

## EFFECT OF POLYIMIDE SUBSTRATE ON THE PHYSICAL PROPERTIES OF ALUMINUM DOPED ZINC OXIDE (AZO) THIN FILMS DEPOSITED BY SPRAY PYROLYSIS TECHNIQUE

M. G. FARAJ<sup>a,\*</sup>, M. H. EISA<sup>a,b\*</sup>

<sup>a</sup>*Department of Physics, Faculty of Science and Health, Koya University, University Park, Kurdistan Region of Iraq*

<sup>b</sup>*Physics Department, College of Science Sudan University of Science Technology, Khartoum 11113, Sudan*

<sup>c</sup>*Department of Physics, College of Sciences. Imam Mohammad Ibn Saud Islamic University (IMSIU) Riyadh 11623, Saudi Arabia*

Aluminum doped zinc oxide (AZO) thin films were set up at 350°C on polyimide (PI) plastic substrates by chemical spray pyrolysis technique. In this commitment, the impacts of aluminum doping on basic, optical and electrical properties are examined as the capacity of dopant fixation, which was differed somewhere in the range of 0 and 7 %. The X-beam diffraction (XRD) designs displayed hexagonal wurtzite precious stone structure with a favored introduction along the (002) plane independent of Al doping fixation. Accordingly, the film's grains diminished in size with expanding Al content. Nuclear power magnifying lens (AFM) estimations are demonstrated that the surface harshness of the films is diminished because of Al doping also. The impacts of the Al content in the ZnO thin film on the optical properties of the relating slight films were contemplated. The energy band gap diminished from 3.3 eV to 3.2 eV for the examined doping fixation run. At long last, electrical examinations uncovered that the film covered with 7 at % Al dopant has a most extreme transporter grouping of  $37.4 \times 10^{20} \text{ cm}^{-3}$ .

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### 1. Introduction

ZnO is a n-type II-VI semiconductor with wide bandgap energy of 3.37eV and an exciton restricting energy of roughly 60 meV at room temperature [1]. ZnO films have numerous focal points, for example, high transmittance in the unmistakable area, low resistivity, and minimal effort [2-4]. The ordinary dopants that are regularly utilized to upgrade the conductivity of ZnO originate from gathering III (B, Al, In and Ga) of the intermittent table. Specifically, aluminum-doped ZnO (AZO) films have been generally utilized as TCO films for their brilliant conductivity [5-8].

Doped zinc oxide thin films can be connected as window layers in sunlight based cells, electrical contacts in fluid gems, sensors, optical coatings, straightforward terminals, the light transmitting diodes among different utilizations [9– 11]. Doped zinc oxide thin film TCOs have been widely considered as non-poisonous and less expensive options to ITO. TCO films are utilized in an expanding number of optoelectronic gadgets, for example, shows and advanced mobile phones, and additionally sun based cells. Aluminum is a standout amongst the most regularly utilized dopants, bringing about superb optoelectronic properties for TCO applications [12].

A Many techniques have been utilized to deliver doped and undoped ZnO thin films, for example, magnetron sputtering, beat laser affidavit (PLD), warm dissipation, sol-gel and shower pyrolysis (SP) [13–17]. Among the previously mentioned systems, splash pyrolysis is a promising

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\* Corresponding author: mohammad.ghaffar@koyauniversity.org

technique because of its minimal effort nature and appropriateness for saving expansive zone thin-films [18].

Research on ZnO testimony on adaptable polymeric substrates, for example, polyimide (PI) is increasing monstrous interests because of their adaptability, light-weight, minimal effort, high temperature opposition (regularly up to 400°C handling temperature), low coefficient of warm development (CTE), low dampness take-up and high dampness discharge qualities, its amazing electrical properties and furthermore expanded voltage perseverance [19,20]. Because of its predominant properties, PI has discovered applications as substrates in adaptable thin-film sun oriented cells, adaptable printed circuits and high thickness interconnects [21,22].

In the present work, the impact of Al consolidation on the properties of ZnO thin films is accounted for. The films were created utilizing compound splash pyrolysis strategies. The impact of Al doping on the basic, optical and electrical properties of the films were researched.

