PROPERTIES OF MULTILAYERED ZnO/Al/ZnO TRANSPARENT FILM ELECTRODES: MODELS AND EXPERIMENTS

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ABSTRACT
ZnO/Metal/ZnO multilayers have been recognized as good candidates for transparent conductive thin films for application in solar cells and optoelectronic devices. One of the important challenges in the experimental design of such structures is the lack of optimum metal thickness range such as Al in ZnO/Al/ZnO multilayers. The present work overcomes this limitation by investigating how Al thickness affects the optical properties of such multilayered films. In this paper, optical simulations are used to explore the possibility of ultrathin, high transparent ZnO/Al/ZnO multilayer films. MultilayeredZnO/Al/ZnO film with mid-Al layer thicknesses between 1 and 10 nm are shown to have optical transmittances between 70 and 90%. Variations in the ZnO thickness between 10 and 100 nm are shown to have little effect on the optical characteristics of the ZnO/Al/ZnO multilayers, for a given Al thickness. The optimum thicknesses for the ZnO and Al layers are predicted and discussed for the design of transparent conducting ZnO/Al/ZnO electrodes. The reliability of the simulation method is verified by comparing the computational and experimental results for a related system, namely, single-layered ZnO thin film deposited on glass substrates.

Keywords: Absorbance, multilayers, reflectance, TCOs, transmittance

INTRODUCTION

Transparent conducting oxides (TCOs) such as indium oxide, tin oxide (TO) and zinc oxide (ZnO) are being used in the layered structures of active/passive optoelectronic devices and components (Rwenyagila *et al*. 2014). These include applications in solar cells and light emitting devices (Feltrin and Freundlich 2008). However, in some cases the TCO conductivities are too low to be adopted as transparent electrodes (Choi *et al.*1999). They, therefore, require improvements in conductivity before they can be used in applications that require low electrical sheet resistances, while retaining good optical properties (Plà *et al*. 2003). Since indium tin oxide (ITO) is the most widely used TCO, one approach that has been proposed is the use of ITO/metal/ITO systems in the enhancement of electrical conductivity at practical optical spectral transmittance (Rwenyagila *et al.*2014). Such multilayer ITO-metal film structures have been shown to have low electrical sheet resistances and higher optical transmittances at relatively low thicknesses that are lower than those of nano-scale single layered ITO films Klöppel *et al.* 2000). The ITO/metal/ITO multilayer films have also been shown to have better durability than single layered metal films (Jung *et al.*2003). However, because of their relatively low abundance (Feltrin and Freundlich 2008) and the increasing market demand of indium as the principal material in Copper-Indium-Gallium-Diselenide (CIGS) solar cells, indium phosphate (InP) for semiconductor devices and liquid crystal displays, ITO is becoming scarce (Rwenyagila *et al*. 2014) and expensive (Bender *et al*. 1998). This has stimulated increasing interest in the improvement of the electrical and optical properties of ZnO as a replacement for ITO based TCO systems. ZnO is a highly abundant (Song *et al.* 2002), low cost and non-toxic material thatis stable against hydrogen plasma and high temperature processes (Chopra *et al*. 1983, Lin *et al.* 2005).

Recent studies show that ZnO/metal/ZnO films can be tailored to have promising transparent conductive properties with film resistivity values that are comparable to those of metal films and transparency as high as that of ZnO (Rwenyagila *et al.* 2014). The ZnO/metal/ZnO film structure is produced by sandwiching a thin layer of a highly conductive metal between the two embedding ZnO layers. The commonly used metals include: gold, silver, copper and aluminum (Rwenyagila *et al* 2015). Aluminum (Al) is the next metal to copper in terms of conductivity (Rwenyagila *et al*. 2014). It also has low enough resistivity to be used in TCO multilayer structures. Compared to copper, silver and gold, which are for expensive metals, Al is cheap (Rwenyagila *et al.*2015) and has also been shown to give good optical and electrical properties, when used as dopant in ZnO film (Luka *et al.* 2011). There is, therefore, great interest in the development of ZnO/Al/ZnO thin films for applications in large area solar cells and light emitting devices. This paper presents the results of computational modelling and simulations of

ZnO/Al/ZnO sandwich structures. The influence of the thicknesses of layers on the optical reflectance, transmittance and absorbance is presented. The accuracy of the numerical method was verified by comparing the numerical and experimental results on a different but related system, namely, single-layered ZnO thin films on glass. The implications of the results are discussed for potential applications of the model multilayered ZnO/Al/ZnO films in solar cells and light emitting devices.

