Indium Tin Oxide (ITO) Thin Films Deposited by MirrorTron Sputtering System on Unheated Poly (Ethylene-Naphthalate) Film Substrate

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ABSTRACT

We performed Indium Tin Oxide (ITO) film formation to develop electrode of flexible solar battery by the MirrorTron Sputtering method. The MirrorTron Sputtering method divides the sputtering chamber and the film formation chamber in order to eliminate damage to specimens by plasma high energy particles. As a result, the ITO film with uniform surface is formed using the MirrorTron Sputtering method by the analysis using the Energy Dispersive X-ray Spectroscopy (EDX) and the Scanning Electron Microscope (SEM). Therefore, the analysis of ITO film is performed quantitatively.

KEYWORDS: Indium Tin Oxide, MirroTron Sputtering, Solar Cell, Poly (Ethylene-Naphthalate) Film

1. INTRODUCTION

In 2009, Barack Obama, President of the United States, declared Energy and Environmental policy called Green New Deal. This is the main provision of economic measures and employment creation for the solar energy industry is planned as a part of it. The solar energy market in the world gained attention.

Currently, power generation efficiency of the solar cell in practical use is about 30 % and the research regarding the improvement of the power generation efficiency has been performed actively in the world. From now on, improvement of the power generation efficiency is gaining attention as a key technology that will lead to the further development of the solar cell industry.

The basic type of the solar cell sold in the market uses crystal system or amorphous materials in general. In these years, dye-sensitised solar cells using TiO_2 [1-3] which is a new construction as a material or solar cells using organic semiconductor material such as phthalocyanine die are proposed. However, general solar cells in the market are considered unable to expect improvement of the power generation efficiency as long as the basic construction and materials are used. Therefore, new usage of general solar cells using current crystals system or amorphous materials are reexamined aiming further diffusion of the solar cells for ordinary households. Furthermore, even though the plasma chemical vapor deposition (CVD) [4-6] is the general

film formation technology of the solar cell, there are various technical problems as a film formation technology for the solar cell.

Therefore, we are researching the development of the film formation technology and the film formation equipment of the flexible solar cell [7,8] which has been gaining attention as a usage of new solar cells in the world. The flexible solar cell can change the shape of the solar battery easily and enables light-weight and large area, which are the features and advantages of the flexible solar cell.

The main purpose of this report is to develop the film formation technology of the battery material having the cutting-edge power generation efficiency in the flexible solar cell field research. This report is to propose Indium Tin Oxide (ITO) film formation technology on Poly (ethylene-naphthalate) (PEN) film by MirrorTron Sputtering method which is the new film formation technology of the sputtering method. Here we report that the ITO film is formed on the PEN film uniformly using the MirrorTron Sputtering method which has been checked as a result of the chemical analysis.

2. EXPERIMENTAL PROCEDURE

Fig.1 shows the outline of the MirrorTron Sputtering system equipment using the MirrorTron Sputtering method. The basic construction of the equipment in this figure is direct current power supply and a magnet, which is similar to the conventional sputtering method. The different point of this equipment is that the metal target and the substrate are arranged to be at a right angle.

Conventional sputtering method has various problems such as the substrate temperature becomes high because the substrate exists in the plasma, ion in the plasma sputters repeatedly, plastic substrates are unable to use because the substrate potential is always in negative condition and the collision of the secondary battery is repeated, and film formation is uneven. However, main features as shown below are improved by the MirrorTron Sputtering method compared to the conventional sputtering method.

① Damage by plasma high energy particles to the specimen is decreased by dividing the sputtering chamber and the film formation chamber and the film formation reproducibility is improved at a high level.

② Very high etching rate film formation is realized compared to the conventional sputtering method.

③ Controllability of the film composition is improved drastically by establishing an oxidation chamber for the sputtering substance.

④ Film formation on a plastic substrate is realized at a low temperature, which was impossible by the conventional sputtering method because the film formation chamber is less subject to plasma.

