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The Effect Of Changing Al_2O_3 Concentrations And Nano Crystal Size On $(Zno)_X$ $(Al_2O_3)_{1-X}$ Thin Films Conductivity And Imaginary Electric Permittivity

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Abstract

In this work, $(ZnO)_x$ $(Al_2O_3)_{1-x}$thin films were deposited on glass substrate, eleven sample were prepared by sol-gel method with zinc oxide concentration ranging from 0.1 to 1 in step of 0.1. The nano crystal size of the deposited micro crystals was found by the XRD technique. The UV-VIS spectrum was used to find the conductivity and the complex electric permittivity. The electric c and activity and the complex electric permittivity increases when the crystal size of $(ZnO)_x$ $(Al_2O_3)_{1-x}$compound increases the electric conductivity attains maximum value of about $35\Omega^{-1}cm^{-1}$, while the maximum value of complex electric permittivity is about 3.06×10^{-2} when the crystal size is minimum and equal $50.662\ nm$ for (ZnO) concentration $x = 0.6$.

The minimum values of electric conductivity and complex electric permittivity are about $33\Omega^{-1}cm^{-1}$ and 2.9×10^{-2} for the crystal size$26.267\ nm$. The conductivity and complex electric permittivity also was found to increase upon increasing (Al_2O_3) concentration. This may be attributed to the fact that the crystal size of (Al_2O_3) alone is $35.483\ nm$ which is greater than the ZnO crystal size which is $27.869\ nm$. This means that increasing crystal size increasing both

electric conductivity and complex electric permittivity. This result can form with proposed theoretical model.

Keywords: glass substrate, XRD technique, UV spectrum, complex electric permittivity, conductivity, nano crystal.

Introduction

Energy problem is of the long standing problem that facing modern civilization. Petroleum fuel which is now widely used is found to cause severe biological hazards [1]. This forces scientists to search for free pollution alternative energy source. The best one is the solar energy, and the most popular converter is the solar cell which convert solar radiation energy to electric energy [2, 3].The commercially available solar cell is that made of Silicon [4,5] which is now widely used in remote and isolated regions. Silicon solar cell has long life time, but unfortunately has low efficiency and high cost [4, 5]. This encourages researchers to try new bath ways to fabricate cheap, efficient solar cells [6, 7]A search for new solar cells alternatives was triggered due to emergence of nano science as a powerful tool that can change matter physical properties [8,9]. One of the new trends to fabricate cheap solar cells is based on nano solar cells. In such cells transparent conducting electrodes as well as polymers and dyes are used in their fabrication [10,11]. The transparent electrodes are usually made from transparent conducting oxides. One of the most widely used one is indium tin oxide (ITO).Although ITO has high transmittance and good conductivity, but unfortunately it is expensive and toxic [12, 13]. This forced researchers to search for cheap non toxic alternatives. One of the best alternatives is the Aluminum Zinc oxides thin films. Such films are cheap and ease controllable resistance, besides having high transmittance these remarkable feature of $(ZnO)(Al_2O_3)$ thin films encourages us to fabricate such films.

Samples Preparation

The preparation of ZnO and Al_2O_3 was made by using Zinc acetate dehydrate $Zn(CH_3COOH)_2.2H_2O$ and Aluminum nitrate monohydrate $Al(NO_3)_3.9H_2O$. The need for surfactant is fulfilled by the use of 2-methoxyethanol (ME) $CH_3OCH_2CH_2OH$. The stock solution was prepared using Zinc acetate 0.1M, dissolved in 300 ml of ethanol in the glass beaker. Then the solution was stirred for 60 min at 80°C until one get milky solution. Drops from 2-methoxyethanol (ME) were added to the solution as stabilizer to get a transparent solution. We get then the Zinc oxide solution. The solution was divided into ten equal volumes of 50 ml in ten beakers. In order to add Al to ZnO, another solution was prepared by dissolving aluminum nitrate monohydrate 0.1M in 50 ml of ethanol. The Al_2O_3 and ZnO were mixed in such a way to make their