Theory of Light Transport in Multilayers

Light Absorption

The optical phenomenological behaviour of multilayered film structures is estimated generally by combining Maxwell's equations and the conditions of continuity at the interfaces of the film layers (Gruber *et al.* 2005). If we start with the form of an exponentially damped wave travelling in the positive *z* direction and conventionally define the complex refractive index N for absorbing materials to be:

$$N = n - ik \quad (1)$$

where n is the *real quantity of refractive index* and *k* is the *extinction coefficient*, the solution to the classical wave equation is readily obtained when the phase vector is assumed to be zero (Bennett 1995). This is given by:

$$E = E_0 exp\left[i\omega\left(t - \frac{Nz}{c}\right)\right] \quad (2)$$

Where *z* is the depth covered by the light wave inside the material, *E* is the *electric field vector* of travelling light wave, *t* is *time*, *c* is the *velocity of light*, and ω is the angular frequency. Accordingly, from the solution, the amplitude of the wave associated with the depth *z* from the interface inside the multilayer film can be obtained (Bennett 1995). It is given by:

$$|E| = E_0 exp\left(-\frac{2\pi kz}{\lambda}\right) \quad (3)$$

where λ is the *wavelength of light* in vacuum. The light intensity, which is proportional to the square of the amplitude, is thus exponentially damped by the absorbing film within the multilayer slab, with absorption coefficient, $\alpha = 4\pi k/\lambda$, obtained as the reciprocal of the distance below an interface at which the amplitude of the wave falls to e^{-1} of its value at the interface.

Reflection and Transmission of Light

Based on the classical model, neglecting chemical interactions of the layers at any flat single interface, the incident light splits into transmitted and a reflected part (Gruber *et al.* 2005). For the non-absorbing materials, the perpendicular and parallel intensity reflection coefficients R_s and R_p are obtained by simply squaring the well-known Fresnel amplitude coefficients r_s and r_p. Similarly, the corresponding intensity transmission coefficients T_s and T_p are calculated from the reflection coefficients. They are given by:

$$T_{s,p} = 1 - R_{s,p} \quad (4)$$

However, in real multilayered film structures, light is transmitted through a slab of materials, where it generally undergoes multiple reflections and interference effects. In this case, the above Fresnel definition is of limited usefulness. However, the intensity transmission coefficient of the sample, T_{sample}, for a slab of non-absorbing materials can be calculated by using a modified form of Fresnel equations. For the non-absorbing film layers, therefore, equation (4) is readily modified from the Airy equation (Bennett 1995, Berning 1960), which leads to:

$$T_{sample} = \left[1 + \left[\frac{4R_{s,p}}{(1-R_{s,p})^2}\right] sin^2\theta\right]^{-1} \quad (5a)$$

and

$$\theta = \frac{2\pi n_l d_l cos\theta_l}{\lambda} \quad (5b)$$

Where n_l is the *refractive index* of the material for a given layer, l, within the multilayered film sample, θ_l is the *angle of refraction* and d_l is the *layer thickness*. For given material-layer-refractive indices, wavelengths and angles of incidence, equation (5) predicts that the transmission of the sample can be varied from maximum to minimum values as $sin^2\theta$ varies between zero and one, with controlled changes of multilayer film thicknesses and/or materials properties.

Since the refractive index is a complex quantity, in the case of an absorbing film inside the slab, different approaches have been proposed (Bennett

1995). One of these involves the use of effective refractive indices in which, both N_{ls} and N_{lp} are complex. They are written according to the Bernings' method, which gives:

$$N_{ls} = N_l cos\theta_l \quad (6)$$

$$N_{lp} = N_l / cos\theta_l \quad (7)$$

where N_l is the *complex refractive index* of the material of layer l and

$$cos\theta_l = \left[\frac{(\alpha_l^2+\beta_l^2)^{1/2}+\alpha_l}{2}\right]^{1/2} - i\left[\frac{(\alpha_l^2+\beta_l^2)^{\frac{1}{2}}-\alpha_l}{2}\right]^{\frac{1}{2}} \quad (8a)$$

$$\alpha_l = 1 + \left(\frac{n_{l-1}sin\theta_{l-1}}{n_l^2+k_l^2}\right)^2 (k_l^2 - n_l^2) \quad (8b)$$

and

$$\beta_l = -2n_l k_l \left(\frac{n_{l-1}sin\theta_{l-1}}{n_l^2+k_l^2}\right)^2 \quad (8c)$$

By inserting equations (6) to (8) into equation (5) and into the corresponding Fresnel R_s and R_p formulae, we can readily substitute the effects of multiple absorptions and reflections in a particular 'z' direction into the multilayer transmission model.

