UV-irradiated sol-gel spin coated AZO thin films: enhanced optoelectronic properties

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ABSTRACT

Thin films of transparent conductive Al doped ZnO (AZO) thin films were produced via sol-gel spin coating route. Structural, optical, and electrical properties were explored for several dopant concentrations. Formation of crystalline AZO was verified by X-ray Diffraction (XRD) Analysis and structural analysis were carried out later from the XRD data. Highest band gap of 3.67 eV was found for 2 mol % AZO thin films. The average transmittance was found to be 84.19% in the visible spectra for the corresponding thin films. 2 mol% AZO also exhibited a minimum resistivity of 2.05 Ω-cm with a maximum value of figure of merit. Prolonged UV irradiation was applied to 2 mol % AZO thin films prior to annealing. It significantly modified the surface morphology of the film and provided shielding near UVA (315–378 nm) spectrum. This also enhanced the conductivity of the thin film by 3-fold compared to non-UV treated sample and decreased optical band gap significantly.

1. Introduction

Doped ZnO thin films have grabbed immense attention as transparent conductive oxide (TCO) for empirical applications in electroluminescent devices and solar cells [1, 2, 3, 4, 5]. ZnO exhibits anisotropic crystalline structure oriented along (002) plane having a direct bandgap about 3.37 eV [6, 7]. Owing low cost, availability, non-toxicity and excellent opto-electronic properties, ZnO thin film would be a good substitute to the extensively utilized indium tin oxide (ITO) thin film [8]. High valence impurities such as Al, Ga, In etc. can further enhance the conductivity of ZnO up to about 10^{-4} S/cm by donor electrons [9, 10]. Such thin films are widely prepared using sol-gel route because of versatile compositional control, affordable cost, homogeneity in molecular level for mixing of liquid precursors and lower crystallization temperature [11]. In alkoxide route, organo-metallic precursors are expensive and sometimes hazardous. Therefore non-alkoxide route is more preferable. ZnO thin film prepared using different types of precursors, solvents and stabilizers are reported in literature to tailor its structural and opto-electronic properties. However, when as solvents 2-methoxy ethanol and as stabilizer mono-ethanolamine (MEA) were used, this results in high c-axis orientation [12, 13]. Many experiments on doped ZnO thin films made by sol-gel processing have been published. However, there remains some difficulties associated with the aqueous sol–gel method, as hydrolysis, condensation, and dry out take place concurrently, resulting in difficulty in reproducing the final product and tailoring target properties.

Apart from that, band gap of ZnO is 3.37 eV corresponding to 378 nm wavelength and distinctive electrooptical properties. The coherent blend of UV absorption with better transparency in the visible range makes it a suitable candidate for optoelectronic devices that needed both the life expectancy and improved efficiency under working conditions [14, 15, 16, 17]. According to some recent papers [18, 19, 20], the effect of UV irradiation altered conductivity, surface morphology and structural properties of zinc oxide thin films significantly. C-Y. Tsay et al. reported two-fold improvement of photocurrent responsivity and a sharp increase in photocurrent under illumination with UVA light with Al doping in ZnO thin films [21]. Y.-K. Tsay reported that UV irradiation results in better crystallinity, solidity, and surface roughness. Their work also showed that, compared to non-UVA treated samples, the films with UV irradiation results in good conductivity [22]. J. Kim et al. showed that the UV irradiation can improve structural properties significantly as well [23]. A. C. Marques et al. investigated the power output density of AZO thin films under UV light and found it to be increased during exposure of the films to UV-light, due to the photo-thermolectric effect [24]. So, further investigation is required to access the quality of thin films after UV irradiation.

Here, Sol-gen spin coating was employed to produce thin films of Al doped ZnO (AZO) were prepared following non-alkoxide sol-gel spin...
coating route and properties were examined thoroughly. Structural features were studied using Field Emission Scanning Electron Microscope (FE SEM) and X-ray Diffraction (XRD), optical properties using Ultraviolet-visible (UV-vis) spectroscopy and electrical properties using Four-Point probe. Dopant concentration yielding the best optical and electrical properties was optimized for future transparent conductors. Effects of extended period of UV irradiation have also been investigated.