## **2. Experimental details**

### **2.1. Materials**

The materials utilized in the arrangement of ZnO and Al doped ZnO thin films were zinc nitrate  $Zn(NO_3)_2 \cdot 6H_2O$  and Aluminum nitrate  $Al(NO_3)_3 \cdot 9H_2O$  from [Applichem Panreac] utilized as got. Polyimide (PI) from Good individual Cambridge restricted was utilized as substrates.

### **2.2. Deposition of AZO thin Film**

ZnO and Al-doped ZnO (AZO) thin films were stored on PI plastic substrates by a chemical spray pyrolysis method (CSP). Right off the bat, the PI plastic substrates were cleaned with ethanol for 10 min to evacuate defilement. After the cleaning procedure, the substrate was flushed with refined water (DI water). The example was then dried under nitrogen ( $N_2$ ) gas. The antecedent arrangement (100 mL) was set up by dissolving 0.1M watery arrangements of zinc nitrate in DI water. To improve the solvency of zinc nitrate, 0.2 mL of nitric corrosive ( $HNO_3$ ) was additionally added to the arrangement. The last arrangement was enthusiastically blended utilizing an attractive stirrer for 30 min at 25 °C. ZnO thin films were saved by splashing the forerunner arrangement onto PI plastic substrates at a temperature of 350 °C. Aluminum nitrate with various groupings of 0, 1, 3, 5 and 7 at % was added to begin answer for aluminum doping.

So as to get uniform thin films, the tallness of the splashing spout and the rate of shower process were kept steady amid the testimony procedure at 27 cm and 5 ml/min separately. Packed nitrogen was utilized to atomize the arrangement at a weight of 1 bar. The thicknesses of AZO thin films on PI plastic substrates were 200 nm, as estimated utilizing an optical reflectometer (Model: Filmetrics F20).

The basic properties of the readied AZO thin films have been examined utilizing a high goals X-beam diffractometer framework (Panalytical Empyrean) with  $CuK\alpha$  radiation ( $\lambda$ ) wellspring of 0.154 nm. The surface morphology of films was affirmed by nuclear power magnifying instrument (AFM) (Model: ULTRA1Objective). The UV–VIS spectra of the thin films were obtained utilizing a VARIAN CARY 50 (Agilent Technologies) spectrometer. At last, the electrical properties of thin films were estimated by Hall estimations (HL5500PC) framework.

## **3. Result and discussion**

### **3.1 Structural properties**

The X-beam diffraction examples of as-saved unadulterated and Al-doped ZnO films at (1, 3, 5 and 7 at%) as shown in Fig.1. It uncovered that all the developed films were polycrystalline in nature, demonstrating three pinnacles comparing to (100) and (101) planes with a favored introduction (002) can be all around listed to the hexagonal wurtzite precious stone structure [23]. All these watched pinnacles are extremely all around coordinated with the tops in the JCPDS card no: 01-089-0510. Regardless of doping fixation, every one of the films demonstrated pinnacles like unadulterated ZnO, which shows that no auxiliary disfigurement happened in ZnO cross-

section upon Al-doping. This affirms the effective substitution of Al particles in Zn cross section destinations in the ZnO framework. The three pinnacles which have a place with the PI polymer are situated at 22.1°, 26.0° and 44.60 as found in Fig. 1. This outcome concurs with recently revealed information [24].

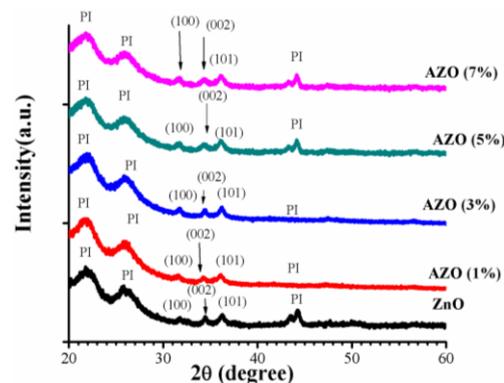


Fig.1. XRD spectra of pure and Al-doped ZnO thin films deposited.

The crystalline grain size ( $t$ ) of the films was determined using Scherrer formula [25].

$$t = \frac{0.9\lambda}{\beta \cos\theta}$$

where  $\beta$  is the full width at half most extreme (FWHM) of the pinnacle,  $\lambda$  is the wavelength of the X-beam of 1.5406 Å, and  $\theta$  is their pinnacle position. In light of the line width of the (002) diffraction top, the crystallite estimate is observed to be in the range between 28.39 to 18.12 nm for ( $x = 0-7$  at %), as appeared table 1. It was seen that the expansion of the doping decline in the crystallite size of the films. This outcome is in great concurrence with recently announced information in other film structures [26].

