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Development of Indium Tin Oxide by Pulsed RF Sputtering Method for Solar Cell Application

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Abstract: The present study has deposited Indium Tin Oxide (ITO) thin films by pulsed RF sputtering technique at low temperature for the application in solar cell. In stead of using RF magnetron sputtering we have used pulsed RF sputtering to deposit ITO thin film. We have been able to deposit about 90 nm thin ITO films with both low resistivity and high transmittance at low substrate temperature (100°C). The effect of Oxygen (O_2) admixture to sputtering gas (Argon) and different pulse modes on the electrical and optical properties of ITO thin films was investigated. We have used different pulse modes to see their effect on material properties. It is found that the increase in O_2 admixture percentage to Ar decreases the growth rate. At certain oxygen percentage film shows low resistivity and high transparency. A minimum value of resistivity (2×10^{-5} Ω cm) and an average transmittance of ~80% in UV-VIS range have been found at 1% of O_2 admixture. During deposition we also monitored plasma by Optical Emission Spectroscopy (OES). By using OES we have studied the emission lines arising due to Indium at 451 nm and Argon neutral at 811.5 nm. Variation of integral peak intensities were studied for different deposition conditions and correlated with material properties.

Key words: ITO, pulsed RF, sputtering, thin film

INTRODUCTION

In amorphous silicon/crystalline silicon Heterojunction (HJ) solar cells, ITO film is frequently used as front contact and anti-reflection coating (Pla *et al*., 2003). For the application in HJ solar cell, many properties including electrical conductivity, optical transparency, depth of film to minimize sheet resistance and reduce optical losses of ITO thin film must be considered. As the sheet resistance of very thin emitter layer of HJ solar cells is very high, ITO contact is necessary to improve the carrier collection in the photovoltaic device. The electrical and optical properties of ITO films strongly depend on process parameters. Transparency and conductivity of this highly degenerate and wide bandgap oxide semiconductor film can be modified by selecting proper deposition conditions. For the improvement of solar cell performance it is necessary to deposit highly transparent and conductive ITO films. Various techniques, such as electron beam evaporation (Rauf, 1996), ion beam assisted deposition (Meng *et al.,* 2008), pulsed laser ablation (Kim *et al.,* 2005), ion implantation (Sawada and Higuchi, 1998) and DC/RF magnetron sputtering (Lee and Park, 2004; El-Akkad *et al*., 2000; Kurdesau *et al*., 2006), are used for the growth of ITO thin films. The main advantage of pulsed RF sputtering technique is that by controlling energy of ions impinging on substrates, it is possible to improve the properties of sputtered thin films. Properly selected pulse rate has greater control over plasma behavior and can eliminate suspended macroscopic particles as may appear in continuous plasma. But pulsed RF sputtering is still not well tested for the deposition of thin ITO film at low temperature. In our present study we successfully deposited about 90 nm ITO thin films with low resistivity and high visible light transparency at low temperature (100°C) by pulsed RF sputtering technique.

MATERIALS AND METHODS

Thin ITO film was deposited by using a pulsed RF (13.56 MHz) sputtering system (ANELVA). During the deposition we have maintained constant target to substrate distance (8 cm). An indium tin alloy (95:5) of 99.99% purity was used as target. Thin films were deposited simultaneously on both intrinsic c-Si (polished) and corning glass substrate. Before the deposition, substrates were cleaned ultrasonically by acetone and alcohol accordingly followed by rinsing with de-ionized water, and then dried with dry N_2 gas. The deposition chamber was evacuated to 1×10^{-6} Torr (base pressure) and the required sputtering pressure inside the chamber was achieved by the introduction of argon gas through mass flow controller. Once the pressure was achieved, the sputtering was carried out in argon and oxygen atmosphere. Oxygen was used as admixture to the sputtering gas and the percentage was varied from 0 to

