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Indium tin oxide (ITO) films have been deposited onto glass substrates at room temperature by ion beam assisted deposition technique (IBAD) at different ion beam currents (80–120 mA). The effect of the ion beam current on films properties has been studied. The films prepared at low ion beam current (80 mA) show an almost amorphous structure. As the ion beam current is increased, films show a preferred orientation along the (222) direction. AFM measurements show that the surface RMS roughness of the film increases as the current goes up. The optical properties have been studied by measuring the transmittance. It has been found that the film prepared at 100 mA current has the highest transmittance. The optical constants of the films have been calculated by fitting the transmittance. The Hall measurements also showed that the film prepared at 100 mA current has one of the lower electrical resistivities. FTIR measurements show that the film prepared at 100 mA current has the highest infra-red reflectance. All the measurements show the 100 mA ion beam current can produce good quality ITO films.

1 Introduction Indium tin oxide (ITO) film, as a transparent conductive oxide material, is known for its excellent transmittance in the visible region and high electrical conductivity. It has been widely used as a transparent electrode in the areas of organic light emitting devices [1], flat panel display devices [2], electrochromic devices [3], electromagnetic shielding [4] and solar cells [5]. Many deposition techniques have been successfully used to produce ITO films, such as sputtering [2, 4], electron beam evaporation [6], sol–gel [7], laser ablation [8], spray pyrolysis [9], chemical vapour deposition [10], and reactive thermal deposition [11]. Generally, to obtain ITO films with low resistivity and high transmittance in the visible region, most of the conventional techniques mentioned above require a high substrate temperature or a post-annealing process. For producing organic luminescent devices, ITO films need to be deposited at room temperature because of the low thermal stability of the organic substrates. However, the ITO films deposited at room temperature using conventional techniques usually show a high electrical resistivity and are not suitable for the applications. Therefore, one of the important technical problems in the manufacturing processes of optoelectronic devices is the development of a low temperature deposition technique. It has been found that the ion beam assisted deposition (IBAD) technique can produce ITO films with low electrical resistivity without extra-heating the substrate [1, 12–16]. Although the bombardment of the ion beam on the substrate will raise the substrate temperature, the temperature will not exceed 100 °C, according to our experiences. By this way one can produce high quality ITO films on polymer substrates. Many parameters can have influences on the properties of the ITO films prepared by IBAD technique, such as ion beam energy, ion beam current, ion beam flow and so on. In order to produce high quality ITO films, these parameters must be optimized. In this work, we report the influence of the ion beam current on the properties of ITO films prepared by IBAD technique.

2 Experimental procedures ITO films were deposited onto the glass substrates at room temperature by ion beam assisted deposition technique using a vacuum coater equipped with an electron beam gun and a Kaufman ion source. ITO powder pellet with a composition of 90 wt% In_2O_3 and 10 wt% SnO_2 was used as the evaporation source material. A 120 mm diameters Kaufman ion source was used to generate oxygen ion beam. The oxygen gas flow was controlled by a mass flow controller and was kept at 40 sccm for all depositions. The deposition rate and the film thickness were monitored and controlled by a quartz crystal sensor which has been linked to an e-beam power supply for automatic controlling. The nominal deposition rate and the thickness were preset at 0.2 nm/s and 200 nm, respectively. The substrate holder was rotated at a speed of 0.3 rounds/s. The angle between the incident oxygen ion beam and the normal of the substrate holder was fixed at 45°. Before the deposition, the chamber was evacuated until a pressure of 1×10^{-3} Pa. After that, the oxygen gas was introduced into the chamber. During the deposition, the dynamic pressure in the chamber was about 2.3×10^{-2} Pa. The film thickness indicated in Table 1 was obtained by fitting the transmittance spectra with semi-quantum model [21, 22]. During all the depositions, the screen voltage and the accelerating voltage were kept constant at 500 V and 250 V respectively. The ion beam current was varied from 80 mA to 120 mA.

The optical transmittance spectra of the films were recorded by a Perklin–Elmer Lambda 900 UV/VIS/NIR spectrometer and the infrared reflectance was tested by a Perkin-Elmer Spectrum GX. Atomic force microscopy (AFM) measurements were made using equipment from Digital Instruments Veeco Metrology Group. The X-ray diffraction was performed in a SHIMADZU XRD-6000 ranging the 2θ values between 20°~60° with a step of 0.05°. The Hall effect was measured using a Lake Shore 665 with a 5 kG magnetic field intensity at room temperature.

