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Substrate Temperature Effects on Optical Properties and Constants of ZnO

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ABSTRACT

The preparation process of ZnO TCO (Transparent Conducting Oxide) thin films have been conducted in this paper. A Q-switched Nd:YAG laser of 7 ns duration and 1 joule of energy was utilized to perform the deposition process of ZnO/glass heterojunction. Up to the optimum oxygen pressure, optical constant and properties of the prepared samples have been investigated. Under oxygen ambient of 300 torrs and without post-deposition heating treatment, the results recorded an electrical sensitivity of 0.0221 Ω.cm. higher transparency of 80% was also listed with a reduction in its value alongside with the reduction of the substrate temperature. Further, the energy bandgap has found to be 3.46 electron-volts.

1. INTRODUCTION

Historically, the beginning of the 20th century was reported the first preparation of transparent conducting oxide (TCO) thin film. Since that time, it goes under significant developments and considered as one of the most important materials that can be seen in everyday applications such as displays [1-7]. The low resistivity of TCO thin film is the most powerful electrical properties (high conductivity) alongside with high transparency in the visible region of the optical spectrum [8-11]. Such distinguished electrical and optical properties of this material were seen as a highly qualified material to be used in a wide range of optoelectronic applications. Zinc Oxide (ZnO) is one of the transparent conducting oxides that got attention because of its distinct properties. Large band-gap, high conductivity, and high thermal and chemical stability are some of ZnO properties [12 – 18]. With such properties of interests, the ZnO thin films mastered the commercial and scientific research fields.

In order to obtain good piezoelectric characteristics, the ZnO films have to be foremost crystallizing in form of quartzite structure. Therefore, the c-axis orientation of the ZnO thin films is becoming a beneficial material used to prepare bulk and surface acoustic-wave devices [19-20]. Besides, wide band-gap of zinc oxide shows a superb transparent material in the range of visible and ultraviolet spectrum. In addition to its lower toxic effect, it becomes an exemplary element enters in the production of ultraviolet blocking sunscreen.

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According to the deposition conditions, in term of substrate temperature and oxygen pressure, the zinc oxide thin films can be classified into three different groups. Firstly, conductive and opaque film group where the ZnO is mixed with the metallic zinc in one solvent. The second group is alike stoichiometric bulk ZnO in its composition which they are lacking conductivity (insulating) with transparent capability. However, the highest importance of groups is the third group which accumulates both higher conductive ability and excellent transparent properties [21-23]. It is worth to annotate that the electrical properties of TCO films increase effectively as a result of increasing temperature from one side and reduction of oxygen pressure from other side. The reason is of dissociation of cadmium oxide formatting of a solid solution of the metal on its oxide.

2. EXPERIMENTAL PROCEDURE
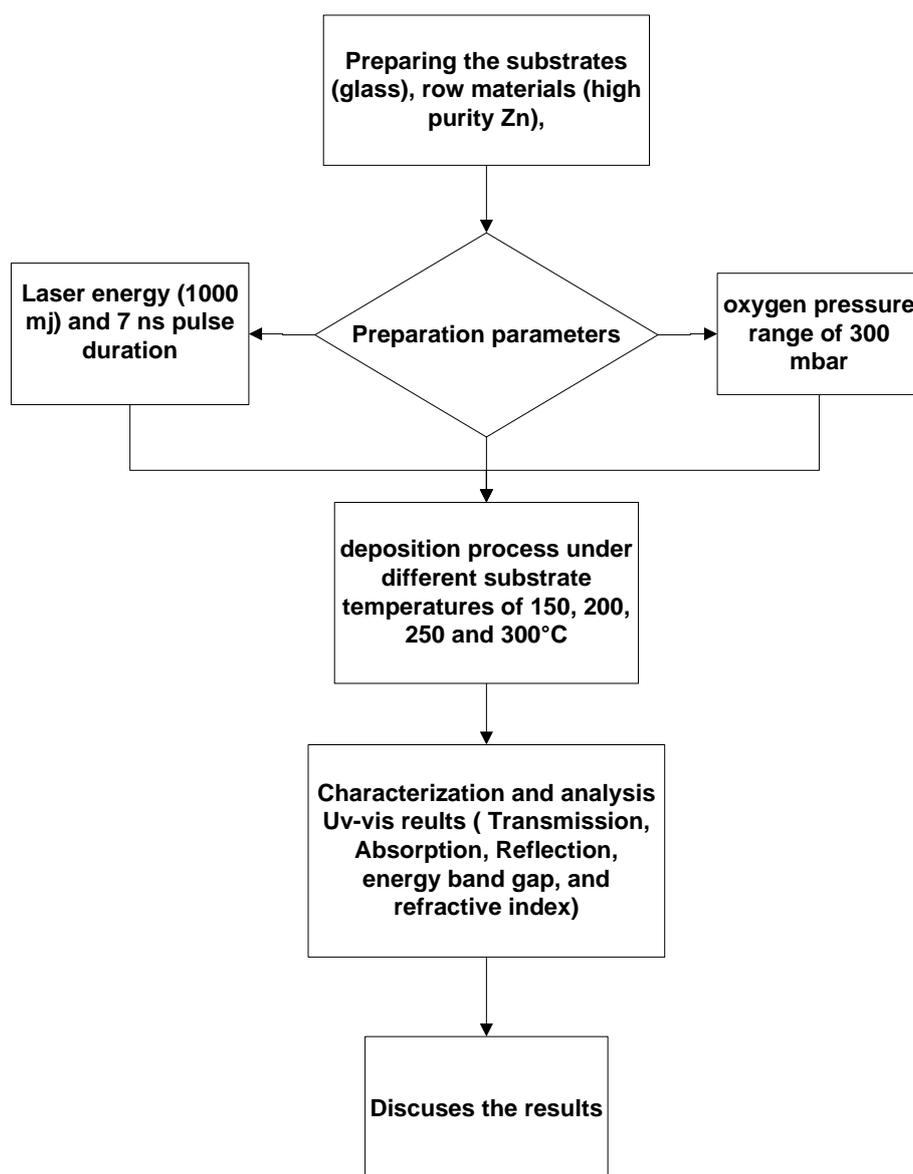
Figure 1. Flowchart of the ZnO nano thin Film preparation process.