MATERIALS AND METHODS

Numerical

The numerical simulations of the multilayered ZnO/Al/ZnO thin film structures were carried out using the Optical Simulator Software, Edition 0.1.8 (CENTURIONI Emanuele, CNR – IMM, Bologna, Italy) (Centurioni 2005). This was used to explore the transparency and the optical properties of model multilayered thin film structures. The Optical Simulator 0.1.8 Software has been developed to realistically simulate the optical characteristics of multilayered film systems (Centurioni 2005). It is a form of modified equation (5) that derives the absorbance, transmittance and reflectance properties of the multilayered films mainly from the refractive indices of the materials and the thicknesses of the film layers. In the case of an absorbing material, the refractive index, which is a complex quantity, is calculated from equation (1). The physical significance of k in the refractive index equation is that on traversing a distance in the film equal to one vacuum wavelength, according to equation (3), the amplitude of the wave decreases by the factor, $\exp(-2\pi k)$. In this study, the layer dimensions and optical characteristics were simulated by building discrete models of discrete film layers within the multilayered ZnO/Al/ZnO thin film structures. The refractive indices of the layers (ZnO and Al) were then incorporated into the optical simulations, within the Optical Simulator Software, which searches for $k$values that satisfy the refractive index equation (1). For a given transmittance, *T*, and reflectance, *R*, the ratio $\xi$is given by:

$$\xi = \frac{T_{s,p}}{1-R_{s,p}} \quad (9)$$

Furthermore, ξ is known to be strongly dependent on *k* (Summonte 1993). Note that, since ξ does not oscillate with photon wavelength (Summonte 1993), the Optical Simulator Software uses equations (5) to (9) to construct the optical reflectance, transmittance and absorbance plots of a given multilayered film system. In this work a four-layered ZnO/Al/ZnO/glass film model was used. The Optical Program was integrated with few refractive indices of different materials. The indices were modified in this work to explore and approximate the optical characteristics of the ZnO/Al/ZnO thin film structures, with well controlled layer thicknesses. The analysis involved modification of the aluminum optical index baseline file created earlier by Schubert (2004). All the layer thicknesses were then varied between 1 nm and 100 nm, while keeping the refractive index of ZnO fixed at 2.02 (Pearton *et al*. 2005).

Experimental

The experimental ZnO film was deposited from a ZnO ceramic target(99.99% purity, Target materials Inc., Columbus, Ohio, USA), having a diameter of 10 cm. Prior to ZnO film deposition, the magnetron chamber was pumped down to below 3×10^{-5} Torr. Film production involved magnetron sputtering on a thoroughly cleaned glass substrate using 100 W Radio Frequency (RF) deposition power. This was done in a pure argon chamber at room temperature and a pressure of 6×10^{-3} Torr. After deposition, the structure and thickness of the films were checked and confirmed using a conventional $\theta - 2\theta$ xpert-pro X-ray diffractometer system (PANalytical B. V., ALMELO, Netherlands) using wavelength of $1.540598 Å Cu\ K_{\alpha 1}$ radiation and a Veeco Dektek 150 Stylus Surface profiler machine, respectively. The X-ray diffraction pattern was obtained from ZnO film sample after annealing in a carbolite tubular furnace at 400°C in oxygen atmosphere for 90 minutes. The experimental measurements of the optical reflectance and transmittance were obtained from a 50 nm ZnO film deposited on glass. This was measured in the wavelength range 200 - 900 nm using an AvaLight-DHc UV-VIS spectrophotometer that was manufactured from Avantes Inc., Broomfield, USA.

RESULTS AND DISCUSSION

Effect of Al Thickness on the Optical Properties of ZnO/Al/ZnO Film

The results of the optical simulations of multilayered ZnO/Al/ZnO film structures are presented in Figures 1–3. The results show the dependence of the optical transmittance, absorbance and reflectance on Al interlayer film thickness. Figure 1 shows the effects of Al thicknesses on the optical properties of multilayered film structures with ZnO thickness fixed at 50 nm, for each of the embedding upper and lower ZnO layers. The results show that a minimum Al layer thickness of between 1 and 10 nm is required for the multilayered ZnO/Al/ZnO film structure to transmit between 68 and 89% of the incident photons. The results also show that both reflectance and absorbance increase with the Al thickness. Hence, to avoid reflecting and absorbing most of the incident light photons, it is important to use mid-Al layers with ultrathin thickness values. However, the Al interlayer thicknesses between a double layered ZnO films must be sufficiently thick to avoid major resistivity limitations (Rwenyagila *et al*. 2014). The design of the multilayered ZnO/Al/ZnO thin films, therefore, requires a balance between conductivity, transmittance and multilayer processing and/or fabrications.