2. Materials and methods

AZO thin films were deposited on a silica glass substrate. Precursor, solvent and stabilizer used were zinc acetate dehydrate (Zn(CH₃COO)₂·2H₂O), 2-methoxy ethanol (MOE) (CH₃OCH₂CH₂OH) and mono-ethanolamine (MEA) ((OHCH₂CH₂)NH₂) respectively. Aluminum nitrate nonahydrate (Al(NO₃)₃·9H₂O) was used as doping agent. MEA to zinc acetate molar ratio was 1.0. Concentration of metal ions in the solution was 0.4 mol/L. Dopant concentration was varied between 1.0 to 3.0 mol percent with respect to ZnO. To obtain a clear, homogeneous, and transparent solution, all of the elements were mixed in a stoichiometric ratio. The sol was stirred for 1 h at 80°C. Dynamic spin coating was used to deposit thin films at a rotation speed of 1000 rpm for 10 s and 3000 rpm for 20 s. After each step, films were dried at 180°C for 5 min. This procedure was repeated 3 times for 3 layers. Finally, at 500°C the films were annealed for 2 h in furnace atmosphere. UV radiation was applied to the sample of highest conductivity prior to annealing at 500°C. The UV source was a 100 W UV-A lamp (LH-360-100-A, UV emission wavelength 365 nm) and the irradiation was performed by illuminating the sample by the UV source in a dark chamber.

Figure 1. XRD of the thin films.

Figure 2. Effect of doping on texture coefficient.

The micro-strain was calculated using Williamson-Hall equation, Williamson Hall Plot is shown in Figure 3:

\[ \beta \cos(\theta) = \frac{K \lambda}{D} + 4\varepsilon \sin(\theta) \] (2)

where, \( D \) is crystal size, \( \lambda \) is CuKα radiation wavelength of (0.15406 nm). Full width at half maximum (FWHM) \( \beta \) is measured in rad and \( \theta \) is half of the Bragg diffraction angle.

With increasing Al concentration, a gradual decrease in major peak intensity was observed which indicates degradation of crystalline quality of ZnO resulted from the substitution of Zn\(^{2+}\) by Al\(^{3+}\) ions (Figure 2). The gradual increase of strain with increasing doping concentration due to variation in ion size of Al with Zn ions (\( r_{Al} = 0.054 \text{ nm} \) and \( r_{Zn} = 0.074 \text{ nm} \)) can be a reason for the deterioration in crystallinity on doping (Table 2) [27, 28]. Also, with excessive doping of 3 mol % Al, the greater

3. Results and discussion

3.1. Effect of doping

3.1.1. Crystallographic structure

The hexagonal wurtzite structure of all the thin films were confirmed from the indexed XRD peaks (ICDD card no. 80-0075). No addition peak was observed for pure Al or Al₂O₃, or any other impurity phase which indicates the well diffusion of Al\(^{3+}\) in the crystal structure of ZnO (Figure 1). High degree of crystallinity of all the films are evident from the peak intensity and their sharp peak width.

Crystal size for all synthesized films were obtained using the Scherrer’s Eq. (1),

\[ D = 0.9\lambda / \beta \cos(\theta) \] (1)

Table 1. XRD data assessment for the thin films.

| Thin Films       | 2θ (degree) of [002] Peak | Average FWHM | Crystallite Size (nm) | Average Micro-strain |
|------------------|---------------------------|--------------|-----------------------|----------------------|
| Undoped ZnO      | 34.3924                   | 0.267        | 30.1                  | 0.293                |
| 1 mol % Al doped ZnO (1% AZO) | 34.4123                   | 0.275        | 28.3                  | 0.348                |
| 2 mol % Al doped ZnO (2% AZO)  | 34.4356                   | 0.283        | 27.5                  | 0.358                |
| 3 mol % Al doped ZnO (3% AZO) | 34.4720                   | 0.298        | 27.1                  | 0.366                |
charge (+3) of aluminum can create more bonds with oxygen which degraded the quality of hexagonal wurtzite structure of Zinc oxide. Here, Al$^{3+}$ substituted Zn$^{2+}$ in the lattice. That means some zinc-oxygen bonds have been replaced by aluminum-oxygen bonds. Also, no phase of aluminum oxide was identified from XRD pattern. It seems that even if any small amount of aluminum oxide was formed, that was not significant and was below the detection limit of XRD.

Preferred orientation were calculated from the Harris’s analysis [24] from texture coefficient as per Eq. (3)

$$\text{TC}_{(hkl)} = \frac{I_{(hkl)} - I_{0(hkl)}}{\sum_{n=-\infty}^{\infty} I_{(hkl)} - I_{0(hkl)}} \times 100\%$$

where, texture coefficient of plane (hkl) is $\text{TC}_{(hkl)}$, $I_{(hkl)}$ and $I_{0(hkl)}$ are the intensity of the planes of the thin films and randomly oriented ZnO powder respectively defined by (ICDD card no. 80-0075); N is diffraction peaks’ number.

