Effect of In-situ He Ion Beam Irradiation on the ITO Films Prepared by IBS Method

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ABSTRACT
Effect of in-situ He ion beam irradiation on the indium tin oxide (ITO) Films prepared at low temperatures by ion beam sputtering (IBS) Method were studied. Optical transmittance and electrical resistivity were then measured. In addition, the theoretical calculation of the Hall mobility and carrier density in ITO films by the Van der Pauw's method is discussed. The films' microstructure were further studied by X-ray diffraction measurements. The transmittance also appeared higher and electrical resistivity lower compared to films not exposed to irradiation, respectively.

Keywords: indium tin oxide, transparent electrical film, argon ion beam, ion beam sputtering method, helium ion beam irradiation.

1. INTRODUCTION
Transparent electrical conductive indium tin oxide (ITO) films are manufactured by doping In_2O_3 with SnO_2. Because of its properties, ITO has been used in a variety of optoelectronic devices including liquid crystal displays and solar cells [1]. Generally, ITO thin films are manufactured either by electron beam evaporation [2] or magnetron sputtering [3]. In both methods, thin films are produced under substrate temperatures of about 300 - 400 ℃. However production on polymer films usually requires a substrate temperature of less than 150 ℃. This makes the conventional process difficult.

Ion Beam Sputtering is yet another procedure for producing thin film without overexposing its surface to high energy particles. In this method, two separate chambers for generating plasma and producing thin film are utilized. Direct contact between the film surface and the plasma is avoided, thus minimizing the rise in substrate temperature during the film growth process. Our research aim is to prepare ITO thin films under low substrate temperatures using the dual ion beam sputtering method and to investigate the influence of irradiating oxygen onto the growing film.

2. DUAL ION BEAM SPUTTERRING DEPOSITION SYSTEM
The ion beam sputtering deposition system used in our study is schematically

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illustrated in Fig. 1. The apparatus is made of two Kaufman type ion guns, one for sputtering and the other for assisting. The Ar sputtering gun is located approximately 45 degrees from the ITO target was used to sputter target material. He gas inserted into the assist gun also located approximately 45 degrees from the substrate was irradiated onto the growing film surface. The aim here is to give substantial energy to the growing film through He. O_2 gas was introduced into the chamber near the substrate. In_2O_3 sintered with 10 %wt SnO_2 was used as the target and a water-cooler was attached to its reverse side. The substrate holder is a rotating type (rotation velocity; 0 - 10 rpm). A substrate heater with a heating potential of up to 450 ℃ was employed. A molecular turbo pump was used as the main vacuum pump and an oil-sealed rotary pump as the auxiliary.

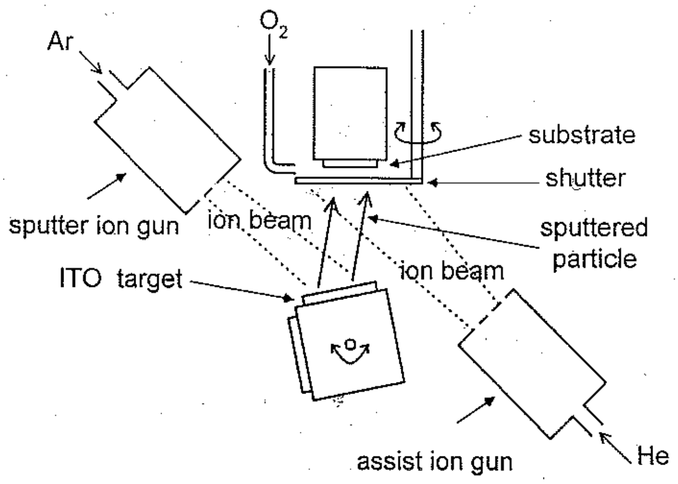

Fig .1 schematic diagram of dual ion beam sputtering deposition system.