total weight equal to a fixed volume, in a total volume at 50 ml. one denoted the weight or amount of ZnO by x. thus the compound is denoted by $(ZnO)_x$ $(Al_2O_3)_{1-x}$. then the solutions have been leaved at room temperature for about 24 hours. Eleven samples were prepared with ZnO concentration x=0.1 to 1.0 with step of 0.1 respectively. The thicknesses of the $(ZnO)_x$ $(Al_2O_3)_{1-x}$ thin films were about 359.3 nm for all samples . The spectra of the $(ZnO)_x$ $(Al_2O_3)_{1-x}$ thin films were displayed as a function of wavelength by UV-visible spectroscopy. The crystal structure of all samples characterized at room temperature using a Philips PW1700 X-ray diffract meter (operated at 40 kV and current of 30 mA). Samples were scanned between 20^o and 90^o at a scanning speed of 0.06 $^oC/s$ using $Cu\ K\alpha$ radiation with λ= 1.5418$\mathring{A}$.

Theoretical Model

X-ray diffraction (XRD) technique is powerful technique, used to determine crystal spacing and type. Recently it is used to determine the nano size crystals in thin films. One can prove this simple here. The diffraction resulted from a unit cell is described by Bragg equation:

$$2d \sin\theta = \lambda \quad (1)$$

With d standing for spacing between crystal plans

$$\theta_r = \sin\theta = {}^{\lambda}/_{2d} \quad (2)$$

One can determine the crystal size by assuming them acting as unit cells, considering the position of the first fringe as a diffraction center for the pattern resulted from crystals, it follows that:

$$2D \sin\theta` = \lambda \quad (3)$$

$$D = X_s \quad (4)$$

$$\theta_r = \sin\theta = \frac{\lambda}{2X_s} \quad (5)$$

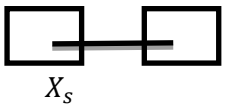

Fig (0.2) Crystal size

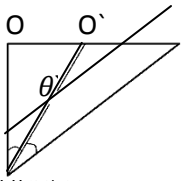

Fig (0.1) crystal diffraction
O = original diffraction center from unit cell
O`= artificial diffraction center from unit cell

From Fig 1

$$\Delta\theta_r = \theta_r` \quad (6)$$

But the radial angel θ is given by

$$\theta_r = \sin\theta \quad (7)$$

Thus

$$\Delta\theta_r = \Delta \sin\theta \quad (8)$$

But

$$\frac{\Delta \sin\theta}{\Delta\theta} = \frac{d \sin\theta}{d\theta} = \cos\theta \qquad (9)$$
$$\therefore \quad \Delta \sin\theta = \cos\theta \, \Delta\theta$$

From equations 5, 6, 8 and 9 then

$$S_s = \frac{\lambda}{2\theta_r`} = \frac{\lambda}{2\Delta\theta_r} = \frac{\lambda}{2\Delta \sin\theta} = \frac{\lambda}{2\cos\theta\Delta\theta} \qquad (10)$$

The conductivity is related to electric permittivity, this relation comes from the definition of the current density J, where

$$J = n\, e\, v = ne\frac{dx}{dt} = \frac{d\, n\, e\, x}{dt} = \frac{dP}{dt}$$
$$= x\frac{dE}{dt} = \mathcal{X}E_0\frac{de^{-iwt}}{dt} = -iw(\mathcal{X}_1 + i\mathcal{X}_2)E \qquad (11)$$
$$= (w\mathcal{X}_2 + iw\mathcal{X}_1)E = \sigma E = (\sigma_1 + i\sigma_2)E$$

Where $x, P, \mathcal{X}_1, \mathcal{X}_2, \sigma_1 \; and \; \sigma_2$ stands for displacement, electric dipole moment, imaginary electric susceptibility, and real beside imaginary conductivity respectively.

