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The effect of Solution Temperature on electrodeposit-ZnO thin film

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Abstract. Zinc Oxide (ZnO) has been successfully electrodeposited on a fluorine doped tin oxide (FTO) coated glass substrates using a simple aqueous solution containing zinc nitrate hydrate by low temperature galvanostatic electrolysis. The solution temperature of zinc nitrate hydrate was varied from 60°C to 75°C in order to investigate the effect of solution temperature on electrodeposit-ZnO thin film. The properties of ZnO film were investigated by X-ray diffraction (XRD), Field-Emission Scanning electron microscope (FE-SEM) and Atomic force microscopy (AFM). The solution temperature shows a significant effect on structural and morphological of deposit-ZnO. The XRD patterns exhibited the increment of (002)-ZnO peak when the solution temperature increased and the highest peak was observed at 75°C. The morphology of ZnO was changed from planar to nanopillar with the solution temperature. In conclusion, ZnO nanopillar with an excellent structural properties was obtained at solution temperature of 75^{o}C.

Introduction

There are many approaches to find new photovoltaic cells, the ideal cell being inexpensive, easy, and environmentally safe to produce with high efficiency and long lifetime. The highest efficiency measured is around 25% for single-crystalline-Si. Thin film chalcogenide cells reach 15-20% efficiency, while dye-sensitized solar cells only reach about 11% efficiency. Multi junction and tandem cells have reached efficiencies higher than 30%[1].

The photovoltaic devices composed on cuprous oxide (Cu_2O) and zinc oxide (ZnO) materials have received broaden attention as a candidate of next generation thin film solar cell due to several advantages such as these materials are abundance and non-toxic. Moreover, the coefficient efficiency of Cu_2O is much lower than single-crystalline Si[2]. In addition, the photovoltaic devices composed of Cu_2O as an absorbing layer have theoretical conversion efficiency of 18 %[3].

The highest conversion efficiency of Cu_2O/ZnO heterojunction, however, was limited at 2.0 % for the MgF_2/ITO/ZnO/Cu_2O heterostructure solar cell reported by A. Mittiga et. al. in 2006 [4]. The Cu_2O layer has been prepared by oxidizing copper sheet using two steps annealing; high temperature (>1100 ^{o}C) and following by 800 ^{o}C. The second highest conversion efficiency is randomly oriented Cu_2O/ZnO heterojunction deposited on FTO glass substrate prepared by electrodeposition method, previously reported in 2007 by M. Izaki et. al. [5], which is twice than that prepared by sputtering method [6]. Increase the heterointerface between the ZnO and Cu_2O is one of the approaches to enhance the conversion efficiency of solar cell, which offer effective charge collection along the thickness direction as well as excellent charge transporting property.

In this work, the ZnO thin film was fabricated using low-temperature electrodeposition technique without any post-heating. The electrodeposition process in aqueous solutions is a well-known technique due to several advantages over thermal oxidation and gas phase deposition technique such as low-fabrication cost, low temperature and ambient pressure processing, controllable film thickness and possible large scale deposition [7-11]. The effect of the solution temperature was investigated in order to obtain a larger surface area for heterojunction formation.

Experimental

The ZnO layer was deposited on FTO coated glass substrate using electrodeposition method. There are three main stages in this project in order to achieve the objective.

A. Phase 1: Substrate Preparation

The FTO glass substrate was cut approximately to 2.5 cm x 1.0 cm. The deposition area is 1.0 cm^2. The FTO-coated glass substrate immersed into acetone about 5 minutes and cleaned using ultrasonic cleaner. After that, the substrate was rinsed using distilled water. Finally, it was polarized in 1M NaOH at current density of +10mA/cm^2 about 60 sec.

B. Phase 2: Fabrication Process

The ZnO film was deposited on FTO-coated glass substrate using a simple aqueous solution containing zinc nitrate hydrate aqueous solution by galvanostatic electrolysis. The solution temperature of zinc nitrate hydrate varied from 60 to 75 °C to investigate the n-ZnO thin film characteristic. A conventional two-electrode cell equipped with Platinum plate counter electrodes used for the deposition. The ZnO electrodeposition galvanostatically carried out at -1.0mA/cm^2 for approximately 60 minutes. The solution temperature was controlled using water bath. The electrodeposition parameter during the growth of ZnO thin films are given in Table 1.

