

Effect of Annealing Temperature on Photovoltaic Parameters of Al doped ZnO Thin Film Solar Cell

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Abstract. Al doped ZnO nanoparticles were prepared by mechanochemical milling process. Al doped ZnO (AZO) films were deposited onto p-Si substrates using spin coating technique. X-ray diffraction (XRD) technique was employed to examine the crystal structure and phase identification of AZO thin film at different annealing temperatures. Microstructural properties of AZO films were observed by Scanning Electron Microscopy (SEM). UV-Vis spectrometer was used to study the optical properties of AZO film at different annealing temperatures. Illuminated I-V characteristics were measured under halogen lamp. From I-V characteristics curves, convection efficiency and fill factor were determined at different annealing temperatures. From results obtained, it meets the special requirements for the development of cost effective thin film solar cell.

Keywords: Mechanochemical milling, optical band gap, photovoltaic parameters, SEM, spincoating ,XRD
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INTRODUCTION

Semiconductor compounds have drawn much attention during the last few years because of their novel optical and transport properties which have great potential for many optoelectronic applications. Zinc oxide (ZnO) is an inexpensive, n type semiconductor of wurtzite structure with a direct energy wide band gap of 3.2-3.3 eV at room temperature, and optical transparency in the visible range. Today, indium tin oxide (ITO) thin films are the most widely used material as a transparent electrode in various optoelectronic devices such as solar cells, liquid crystal displays (LCDs) organic light – emitting diodes (OLEDs) and other flat panel displays (FPDs) [1,2]. Moreover, most II–VI semiconductors are direct band gap materials, and their properties (such as electrical resistivity, piezoelectricity and structure) can be regulated or controlled by using different dopants [3]. Accordingly the compound has wide applications in chemical sensors, heterojunction solar cells, electrophotography, surface acoustic wave devices, conductive transparent conductors and many others. However, pure ZnO thin films lack stability in terms of thermal edging in air or corrosive environments [4]. Therefore polycrystalline ZnO films have been doped with group II and group III metal ions such as indium (In), aluminum (Al), gallium (Ga), copper (Cu) cadmium (Cd) etc, to enhance their structural, optical and electrical properties [5]. Doping is particularly

done to get high transparency, stability and high conductivity. Aluminum doping is particularly stable for this purpose. Aluminum doped ZnO (AZO) thin films have high transmittance in the visible region, and a low resistivity, and the optical band gap can be controlled by using Al doping amount [6]. And they have got potential applications in solar cells [7]. Different physical and chemical techniques have employed for aluminum incorporation in ZnO. These include dc and rf magnetron sputtering, pulsed laser ablation[8], chemical vapor deposition, chemical beam deposition [9], sol gel [10], electroless technique [11], spray pyrolysis [12] among others. The conventional physical techniques generally produce good quality transparent films. However they are very expensive and are difficult to carry out in the industrial level. Chemical deposition techniques, on the other hand are relatively low cost processes and can be easily scaled up for industrial applications. Since the last two decades, chemical techniques have come out to be a good alternative for material preparation in thin film form [13]. Among various chemical methods employed so far to deposit AZO films, the sol – gel process is one of the versatile methods to prepare thin film–supported nano-sized particles without complicated instruments such as chemical vapor deposition (CVD). It is simple, inexpensive, and has a general advantage of large area deposition and uniformity of the films thickness [14]. In this paper,

temperature dependence on cell performance of AZO thin film cell was observed.

EXPERIMENTAL

Aluminum oxide (Al_2O_3), and zinc oxide (ZnO) were firstly observed by XRD to examine the purification. They were separately ball milled for 5h to get more contact points with other molecules. And then, the mixture of $(1-x)$ ZnO and (x) Al_2O_3 were prepared according to the stoichiometric composition of $X=12$ mol%. After that, the powder was ground by agate mortar for 1h. It was also repeated by ball milling method to reduce the particle size. The time interval was set for 5h. Sample was milled by grinding bowl (volume of 100 g) and grinding balls (20 balls, 5mm diameter) at 300 rpm in air atm. Three stages mesh – sieve was employed to form uniform particle size of $\text{Zn}_{1-x}(\text{Al})_x\text{O}$ powder. Then the fine powder was annealed at 500°C and confirmed by XRD and SEM. After that the AZO powder was mixed with methoxythanol solvent at 150°C by using magnetic-stirrer to become precursor solution.

Next, they were coated onto Si substrate by Single Wafer Spin Processor (MODEL WS-400 BZ 6 NPP/LITE). This machine was set by LCD display with constant rate of 3000 rpm at spin time interval of 30 sec. After fabrication, all fabricated AZO films were annealed at 600°C , 700°C , 800°C for 1h. The films were analyzed by X-ray diffraction (XRD), Scanning Electron Microscopy (SEM) and I-V characteristics were measured.