3 Results and discussion With the experimental conditions we mentioned before, it can be seen from Table 1 that all the deposited films have a similar thickness around 220 nm.

The X-ray diffraction patterns of the ITO films prepared at different oxygen ion beam currents are shown in Fig. 1. The assigned peaks are related to the cubic In_2O_3 structure [17]. We could not find any tin oxide phases in our samples. The films prepared at different ion beam currents show a preferred orientation of the crystallites towards the (222) crystallographic plane. However, the film prepared at 80 mA ion beam current shows very weak dif-

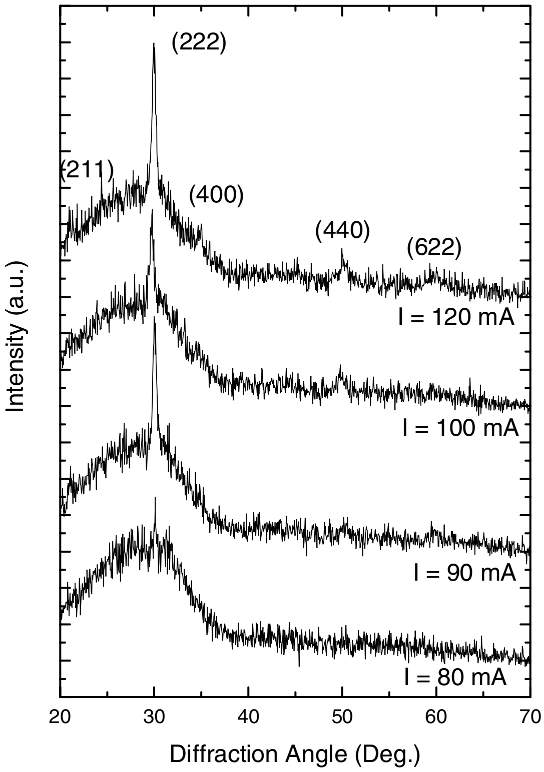

Figure 1 XRD spectra of the ITO films deposited at different ion beam currents.

fraction peak intensity. It means that this ion beam current is not enough to form the ITO films with the good polycrystalline structure. Usually, the ITO films deposited by sputtering show a (400) preferred orientation [2, 18] and the ITO films deposited by evaporation show a (222) preferred orientation [6, 11, 14, 16]. Our results agree with these previous works. However, it has been found that the crystal orientation of the films not only depends on the deposition technique, but also on the deposition parameters. For example, even for sputtered ITO films, in some deposition conditions, a preferred orientation along (222) or (440) directions can be obtained although most of sputtered ITO films give a preferred orientation along (400) direction [19, 20]. So far, there is no definite explanation to these phenomena. One of the possible explanations is the rela-

Table 1 Properties of ITO films prepared at different ion beam currents.

sample	I1	I2	D2	A
ion beam current (mA)	80	90	100	120
thickness (nm)	236	223	215	228
sheet resistance (Ω/square)	117	56	51	101
resistivity ($\times 10^{-3}$ Ω cm)	13	6	5	10
carrier concentration ($\times 10^{20}$ cm^{-3})	3.6	3.1	2.7	0.8
Hall mobility (cm^2/V S)	6	17	21	35
surface roughness by AFM (nm)	1.1	1.3	1.4	3.2

tion between the adatoms and clusters energies on the substrate with the crystal orientation. High energy adatom and cluster are favourable for the formation of the (400) orientated films and low energy adatom and cluster are favourable for the formation of (222) orientated films. It can be suggested that low adatom and cluster energy form an amorphous structure film, as the energy is increased, the (222) orientated film will be formed and the (400) orientated film will be formed with the further increase of the energy. From Fig. 1 it can be seen that the ITO film prepared at 80 mA ion beam current has an amorphous structure. The polycrystalline structure towards the (222) direction only can be formed at ion beam current higher than 90 mA. Low ion beam current (80 mA) means that a smaller number of ions can reach the substrate, resulting in a low adatom and cluster energy on the substrate and then an amorphous structure. As the ion beam current is increased, more ions can reach the substrate and the adatom and cluster energy increase, forming a polycrystalline structure.