A pure zinc oxide (ZnO) was deposited on a glass substrate under oxygen pressure range of 300 mbar. A pulsed Nd:YAG laser source of 1.064 μm wavelength and 7 ns pulse width was firstly

used to ablate the high purity (99.99% purity) zinc. To ensure an accurate ablation for the target material, a lens of 20 cm focal length has been installed in front of the laser source at the angle of incidence of 45 degrees with a rotation speed of 1 rpm. 50 laser pulses are applied to the target to perform the deposition process under different substrate temperatures of 150, 200, 250 and 300°C. All the preparations and characterization processes are presented by the flow chart in the Figure 1.

A 200 nm film thickness was optically recorded using a stylus profilometer. Other optical properties such as transmittance and reflectance were investigated between the spectral the range between 0.3μm to 1μm using double beam spectrometer. The incident photon energy as a function of the wavelength can be obtained using equation (1) [24-26]:

$$E_g(eV)=1240/\lambda(nm) \tag{1}$$

where λ is wavelength. The Tauc equation (2) was used to find the energy band gap as a function of the absorption coefficient and excitation of the transmission [27-29]:

$$(\alpha h\nu)=B (h\nu-E_g)r \tag{2}$$

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) [30-32]:

$$\alpha=2.303(A/t) \tag{3}$$

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) [33-35]:

$$K = \alpha \lambda / 4\pi \tag{4}$$

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) [36-38]:

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

Knowing that R represents the reflectance which can be calculated from (R = 1-T-A), or from equation (6) [39-43]:

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

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

3. RESULTS AND DISCUSSION
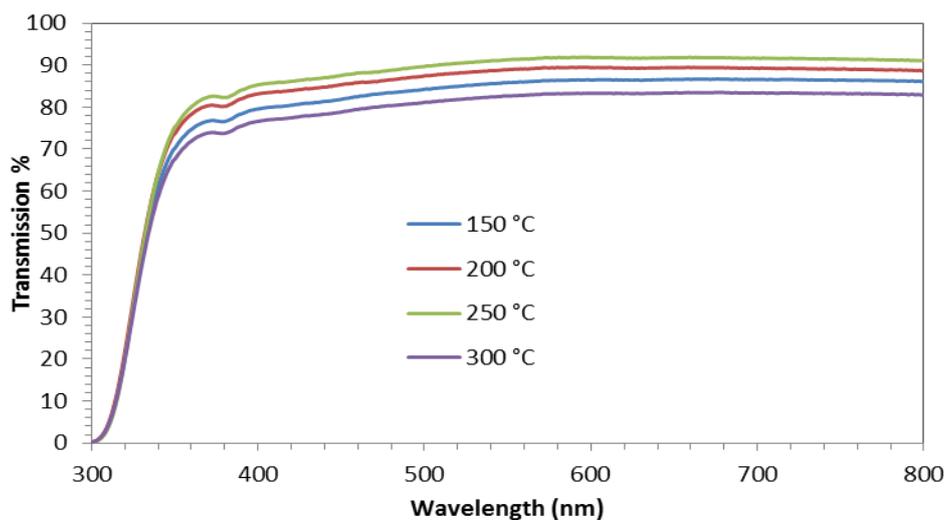
Figure 2. The Transmission curve as a function of the wavelength ranges between 300 nm to 800 nm.

