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Large-Area ITO Deposition Technology and Its Application to Flexible Film Type Solar Cells

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We have been developing large-area flexible film type solar cells. We employed a simplified process based on roll-to-roll apparatus in order to reduce the production cost through mass production. In this process, we deposit each layers of solar cells stepwise in several chambers. Indium tin oxide (ITO) film is used as a transparent conductive electrode in the solar cells. Low resistive and high transparent ITO films should be deposited with good thickness uniformity in an area of 400mm by 800mm. Scanning magnet type sputtering apparatus was applied to improve the thickness uniformity which enabled us to deposit ITO film with a thickness uniformity of $\pm 1.5\%$ in the large area. ITO films deposited by the scanning magnet sputtering apparatus has slightly higher resistivity than that deposited by the conventional fixed magnet sputtering apparatus. ITO films deposited by the scanning method have accumulated structure of layers deposited in and out of the erosion areas. The optimum deposition conditions are different between in and out of the erosion, hence both conditions are not satisfied at the same time. The scanning magnet type sputtering apparatus achieves excellent film thickness uniformity in the large area at the sacrifice of optoelectric properties. The optimized lowest resistivity of ITO films deposited by the scanning magnet type sputtering is $2.4\times10^{-4}\Omega\cdot$cm.

Key words: ITO, Large-Area, Solar Cells, Resistivity

1. FLEXIBLE FILM SUBSTRATE AND ROLL-TO-ROLL PROCESS

Amorphous silicon (a-Si) and related thin films such as amorphous silicon germanium (a-SiGe) are promising materials for low cost solar cells. To make it real, however, a large-scale mass production of large-area solar cells is an essential requirement. Our approach at Fuji Electric to the practical mass production is to develop a simplified process based on roll-to-roll apparatus by adopting flexible substrate[1,2]. In this production process, we employed a roll of heat-resisting plastic film substrate. The advantageous roll-to-roll process offers large-area deposition, high throughput and excellent reproducibility because the whole cell structure can be fabricated without exposure to air and the deposition chamber can be maintained under a vacuum until the completion of roll processing.

The primarily important deposition apparatus named "stepping-roll" apparatus has been developed to deposit layered device structures. The stepping-roll apparatus has 13 chemical vapor deposition (CVD) chambers and 2 magnetron sputtering chambers in one common vacuum chamber. A photograph of the apparatus is shown in Fig. 1. An unwinding roll is set one side of the apparatus and a winding roll is set the other end. Polyimide heat-resisting film substrate is moved intermittently from one deposition chamber to the next deposition chamber. The stepping-roll apparatus can reduce contamination in a-Si (a-SiGe) films because each deposition chamber is separated in the common chamber, which is crucial for solar cell performance. We can produce more than several hundreds of large-area solar cells with a size of 400mm by 800mm using a rolled film without breaking a vacuum. A solar cell is fabricated every 2 or 3min in one step cycle with high reproducibility.

2. DEVICE STRUCTURE AND ITO FILM

We have developed a new series-connection device structure named SCAF (Series-Connection through Apertures formed on Film) as shown in Fig. 2[1,2]. In the structure, photo-generated electric current is efficiently collected by an indium tin oxide (ITO) layer and introduced to the reverse side electrode through holes formed on films, which reduce the resistance loss in the ITO film. Figure 3 shows a layered structure of a-Si/a-SiGe tandem solar cells fabricated on one side of the electrode. ITO film is placed on the top of the device. Incident light goes into the a-Si and a-SiGe intrinsic layers (i-layers) through the transparent ITO layer. Absorption coefficients of ITO films should be low in the wavelength region between 300 and 1000nm to reduce absorption loss and obtain higher generated current, resulting in higher conversion efficiency.