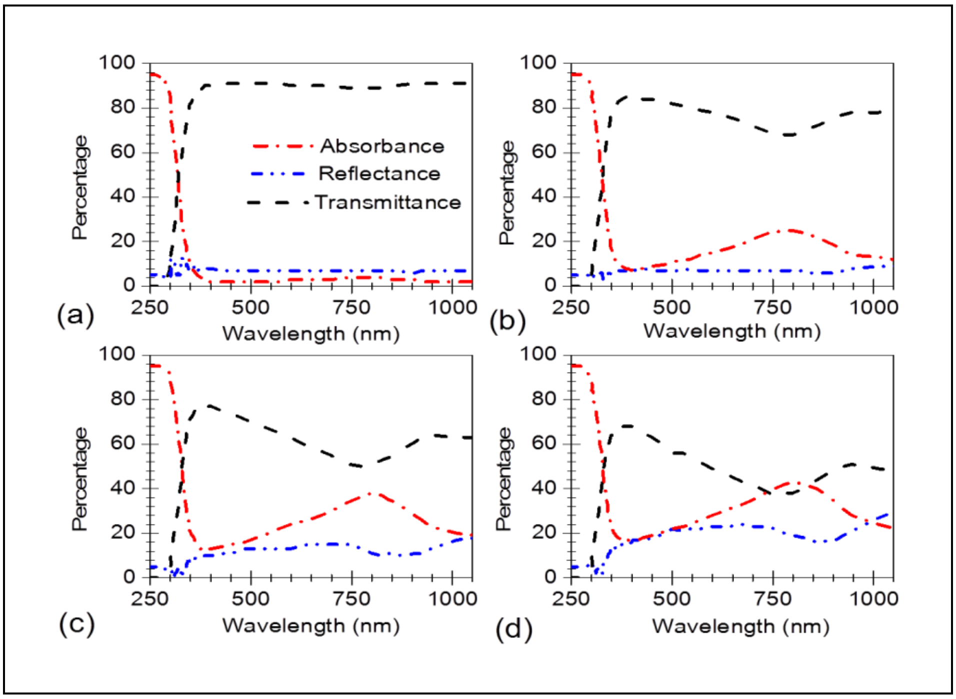

Figure 1: Simulated optical properties of ZnO/Al/ZnO films for different mid-Allayer thicknesses of: (a) 1 nm (b) 10 nm (c) 20 nm (d) 30 nm embedded in 50 nm ZnO layers.

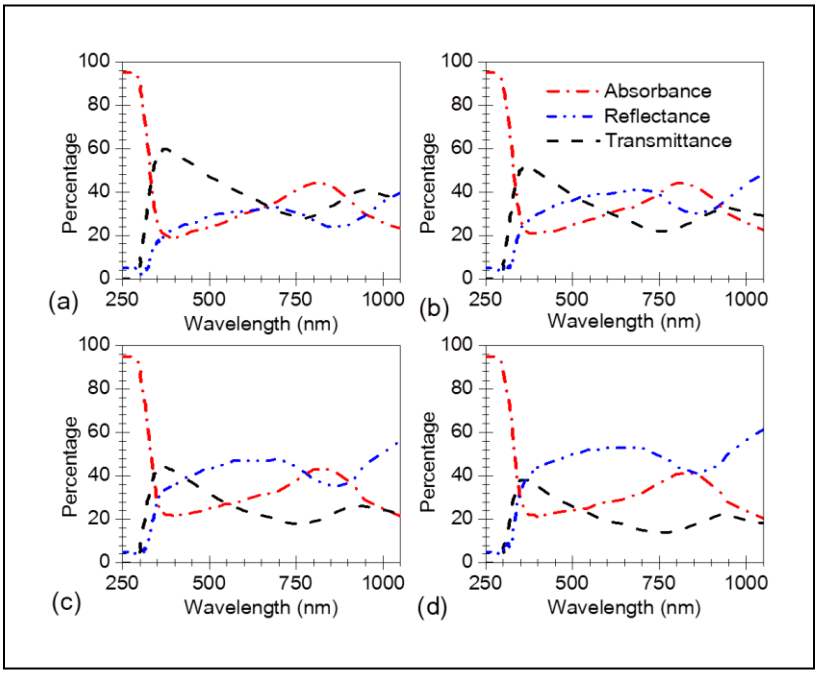

Figure 2: Simulated optical characteristics of ZnO(50 nm)/Al/ZnO(50 nm) for: (a) 40 nm, (b) 50 nm, (c) 60 nm and (d) 70 nm mid-layer Al thicknesses.