Under different substrate temperatures, the transmission spectra of the prepared thin films have been examined and analyzed. Figure 2 displays the transmission of the prepared films as a function of wavelength. The Figure 2 shows that the average transmission ratio of the prepared samples increases as the temperature of the substrate goes up to 250°C then back down in 300°C as a result of effects the heating on the transparency of thin films [44]. 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 81%. The transmission curves are found to be about 86%, 88%, 91% and 82% for 150, 200, 250 °C and 300°C of temperatures respectively. Furthermore, this change, as a result, of the improved the homogeneity and the uniformity of the structure alongside with the increasing of film thickness and the change in the grains size a very sharp cut-off is also presented at the wavelength 340 nm. Higher transmission values indicate that the prepared thin films are in crystalline form.
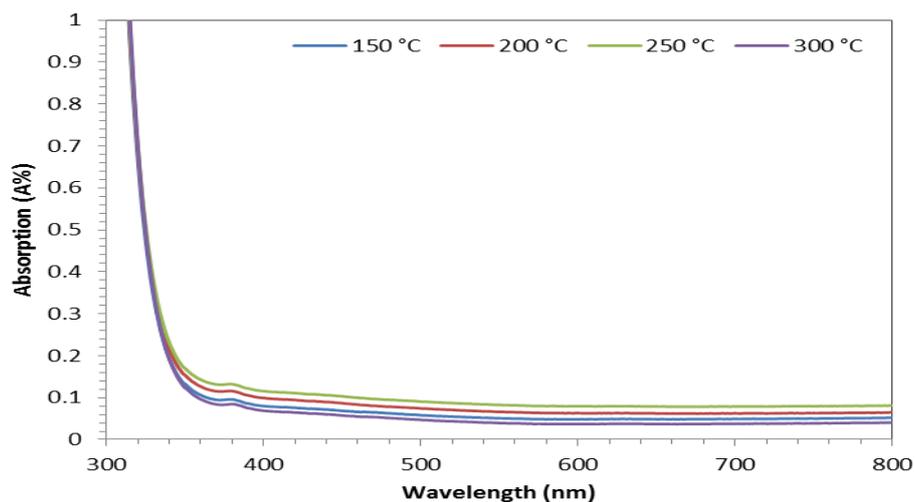
Figure 3. Optical absorption curve as a function of wavelengths under two substrate temperatures.

Figure 3 presents the absorbance spectra of the deposited ZnO thin films under substrate temperatures of 150, 200, 250 °C and 250°C. The 250 °C substrate temperature shows a higher

absorption rate with a notable shift towards lower wavelength region. This indicates that the optical energy band-gap of the prepared films is expanding.
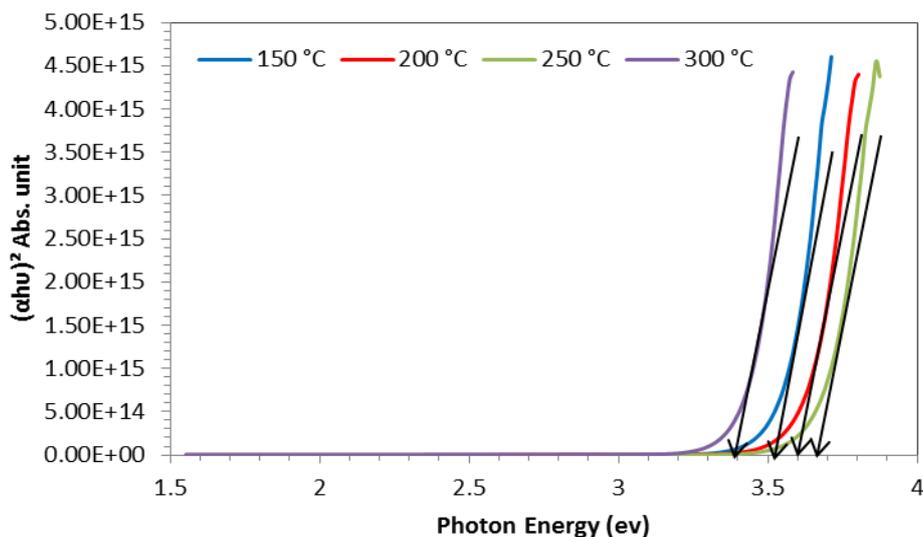
Figure 4. The optical band-gap energy as a function of photon energy for two substrate temperatures.

