Photoelectric properties of ITO thin films deposited by DC magnetron sputtering*

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Abstract: As anti-reflecting thin films and transparent electrodes of solar cells, indium tin oxide (ITO) thin films were prepared on glass substrates by DC magnetron sputtering process. The main sputtering conditions were sputtering power, substrate temperature and work pressure. The influence of the above sputtering conditions on the transmittance and conductivity of the deposited ITO films was investigated. The experimental results show that, the transmittance and the resistivity decrease as the sputtering power increases from 30 to 90 W. When the substrate temperature increases from 25 to 150 ℃, the transmittance increases slightly whereas the resistivity decreases. As the work pressure increases from 0.4 to 2.0 Pa, the transmittance decreases and the resistivity increases. When the sputtering power, substrate temperature and work pressure are 30 W, 150 ℃, 0.4 Pa respectively, the ITO thin films exhibit good electrical and optical properties, with resistivity below 10^{-4} Ω·cm and the transmittance in the visible wave band beyond 80%. Therefore, the ITO thin films are suitable as transparent electrodes of solar cells.

Key words: ITO thin films; DC magnetron sputtering; optical properties; electrical properties

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1. Introduction

Nowadays, one of the most important issues for the thin films solar cells such as a-Si : H and CIGS is the enhancement of efficient light trapping[1, 2]. Indium tin oxide (ITO) is an n-type semiconductor with good electrical conductivity and high transparency in the visible spectra[3, 4], so it is used not only as anti-reflecting thin films but also as electrodes of solar cells. As anti-reflecting thin films and electrodes of solar cells, the ITO films with high quality should be deposited with lower power at lower substrate temperature[5]. Because high temperature and high sputtering power will degrade the interface of the hetero-junction and the underlying device structure, due to the inter-diffusion between the adjacent layers. However, in the most reports, ITO thin films are sputtered at relatively higher temperatures and sputtering powers or they are deposited at room temperature and then annealed at higher temperatures[6–9], these will damage inter-diffusion across hetero-junction interface of thin films solar cells or the underlying electronic device structure. Therefore, in this paper ITO films were deposited by DC sputtering at lower temperatures and lower sputtering power, and the influence of the deposition parameters on the characteristics of ITO thin films were investigated in detail.

2. Experiment

The glass substrates were cleaned by deionized water, acetone, ethanol, deionized water in turn, and then dried by ovens. ITO films were prepared on the glass substrates by DC magnetron sputtering system (WYCD-600). The target is indium-tin oxides (In_2O_3 : SnO_2 = 90 : 10 wt. %) with a purity of 99.99%. The samples were mounted on a holder without circumgyrating, and the distance of the target-substrate is 8 cm. The base pressure was about 5×10^{-5} Pa, and the flow rate of Ar (99.99%) was kept at a constant value of 10 sccm controlled by a mass flow controller. The substrate temperature (T_s) was varied in the range 25 ℃ $\leqslant T_s \leqslant$ 150 ℃ using IR heater (quartz halogen lamp) and measured using a chromel-alumel thermocouple. Before sputtering ITO films on the substrates, the target was presputtered for about 5 min with a shutter covering the target in order to remove the surface oxide layer.

Twelve ITO thin films samples with thickness of 150 nm were deposited on glass substrates by changing the sputtering conditions. Table 1 lists the sample names and their deposition conditions.

Optical measurements of the ITO thin films were performed in the wavelength range from 300 to 800 nm at room

Table 1. Sample names of ITO thin films with different deposition conditions.

Sample	DC power (W)	Substrate temperature (℃)	Sputtering work pressure (Pa)
ITO-1	30	25	1.6
ITO-2	50	25	1.6
ITO-3	70	25	1.6
ITO-4	90	25	1.6
ITO-5	30	60	1.6
ITO-6	30	90	1.6
ITO-7	30	120	1.6
ITO-8	30	150	1.6
ITO-9	30	150	2.0
ITO-10	30	150	1.2
ITO-11	30	150	0.8
ITO-12	30	150	0.4

