

EFFECTS OF SUBSTRATE TEMPERATURES ON OPTICAL PROPERTIES AND CONSTANTS OF ZnO PREPARED BY PLD

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ZnO/quartz heterostructure was deposited of a high purity Zn target in the presence of substrates temperatures. The optical properties and the optical constant of these film has been investigated reaching to use for Optoelectronics applications, formed at 300 torr oxygen ambient, showed an electrical resistivity of 0.0221 Ω .cm, without using post-deposition heat treatment. The Optical property shows high transparency 90 % and found to decreases sharply with the decreasing of the temperatures of the substrate. The value of the energy gap (Eg) of the prepared films is around 3.66 eV. The deposit films were analyzed using the UV-visible.

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1. Introduction

The (TCOs) Transparent Conducting Oxide nano and micro films are electrically conductive Materials that are highly transparent in the visible range of wavelengths as well as electrically conductive [1-4]. The importance of this material can be returned to the twentieth century when the first films were recorded with their unique properties [5, 6].

This material has found a wide application in optoelectronic, nanoelectronics and photonics devices such as SAW, the optical switching, the solar cells, Liquid Crystal Display (LCD), heat mirrors and many others different applications depending on the type of material [7-14]. The aim of many manufacturers, beyond the fabrication Of TCOs, is to achieve stable characteristics of the operations of coating a large area with the high value of the transmittance and a low film resistance within the values of the visible region of the electromagnetic spectrum. It is worth to annotate that the electrical properties of TCO films increases effectively as a result of increasing temperature from one side and reduction of oxygen pressure from other side.

The reason is of the disintegration of the zinc oxide formatting of a solid solution of the metal on its oxide.. The only way to get a good transparent conductors is to create a degeneration of the electron in the wide band gap value around or more than 3eV oxide by the introduction of a controlled of non - stoichiometry and/or by choosing a suitable dopant [15-19]. The characteristics of thin film ZnO are currently of great commercial and scientific significance.

The ZnO crystallizes primarily in the wurtzite structure, which carries strong piezoelectric properties. For this reason, the zinc oxides (caxis-oriented) nano and microfilms are of interest in the fabricating of surface acoustic wave (SAW), optoelectronics, and Photonics devices [20-23] beside that, the wide band gap of this material makes it an excellent visible light transparent and UV absorbing material. This, together with its low toxic effect, makes it an ideal material to use in sunscreen as a UV blocking element. Depending on the deposition conditions (substrate temperature and oxygen pressure), the ZnO films can be divided into three groups, the first group belongs to a mixture of ZnO and metallic zinc; and these films are conductive and opaque, the

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second group of films has a composition close to stoichiometric bulk ZnO; they are non-conductive but transparent, while films of the third group is of high importance in optoelectronic applications since they are transparent and conductive films [24-28]. The electrical conductivity of these films increases in the temperatures increasing and reducing partial oxygen pressure [29-33].

This work present effect of substrates temperatures on the structural, optical, and surface morphology, of ZnO nanoparticles prepared using RPLD technique.

2. Experiment

The undoped, high purity, and high crystallization ZnO films were successfully deposited on the extra cleaned quartz sheets at (300, 400) C°. The target was ablated using Q- switched Nd:YAG pulsed laser of (FWHM) equal 7 ns and $\lambda = 1.064 \mu\text{m}$, which Focus on a converged lens with a high degree of purity of Zinc. The deposited films were created by using 40 laser shots and deposited at different substrate temperature.

A 200 nm film thickness was optically recorded using a stylus profilometer. The other optical characteristics such as the transmittance T%, and the reflectance R%, were investigated between the spectral the range between 0.3 μm to 1 μm using double beam spectrometer. The photon energy incident as a function of λ (the wavelength) can be obtained using the equation (1) [34, 35]:

$$E_g(eV)=1240/\lambda(nm) \quad (1)$$

where λ is wavelength. The Tauc equation (2) was used to find the energy band gap as a function of absorption coefficient and excitation of the transmission [36, 37]:

$$(\alpha h\nu)=B (h\nu-E_g)^r \quad (2)$$

where r is a constant that can take any integer value based on the raw material and the optical transmission values $\alpha h\nu$. B is a constant inversely proportion tit the value of r . The energy band gap value can be obtained from the intercept of both curves of $(\alpha h\nu)^{1/r}$ versus $(h\nu)$ extrapolated at $(\alpha h\nu)^{1/r}=0$.