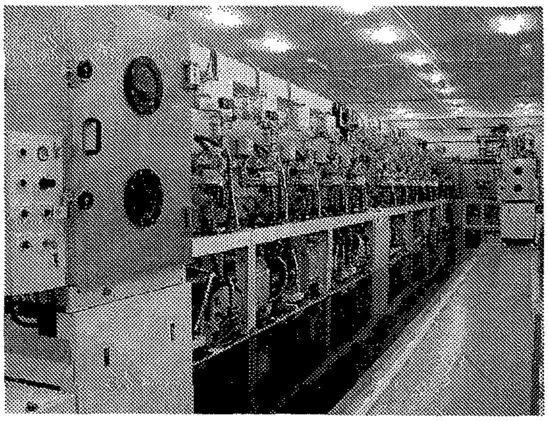

Fig. 1 Stepping-roll apparatus for flexible film type solar cell production using heat-resisting plastic film substrates. The apparatus has 13 CVD and 2 magnetron sputtering chambers.

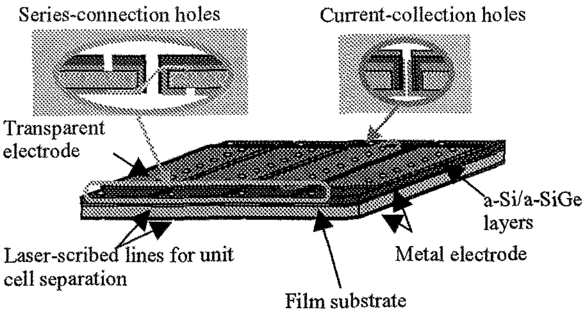

Fig.2 SCAF cell structure.

The ITO layer also has a role of an anti-reflection coating. Figure 4 shows the calculated quantum efficiency (a) and short circuit current density (Jsc) (b) depending on ITO film thickness. As shown in Fig. 4, ITO film thickness control the window position of low reflectance region and shift the peak of quantum

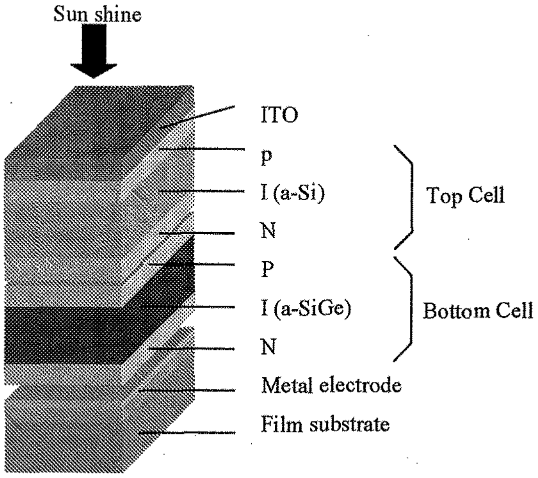

Fig.3 Structure of a-Si/a-SiGe tandem solar cell. ITO film is placed on the top of the device. The ITO film should show excellent transparency and conductivity.

efficiency, which introduce the change of Jsc. We need to control the film thickness uniformity in approximately ±5% in order to optimize Jsc.

On the other hand, the color uniformity is primarily important for application of solar cells to rooftop. The shift of the low reflectance window corresponds to the change in the color of the solar cells, which can be controlled by the ITO film thickness. We need to achieve a film thickness uniformity of at least ±3% in the large-area to obtain uniform colored solar cells.