Texture co-efficient having values greater than one (unity) implies a greater degree of preferred orientation. As X-ray intensities depend on atomic structure factors, the deviation in the TC values from unity means change in atomic density along that plane [31]. The values of texture coefficient indicate preferred orientation along (002) plane for both undoped and doped films (Figure 2). The preferred orientation towards c axis gradually decreased with the increasing doping concentration. This indicates that the strong (002) orientation reduced with the substitution of Al in ZnO lattice. The greater values of TC to a particular plane corroborates with the increase in planer density along that plane [29, 30].

### 3.1.2. Optical properties

Figure 4 shows transmission and absorbance spectra in the wavelength range 300–1000 nm for undoped ZnO and AZO thin films prepared with various doping concentrations. Average transmittance of undoped ZnO was 86.28% and was dropped slightly for all the doped thin films. However, in the visible spectra, all films showed transparency greater than 80% and a sharp absorption edge in the 350–400 nm wavelength range, which can be credited to the intrinsic band gap of the films [31]. The rapid decline in transmittance near the fundamental absorption edge is the corroboration of the superior crystallinity of the films [5]. The superior crystalline nature with comparatively lower defect density makes it suitable candidate for optoelectronic devices [32, 33].

The estimation of direct band gap energy was obtained using the use of Tauc’s law [34, 35].

$$\alpha h \nu = C (h \nu - E_g)^{1/2}$$

(4)

Here, $\alpha$ is coefficient of optical bandgap, energy of incident photon is $h \nu$, C is a constant and $E_g$ is optical band gap. By extrapolating the linear portion, the band gap values were estimated from the $(\alpha h \nu)^2$ vs $E_g$ plots (Figure 3).

Band gap values on doping were higher than that for undoped ZnO (Figure 5). Band gap increased with increasing Al addition showing maximum band gap of 3.67 eV for 2% AZO. This increase can be attributed to the degenerate doping of Al. When Zinc oxide is doped with Al, the lowest band gets saturated with free electrons. Fermi level remains between conduction and valence bands for nominally doped semiconductors. However, Fermi level can is pushed into the conductive band minimum for degenerate doping, such as for Al. Therefore, the next available empty state now becomes the suitable candidate for higher unoccupied conduction state. This raises the lowest optical transition energy and the gap in the optical band. This phenomenon is known as the Moss-Burstein shift [36]. For further addition of Al above 2 mol %, bandgap was decreased. This decrease of $E_g$ is believed due to the separation of excess Al atoms at grain limits as well as by the presence of the interstitial defect states [37]. This decrease also can be attributed to the exchange interactions between sp-d bands.

### 3.1.3. Film resistivity

Figure 6 depicts the difference in resistivity of undoped and AZO thin films with different doping concentrations. The resistivity was dropped

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Table 2. Texture coefficient and film thickness for ZnO and AZO thin films.

| Thin Films         | Texture Coefficient, TC(hkl) | Film Thickness (nm) |
|--------------------|-----------------------------|---------------------|
| Undoped ZnO        | (002) | (100) | (101) | 393 nm |
| 1 mol % Al doped ZnO (1% AZO) | 2.56 | 0.20 | 0.22 | 287 nm |
| 2 mol % Al doped ZnO (2% AZO) | 2.30 | 0.38 | 0.31 | 227 nm |
| 3 mol % Al doped ZnO (3% AZO) | 2.06 | 0.65 | 0.28 | 225 nm |

Figure 3. Williamson Hall Plot of AZO thin Films.

Figure 4. Optical transmittance spectra (left) and absorbance spectra (right) of ZnO and AZO thin films.

Figure 5. Band gap increased with increasing Al addition showing maximum band gap of 3.67 eV for 2% AZO.
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significantly with 1 mol % Al addition because of the excess carrier concentration resulted from Al doping and decreased further showing minimum resistivity of 2.05 Ω-cm for 2% AZO. Later, resistivity was increased with further Al addition. For such addition, carrier concentration decreases since increasing dopant might have formed impartial defects. Also these atoms do not provide free electrons. Decrease in crystallinity may also offset the effect of increased carrier concentration by decreasing carrier mobility leading to the declined conductivity for 3 mol percent doping [38].