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 $(\alpha h v)^2$ versus the photon energy given in Figure 4 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.
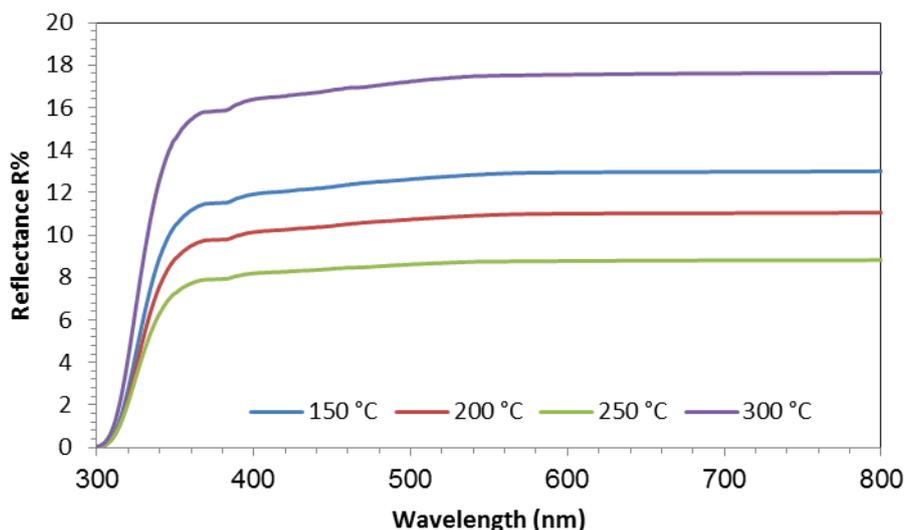
Figure 5. Reflectance as a function of wavelength range under two substrate temperatures.

A double beam ultraviolet-visible spectrometer was used to measure the reflectivity of the prepared ZnO samples. Figure 5 presents the reflectance spectra as a function of wavelength range defined previously. As the substrate temperature increases it is clear to notice that the optical reflection is gradually decreasing. Such reduction comes from the increasing of the 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.
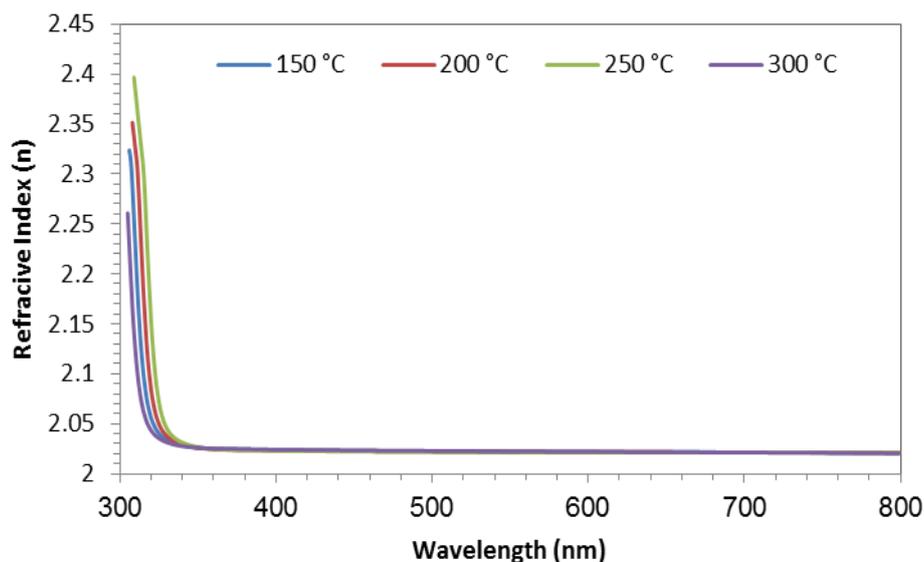
Figure 6. Refractive index as a function of wavelength range for two substrate temperatures.

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

Substrate temperature (C°)	E_g measured (eV)	n measured	$T\%$
150	3.51	2.32	86
200	3.60	2.35	88
250	3.68	2.40	91
300	3.39	2.26	82

The values of optical constants such as the refractive index *n* were calculated using the transmittance data spectrum given in figure (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 2.26 at 300 ᵒC to 2.40 at 250 ᵒC at the wave length of 340 nm as present in figure (6) and tabulated in Table 1. The reason behind the increase 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 increase 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 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.

4. CONCLUSION

ZnO thin films were successfully deposited on glass substrates using low-cost reactive pulsed laser deposition method at different substrates temperature. The effect of substrates temperature on the 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. The maximum value of the transmissions was found approximately 91% at

250 ^{o}C. The value of the refractive index presented the highest value of 2.26, whereas 2.40 that is appropriate for the optoelectronics devices application.

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