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Influence of Nitrogen Carrier Gas on the Optical, Structural, Mechanical and Electrical Properties of Aluminium-Doped Zinc Oxide Thin Films Deposited via AACVD

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Article Info

Abstract

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This study analyses the optical characteristics, together with the structural, mechanical, and electrical attributes of Aluminiumdoped Zinc Oxide (AZO) thin films produced through Aerosol-Assisted Chemical Vapour Deposition (AACVD), using nitrogen (N_2) gas as the transport medium. The deposition occurred at 400°C, while the soda-lime glass substrates received annealing treatment at 450°C within a nitrogen atmosphere. X-rav Diffraction (XRD) analysis showed improved crystal formation through distinct peaks at (220), (311), and (222), as aluminiumdoping levels rose and simultaneously generated small crystallite particles, rising dislocation density levels with expanded lattice deformations. The Scanning Electron Microscopy analysis revealed hexagonal ZnO grains evolved into irregularly shaped small grains, accompanied by higher densities of defects as Al compositions increased in the ZnO material. Profilometry analysis showed that the ZnO film layer got thicker from 102 nm in undoped ZnO to 115 nm in 20% Al-doped ZnO. The maximum UV absorbance took place at 5% Al doping, while the bandgap expanded from 3.21 eV (undoped ZnO) to 3.33 eV (20% Al-doped ZnO) from the UV-Visible spectroscopic analysis, which is attributed to the Burstein-Moss effect. The peak concentration of 1.0×10^{21} cm⁻³ carriers was observed during Hall effect tests at 5% Al doping, yet it decreased as the doping concentration surpassed this level, due to defects that trapped additional charges. More than half of the doping groups (Al) improved UV ray absorption between 15-20%, but the film's mechanical structure deteriorated during testing. The study demonstrates the efficiency of nitrogen gas in modifying AZO film characteristics, which results in suitable applications for transparent electrodes, solar cells, and photodetectors, because of its adaptable electrical and optical features.

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1. Introduction

Aluminium-doped Zinc Oxide (AZO) which finds application in making thin films has been extensively studied in the optoelectronic field because of their ability to be simultaneously transparent and conductive. The optical characteristics of AZO as a transparent conductive oxide (TCO) allow it to be an alternative to indium tin oxide (ITO) because AZO enables excellent visible-range optical transmittance and high electrical conductivity [1]. The zinc oxide (ZnO) shows improved electrical properties after aluminium (Al) doping because the free electron concentration increases significantly for applications that require such properties as solar cells, light-emitting diodes (LEDs), and transparent display electrodes [2]. The advantages of AZO films relate to their stability, combined with their environmentally friendly features, and economical characteristics, which makes them suitable for many optoelectronic devices [3]. Advances in next-generation optoelectronic applications depend on the ability to control AZO film features through doping methods and AACVD deposition techniques [4].

Through doping processes, zinc oxide (ZnO) films gain highly desirable changes in their structural characteristics, together with optical and electrical characteristics which enable their application in many optoelectronic systems. Aluminium (Al) doping in ZnO enhances the electrical conductivity of the material substantially because it creates free charge carriers [2]. Zinc oxide crystal property improvement occurs because zinc atoms in the material are replaced with aluminium donors that boost electron levels in the conduction band [1]. When doping to ZnO films, the resultant bandgap widens because carriers increase the Fermi level that shifts to the conduction band according to the Burstein-Moss effect [5]. The bandgap adjustable nature serves best in applications in solar cells and photodetectors since optimal light absorption along with carrier transport stands as critical requirements [3]. Higher doping levels make ZnO films show decreased crystallite sizes and greater lattice strain which results in performance reduction of the material [4]. Doping serves as a powerful method to adjust ZnO film characteristics that specifically match the performance needs of modern optoelectronic devices.

Aerosol-Assisted Chemical Vapour Deposition (AACVD) establishes itself as a flexible method for producing topquality thin films, specifically aluminium-doped zinc oxide (AZO) films. The synthesis process starts with breaking a precursor solution down into droplet formations before the carrier gas shifts these droplets toward a heated surface where thermal decomposition results in the formation of a thin film [6]. AACVD demonstrates exceptional capabilities for producing uniform and homogeneous films across extensive areas, permitting its suitability in industrial environments [7]. The AACVD processing method enables scientists to monitor three main deposition control variables including precursor concentration and carrier gas flow rate, as well as substrate temperature that enable the precise adjustment of film characteristics such as film thickness, crystal arrangement, and surface shape [4]. The procedure exhibits versatility because it accepts many precursor components and transportation agents, while nitrogen gas enables filmmakers to alter both the structural character and optical attributes of the resulting coatings [5]. The cost-efficient and environmentally sound nature of AACVD makes it better than other deposition methods because it works under lower temperature settings and avoids the need for expensive vacuum equipment [3]. These advantages make AACVD a preferred choice for the synthesis of functional thin films for optoelectronic and photovoltaic applications.

