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Study on the electrical and optical properties of Ag/Al-doped ZnO coatings deposited by electron beam evaporation

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Abstract

A layer of silver was deposited onto the surface of glass substrates, coated with AZO (Al-doped ZnO), to form Ag/AZO film structures, using e-beam evaporation techniques. The electrical and optical properties of AZO, Ag and Ag/AZO film structures were studied. The deposition of Ag layer on the surface of AZO films resulted in lowering the effective electrical resistivity with a slight reduction of their optical transmittance. Ag (11 nm)/AZO (25 nm) film structure, with an accuracy of ±0.5 nm for the thickness shows a sheet resistance as low as $5.6 \pm 0.5\ \Omega$/sq and a transmittance of about $66 \pm 2\%$. A coating consisting of AZO (25 nm)/Ag (11 nm)/AZO (25 nm) trilayer structure, exhibits a resistance of $7.7 \pm 0.5\ \Omega$/sq and a high transmittance of $85 \pm 2\%$. The coatings have satisfactory properties of low resistance, high transmittance and highest figure of merit for application in optoelectronics devices including flat displays, thin films transistors and solar cells as transparent conductive electrodes.

Keywords: Al-doped ZnO; Ag; TCO; Optical and electrical properties

1. Introduction

Nowadays Al-doped ZnO thin films are investigated as transparent conductive electrodes for use in optoelectronics devices including flat displays, thin films transistors, solar cells because of their unique optical and electrical properties [1–3]. For the use as transparent conductive electrodes, a film has to have low resistivity, high absorption in the ultra violent light region and high optical transmission in the visible region. Different technologies such as electron beam evaporation, chemical vapor deposition, laser evaporation, DC and RF magnetron sputtering [4–8] have been reported to produce thin films of AZO with adequate performance for applications. In order to optimize the optical and electrical characteristics, these techniques usually are applied in combination with temperature annealing (during or after deposition process) [9–11]. Annealing procedures increase the optical transmittance and reduce the defects of the crystalline structures (vacancies and interstitial impurities). Then, free carrier density can be reduced and hence the reduction of film electronic conductivity can occur. In order to increase the electronic conductivity and also to retain the high transmittance, thin silver layer can be deposited on AZO. In this study we select the silver (Ag) film coated AZO because the Ag starts with a low resistivity of $2 \times 10^{-6}\ \Omega$ cm. The sheet resistance of an Ag layer proportionally decreases with the Ag thickness. However, thicker Ag films deteriorate optical properties in the visible wavelength region such as transmittance, reflectance and reflective color. To find a way out of these difficulties, optimization of the electrical resistivity of Ag thin film along with optical properties is essential for a practical use. Recently, it is also known that electrical resistivity of Ag thin film changes with its under layer materials; the resistivity of Ag becomes low when Ag is deposited on ZnO undercoats [12]. We investigated the effect of AZO undercoats on the electrical resistivity of Ag thin films. The study of multilayer system Ag/AZO produced by e-beam allowed us to determine the optical characteristics of the AZO films and to produce a transparent and conductive electrodes on highly transparent glass substrates.

2. Experimental

Thin films of Ag/AZO structures were deposited on glass substrates (corning eagle 2000 glass) in an e-beam evaporation

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system. The multilayer films were successively formed on glass substrates without vacuum break using an Al-doped zinc oxide sintered target [ZnO (OSAKA, 99% purity) doped with 2 wt% Al_2O_3 (Alcoa, 99.7% purity), pressed and sintered at 1400 °C for 2 h] and metal Ag chips (99.999% purity). The e-beam chamber was pumped down to 9×10^{-7} Torr prior to deposition. The substrate temperature was measured using a thermocouple gauge. The variation of substrate temperature during deposition was maintained within ±5 °C. Substrate temperature was controlled in the range 50–250 °C. Deposition of AZO and Ag films were performed at a pressure of 2×10^{-5} Torr in the evaporation chamber. AZO films were deposited at 4 kV and 20–30 mA at a substrate temperature of 200 °C. Ag films were deposited at 8 kV and 20 mA at a substrate temperature of 30 °C. Specimens with different thickness fabricated at different conditions were prepared and characterized.

The thickness of the film was measured using a surface profiler (Alpha-step 500, TENCOR, USA) and on line thickness measurement system which was further confirmed by cross-sectional SEM observation within an accuracy of ±0.5 nm. Conventional θ–2θ XRD studies on the films were carried out in Regaku (D/MAX 2500, Japan) diffractometer using Cu Kα radiation to investigate the crystalinity and crystal orientation of the films. Surface morphology was observed by field emission scanning electron microscope (FESEM, XL-40, The Netherlands). Sheet resistance was measured using four-point probe method. Optical transmittance was measured in the range 300–800 nm by UV–Vis–IR spectrophotometer (Hewlett Packard 8452A, Palo Alto CA, USA).

