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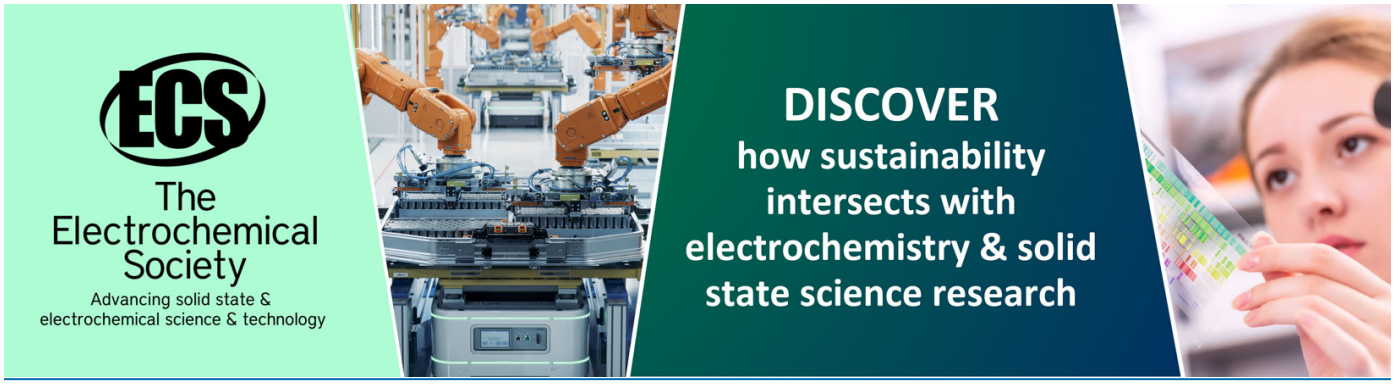

Optimization of Al-doped ZnO films by RF magnetron sputtering at room temperature for Cu (In, Ga) Se_2 solar cells

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Abstract. In this work, Al-doped zinc oxide (AZO) thin films were deposited by RF magnetron sputtering with various RF power at room temperature. The effect of RF power on the structural, electrical, and optical properties of AZO thin films were investigated by XRD, SEM, UV–Vis–NIR spectroscopy and Hall measurements. The lowest resistivity of 1.8×10^{-3} Ω·cm was obtained at the highest RF power of 450 W. The average optical transmittance is about 90% in the visible range and above 80% in the range of 300-2000 nm. CIGS thin-film solar cells were prepared using the AZO films as the windows layer and an efficiency of 15.36% in CIGS solar cell has been achieved.

1. Introduction

Transparent conductive oxides (TCOs) have received wide attention due to high transparency and low resistivity, which are applied widely in various optoelectronic devices, such as photovoltaic devices, flat-panel displays, and light-emitting diodes [1-6]. At present, Compared with the ITO, AZO is a promising material because of its low cost, high transmittance, and low resistant. To improve the optoelectrical properties of AZO, a thermal process commonly performed such as high substrate temperature in the deposited process or annealing after deposited [7-10]. As previous research, Mosbah et al. reported the resistivity decreases from 5×10^{-4} to 3×10^{-5} Ω·cm with the deposition temperature, the transmittance also decreases with the deposition temperature [11]. Wang et al. reported that the optoelectrical properties of AZO films are improved by annealing in range RT to 200°C [12]. Kim et al. investigated the effect of post-annealing time on the properties of sputtered AZO films at 300 °C. The results showed that the improvement of electrical properties with elongate in the post-annealing time [13]. Zhu et al. reported that the optoelectrical properties of AZO films were improved involving H_2 in the sputtering ambient at RT and 100 °C [14]. That limited the application in flexible devices and temperature sensitive devices, such as flexible devices, CIGS solar cells and polymer substrates [15]. For the CIGS solar cells, the PN junction of solar cells consisted of CIGS/CdS/i-ZnO/AZO, which is crucial for the performance of the solar cells, is sensitive to the temperature [16]. The performances of the CIGS solar cells will deteriorate if the annealing temperature above 150, there is necessary to optimize the properties of AZO deposited at room temperature.

AZO films have been prepared by the various method including sol-gel coating[17], spray pyrolysis[18-19],evaporation[20],chemical vapor deposition[21], atomic layer deposition[22], pulsed laser deposition [23],electrospinning[24], DC magnetron sputtering[25], and RF magnetron

sputtering[26]. Among those techniques, RF magnetron sputtering is considered to be suitable for large-scale industrial production due to high reproducibility, high uniformity, and high deposition rate [27].

