

Elaboration and Characterization of Transparent Conductive Oxide Thin Films and the Effect of Doping

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Abstract: This study presents the synthesis and characterization of Al-doped ZnO (AZO) thin films on glass substrates via spray pyrolysis. The influence of Al doping (3%, 5%, 7%) on structural, optical, and electrical properties was analyzed. XRD confirmed a polycrystalline wurtzite structure with a (002) orientation, ensuring successful Al incorporation. UV-Vis spectroscopy showed enhanced transparency (70% to 78%), with the bandgap widening from 3.23 eV to 3.32 eV due to the Burstein-Moss effect. Hall Effect measurements revealed improved conductivity ($3.37 \times 10^{-1} \Omega^{-1} \text{cm}^{-1}$) at 3% doping, though mobility saturation limited further gains at higher levels. These results highlight AZO as a cost-effective alternative to ITO for solar cells and optoelectronics.

Keywords: ZnO; Thin films; Al doping; Spray pyrolysis.

Introduction

Zinc oxide (ZnO) is a widely studied transparent conductive oxide (TCO) due to its excellent optical and electronic properties. With a direct bandgap of ~ 3.34 eV and high exciton binding energy (60 meV), it is ideal for optoelectronic applications such as solar cells, LEDs, and sensors (Fan et al., 2017; Ouhabi et al., 2018; Pham, 2023). However, pure ZnO has low conductivity and suboptimal transparency due to limited carrier concentration (Van de Walle, 2009; Lee et al., 2023).

Doping, particularly with aluminum (Al), significantly enhances ZnO's electrical and optical properties. Al-doped ZnO (AZO) films offer improved conductivity, transparency, and stability, making them viable alternatives to indium tin oxide (ITO) for photovoltaic and optoelectronic devices (Liu et al., 2010; Petrov et al., 2023; Miloua et al., 2012; Amroun et al., 2020). Spray pyrolysis, a cost-effective and scalable deposition method, enables precise control over AZO film properties, making it suitable for large-area coatings (Otieno, 2023).

This study examines the impact of Al doping (3%, 5%, 7%) on ZnO thin films synthesized via spray pyrolysis. XRD, UV-Vis spectroscopy, Hall effect measurements, and XPS are used to analyze structural, optical, and electrical properties. The goal is to optimize doping concentration for superior conductivity and transparency, maximizing ZnO-based solar cell efficiency.

Method

Al-doped ZnO (AZO) thin films were deposited via spray pyrolysis at different doping concentrations (3%, 5%, 7%). Zinc acetate dihydrate (0.15 M) was dissolved in 75 mL of doubly distilled water and stirred at room

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temperature. Aluminum nitrate nonahydrate was then added, and the solution was homogenized before deposition.

The deposition was performed at $375 \pm 10^\circ\text{C}$, with a solution flow rate of 1.75 mL/min and a nozzle-to-substrate distance of 28 cm to ensure uniform film formation. The estimated thickness values were 289 nm (ZnO), 281 nm (AlZO-3.00), 285 nm (AlZO-5.00), and 280 nm (AlZO-7.00), determined using the Seed Preprocessing Pattern Search (spPS) technique. The spray pyrolysis system used for the deposition process is schematically illustrated in Figure 1.

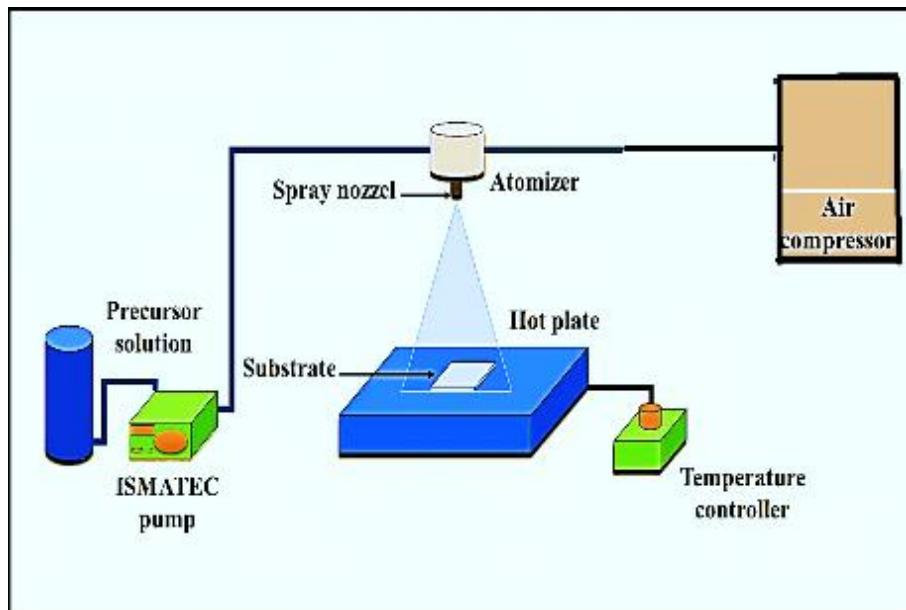


Figure 1. Schematic diagram of spray pyrolysis system used for the preparation of ZnO thin films.