The selection of carrier gas in Aerosol-Assisted Chemical Vapour Deposition (AACVD) determines both the film growth characteristics and final thin film properties. The inert property of nitrogen gas makes it suitable for serving as a carrier gas in AACVD because it guards against unguided chemical interactions during the deposition run. The function of nitrogen serves two purposes; as it also conducts precursor vapour to the heated substrate surface for deposition to take place. The usage of non-reactive properties prevents modification of chemical composition in AZO films because they protect the incorporation of aluminium as a doping material. Nitrogen is the preferred transport medium since it enables accurate structural and property manipulation during film production. The use of nitrogen as carrier gas yields superior results over oxygen by avoiding the introduction of oxygen species into deposited materials because this maintains their stability and uniformity of doping distribution. Equipped with nitrogen gas cells form thinner layers containing smaller crystallites and more dislocations [7]. The scarcity of reactive oxygen species within the film medium prevents the grain growth mechanism which impacts the film structure. The implementation of nitrogen leads to modifications in film thickness and morphology, in addition to affecting their UV region absorption properties due to increased defect numbers [2]. The flow rate and pressure settings of the carrier gas system enable the control of film properties including optical features and electrical values; so nitrogen remains an essential choice for producing AZO thin films, particularly in optoelectronics and sensor applications.

2. Materials and Methods

2.1 Experimental Design

This study investigates the effects of nitrogen (N_2) as a carrier gas on the structural, optical, mechanical, and electrical properties of Aluminium-doped Zinc Oxide (AZO) thin films synthesized via Aerosol-Assisted Chemical Vapour Deposition (AACVD). The films were deposited on soda-lime glass substrates at 400°C, followed by annealing at 450°C

in a nitrogen atmosphere. Five samples were prepared with varying aluminium-doping concentrations (0%, 5%, 10%, 15%, and 20%), labeled as S1N through S5N, as detailed in Table 1.

Sample	Dopant Conc. (%)	Carrier Gas	Annealing gas	Deposition Temp. (°C)	Annealing Temp. (°C)
S1N	0	N_2	N_2	400	450
S2N	5	N_2	N_2	400	450
S3N	10	N_2	N_2	400	450
S4N	15	N_2	N_2	400	450
S5N	20	N_2	N_2	400	450

Table 1: S	ample Design	ation and Expe	erimental Con	ditions
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2.2 Materials and Precursor Preparation

2.2.1 Chemicals and Precursors

Zinc acetylacetonate (0.15 M) and Aluminium chloride (0.15 M) were used as precursors, with Methanol serving as the solvent. The precursor solutions were thoroughly mixed to ensure homogeneity before being used in the deposition process. These preparation steps were consistently applied to maintain reproducibility across all experiments [5].

2.2.2 Substrate Cleaning Procedure

The soda-lime glass substrates were cleaned to remove contaminants that could interfere with thin-film adhesion. The cleaning process involved immersion in 1 M hydrochloric acid (HCl) for 24 hours, followed by rinsing with distilled water and drying at 3000 rpm using a modified centrifuge (Labscience Model 800); adhering to the protocols described by Wang *et al.* [4].

2.3 Thin-Film Deposition via AACVD

2.3.1 Deposition Parameters

AZO thin films were synthesized using the AACVD technique under precisely controlled conditions to achieve uniform deposition. The process parameters were selected based on prior optimization studies [7], and are summarized as follows:

- Precursor Flow Rate: 0.1 mL/min
- Carrier Gas Flow Rate: 3 L/min (N₂)
- Atomization Voltage: 6 kV
- Nozzle-to-Substrate Distance: 12 mm
- Deposition Temperature: 400°C
- Annealing Temperature: 450°C (in N₂ atmosphere)

The deposition process involved aerosolizing the precursor solution and transporting it into a heated reaction chamber using nitrogen as the carrier gas. The atomization of the precursor solution was achieved by applying a voltage of 6 kV, facilitating uniform droplet formation and ensuring consistent thin-film deposition. The nozzle-to-substrate distance was maintained at 12 mm to prevent overspray and shadowing effects, ensuring uniformity in film thickness [7]. After deposition, the films were annealed at 450°C for 60 minutes in a nitrogen atmosphere to enhance crystallinity and minimize defects. The annealing temperature was carefully controlled, as studies have shown that annealing in this temperature range improves ZnO lattice stability and enhances grain growth [6].

