Highly Oriented ZnO:Al Thin Films as an Alternative Transparent Conducting Oxide (TCO) for Windows Layer of Solar Cells

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Highly Oriented ZnO:Al Thin Films as an Alternative Transparent Conducting Oxide (TCO) for Windows Layer of Solar Cells

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Abstract. Aluminum doped zinc oxide (ZnO:Al) thin films were deposited on corning glass substrates using DC magnetron sputtering at various growth temperatures (27°C to 400°C). X-ray diffraction spectroscopy (XRD) analyses showed the crystal structure of ZnO:Al thin films was wurtzite with c-axis orientation. By increasing the growth temperature, the crystal size and the crystal stress increased, while the resistivity of films decreased. Crystal size increased from 35 nm to 52 nm, the stress increased from -7.689 GPa to -5.126 GPa, while the resistivity decreased from 6.29×10^{4} Ωcm to 4.05×10^{3} Ωcm. Generally, the quality of crystal was enhanced as the increase of growth temperature.

Introduction

Recently, studies on zinc oxide (ZnO) as an alternative transparent conducting oxide (TCO) for window layer of solar cell are more extensive than the studies on indium tin oxide (ITO) [1-3]. ZnO thin films is a promising alternative material to TCO, because it is accessible, low cost, has a high optical transmittance, high electrical conductivity, nontoxic, anisotropy in crystal structure [3-5]. Besides, ZnO film can be deposited at low temperature and has a good stability. Moreover, n-type semiconductor doped zinc oxide shows stable electrical and optical properties [6]. However, the transmittance and conductivity of ZnO based-TCO are still lower than those of ITO. The challenge is how to enhance the properties of ZnO based-TCO so that it becomes comparable to the ITO based-TCO. In order to enhance the transmittance and the conductivity of ZnO based-TCO, the ZnO films properties need to be manipulated. One way to manipulate the properties of material is by introducing a suitable doping material to the host material. One of the most optimum dopants from group IIIA (B, Al, Ga, In) in the periodic table is aluminum (Al). In this research, therefore, we grew ZnO:Al thin films and investigated the effect of growth temperature on the structure and the electrical resistivity of ZnO:Al thin films.

To date, thin films could be deposited using many different methods, such as MOCVD, sol-gel dip-coating RF magnetron sputtering and pulsed laser deposition [1-11]. Among these methods, sputtering is one of reasonable thin film deposition methods that can be considered. It is because sputtering can be used to produce thin films from materials that have high melting points [1,4]. Besides, the layer thickness can be controlled accurately and deposition process can be carried out at relatively low temperatures with high deposition rate [2]. In this research, ZnO:Al thin films were deposited at low growth temperature using a home-made DC magnetron sputtering system.

Experimental

A mixture of ZnO (99.999%) and Al_2O_3 (99.999%) was employed as pellet made-materials then used as a sputtered target. The amount of Al_2O_3 added to the target was 2 wt%. The target ZnO:Al was heated in a furnace at 750°C for 2.5 h. Corning glasses (used as substrates) were cleaned through ultrasonic bath in an acetone and methanol, respectively, for 15 minutes and then blown dried with oxygen before being admitted to the sputtering system, and were grown using the DC magnetron sputtering. The DC power was 40 W. Argon gas was used as sputter gas that was set at working pressure of 500 mTorr with a variety of growth temperatures ranging from room temperature, 200°C, 300°C, and 400°C. The crystal structure of ZnO:Al films was examined using XRD with CuKα1 radiation (λ = 1.5406 Å) at 40 kV and 20 mA. JCPDS CAS (Number 1314-13-2, CuKα1) was used as an internal X-ray standard. Two-point probe configuration of I-V measurement was used for electrical measurements of the ZnO:Al thin films.

Results and Discussion

The X-ray diffraction spectra (XRD) of ZnO:Al films deposited at different growth temperatures are shown in Fig. 1. It can be seen that the intensity of the (002) peak increases along with the increase of the substrate temperature. This result notices that enhancing the growth temperature will improve the crystallinity of films [3-6].

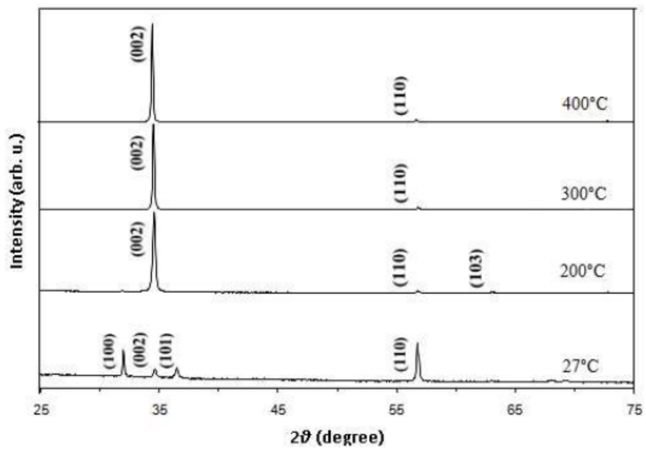

Figure 1. X-ray diffraction spectra of ZnO:Al films deposited at different growth temperatures.