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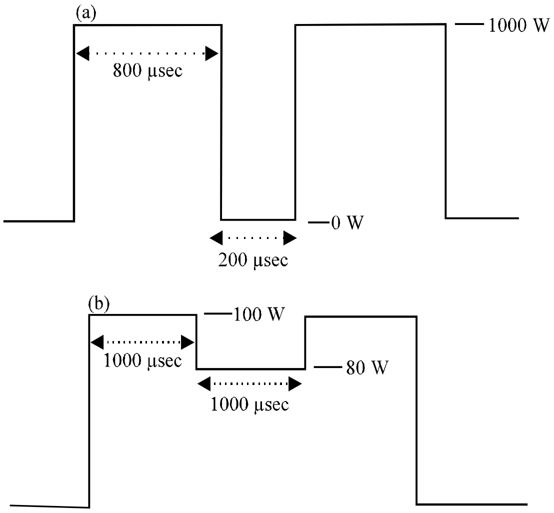

Fig. 1(a-b): Two types of pulse modes used in this study, (a) On-off, (b) High-low

6%. To remove the oxide layer formed on the In-Sn surface, the target was pre-sputtered in argon atmosphere for 10 min with a shutter on the substrate. The oxygen was introduced again after pre-sputtering and shutter was removed for the deposition of the film on the substrate. Films were deposited in two pulse modes: (I) on-off , Fig. 1a (2) high-low, Fig. 1b.

Thickness and refractive index was measured by an Ellipsometer (Five lab MARY-102) and sheet resistance was measured by four-point probe method. Sheet resistances were converted to resistivity using the measured thickness value. Optical transmission and reflectance were recorded by Hitachi U-4000 UV/Vis spectrophotometer. An optical emission spectroscopy system is installed on the multi-source sputtering chamber. The optical emission system consists of a ¼ m spectrograph coupled with a 1024 element photo diode array, a detector, fiber optics. Light for OES analysis is collected from the plasma by fiber optics. Three vacuum-compatible fiber optic cables are located inside the deposition chamber. The OES system was connected with a personal computer for scanning and data acquisitions. Each spectrum was obtained by averaging 10 exposures of 10 sec each.

RESULTS AND DISCUSSION

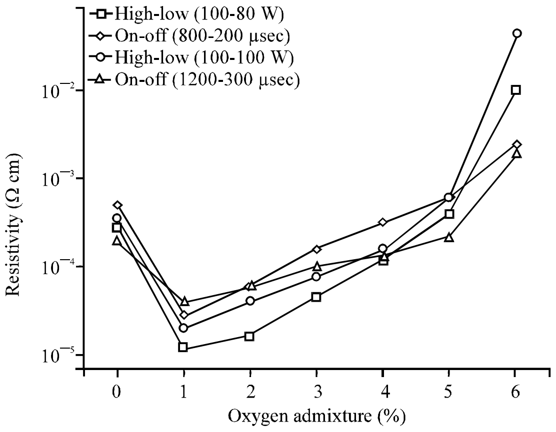

Fig. 2: Variation of deposition rate with oxygen admixture for different modes

Table 1: Refractive indices for different pulse modes

	Refractive index			
Oxygen (%)	On-off (800-200 μsec)	High-low (100-80 W)	On-off (1200-300 μsec)	High-low (100-100 W)
0	2.078	2.079	2.077	2.080
1	2.084	2.082	2.091	2.090
2	2.083	2.091	2.092	2.085
3	2.090	2.097	2.088	2.092
4	2.088	2.092	2.084	2.080
5	2.091	2.092	2.088	2.085
6	2.080	2.088	2.090	2.092