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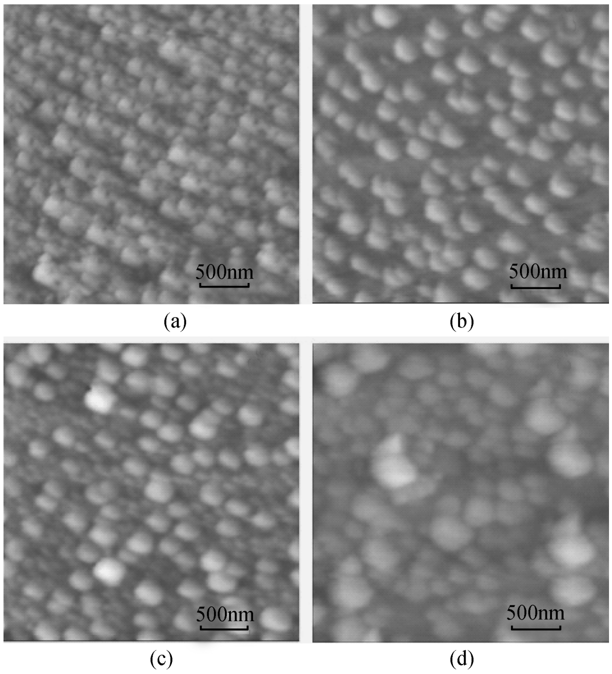

Fig. 1. AFM images of ITO thin films deposited under different sputtering powers. (a) 30 W. (b) 50 W. (c) 70 W. (d) 90 W.

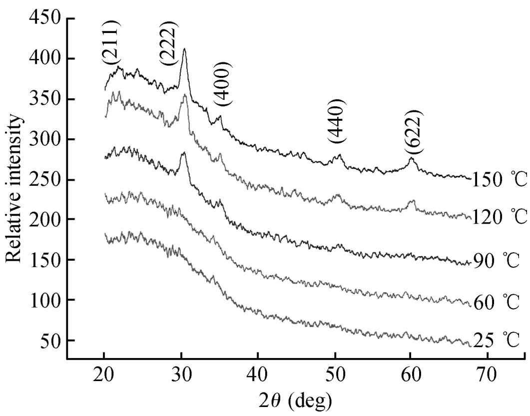

Fig. 2. XRD patterns of ITO films sputtered at various substrate temperatures.

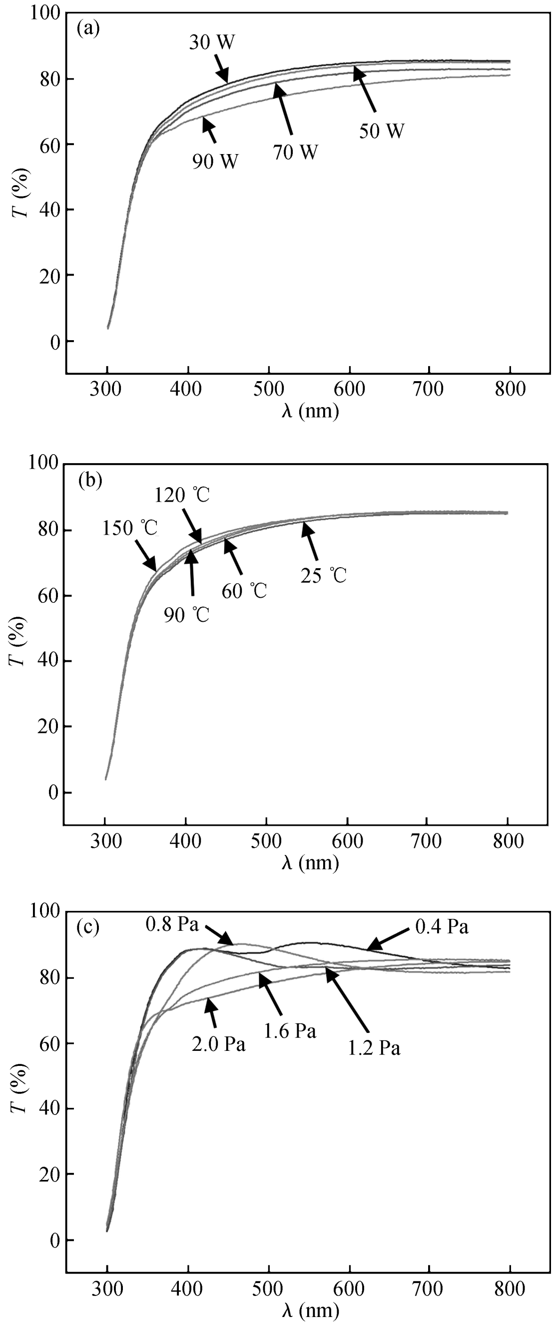

Fig. 3. Transmittance spectra of the ITO films deposited at different sputtering conditions. (a) At different sputtering powers. (b) At different substrate temperatures. (c) At different work pressures.

temperature. UV–VIS spectra were obtained from PerkinElmer Lambda 900 UV/VIS/NIR spectrometer. The electrical measurements were made using an HMS-3000 Hall measurement system. Film thickness was measured with a stylus surface profiler. X-ray diffraction (XRD) patterns were recorded using a diffractometer (PANalytical, X'Pert, CuKα, λ = 1.5406 Å). The surface morphological examinations were carried out by a CSPM5000s atomic force microscope system (AFM) in the contact mode.

