Effect of Substrate Temperature on the Electrical Properties of Al-doped Zinc Oxide Films Deposited on Polyethylene Terephthalate

Mohammad G. Faraj

Department of Physics, Faculty of Science and Health, Koya University, Koya KOY45, Kurdistan Region - Iraq

Abstract—**To prepare homogeneous thin films of zinc oxide (ZnO) doped with aluminum (Al) on a polyethylene terephthalate (PET) substrate at different temperatures (200–250°C), the process is carried out by utilizing the chemical spraying pyrolysis approach. A study of the effects of substrate temperature on the Al-doped Zinc Oxide (AZO) films' electrical characteristics and roughness is performed. The measurements of atomic force microscopy show that the root mean square roughness of the AZO films is increased with the increase of PET substrate temperature. Hall measurements show that the electrical resistivity decreases as the substrate temperature increases. On the increment of substrate temperature, there is an increase in the carrier concentration value from 9.98×10^{19} to 5.4×10^{20} cm^{-3} and an increase in the carrier mobility value from 5.5 to 9.76 cm^2.(V. S)$^{-1}$.**

***Index Terms*—Al-doped zinc oxide, Chemical spray pyrolysis, Electrical properties, Hall measurements, Polyethylene terephthalate.**

I. Introduction

Zinc oxide (ZnO) is an important semiconductor material due to its wide applications in many different fields. Recently, ZnO has received an increasing attention by the researchers. At room temperature, ZnO is an n-type semiconductor with a direct band gap and high energy exciton bonds (Faraj and Ibrahim, 2011). ZnO has many applications because it is cheap, stable, readily available, and secure. These properties make it one of the best materials to be used in optoelectronics. Several research works have been previously reported to study ZnO (Faraj and Taboada, 2017; Faraj and Taboada, 2017; Faraj and Esia, 2019).

Post deposition treatments can easily be used to tune the physical properties of ZnO, such as the incorporation of gallium (Ga), tin (Sn), and aluminum (Al) (Park, Ikegami and Ebihara, 2006; Tynell, et al., 2013).

Researchers have employed gallium, aluminum, and indium as dopants to create n-type ZnO films with excellent quality and conductivity (Myong, et al., 1997; Assuncao, et al., 2003). Al doped ZnO films is considered to be one of the best elements for optoelectronics due to its excellent electrical conductivity (Gong, Lu and Ye, 2010; Raghu, et al., 2017). Several methods were used in the literature to prepare Al-doped Zinc Oxide (AZO) thin films, such as magnetron sputtering, atomic layer deposition, and spray pyrolysis (Zhou, et al., 2007; Kaurn, Mitra, and Yadav, 2015; Ayouchi, et al., 2003).

Plastic substrates are being used more and more in optoelectronics applications, particularly for energy-producing devices like solar cells (Ryu, Cha and Jung, 2010), as they can offer benefits including light weight, high shock resistance, and scalable roll-to-roll preparation processes. Due to their flexibility, light weight, and low cost, AZO films on flexible polymeric substrates as polyethylene terephthalate (PET) for such intended applications are in particular attracting a lot of attention (Jeong, et al., 2010). New to the ongoing work is the preparation and characterization of AZO films fabricated on PET plastic substrates with a chemical spraying pyrolysis technique at different temperatures (200–250°C).

In this study, AZO thin films are deposited on PET plastic substrates by chemical spray pyrolysis at different substrate temperatures (200°C, 225°C, and 250°C) with steps of 25°C. Substrate temperature influences the roughness and electrical characteristics of the AZO films.

II. Experimental Procedure

A. Materials

In this experiment, PET plastic substrates were acquired from Penfibre Sdn. Bhd., company. Materials used in the preparation of AZO thin films include Zinc nitrate ($Zn(No_3)_2.6H_2O$; 99.0%) and aluminum nitrate $Al(NO_3)_3.9H_2O$; 99.9%) were obtained from Sigma-Aldrich.

B. Deposition of AZO Thin Film

AZO thin films were preserved on PET plastic substrates using a chemical spray pyrolysis process. The PET plastic

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Corresponding author's e-mail: mohammad.ghaffar@koyauniversity.org

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substrates were immediately washed with ethanol for 10 min to remove contamination. After cleaning, purified water is used to rinse the substrate (DI water). After that, the sample was dried using nitrogen (N_2) gas. By dissolving 0.1 M watery arrangement of zinc nitrate in DI water, the antecedent arrangement (100 mL) was created. Then, 0.2 mL of nitric corrosive (HNO_3) was added to the arrangement to increase the zinc nitrate's solvency. The final mixture was vigorously mixed for 30 min at 25 °C with an appealing stirrer.

The precursor arrangement was split onto PET plastic substrates at a temperature of preserved ZnO thin films (200°C, 225°C, and 250°C). To begin the investigation into aluminum (Al) doping, aluminum nitrate in various groups of 3% was introduced. To obtain homogeneous thin films, the height of the splashing spout and the rate process were kept constant throughout the testimony operation, at 27 cm and 5 ml/min, respectively.

The arrangement was atomized using packed nitrogen at a weight of 1 bar. Using an optical reflectometer (Model: Filmetrics F20), the estimated thickness of the samples was 300 nm. Using the Hall measurement (HL5500PC) system, hall effect measurement was carried out to determine the electrical properties of AZO thin films, including resistivity, carrier type, mobility and concentration.

Hall mobility (μ_H) is calculated using the relation shown below (Bube; 2003):

$$\mu_H = \sigma . R_H \tag{1}$$

Where σ is the electrical conductivity at room temperature and R_H is the Hall coefficient, whereas the carrier concentration (n) can be determined using the relation (Bube; 2003):

$$n = \frac{1}{R_H . e} \tag{2}$$

Where e is the electron charge.

