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Letter

Deposition of Ag-based Al-doped ZnO multilayer coatings for the transparent conductive electrodes by electron beam evaporation

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Abstract

Silver-based Al-doped ZnO (AZO) multilayer coatings were prepared on glass by e-beam evaporation techniques. Optimization of the deposition conditions of both AZO and Ag layers were performed for better electrical and optical properties. The properties of the multilayers were affected by the deposition process of both AZO and Ag layers. The best multilayer coatings exhibit low sheet resistance of 5.34 Ω/sq and transmittance of more than 85%. The coatings have satisfactory properties of low resistance and high transmittance for application as transparent conductive electrodes.

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

1. Introduction

Al-doped ZnO thin films show low electrical resistance and high transmittance in the visible range of the spectrum. They are used as transparent conductive electrodes in optoelectronics devices including flat displays, thin films transistors and solar cells [1–3]. Different technologies such as electron beam evaporation [4], sol–gel [5], chemical spray [6], pulsed laser deposition [7], DC and RF magnetron sputtering [8–10], etc. 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 of sample (during or after deposition process). 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 (Ag) layer can be deposited on AZO.

The multilayer structure having an Ag metal layer was initially investigated for the application of low emissivity coatings. Most of the studies were concentrated on the durability and thermal stability of such films [11–13]. Only a few reports are referred to the possibility of its usage as a very low resistance electrode [12,13].

It is also well known that the optical and electrical properties of very thin metal films depend considerably on their structure [14]. To get bulk like properties, the Ag films should form a continuous structure, although they must be thin for high transmittance. In this study, we select the Ag film coated on AZO because the Ag starts with a low resistivity of $2 \times 10^{-6}\,\Omega\,\mathrm{cm}$. Ag metal films having lowest resistivity were chosen to improve the electrical property of the multilayer. Multilayer and single layer AZO and Ag films were prepared and optimized the condition of deposition process for practical use of TCO. The influence of preparation process on the properties of the film was studied.

2. Experimental

Thin films of Ag, AZO and multilayer (AZO/Ag/AZO) were deposited on glass substrates (corning eagle 2000 glass) in an e-beam evaporation system. The films were

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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 variation of substrate temperature during deposition was maintained within ± 5 °C. Substrate temperature was controlled in the range of 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 20–50 mA at different substrate temperature. Ag films were deposited at 8 kV and 20 mA at different substrate temperature.

The thickness of the film was measured using a surface profiler (Alpha-step 500, TENCOR) and on line thickness measurement system, which was further confirmed by cross-sectional SEM observation. Surface morphology was observed by field emission scanning electron microscope (FE-SEM, XL-40). Sheet resistance was measured using four-point probe method. Optical transmittance was measured in the range of 300–800 nm by UV–VIS–IR spectrophotometer (Hewlett Packard 8452A, Palo Alto, CA).

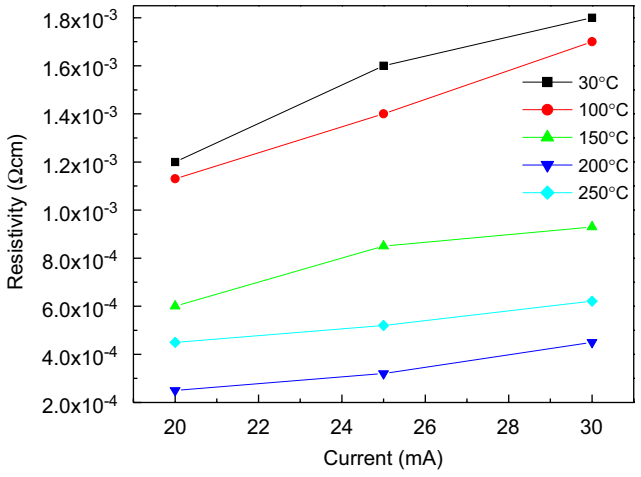

Fig. 1. Resistivity of AZO film as a function of electron beam current at a substrate temperature of (a) 30, (b) 100, (c) 150, (d) 200 and (e) 250 °C.

3. Results and discussion

Generally, AZO films deposited at high substrate temperature show improved electrical and optical properties [2,6,7]. However, for practical application, the film should be formed at low substrate temperature. Fig. 1 shows the resistivity of AZO films deposited at different substrate temperature as a function of electron beam current. There is an increase of resistivity with increase of beam current. Film deposited at a substrate temperature of 200 °C shows lowest resistivity at all beam currents. The resistivity range of AZO films deposited at 200 °C was $2.5–4.6 \times 10^{-4}\,\Omega$ cm, but the film deposited at room temperature showed higher resistivity. This was caused by a low carrier concentration due to the low activation rate of Al atoms at a low temperature. As lower layer affects the Ag film property [15], the surface morphology was observed using a FESEM. The growth of thin film

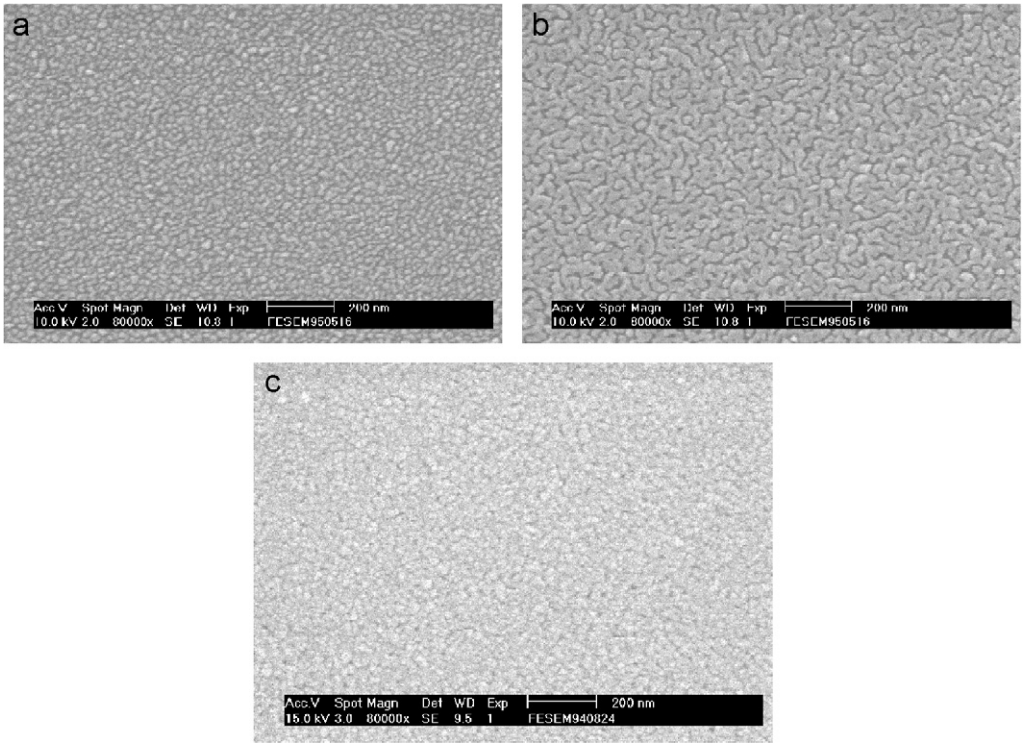

Fig. 2. SEM micrograph of Ag thin film deposited on AZO with various deposition times: (a) 10, (b) 20 and (c) 30 s.