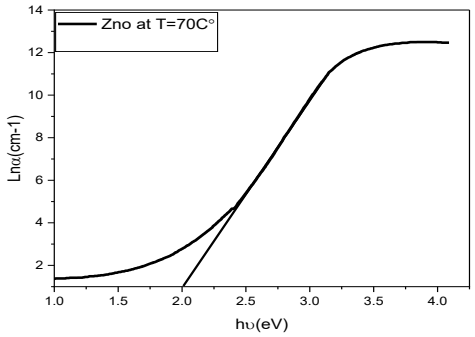


Figure 9. shows $(lin(\alpha))$ versus $(h\upsilon)$ at 70°C.

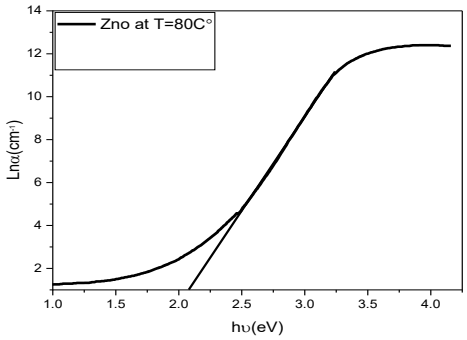


Figure 10. shows $(lin(\alpha))$ versus $(h\upsilon)$ at 80°C

3.6. Electrical conductivity (σ):

Electrons and holes in semiconductors are charge carriers, as they are responsible for transporting electric current within the material. The density of these carriers is an important property of semiconductors, as it determines their electrical conductivity. We measured the electrical conductivity values using a Fisher scope mms. The results shown in Tables (3 and 4) show that the electrical conductivity increases with increasing temperature. This is explained by the fact that increasing temperature causes some bonds to break and electrons to be released, leaving spaces called positive holes. This leads to an increase in the number of free electrons and positive holes, which results in an increase in electrical conductivity [9].

Table 3. shows the electrical conductivity at 70°C and different times.

Time(min)	Conductivity (ms/m)
1	10.79
2	12.25
3	13.304
4	14.53

Table 4. shows the electrical conductivity at 80°C and different times.

Time(min)	Conductivity (ms/m)
1	12.312
2	13.42
3	14.821
4	16.163

CONCLUSION

Zinc oxide thin films were successfully fabricated using the dip-deposition technique. The experimental results confirmed that the structural and optical properties of the prepared films were consistent with those of standard zinc oxide materials. It was observed that the film thickness increased with increasing deposition temperatures and immersion times in the sol-gel solution. The optical transmittance spectra showed similar patterns across all samples, with transmittance increasing with increasing deposition temperature. In contrast, the absorption spectra indicated an increase in the absorption coefficient with increasing temperature. The energy gap values showed a decrease with increasing temperature. In addition, the Urbach energy was found to increase with temperature. Electrical characterization revealed that the conductivity

of the ZnO films improved with increasing immersion times.

These results demonstrate that temperature and immersion duration significantly influence the structural, optical, and electrical properties of ZnO thin films deposited by the immersion method.

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