The raising of the intensity is influenced by the number of reflected areas in the composition of atoms inside the films. The interference from diffracted waves on the larger reflected area would combine each other and made the intensity becomes higher. As a result, the higher the substrate temperature it leads to improving of diffracted patterns intensity. It is because the increase of substrate temperature affects the formation of atoms in the ZnO:Al film. The increase of the substrate temperature causes the increase of kinetic energy and the momentum of the atoms. Atoms with high kinetic energy and momentum will easily migrate and arrange themselves on the surface of substrate and it therefore produces large area ZnO:Al film. The XRD spectra showed that the films are polycrystalline in wurtzite structure. The lattice spacing, d of (002) peak achieved at various growth temperatures (27°C-400°C) is 2.590 Å. The c-lattice parameters were estimated using Eq. (1), while the crystal size along the (002) peak was evaluated using Eq. (2) [5].

$$\frac{1}{d^2}=\frac{4}{3}\left(\frac{h^2+hk+k^2}{a^2}\right)+\frac{l^2}{c^2} \quad (1)$$

where hkl is the Miller index, a and c are the lattice parameters.

$$D=\frac{0.9\lambda}{\beta\cos\theta} \quad (2)$$

where D is the crystal size, λ is the wavelength of X-ray, β is the FWHM, θ is the diffraction angle.

Based on the XRD results, we can see that the crystal size of the films increases from 35 nm to 52 nm along with increase of the substrate temperature from 27°C to 400°C. The larger crystal size showed the higher opportunity of sputtered atoms to occupy the correct site in the crystal lattice, so the crystal quality became higher [6]. The alteration of crystal size indicates the changing of atoms arrangement with similar orientation. Therefore, the transformation of the crystal size indicated the occurrence of lattice strain in the films. Lattice strain, ε, of the film was determined using Eq. (3) while the stress, σ, was determined using Eq. (4) [5,7] and the results are presented in Table 1.

$$\varepsilon = \frac{\beta}{4\tan\theta} \tag{3}$$

$$\sigma = -233\varepsilon \tag{4}$$

Table 1. Summary of analized X-ray ZnO:Al films deposited by various growth temperature

Growth temperature (°C)	Pos 2θ (°)	d- Spacing (Å)	c-Lattice (Å)	FWHM (°)	Crystal size (nm)	Lattice strain	Stress (GPa)
27	34.5918	2.590	5.18	0.4145	35	0.033	-7.689
200	34.5918	2.590	5.18	0.3869	37	0.031	-7.223
300	34.5918	2.590	5.18	0.3316	43	0.026	-6.058
400	34.5918	2.590	5.18	0.2764	52	0.022	-5.126

Lattice strains of the films indicated the occurrence of stresses in the films. The stresses of the films, which were -7.689GPa, -7.223GPa, -6.058GPa, and -5.126GPa, respectively, decreased by increasing the growth temperature. Stress of crystal influenced the quality of the crystal. The crystal quality of the films is improved when the stress is increased. The stress magnitude is proportional to the force magnitude between the bindings of the atoms. Therefore, film with higher stress produces stronger bounding forces between the atoms. The magnitude of binding force between the atoms are proportional to the binding energy of the crystal i.e. energy is required to separate the atoms in the crystal. The contribution of the vibration in the formation of the crystals showed the regularity of the atoms arrangement in the crystal. The atoms in this structure are not able to move freely because of those binding forces. The atoms that constructed the crystal always vibrate in their equilibrium position, so that higher stress level produces more regular arrangement of atoms configuration in the films. Hence, the crystals quality of the films is improved when the stress of crystal increases. The summary of X-ray analyses of ZnO:Al films deposited by various substrate temperatures is shown in Table 1.

Table 2. Electrical resistivities of Al-doped ZnO films deposited at different substrate temperatures.

Sample	Substrates temperature (°C)	Electrical resistivity (Ω.cm)	Electrical conductivity ((Ω.cm)$^{-1}$)
ZnO	27	2.49 x 10^6	4,02 x 10^{-7}
ZnO:Al	27	6.29 x 10^4	1.59 x 10^{-5}
	200	6.29 x 10^4	1.59 x 10^{-5}
	300	1.07 x 10^4	9.35 x 10^{-5}
	400	4.05 x 10^3	2.47 x 10^{-4}

The electrical resistivities of ZnO:Al films are listed in Table 2. We can see that the electrical resistivity decreases due to the increase of substrate temperature. The effect of the growth temperature on the electrical resistivity could be connected to the transformation of the microstructure of the film. Chaabouni, et al. explained that the electrical properties depend on the elementary properties of the grain quality. The substrate temperature was not proportional with scattering process on the grain boundaries [11]. Increasing substrate temperature leads to reducing scattering process on the grain boundaries. Yang et al. reported that when the substrate temperature increases, the grain size of crystal will increase to cause less scattering on the grain boundaries [8]. The less scattering on the grain boundaries leads to the enhancement of mobility and decrease of resistivity [7-8]. The conductivity of ZnO film is conversely proportional to the film resistivity and directly related to the number of electrons. These electrons are generated by the ionization of the interstitial zinc atom and the oxygen vacancies affect the electrical conductivity of ZnO crystals [7-9]. The ZnO:Al films deposited under higher temperatures may have much carrier concentration in the conduction band so that the electrical conductivities of the ZnO:Al thin films become higher.

Summary

We have investigated the effect of growth temperature on structures and resistivity of ZnO:Al thin films deposited using DC magnetron sputtering. X-ray diffraction spectroscopy (XRD) analysis showed that the crystallinities of films are enhanced with increasing growth temperature from 27°C to 400°C. The crystal structures of ZnO:Al thin film were wurtzite with c-axis orientation. By increasing the growth temperature, the crystal size of films enhanced from 35 nm to 52 nm, while the resistivities of films decrease from 6.29×10^4 Ω.cm to 4.05×10^3 Ω.cm.

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