

A.C. Conductivity and Dielectric Study of ZnO Thin Films

Ziad T. Khodair, Nabeel A. Bakr, Nedhal A. Mahmood, Kadhim A. Adem

Abstract— Aluminum/ zinc oxide/Aluminum polycrystalline thin films (Al/ZnO/Al) with thickness of 100 nm have been prepared at room temperature by thermal evaporation technique. The films have been annealed at different annealing temperatures (373,423,473) °K. A.C. conductivity $\sigma_{a.c.}(\omega)$ of the prepared thin films has been measured in the frequency range of (0.1 - 400) KHz at room temperature. The results reveal that $\sigma_{a.c.}(\omega)$ obey the relation $\sigma_{a.c.}(\omega) = A\omega^s$ and the exponent (s) was found to decrease by increasing the temperature. The values of (s) of the investigated thin films lie between 0.77 and 0.86. The data were analyzed in terms of different models of A.C. conduction. It was found that the correlated barrier hopping (C.B.H.) is the dominant conduction mechanism. The dependence of dielectric constant (ϵ') and loss factor ($\tan \delta$) on frequency and annealing temperatures have been analyzed too.

Index Terms— A.C. Conductivity, Dielectric Constant, Electrical Properties, ZnO thin films.

I. INTRODUCTION

In the group of II-VI compound semiconductors, Zinc oxide (ZnO) has received intense attention due to its remarkable combination of physical and optical properties. Its wide band gap (3.37 eV at room temperature), high exciton binding energy (60 meV), and its diverse growth morphologies, make ZnO a key material in the fields of nanotechnology and wide band-gap semiconductors [1]. ZnO is one of the most prominent metal oxide semiconductors. It is an n-type semiconductor of hexagonal (wurtzite) structure with a direct energy wide band gap of about 3.37 eV at room temperature [2]. It is exhibited unique electrical properties which makes it potentially useful for applications such as in UV light emitting diodes (LEDs)[3], photovoltaic devices[4], optical wave guides[5], gas sensors, etc [6].

Also, ZnO thin films have interested as transparent conductor, because of its wide band gap and high transmission in the visible range, as well as ZnO thin films can take place of SnO₂ and ITO because of their electrical and optical properties and its excellent stability which has been mentioned widely[7,8]. In the present study, the A.C. conductivity, dielectric constant (ϵ') and loss factor ($\tan \delta$) of

ZnO thin films were measured as a function of frequency and temperature with a view to determine the possible conduction mechanism. The effect of annealing temperatures on both conductivity and dielectric constant has been also discussed.

II. EXPERIMENTAL PROCEDURE

Thin films of the ZnO were prepared by thermal evaporation technique at high pressure about 10⁻⁶ mbar using Balser coating system (E-306). The measurement was carried out on sandwich structure form. Metal (Aluminum) thick film (2000Å) was evaporated on glass substrate as a base electrode followed by the sample and finally the second gold electrode was evaporated. Effective area was about 0.925 cm². Hp. 4274 A and 4275 programmable automatic RCL (model, Hewlett. Packard) meter was used for Measurements A.C. conductivity ($\sigma_{a.c.}$), dielectric constant (ϵ') (real and imaginary parts) and loss factor ($\tan \delta$) of the investigated thin films were obtained in the frequency range of (0.1 - 400) kHz. Samples were treated at different annealing temperatures.

III. RESULTS AND DISCUSSION

Figure (1) shows the variation of capacitance as a function of angular frequency at different annealing temperatures for ZnO thin films. Noticeably, the decrease in capacitance is very rapid in the initial frequency. This decrease can be accounted by increasing in the space charge region at the electrodes. Also, it can be seen that the capacitance increases as the annealing temperature increases.

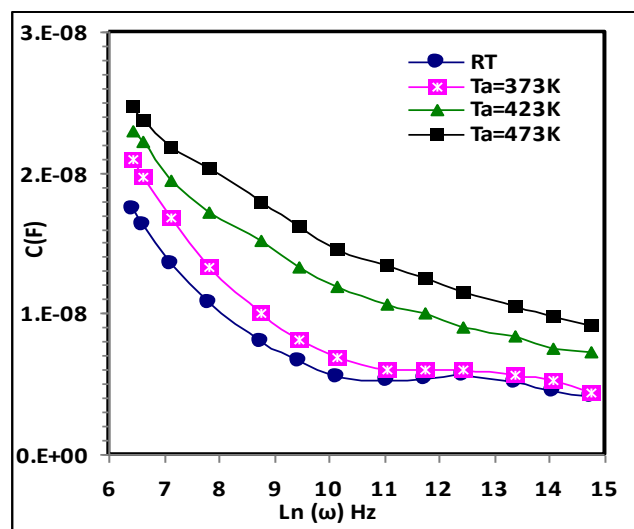


Fig.(1) Variation of capacitance with angular frequency for ZnO thin films at different annealing temperatures

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Ziad T. Khodair, Department of Physics, College of Science, University of Diyala, Diyala, Iraq.