Thus the conductivity is given by:

$$\sigma_1 = w\mathcal{X}_2 \qquad (12)$$

And the imaginary dielectric constant is given by

$$\varepsilon_2 = \varepsilon_2\mathcal{X}_2 = \frac{\varepsilon_0\sigma_1}{w} \qquad (13)$$

Results and Discussion

The x-ray diffraction pattern in Fig (0.1), is used to find the space between two successive planes, the crystal size of nano particles x_s, and the crystal densityρ. The relation between the ZnO concentration x on one side and crystal sizex_s, plane spacing a, and matter density ρ are exhibited in table (1). Fig (0.2) shows that the nano crystal size has maximum values at concentrations $x = 0.6, 0.3$, and 0.1, where they take values 40.6, 39.2, and 38.9 nm respectively. Whereas it attains minimum values of 62.2 nm at x=0.5.

The plane spacing d variation is not large, where it attains minimum value of 0.1945 nm and maximum value of 0.2827 at concentration x=0 and x=0.1 respectively. [see table (1) and Fig (3)]

The empirical relations for the imaginary dielectric constant and the electrical conductivity spectra versus the wavelength λ for all samples have been displayed in Figs (6) and (8).

It very interesting to note that the electrical conductivity $\sigma$and imaginary electric susceptibility have minimum values for x = 0.5 and maximum values for x = 0.6. The crystal size is very small and equal to 26 nm for x = 0.5 where both σ and $\mathcal{X}_2$ attains minimum values of about $33\Omega^{-1}cm^{-1} \; and \; 2.9 \times 10^{-2}$ respectively. The crystal size is very large and equal 40 nm for x = 0.6, where s and c 2 have maximum values of

about $35\ \Omega^{-1}cm^{-1}\ and\ 3.06 \times 10^{-2}$ respectively.This may be explained by bearing in mind that when the crystal size is large the spaces between them become very small. This is since the size of the glass substrate surface size is the sum of crystals sizes and spaces between them. These small spaces decreases resistance and increases conductivity. Since (see equation 12) $\mathcal{X}_2 = \sigma$, it follows that the imaginary dielectric constant $\varepsilon_2 = \varepsilon_0 \mathcal{X}_2$increases also. The large crystal size also increases the internal field $E_i$also. This increases the electrons velocity v, which increases conductivity σ,according to the equation

$$J = \sigma_0(E + E_i) = \sigma_0(E + C_0E) = \sigma_0(1 + C_0)E = \sigma E = n\ e\ v$$

This also increases ε_2 according to equation (13).

Increasing Al_2O_3 concentration increases the conductivity also. This may be related to the role of it in increasing the crystal size. According to table (1) below, the crystal size of (Al_2O_3) only is about 35 nm, while the size of ZnO alone is about 27nm.

Table (1) some of crystallite lattice parameter (c-form, a, b, c, $\beta, \alpha, \gamma, \rho, X_s(nm) and\ d-spacing$) of all samples that made by $(ZnO)_x\ (Al_2O_3)_{1-x}$

Sample	C-form	A	B	C	α	β	γ	ρ	X_s	D
$(ZnO)_{0.9}(Al_2O_3)_{0.1}$	Hexagonal	5.68	5.68	13.71	90	90	120	6.2756	40.6	0.244
$(ZnO)_{0.8}(Al_2O_3)_{0.2}$	Orthorhombic	7.93	7.96	11.71	90	90	90	6.647	27.2	0.213
$(ZnO)_{0.7}(Al_2O_3)_{0.3}$	Hexagonal	5.68	5.68	22.52	90	90	120	3.2255	27.2	0.213
$(ZnO)_{0.6}(Al_2O_3)_{0.4}$	Triclinic	5.00	5.18	4.88	97.5	118.74	184.74	40.15	40622	0.244
$(ZnO)_{0.5}(Al_2O_3)_{0.5}$	Hexagonal	3.047	3.037	57.26	90	90	120	1.1069	26.267	0.2602
$(ZnO)_{0.4}(Al_2O_3)_{0.6}$	Hexagonal	4.83	4.52	7.38	90	90	120	0.9169	27.157	0.213
$(ZnO)_{0.3}(Al_2O_3)_{0.7}$	Triclinic	2.86	2.85	2.86	59.6	59.9	59.72	5.053	39.24	0.2434
$(ZnO)_{0.2}(Al_2O_3)_{0.8}$	Monoclinic	5.93	4.77	6.56	90	1172	90	1.8520	38.972	0.2826
$(ZnO)_{0.1}(Al_2O_3)_{0.9}$	Hexagonal	5.57	5.57	8.64	90	90	120	3.6473	38.973	0.2827
$(ZnO)_{0.0}(Al_2O_3)_{1.0}$	Monoclinic	9.22	7.54	10.35	90	109.2	90	1.2827	35.483	0.1945
$(ZnO)_{1.0}(Al_2O_3)_{0.0}$	Hexagonal	3.24	3.24	5.21	90	90	120	5.6779	27.865	0.215