Atomic force microscopy (AFM) (model: ULTRA1Objective model) was used to examine the surface morphology of the AZO films). The temperate of the all samples were measured by electric hot Plate (model: Onilab). The melting point of PET plastic is 254°C (Faraj, Ibrahim and Ali, 2011), making the temperatures used in this study for AZO film deposition on PET plastic substrates appropriate.

III. Results and Discussion

Fig. 1 presents AFM images of the surface morphologies of AZO deposited on PET plastic substrates at different substrate temperatures. The surfaces of the AZO thin film products were smooth. For films with substrate temperatures of 200°C, 225°C, and 250°C, the estimated root mean square (RMS) surface roughness of the AZO samples was 12.3 nm, 19.64 nm, and 38.87 nm, respectively. As the substrate temperature rises, the RMS surface roughness also rises. Surface roughness was always brought on by the conical features that could be readily seen on the sample surface. It is crucial to remember that surface smoothness is a highly desired property for coatings used in optical applications since it lowers the reflection loss caused by surface scattering caused by roughness. This behavior is similar to earlier reports (Ghorannevis, et al., 2015; Faraj, et al., 2011).

Hall measurements were used to measure the electrical parameters. According to the electrical analysis, the films degenerate and exhibit n-type electrical conductivity. Fig. 2 shows the electrical resistivity of the AZO films as a function of substrate temperature. The film resistivity decreases dramatically as the substrate temperature is increased from 200°C to 250°C in 25°C steps. The evaluated film resistivity's were 18.54×10^{-2}, 3.2×10^{-2}, and 1.14×10^{-2} (Ω.cm) at different temperatures (200°C, 225°C, and 250°C). The Hall mobility and carrier concentration of the AZO films depend on the substrate temperature, as shown in Table 1. As the substrate temperature rises, the Hall measurement reveals an increase in the carrier concentration value from $9.98\ 10^{19}$ to $5.4\ 10^{20}$ cm^3 with a mobility value in the range of 5.5–9.76 cm^2 $(V.\ S)^{-1}$, as shown in Table 1.

The increased mobility of the charge carriers most likely causes the decrease in resistivity. This behavior agrees with that reported in the literature (Hu, et al., 2014; Wang, Li and Zhang, 2009; Zhao, et al., 2020). Since all samples exhibit low resistance and high carrier concentration, transparent front contacts for solar cells are possible.

Table I
The Dependence of the Hall Mobility and Carrier Concentration of the Al-doped Zinc Oxide Films on the Substrate Temperature

No.	T (C)	Hall mobility (cm^2/V.S)	Carrier concentration (cm^{-3})
1	200	5.5	9.98×10^{19}
2	225	8.3	1.6×10^{20}
3	250	9.76	5.4×10^{20}

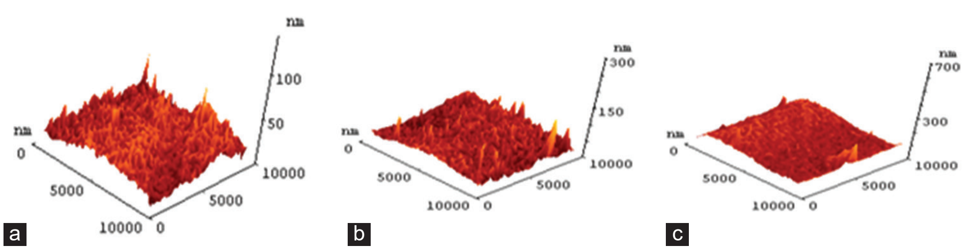

Fig. 1. Atomic force microscopy surface images of Al-doped zinc oxide films as a function of substrate temperature: (a) 200°C, (b) 225°C and (c) 250°C.

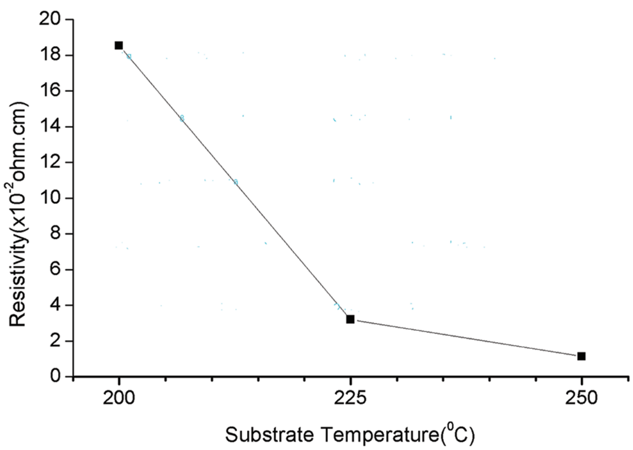

Fig. 2. Resistivity of Al-doped zinc oxide films as a function of substrate temperature.

IV. Conclusions

Chemical spray pyrolysis was used to deposit zinc oxide-doped aluminum (AZO) thin films at various substrate temperatures on PET plastic substrates. AFM and Hall measurements were used to examine the properties of AZO thin films. The effects of the substrate temperature on the roughness and electrical properties of the AZO films were studied. From AFM images, it was found that the RMS roughness of the AZO film surface increased as the substrate temperature increased. From the Hall measurements, it was found that AZO films with different substrate temperatures always exhibit n-type conductivity. From the investigations, it can be concluded that AZO thin film PET plastic substrates would be acceptable for use as transparent front contacts in thin film solar cells.

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