To quantify the optoelectronic properties, figure of merit ($\phi$) was calculated using the following equation [39]:

$$\phi = \frac{T^{10}}{\rho}$$

where, $T$ is the transmittance value at 650 nm wavelength [40] and $\rho$ is the volume resistivity. The maximum value of $\phi$, best fusion of lower resistivity and higher transmission results in TCO films with comparatively better performance or quality [35]. The figure of merit was first decreased with initial addition of Al since the decrease in resistivity could not offset the decrease in transmittance compared to undoped ZnO. However, highest value of $\phi (0.94 \times 10^{-8} \Omega^{-1} \cdot \text{cm}^{-1})$ was obtained for 2% AZO film which is in well acceptable range for TCO to be used in optoelectronic applications [41, 42].

### 3.2. Effects OF UV irradiation

#### 3.2.1. Crystallographic structure

The lengthy period of UV treatment has significant changes in crystal structure of 2% AZO thin film (Table 3). The average crystallite size increased with the UV treatment along with the increase in Texture coefficient along (002) plane. That indicates the tendency of the crystals to grow along (002) plane increased with prolonged UV treatment. UV treatment could also initiate the recrystallization of the grains causing rise in surface roughness of after irradiation which is evidenced in Table 4.

Average FWHM of (002) peak is reduced with the advent of UV treatment (Figure 7) and subsequently increase of the crystallite size was from 27.5 nm to 30.8 nm. The Braggs angle shifted towards higher values along with the increase in micro strain in the structure which suggest that the onset of UV treatment created compressive stress in the structure of AZO thin films [43, 44]. After the UV treatment, the increase in texture coefficient in (002) direction establishes the preferential c axis orientation of the crystals. So, the crystallographic properties improved in the (002) direction as the XRD peak intensity increased with the decrease in FWHM values from 0.283 to 0.263. These results indicate that crystallinities towards c axis improved after prolonged UV treatment.

#### 3.2.2. Optical properties

From the comparison of the transmission spectra for UV treated and non-UV treated 2% AZO thin films it is evident that absorption edge shifted to higher wavelength for the case of UV treated sample (Figure 8). This corresponds to the decreased optical band gap of 3.59 eV. This decrease in band gap with increasing crystallite size is well expected due to quantum confinement effect. The protracted UV exposure caused the increase in RMS surface roughness from 18.29 nm to 22.45 nm. 2 mol% AZO UV treated sample shows an average transmittance of 82.20% in visible region compared to 84.19% transparency in non-UV treated 2% AZO films which can be attributed to the increased RMS value and greater light scattering effect.

Upon reaching the visible spectra from UVA spectra there is a sudden increase in transparency in the thin film. This sudden change in transparency in UV region might be helpful in constructing various sensors.

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### Table 3. Experimental results from XRD analysis of UV treated vs non-UV treated 2% AZO films.

| Thin Films | $2\theta$ (degree) | Average FWHM | Crystallite Size (nm) | Average Micro Strain | Texture Coefficient of (002) |
|------------|--------------------|---------------|-----------------------|----------------------|-----------------------------|
| 2% AZO     | 34.4356            | 0.283         | 27.5                  | 0.358                | 2.30                        |
| 2% AZO UV-treated | 34.4534          | 0.263         | 30.8                  | 0.396                | 2.33                        |

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![Figure 5](image_url)  
*Figure 5. $(\alpha h \nu)^2$ vs $E_g$ plot to calculate optical energy bandgap.*

![Figure 6](image_url)  
*Figure 6. Volume resistivity and figure of merit with doping.*
After UV treatment the 2% thin films showed greater capacity to block UVA radiation by 42.49% compared to non-UV treated samples which blocks by 33.35% as shown in Table 4. UV blocking capabilities of the thin films were calculated using Eq. (6). The overall light transmittance in each wavelength band is measured by the \( \tau_{\lambda,\lambda_0} \) value, where \( \lambda \) and \( \lambda_0 \) is the range of the wavelength, which is evaluated, \( \Phi_{\text{AM} 1.5} (\lambda) \) is solar light intensity distribution function. Data was taken from National Renewable Energy Laboratory (NREL), weighted average is between 315 and 378 nm and \( T(\lambda) \) is the weighted average of transparency in the 315 nm–378 nm wavelength for estimating UV blocking.

\[
\tau_{\lambda,\lambda_0} = \frac{\int_{\lambda_0}^{\lambda} T(\lambda) \cdot \Phi_{\text{AM} 1.5} (\lambda) \cdot d\lambda}{\int_{\lambda_0}^{\lambda} \Phi_{\text{AM} 1.5} (\lambda) \cdot d\lambda} \times 100
\]  

(6)

### 3.2.3. Film resistivity

Prolonged UV irradiation caused noticeable improvement in the conduction of 2% doped films (Table 4). UV exposure resulted in the enhanced conductivity. AZO thin films have a persistent photoconductivity because of activation due to light irradiation. UV irradiation for a long period affects excitation and activation of conduction band electrons and thus upgrades the electrical conductivity [46].

