Resistivity Measurement of ZnO:Al films for Solar Cell

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Abstract

Aluminium doped Zinc Oxide films were deposited on glass slide by RF magnetron sputtering using a ZnO target mixed with Al₂O₃. All the films were growth in room temperature without intentional heating. The resistivity of the ZnO:Al films were measured using van der Pauw method in terms of the preparation conditions such as RF power, working pressure, deposition time, O₂ content in sputtering gas and target-substrate distance. Resistivity of the deposited films shows the following behaviours: decreases with the increasing RF power and film thickness while increase with increasing target substrate distance, and O₂ content in sputtering gas. Resistivity for films prepared in different working pressure decreases with the Argon pressure but increased after the optimal pressure of 45mTorr.

Keywords: Resistivity, RF magnetron sputtering, ZnO:Al.

Introduction

Zinc oxide thin film have been investigated in recent years because of their good electrical and optical properties, abundance in nature, relatively low cost, and non-toxicity thus possessing many advantages compared to other transparent conducting oxides. ZnO is a group II-VI binary n-type semiconductor with a direct wide band gap at 3.3 eV and a hexagonal wurtzite crystal structure. It can be deposited at relatively low room temperature [1], stable in hydrogen plasma and more transparent than Indium doped Tin Oxide (ITO) or Tin Oxide (TO) in the wavelength of 400 to 800nm. Various deposition techniques such as dc / rf sputtering [2-3], metal-organic chemical vapour deposition [4], electron beam evaporation [5], spray pyrolysis [6] etc had been used to deposit ZnO films. These techniques produce high quality ZnO films that are suitable for many applications [7]. Among the deposition techniques used to prepare ZnO thin films, magnetron sputtering is considered the most suitable technique due to the inherent ease in controlling the deposition parameters [8], and the possibilities to obtain a good orientation and uniform films [1]. In this paper we report our studies on resistivity of ZnO thin films prepared by RF magnetron sputtering.

Materials and Methods

ZnO:Al films were prepared on glass substrate by RF magnetron sputtering from a 2wt% aluminium doped zinc oxide ceramic target (99.999% purity). Soda lime glass slide was use as substrate. The substrates were ultrasonically cleaned with a sequence of chromic acid, deionised water, acetone, alcohol and deionised water. It is then blown to dry using nitrogen gas. The base pressure of the chamber was ~1.5 x10⁻⁵Torr. Sputtering was carried out at a working pressure in the range from 30 mTorr to 80 mTorr with high purity argon and oxygen gas (99.999%) as the working gas. The distance between the target and substrate was 30 mm to 60 mm. Before each deposition, a 10 min pre-sputtering in argon ambient was allowed to clean the target surface and to let the plasma become stable [9]. RF power in range of 50 W to 250 W was used to deposit the ZnO:Al films. All the films are growth in room temperature without any intentional heating, however the substrate temperature
increased due to bombardment by the sputtering ion and the final substrate temperature reached 70°C with RF power 50W.

After deposition, glass substrates were cut into 12mm × 12mm square. Ohmic contact was fabricated by thermal evaporating small quarter dot of aluminium at four corners of the deposited ZnO:Al films. Dimension of the contact electrode was kept small relative to the specimen size. Resistivity of the deposited film was measured using Van der Pauw method in dark [10-12]. The sample was connected to Keithley 6512 electrometer and Keithley 220 constant current source through JFET op amp as unity gain amplifier to minimize leakage current and circuit time constants [13-14]. The circuit was shown in figure 1. Thickness of the films was measured using a Dektak® surface profiler.

Results and Discussion

Effect of RF power
Figure 2 shows the growth rates dependency of the ZnO:Al thin films on RF power. In this case, the argon gas pressure was kept constant at 40mTorr. The growth rate increases with increasing RF power. It can be explained that with higher sputtering power, it generate more argon ions in the plasma and the bombardment on the target is increased as well, the probability that sputtered ion arrive at the substrate increase, therefore the growth rate also increases. The effect of RF power on the thin film resistivity is shown in figure 3. The resistivity of the films decreases as the RF power increased. The lowest resistivity was obtained at RF power 250W. It can be reasoned that higher RF power tensed to produce film with higher crystallinity as the atoms produced have larger driving force to migrate to more suitable lattice sites and result in more perfect crystals. [15] The same effect had been observed by Song et al [16] where increasing in the RF power improves the crystallinity of the film.

