

# Annealing temperature effect on the aluminium doped ZnO films for transparent electronics

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Transparent conducting Al-doped ZnO thin films were prepared on glass substrates by spin coating method. The effects of an annealing treatment on electrical and optical properties were investigated for 1.5 atomic percentage of aluminum. The average optical transmittance of 90 % in the visible range was obtained exploring the potential of these films for transparent electronics. The minimum electrical resistivity of  $0.247 \Omega \text{ cm}$  was obtained. We realize the optical band gap of 3.3 eV for 1.5 atomic percentage of aluminum.

(Received August 1, 2008; accepted August 14, 2008)

*Keywords:* Spin coating, Al doped ZnO, Transparent electronics

## 1. Introduction

Transparent conducting oxide (TCO) films are preferably used in optoelectronic devices such as touch panels, flat panel displays (FPD), and thin film solar cells [1,2]. For display applications, TCO films of high optical transmittance in the visible region and high electrical conductivity are required. Indium tin oxide (ITO) has been commercially used for the flat panel display industry [3, 4]. However, impurity-doped zinc oxides (ZnOs) are widely accepted as an alternative for ITO-based TCO because of the advantages of low cost, resource availability and nontoxicity. Zinc oxide is accepted as wide band gap semiconductors as an alternative to GaN because it has band gap of 3.37 eV at room temperature. Besides this, it has a large free exciton binding energy (60 meV), and high mechanical and thermal stabilities. With enhancement in the quality and control of conductivity in bulk, ZnO found its potential as a material for short wavelength light emitters and transparent electronics [5-7].

Generally, undoped ZnO thin films typically show evidence of n-type conduction. In particular, an Al-doped ZnO transparent conductive oxide (TCO) film has high transmittance in the visible region, and a low resistivity, and by doping it with Aluminum its optical band gap can be controlled [8,9]. However, Al-doped ZnO shows excellent transparency over the entire visible spectrum and has better transport properties than tin oxide due to higher electron mobility. There are several methods used for the fabrication of ZnO thin films such as pulsed laser deposition [10], sputtering [11-12], molecular beam epitaxy [13], and sol-gel process [9]. Among these methods, the sol-gel technique has several advantages over the others such as low cost, simplicity; homogeneity and large area films can be prepared at lower temperature.

In the present work, aluminum-doped zinc oxide films are deposited on glass substrate using sol-gel spin coating

process. Many factors can affect the properties of aluminum doped ZnO films prepared by sol gel method. Among them, Aluminum composition and annealing temperature have significant effect on the optical and electrical properties of the film. We have already reported effect of aluminum doping on the properties of the films [9]. Here, we have investigated effect of annealing temperature on the Aluminum doped ZnO films prepared by sol-gel method on the glass substrate by considering 1.5 atomic percentage of Aluminum.

## 2. Experimental procedure

In our process, 2-methoxyethanol and ethanolamine were used as solvent and reagent respectively and the aluminum nitrate nonahydrate was used as the doping element. The zinc acetate and aluminium nitrate nonahydrate were mixed together in 2-methoxyethanol. The mixture was vigorously stirred on hot plate with magnetic stirrer maintained at  $\sim 600^\circ\text{C}$  and the ethanolamine was added drop wise in the solution. After stirring for half an hour, the solution became fully transparent. The solutions were prepared for the 1.5 at% of aluminum. The obtained solutions were used to deposit thin films. The Al doped ZnO thin films were deposited on the glass substrate. The deposition process was carried out on spin coater maintained at constant rotation. It was ensured that the constant thickness of coating could be attained per coat. The samples were coated repeatedly for the six times. After each coating, the sample was heat treated at  $350^\circ\text{C}$  for three minute and cooled down at room temperature before applying new coat. This preheat treatment is necessary for evaporation of the organic group contents present in the film. These deposited samples were annealed at different temperature in open air for one hour to investigate annealing temperature effect on the electrical and optical properties of Al doped ZnO films.

