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The influences of high energetic oxygen negative ions and active oxygen on the microstructure and electrical properties of ZnO:Al films by MF magnetron sputtering

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ABSTRACT

In this paper, ZnO:Al transparent conducting films were prepared on glass substrate by magnetron sputtering from Al doped ZnO ceramic targets. By measuring and analyzing the structure and electrical properties of films in front of targets at different target-to-substrate distance, it was concluded that the bombardment of energetic oxygen negative ions decreased with increasing target-to-substrate distance, dominating variation of resistivity and the microstructure in erosion area, while numbers of active oxygen decrease with increasing target-to-substrate distance, explaining variation of resistivity in non-erosion area. The influence of target-to-substrate distance on electrical and microstructure properties of ZnO:Al films on drum was also investigated in order to confirming our result. The result indicated that both energetic oxygen negative ions and numbers of active oxygen determined the properties of films on drum. While target-to-substrate distance is less than 95 mm, the numbers of energetic oxygen ions are the key factor and vice versa. The optimum resistivity of post-annealed films on drum was $5.1 \times 10^{-4}\ \Omega$ cm at target-to-substrate distance of 95 mm.

1. Introduction

Transparent conducting oxide (TCO) films have been extensively studied because of increasing use of optoelectronic device, like solar cell, and flat plane displays [1–3]. ZnO:Al (AZO) thin films, as alternative materials for expensive ITO (In_2O_3:Sn) thin films, have many advantages, such as cheap value, abundant resource [4], non-toxicity [5], and good stability in hydrogen plasma [6]. Magnetron sputtering is a well-established technique for preparing high-quality films and suitable for industrial production. However, the optimum resistivity of AZO films is still higher than resistivity of ITO films. There were two distinct explanations for the origin of high resistivity: the energetic oxygen negative ions [7] and the nonuniformity of active oxygen reaching the substrate [8,9]. Generally, there are three different particles from target besides Zn and Al atoms: positive ions (Ar^+) [10], neutral particles (Ar, O) [11], negative ions (O^{2-}, O^-) [12] and clusters. The positive ions are favorable for films growth by improving the mobility of particles on the surface while the negative ions with high energy cause the damage of films quality. The change of high energetic ions with different target-to-substrate distance (TSD) has been hardly reported as our knowledge.

In this paper, the spatial distribution of resistivity at different TSD was systematically studied. The influence of high energetic oxygen negative ions and numbers of active oxygen on the microstructure and electrical properties of AZO films were investigated systematically at different TSD. Moreover, the films on drum at different TSD were also deposited on glass slides, confirming our conclusion above. The resistivity of films on drum was $5.1 \times 10^{-4}\ \Omega$ cm after annealing in H_2 atmosphere.

2. Experiment

The AZO films were deposited on glass slides and K9 glasses in a dual-target mid-frequency magnetron sputtering apparatus (RAS-1100C, SHINCRON CO., LTD.), which has been described elsewhere [13]. Two rectangular AZO (2.2 wt% Al_2O_3) ceramic targets with size of 127 mm × 457 mm were placed side by side next to each other with a distance of 36 mm to form dual-target magnetron sputtering. The chamber was pumped down to lower than 2.0×10^{-4} Pa. Then the Ar was imported as working gas with working pressure about 0.3 Pa and no oxygen gas was intentionally imported. The sputtering power density was about 5.2 W/cm^2. First of all, the glass slides were placed in front of the targets to analyze the change

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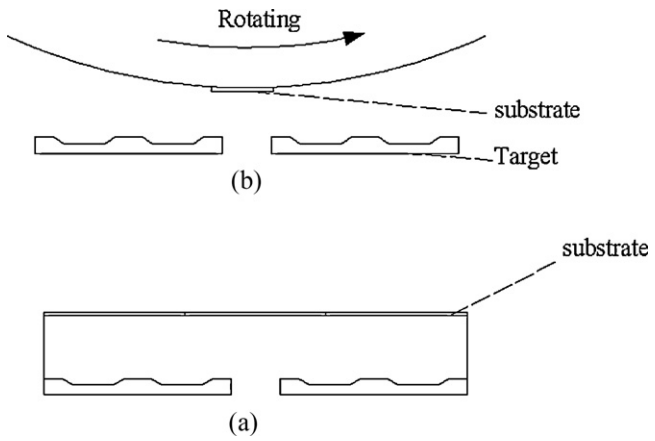

Fig. 1. The sputtering schematic diagram of different ways. (a) Sputtering films in the fixed clamp; (a) sputtering films in the rotating drum.

of energetic oxygen negative ions, numbers of active oxygen, spatial distribution of resistivity at different TSD (shown in Fig. 1a). The target-to-substrate distance varied from 62 mm to 140 mm, every glass slide was divided into small glass slides with size of 1.5 cm × 1.5 cm for measuring resistivity of films. Moreover, the glass slides were also placed on drum with high rotating rate during sputtering at different TSD (shown in Fig. 1b). The films on drum were annealed in H_2 atmosphere at 450 °C for 30 min with heating rate 5 °C/min of in order to improving the resistivity.