The examination of the absorption coefficient for all used wavelengths was found from equation (3) [38, 39]:

$$\alpha=2.303(A/t) \quad (3)$$

where A and t are the absorbance and the transmittance, respectively. Equation (4) explains the relation between the absorption coefficient (α) and the excitation ratio (K) [40, 41]:

$$K = \alpha \lambda / 4\pi \quad (4)$$

Other constants such as the refractive (n), ϵ_r and ϵ_i which they relate to the real and imaginary parts of the dielectric constants, the conductivity (σ) are used to be calculated from equation (5) [42, 44]:

$$n = \{[4R / (R-1)]-K^2\}^{1/2} - [(R+1) / (R-1)] \quad (5)$$

Knowing that R represents the reflectance which can be calculated from ($R = 1-T-A$), or from equation (6) [45-47]:

$$n=n_s \left(\frac{1+\sqrt{R}}{1-\sqrt{R}} \right)^{1/2} \quad (6)$$

where n_s denotes to the substrate refractive index. It is an important to mention that the refractive index of the quartz substrate is considered to be 1.51.

3. Results and discussion

The XRD pattern at the optimum substrate temperature (400 C°) are shown in Fig. 1. From this figure, we can recognize that the peaks appear at $2\theta = 31.7, 36.2, \text{ and } 37.8$ in the spectra of the ZnO films corresponding to the reflection from (100), (101), and (102) planes. The formation of the ZnO films was confirmed by the reflections from the given planes and the diffraction angle given above.

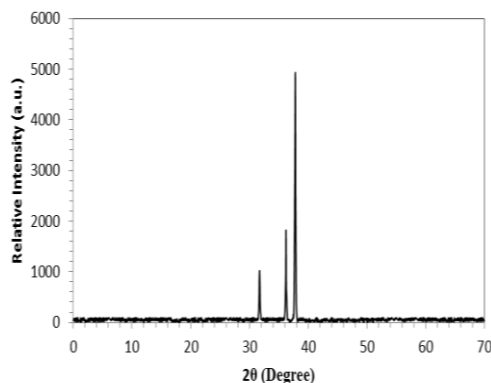


Fig. 1. The XRD pattern of the prepared samples at 400 C°.

Under two substrate temperatures, the transmission spectra of the prepared thin films have been examined and analyzed. Fig. 2 shows the transmission of the prepared films as a function of wavelength. The Fig. 2 shows that the average transmission ratio of the prepared samples increases as the temperature of the substrate goes up. It can be concluded that all prepared samples have high transparency at the visible region of the optical spectrum with average values reaches up to 90%. The transmission curves are found to be about 80% and 90% for 300°C and 400°C of temperatures, respectively. Further, a very sharp cut-off is also presented at the wavelength 340 nm. Higher transmission values indicates that the prepared thin films are in crystalline form

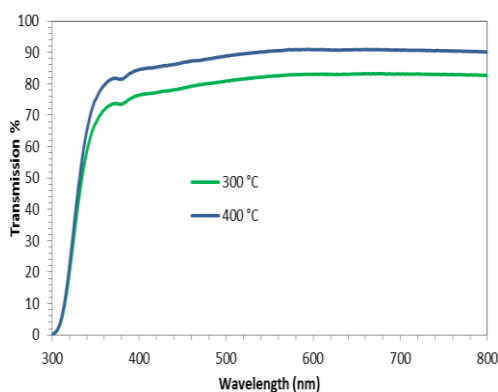


Fig. 2. The Transmission curve as a function of the wavelength ranges between 300 nm to 800 nm.

It is known that the sharp decrease in transparency of Zinc oxide films in UV and slightly decrease IR regions is caused by fundamental light absorption and by free – carrier absorption respectively, this agrees with the result in similar work [2, 48].

Previously, we referred to Tauc equation (2) to be used for optical band-gap calculation of the prepared ZnO films. The extrapolating of the straight lines of the plot $(\alpha h\nu)^2$ versus the photon energy given in Fig. 3 is used to determine the band-gap energy. Comparing between both

substrate temperatures, the difference in the band-gap energy bands is comparatively small. Table 1 gives more details of the optical band-gaps for samples prepared at two substrate temperatures.

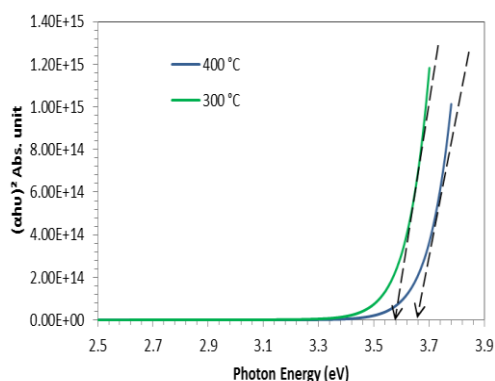


Fig. 3. The optical band-gap energy as a function of photon energy for two substrate temperatures.