![Van der Pauw measurement by using unity gain buffers and differential measurement.](image)

![Figure 2: Dependence of ZnO:Al thin film growth rate as function of RF power.](image)
Figure 3: Dependence of ZnO:Al film resistivity as a function of RF power.

Effect of Argon gas pressure
The effect of argon gas pressure on resistivity of ZnO:Al films prepared with 2wt% Al₂O₃ target at RF power 150W is shown in figure 4. The figure shows that the resistivity of sputtered film decreases with the argon pressure but increases after the optimal pressure of 45mTorr. The increase of resistivity is due to the decrease of carrier concentration and mobility in the sample prepared at higher working pressure [7]. The decrement of mobility is ascribed to the decrease in grain size when higher argon pressure is used [17]. High resistivity of sputtered films at low pressure is due to low intensity of plasma in the sputtering chamber. This fact is support by low growth rate of ZnO:Al films when low argon pressure was used.

Figure 4: Dependence of ZnO:Al films resistivity as function of argon gas pressure.

Effect of thickness / deposition time
Thickness of film increases linearly with deposition time. Figure 5 shows the thickness dependence of the films resistivity. All films were deposited at RF power 150W in pure argon ambient. With increasing film thickness, resistivity decreases with the thickness. The resistivity decreases rapidly when thickness ranges from 139.4 nm to 257.5 nm. When the thickness is more than 437.6 nm,
resistivity decreases slowly to a nearly constant value of 7.08 \times 10^{-2} \Omega \text{cm}. The reduced resistivity is ascribed to the improved crystallinity as the ZnO molecules have more time to growth and arrange. The grain size increased and reduced the grain boundary scattering. This will increases the carrier lifetime and, consequently increases the carrier mobility with increasing thickness [18-19].

![Graph](image1)

**Figure 5:** Dependence of ZnO:Al films resistivity as function of thickness.

![Graph](image2)

**Figure 6:** Dependence of ZnO:Al films resistivity as function of oxygen content in sputtering gas.

**Effect of oxygen content in sputtering gas**

Figure 6 shows the oxygen content dependence of the resistivity of films under condition RF 150W, working pressure 45mTorr. The resistivity of the film was strongly influenced by oxygen content during the film preparation. ZnO:Al film deposited using pure Argon ambient yield a lowest resistivity of 1.41 \times 10^{-1} \Omega \text{cm}. Films deposited using oxygen content more than 10% shows high resistivity. Resistivity of film is nearly constant for films prepared using oxygen content 20-50%. This can be explained by the fact that at pure argon ambient or low oxygen content, the films with large number of deficiencies are deposited which resulting in high carrier concentrations. With increasing oxygen content, the oxygen vacancies were filled, and reduced the carrier concentration [20]. Jeong et al suggested that films prepared using high oxygen content was formed at the same condition and was very close to stoichiometry. [21]. This suggest that to produce a highly conductive film from a ZnO:Al ceramic target, it it's better to sputter in pure Argon ambient [22].
Effect of target substrate distance
Figure 7 shows an overview on how the resistivity of sputtered films is influenced by target substrate distance in the sputter process. The sputtering condition is RF power 150W at 45mTorr pure argon ambient. The resistivity of films decreases with decreasing distance. This can be reasoned that the crystallinity of the resulting films is improved and the crystallite size becomes larger when the target to substrate distance is decreasing.

![Figure 7: Dependence of ZnO:Al films resistivity as Target Substrate Distance.](image)

Conclusion
Transparent conductive aluminium doped zinc oxide thin film was successfully deposited on glass substrate using RF magnetron sputtering from ceramic ZnO:Al$_2$O$_3$ target. Effect of deposition parameters such as RF sputtering power, working pressure, deposition time, target substrate distance and oxygen content in sputtering gas toward films resistivity was studied. We observed those parameter were critical to acquire the optimum electrical properties of ZnO:Al thin films. The lowest resistivity we obtained for room temperature as-deposited films in this experiment was $1.44 \times 10^{-2} \ \Omega \text{cm}$. The sample prepared in this experiment can be useful for solar cells and other applications.

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References