In the present work, AZO thin films were prepared using the RF magnetron sputtering at room temperature with various RF power. The influence of RF power on structural, morphological, optical and electrical properties of the AZO thin films is investigated

2. Experimental

All AZO thin films were prepared on soda-lime glass (SLG) substrates and Mo-coated SLG at room temperature in a magnetron sputtering system as described detaily in our previous work [28]. Pure argon (Ar) gas flux of 30 sccm was used as the sputtering gas. The working pressure was kept constant at 0.1 Pa. The sputtering RF power was adjusted 250 to 450W with a step 50W. The film thickness was controlled at approximately 350 nm by adjusting the sputtering time. Solar cells were fabricated with the structure of SLG/Mo/CIGS /CdS/i-ZnO/AZO/Al. CdS layers with a thickness of about 50 nm were deposited by chemical bath deposition at 70 °C. The i-ZnO and AZO layers by RF magnetron sputtering were deposited in sequence. The Al contacts were finally formed by electron beam evaporation with a mask. The structures, morphologies, electrical, and optical properties of AZO films were investigated by Bruker D8 Advance, FEI NOVA NANOSEM 450, Hall measurement with Van der Pauw method, and Hitachi U-4100, respectively. The performance of solar cells was recorded under the standard test conditions (AM 1.5G, 100 mW/cm^2, and 25 °C).

3. Results and Discussion

Fig.1 shows the X-ray diffraction spectrum of ZnO films deposited on Mo-coated SLG at 250, 300, 350, 350, 400 and 450W. It is observed that the XRD pattern (excepted defined Mo peak) has four well define peak at 33.95°, 35.83°, 47.04°, and 62.34°, corresponding to (002), (101), (102) and (103) respectively. These are good agreement with standard JCPDS card #51-0037 of the hexagonal wurtzite structure of AZO films. All the samples exhibited both (002) preferential orientations. The intensities of (002) diffraction peaks got stronger with the increasing RF power from250 to 450 W, indicating a preferred polycrystal orientation along the [001] direction with the c-axis perpendicular to substrate surface [29].

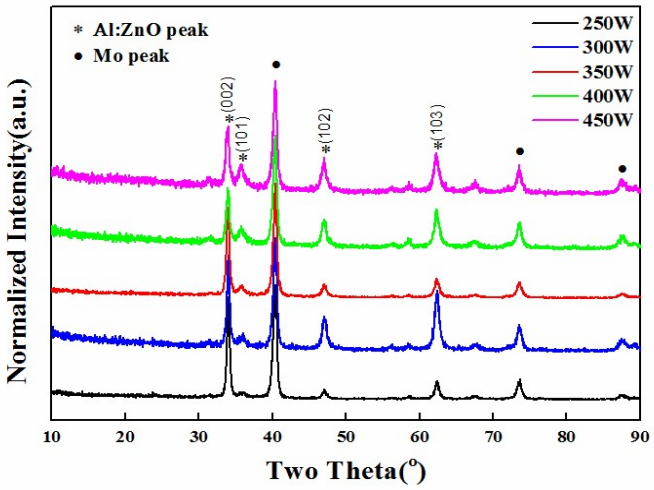

Figure 1. X-ray diffraction patterns of IZO films deposited with various RF power

Fig.2 shows the surface morphology SEM images of the AZO films deposited on SLG by different RF power. Clusters of grains exist in the sample deposited at 250 W, suggesting this film is rough and inhomogeneous and some small pinholes exist, as shown in Fig.2a. Increasing the RF power to 300 W, the film surface shows a more significant and regular grain increase (Fig.2b). When the RF power

increased to 350 W, the film is composed of large and the average grain size is about 2 μm, as revealed in Fig. 2c. As the RF increased to 400 and 450 W, It was observed the grains size becomes smaller and the surface microstructure becomes relatively smooth.