A double beam ultraviolet-visible spectrometer was used to measure the reflectivity of the prepared ZnO samples. Fig. 4 presents the reflectance spectra as a function of wavelength range defined previously. As the substrate temperature increases it is clearly to notice that the optical reflection is gradually decreasing. Such reduction is come from the increasing of layer thickness of the prepared samples. Depending on the energy of the incident photon, film thickness, the deposited particle of the films and the bound and lose electrons, light beam might be reflected and then transmitted.

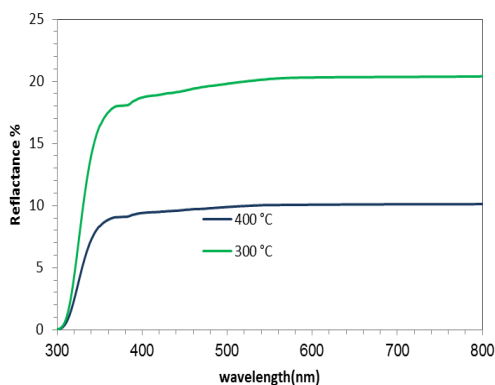


Fig. 4. Reflectance as a function of wavelength range under two substrate temperatures.

The values of optical constants such as the refractive index n were calculated using the transmittance data spectrum given in Fig. 2 by using the envelope method. The value of (n) was calculated using the transmittance spectrum. There is a notable change in the value of the refractive index from 2.33 to 2.39 at 340 nm as present in Fig. 5 and tabulated in Table 1. The reason behind the increasing of the refractive index is back to the reduction in the particle size of the zinc oxide during the deposition process. This is due to the increasing in the transmission value along with the decreasing of the film thickness. Comparing the effect of both substrate temperatures on the value of the refractive index, it can be annotate that there is a reduction in the value as long as the temperature goes up. Hence, this behavior may be attributed to a decline of the film thickness which resulting in low reflection.

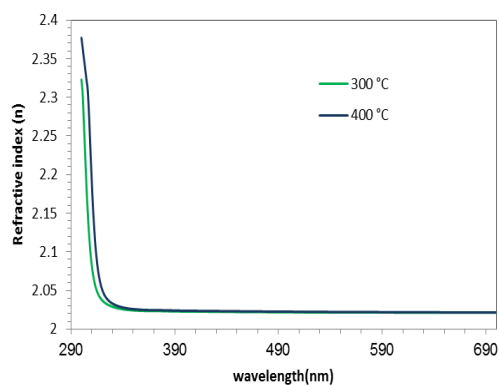


Fig. 5. Refractive index as a function of wavelength range for two substrate temperatures.

Table 1. The energy band gaps and refractive index corresponding to substrates temperature of ZnO nanostructures.

Substrate temperature (C ^o)	E _g measured (eV)	n measured	T %
300	3.58	2.33	80
400	3.66	3.39	90

Fig. 6 shows the surface morphology of deposited ZnO nanostructure on the quartz substrate at optimum preparing condition (400 C^o) resulted in highly uniform ZnO Nanoparticles thin film and its average grain size of about (93.8) nm. The surface roughnesses of the deposited thin film are about (0.90 nm) which reflect a high quality thin film. Such result opens a new gate toward introducing such films into optoelectronic application, e.g. fabrication of thin film solar cell detectors, and others.

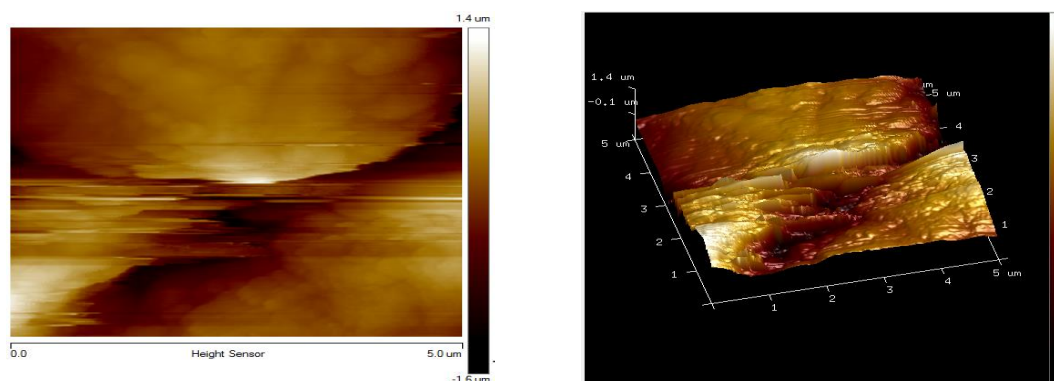


Fig. 6. Surface morphology of ZnO thin film prepared by RPLD.