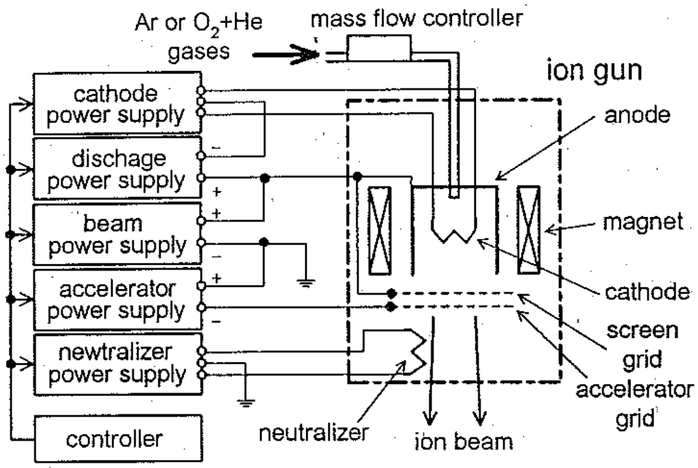

Fig. 2 Schematic diagram and electric system of Kaufman type ion gun.

Two ϕ 3 cm Kaufman type ion guns (3.0-1500-100, Iontech Inc.) generated by two independent power supplies (MSP-3000FC, Iontech Inc.) are the main components in our experimental apparatus. Fig. 2 shows the schematic diagram and electric system of the Kaufman type ion gun. Thermal electrons released from the cathode made their way to the anode in a spiral orbit. The impact from the collision ionized the gas into plasma, consequently the ion from the plasma was extracted with a multi-hole screen grid and accelerator grid under high vacuum pressure. The screen grid bears the same electric potential as the cathode but the accelerator grid has relatively lower electric potential. We can therefore conclude that the energy of the ion beam depended on the beam voltage. A neutralizer was fixed on the front part of the accelerator grid. Thermal electrons were released from the neutralizer and these electrons neutralized the ion beam, preventing charge-up on the target and other parts of the apparatus.

3 EXPERIMENTAL DETAILS

ITO films were prepared on Corning glass (#7059) substrates. Experiments were carried out at room temperature, sputter ion gun Ar gas flow rate was 4 ccm, beam voltage; 1000 V and beam current; 30 mA. The assist ion gun He gas flow rate was 100 ccm, beam voltage; 70 or 150 V and beam current; 2 mA. O_2 gas flow rate was 2 - 8 ccm. The O_2 partial pressure was 9.0×10^{-4} - 2.7×10^{-3} Pa, background pressure was 1.1×10^{-3} Pa and the total pressure was approximately 4.0×10^{-2} Pa.

Average visible transmittance was measured with a spectrophotometer (UV-3100, Shimazu Co., Ltd.), with a wavelength range of 400 ~ 800 nm. Electrical resistivity was measured by the four-point probe method. · Hall mobility and carrier density were obtained using the van der Pauw's method (magnetic flux density: 0.68 T) [4]. · Film thickness was measured using a multiple-beam interferometer (BM-4, Mizojiri Optical Co., Ltd.) and microstructure of the film was observed with a field emission scanning electron microscopy, FE-SEM (S-900, Hi-tachi, Ltd.). X-ray diffraction was performed with 40kV-30mA CuKα radiation (M03X-HF, Mac Science Co., Ltd.) using the θ-2θ method.

4 RESULTS AND DISCUSSIONS

4.1 Optical and electrical properties of the film

Fig. 3 shows variations in electrical resistivity and optical transmittance relating to the assisted ion beam voltage and O_2 gas flow rate. The triangular points indicate the properties of the films which were prepared with regular gas flow and without irradiation from the assist ion gun. The transmittance was 80 - 90 % and improved with increasing O_2 gas flow rate. On the other hand, electrical resistivity fell to its lowest value when beam voltage was 70 V and decreased with an increase in O_2 gas flow rate. When the assist He ion beam was irradiated onto the surface of the growing film with a beam voltage of 70 V and an O_2 gas flow rate of 6 ccm, we observed a minimum value 7.0×10^{-4} Ωcm. However an increase was recorded when the assist beam was irradiated at 150 V.