In this experiment, ITO target of 100 x 500 mm^2 in size and plasma gas of Ar (50 sccm) + O_2 (2.0 sccm) mixture gas is used for the film formation. Each condition of the thin film formation is: film formation rate 150 nm/min, plasma electric power 1.6 kW, film formation duration 2700 sec, magnetic field strength at the center 25 mT, film formation temperature 100℃, and achieved gas pressure 3.72 x 10^{-5} Pa.

Fig.2 shows the constitutional formula of Poly (ethylene Naphthalate) (PEN) film (Teijin DuPont Films; Q51) used for the plastic substrate which is used for the film formation. ITO film is deposited on the PEN film to use as the electrode material for the solar cell. The feature of the PEN film is that it has superior heat resistance and mechanical characteristics.

The crystal architecture of the ITO thin film surface formed by the MirrorTron Sputtering method is observed visually using the Scanning Electron Microscope (SEM) (Hitachi; S-4100). Also the ITO thin film surface is analyzed quantitatively by the Energy Dispersive X-ray Spectroscopy (EDX) (Shimadzu Co.; EDX-720) analysis.

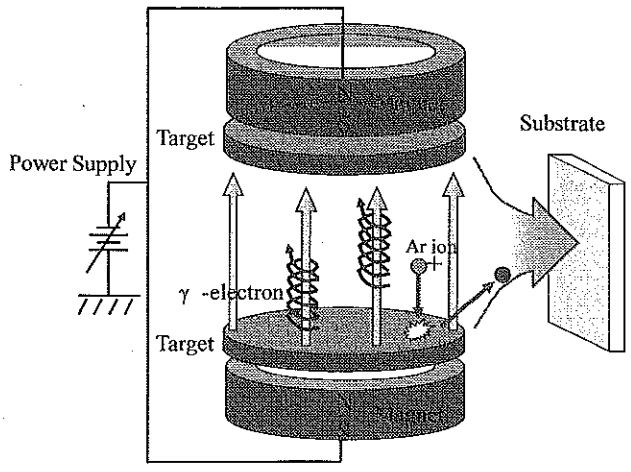

Fig.1 Schematic of MirroTron Sputtering system.

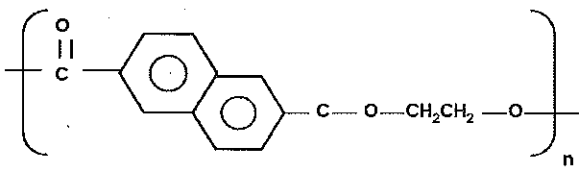

Fig.2 Chemical structure model of the PEN film.

3. RESULTS AND DISCUSSION

Fig.3 shows the SEM image of the ITO thin film surface formed by the MirrorTron Sputtering method observed by SEM. It shows that the crystal construction of the formed ITO thin film surface is uniform.

Fig.4 shows qualitative analysis spectrum (element: Ti-U) when the ITO film surface formed by the MirrorTron Sputtering method is analyzed quantitatively using thin film FP method with the EDX analysis. In the line (a) indicates spectrum of qualitative analysis of the non-deposited PEN film as a reference. In the line (b) indicates spectrum of qualitative analysis of the deposited ITO thin film. When the lines (a) and (b) are compared, spectrum of peaks of -InKa, -SnKa, -InKb and –Inkb2 are notable in the line (b). This show that all spectrums are the result of the element analysis spectrum regarding the ITO thin film and the formed ITO thin film can be evaluated qualitatively using the MirrorTron Sputtering method.

Fig.5 shows qualitative analysis spectrum (element: Na-Sc) when the ITO film surface formed by the Mirrortron Sputtering method is analyzed quantitatively using thin film FP method with the EDX analysis. The **Fig.4** (a) indicates spectrum of qualitative analysis of the non-deposited PEN film as a reference and the figure (b) indicates spectrum of qualitative analysis of the formed ITO thin film. When the lines (a) and (b) are compared, spectrum of peaks of -InLa, -SnLa InLb1 –InLb2 and –InLg1 are notable in the line (b). All spectra indicate the element analysis spectrum related to ITO thin film.