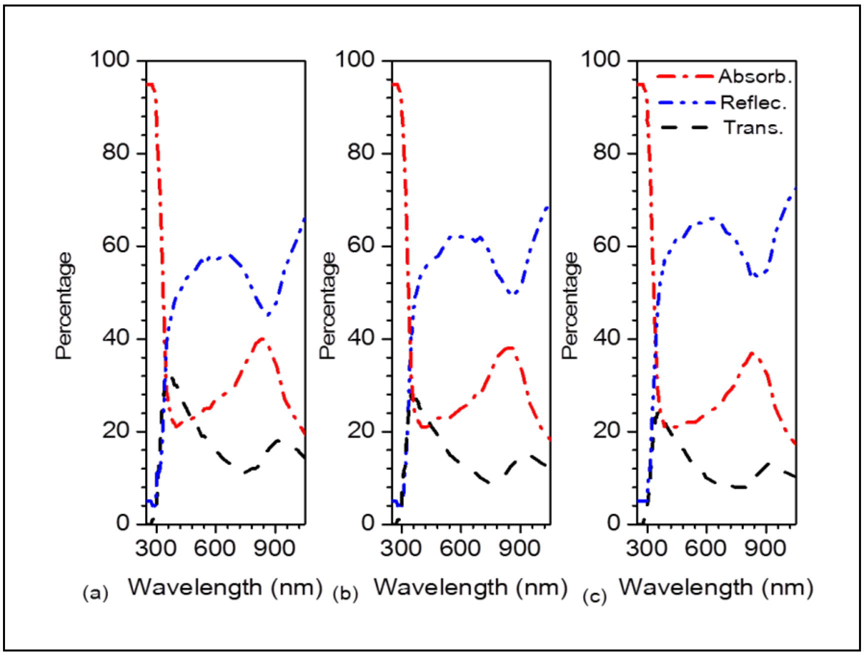

Figure 3: Simulated dependence of the optical properties of ZnO(50 nm)/Al/ZnO(50 nm) film on different Al thicknesses of: (a) 80 nm (b) 90 nm (c) 100 nm.

The simulated results for the thick Al interlayers between 40 and 100 nm are presented in Figures 2 and 3. It is clear from these plots that, any attempt to increase the thickness of mid-Al layer leads to increased light absorption and reflection values. This can be associated to the high optical reflection and absorption coefficients of Al as a result of its metallic complex refractive index and the nearly zero optical band gap energy. Hence, increasing Al thickness results in a serious deterioration of the

ZnO/Al/ZnO multilayer film transmission quality for the Al layer thicknesses between 10 nm and 100 nm. However, for Al thicknesses between 1 and 10 nm, the multilayer film reflection and absorption are low and less significantly affected by the changes in the Al interlayer thickness.

The effects of Al interlayer thicknesses between 1 and 10 nm were also explored for a fixed ZnO double-layer thickness of 50 nm. The result is presented in Figure 4. It is observed that, for Al interlayer thicknesses between 1 and 3 nm, the multilayer film transmission efficiency is not significantly affected by changes in Al layer thickness. Furthermore, the predicted high values of optical transmittances for the multilayered ZnO/Al/ZnO films with ultrathin Al layer between 1 – 3 nm are generally desirable for the improved optical performance of the transparent electrode films. However, the ultrathin mid-Al layer (between 1–3 nm) might have detrimental effects on the conductivity of the model transparent electrodes. Hence, increasing the Al interlayer thicknesses to between 3 and 10 nm might be necessary to ensure the required balance between conductivity, absorbance and transmittance for practical applications of the model multilayers as transparent electrodes for applications in electronics, optoelectronic devices and their components (Rwenyagila *et al.* 2014).