RESULTS AND DISCUSSION

XRD Analysis

The structure of AZO powder and thin films were characterized by X-ray diffraction (XRD) system. Figure 1 showed the XRD profile of Al (12 mol %) doped ZnO powder. Nine diffracted peaks were formed on observed XRD profile and well matched with those of ZnO standard. Thus Al ion was absolutely occupied by Zn-site of ZnO lattice. The most intense peak was examined to be (101) collection. The XRD spectrum of AZO thin film at different annealing temperatures (600°C , 700°C and 800°C) was given as Figure 2. As the detail analysis of film analysis by XRD, AZO film was successfully formed onto substrate at different annealing temperatures. Table 1 showed some crystallographic parameters of AZO powder and films.

TABLE 1. Some crystallographic information about the Al doped ZnO

Sample	Intensity	FWHM	Crystallite size (nm)
AZO (Powder)	533	0.214	46.26 nm
AZO Film (600°C)	199	0.193	43.28
AZO Film (700°C)	220	0.213	39.21
AZO Film (800°C)	81	0.206	40.26

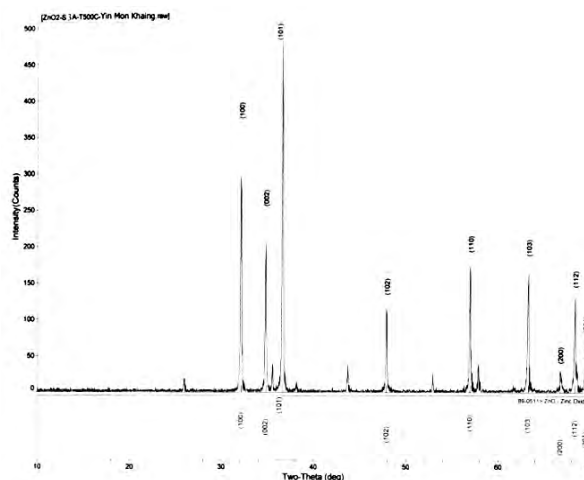


FIGURE 1. XRD profile of Al(12mol%) doped ZnO Powder.

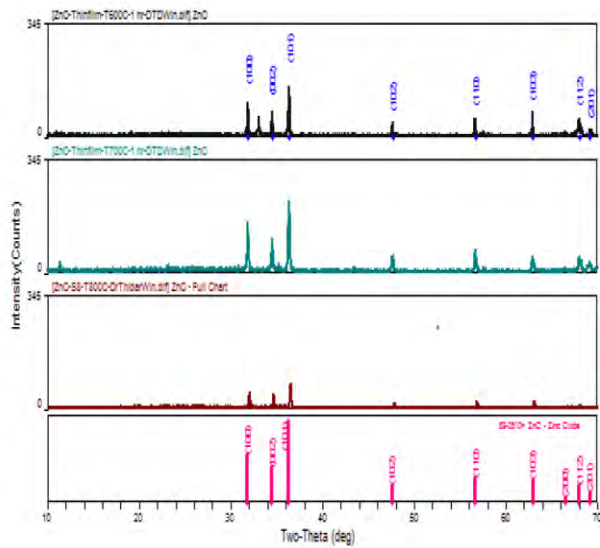


FIGURE 2. The XRD patterns of the AZO films annealed at 600,700 and 800°C.

SEM Analysis

The microstructures of grown samples were investigated by scanning Electron Microscopy. Figure 3 showed SEM image of AZO thin film with magnifications ten thousands. SEM image indicated the normal grain distribution and some were porous structure. The majority grain on SEM image with colourful are Al doped ZnO. The others are impurities. The hole among the grain are porosity. The grain size was measured to be 171 nm at powder AZO. The ratios of grain size(nm) of AZO films are about 160 nm, 208 nm and 250 nm as the temperature amount increases from 600,700 and 800°C, respectively. The modifier Al ion were helpful in increasing and the grain size increase with increase of annealing temperature.

Optical Properties Analysis

Figure 4 indicated the optical transmittance spectrum of AZO film at different annealing temperatures. The wavelength range of 300-800nm was scanned for all films. The maximum wavelengths were found to be 716 nm, 717 nm and 759 nm for AZO film at 600°C, 700°C and 800°C respectively.

The optical band gaps were determined to be 3.11 eV, 3.13 eV and 3.16 eV for respective films. These values were collected and listed in Table 2.

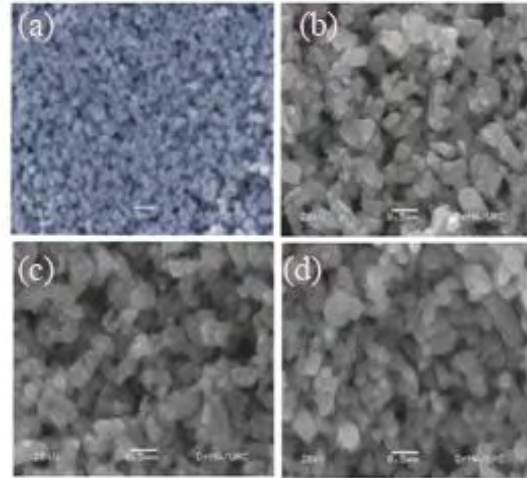


FIGURE 3. SEM images of (a)AZO powder (b)AZO film at 600°C(b)AZO film at700°C(c) AZO film at 800°C.