Hall coefficient were evaluated using Van Der Pauw method [47]. 2% AZO UV treated thin films have excess carrier concentration may be due to ionization of oxygen vacancies which work as donor sites in the AZO lattice. With the prolonged UV light absorption electron–hole sets are

| Thin Films                  | Fraction of UV Light Blocked (%) | Average Transmittance in Visible Region (%) | Optical Bandgap (eV) | Conductivity (S/cm) | RMS Surface Roughness (nm) | References                      |
|-----------------------------|----------------------------------|--------------------------------------------|----------------------|---------------------|---------------------------|--------------------------------|
| 2% AZO                     | 33.35                            | 84.19                                      | 3.67                 | 0.3806              | 18.29 nm                  | Current Research              |
| 2% AZO UV-treated          | 42.49                            | 82.70                                      | 3.59                 | 1.177               | 22.45 nm                  | Current Research              |
| Undoped ZnO                | 73                               | 79.8                                       | 3.57                 | -                   | 31 nm                     | [24]                           |
| 2% Doped ZnO               | -                                | 86                                         | 3.29                 | 9.52                | -                         | [27]                           |
| Undoped ZnO with 25 Hours UV treatment | 40–45                            | 85                                         | 3.24                 | -                   | 13.23                      | [22]                           |

### Table 4. Optical Properties summary of 2% AZO sample before and after UV treatment with reference to relevant references of Similar work.

Figure 7. XRD patterns of films before and after UV treatment.

Figure 8. (a) Absorbance spectra, (b) transmittance spectra, band gap of (c) non-UV treated and (d) UV-treated 2% AZO.
created by UV light assimilation at beginning. Following hole diffusion to the grain boundaries, the chemisorbed species existing at these locations are oxidized, which might result in their desorption [48]. Because this process solely consumes holes, a buildup of electrons develops, leading to an growth in carrier density. As a result of the increased number of surface donors generated by UV irradiation, the electrical conductivity of the material is improved. Furthermore, it can decompose organic substances such as methoxy groups via photocatalytic reactions when exposed to UV light. These decompositions can result in photo-induced ion doping and the generation of free electrons on the 2% AZO UV treated thin film [49]. The availability of free electrons may influence the increase in conductivity in UV-treated thin films.

3.2.4. Morphology of film surface

The micrographs demonstrated granular morphology of all the films with polycrystallinity in nature where each grain was formed due to multiple crystallite agglomeration (Figure 9). Ganglia-like patterns were found at low magnification for both pure ZnO and AZO thin films, which are typical of such films. The mentioned pattern development is driven by stress relief because of solvent evaporation during the drying process [40]. The average film thickness of the 2% AZO UV treated sample was 212 nm, while it was 237 nm for the non-UV treated sample. It validates the enhancing of the solvent evaporation process later on after it was put into UV chamber [50] and reduced the film thickness. Because the energy came from the UV light irradiation, it could move the molecules and could enhance the solvent evaporation process after putting it into UV chamber.

From the 3D images of the AZO samples in Figure 10 (b, d) it was detected that surface roughness intensified on UV treatment causing blockade of UV rays absorption into the sample which led to higher UV reflection.

4. Conclusion

In this study, all films were made following sol-gel spin coating and its properties were characterized afterwards. Varying concentration of Al,
2% AZO annealed at 500 °C exhibited maximum optical band gap of 3.67 eV with a transmittance of 84.19% and minimum volume resistivity of 2.05 Ω·cm. Low volume resistivity along with the high transmittance results in the high value of figure of merit for 2% AZO which makes it most suitable to be used in optoelectronic applications. The UV exposure results on this doped AZO thin film showed a significant increase in conductivity with decreased band gap and better UVA protection capabilities under UV source. The UV irradiation shows significant change in RMS surface roughness and film thickness.

Declarations

Author contribution statement

Md. Irfan Khan, Tasratur Reaj Naha: Performed the experiments, analyzed and interpreted the data, wrote the paper. Md. Muktidar Billah: Conceived and designed the experiments, analyzed and interpreted the data, contributed reagents, materials, analysis tools or data.

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The authors declare no conflict of interest.

Additional information

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