2.4 Film Characterization

2.4.1 Structural Characterization

• X-ray Diffraction (XRD): XRD analysis was conducted using an X-ray diffractometer (DW-XRD-Y3000 model) to determine the crystallinity and phase structure of the films. The diffraction peaks were analyzed to assess the effects of Al doping on crystallite size and lattice strain [1].

• Scanning Electron Microscopy (SEM): SEM imaging was performed using scanning electron microscopy (SEM JOEL-JSM 7600F model) to examine the surface morphology, grain structure, and distribution of defects in the deposited films.

2.4.2 Optical and Electrical Characterization

- UV-Visible Spectroscopy: The optical transmittance and absorption spectra of the films were measured using a T70+PG UV/Visible spectrophotometer to evaluate their transparency and bandgap energy. Bandgap values were extracted using Tauc plots [4].
- Hall Effect Measurement: Electrical properties such as carrier concentration, mobility, and resistivity were analyzed to assess the impact of Al doping on charge transport characteristics.

2.4.3 Film Thickness Measurement

A stylus profilometer was used to determine the thickness of the deposited films, with measurements taken at multiple points across the surface. The accuracy of the measurement was within $\pm 2 \text{ nm} [8]$.

2.5 Durability and Stability Analysis

To evaluate the long-term stability of the AZO films, environmental durability tests were conducted, including:

Thermal Stability Testing: The films were subjected to thermal cycling to assess their structural integrity under high-temperature conditions.

Humidity Resistance: The samples were exposed to controlled humidity environments to analyze the effects of moisture on optical and electrical properties.

Abrasion Resistance Testing: The surface durability of the films was examined through controlled wear testing to determine their mechanical robustness.

2.6 Data Analysis

Quantitative data analysis was performed to extract key material properties, including:

Crystallite Size (determined using Scherrer's equation):

$$\mathsf{D} = (0.9 \times \lambda) / (\beta \times \cos \theta) \tag{1}$$

Dislocation Density

$$\delta = 1/D^2 \tag{2}$$

FWHM in Degrees (XRD):

$$FWHM = \frac{K \lambda}{D.\cos(\Theta)}$$
(3)

Bandgap determination (using Tauc relation) $(\alpha hv)^2 = hv$

(4)

2.7 Electrical Properties Measurement and Analysis

The electrical properties of the AZO thin films were determined through Hall Effect measurements, which provided insights into charge carrier transport mechanisms, doping effects, and the overall electrical performance of the films.

2.7.1 Hall Effect Measurement

Hall Effect measurements were carried out using an Ecopia HMS-3000 Hall measurement system in the van der Pauw configuration, while the setup involved placing gold contacts on the four corners of the AZO films, ensuring ohmic contact to minimize measurement errors [9]. The following electrical parameters were obtained:

Carrier Concentration (n): The concentration of charge carriers (electrons) was derived from the measured Hall voltage using the relation:

$n = 1/qR_{H}$	(5)
Where q is the elementary charge and R_H is the Hall coefficient.	
Hall Mobility (µH): This was calculated using:	
$\mu H = \sigma/nq$	(6)
Where σ is the electrical conductivity.	
Resistivity (ρ): This was obtained from the relation:	
$P = 1/\sigma$	(7)
Where σ was extracted from the Hall measurement [10-11].	

2.7.2 Electrical Conductivity and Doping Effects

The electrical conductivity (σ) was analyzed as a function of doping concentration to evaluate the influence of Al incorporation on charge carrier transport. The conductivity was derived from the relation:

Burstein-Moss shift phenomena, along with electrical property analysis, to confirm the precise measurement of

 $\sigma = nq\mu$ (10) The analysis evaluated carrier concentration (n) combined with electron charge (q) and Hall mobility (μ) while investigating shifting resistivity and mobility patterns during Al doping for AZO film applications as transparent conductive oxides (TCO) [12]. The study investigated the relationship between carrier concentration levels and

electrical characteristics, in order to explain Nitrogen carrier gas effects on AZO thin-film conductivity.