### 3. Results and discussion

In the light of emerging application of aluminum doped ZnO for transparent electronics, the most significant results have been reported here to investigate effect of annealing temperature on the electrical and optical properties of the films. Fig. 1 shows effect of annealing temperature on the resistivity of the aluminum doped ZnO films for 1.5 atomic percentage of aluminum. It reveals that the resistivity of the films is continuously increasing with increase in the annealing temperature. This increase in the resistivity has been attributed to the incorporation of oxygen atoms during the annealing process [14]. It results in the reduction of the electron concentration in the aluminum doped ZnO films. According to the scattering mechanism of transparent conducting described by Pei et. al. [15], ionized impurity scattering seemed to be dominant in the low temperature range. At the high temperature range, the grain boundary scattering introduces the increase in the resistivity and thereby reduces the mobility of the carriers. For the temperature range between 350°C to 450°C, the resistivity value was slightly increased from 0.247 to 1.23 Ωcm. At 500 °C, the resistivity was 9.05 Ωcm. This sudden increase in resistivity at 500 °C was also appeared due to the glass substrate. As glass substrate gets softens at this temperature, the impurities gets added in to the film and degrades the properties of transparent conducting film [5].

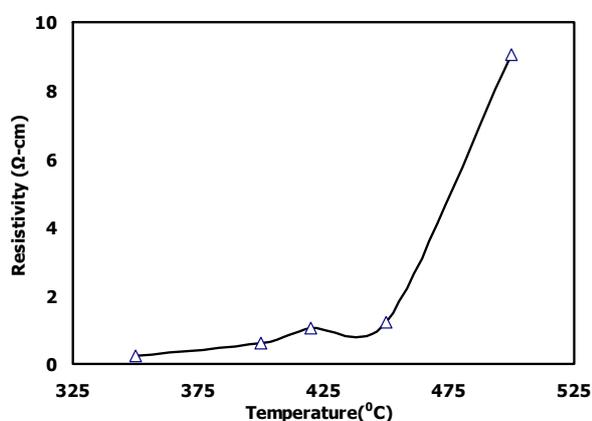


Fig. 1. Annealing temperature effect on the resistivity of the films.

The optical transmittance spectra of Aluminum doped ZnO films were recorded between 350 to 800 nm wavelength ranges. Fig. 2 shows the optical transmission spectra of aluminum doped ZnO film annealed at different temperatures. The oscillations appeared in the spectra did principally due to the interface effect owe to reflection at the interface of film and the substrate. The sharp fundamental absorption edges were observed in all the spectra and average transmission of 90 % has been perceived in the visible range. It conforms that, the deposited Aluminum doped ZnO films having good surface quality and homogeneity [6]. It indicates that the

films were weakly absorbing and the substrate was completely transparent at the visible wavelength range.

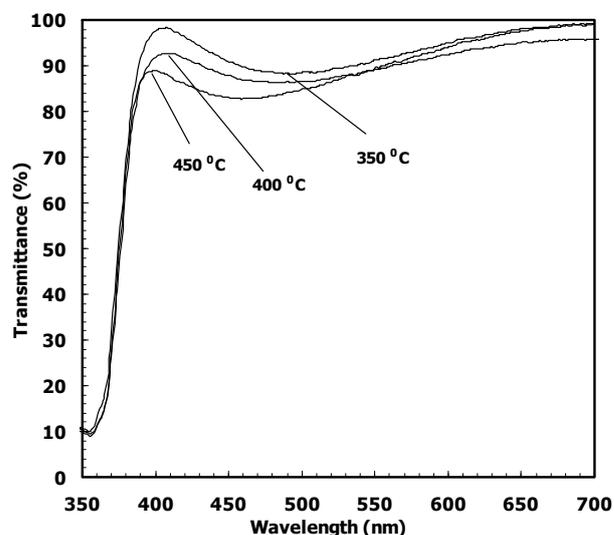


Fig. 2. Transmission spectra of Al doped ZnO films (350-700 nm).

