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Low-resistivity indium tantalum oxide films by magnetron sputtering

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ABSTRACT Low-resistivity Ta-doped In_2O_3 (InTaO) films from ceramic targets of In_2O_3 doped with 2, 5, and 10 wt % Ta_2O_5 were deposited on Corning glass # 1737 substrates by magnetron sputtering. The electrical and optical properties of these films were studied. The carrier type of InTaO films was found to be *n*-type. The resistivity, carrier density, and Hall mobility of InTaO films were in the range of $0.28–200.2 \times 10^{-4}\ \Omega$ cm, $0.2–7.4 \times 10^{20}$ cm^{-3}, and 3–31 cm^2 V^{-1} s^{-1}, respectively. A minimum resistivity of $2.8 \times 10^{-4}\ \Omega$ cm with a mobility of 31 cm^2 V^{-1} s^{-1} and a high transparency of 85% in the visible were achieved for the InTaO thin films doped with 5 wt % Ta_2O_5.

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Transparent conductive oxides (TCOs) are unique materials that exhibit mutually exclusive material properties simultaneously – low electrical resistivity and high optical transparency. Generally, TCOs have a wide band gap ($E_g > 3$ eV), which is responsible for their optical transparency, and have relatively large carrier densities (10^{20}–10^{21} cm^{-3}), which account for their low electrical resistivity. By utilizing these unique properties, TCOs are widely used in a variety of applications such as optoelectronics, smart windows, antistatic coatings, and solar cells [1–3]. Among various TCOs, Sn-doped In_2O_3 (ITO) is the most widely used for optoelectronic applications because ITO thin films have high transmittance and low resistivity. The reported minimum resistivity of ITO is smaller than 0.1 mΩ cm [4]. However, new materials with improved transparency and good conductivity would be of great interest in the rapidly developing technology of optoelectronics. In search of an alternative TCO with a higher transmittance in the visible and a lower resistivity, we have investigated the electrical and optical properties of Ta-doped In_2O_3 (InTaO) thin films. In this study, film properties such as resistivity, carrier density, mobility, and optical transparency were investigated by varying the sputtering conditions such as the deposition temperature (T_S) and the oxygen partial pressures (p_{O_2}). In this paper, we report the structural, electrical, and optical properties of InTaO films prepared by either dc or rf magnetron sputtering. Through doping and deposition-parameter controls, a minimum resistivity of 0.28 mΩ cm was obtained. Although this resistivity value is slightly higher than that of ITO, InTaO is found to be one of the most conducting TCOs.

The targets, which were sintered disks containing 2, 5, and 10 wt % of Ta_2O_5 mixed with In_2O_3, were 2″ in diameter and 0.25″ in thickness. The densities of the targets used were estimated to be 60%–70% of the theoretical ones. Of the Ta-doped In_2O_3 (InTaO) films on Corning glass # 1737 substrates, the In_2O_3 targets doped with 2 wt % and 10 wt % Ta_2O_5 were deposited by dc sputtering and the In_2O_3 target doped with 5 wt % Ta_2O_5 by rf sputtering. The base pressure of the sputtering chamber was 1×10^{-6} Torr and the target-to-substrate distance was 7 cm. The depositions of InTaO films were performed with mixed argon gas and oxygen gas (the oxygen partial pressure, p_{O_2}, was varied from 0 Torr to 10×10^{-5} Torr) at a total pressure of 5 mTorr. The InTaO film thickness was measured by a profilometer (Alpha-Step 500 surface profiler, KLA-Tencor). The compositions of the InTaO films were measured by energy-dispersive spectroscopy (EDS) and were in agreement with the target composition within experimental errors. The crystal structure of the films was analyzed with X-ray diffraction (XRD) with Cu K_α radiation. The resistivity (ϱ), carrier density (n_H), and Hall mobility (μ_H) were measured with the van der Pauw method [5] at room temperature in a magnetic field of 3 kOe. The temperature dependence of the resistivity was measured by a standard four-point-probe technique. The optical transmission was measured using a spectrometer equipped with an optical multi-channel analyzer.