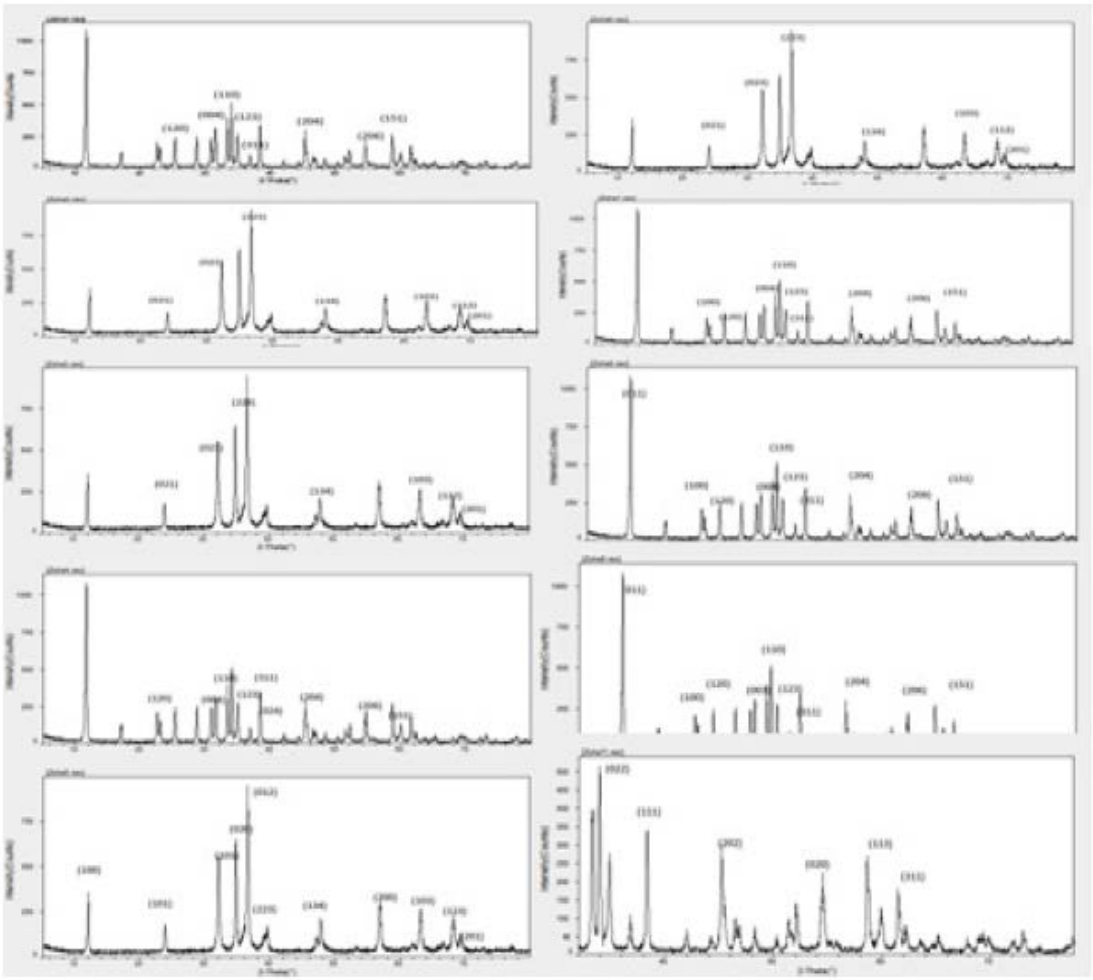

Fig (1) XRD diffraction pattern for all $(ZnO)_x(Al_2O_3)_{1-x}$ samples

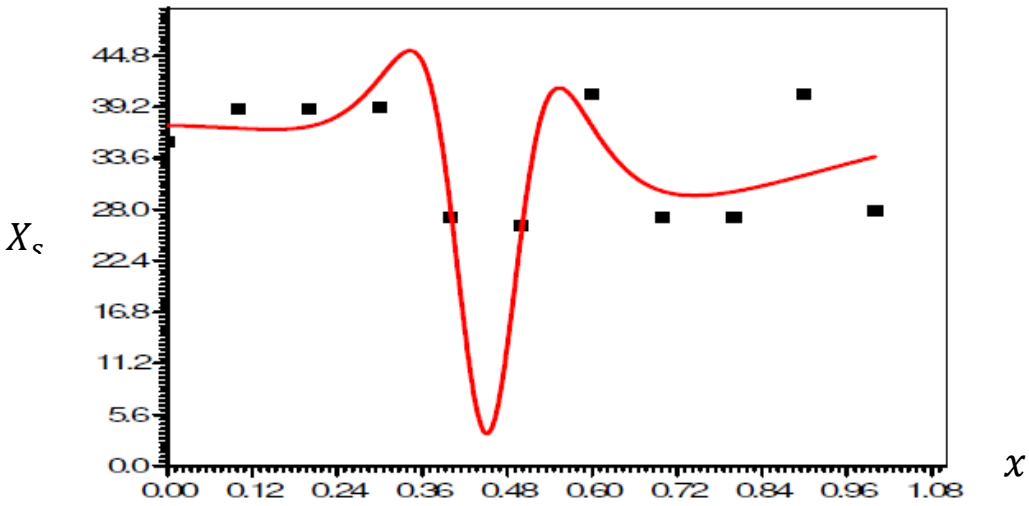

Fig (2) dependence of the crystal size X_s on ZnO concentration x