The structural, morphological, optical, and electrical properties were analyzed using:

- X-ray Diffraction (XRD): Bruker D2 Phaser with $\text{CuK}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$) for crystalline structure and phase analysis.
- Scanning Electron Microscopy (SEM) & Energy-Dispersive X-ray Spectroscopy (EDX): Jeol JSM 5800 and IXRF Model 550i for surface morphology and elemental composition.
- Optical Transmittance: JASCO 570 UV-Vis-NIR spectrophotometer (200–2500 nm).
- Electrical Properties: Hall effect measurements (ECOPIA HMS-5000) for conductivity, carrier concentration, and mobility.
- X-ray Photoelectron Spectroscopy (XPS): Kratos Axis Ultra with $\text{Al K}\alpha$ (1486.6 eV) radiation, using C 1s at 284.5 eV for charge correction.

Results and Discussion

Structural Properties and Morphology

The X-ray diffraction (XRD) spectra of undoped and Al-doped ZnO thin films with different concentrations are illustrated in Figure 2. The XRD analysis confirms the *polycrystalline nature* of the films, with all peaks matching the *wurtzite ZnO structure* (JCPDS No. 36-1451). No secondary phases of aluminum or aluminum oxide were detected, indicating the *successful Al incorporation*. The films exhibit a *preferred (002) orientation*, with the *c-axis perpendicular* to the substrate, consistent with previous studies.

To determine the crystallite size (D) of the films, the Scherrer formula (Eq.1) is applied, which relies on measuring the full width at half maximum (FWHM) of the diffraction peaks b.

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

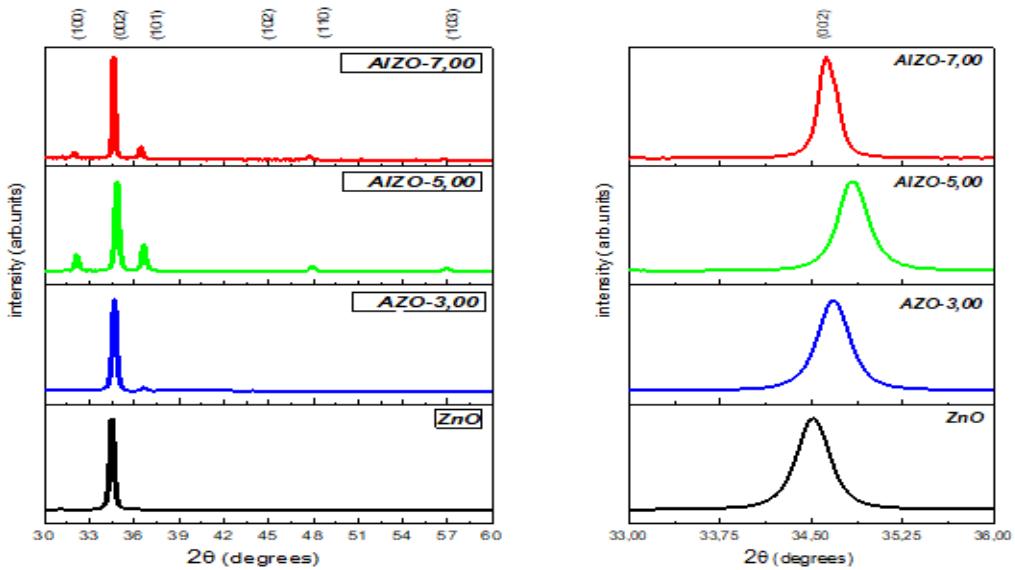


Figure 2. XRD of thin layers of ZnO and AlZO different concentrations of aluminum and Magnified and higher solution XRD pattern.

The chemical composition of pure ZnO and Al-doped ZnO thin films (3.00%, 5.00%, and 7.00%) deposited on glass substrates via the spray pyrolysis technique was analyzed using energy-dispersive X-ray spectroscopy (EDX), as shown in Figure 3.