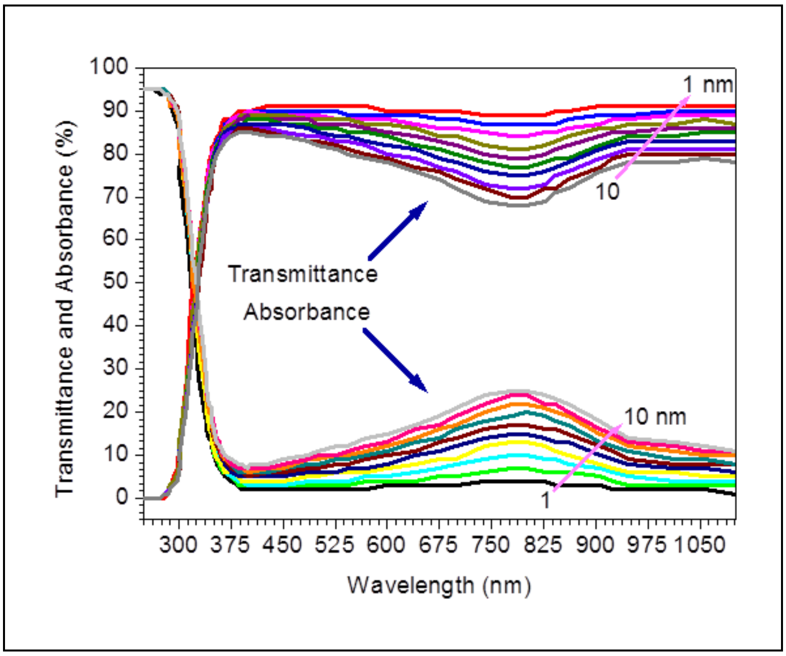

Figure 4: Simulated Transmittance and absorbance for ZnO/Al/ZnO films with Al interlayer thickness between 1 and 10 nm embedded in 50 nm ZnO layers.

Fig. 5 shows the simulated transmittances obtained for the transparent multilayered ZnO (25nm)/Al/ZnO (25nm) film structures with Al interlayer thicknesses purposely based on 3-8 nm. Within this range, the average multilayered film transmittance values are between 79 and 84%, while the average absorbance values are between 9 and 13%. Moreover, the predicted thickness values in thinner mid-Al interlayer films (between 3 – 8 nm), for wavelengths between 400 and 900 nm are generally in good agreement with earlier experimental results obtained for other multilayered ZnO/metal/ZnO systems with similar interlayer (Rwenyagila *et al.* 2015). This indicates that such thicknesses correspond to practical mid-Al interlayer thickness range at which the transmittances are sufficient for potential applications of the multilayered film structures in solar cells and light emitting devices. It also suggests that, the ZnO/Al/ZnO films may be fabricated as alternatives to the costly ITO based anodes in solar cells and light emitting devices and components.

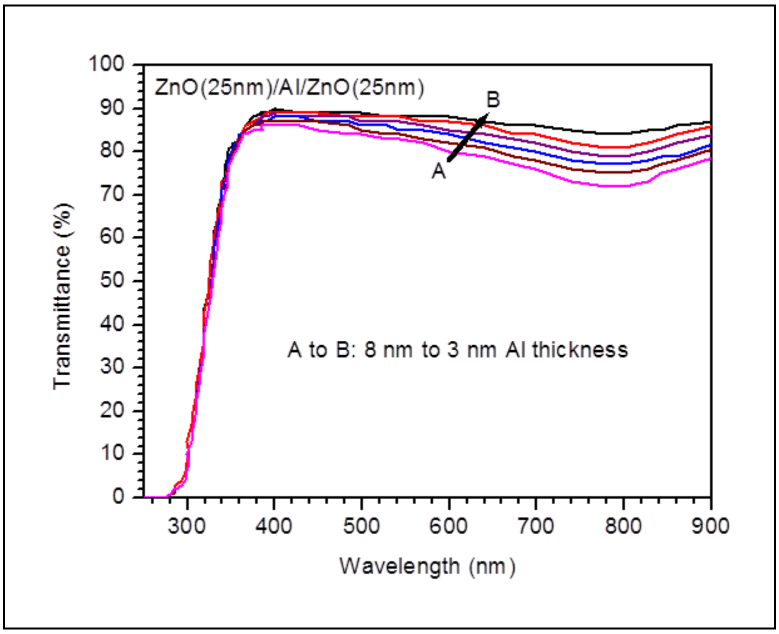

Figure 5: Simulated optical transmittance curves illustrating the dependence of ZnO(25nm)/Al/ZnO(25 nm) multilayer on the thickness of Al interlayer between 3 and 8 nm.

Effects of ZnO on the Optical Properties of ZnO/Al/ZnO Film

Figure 6 shows the predicted effects of ZnO film thickness on the optical properties of multilayered ZnO/Al/ZnO film structures. In the case of structures with fixed mid-Al thickness of 5 nm, the transmission output characteristics of the model multilayered film structures are nearly independent of ZnO thickness between 10 and 100 nm. These can be explained by the lower optical absorption and reflection coefficients of ZnO, compared to those of aluminum. The associated absorbance and reflectance (not shown in this paper) were also low and not significantly affected by changes in ZnO thickness from 10 to 100 nm.