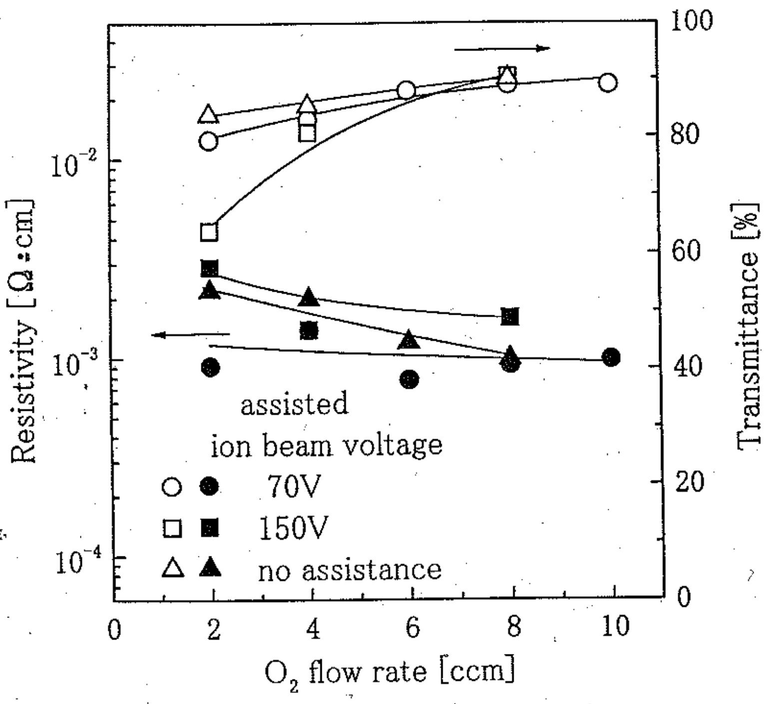

Fig. 3 Variations in electrical resistivity and optical transmittance relating to with the assisted ion beam voltage and O_2 flow rate.

Fig. 4 shows variations in carrier density and Hall mobility relating to assisted ion beam voltage and O_2 gas flow rate. Hall mobility improved with an increasing O_2 gas flow rate and decreased with a higher beam voltage. On the other hand, carrier density grew when the assist beam was irradiated and became maximized with a beam voltage of 70 V. It was concluded that a decrease in the electrical resistivity with the assisted ion beam voltage of 70 V and an O_2 gas flow rate of 6 ccm results in the increase of carrier density.

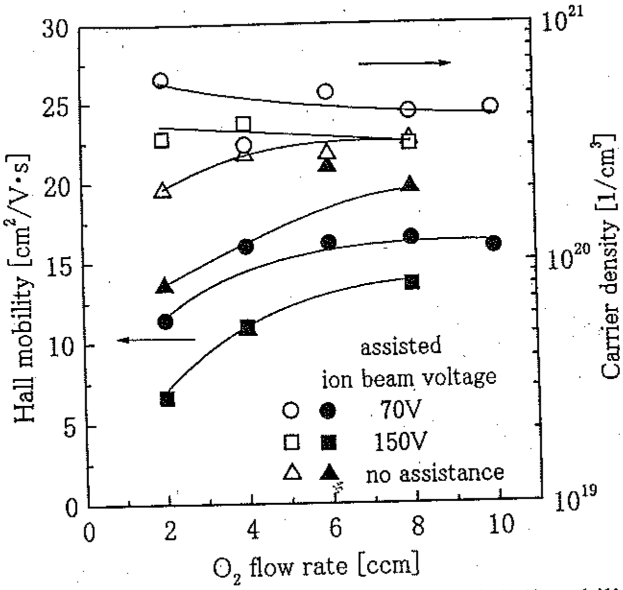

Fig. 4 Variations in carrier density and Hall mobility relating to assisted ion beam voltage and O_2 flow rate.

4.3 Analysis of crystal structure using X-ray diffraction method

Fig. 5 (a) - (c) show typical X-ray diffraction profiles of the ITO films prepared with (a) no assistance, with assisted ion beam voltage of (b) 70 V and (c) 150 V. As a reference, peak positions of the In_2O_3 crystal powder are included. The measured peak positions were in good agreement with the In_2O_3 crystals, although were seen to shift slightly to the lower angle side. It was therefore assumed that ITO crystals could be obtained.

Fig. 6 shows the variation in crystal grain size with the assisted ion beam voltage. Values were calculated using Scherrer's equation. Grain size lessened with an increased beam voltage. The grain boundary appeared to grow wider, which could be attributed to a drop in Hall mobility and carrier density.