Nabeel A. Bakr, Department of Physics, College of Science, University of Diyala, Diyala, Iraq.

Nedhal A. Mahmood, Department of Physics, College of Science, University of Diyala, Diyala, Iraq.

Kadhim A. Adem, Department of Physics, College of Science, University of Baghdad, Baghdad, Iraq.

Figure (2) shows the dependence of $\text{Ln}(\sigma_{a.c})$ on $\text{Ln}(\omega)$ for thermal evaporated ZnO thin films in the frequency range of (0.1 - 400) kHz at RT and annealed at different annealing temperatures (373,423,473) °K.

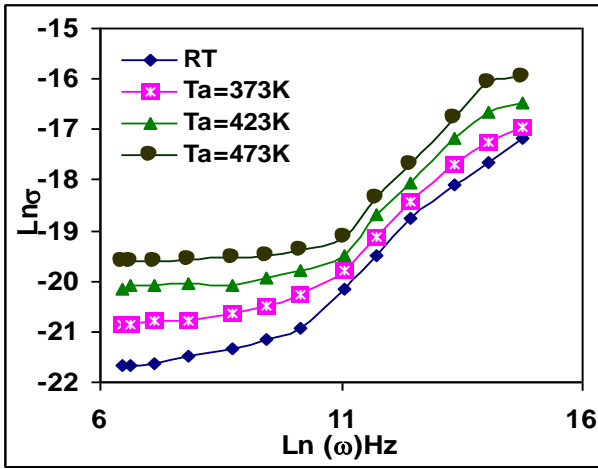


Fig.(2) Frequency dependence of A.C. Conductivity $\sigma_{a.c}$. for ZnO thin films at various annealing temperatures.

All samples follow a common pattern where $\sigma_{a.c}$ increases with increasing of angular frequency. Obtained data reveal that the A.C. conductivity follows the well known relation [9]:

$$\sigma_{a.c}(\omega) = A \omega^s \dots\dots\dots(1)$$

where A is constant and $s (\leq 1)$ is the frequency exponent. The phenomenon has been ascribed to relaxations caused by the motion of electrons or atoms. Such motion can involve hopping or tunneling between equilibrium sites [10].

The behavior of the exponent (s) with temperature can help in determining the possible of conduction mechanism and can be estimate from the slope of the curves plotted between $\text{Ln}\sigma_{a.c}(\omega)$ versus $\text{Ln}(\omega)$ as declared in figure (3) found to be less than unity for all prepared films and decreases with increasing of annealing temperature. That means the exponent (s) fits C.B.H. model given by Elliott [11] from which A.C. conductivity occurs between two sites over the barrier separating between (D^+D^-) defect centers in the band gap. This leads to greater loss in the dielectric and $\sigma_{a.c}(\omega)$ is dominating.

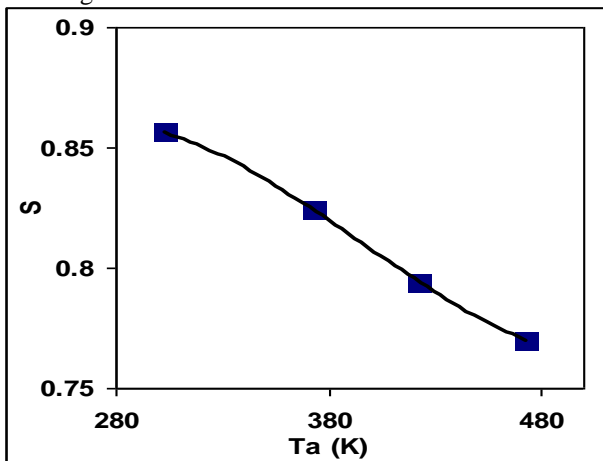


Fig.(3) Temperature dependence of the exponent (s) for ZnO thin films

Representative plot of real (ϵ_1) and imaginary (ϵ_2) parts of dielectric constants versus frequency at different annealing temperatures are shown in figures (4) and (5) respectively. The general features are the rapid decrease of dielectric constants with increasing frequency. Very large values of (ϵ) are observed, especially at High annealing temperatures. Most of this behavior arises from electrode polarization effects.