Figure 1 shows the XRD (θ–2θ) diffraction patterns of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 grown at a deposition temperature (T_S) of 350 °C. The partial pressures of the oxygen (p_{O_2}) used during the deposition of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 0×10^{-5} Torr, 0×10^{-5} Torr, and 2×10^{-5} Torr, respectively. The oxygen partial pressures

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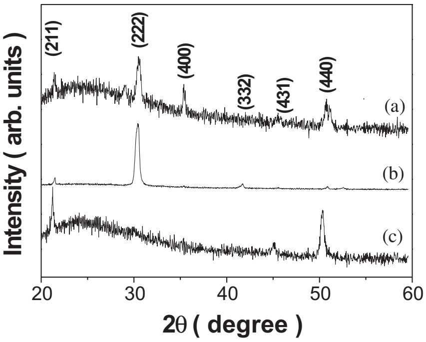

FIGURE 1 X-ray diffraction patterns of InTaO films doped with (a) 2, (b) 5, and (c) 10 wt % of Ta_2O_5 grown at substrate temperature (T_S) of 350 °C. The partial pressures of oxygen (p_{O_2}) used during the deposition process of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 0×10^{-5} Torr, 0×10^{-5} Torr, and 2×10^{-5} Torr, respectively

were chosen because the InTaO films grown at T_S of 350 °C with varying p_{O_2} have minimum resistivity values (Fig. 2). The XRD pattern of the InTaO film shows the characteristic peaks of the cubic bixbyite structure. Each peak in the diffraction patterns of the films was indexed based on the cubic bixbyite structure. With the assumption that $InTaO_3$ films have a cubic bixbyite structure, the lattice parameter was obtained. The values of the lattice parameters of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 1.016 nm, 1.024 nm, and 1.024 nm, respectively. The values of the lattice parameters for the InTaO films were similar to those (1.01–1.02 nm) for the 10% tin-doped In_2O_3 (ITO) films [6].

The electrical properties of the InTaO films were found to depend on the oxygen partial pressure p_{O_2}. Figure 2 shows the dependences of the electrical resistivity of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 grown at 350 °C on the deposition oxygen partial pressure. The resistivity minima of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were located at low oxygen partial pressures p_{O_2} of 0×10^{-5} Torr, 0×10^{-5} Torr, and 2×10^{-5} Torr, respectively. With further increasing oxygen partial pressure, the resistivity increased. It is believed that the low resistivity of InTaO films grown at a low p_{O_2} is related to the creation of oxygen vacancies. On the other hand, the high resistivity of the InTaO films grown at a high p_{O_2} may be explained by the decrease in the carrier density due to a decrease in oxygen vacancies. The minimum resistivity value of the InTaO films doped with 5 wt % Ta_2O_5 was found to be as small as 0.28 mΩ cm. This magnitude is slightly larger than that (∼ 0.1 mΩ cm) of the optimally prepared ITO films [4], and is smaller than that of most TCO materials such as $GaInO_3$, $ZnSnO_3$, and ZnO-In_2O_3 [7–9]. In addition, there is a great potential to further decrease the resistivity of InTaO films by optimizing the doping level and growth details. Although very small resistivity values for InTaO films have been observed, the origins of the low resistivity in the present materials are not clear yet. The carrier type of InTaO films was found to be *n*-type. The measured Hall mobilities and carrier densities of the InTaO films were in the range of 3–31 $cm^2 V^{-1} s^{-1}$ and 0.2–6.4 × 10^{20} cm^{-2}, respectively. The Hall mobility and carrier density of the InTaO film with a resistivity of 0.28 mΩ cm were 31 $cm^2 V^{-1} s^{-1}$ and 6.4×10^{20} cm^{-2}, respectively.

