

Growth of ZnO:Al transparent conducting layer on polymer substrate for flexible film typed dye-sensitized solar cell

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ABSTRACT

Aluminium-doped zinc oxide (ZnO:Al) conducting layer was deposited on polyethylene terephthalate (PET) substrate by r.f. magnetron sputtering to investigate the possible application of the film as transparent conducting electrode for film-typed dye-sensitized solar cells (FT-DSCs). The structural and electrical properties of ZnO:Al thin film were influenced significantly by the position of PET substrate and by also gas pressure. The minimum resistivity of $2.1 \times 10^{-3} \Omega \text{ cm}$ was obtained at gas pressure of 5 mTorr. The deposition rate of ZnO:Al film at 5 mTorr of gas pressure was 248 nm/min. The efficiency of prepared FT-DSCs sample with ZnO:Al electrode showed about 2.1% (FF: 0.54, V_{oc} : 0.73 V, J_{sc} : 5.3 mA/cm²).

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1. Introduction

Recently film-typed dye-sensitized solar cells (DSCs) attract much interests with increasing applications for a smart window, a wearable PC as a supplementary power supplier and a winter clothes due to its flexibility and transparency [1]. In the film-typed DSCs with a ZnO:Al film, which serves as transparent conducting electrode, has emerged as one of the most promising transparent conducting films since it is inexpensive, mechanically stable, and highly resistant to deoxidation [2]. In this case ZnO:Al film must be deposited on the flexible polymeric substrate, replacing for glass substrate. In fabricating the ZnO:Al film by dc or r.f. sputtering method the substrate heating is considered as one of the essential processes to obtain the high electrical and optical properties [3]. However, in the case of using polymer substrate, we cannot use the substrate heating due to the poor thermal resistance of polymer substrate. If, without substrate heating, the film shows an incomplete reaction and rough films are obtained. Moreover, the deposition rate of ZnO:Al film deposited on the polymeric substrate cannot be comparable with that deposited on glass substrate [4]. However, systematic studies on the growth of ZnO:Al transparent conducting layer on polymeric substrate cannot be performed. In this study ZnO:Al thin film was deposited on PET substrate by r.f. magnetron sputtering method. The effects of gas pressure on the morphological and electrical properties of ZnO:Al thin film were studied. Especially the position of PET substrate was studied to im-

prove the electric property of ZnO:Al film and to increase also the deposition rate. One inched FT-DSC with ZnO:Al electrode was fabricated, and possible application of ZnO:Al thin film to FT-DSCs was verified from some properties of FT-DSC such as fill-factor and photo-electric conversion efficiency.

2. Experiments

Fig. 1 shows schematic diagram of experimental setup. In order to improve the deposition rate and to protect the PET substrate from direct contact of thermal energy of highly energized particles in the plasma body the substrate was placed 6 cm apart from the target as shown in Fig. 1. ZnO:Al films were prepared on PET (30 mm × 30 mm × 0.25 mm) by r.f. magnetron sputtering. The target was a mixture of ZnO (99.99%) and Al₂O₃ (99.99%), pressed on a copper saucer with a diameter of 3 in. The PET substrate was cleaned prior to each deposition in an ultrasonic cleaner and dried by nitrogen flow. To maintain the same target condition for each deposition, the target was also sputtered in pure argon (99.999%) environment prior to each deposition with the shutter covering the substrate. Thickness and sputter yield of the ZnO:Al films were measured at a given experimental condition using a DEKTAK 300 alpha-step or direct measurements from SEM photographs. Crystalline structures of the films were characterized by a X'Pert PRO MPD (Multi-Purpose High Resolution X-ray Diffractometer). Hall mobility and carrier concentration were measured by a HEM-2000 (Hall Effect Measurement System).

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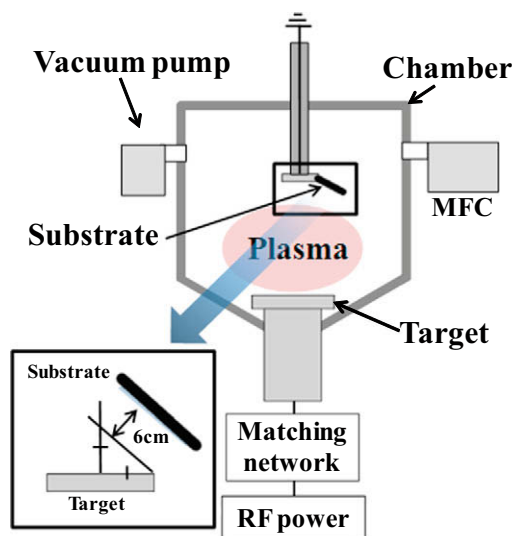


Fig. 1. Schematic diagram of experimental setup.