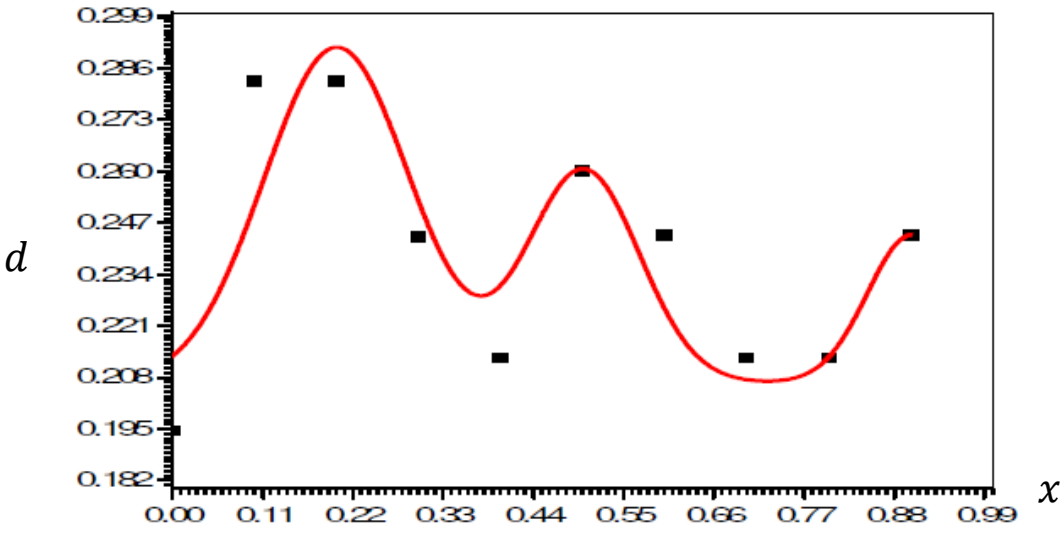

Fig (3) dependence of distance between two planes d on ZnO concentration x

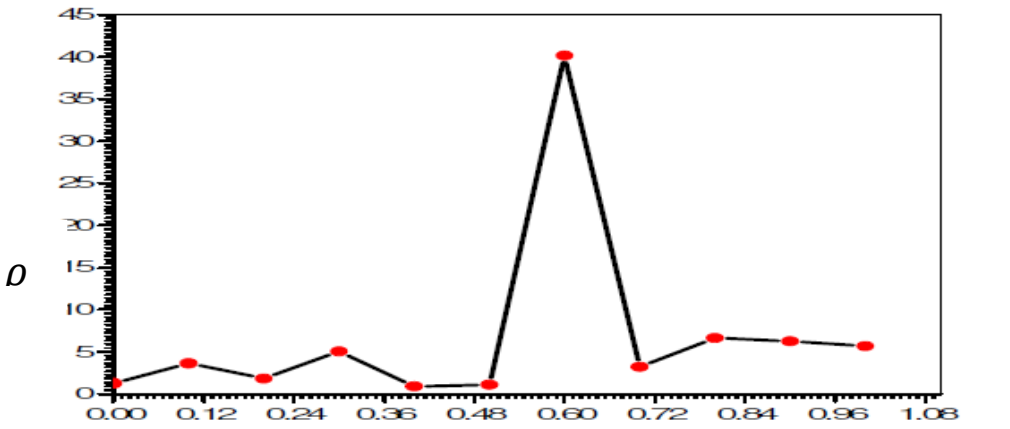