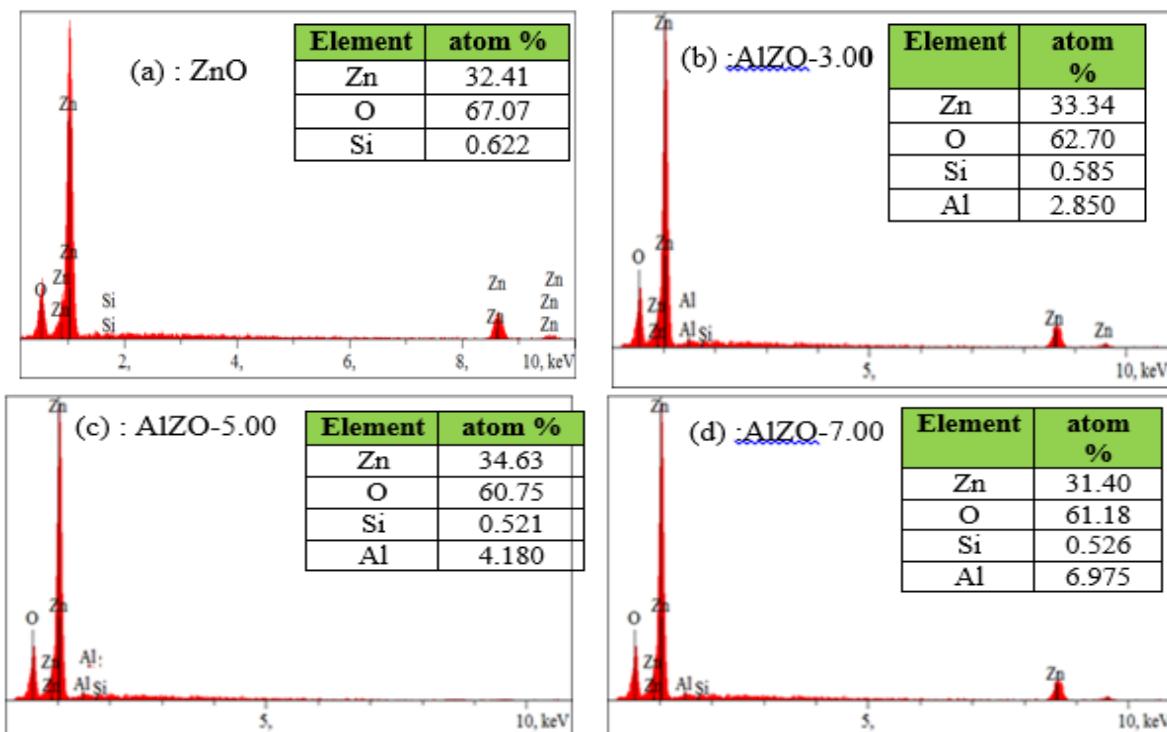


Figure 3. Energy dispersive X-ray (EDX) spectra of the obtained films, (a) ZnO, (b) AlZO-3.00, (c) AlZO-5.00 and (d) AlZO-7.00.

Fig.3a confirms the presence of Zn and O in pure ZnO films, while Fig.3b–3d validate the incorporation of Al in AlZO-3.00, AlZO-5.00, and AlZO-7.00 films. The absence of additional peaks supports the successful Al doping, consistent with XRD results. Si traces originate from the glass substrate. The atomic percentages in Fig.3 align with the intended doping levels, confirming the doping process's effectiveness. X-ray photoelectron spectroscopy (XPS) was performed to analyze the chemical states of elements present in the thin films. The XPS survey spectrum of the AlZO-5.00 sample is shown in Fig.4, revealing the presence of zinc (Zn), oxygen (O), and aluminum (Al).

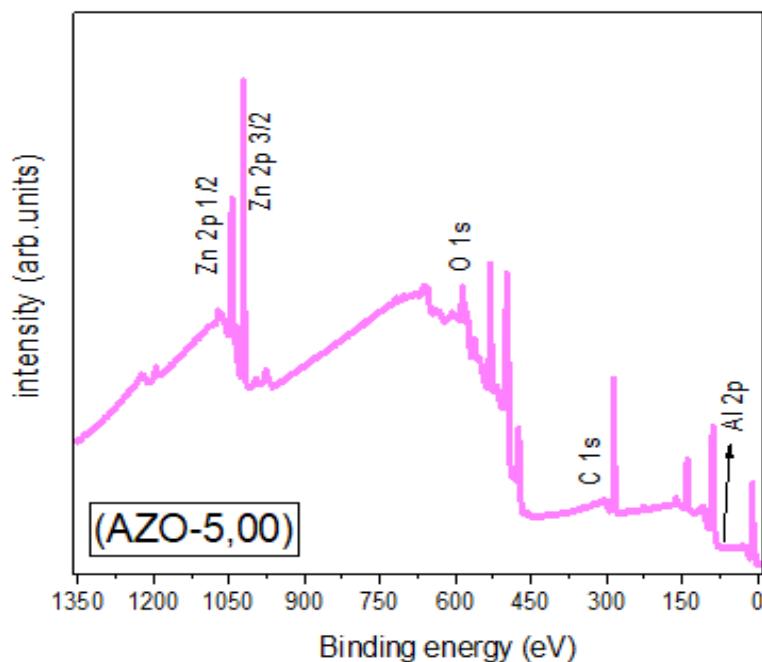


Figure 4. XPS full survey scans of AlZO-1.00 thin films.