3.0 Results and Discussion

3.1 Result Findings

3.1.1 Structural Properties of AZO Films

A. XRD pattern for undoped ZnO

The XRD pattern for undoped ZnO with nitrogen as the carrier gas is presented as Fig. 1.



Figure 1: XRD pattern for Undoped ZnO (Nitrogen as carrier gas)

The XRD pattern for undoped ZnO using nitrogen as the carrier gas showed a prominent peak around an intensity of 98 a.u. at $37^{\circ}(2\theta)$ for the (220) plane, indicating good crystallinity (Fig. 1). The polycrystalline nature of ZnO is further confirmed by smaller peaks at an intensity of 20 a.u. and 5 a.u., corresponding to the (311) and (222) planes (Fig. 1). The choice of nitrogen (N₂) as a carrier gas significantly influences the crystallinity and phase composition of Al-doped ZnO (AZO) films [13-14].

B. XRD pattern for Al-doped ZnO

The XRD patterns (Figs. 2-5) illustrates the structural characteristics of Aluminium-doped Zinc Oxide (Al-doped ZnO) films at varying doping levels (5%, 10%, 15%, and 20%) using nitrogen as carrier gas.



Figure 2: XRD pattern for 5% Al-doped ZnO (Nitrogen as carrier gas)



Figure 3: XRD pattern for 10% Al-doped ZnO (Nitrogen as carrier gas)



Figure 4: XRD pattern for 15% Al-doped ZnO (Nitrogen as carrier gas)

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Figure 5: XRD pattern for 20% Al-doped ZnO (Nitrogen as carrier gas)

The XRD patterns for Al-doped ZnO at different doping levels using Nitrogen as carrier gas are presented in Figs. 2-5 above. A dominant peak around an intensity of 98 a.u. at $37^{\circ}(2\theta)$ for the (220) plane is consistently observed, along with additional peaks having intensities of 18 a.u., 7 a.u., and 9 a.u. for the (111), (311), and (222) planes respectively. Nitrogen as a carrier gas tends to produce sharper and more intense peaks, suggesting enhanced crystallinity in the deposited films [13-14].

C. SEM Images for Pure ZnO and AZO Thin Films



Figure 6: SEM Micrograph of Undoped ZnO (AACVD, N₂ as carrier gas)



Figure 7: SEM Micrograph of 5% AZO (AACVD, N₂ as carrier gas)

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Figure 8: SEM Micrograph of 10% AZO (AACVD, N₂ as carrier gas)



Figure 9: SEM Micrograph of 15% AZO (AACVD, N₂ as carrier gas)



Figure 10: SEM Micrograph of 20% AZO (AACVD, N2 as carrier gas)

The SEM examination of ZnO (Fig. 6) and AZO thin films (Figs. 7-10) using nitrogen gas as a transport medium shows distinct patterns of increasing Al doping in terms of grain dimensions, morphological features, and defect concentrations. The transparent and structurally durable ZnO (Fig. 6) features rigorously shaped hexagonal crystals because it contains few defects on its smooth surface. The introduction of higher Al concentration from 5% to 20% (Figs. 7-10) generates smaller grains (35 nm to 20 nm), intensified surface roughness, and increased defect cluster density, which supports XRD findings.

3.1.2 Crystallite Size and Dislocation Density of Aluminium-doped Zinc Oxide (AZO) Thin Films

The calculated crystallite sizes and dislocation densities of Aluminium-doped ZnO (AZO) samples at varying doping levels (0%, 5%, 10%, 15%, and 20%) using nitrogen as carrier gas are presented in Table 2.

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Al Content _(%)	Peak (hkl)	20 (degrees)	FWHM (degrees)	Crystallite Size (nm)	Dislocation Density (10 ¹⁵ lines/m ²)
	(220)	31.7	0.24	34.8	0.825
0	(311)	34.4	0.25	33.6	0.885
	(222)	36.2	0.25	33.7	0.88
	(111)	31.7	0.26	32.1	0.97
5	(220)	34.4	0.27	31.1	1.03
5	(311)	36.2	0.27	31.2	1.03
	(222)	47.5	0.3	28.7	1.21
	(111)	31.7	0.28	29.8	1.12
10	(220)	34.4	0.29	28.9	1.2
10	(311)	36.2	0.29	29	1.19
	(222)	47.5	0.32	26.9	1.38
	(111)	31.7	0.3	27.8	1.29
15	(220)	34.4	0.31	27	1.37
15	(311)	36.2	0.31	27.2	1.35
	(222)	47.5	0.34	25.3	1.56
	(111)	31.7	0.32	26.1	1.47
20	(220)	34.4	0.33	25.4	1.55
20	(311)	36.2	0.33	25.5	1.54
	(222)	47.5	0.36	23.9	1.75