Fig (4) Dependence of the density ρ on ZnO concentration x

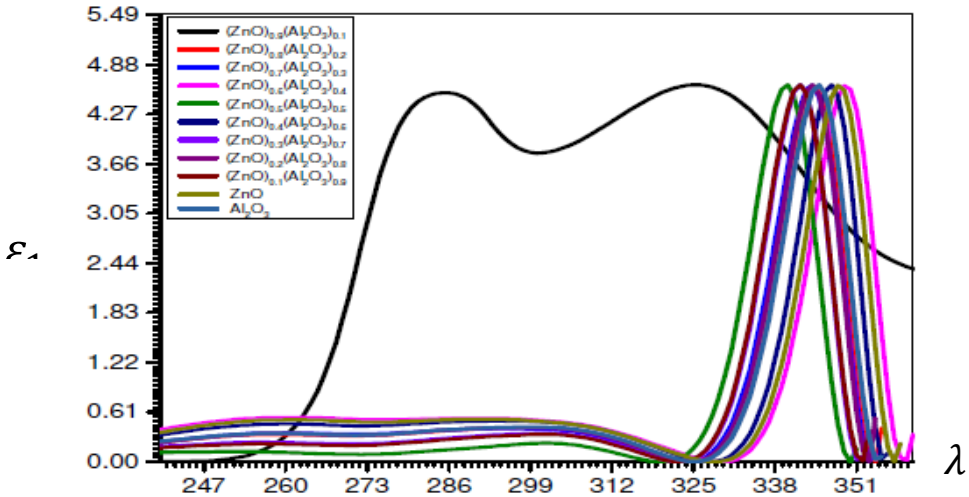

Fig (5) The relation between real dielectric constant and wavelengths of $(ZnO)_x(Al_2O_3)_{1-x}$ samples