Here we have analyzed the effect of oxygen admixture and different pulse modes on electrical and optical properties o f ITO thin films. Few of the results are presented and discussed below. Here various percentage of oxygen (from 0 to 6%) was used to the sputtering gas argon. For all the depositions working gas pressure maintained at 5 m Torr and substrate temperature 100°C. Two types of pulses are used for on-off and high-low mode. For on-off mode pulse on time duration (t_{ON}) is 800, 1200 and off time is 200, 300 μsec, respectively. All the depositions in this mode have been carried out at 100 W power. For high-low mode two types of pulses with 100-80 W and 100-100 W are used. Deposition time was adjusted so that the thicknesses of the deposited films are about 90 nm. Figure 2 shows the influence of oxygen admixture on deposition rate. We see that for pure argon gas deposition rate is low but it becomes high when little amount of oxygen is added to it. Interesting thing is that for all the cases deposition rate decreases with the increase in percentage of oxygen. Also it is obvious from Fig. 2 that the deposition rate is less for on-off mode in comparison to high-low mode. Decrease in deposition rates with the increase in oxygen percentage was attributed to the oxidation of target surface (Buchanan *et al.*, 1981), to the possibility of resputtering of the film by energetic particles in plasma and to the compensation of deviated stoichiometry by the reacting gas (Mehra and Rhodesm, 1986). Measured refractive indices are shown in Table 1. It is seen that

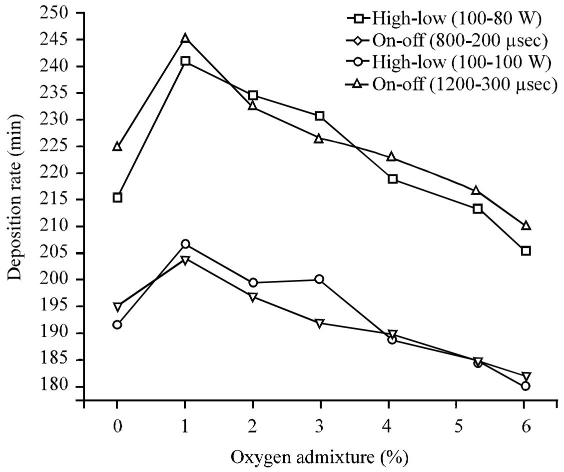

Fig. 3: Effect of oxygen admixture on resistivity for different modes

there is no general trend in the change but for most of the samples refractive index lie within the range 2.075 and 2.095.

Figure 3 shows the influence on the resistivity of the grown film due to oxygen admixture at a fixed pressure and substrate temperature for different pulse modes. We see that the material deposited without oxygen doesn't give the lowest resistivity. But if we add little amount of oxygen to argon, resistivity decreases for all the cases.

For our present range of deposition conditions lowest resistivity is around 2×10^{-5} Ω cm and it is deposited in High-low mode with 1% of oxygen added with argon. This value is very good in comparison to the ITO deposited by other methods. This is probably due to the fact that in our study we have properly selected the pulse rate of time-modulated plasma. In plasma deposition sometimes suspended macroscopic particles deteriorates film property. From Fig. 3 one can also observe that for all the cases minimum resistivity is found at the admixture of 1 or 2% of oxygen. Resistivity again increases with the increase of oxygen percentage. Minimum of the resistivity at a certain oxygen percentage caused by the fact that an exact amount of reactive oxygen needed in order to oxidize the metal atoms arriving at the substrate to a slightly substoichiometric film (oxygen-to-metal ratio approx. 1.5). Smaller amount of oxygen lead to opaque films due to un-oxidize metal clusters, larger amount of oxygen cause fully oxidized films, with high resistivity (Mientus and Ellmer, 2001). Also it is cleared that, the variation in the resistivity of ITO thin film is related to the variation in carrier concentration of the film, which leads to the variation of oxygen vacancies possibility of incorporation of oxygen atoms into the growing ITO thin films (Kim *et al*., 2003).