4. Conclusion

The prepared film by low-cost reactive pulsed laser deposition technique on the quartz substrates at different substrate temperatures. The effect of substrate temperature on the structural, morphological, optical properties and optical constant of deposited ZnO nanostructures

has been studied. The optical band gap energy of ZnO found to be decreasing as the substrates temperature increases. The optical constant (n) was decreased reveal the effect of substrates temperature.

References

- [1] Y. S. Choi, J. Y. S. I. M. Lee, *JPN J. Appl. Phys.* **42**, 1560 (2003).
- [2] E. T. Salem, Raid A. Ismail, Makaram A. Fakhry, Yushamdan Yusof, *Int. J. Nanoelectronics and Materials* **9**, 111 (2016).
- [3] Farah T. M. Noori, A. Kadhim, N. D. Hamza, *International Journal of Nanoelectronics and Materials* **11**(3), 347 (2018).
- [4] Khalid S. Shibib, Mohammed A. Munshid, Tahir, M.M., *Thermal Science* **13**(4), 143 (2009).
- [5] Evan T. Salem, Makram A. Fakhry, Hala Hassen, *Int. J. Nanoelectronics and Materials* **6**, 121 (2013).
- [6] H. Faber, S. Das, Y. Lin, N. Pliatsikas, K. Zhao, T. Kehagias, G. Dimitrakopoulos, A. Amassian, Panos A. Patsalas, and Thomas D. Anthopoulos, *Science Advances* **3**(3), e1602640, DOI: 10.1126/sciadv.1602640 (2017).
- [7] Khalid S. Shibib, Mohammed A. Munshid, *Termal Scince* **14**(1), 49 (2010).
- [8] Abdulrazaq, O.A., Saleem, E.T., *Turkish Journal of Physics* **30**(1), 35 (2006)
- [9] Joshua J. Robbine Colin A. Wolden, *Applied Physics Letters* **83**(19), 3933 (2003).
- [10] Agool, I.R., Salem, E.T., Hassan, M.A., *International Journal of Modern Physics B* **25**(8), 1081 (2011).
- [11] Khalid S Shibib, Mayada M Tahir, Haqi I Qatta, *Pramana* **79**(2), 287 (2012).
- [12] E. Millon, O. Albert, J. C. Lonlergue, *Jornal of Apllied Physics*, **88**(11), 6937 (2000).
- [13] Salem, E.T., Agool, I.R., Hassan, M.A., *International Journal of Modern Physics B* **25**(29), 3863 (2011).
- [14] AS Ibraheam, JM Rzaij, Makram A Fakhri, AW Abdulwahhab, *Materials Research Express* **6**(2), 055916 (2019).
- [15] X. Wei, R. Zhao, M. Shao, X. Xu and J. Huang, *Nanoscale Res Lett.*, **8**(1), 112 (2013).
- [16] Raid A. Ismail, Bassam G. Rasheed, Evan T. Salim, Makram A Alhadithi, *Journal of Materials Science: Materials in Electronics* **18**(4), 397 (2007).
- [17] Khalid S Shibib, Mohammad A Munshid, Kadim A Hubiter, *Pramana* **81**(4), 603 (2013).
- [18] Raid A. Ismail, Bassam G. Rasheed, Evan T. Salim, Makram A Alhadithi, *Journal of Materials Science: Materials in Electronics* **18**(10), 1027 (2007).
- [19] Zaid T. Salim, U. Hashim, M. K. Md Arshad, Makram A. Fakhri, *Int. J. Appl. Eng. Res.* **11**, 8785 (2016).
- [20] Makram A Fakhri, *Int. J. Nanoelectronics and Materials* **9**(1), 93 (2016).
- [21] G. Kaur, A. Mitra, K. L. Yadav, *Progress in Natural Science: Materials International* **25**(1), 12 (2015).
- [22] E. T. Salim, S. M. Al Wazny, M. A. Fakhri, *Mod. Phys. Lett. B* **27**(16), 1350122 (2013).
- [23] K. M. Sandeep, S. Bhat, S. M. Dharmaprakash, *Journal of Physics and Chemistry of Solids* **104**, **36** (2017).
- [24] Zaid T Salim, U Hashim, MK Md Arshad, 2016 IEEE International Conference on Semiconductor Electronics (ICSE), 5 (2016) DOI: 10.1109/SMELEC.2016.7573577
- [25] Salim, E.T., *Indian Journal of Physics* **87**(4), 349 (2013).
- [26] A. V. Singh, M. Kumar, R. M. Mehera, *J. Indian Inst. Sci. SRP.* **81**, 527 (2001).
- [27] E. Muchuweni, T. S. Sathiaraj, H. Nyakoty, *Heliyon* **3**(4), e00285 (2017).
- [28] Makram A. Fakhri, F. Hattab, *Engineering Sciences (FNCS)*, 2012 First National Conference IEEE, **2012**, 1 (2012).
- [29] C. Gumu, O. M. Ozkendir, H. Kavak, Y. Ufuktepe, *J. Optoelectron. Adv. M.* **8**(1), 299 (2006).
- [30] A. Nakrela, N. Benramdane, A. Bouzidi, Z. Kebbab, M. Medles, C. Mathieu, *Results in Physics* **6**, 133 (2016).