Optical Properties

The optical transmittance spectra of ZnO, AlZO-3.00, AlZO-5.00, and AlZO-7.00 thin films are presented in Figure 5.

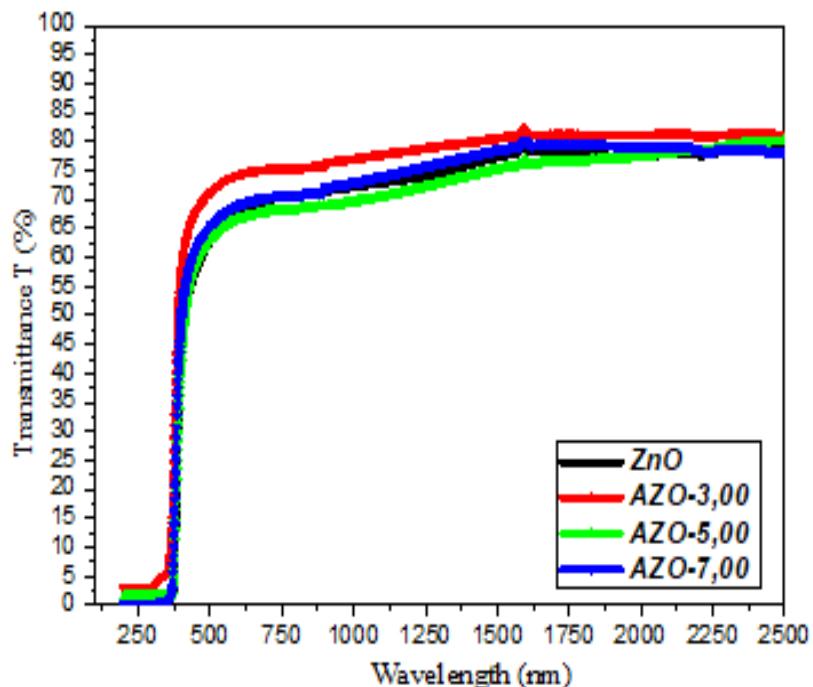


Figure 5. Transmittance spectra of the obtained films.

The curves indicate a clear increase in optical transparency with Al doping. The transmittance improves from 70% in the undoped ZnO film to 78% in the AlZO-3.00 thin film, reflecting an 8% enhancement in the visible range (380–550 nm). This improvement is due to the incorporation of Al, which alters the electronic structure and minimizes light scattering. ZnO is classified as a direct bandgap semiconductor, $n = 2$ is used in the optical bandgap calculations. $(\alpha h\nu)^2$ curves are plotted in Figure 6.

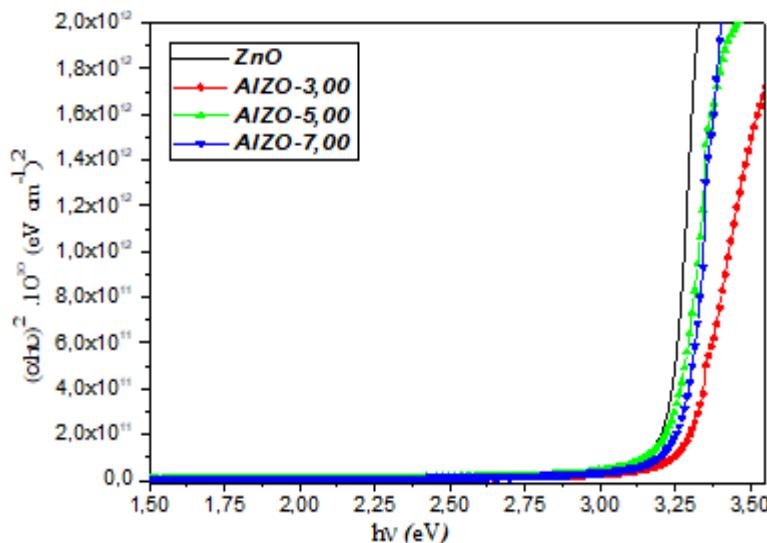


Figure 6. Variation of $(\alpha h\nu)^2$ versus energy of the obtained films.