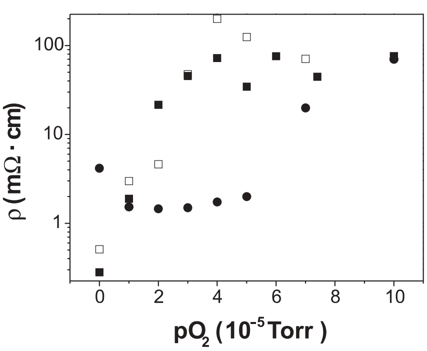

FIGURE 2 Dependence of the electrical resistivity of InTaO films doped with (□) 2, (■) 5, and (●) 10 wt % of Ta_2O_5 grown at substrate temperature of 350 °C on partial pressure of oxygen (p_{O_2})

The deposition temperatures were found to affect the electronic properties of the InTaO films. Figure 3 shows the dependence of the electrical resistivity (ϱ) of the InTaO films on the deposition temperature (T_S) varying from 20 °C to 350 °C. The deposition oxygen partial pressures of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 0×10^{-5} Torr, 0×10^{-5} Torr, and 2×10^{-5} Torr, respectively. The resistivity of the InTaO films grown at $T_S = 25$ °C doped with 2, 5, and 10 wt % Ta_2O_5 were 0.9 mΩ cm, 4.1 mΩ cm, and 8.4 mΩ cm respectively: the magnitude of the resistivity increased with the increase of the doping level. The resistivity decreased with increasing T_S. The resistivity of the InTaO films grown at 350 °C doped with 2, 5, and 10 wt % Ta_2O_5 were 0.51 mΩ cm, 0.28 mΩ cm, and 1.25 mΩ cm, respectively. It is interesting to note that the resistivity difference $\Delta\varrho$, which is defined as $\varrho(T_S = 25\,°C) - \varrho(T_S = 350\,°C)$ where $\varrho(T_S = 25\,°C)$ and $\varrho(T_S = 350\,°C)$ are the resistivity of the InTaO films grown at $T_S = 25$ °C and $T_S = 350$ °C, respectively, increased with increasing doping. The deposition-temperature dependence of $\Delta\varrho$ may be related to the deposition-temperature dependence of the films' crystallinity and oxygen vacancies and the diffusion of Ta atoms from the interstitial locations and grain boundaries into the indium cation sites. The dependence of the crystallinity and oxygen vacancies on T_S may be similar for all doping levels, and therefore this may not explain the doping dependence of $\Delta\varrho$. For higher doping, it is expected that a higher deposition temperature is required for the Ta dopants' diffusion from the interstitial locations and grain boundaries into the indium sites, Ta atoms substituting In atoms. When Ta substitutes In, Ta in an InTaO film donates two electrons to the conduction band because In has a valence state of three and Ta has a valence state of five. Thus, the deposition-temperature dependence of the Ta diffusion effect is expected to be relatively large for highly doped films and small for lowly doped films. For this reason, with increasing deposition temperature the carrier density of highly doped InTaO films may in-

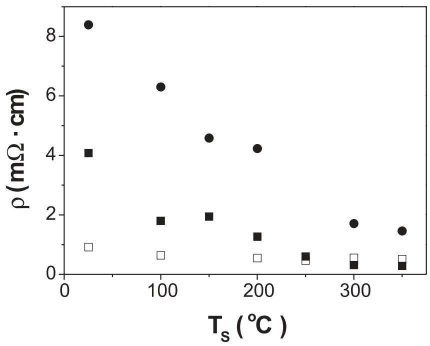

FIGURE 3 Dependence of the electrical resistivity of InTaO films doped with (□) 2, (■) 5, and (●) 10 wt % of Ta_2O_5 on substrate temperatures. The partial pressures of oxygen (p_{O_2}) used during the deposition process of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 0×10^{-5} Torr, 0×10^{-5} Torr, and 2×10^{-5} Torr, respectively

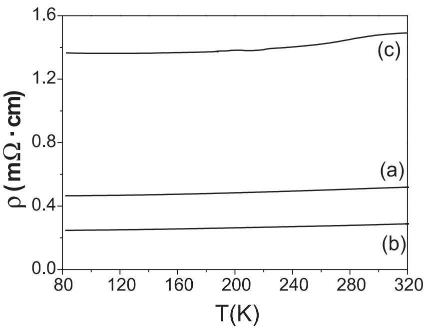

FIGURE 4 Temperature dependence of the resistivity of InTaO films doped with (a) 2, (b) 5, and (c) 10 wt % of Ta_2O_5 grown at 350 °C. The partial pressures of oxygen (p_{O_2}) used during the deposition process of the InTaO films doped with 2, 5 , and 10 wt % Ta_2O_5 were 0×10^{-5} Torr, 0×10^{-5} Torr, and 2×10^{-5} Torr, respectively