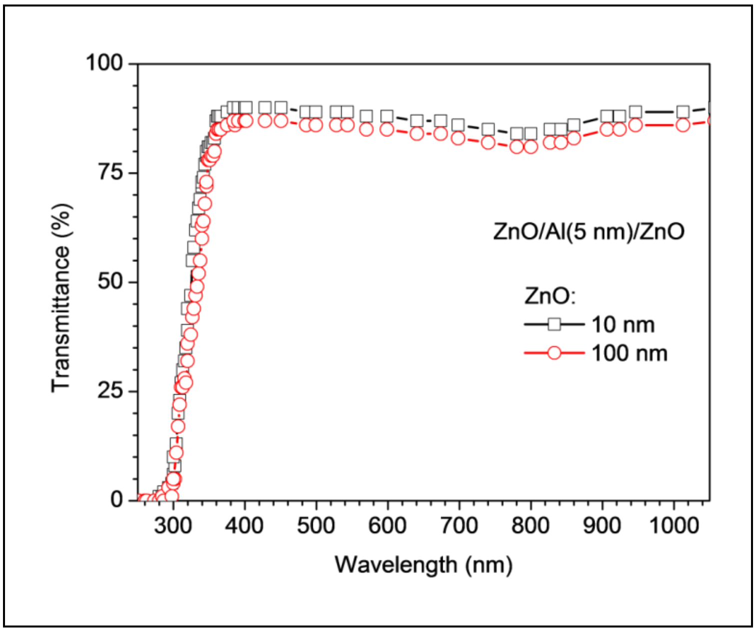

Figure 6: Effects of ZnO thickness on the transmittance of ZnO/Al/ZnO multilayer.

Experimental Validation of Numerical Model

In order to test the validity of the numerical method used in the simulations, the numerical simulations were compared with experimental measurements that were performed on pure ZnO thin film deposited on glass. Figure 7 shows the XRD pattern that was obtained after annealing the ZnO film samples at 400°C for 90 minutes in air. Clearly, the XRD results show that the ZnO films are crystalline with preferred Wurtzite (002)-oriented ZnO crystallite structures. The XRD pattern (Figure 7) is also in good agreement with published diffraction patterns for ZnO films (Song *et al.* 2002, Rwenyagila *et al.* 2014).

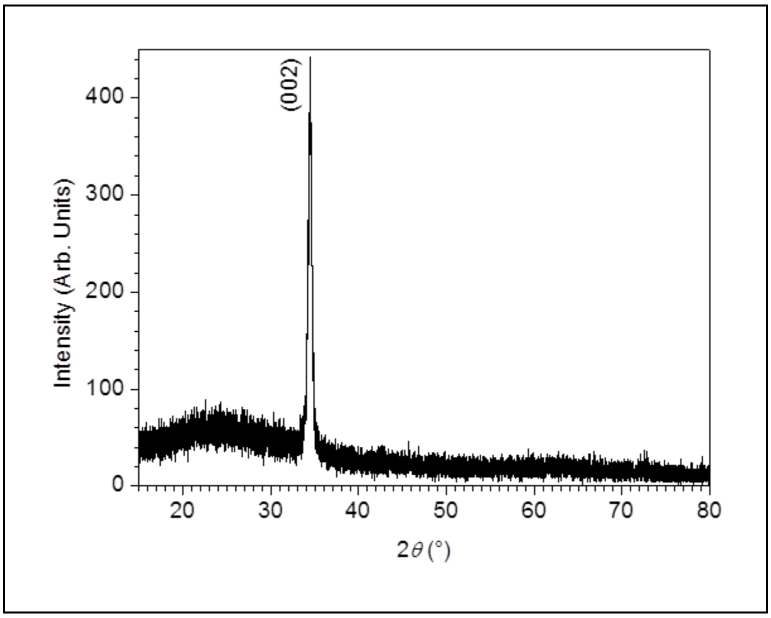

Figure 7: XRD pattern of 50 nm ZnO thin film deposited on glass after annealing for 90 minutes at 400°C in air.

Optical Transmission of ZnO Thin Film: Model and Experiment

Figure 8 presents the experimental and simulated transmittance and reflectance profiles for a single-layered 50 nm thick ZnO film on glass substrates. In comparison, both simulated and experimental optical transmittance and reflectance spectra were generally in agreement. Furthermore, the agreement between the predicted and measured optical transmittance values was generally good in the high wavelength region of the spectra with wavelengths between 400 and 900 nm. The differences observed between the two profiles, in this region of the spectra can be attributed to artefacts in the spectrophotometer used. For example, based on the working mechanism of spectrophotometers, which is described in the theory section by equation (5), light undergoes generally a π phase shift upon reflections at interfaces (Bennett 1995). Such phase shifts were not captured in the simulations and can readily cause small discrepancies between the simulations and the experimental optical profiles. However, there are also inconsistencies between the simulated and measured transmittance values in the lower region of the spectra with wavelength between 280 and 340 nm. These differences can be attributed to the role of defects in the experimental ZnO films. For example, it is well known that the non-ideal structure of experimentally prepared materials can affect considerably their optical constants (Bennett 1995, Rwenyagila *et al.* 2015). As consequences, the optical band gap energy and the absorption characteristics of the experimentally prepared ZnO thin film becomes slightly different to those of a perfect ZnO crystal (Rwenyagila *et al*. 2014), that were assumed in the simulations of the ZnO/Al/ZnO films.