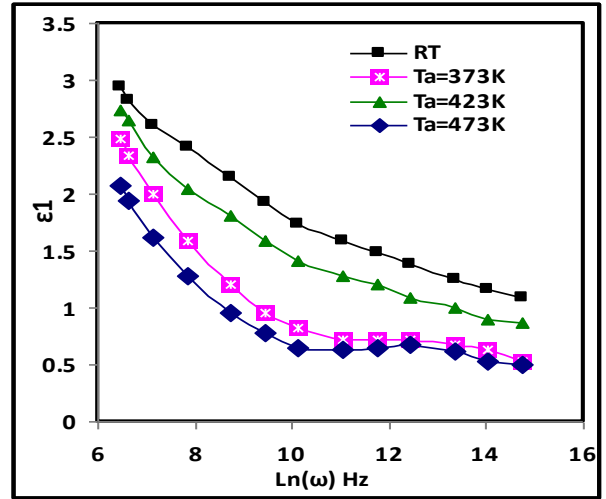


Fig.(4) Frequency dependence of ϵ_1 of dielectric constant for ZnO thin films at various annealing temperatures.

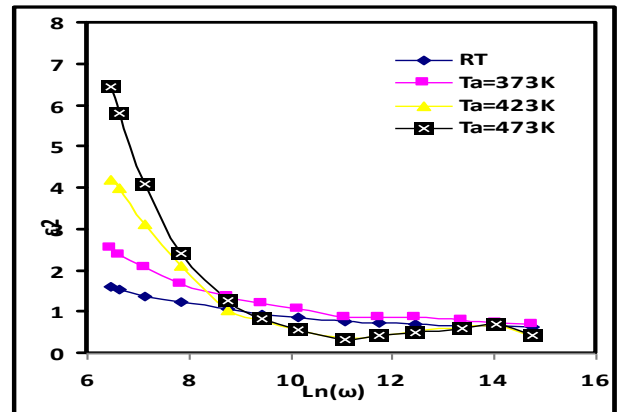


Fig.(5): Frequency dependence of ϵ_2 of dielectric constant for ZnO thin films at various annealing temperatures.

Figure (6) shows the frequency dependence of loss factor ($\tan \delta$) at different annealing temperatures. It can be seen that ($\tan \delta$) decreases with increasing of frequency for films annealed at (474 and 423) °K while the behavior of the films is non systematic for temperatures (RT and 373) °K. Moreover, the increase of annealing temperatures leads to increase of ($\tan \delta$). This may be related to the fact that, at high temperatures the loss is dominated by thermally activated electron hopping, whereas at the low-temperature region such activated process is frozen out, resulting in a decrease of (ϵ) at low temperature [12].

Also, it may be noted that, σ_{ac} is related to (ϵ) and $(\tan \delta)$. Therefore as the temperature increases, the conductivity of thin film, and hence both (ϵ) and $(\tan \delta)$ would increase.

The relation between the real part (ϵ_1) and imaginary part (ϵ_2) of the dielectric constant was plotted for all samples. Figure (7) represents the variation of ϵ_1 and ϵ_2 for ZnO thin films at different annealing temperatures. Similar behavior graphs were obtained for all samples.

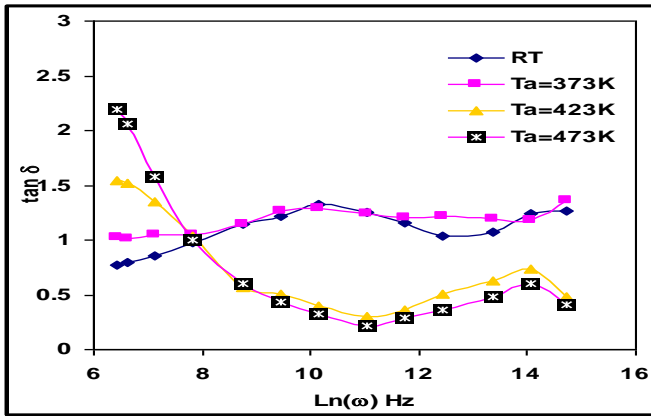


Fig.(6): Frequency dependence of $\tan \delta$ for ZnO thin films at various annealing temperatures

The behavior of these Cole-Cole diagrams is characterized by semicircles originating for all ZnO samples indicating that a single relaxation mechanism is present [13]. Such pattern tells us about the electrical processes occurring within the sample and their correlation with the sample microstructure. From these figures, (q) angle can be determined and quiescently the polarization angle can be estimated from the relation $\alpha=2q/\pi$.

The polarization can be determined from the relation $P= \alpha E$ where E is the electric field. The variation of (P) with annealing temperature for ZnO samples is shown in Fig. (8). It is clear that the value of (P) increases with increasing temperature. This indicates that the dielectric dispersion is of poly type [14].The same behavior was observed for all other samples.