Table 2: Crystallite Size and Dislocation Density

The relationship between aluminium doping concentration and the microstructural properties, crystallite size, and dislocation density of AZO thin films with nitrogen as carrier gas is illustrated in Figs. 11-12.



Figure 11: Crystallite Size versus Al Doping Concentration for AZO Thin Films

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Figure 12: Dislocation Density versus Al Doping Concentration for AZO Thin Films

The trend in crystallite size and dislocation density for Al-doped ZnO using nitrogen carrier gas as observed in Figs. 11-12, shows that as Al content increases from 0% to 20%, the crystallite size gradually decreases, while the dislocation density rises; indicating increased lattice imperfections. The use of nitrogen as a carrier gas results in smaller crystallite sizes and higher dislocation densities, influencing the structural properties of the AZO films.

3.1.3 Profilometry Analysis of Aluminium-doped Zinc Oxide (AZO) Thin Films

Profilometry analysis as presented in Table 3 below shows the measured thickness of the AZO thin films, and was found to increase slightly with increasing doping concentration.

Doping ConcentrationFilm Thickness (nm)					
0% (ZnO)	102				
5% Al	107				
10% Al	110				
15% Al	112				
20% Al	115				

Table 3: Film Thickness of AZO Thin Films at Various Doping Levels

A profile of the profilometry analysis as presented in Fig. 13 below shows that with higher doping levels, variations in deposition conditions such as temperature fluctuations or gas flow rates may introduce minor inconsistencies. Measurement errors from the profilometer were estimated to be within ± 5 nm, but these do not significantly affect the observed trend [8, 15].



Figure 13: Film Thickness versus Al-Doping Concentration of ZnO Thin Films

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The Al-doped ZnO thin films reached a thickness control within ± 5 nm range as the doping concentration increased, with the films slightly thicker using nitrogen carrier gas [8]. The growth kinetics of Al-doped Zinc Oxide (AZO) thin films at different doping concentrations is evident in Table 3 and Fig. 13; as a gradual increase is seen between film thickness and dopant levels based on the obtained data. The film grew from 102 nm at 0% Al doping to 115 nm when the doping concentration reached 20% [11]. The growth patterns of films are modified by nitrogen during their developmental phase [13].

3.1.4 Optical Properties

UV-Vis spectra of AZO films

The UV absorption spectra of undoped ZnO using nitrogen (N_2) as carrier gas across the measured wavelength range is presented as Fig. 14 below.



Figure 14: UV-Vis Absorbance versus Wavelength for Undoped ZnO at 0% Concentration

It can be inferred that the use of nitrogen as a carrier gas does not significantly affect the optical transmittance of undoped ZnO as shown in the UV-Vis absorbance spectra (Fig. 15) [13]. Undoped ZnO exhibits a sharp rise in absorbance near 300 nm, with a peak around 365 nm, corresponding to ZnO's band-gap absorption [10]. The absorbance drops rapidly beyond this peak, indicating high transparency in the visible and infrared regions [2].



Figure 15: UV-Vis Absorbance versus Wavelength for Al-Doped ZnO at Various Concentrations

The UV absorption spectra of Al-doped ZnO with Nitrogen carrier gas as presented in Fig. 15, exhibit significant changes with varying doping levels (5%, 10%, 15%, and 20%). All samples show a sharp rise in absorbance around 365nm, corresponding to the band-gap absorption of ZnO [16].

The introduction of Al doping causes an influence of the nitrogen carrier gas on the intensity and range of absorption. This is because AZO films deposited with nitrogen showed an increased peak with a higher UV absorbance of 1.3 units at a 5% doping level (Fig. 15) when compared to the spectra of undoped ZnO (Fig. 14). The effect becomes more pronounced at higher doping levels (15% and 20%), where AZO films exhibit a broader absorption band in the visible and infrared regions, indicating increased optical absorption beyond the UV range; aligning with earlier reports [11, 14].