The (222) diffraction peak for the ITO films prepared at different ion beam currents has been fitted. The distance between (222) crystal plane has been calculated and compared with standard value ($d_0 = 0.2921$ nm) [17]. It is obvious that comparing the measured (222) peak position with the standard (222) peak position to get the information on stress in the ITO films is not an ideal way for the following reasons: (1) The ITO lattice parameter depends on the concentration of Sn that is substitutional on In sites and (2) although textured, the film is polycrystalline and biaxial stress will not uniformly affect the 222 interplanar spacing. As we use the same ITO powder pellet as evaporation source for different ion beam currents, so the Sn concentration in ITO films could be same for all the samples, so the effect of the first reason could be same for all the samples. Although the biaxial stress will not uniformly affect the 222 interplanar spacing, as we only discuss qualitatively the stress in ITO films, it will not affect the conclusion. Based on these arguments, we still use this method to estimate the stress type in ITO films. It has been found that all the samples have values bigger than standard value which indicates that all the films are subjected to the compressive stresses. The crystalline dimension along the (222) direction has also been estimated by fitting the XRD spectra. The results show the crystalline dimension along the (222) direction is about 20 nm for all the samples, and does not change with different ion beam currents.

The surface morphologies of the ITO films have been studied by AFM as shown in Fig. 2. The surface room-mean-square (RMS) roughness has been calculated for all the samples. The results are given in Table 1 and shown in Fig. 3. It can be seen very clearly that the surface roughness has a sharp increase when the ion beam current is increased from 100 mA to 120 mA. Therefore, in order to obtain films with a relative smooth surface, the ion beam current should not be higher than 100 mA.

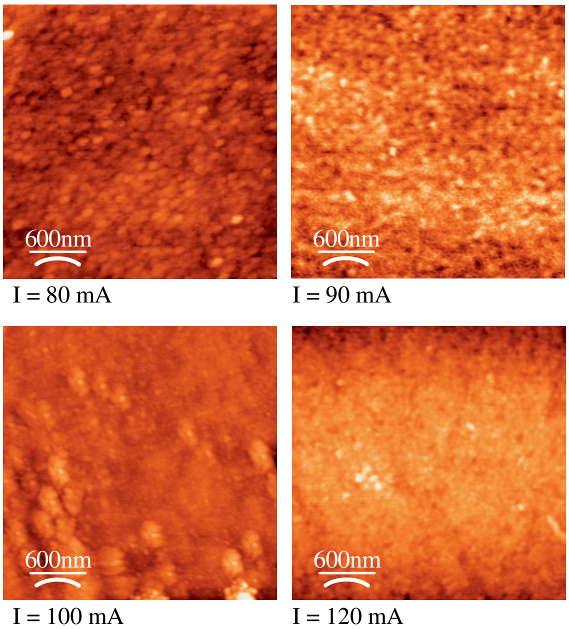

Figure 2 (online colour at: www.pss-a.com) AFM images (3 μm × 3 μm) of the surface of the ITO films deposited at different ion beam currents.

The transmittance of the films has been given in Fig. 4. It shows that the transmittance of the films, both in the visible and near IR regions, increases as the ion beam current is increased. It is well known that when the ITO pow-

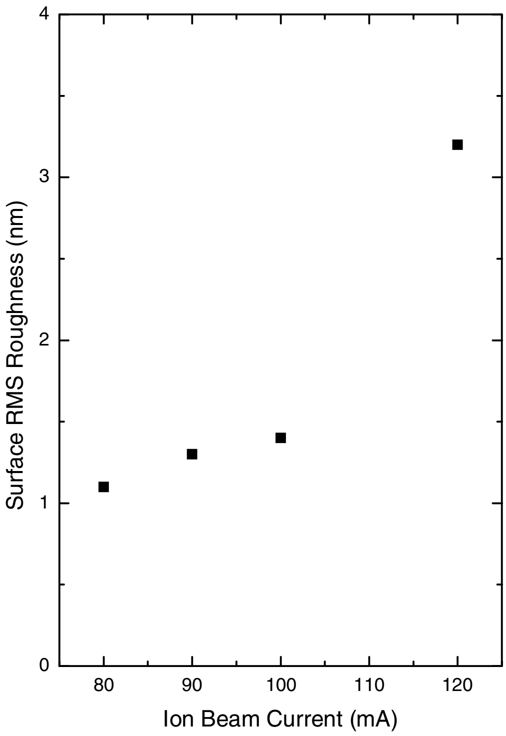

Figure 3 Surface root mean square (RMS) roughness of ITO films deposited at different ion beam currents.