3. Results and discussion

3.1. Structure and morphology

Figure 1 shows AFM images of ITO thin films deposited at different powers (samples ITO-1 to ITO-4). The surfaces of these samples seem to be formed by small nanosized grains, and the surface morphology of the films varies significantly with the deposition power. The roughness increases with the increasing of deposition power, seen in Table 2. Therefore, it is obvious that the sputtering power has some influences on the morphology of the films.

Figure 2 shows the XRD patterns of ITO films (samples ITO-1, ITO-5 to ITO-8) deposited at different substrate temperatures. Sharp and intense peaks are observed in XRD patterns of ITO films with a relatively high deposition temperature. The films deposited at temperatures below 60 ℃ are amorphous. When the deposited temperature exceeds 90 ℃, the films exhibit several obvious XRD peaks corresponding to the planes (211), (222), (400), (440) and (622) of ITO with the structure of JCPDS-ICDD No. 050848 (cubic bixbyite structure).

Table 2. Physical properties of the ITO films deposited at different sputtering powers.

Sample	DC power (W)	RMS roughness (nm)	Resistivity (Ω·cm)	Mobility ($cm^2/(V·s)$)	Carrier concentration (cm^{-3})
ITO-1	30	8.9	7.77	0.9	-8.926×10^{17}
ITO-2	50	10.8	1.1×10^{-2}	14.31	-3.966×10^{19}
ITO-3	70	13.6	2.231×10^{-3}	18.01	-1.554×10^{20}
ITO-4	90	23.1	1.096×10^{-3}	16.70	-3.411×10^{20}

Table 3. Semiconducting properties of the ITO films deposited at different substrate temperatures.

Sample	Substrate temperature (℃)	Resistivity (Ω·cm)	Mobility ($cm^2/(V·s)$)	Carrier concentration (cm^{-3})
ITO-1	25	7.77	0.9	-8.926×10^{17}
ITO-5	60	0.5449	2.228	-5.141×10^{18}
ITO-6	90	7.651×10^{-2}	4.162	-1.960×10^{19}
ITO-7	120	1.520×10^{-2}	8.54	-4.808×10^{19}
ITO-8	150	3.346×10^{-3}	14.45	-1.291×10^{20}

Table 4. Semiconducting properties of the ITO films deposited at different work pressures.

Sample	Gas pressure (Pa)	Resistivity (10^{-4} Ω· cm)	Mobility ($cm^2/(V·s)$)	Carrier concentration (cm^{-3})
ITO-12	0.4	3.574	24.04	-7.264×10^{20}
ITO-11	0.8	6.007	17.63	-5.892×10^{20}
ITO-10	1.2	12.2	15.42	-3.32×10^{20}
ITO-8	1.6	33.46	14.45	-1.291×10^{20}
ITO-9	2.0	76.23	11.58	-7.07×10^{19}

With increasing the substrate temperature, the intensity and the full width at half maximum of diffraction peak (222) become strong, indicating that the ITO films grow with a strongly preferred (222) orientation with better crystallinity.

3.2. Optical characterization

The optical transmittance of the films was recorded as a function of wavelength in the wavelength range 300–800 nm. Figures 3(a), 3(b) and 3(c) show transmittance spectra of the ITO films deposited at different sputtering conditions. In Fig. 3(a), the transmittance of the films is decreased in the visible wave band with increasing the sputtering power. Because the high-energy plasma bombards thin films during sputtering, some ITO may be decomposed into metal, thus the transmittance of the films decreases. As anti-reflecting thin films and electrodes of solar cells, ITO thin films should have high transmittance, therefore the sputtering power of 30 W is appropriate. In Fig. 3(b), the transmittance increases slightly with increasing the substrate temperature from 25 to 150 ℃, but the variation of the transmittance is not obvious.

Figure 3(c) shows the transmission spectra of the ITO films deposited at different work pressures. The result shows that an optimum optical transmittance is obtained from the ITO film deposited at 0.4 Pa pressure. At lower pressure, the ion energy distribution is sharper and the average energy is higher, therefore the influence of ion bombardment is more significant. At the same time, low pressure leads to a long mean free path of Ar^+ with high kinetic energy, and the atoms sputtered from the target have higher kinetic energies, thereby the crystallization of the thin films is better, and the transmission is higher.