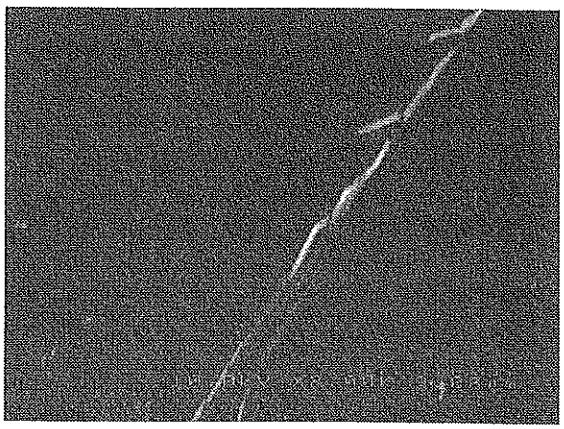

Fig.3 Photograph of SEM image (ITO thin film).

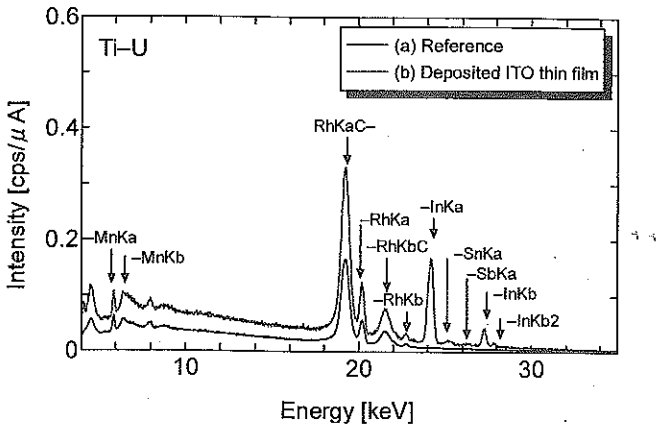

Fig.4 EDX spectrum in the region from Ti-U.

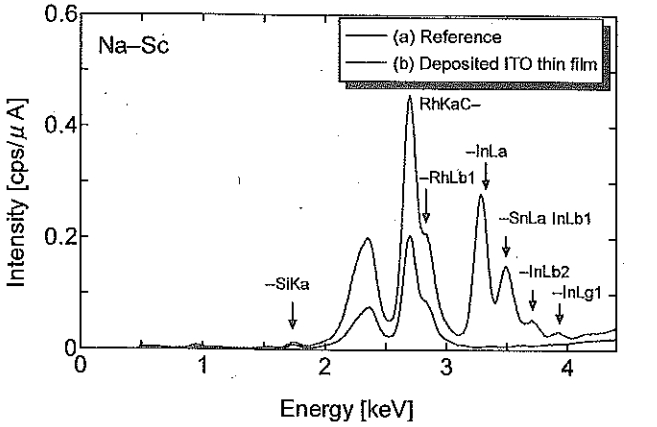

Fig.5 EDX spectrum in the region from Na-Sc.

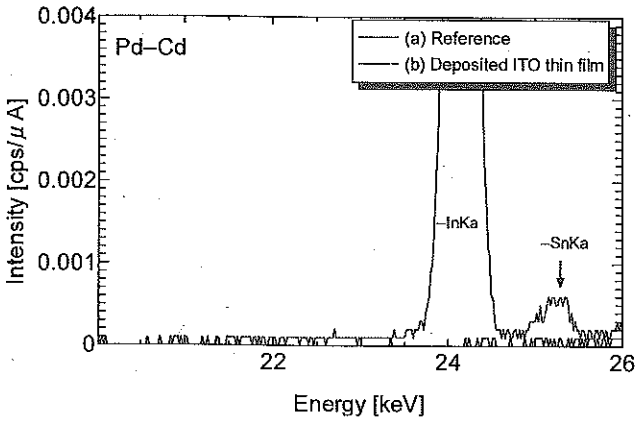

Fig.6 EDX spectrum in the region from Pd-Cd.