TABLE 2. UV – Vis analysis of λ_{max} and optical band gap from AZO films at various temperatures .

Sample	λ_{max} (nm)	Optical band gap (eV)
600°C	716	3.11
700°C	717	3.13
800°C	759	3.16

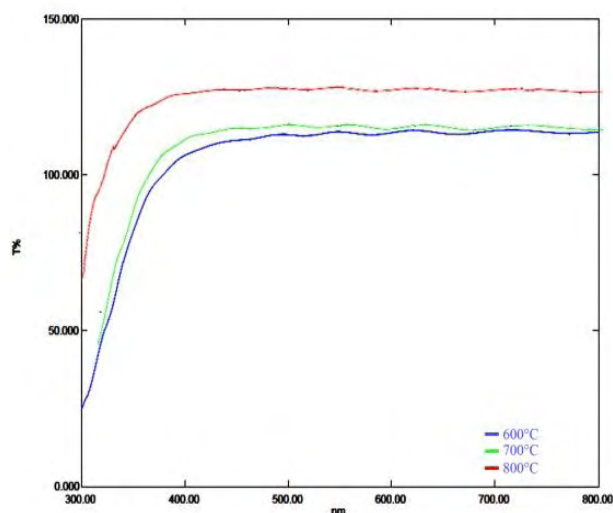


FIGURE 4. Optical transmittance spectrum of AZO films at different annealing temperatures.

I-V Analysis

Dark I-V characteristics were measured and plotted as Figure 5(a). At the forward bias region, the current did not allow to flow before threshold voltage (lead space) without any trend of saturation. The dark I-V curve indicated the leakage current formation during processing. Important diode parameters such as zero-bias barrier height and ideality factor were determined. Figure 5(b) showed the illuminated I-V characteristics of AZO thin film cells. The negative current was formed at positive voltage in fourth quadrant of the circle. By rotating the current axis with 180° , V_{oc} - I_{sc} variation was formed and shown in Figure 5(c). From V_{oc} - I_{sc} characteristics curve, conversion efficiency (η_{con}) and fill factor (FF) were evaluated. Table 3 described the saturation current (I_s), zero-bias barrier height (ϕ_{bo}), ideality factor (η), conversion efficiency (η_{con}) and fill factor (FF) of AZO film at different annealing temperatures.

CONCLUSION

Growth and characteristics of AZO thin film have been studied at different annealing temperatures. As a results of XRD, Al-ion was absolutely occupied by Zn-site of ZnO lattice. From SEM result, it was found that all SEM images were porous structure. As a result of UV-vis spectroscopic measurement, the optical

band gap increased with increasing annealing temperature. The difference in band-gap was caused by the existence of grain boundary with different forms. All V_{oc} - I_{sc} were indicated the solar cell nature of AZO thin film. The largest value of efficiency (6.22%) was formed at 800°C .

TABLE 3. The saturation currents (I_s), Ideality factor (η), zero-bias barriers height (ϕ_{bo}), efficiency (%), fill factor (FF) of AZO thin films with process temperatures

Substrate Temperature ($^\circ\text{C}$)	I_s (mA)	ϕ_{bo}	Ideality Factor (η)	Efficiency (%)	fill factor (FF)
600	0.07	0.34	1.49	1.0	0.32
700	0.1	0.33	1.49	3.74	0.27
800	0.06	0.34	1.51	6.22	0.28

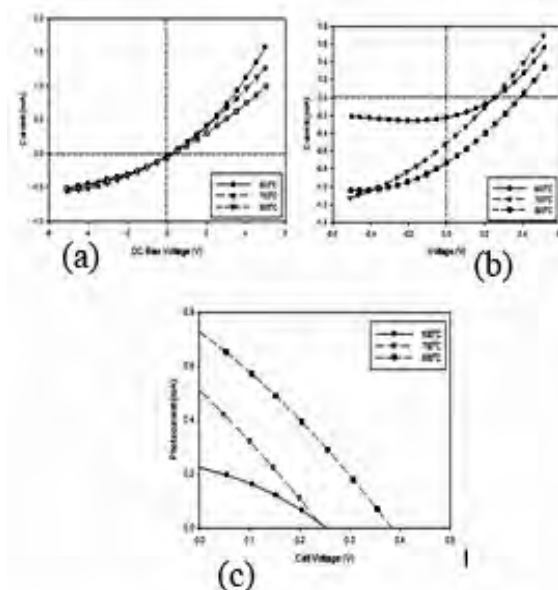


FIGURE 5. (a) Dark I-V characteristics (b) Illuminated I-V characteristics (c) V_{oc} - I_{sc} variation curve of AZO thin film at different annealing temperatures.

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