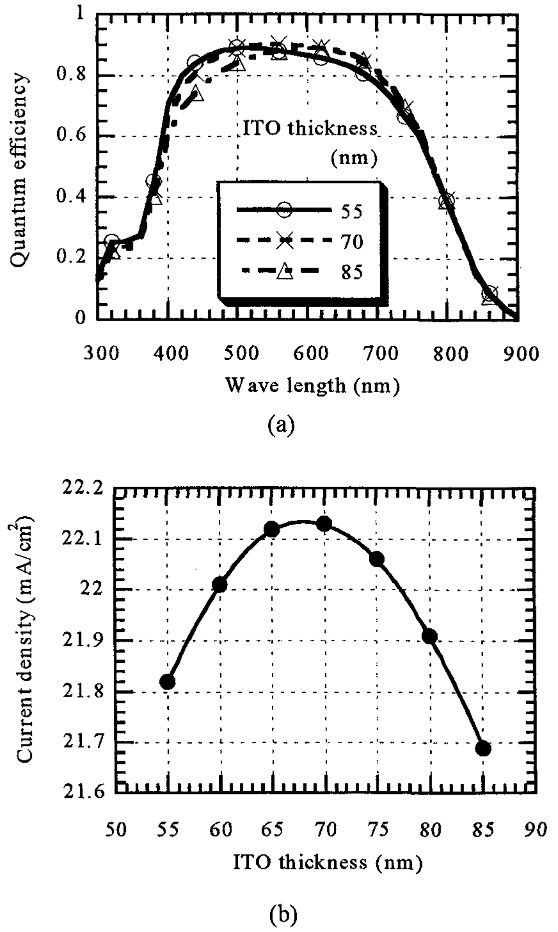

Fig.4 Calculated quantum efficiency and short circuit current density (Jsc) depending on ITO film thickness. ITO film thickness uniformity should be controlled in ±5% from a point of Jsc control, and in ±3% from a point of appearing color control.

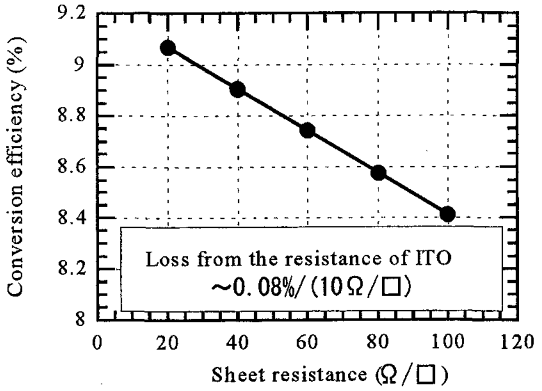

Fig.5 Calculated solar cell conversion efficiency as a function of ITO sheet resistance.

As a transparent conductive electrode, low resistivity is also required as well as transparency and anti-reflecting effect. SCAF cell efficiency is affected by ITO sheet resistance as shown in Fig. 5. Our target of the ITO resistivity is $2\times10^{-4}\Omega\cdot$cm when the ITO film thickness is 70nm.

3. LARGE-AREA ITO FILM DEPOSITION

ITO films were deposited by two types of magnetron sputtering chambers for comparison in this study. One is the large-area scanning magnet type magnetron sputtering chamber which is put in the stepping-roll apparatus shown in Fig. 1[3]. The other is the conventional fixed magnet type magnetron sputtering chamber. The scanning magnet type apparatus is superior to the conventional fixed magnet type apparatus from an aspect of large-area uniform film deposition. Both sputtering apparatus have a DC power source for generating plasma because DC magnetron sputtering achieves much higher deposition rate such as 50nm/min than RF magnetron sputtering. The film thickness is set at approximately 70nm in order to obtain the highest anti-reflecting effect. The detailed sputtering conditions are described in Table I.

ITO film thickness is measured by reflectometry and stylus method. Sheet resistance of the films was measured by a conventional four probe method.

Table I Sputtering conditions

Pressure (Pa)	0.25 ~0.4
Temperature(℃)	200~250
SnO_2 content(wt%)	1~10
O_2 content(%)	0~2
Thickness (nm)	70

3.1 FILM THICKNESS UNIFORMITY

We used the large-scale magnet scanning type sputtering chamber in the stepping-roll apparatus with a technique of controlling the magnet scanning speed to obtain uniform large-area ITO films. Figure 6 shows the film thickness uniformity in the direction of magnet scan. We obtained a good film thickness uniformity of ±1.5%, which is quite difficult to achieve using the conventional fixed magnet type apparatus.