With the help of envelope method, the refractive index of these aluminum doped ZnO films were determined [16] and are depicted as a function of wavelength in the Fig. 3 for different annealing temperatures. It was realized from our results that the refractive index of the films for all the annealing temperatures decreases with increase of wavelength. The absorption coefficient was determined as a function of wavelength as illustrated in Fig. 4 for different values of annealing temperatures. The little variation was found in the absorption coefficient value due to the effect of annealing temperature. The optical band gap energy was deduced for each aluminum doped ZnO film. Fig. 5 shows the graph of optical band gap energy as a function of annealing temperature.

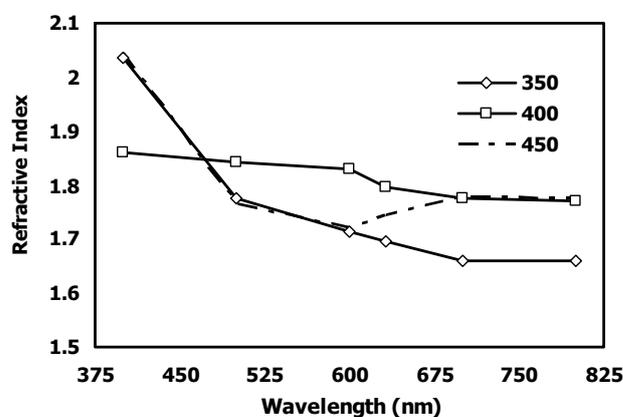


Fig. 3. Refractive index versus wavelength of Al doped ZnO films.

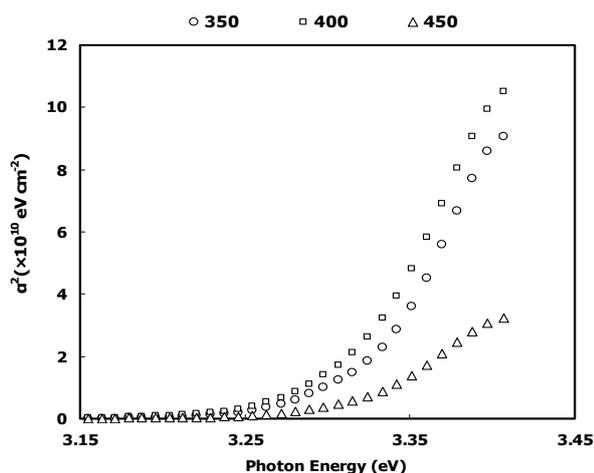


Fig. 4. Absorption coefficient versus photon energy for 1.5 at% of Al.

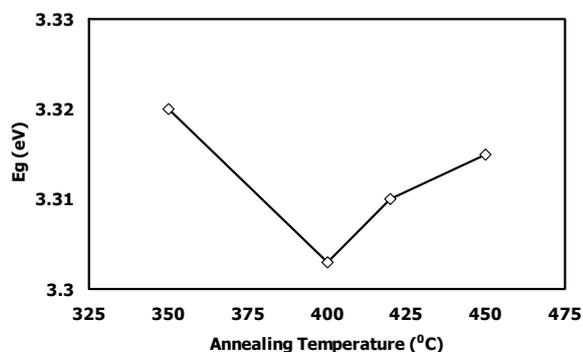


Fig. 5. Variation of band gap with annealing temperature.

The little increase in the band gap was found for deposited aluminum doped ZnO films with an effect of annealing temperature.

#### 4. Conclusions

The highly transparent films of aluminum doped ZnO were successfully deposited on glass substrate by sol gel method for 1.5 atomic percent of aluminum at various annealing temperature using 2-methoxyethanol. Films showed good optical transparency in the visible range. Little increase in the band gap was observed with respect to change in the annealing temperature. The increased annealing temperature results in the increase in resistivity. The minimum value of resistivity obtained was 0.247 Ωcm. We have investigated effect of annealing temperature on the electrical resistivity and the optical properties of the films to explore the potential of aluminum doped ZnO films for the transparent electronics.

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