3.3. Electrical properties

Semiconducting properties of the films were measured by the Hall measurement system at room temperature. Table 2 exhibits the electrical properties of the ITO thin films deposited at different sputtering powers. The negative symbol of the measured carrier concentration indicates that the films are of n-type conductivity. The resistivity decreases as the sputtering power increases from 30 to 90 W, and this might be due to the better crystallinity of the films caused by the bombardments of high-energy particles (argon positive ions (Ar^+)) at the surface of the target. As the bombarded atoms are accelerated from the cathode sheath to the substrate, the films with better crystallinity could be deposited at the larger sputtering power. The better crystallinity of the films results in lower resistivity.

Table 3 gives the electrical properties of ITO thin films deposited at different substrate temperatures. With the increasing of the substrate temperature, the resistivity decreases from 7.77 to 3.346×10^{-3} Ω·cm, the electron mobility increases from 0.9 to 14.45 $cm^2/(V·s)$ and the carrier concentration increases from 8.926×10^{17} to 1.291×10^{20} cm^{-3}. The improvement of the electrical properties of the films with increasing the substrate temperature is mainly due to enhanced crystallization of the films. Combined with Fig. 2, there is a correlation between the conductivity and the crystallinity. Hence increasing the substrate temperature is considered to be a pathway for high-quality ITO films.

Table 4 shows the semiconducting properties of the ITO films deposited at different work pressures. It is found that the resistivity of the ITO thin films increases with increasing the work pressure from 0.4 to 2 Pa. When the work pressure is increased, neutral gas and ionized gas ions will frequently hit each other, so kinetic energy of Ar+ bombarding the target diminishes owing to scattering process. The atoms with small energies deposited at the substrates reduce the active reaction of ITO, which will produce a number of indium tin subcompounds, and affect the crystalline properties of the films. Therefore the resistivity of the ITO thin films will increase with increasing the work pressure.

4. Conclusion

ITO thin films were deposited on glass substrates by DC magnetron sputtering under various conditions. It was confirmed that the electrical and optical properties of ITO thin films strongly depended on the sputtering power, substrate temperature and work pressure. Based on the required resistivity and transmission properties of anti-reflecting thin films and electrodes of solar cells, an optimum sputtering power for depositing ITO films is 30 W. For improvements in conductive and transparent properties, the substrate temperature should be increased to 150 ℃ and the work pressure should be decreased to 0.4 Pa, this being supported by the structural, optical and electrical evidence obtained from X-ray diffraction experiments，UV–VIS spectrometer, and Hall measurement system. These films exhibit low resistivity ($3.574 \times 10^{-4} \Omega \cdot cm$) and approximately 86% average transmittance. They are good for anti-reflecting thin films and electrodes of solar cells.

References

[1] Chopra K L, Major S, Pandya D K. Transparent conductors—a status review. Thin Solid Films, 1983, 102: 1

[2] Miro Z. Thin film solar cells: fabrication, characterization and applications. John Wiley & Sons, Ltd, 2006

[3] Salehi A. The effects of deposition rate and substrate temperature of ITO thin films on electrical and optical properties. Thin Solid Films, 1998, 328: 72

[4] Kim H, Gilmore C M, Pique A, et al. Electrical, optical, and structural properties of indium-tin-oxide thin films for organic light-emitting devices. J Appl Phys, 1999, 86: 6451

[5] Romeo A, Terheggen M, D Abou-Ras. Development of thin-film Cu(In,Ga)Se_2 and CdTe solar cells. Progress in Photovoltaics: Research and Applications, 2004, 12: 93

[6] Bender M, Seelig W, Daube C, et al. Dependence of oxygen flow on optical and electrical properties of DC-magnetron sputtered ITO films. Thin Solid Films, 1998, 3326: 72

[7] Hu Y L, Diao X G, Wang C, et al. Effects of heat treatment on properties of ITO films prepared by RF magnetron sputtering. Vacuum, 2004, 75: 183

[8] Teixeira V, Cui H N, Meng L J, et al. Amorphous ITO thin films prepared by DC sputtering for electro chromic applications. Thin Solid Films, 2002, 420: 70

[9] Kim S S, Choi S Y, Park C G, et al. Transparent conductive ITO thin films through the sol–gel process using metal salts. Thin Solid Films, 1999, 347: 155