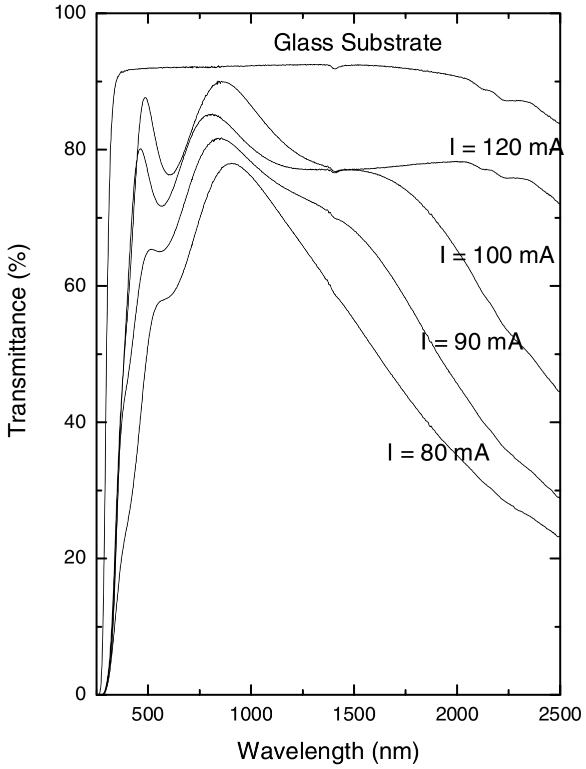

Figure 4 Specular transmittance as a function of the wavelength for the ITO films deposited at different ion beam currents.

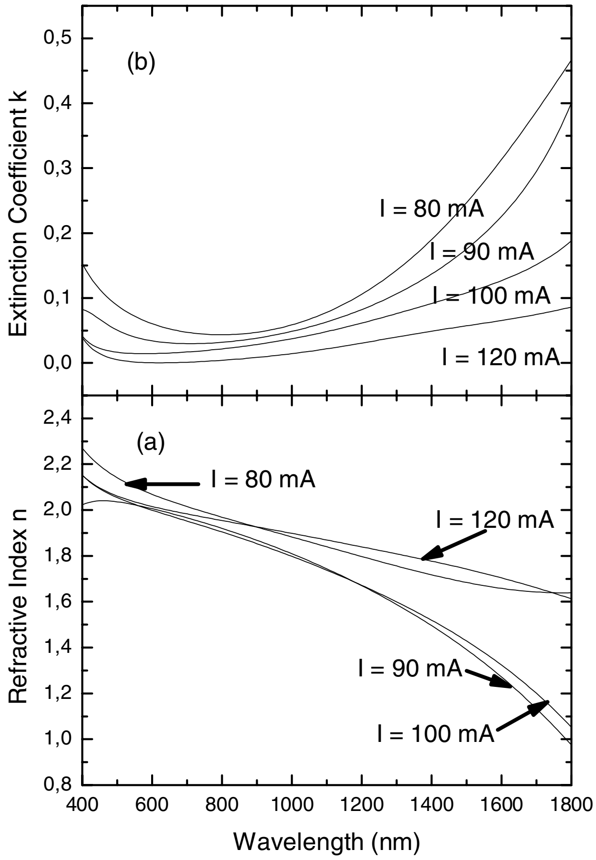

Figure 5 Refractive index (a) and extinction coefficient (b) as a function of the wavelength for the ITO films deposited at different ion beam currents.

der pellet is evaporated by e-beam, the oxygen will be lost and an incomplete oxidized ITO films will be formed on the substrate. These incomplete oxidized ITO films usually consist of many oxygen vacancies resulting in a low transmittance and high electron concentration as the oxygen vacancy is the donor of the electrons. The introduction of the oxygen ion beam will compensate the loss of the oxygen during the evaporation process and improve the transmittance of the deposited ITO films. When the ion beam current is low, the amount of the oxygen ions reaching the substrate is not enough to compensate the loss of the oxygen during the evaporation process and will result in a low transmittance. As the ion beam current is increased, the amount of the oxygen ions reaching the substrate is also increased, then reducing the oxygen vacancies and improving the transmittance, as shown in Fig. 4. As the proportion of oxygen vacancies is reduced, the electron concentration will also be reduced. The increase of the transmittance in the near IR region with oxygen ion beam current confirms this fact.