3. Results and discussion

Fig. 1 shows the XRD spectra of AZO (25 nm) and Ag/AZO (25 nm) films for different thickness of Ag layer. The spectrum of the as deposited AZO film exhibit the existence of (0 0 2), (1 0 3), (1 0 2) ZnO crystal planes. There is no Al diffraction peak appeared in the pattern. However, in the spectrum of Ag/AZO film, silver has (1 1 1) orientation and there is a small shift of ZnO peaks towards lower diffraction angle. Shift of (0 0 2) peaks towards the lower diffraction angle with the addition of Ag layer indicates that there is small distortion of crystallites

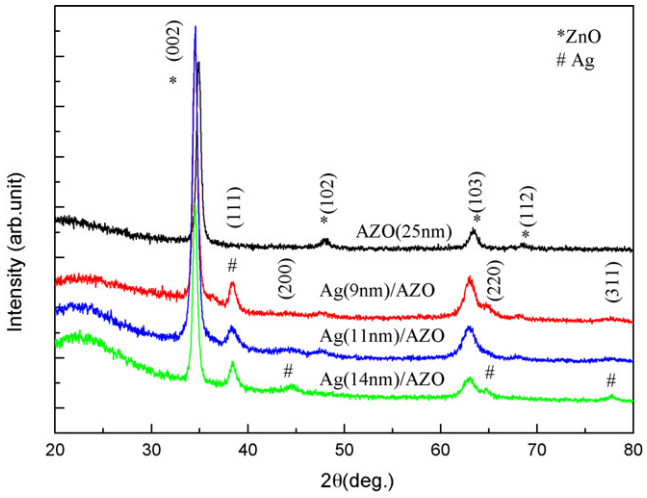

Fig. 1. XRD patterns of AZO and Ag/AZO thin films for fixed thickness of AZO layer (25 nm).

[13,14]. With the increase of thickness of Ag layer, more intense (2 0 0), (2 2 0), (3 1 1) Ag peaks are also developed. Addition of Ag layer enhanced the polycrystalinity of AZO layer. The oriented Ag films with their (1 1 1) planes parallel to the substrate can be deposited on preferentially grown AZO undercoats. The SEM cross-sectional view of the Ag (11 nm)/AZO and Ag (14 nm)/AZO thin films shown in Fig. 2 indicates that the layer structures are not parallel to the surface of substrate. There is interdiffsusion of Ag and AZO which may be due to higher reactivity of Ag [15]. Interdiffusion of particles changes the morphology films which can affect the photo-electric properties of the films. So systematic search was carried out to determine the effect of each layer on the optical and electrical properties of the films. The most crucial factor that affects the performance of the coating is the homogeneity of the metal layer as the thickness of Ag layer is not allowed beyond a certain threshold for high transmittance in the visible region.

Fig. 3 shows the sheet resistance of Ag/AZO films with the thickness of silver layer. There is a report that silver film does not show significant conductivity until its thickness exceeds few nm, e.g. >30 nm, when it becomes continuous and its resistivity decreases with thickness [16–18]. However, this thickness of silver film depends on the deposition process and

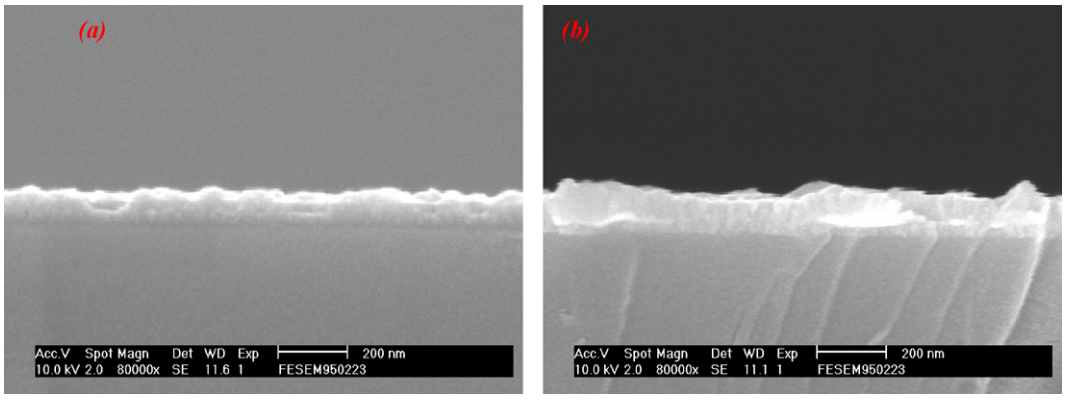

Fig. 2. SEM cross-section view of: (a) Ag (11 nm)/AZO/glass and (b) Ag (14 nm)/AZO/glass film.