Table 1. The  $2\theta$  values, inter planar spacing and lattice parameter of Al doped ZnO thin films as a function of Al content are shown.

Al at. %	$2\theta$ value ( $^{\circ}$ )	Crystalline grain size ( $t$ ) nm	Inter planner ( $d$ ) Å <sup>0</sup>	Crystal plane (hkl)
0	34.5124	28.39	2.6145	(002)
1	34.4124	24.17	2.6137	(002)
3	34.3235	22.43	2.6121	(002)
5	34.2531	19.31	2.6102	(002)
7	34.1875	18.12	2.6083	(002)

The between planer dividing and their cross section consistent are diminished with expanding doping focus as appeared Table 1. The decline in the grid consistent is ascribed to the sl substitution of Al contamination particles in the place of Zn ionic locales in ZnO lattice, which causes a little rebel on account of thevarious ionic span of Al particles contrasted with Zn particles, as recently revealed [27].

AFM pictures of the surface morphologies of the ZnO and ZnO: Al thin filmsare shown in Fig. 2. The rms harshness of the unadulterated ZnO and (1, 3, 5 and 7 at %) Al-doped ZnO thin films were observed to be around 29.4, 23.7, 20.3, 17.24 and 15.46 nm, separately. The AFM

results uncover that the surface harshness diminishes with expanding Zn fixations. This outcome is concurs with recently detailed outcomes [28].

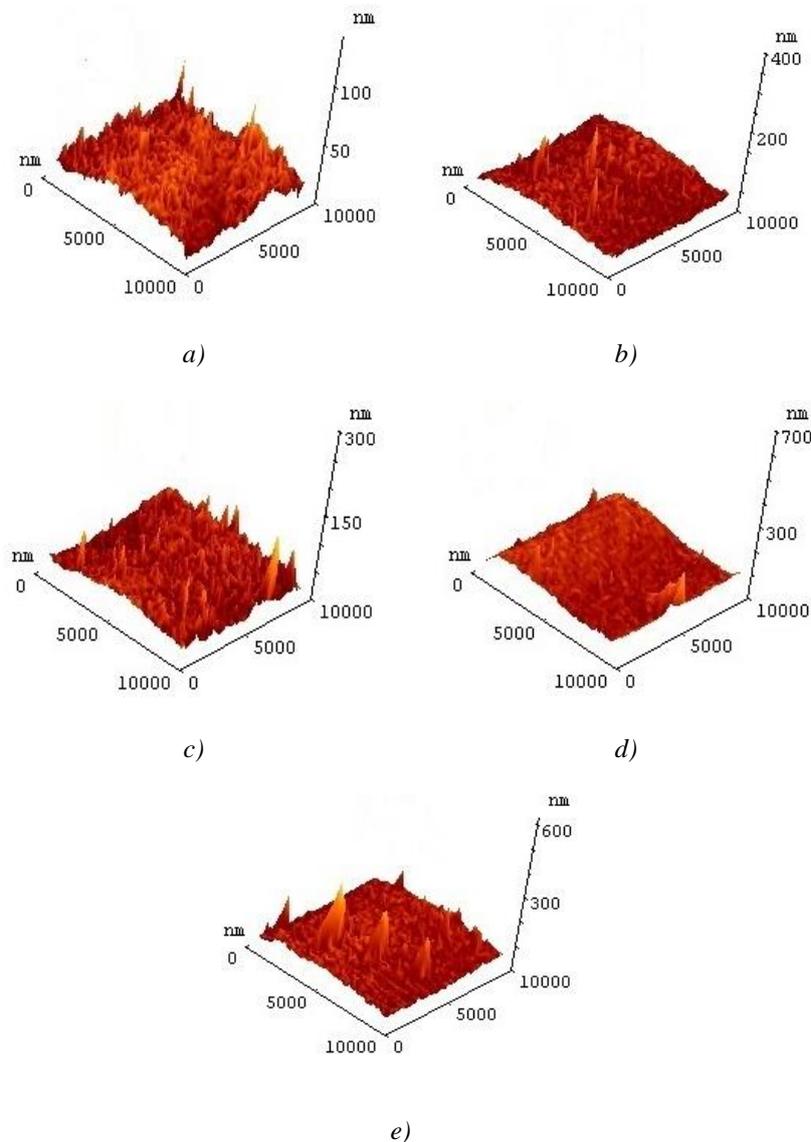


Fig.2. AFM analysis of films with (a) undoped ZnO, (b) Al (1 at %), (c) Al (3 at %), (d) Al (5 at %) and (e) Al (7 at %).