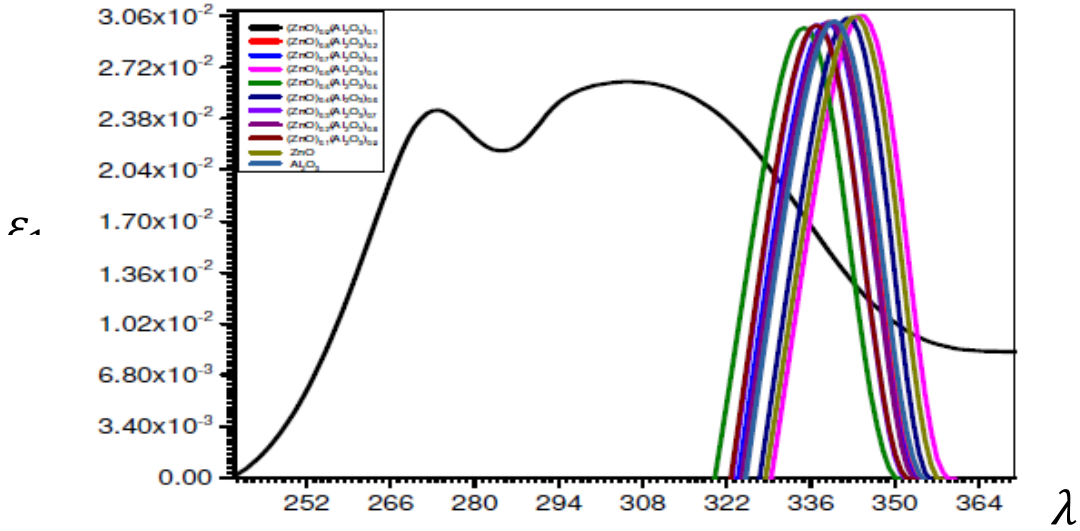

Fig (6) The relation between imaginary dielectric constant and wavelengths of $(ZnO)_x(Al_2O_3)_{1-x}$ samples

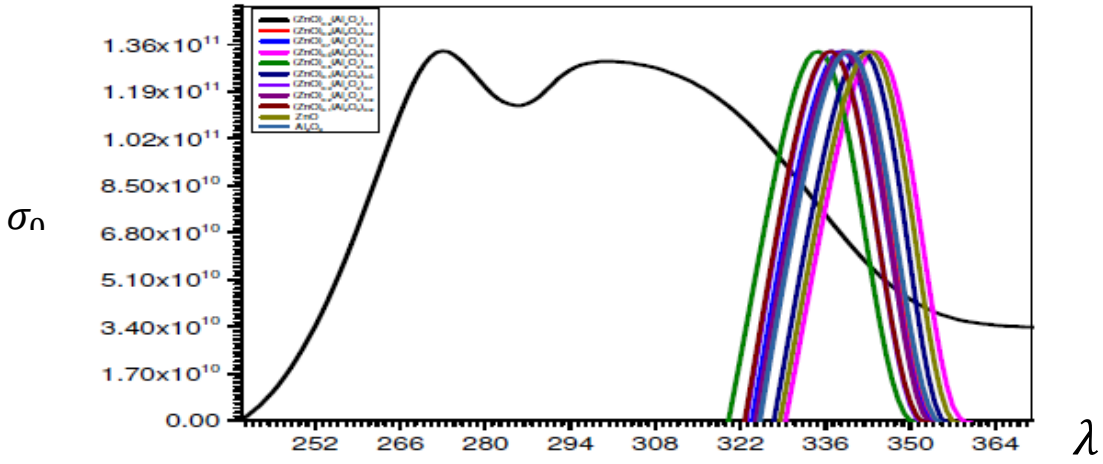

Fig (7) The relation between optical conductivity and wavelengths of $(ZnO)_x(Al_2O_3)_{1-x}$ samples

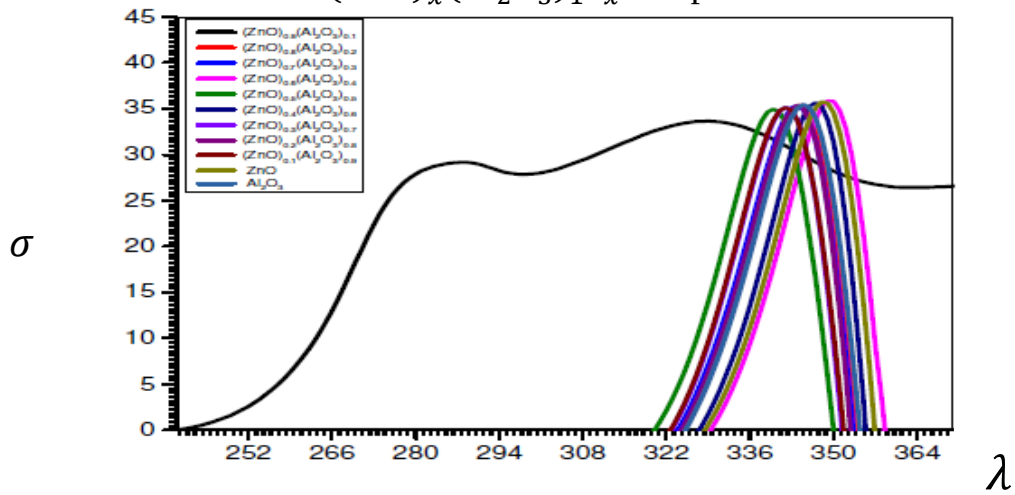

Fig (8)The relation between electrical conductivity and wavelengths of $(ZnO)_x(Al_2O_3)_{1-x}$ samples