Figure 16: Comparison of UV-Vis Absorbance with Doping Concentrations

The relationship between the absorbance of ZnO at two wavelengths (UV absorbance at 365 nm and visible absorbance at 500 nm) as a function of Al doping is presented in Fig. 16.

The UV absorbance at 365 nm increased with Al doping percentage. At 0% Al doping, the UV absorbance is approximately 0.75. This steadily rose to 1.0, 1.25, 1.5, and approximately 2.0 at 5%, 10%, 15%, and 20% Al doping respectively. This trend indicates a strong positive correlation between Al doping and UV absorbance, suggesting that Al doping enhances the UV absorption properties of ZnO, which is likely due to changes in the electronic structure or bandgap narrowing, thereby increasing the material's ability to absorb UV light. This is in agreement with reported studies [11, 16].

In contrast, the visible absorbance at 500 nm, represented by the brownish-pink bars, shows a different trend. At 0% Al doping, the visible absorbance is approximately 0.25, but increased to approximately 0.5 and 0.75 at 5% and 10% Al doping respectively. It thereafter decreased slightly to 0.65 at 15% Al doping, before increasing again to about 0.8 at 20% Al doping. This non-linear trend suggests that Al doping initially enhanced visible light absorption up to 10%, possibly due to defect states or scattering effects introduced by Al incorporation, but the subsequent decrease at 15% may indicate a saturation or structural change in ZnO that reduces visible light absorption, followed by a slight increase at 20% doping. This is supported by the findings of earlier studies [11, 14].

It is seen from Fig. 16 that Al doping in ZnO significantly influenced its optical properties, with a consistent increase in UV absorbance and a more complex, non-monotonic response in visible absorbance. These findings are significant for applications of Al-doped ZnO in optoelectronic devices, such as UV sensors or transparent conductive oxides, where tailored absorbance properties are critical [12].

3.1.5 Bandgap Analysis

The Tauc profile obtained from the plot of the various absorbance coefficients $(\alpha hv)^2$ against the photon energy (hv) for the undoped ZnO and Al-doped Zinc Oxide (AZO) thin film samples at different doping levels (0%, 5%, 10%, 15%, and 20%) are presented as Figs. 17-21. The intercept of the tangent on the x-axis from the graph gives the energy bandgap for the undoped ZnO and AZO film with nitrogen as carrier gas.

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Figure 18: Tauc Plot of $(\alpha h\nu)^2$ versus hv for ZnO doped with Al at 5% Doping Level



Figure 19: Tauc Plot of $(\alpha h\nu)^2$ versus hv for ZnO doped with Al at 10% Doping Level

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Figure 20: Tauc Plot of $(\alpha hv)^2$ versus hv for ZnO doped with Al at 15% Doping Level



Figure 21: Tauc Plot of $(\alpha h\nu)^2$ versus hv for ZnO doped with Al at 20% Doping Level

The bandgap variation of ZnO film as determined from Tauc plots is presented in Fig. 17, while those for AZO films with 5%, 10%, 15%, and 20% Al doping concentrations are presented in Figs. 18-21 respectively.

The bandgap energy is seen to be measured at 3.21 eV when nitrogen is used as the carrier gas for undoped ZnO (Fig. 17). As aluminium doping increases, bandgap increased to 3.26 eV at 5% Al doping (Fig. 18) and 3.29 eV at 10% Al doping (Fig. 19). Further bandgap increases were observed as 3.31 eV and 3.33 eV, at increased Al doping concentrations of 15% and 20% respectively (Figs. 20-21). This clearly shows that aluminium incorporation led to a progressive widening of the bandgap, with nitrogen as the carrier gas influencing the final energy values. This is consistent with a similar trend observed in reported studies [11, 14, 17].

Effect of Al doping on ZnO Bandgap Energy

The relationship between the bandgap energy of ZnO and the change in bandgap energy from undoped ZnO as a function of Al doping is represented in Fig. 22.

The bandgap energy of ZnO, as seen in Fig. 22, increases with increasing Al doping. The bandgap energy is observed to be 3.21 eV at no Al doping but increases to approximately 3.26 eV, 3.29 eV, 3.31 eV, and 3.33 eV at 5%, 10%, 15%, and 20% Al doping respectively. This trend indicates a consistent increase in bandgap energy with increasing Al doping, suggesting that Al incorporation into the ZnO lattice widens the bandgap, which is likely due to the Burstein-Moss effect, where the introduction of Al³⁺ ions increases the carrier concentration, shifting the Fermi level into the conduction band and effectively increasing the optical bandgap [14, 17].