### 3.2. Optical properties

The impact of Al-doping levels on the optical properties of splashed AZO thin films was considered from the optical estimations. Fig. 3 demonstrates the optical transmittance versus wavelength spectra of AZO films taken in the wavelength go, 300– 1000 nm.

The undoped ZnO films demonstrated an optical transmittance of > 70% while the transmissions of the AZO films increments with expanded the substrate temperature. The Al doped ZnO films with higher dopant fixation (7 at %) showed marginally high optical transmittance (> 80%). The variety in the optical transmittance of the films in the noticeable district relates to the progressions in both the crystalline size and the surface harshness of the films on Al-doping [29].

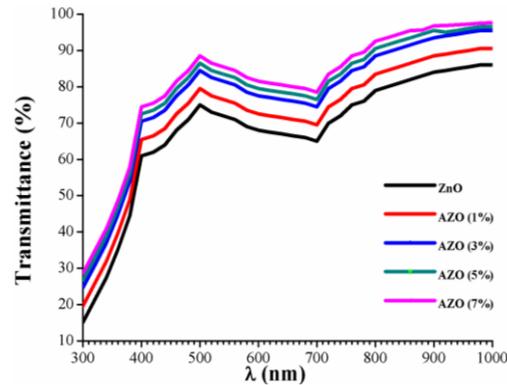


Fig. 3. Transmittance spectra of AZO films with Al doping varying from 0 to 0.7 as a function wavelength.

It is well known that ZnO is a direct-band-gap semiconductor. Thus, the absorption coefficient in the region of strong absorption obeys the following equation [30].

$$\alpha = \frac{k}{h\nu} (h\nu - E_g)^{\frac{1}{2}} \quad (3.2)$$

where  $k$  is steady,  $h$  is the Planck constant,  $\nu$  is the radiation recurrence, and  $E_g$  is the band gap energy. From the straight line of the plots  $(\alpha h\nu)^2$  versus photon energy ( $h\nu$ ) the band gap of AZO thin films saved at 250, 265, 280 and 295°C (see Fig.4) were observed to be 3.3, 3.28, 3.25, 3.23 and 3.20 eV, separately. From such plots, the optical band gap estimations of AZO thin films are found to diminish as the doping expanded. The doping prompted counterbalance the retention edge towards low energies and the explanation behind this is to make topical dimensions beneath the conduction band as they are directed to retain photons of lower energy and increment in electronic changes and after that decline in the estimations of the optical energy gap [31]. These qualities go amiss from the standard mass esteem, 3.2 eV [32].