Conclusion

The electric conductivity and imaginary electric permittivity of $(ZnO)_x(Al_2O_3)_{1-x}$ thin films are maximum when the crystal size is maximum, at $x = 0.6$, while they are minimum when the crystal size is minimum at $x = 0.5$. Also the electric conductivity and imaginary electric permittivity are found to be minimum upon increasing (Al_2O_3) concentration. This may be related to the fact that the size of crystal formed from (Al_2O_3) alone is larger than that of (ZnO) crystals. Generally speaking the conductivity and permittivity increase when the crystal size increase. This result can form with theoretical relation.

References

[1] Nelson, The Physics of Solar Cell, *Imperial College Press*, London, UK, 2003.

[2] L. A. Dobrzanski, L. Wosinka, B. Bolzanka, and A. Drygala, "Comparison of Electrical Characteristics of Silicon Solar Cells," *Journal of Achievements in Material and Manufacturing Engineering*, vol. 18, pp. 215-218, 2006.

[3] M. Chegaar, Z. Ouennoughi, F. Guevhi, and H. Langueur, "Determination of Solar Cells Parameters under Illuminated Conditions," *The Journal of Electron Device*, vol. 2, pp. 17-22, 2003.

[4] J. D. Servaites, M. A. Ratner, and T. J. Marks, "Practical Efficiency Limits in Organic Photovoltaic Cells: Functional Dependence of Fill Factor and External Quantum Efficiency" *Applied Physics Letters*, vol. 95, no. 16, Article ID 163302, 3 pages, 2009.

[5] A. Subrahmanyan and N. Balasubramanian, "Studies of The Photovoltaic Behavior of Indium Tin Oxide (ITO)/Silicon Junctions Prepared by the Reactive Thermal Evaporation Technique," *Semiconductor Science and Technology, vol. 7, no. 3, Article 07, pp. 324-327, 1992.*

[6] Stroble, G. R. 2007, The Physics of Polymers Concepts for Understanding Their Structures and Behavior, p293, ISBN: 978-3-540-2578-8.

[7] Ettersson, H., Gruszecki, T., 2001, Long-Term Stability of Low-power Dye-sensitized Solar Cells prepared By Industrial Methods, Solar Energy Materials & Solar Cells, 701, 203-212.

[8] NimaParsiBenehkohal (2013) Innovations in Electrophoretic Deposition of Nanotitania-Based Photoanodes for Use in Dye-sensitized Solar Cells, Ph. D Thesis McGill University Montreal, QC, Canada.

[9] Nazeeruddin MD. K., Baranoff E., Gratzel M (2011) Dye-Sensitized Solar Cells: A brief Overview, Sol. Energy, vol. 85, pp. 1172-1178.

[10] HaraldHoppe N. SerdarSaricftci 2 Polymer Solar Cells AdvPolymSci DOI 10.1007/12_2007_121 © Springer-Verlag Berlin Heidelberg Published Online: 17 OCT. 2007 P2.

[11] Murat Atesa,, TolgaKarazehira and A. SezaiSaracb (2012) Conducting Polymers and Their Applications, Current Physical Chemistry, 2012, 2, 224-240.

[12] M. Nisha, S. Anusha, A. Antony, R. Manoj, and M. K. Jayaraj, "Effect of Substrate Temperature on the Growth of ITO Thin Films," *Applied Surface Science*, vol. 252, no. 5, pp, 1430-11435, 2005.

[13] N. Manavizadeh, F. A. Boroumand, E. Asl-Soleimani et al., "Influence of Substrates on the Structural and Morphological Properties of RF Sputtered ITO Thin Films for Photovoltaic Application.," Thin Solid Films, vol. 65, no. 3, pp. 1270-1280, 1989.