- [31] R. Vinodkumar, K. J. Leth, P. R. Arunkumar, Renju R. Krishnan, N. Venugopalan Pillai, V. P. Mahadevan Pillai, Reji Philip, *Materials Chemistry and Physics* **121**, 406 (2010).
- [32] C. Asma, B. Boumdienne, C. Meriem, *Int. J. Nanoelectronics and Materials* **9**, 103 (2016).
- [33] A. Ayeshamariam, M. Kashif, V. S. Vidhya, M. G. V. Sankaracharyulu, V. Swaminathan, M. Bououdina, M. Jayachandran, *Int. J. Nanoelectronics and Materials* **9**, 49 (2016).
- [34] Makram A Fakhri, M Halim A Wahid, Ban A Badr, Suad M Kadhimi, Evan T Salim, Uda Hashim, Zaid T Salim, *EPJ Web of Conferences* **162**, 01004 (2017).
- [35] Marwa A Dawood, Makram A Fakhri, Farah G Khalid, Omer S Hassan, Mustafa S Abdulla, Abdulrahman A Ahmed, Saad A Abduljabar, *IOP Conference Series: Materials Science and Engineering* **454(1)**, 012161 (2018).
- [36] Makram A Fakhri, Evan T Salim, M. H. A. Wahid, U Hashim, Zaid T Salim, Raid A Ismail, *Journal of Materials Science: Materials in Electronics* **28**, 11813 (2017).
- [37] Evan T. Salim, Jehan A. Saimon, Marwa K. Abood, Makram A. Fakhri, *Materials Research Express* **4(10)**, 106407 (2017).
- [38] Fakhri, M.A., Al-Douri, Y., Hashim, U., Salim, E.T. *Solar Energy* **120**, 381 (2015).
- [39] Zaid T Salim, U Hashim, MK Md Arshad, Makram A Fakhri, Evan T Salim, *Materials Research Bulletin* **86**, 215 (2017).
- [40] B. A. Bader, N. H. Numan, F. G. Khalid, M. A. Fakhri, A. W. Abdulwahab, *Journal of Ovonic Research* **15(1)**, 53 (2019).
- [41] Zaid T. Salim, U. Hashim, M. K. Md Arshad, Makram A. Fakhri, Evan T. Salim, *Microelectronic Engineering* **179**, 83 (2017).
- [42] Mariam M Hassan, Makram A Fakhri, Salah Aldeen Adnan, *IOP Conference Series: Materials Science and Engineering* **454(1)**, 012172 (2018).
- [43] Haleemah T Halboos, Evan Tareq Salim, *IOP Conference Series: Materials Science and Engineering*, 454(1), 012174 (2018).
- [44] Makram A Fakhri, Ahmed W Abdulwahhab, Suad M Kadhimi, Marwa S Alwazni, Salah A Adnan, *Materials Research Express* **6(2)**, 026429 (2018).
- [45] Marwa A.Hassan, Mahasin F. Hadi Al-Kadhemy. Evan T. Salem, *Int. J. Nanoelectronics and Materials* **8**, 69 (2015).
- [46] Evan T Salim, Raid A Ismail, Makram A Fakhri, BG Rasheed, Zaid T Salim, *Iran J Sci Technol Trans Sci.* **1** (2018). <https://doi.org/10.1007/s40995-018-0607-8>
- [47] Muataz Hameed Salih Yaseen Jurn, Fareq Malek, Sawsen Abdulahadi Mahmood, Wei Wen Liu, Mukram Al-Hadethy, *Key Engineering Materials* **701**, 57 (2016)
- [48] Makram A Fakhri, Y Al-Douri, Evan T Salim, Uda Hashim, Yushamdan Yusof, EeBee Choo, Zaid T Salim, Yaseen N Jurn, *ARPN J. Eng. Appl. Sci.* **11**, 4974 (2016)