Table 1 Fabrication parameters

Deposition time (min)	Concentration (M)	Current Density (mA/cm^2)	Temp. (^{o}C)
60	0.08	-1	60, 65, 70, 75

C. Phase 3: Deposit Characterisation

The investigation of the structural and morphological of electrochemical deposit-n-ZnO thin film will be carried out using X-Ray diffractometer (XRD), Field-Emission Scanning electron microscopy (FE-SEM), and Atomic force microscopy (AFM), respectively.

Results and Discussion

A. Structural Characteristic

Fig. 1 shows the XRD patterns of FTO glass substrate and samples prepared at solution temperature of 60, 65, 70 and 75 °C. There are several peaks corresponding to ZnO was observed in addition to substrate peaks indicating that the ZnO thin film was successfully deposited. This result is consistent with the previously reported [5, 8, 11]. Moreover, no other peaks were seen on the XRD patterns. The (002)-ZnO peak was increased with the solution temperature preparation and the highest peak was observed at solution temperature of 75°C. According to this result, the solution temperature shows a significant effect of structural properties of ZnO. It can be concluded that the quality of ZnO thin film is depending on the solution temperature.

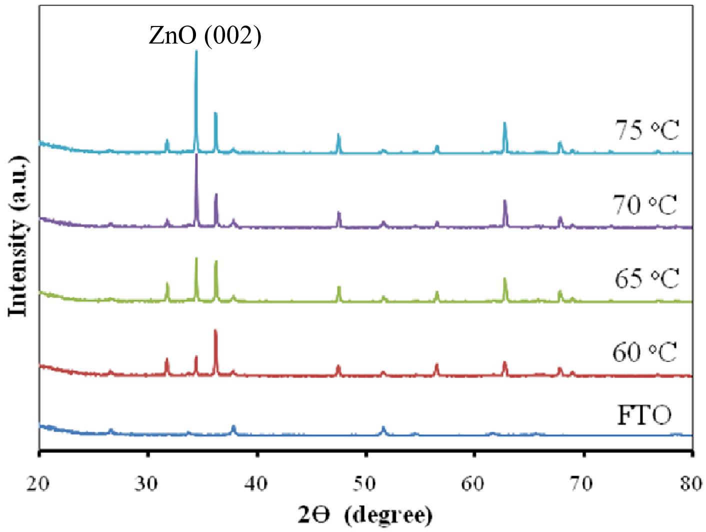

Fig. 1 XRD patterns of ZnO thin film prepared at solution temperature of 60, 65, 70 and 75 ^{o}C.

B. Morphological Characteristic

Fig. 2(a)-(d) show the FE-SEM images of the ZnO thin films deposited under the temperature of 60, 65, 70 and 75°C, respectively. The typical hexagonal columnar grains of ZnO could be observed in all FE-SEM images[5], confirmed that the successfully fabrication of ZnO on FTO glass substrate. From the figures, the morphology of ZnO thin film changed with solution temperature from planar to the nanopillar. The average grain sizes for the ZnO thin films were also changed with the solution temperature, as summarized in Table 2. This result shows that the average grain size decreases when the solution temperature increased. In conclusion, the larger surface of ZnO could be obtained at high solution temperature.