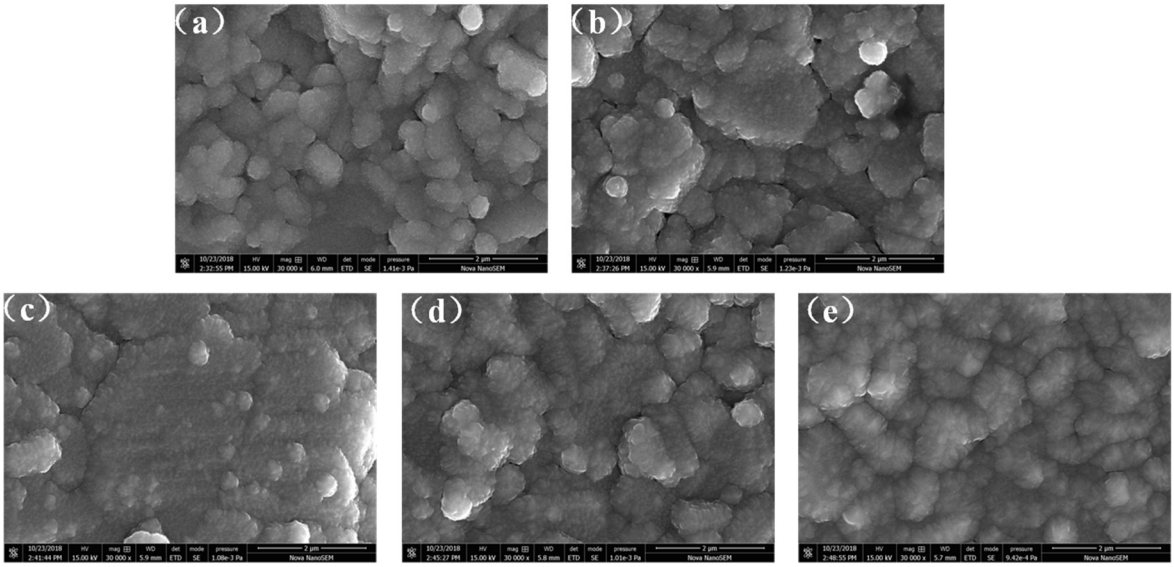

Figure 2. SEM morphology images of AZO films deposited with various RF power, (a) 250W, (b) 300W, (c) 350W, (d) 400W, (e) 450W

The transmittance spectra of AZO films deposited with various RF power are shown in Fig. 3. the average transparency of all the films was around 90% within the visible region of the spectra and the oscillatory character of the curves is due to interference effects. With increasing sputtering power, the average transparency sharply decreases in the near-infrared (NIR) spectral region. In general, the transmittance of AZO films decreased with the RF power due to the increase in reflectance. It can be explained to the plasma resonance of electron gas in the conduction band [3, 30].

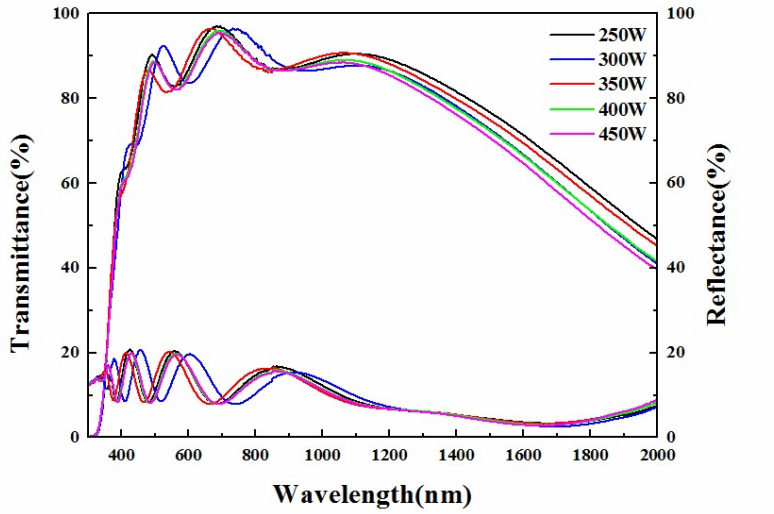

Figure 3. Transmission spectra of AZO films deposited with various RF power

The electrical properties of AZO films deposited at various RF power were analyzed via the Hall effect measurement employed by the van der Pauw method. The resistivity (ρ), mobility (μ), and carrier concentration (n) of the prepared AZO films are presented in Fig. 5. The ρ decreases with increasing RF power can be ascribed to the increase in the carrier concentration caused by interstitial Al ion, the lowest resistivity of 1.8×10^{-3} Ω·cm was obtained at the highest RF power of 450 W. Highest mobility value of 10.05 cm^2/Vs is observed for the AZO deposited at 350 W. The similar trend was reported by Vidhya et al. and Kim et al. [31, 32].

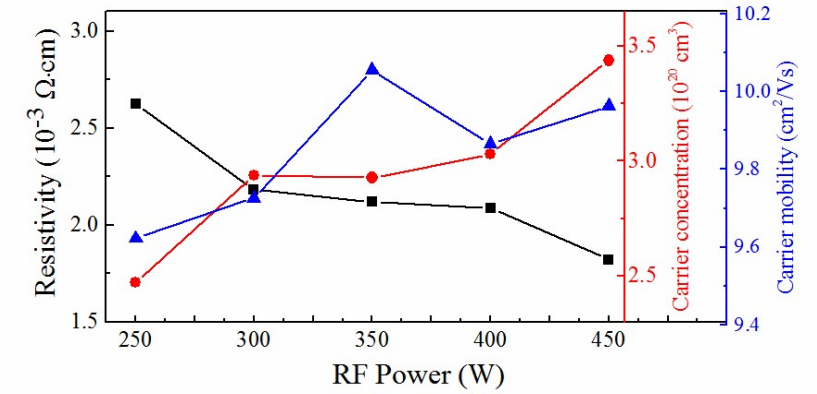

Figure 4. Resistivity, carrier concentration and mobility of AZO films deposited as a function of RF power.