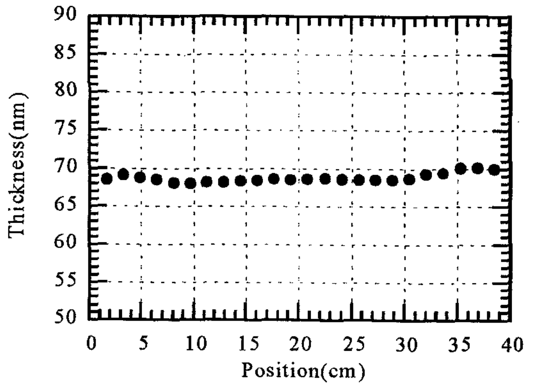

Fig.6 ITO film thickness uniformity in the direction of magnet scan. ITO film is deposited by the scanning type magnetron sputtering chamber in the are of 400mm by 800mm.

3.2 ELECTRIC AND OPTICAL PROPERTIES

We optimized the target composition, oxygen pressure in sputtering gas using the two types of sputtering chambers, and found that optimum deposition conditions are different between the two.

Figure 7 shows sheet resistance depending on oxygen concentration in the sputtering gas. Each type of deposition chamber has a target with the optimum SnO_2 composition (7.5% for scanning magnet type apparatus and 10% for fixed magnet type apparatus). Figure 7 shows that lower resistivity is obtained by the fixed magnet type chamber than by the scanning magnet type chamber.

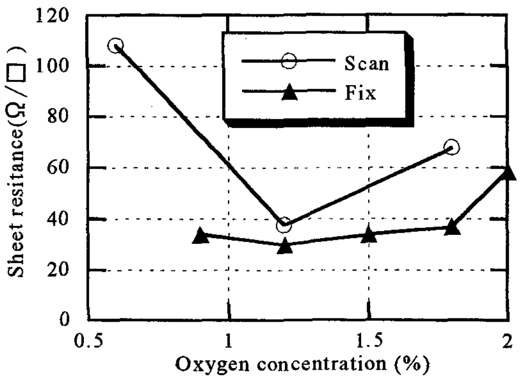

Fig.7 Sheet resistance dependence of fixed and scanning magnet apparatus

In order to investigate the difference between the scanning magnet type chamber and the fixed magnet type chamber, we deposited ITO films with and without scanning the magnet using the same apparatus. Figure 8 shows the sheet resistance distribution in the film around the erosion without scanning the magnet. Figure 8 clearly shows that the electric property of the film is different between in and out of the erosion areas.

Figure 9 and 10 shows resistivity and transmittance as a function of oxygen concentration in the sputtering gas. As shown in Fig. 9, resistivity have a minimum value with the oxygen concentration of 1.2% in the case of scanning magnet type apparatus, whereas 0% and 1.2% are the optimum oxygen concentrations in and out of the erosion areas, respectively, in the case of fixed magnet type apparatus. These results suggest that

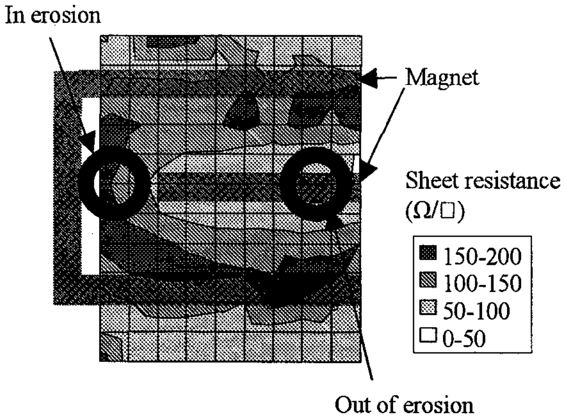

Fig.8 Sheet resistance distribution.

oxygen concentration in the films deposited at inside and outside of the erosion is different. More oxygen may be introduced by the plasma, which is confirmed by Fig. 9 and 10. Although it is generally said that ITO films deposited at the erosion are degraded by oxygen ion[4], ITO films deposited inside and outside of the erosion area show comparable low resistivity, which suggests that ITO films are scarcely degraded by the ion incidence. Sheet resistance of 70nm thick ITO films deposited by the scanning magnet type sputtering is 37Ω/□, while sheet resistance of ITO films deposited by the fixed magnet type sputtering in and out of erosion areas are 34Ω/□ and 29Ω/□, respectively.