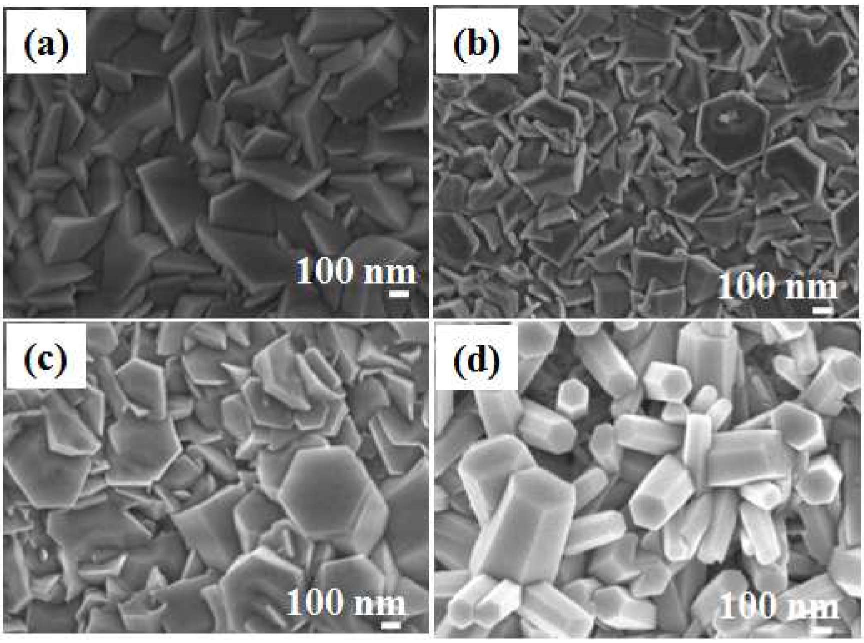

Fig. 2 FE-SEM images of ZnO thin film prepared at solution temperature of (a) 60, (b) 65, (c) 70 and (d) 75 ^{o}C.

Fig. 3 shows the roughness of ZnO thin film fabricated at solution temperature of 60, 65, 70 and 75°C. It was observed that the ZnO thin film roughness increased with the solution temperature. This roughness was taken using Atomic Force Micrograph (AFM) measurement and the AFM three-dimensional (3D) picture of the ZnO thin film shown in Fig. 5. According to this result, the ZnO roughness is consistant with the morphological properties shown in Fig. 2. When the morphology of ZnO changed from planar to nanopillar, the roughness was increased, as predicted. This result shows that the larger surface of ZnO was obtained at higher solution temperature.

Table 2 Average Grain Sizes of ZnO Thin Films

No.	Deposition temperature (°C)	Average grain size (nm)
1	60	439.3
2	65	328.8
3	70	321
4	75	218.6

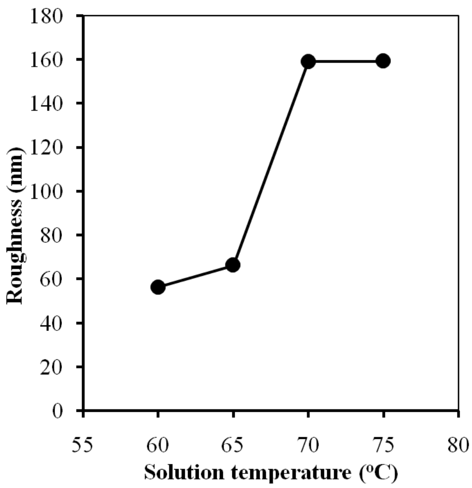

Fig. 3 Roughness of ZnO Thin Film fabricated at solution temperature of 60, 65, 70 and 75 ^{o}C.

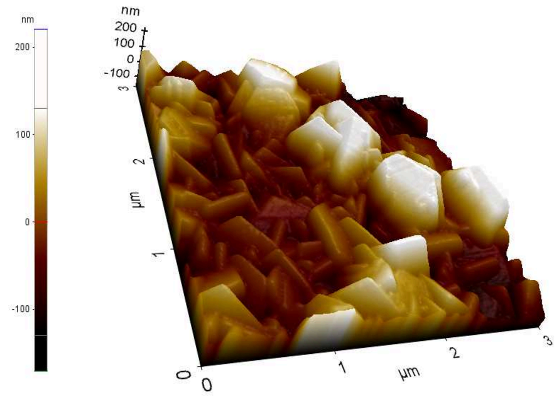

Fig. 4 AFM image of ZnO prepared at solution temperature of 65 ^{o}C.

Conclusion

ZnO thin films were successfully electrodeposited onto FTO glass substrates by the confirmation of XRD and FE-SEM images. The XRD patterns exhibited the increment of (002)-ZnO peak when the solution temperature increased. Moreover, the highest peak of ZnO(002) was observed at 75°C. The morphology of ZnO was changed from planar to nanopillar with the solution temperature. In conclusion, ZnO nanopillar with an excellent structural properties was obtained at solution temperature of 75^{o}C. Although some improvement is needed (increase the solution temperature), the result obtained in this project will open a new door for enhancement of conversion efficiency of heterojunction thin film fabrication.

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