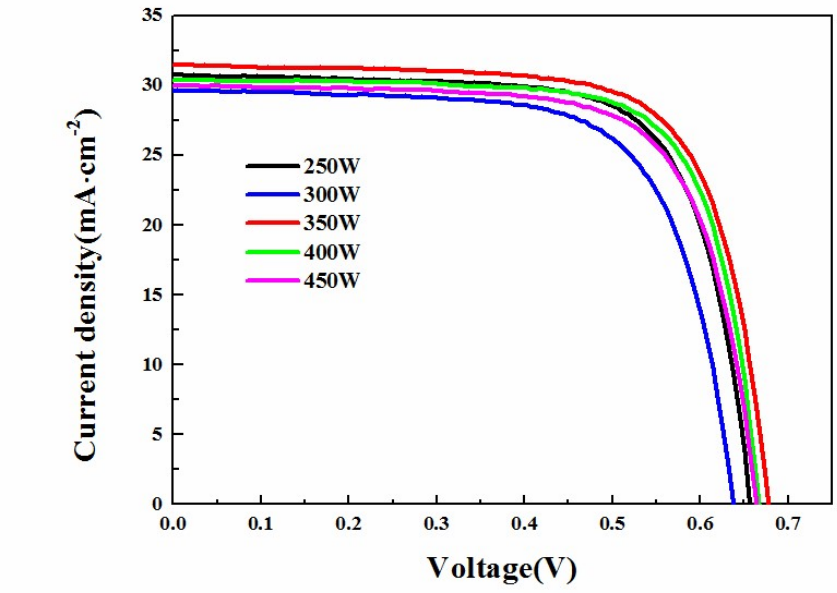

Figure 5. The J–V curve parameters of CIGS solar cells with various RF power

Table 1. The device parameters of CIGS solar cells based on different RF power. Diode parameters determined from current-voltage curves. J_0 is the reverse saturation current density and A the ideality factor.

Sputtering power	Voc(mV)	Jsc(mA·cm^{-2})	FF(%)	η(%)	G(mS·cm^{-2})	Rs(Ω·cm^{-2})	A	j_0(mA·cm^{-2})
250W	656	30.74	71.89	14.05	2.11×10^{-5}	16.27	1.41	4.70×10^{-9}
300W	632	29.63	69.96	13.10	4.30×10^{-5}	3.31	1.51	3.70×10^{-9}
350W	674	31.46	72.39	15.36	1.45×10^{-5}	13.70	1.23	1.78×10^{-9}
400W	662	30.38	74.05	14.90	9.89×10^{-6}	43.82	1.73	8.59×10^{-9}
450W	662	29.95	71.62	14.21	1.93×10^{-5}	45.76	1.79	1.77×10^{-8}

Solar devices with the structure of glass/Mo/CIGS absorber/CdS/i-ZnO/AZO/Al grid were fabricated to study the influence of the sputtering power on the CIGS solar cells. Fig. 5 shows the current density–voltage (J-V) curve for individual best CIGS solar cells with AZO window layers deposited at different sputtering power. The parameters derived from each J-V curve were listed in Table 1. As expected from the results of the optical measurements of the AZO films, Jsc decreases with increasing RF power. This is the agreement with the optical properties trend with the RF power. The highest efficiency of 15.36% has been achieved for CIGS solar cells with AZO films, in which the V_{OC}, $J_{SC,}$ and FF are 674 mV, 31.46 mA/cm^2 and FF= 72.39%, respectively.

4. Conclusion

The AZO thin films were prepared by magnetron sputtering with different RF power on SLG and on CIGS solar cells at room temperature. From the XRD examination, the structure of all AZO films was preferred (002) preferential orientations. Hall effect measurements show that the optimal hall mobility of 10.05 cm^2/Vs and resistivity of 2.12×10^{-3} Ω•cm were obtained at the power of 350 W. Furthermore, with the increasing RF power the transmittance of AZO thin films decreased. The results demonstrated that it is key to balance the optical and electrical properties as AZO films applied in photovoltaic devices.

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