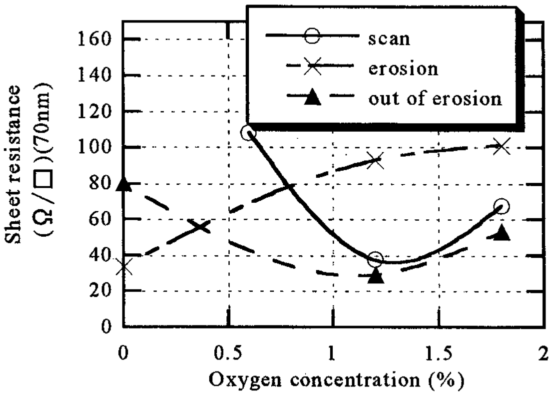

Fig.9 Sheet resistance as a function of oxygen concentration in the sputtering gas.

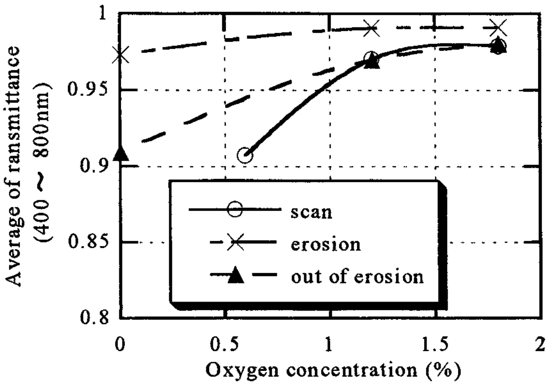

Fig. 10 Average transmittance between 400nm and 800nm as a function of oxygen concentration in the sputtering gas.

The large-area ITO films deposited by the scanning magnet type sputtering apparatus may have accumulated structure of layers deposited in and out of the erosion areas as shown in Fig. 11. As mentioned above, the optimum deposition conditions are different between in and out of the erosion areas, which may make the resistivity of the ITO films deposited by the scanning magnet type sputtering slightly higher than that of the films deposited by the conventional fixed magnet type sputtering apparatus. In the scanning magnet type sputtering, both deposition conditions in and out of the erosion areas are not satisfied at the same time. The scanning magnet type sputtering apparatus achieves excellent film thickness uniformity in the large-area at the sacrifice of optoelectric properties.

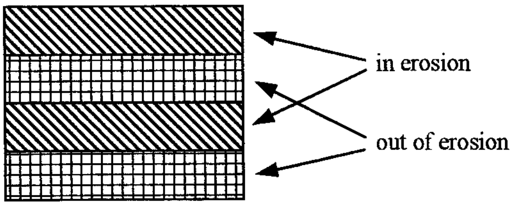

Fig. 11 ITO film structure

4. CONCLUSION

We applied the scanning magnet sputtering apparatus to the deposition of large-area ITO films for flexible film type amorphous solar cells. The method enabled us to deposit ITO film with a good uniformity of ±1.5% in the area of 400mm by 800mm. ITO films deposited by the scanning magnet sputtering apparatus has slightly higher resistivity than that deposited by the conventional fixed magnet sputtering apparatus. ITO films deposited by the scanning method has accumulated structure of layers deposited in and out of the erosion area. Although the optimum deposition conditions are different between in and out of the erosion, both conditions are not satisfied at the same time. The scanning magnet type sputtering apparatus achieves excellent film thickness uniformity in the large-area at the sacrifice of optoelectric properties. The optimized lowest resistivity of an ITO film is $2.4\times10^{-4}\Omega\cdot\text{cm}$.

5. ACKNOWLEDGEMENTS

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