To obtain the optical constants and the thickness of the ITO films prepared at different oxygen ion beam currents, the transmittance of these films has been fitted using semi-quantum model [21, 22] combined with Drude model. The dielectric function can be described as follows:

$$\varepsilon(\omega) = \varepsilon_\infty \prod_j \frac{\omega_{j\mathrm{LO}}^2 - \omega^2 - i\gamma_{j\mathrm{LO}}\omega}{\omega_{j\mathrm{TO}}^2 - \omega^2 - i\gamma_{j\mathrm{TO}}\omega} + \frac{\omega_p^2}{-\omega^2 + i\gamma_d\omega}\,. \qquad (1)$$

The first term is the semi-quantum model and it represents the dielectric function as a product of individual oscillator terms. For each term there are four parameters, where $\omega_{j\mathrm{TO}}$, $\gamma_{j\mathrm{TO}}$, $\omega_{j\mathrm{LO}}$, $\gamma_{j\mathrm{LO}}$ are the resonance frequencies and damping constants of the transverse and longitudinal optic modes, respectively. ε_∞ is the “high-frequency” contribution to the dielectric function. The second term is the Drude model which is used to modify the free electron properties. The refractive index and extinction coefficient obtained by this fitting are shown in Fig. 5. The extinction coefficient of the ITO films decreases gradually as the ion beam current is increased from 80 mA to 120 mA, in agreement with the variation of the transmittance, as shown in Fig. 4. It can also be seen from Fig. 4 that the transmittance of the ITO films increases gradually with the ion beam current, resulting in the decrease of the extinction coefficient as shown in Fig. 5b. The refractive index does not show a regular variation with the ion beam current. The films prepared at 80 mA and 120 mA given a high refractive index and the films prepared at 90 mA and 100 mA shown a low refractive index. The refractive index is related with the film packing density. The film with high packing density will have high refractive index. That means the films prepared at 80 mA and 120 mA have

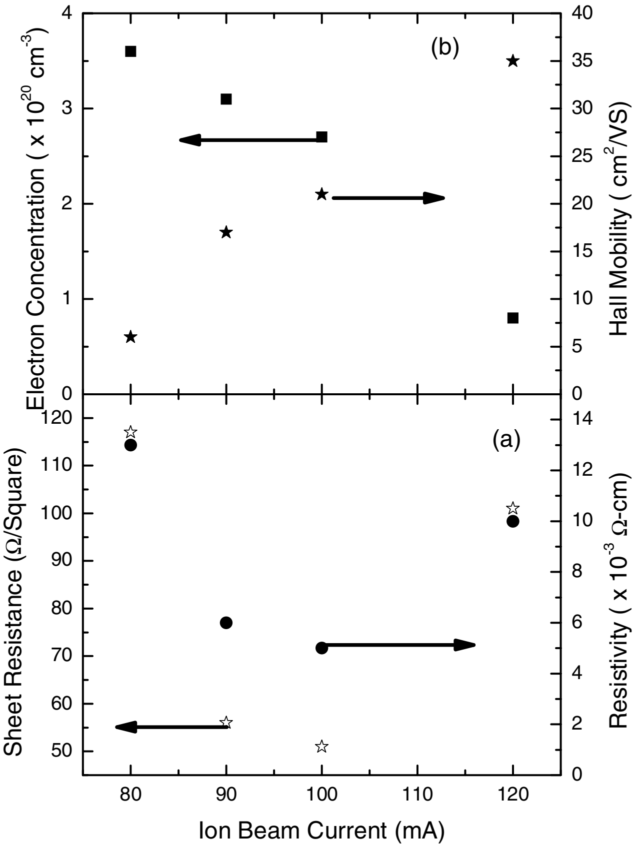

Figure 6 Sheet resistance and resistivity (a), and carrier concentration and Hall mobility (b) of ITO films deposited at different ion beam currents.