Ta doping concentration (wt %)	Resistivity (10^{-4} Ω cm)	Carrier density (10^{20} cm^{-3})	Mobility (cm^2 V^{-1} s^{-1})	Energy gap E_g (eV)	Mean transmission (%) (400–700 nm)
0.02	5.10	4.4	18	3.7	82
0.05	2.8	7.4	30	3.8	85
0.10	12.5	2.7	15.5	3.7	80

TABLE 1 Electrical and optical properties of InTaO films deposited at 350 °C

crease more rapidly than that of lowly doped InTaO films, as observed: the carrier density of the InTaO films doped with 2 wt % Ta_2O_5 was found to decrease from 6.6×10^{20} cm^{-3} at T_S of 25 °C to 3.6×10^{20} cm^{-3} at T_S of 350 °C with increasing T_S, whereas the carrier density of the InTaO films doped with 10 wt % increased from 1.5×10^{20} cm^{-3} at T_S of 25 °C to 2.7×10^{20} cm^{-3} at T_S of 350 °C with increasing T_S. $\Delta\varrho$ is, therefore, expected to be large for highly doped films and small for lowly doped films. As T_S increases, the crystallinity of the film improves, and the improvement of the crystallinity results in a decrease in the microscopic imperfections; thus the mobility increases. The values of the mobility of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 at T_S of 25 °C were 7 cm^2 V^{-1} s^{-1}, 4 cm^2 V^{-1} s^{-1} and 5 cm^2 V^{-1} s^{-1} respectively, and those of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 at T_S of 350 °C were 21 cm^2 V^{-1} s^{-1}, 31 cm^2 V^{-1} s^{-1}, and 15.5 cm^2 V^{-1} s^{-1}, respectively.

Figure 4 shows the temperature dependence of the resistivity of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 grown at 350 °C (XRD patterns are shown in Fig. 1). The InTaO films exhibit a metallicbehavior with relatively weak temperature dependence. The values of $\varrho(80\,K)$ for the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 1.34 mΩ cm, 0.25 mΩ cm, and 0.45 mΩ cm, and the values of $\varrho(295\,K)$ for the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 were 1.47 mΩ cm, 0.28 mΩ cm, and 0.51 mΩ cm, where $\varrho(80\,K)$ and $\varrho(295\,K)$ are the resistivity at 80 K and 295 K. For the 2, 5, and 10 wt % Ta_2O_5-doped films, the relative resistivity values defined as $\varrho(80\,K)/\varrho(295\,K)$ were 0.91, 0.88, and 0.93, respectively. This shows that the smaller the resistivity of a film, the smaller the value of $\varrho(80\,K)/\varrho(295\,K)$ for the film. In other words, the 5 wt % Ta_2O_5-doped InTaO has the least temperature-independent contribution coming from the grain boundaries and lattice imperfections to the resistivity.

The properties of the InTaO films doped with 2, 5, and 10 wt % Ta_2O_5 grown at 350 °C are summarized in Table 1. All the transmission values were normalized by the transmission of a bare glass substrate. The values of the energy gap, E_g, were determined by extrapolations of the straight regions of the plots of the square of the absorption coefficient α^2 vs. photon energy $h\nu$. The absorption coefficient α was determined by the equation $\alpha = \ln(1/T)/d$, where T is the transmission and d is the film thickness [10]. The calculated band-gap energies and mean transmissions are listed in Table 1.

In summary, we have shown that Ta-doped In_2O_3 films with excellent electrical and optical properties are able to be grown by magnetron sputtering from In_2O_3 targets doped with 2, 5, and 10 wt % Ta_2O_5. The substrate temperature and oxygen partial pressure dependence of these films have been studied. The minimum resistivity of InTaO films was 0.28 mΩ cm with more than ∼ 85% transparency in 400–700 nm. These features make InTaO films a potential candidate for low-resistivity TCOs in optoelectronics and other applications.

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