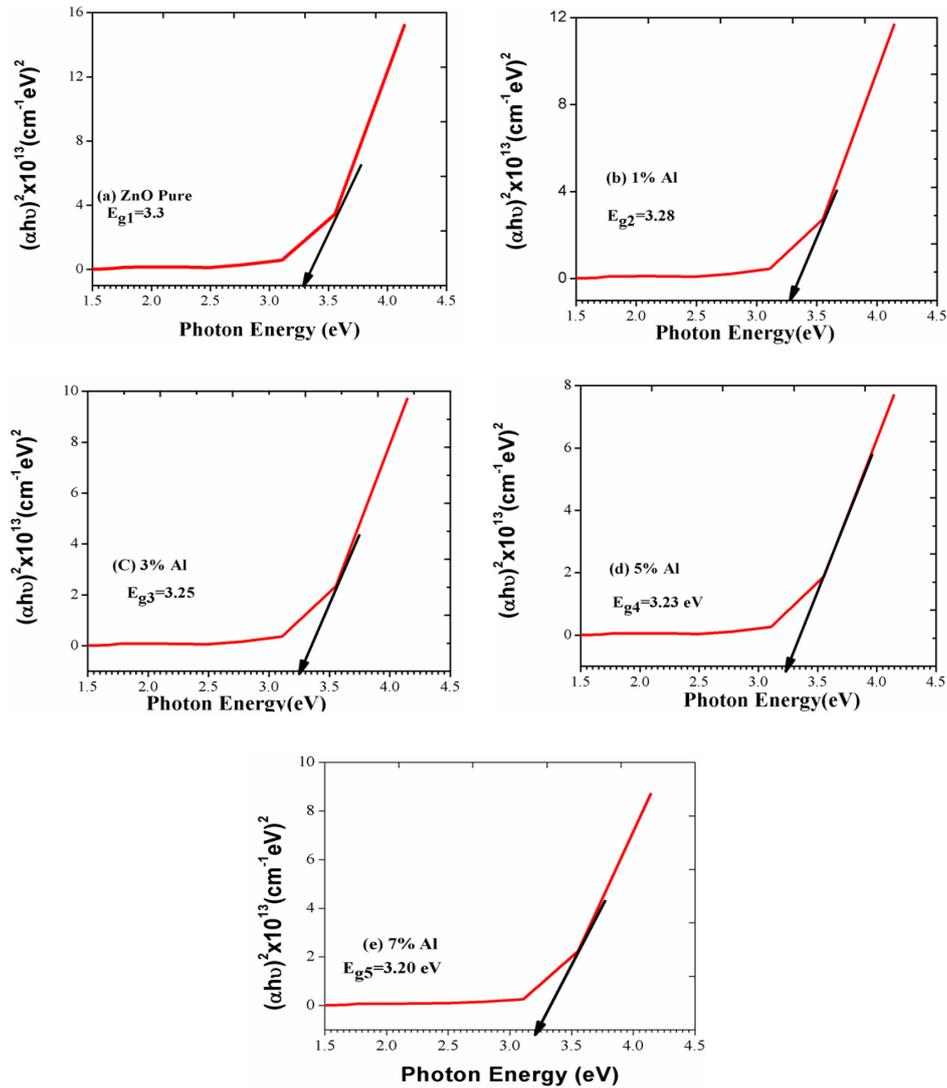


Fig. 4. Plot of  $(\alpha h\nu)^2$  as a function of photon energy for AZO films deposited on PI substrate at different doping Al.

### 3.3. Electrical properties

The reliance of the electrical resistivity and the transporter fixation on the Al substance of ZnO thin film are appeared in Table 2. The resistivity of all films declines with expanding Al fixations because of the increment in carrier focus and in view of substitutional joining of Al particles in the ZnO structure [33].

Table 2. Dependence of electrical resistivity and carrier concentration on the Al content of the ZnO films.

Al content (at %)	Resistivity ( $\Omega.m$ )	Carrier concentration ( $cm^{-3}$ )
0	$32.34 \times 10^{-2}$	$6.3 \times 10^{20}$
1	$23.24 \times 10^{-2}$	$8.5 \times 10^{20}$
3	$18.54 \times 10^{-2}$	$10.1 \times 10^{20}$
5	$9.65 \times 10^{-2}$	$20.3 \times 10^{20}$
7	$4.35 \times 10^{-2}$	$37.4 \times 10^{20}$

#### 4. Conclusions

In the present paper, the effect of differed Al-doping focus (0– 7 at %) on basic, optical and electrical properties of ZnO thin films were considered. Aluminum doped zinc oxide (AZO) thin films with an ostensible 200 nm thickness were set up at 350°C on polyimide (PI) plastic substrates by synthetic splash pyrolysis technique. It is found from the XRD examination that the precious stone nature of the films was disintegrated with the expansion of Al doping focus. Nuclear power magnifying lens (AFM) estimations are demonstrated that the surface harshness of the films is diminished because of Al doping too. of the electrical outcomes showed that the AZO thin film is a n-type semiconducting material. The electrical investigations uncovered that the film covered with 7 at% Al dopant has a most extreme bearer centralization of  $37.4 \times 10^{20} \text{ cm}^{-3}$ . From these outcomes, it is demonstrated that Al=7 at.% is the upgraded doping fixation for ZnO films so as to improve its physical properties appropriate for sun based cell application.

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