3. Results and discussion

From our previous research [4] an increase in sputtering power improved the electrical and morphological properties of ZnO:Al film, and the lowest electrical resistivity was obtained at around 200 W in sputtering power. Thus, in this experiment all the films were deposited at 200 W of sputtering power. In this paper, the effect of Ar gas pressure are mainly studied to improve the structural and electrical properties of ZnO:Al film deposited on the PET substrate. Fig. 2 shows the effect of gas pressure on the X-ray diffraction patterns of ZnO:Al film. As well known in Fig. 2, ZnO:Al films show the (0 0 2) preferred orientation regardless of gas pressures, and they are located at $2\theta = 34.5^\circ$, which are very close to that of the standard ZnO crystal (34.45°). The very weak (0 0 4) peaks also

appeared at near $2\theta = 72$ up to 10 mTorr. But no additional (0 0 4) peaks were observed as gas pressure increases from 15 mTorr to 25 mTorr. It is clearly seen in Fig. 2 that (0 0 2) peak intensity of the film deposited at 5 mTorr is the strongest, compared to the films deposited at other gas pressures. However, the peak intensity decreases when the film was deposited at higher gas pressure than 5 mTorr, and the peak decreases significantly from 15 mTorr. This result could be related to an increase in energy of sputtered atom arriving at the substrate because the decrease in gas pressure makes a mean free path of the sputtered atom long. That is, the sputtered atoms with energy obtained as a result of interchange with that of positive ion in the plasma can easily arrive at substrate without collision with Ar gas molecule as gas pressure decreases. It is also expected that as gas pressure decreases from 25 mTorr to 5 mTorr, the energy of electron in the plasma increases due to longer mean free path. This highly energized electron bombards the growing film on substrate, providing thermal energy for the deposited atoms. This increasing electron energy can be utilized to promote the sputtered atoms to grow in a particular order, resulting in the strong (0 0 2) preferred orientation. That is, the appropriate gas pressure induces an improvement in the crystallinity of the film. In this study the best condition of gas pressure was 5 mTorr.

Table 1 shows the full width at half maximum (FWHM) of the (0 0 2) peak and the grain size of the film as function of gas pressures. The grain sizes of the films were calculated using Scherrer's formula [5]. In the figure it is well known that the grain size at 5 mTorr at (0 0 2) preferred orientation is bigger than those at 1 mTorr and 10 mTorr. Although the grain size at 20 mTorr was the biggest, the crystalline grain grew very nonuniformly at 20 mTorr as shown in Table 1. Therefore the XRD peak cannot be strong at this gas pressure condition. The 6.1 nm of crystalline size was obtained at 5 mTorr of gas pressure. On the other hand, in general, the deposition rate of ZnO:Al film deposited on the polymeric substrate cannot be comparable with that deposited on glass substrate [6]. Therefore from our preliminary study the substrate was placed 6 cm apart from the target as shown in Fig. 1, to

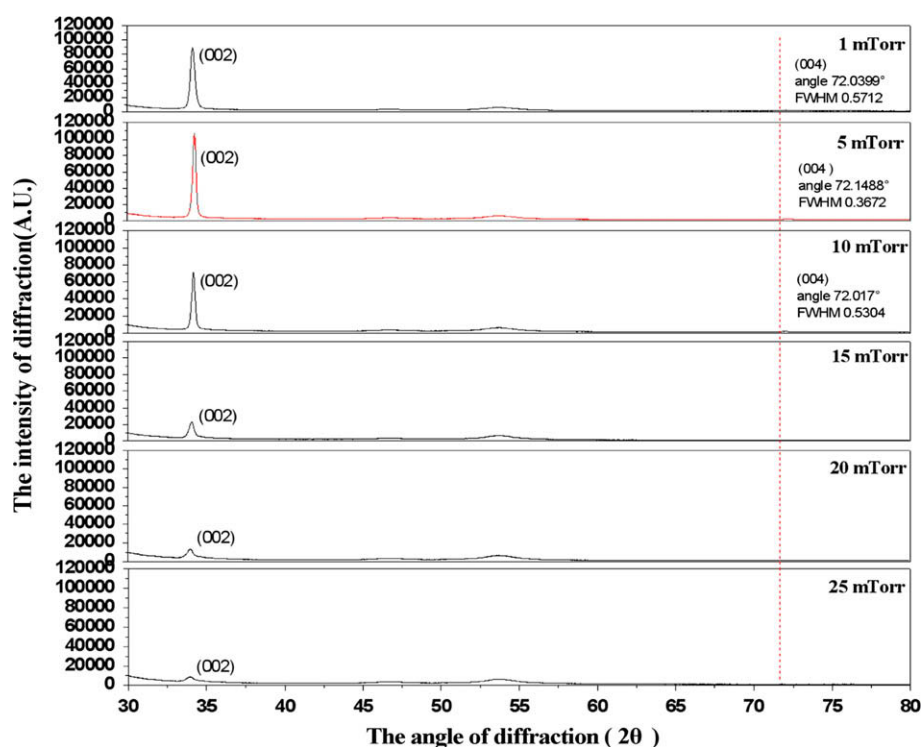


Fig. 2. XRD patterns of the ZnO:Al films deposited at conditions of different gas pressures.

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