Fig.6 shows qualitative analysis spectrum (element: Pd-Cd) when the ITO film surface formed by the Mirrortron Sputtering method is analyzed quantitatively using thin film FP method with the EDX analysis. In the line (a) indicates spectrum of

Table 1 Quantitative analysis of EDX

Layer 1	Poly (ethylene Naphthalate)
	15.200 mg/cm^2
Si	0.051 %
Sb	0.010 %
Mn	0.007 %
$C_{10}H_8O_4$	99.916 %
Layer 2	Indium Tin Oxide
	84.747 μg/cm^2
In	95.774 %
Sn	4.226 %

qualitative analysis of the non-deposited PEN film as a reference and the line (b) indicates spectrum of qualitative analysis of the formed ITO thin film. The lines (a) and (b) are compared, spectrum of peaks of -InKa, -SnKa are notable in the line (a). All spectra indicate the element analysis spectrum related to PEN film. In the line (b), information of the element analysis spectrum of PEN film is disappeared because the ITO thin film is deposited using the PEN film as a plastic substrate.

Table 1 shows qualitative analysis spectrum when the ITO film surface formed by the MirrorTron Sputtering method is analyzed quantitatively using thin film FP method with the EDX analysis. The ITO thin film is deposited, referring the non-deposited PEN film in the **Table 1**. Layer 1 is a value data of the reference (PEN film: 15.200 mg/cm^2) and Layer 2 is a value data of the ITO thin film. It is confirmed that the ITO thin film surface formed by the MirrorTron Sputtering method is 88.308 μg/cm^2 thin film and the rate of the ITO thin film component is 95.774 % of In and 4.226 % of Sn.

4. CONCLUSION

The ITO thin film is produced on the PEN film by the MirrorTron Sputtering method successfully with this research. As a result of surface observation using the SEM, it is confirmed that a uniform thin film is formed. Also, as a result of quantitative analysis on the formed ITO thin film surface with the EDX analysis, the ITO thin film surface formed by the MirrorTron Sputtering method is a thin film of 88.308μg/cm^2.

The thin film production technology on the film substrate did not realize by the conventional sputtering method. However, material thin film production technology on the PEN film for the flexible solar cell has been realized using the MirrorTron Sputtering method.

From now on, we will start researching the flexible solar cell thin film material to realize high power generation efficiency by depositing plural layers of thin film materials used for the flexible solar cell on the PEN film using the MirrorTron Sputtering method.

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REFERENCES

[1] **T.Yuji, H.Akatsuka, N.Mungkung, B.W.Park and Y.M.Sung**; *Vaccum*, **Vol.83**-1, 2008, pp.124-127.

[2] **G.Syrrokostas, M.Giannouli and P.Yianoulis**; *Renewable Energy*, **Vol.34**-7, 2009, pp.1759-1764.

[3] **M.C.Kao, H.Z.Chen, S.L.Young, C.Y.Kung and C.C.Lin**; *Thin Solid Films*, **Vol.517**-17, 2009, pp.4741-4744.

[4] **T.H. Chung, M.S. Kang, C.J. Chung, Y. Kim**; *Current Applied Physics*, **Vol.9**-3, 2009, pp.598-604.

[5] **H. Wolf, Z. Pajkic, T. Gerdes, M. Willert-Porada**; *Journal of Power Sources*, **Vol.190**-1, 2009, pp.157-161.

[6] **K.Baba, R.Hatada, S.Flege and W.Ensinger**; *Surface and Coatings Technology*, **Vol.203**-17-18, 2009, pp.2747-2750.

[7] **M.Ikegami, J.Suzuki, K.Teshima, M. Kawaraya and T.Miyasaka**; *Solar Energy Materials and Solar Cells*, **Vol.93**-6-7, 2009, pp.836-839.

[8] **F.-J.Haug, T.Söderström, M.Python, V. Terrazzoni-Daudrix, X.Niquille and C.Ballif**; *Solar Energy Materials and Solar Cells*, **Vol.93**-6-7, 2009, pp.884-887.