Resistivity was measured by four-probe method. Carrier concentration and mobility was calculated from Hall effects measured by van der Pauw method. The film thickness was calculated by Maclod method and the transmittance and reflectance spectrum measured by UV-VIS-IR4100 spectrophotometer. The film thickness is about 400–600 nm, which is nearly no effect on resistivity [14]. The structural properties of AZO films were examined by X-ray diffraction (XRD) and scanning electronic microscopy (SEM).

3. Results and discussion

3.1. The spatial distribution for films in front of target

Considering the symmetry, the spatial distribution of AZO films only at one side of dual targets is measured. O position is the center of the dual targets and D position has a distance of 7.5 mm offset O position, A and C position is the erosion area while B position is center of two erosion area, as shown in Fig. 2a.

Fig. 2a also shows the spatial distribution of resistivity of films as a function of TSD. There are two different trends for variation of resistivity at erosion area (A and C position) and non-erosion area. For example, in A position, resistivity decreases until TSD = 120 mm (C position, until TSD = 100 mm) and then slightly increases (shown in Fig. 2b). On the other hand, resistivity increases with increasing TSD at non-erosion area in D position (shown in Fig. 2c, except for A

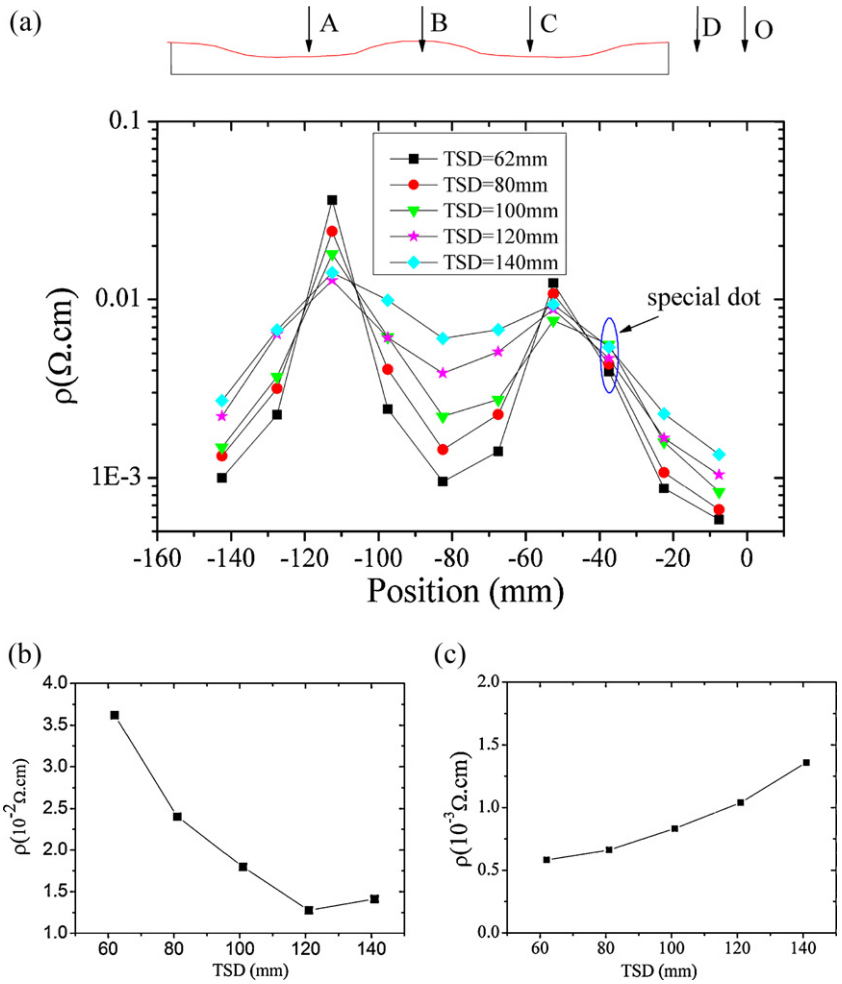

Fig. 2. The spatial distribution of resistivity as a function of different TSD and variation of film resistivity for different area: (a) the spatial distribution of resistivity; (b) A position; (c) D position.