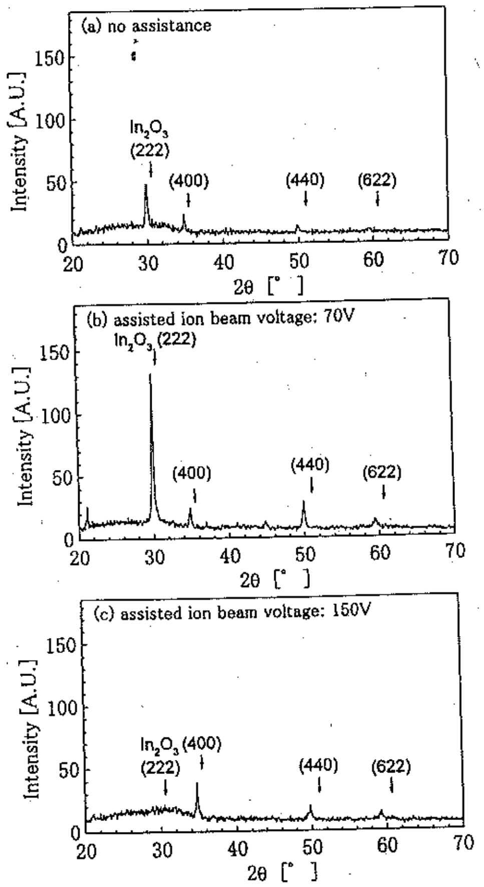

Fig. 5 Typical X-ray diffraction profiles of ITO films. (a) no assistance, (b) assisted ion beam voltage: 70 V, (c) 150 V.

Owing to an increase in beam voltage irradiated onto the surface of the growing film, the ITO crystal structures improved and transmittance increased. However, more damage on the film surface occurred, inhibiting grain size and widening the grain boundary, thus adversely effecting the Hall mobility and carrier density of the film as a whole.

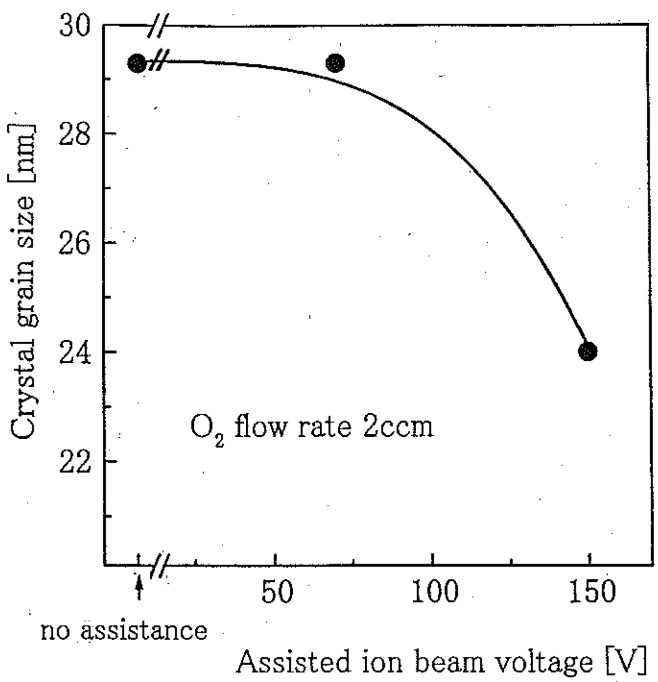

Fig. 6 Grain size calculated from (222) peak shown in Fig.5

5. CONCLUSION

Preparation of ITO films at low temperatures using dual ion beam sputtering and the effects of irradiated growing film surfaces with He were studied. The results obtained are as follows:

1) Irradiation of an assisted He ion beam with a beam voltage of 70 W resulted in higher quality ITO crystal structures even when prepared at room temperature. Transmittance also appeared higher and electrical resistivity lower compared to films not exposed to irradiation, respectively.
2) With an increasing beam voltage from the assisted ion beam, damage on the film became more apparent. Due to the diminution in crystal grain size and a widening of the grain boundary, the cumulative Hall mobility and carrier density of the film decreased while resistivity increased.

Our next task is to prepare ITO films without causing damage to their surface by controlling the assisted ion beam and manipulating the oxygen atoms that pass through them. Furthermore, by measuring the amount of ions and neutrons present in the ion beam, we plan to conduct study into its role in determining agent of the crystal structure.

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