As illustrated in Fig.7, the optical bandgap expands from 3.23 eV in the undoped ZnO film to 3.32 eV in the 3% Al-doped ZnO film. This increase is due to the Burstein-Moss effect, where a higher carrier concentration shifts the Fermi level, causing an apparent widening of the bandgap.

Electrical Properties

The electrical properties of Al-doped ZnO thin films were analyzed via Hall Effect measurements, with the results summarized in Table 3.

Table 3. Electrical parameters of ZnO and Al-doped ZnO thin films with different concentrations.

Doping level (wt.%)	Mobility ($\text{cm}^2/\text{V}\cdot\text{s}$)	Carrier concentration (cm^{-3})	Conductivity ($\Omega\cdot\text{cm}$) $^{-1}$
0% – ZnO	2.55×10^{-1}	-6.85×10^{16}	3.95×10^{-3}
3% – AlZO-3,00	5.89×10^{-1}	-4.02×10^{18}	3.37×10^{-1}
5% – AlZO-5,00	3.56×10^{-1}	-3.78×10^{18}	2.89×10^{-1}
7% – AlZO-7,00	4.78×10^{-1}	-1.60×10^{18}	2.98×10^{-2}

The conductivity and carrier concentration of ZnO and Al-doped ZnO thin films with varying doping concentrations are presented in Figure 7

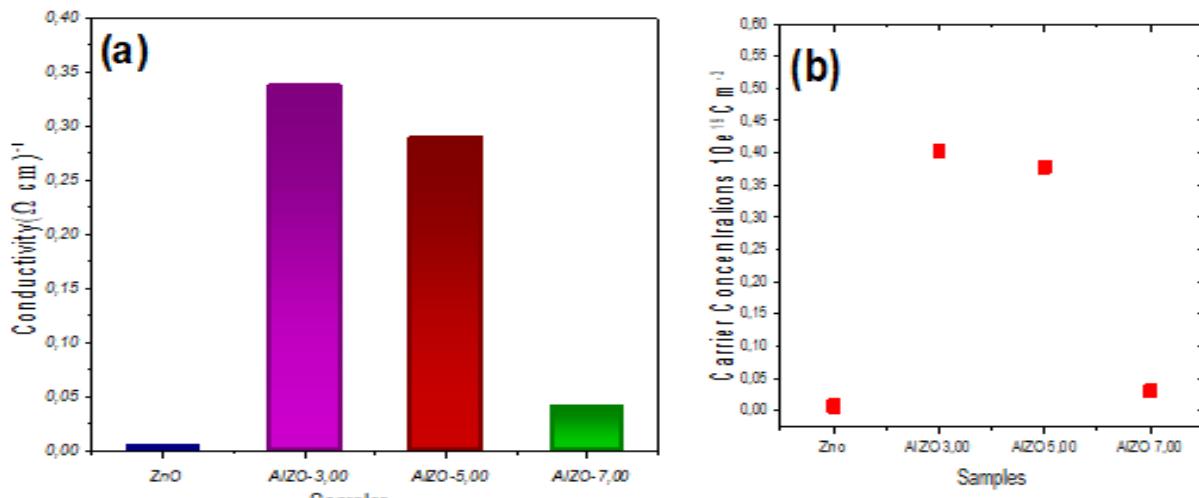


Figure 7. The functional properties of ZnO thin films

Conclusion

In this study, Al-doped ZnO (AZO) thin films were successfully synthesized on glass substrates using the spray pyrolysis technique. Structural analysis confirmed the formation of a polycrystalline wurtzite ZnO phase with strong (002) orientation, demonstrating effective aluminum incorporation without secondary phases. The crystallinity slightly improved at low doping concentrations, while excessive Al content induced lattice strain. Optical investigations revealed a significant increase in transmittance up to 78% and a widening of the optical bandgap from 3.23 eV to 3.32 eV due to the Burstein–Moss effect. Electrical measurements indicated that 3% Al doping yielded the highest conductivity and carrier concentration, whereas higher doping caused mobility reduction. These findings confirm that moderate Al doping effectively enhances both transparency and electrical performance. Therefore, AZO thin films are promising candidates to replace indium tin oxide (ITO) in low-cost, transparent electrodes for solar cells and optoelectronic applications. Future work will focus on optimizing deposition parameters and exploring other dopants to further improve film uniformity and long-term stability.

Scientific Ethics Declaration

* The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

Conflict of Interest

* The authors declare that they have no conflicts of interest

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