higher packing densities than the films prepared at 90 mA and 100 mA. Generally, an amorphous film has a compact structure and a high packing density; a polycrystalline film has a columnar structure and a low packing density. That is why films prepared at 80 mA ion beam current have a high refractive index. At the high ion beam current (120 mA), the film structure may be improved, the number of the void may decrease and results in a small increase of the refractive index. In fact, it can be seen that the variation of the refractive index in the visible region is small, the big divergence only happens in the infrared region. The variation of the refractive index in the infrared region results not only from the difference of the packing density, but also from the behaviour of the free carriers. Both the carrier concentration and mobility will influence the film refractive index in this region. The electrical properties of the ITO films prepared at different ion beam currents are given in Fig. 6. It can be seen from Fig. 6a that the electrical resistivity and the sheet resistance of the ITO films decrease as the ion beam current increases and reach the minima values at 100 mA ion beam current, and then increase as the ion beam current is increased further. The Hall measurements show that although the electron concentration in ITO films decreases as the ion beam current is increased, the Hall mobility increases as the ion beam current is increased. The electrical resistivity is related with both the electron concentration and the Hall mobility. Although the ITO film prepared at 100 mA ion beam current has a low electron concentration, it has a high Hall mobility, resulting in a low electrical resistivity. The decrease of the electron concentration with the ion beam current can be related with the disappearance of the oxygen vacancies. As the ion beam current is increased, the amount of the oxygen ions reaching the substrate also increases; it will compensate the loss of the oxygen during the evaporation process and reduce the amount of the oxygen vacancies, producing then a low electron concentration. The Hall mobility is related with the film structure. As the ion beam current is increased, the oxygen vacancies vanish, the scattering possibility of the carriers by oxygen vacancies decreases and results in an increase of the Hall mobility, as shown in Fig. 6b.

The reflectance of the ITO films prepared at different ion beam currents in the infer-red region is given in Fig. 7. This feature is very important for the application of the ITO films in the electromagnetic shield area. It can be seen that only the films prepared at 90 mA and 100 mA give high reflectance in IR region. There is a very simple relation between the infrared reflection R and the sheet resistance $R_{\square}$ [23, 24]:

$$R_{\mathrm{IR}} = (1 + 2\varepsilon_0 c_0 R_{\square})^{-2} . \quad (2)$$

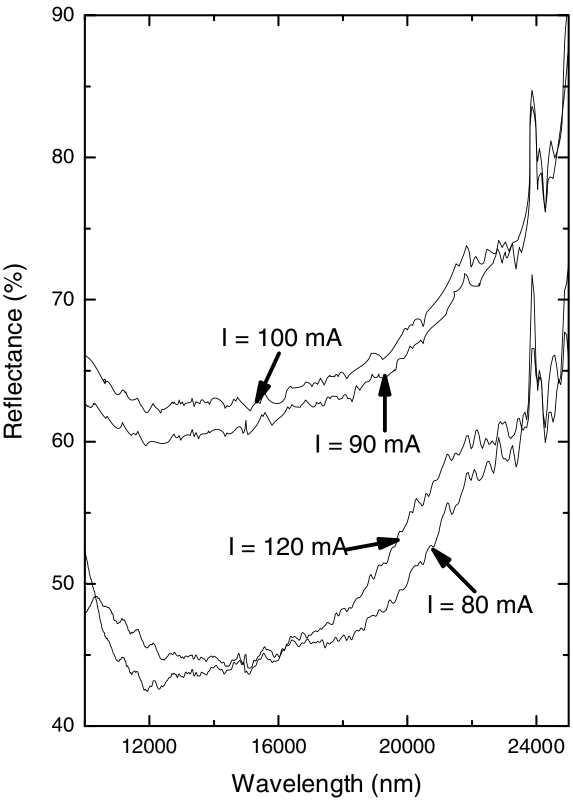

Figure 7 Infrared reflectance spectra for ITO films deposited at different ion beam currents.

It can be seen that the high sheet resistance will result in a low infrared reflection. Table 1 shows that the films prepared both at 80 mA and 120 mA have high sheet resistance and then a low reflectance in the infrared region.

4 Conclusions ITO films have been deposited onto glass substrates at room temperature by ion beam assisted deposition technique at different ion beam currents (80–120 mA). It has been found that the ion beam current has a great influence on ITO films properties. The lowest electrical resistivity (5×10^{-3} Ω cm) with good transmittance has been obtained for the ITO film prepared at 100 mA ion beam current. It gives the way to deposit ITO films with good qualities without extra heating of the substrate during the deposition process. The FTIR measurements also show that the film prepared at 100 mA has the highest reflectance in the IR region. Therefore, the ITO film prepared at 100 mA ion beam current shows a good quality, high transmittance in the visible region, low electrical resistivity and high infrared reflectance, and then seems to be the most adequate for the applications both in the transparent conductive electrode area and in the electromagnetic wave shield area.

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