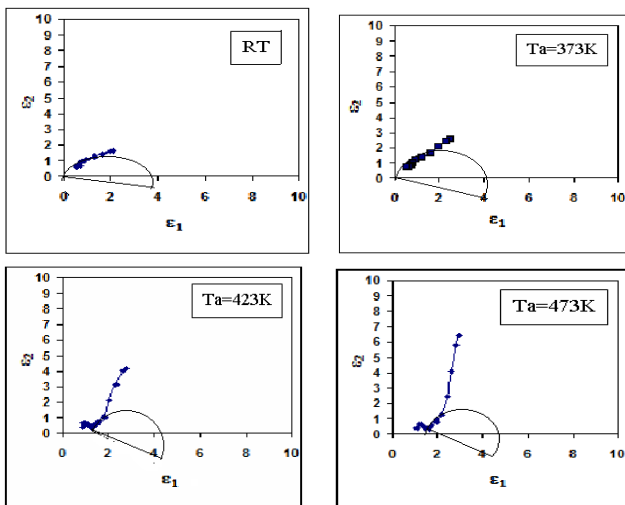


Fig.7. Cole-Cole plot for ZnO thin films at different annealing temperatures

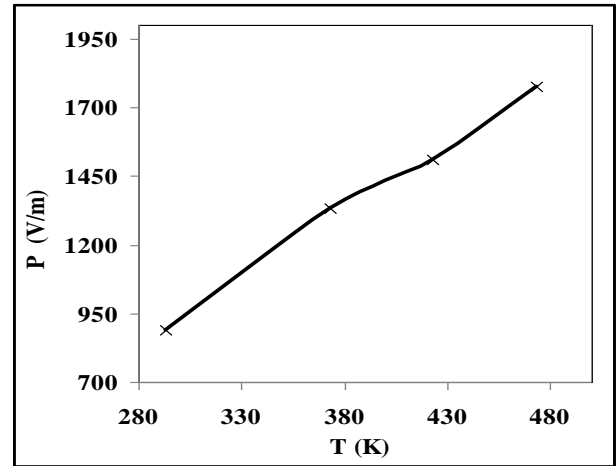


Fig.(8) Variation of polarization with annealing temperatures for ZnO thin films

IV. CONCLUSIONS

The A.C. conductivity for all samples showed strong temperature dependence where the exponent (s) decreases by increasing annealing temperature and it was related to the correlated barrier hopping type (CBH) mechanism. The dielectric constant (ϵ) and loss factor $(\tan \delta)$ were found to be influenced by annealing temperatures. The Cole-Cole diagrams showed semicircles with centers below the abscissa axes which confirm the existence of a distribution of relaxation times.

REFERENCES

- [1] G Mónica Morales Mas'ís " Fabrication and Study of ZnO Micro- and Nanostructures" M.Sc thesis, Wright State University, Dayton-Ohio (2007).
- [2] Ziad T. Khodair "Design and Fabrication Nanostructures growth of (ZnO:Fe) Compound by APCVD Technique and Study Some Physical Properties and Deposition Parameters" Ph.D Dissertation, Baghdad University, College of Education,(2011).
- [3] C. R. Gorla, N. W. Emanetoglu, S. Liang, W. E. Mayo, Y. Lu, M. Wraback and H. Shen, J. Appl. Phys. 85, p.2595(1999).
- [4] T. Minami, T. Yamamoto and T. Miyata, Thin Solid Films 366, 63(2000).
- [5] D. C. Look, Mater. Sci. Eng. B 80, 383 (2001).
- [6] C. Klingshirn, Phys. Stat. Sol. (b) 244, 3027 (2007).
- [7] D. C. Look "Recent advances in ZnO materials and devices" Materials Science and Engineering B 80, p.383-387(2001).
- [8] F. K. Shan and Y. S. Yu "Band gap energy of pure and Al-doped ZnO thin films", Journal of the European Ceramic Society 24, p.1869-1872, (2004).
- [9] C.A. Hogarth, M.H. Islam and S.S.M.S. Rahman, J. of Materials Science 28,p. 518 (1993).
- [10] S.R. Elliott, Advances in Physics, 36(2), p.135 (1987).
- [11] S.R. Elliott, "Philo. Mag.", 36,p. 1291, (1977).
- [12] A.R. Long and W.R. Hogg, J. of Non-Cryst. Solids 59-60, 1095 (1983).
- [13] E. Barsoukov and J. R. Macdonald, Impedance Spectroscopy, Wiley-Interscience (2005).
- [14] M. A. Al-Muraikhi, Ph.D dissertation, Ain Shams University, Egypt (1987).