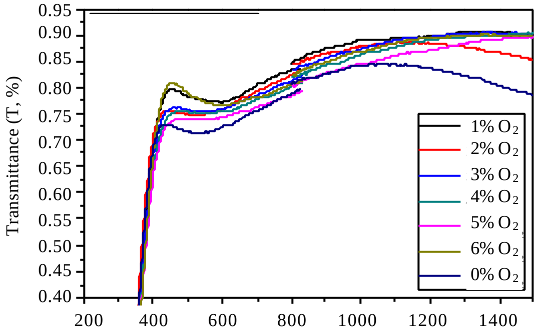

Fig. 4: Transmittance spectra of films prepared at different oxygen percentage for high-low mode (100-80 W)

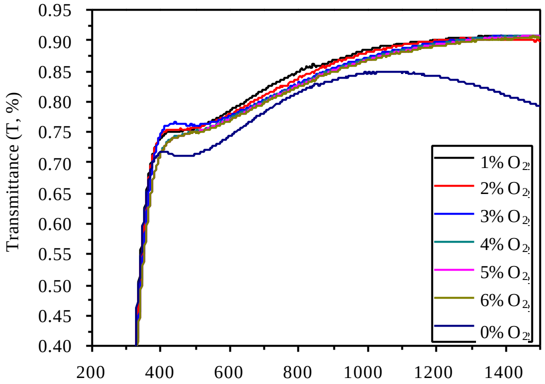

Fig. 5: Transmittance spectra of films at different oxygen percentage for on-off mode (800-200 μsec)

Figure 4 and 5 show the optical transmittances of the ITO films grown with different oxygen admixtures. Among four sets of deposited samples we have shown two sets of results -one is for on-off mode and another is for high-low mode. Because for both the modes two sets of samples show similar trend in transmittance curve with the change in oxygen percentage. From Fig. 4 and 5 we can see that the films deposited with 1% of oxygen show highest transmittance. Although the difference of transmittances among films deposited with different oxygen percentage are not very high, still we can identify that the film deposited at 1% of oxygen gives highest transmittance. Normally there is a trade off between resistivity and transmittance of the films deposited by DC or RF sputtering. But in this case we get highest transmittance for the lowest resistivity samples. Also we notice that the transmittance increases with the introduction of oxygen with argon. The *in situ* plasma diagnostics was done by

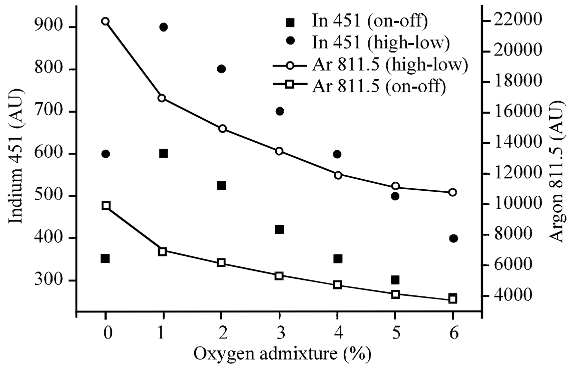

Fig. 6: Variation of spectral line intensities of indium (451 nm) and argon (811.5 nm) with oxygen admixture of different modes

recording the optical emission spectra. Among different lines we have chosen Indium line at 451 nm and Argon line at 811.5 nm. Figure 6 shows variation of integral peak intensity of Indium line (In 451) and Argon line (Ar 811.5) with the change in oxygen percentage. From this figure we can observe that with the introduction of oxygen, In 451 increases and then it decreases with the increase of oxygen percentage. This behavior is same for both on-off and High-low mode. Now if we compare the variation of In 451 with deposition rate (Fig. 2) we see that these two variations show similar trend. This demonstrates that the variation of deposition rate is proportional to the variation of In 451. Intensity of Ar 811.5 continuously decreases with the increase of oxygen admixture. This decrease of intensity is due to the decrease of argon partial pressure. We don't find any correlation between Ar 811.5 and material properties.

CONCLUSION

In this study we have used pulsed RF sputtering at low temperature to deposit ITO thin films of about 90 nm thin for the application in solar cell. Sometimes there is a trade off between electrical and optical properties of ITO film. In our present study we have tried to optimize the electrical and optical properties of the film deposited by varying the deposition conditions at low substrate temperature. It is obvious from the results that good quality ITO films can be deposited by pulsed RF sputtering method. The film quality can be improved further by widening the range of deposition conditions. By observing the optical emission spectra of the plasma, few of the film properties may be predicted.

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