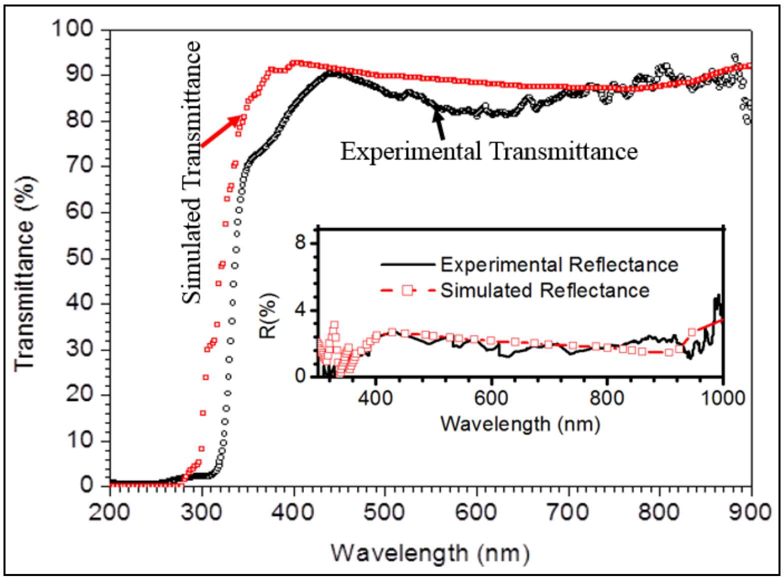

Figure 8: Experimental and simulated optical properties illustrating the transmittance and reflectance (insert Figure) profiles for a 50 nm ZnO film on glass substrates.

Implications of the Results

The practical implications of the current work are quite significant. First, experiments Klöppel *et al.* 2000, Rwenyagila *et al.* 2014) have revealed that TCO/metal/TCO sandwich structures can significantly enhance the figure of merits and the performance of TCOs. As a result one of the important challenges in the experimental design of such structures is the lack of optimum metal thickness range such as Al in ZnO/Al/ZnO samples. The present work overcomes this limitation by investigating how Al thickness affects the optical properties of such multilayered films. The numerical results obtained for a related film system, namely, single-layered ZnO thin film deposited on glass substrates were generally in agreement with the actual experimental transmittances and reflectance of the model film. This suggests that for the ZnO and the other multilayered films with known optical constants, the numerical method presented in this paper can be used to obtain realistic designs of optical properties of nano-films. However, further work is required to test the optical and electrical performance of the model composite ZnO/Al/ZnO film in actual solar cells and light emitting devices. Such work should explore both the electrical models of the ZnO/Al/ZnO films along with measurements of the electro-optical properties of the multilayer, and the current-voltage characteristics, fill factors, power conversion and external quantum efficiencies, degradation mechanisms and the long term performance characteristics of solar cells and light emitting devices fabricated with ZnO/Al/ZnO composite films on glass substrates. These are clearly some of the challenges and opportunities for future work. Such work is particularly important due to the limited abundance and availability of indium (used in ITO films) for the wide range of applications in layered electronic structures and components.

CONCLUSIONS

The results of computational simulations of the optical properties of a four layered ZnO/Al/ZnO/glass thin film structure were discussed. These structures have the potential for applications as improved TCOs in solar cells, light emitting diodes and other optoelectronic devices and components. Predictions of absorbance, reflectance and transmittance are obtained for Al interlayer film with thicknesses between 1 and 100 nm. The results suggest that lower thicknesses of Al interlayer result in improved spectral transmittance of the model multilayer ZnO/Al/ZnO film. However, from considerations of both optical and electrical limitations, a highly transmitting thin film structure system with ultrathin Al interlayer thicknesses between 3 and 8 nm is proposed. Within this range, the optical simulations showed that variations in the Al thicknesses result in reasonable transmittances between 79 and 84%. The simulated results obtained for pure 50 nm ZnO film on glass substrate are consistent with experimental transmittance and reflectance measurements in single-layered ZnO of the same thickness.

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