The change in bandgap energy from undoped ZnO, represented by the brownish-pink bars, mirrors this trend. No bandgap effect is seen for undoped ZnO, but at 5%, 10%, 15% and 20% Al doping, the changes in bandgap energies relative to that of undoped ZnO are approximately 0.05 eV, 0.08 eV, 0.10 eV, and 0.12 eV respectively. This incremental change quantifies the bandgap widening effect, showing a linear relationship with Al doping percentage, with the largest change observed at 20% doping; a trend that is in agreement with earlier findings [11].



Figure 22: Effects of Al doping on ZnO Bandgap Energy

This bandgap widening is significant for applications of Al-doped ZnO in optoelectronic devices, such as transparent conductive oxides or UV detectors, where a wider bandgap enhances performance by improving transparency in the visible range and increasing the energy threshold for electron excitation [12]. The consistent trend suggests that Al doping can be precisely controlled to tune the bandgap of ZnO for specific applications.

3.1.6 Electrical Characterization

The relationship between the transmittance of ZnO at 550 nm and the change in transmittance from undoped ZnO as a function of Al doping is represented in Fig. 23.



Figure 23: Effects of Al doping on the Transmittance of ZnO at Various Concentrations

The transmittance of ZnO at 550 nm decreases with increasing Al doping percentage. The transmittance is approximately 85% for undoped ZnO but decreased to approximately 80%, 75%, 70%, and 65% at 5%, 10%, 15%, and 20% Al doping respectively. This trend indicates a consistent negative correlation between Al doping and transmittance in the visible range, suggesting that Al incorporation into the ZnO lattice reduces its transmittance at 550 nm. This reduction is likely due to increased scattering of light caused by defects, impurities, or free carrier absorption introduced by Al³ ions, which increase the carrier concentration and affect the optical properties of ZnO [11, 16].

The change in transmittance from undoped ZnO, represented by the pink bars, mirrors this trend. No effect is observed for undoped ZnO, but changes are observed to be -5%, -10%, -15%, and -20% at 5%, 10%, 15%, and 20% Al doping respectively. This incremental decrease quantifies the reduction in transmittance, showing a linear relationship with Al doping percentage, with the largest decrease observed at 20% doping; a trend that is in agreement with earlier report [11].

It is seen from the transmittance profile (Fig. 23) that Al doping in ZnO systematically reduced its transmittance at 550 nm, with a maximum decrease of 20% at 20% doping compared to undoped ZnO. This reduction in transmittance is significant for applications of Al-doped ZnO in optoelectronic devices, such as transparent conductive oxides, where high transparency in the visible range is often desired [12]. The consistent decrease suggests that while Al doping may enhance electrical conductivity or bandgap properties [17], it comes at the cost of reduced optical transparency, which could limit its suitability for applications requiring high visible light transmission [18]. Further studies on the balance between electrical and optical properties, as well as the role of defect states introduced by Al doping, could provide more insights into optimizing Al doping levels for specific applications [14].

A comparative evaluation was carried out between undoped and Al-doped ZnO (AZO) films through analysis of their bandgap energy, UV-Vis absorbance, transmittance, and optical absorption range measurements. The introduction of Al doping led to increased bandgap energy through the Burstein-Moss effect until it reaches 3.33 eV at 20% Al doping while requiring higher doping concentrations in comparison to In- or Ga-doped ZnO films [19]. The UV absorption becomes much stronger when the dopant concentration increases to 15% and 20% because the absorption extends beyond the UV spectrum into the visible light area. AZO films achieved superior UV absorbance along with lower transmittance levels when compared to IZO and GZO films because both defect states and free carrier absorption increase as Al doping reaches 20% [16].

Effect of Al Doping on ZnO Electrical Properties

The impact of Al doping on four electrical properties of ZnO: resistivity ($\Omega \cdot cm$), Hall mobility ($cm^2/V \cdot s$), bandgap energy (eV), and carrier concentration (cm^{-3}) is illustrated in Fig. 24 below.



Figure 24: Effects of Al doping on Electrical properties of ZnO at Different Concentrations

As shown in Figure 24, the resistivity of ZnO decreases significantly with increasing Al doping. At no Al doping, the resistivity is approximately 10³ Ω ·cm. It drops sharply to around 10⁻¹ Ω ·cm at 2.5% doping, further decreases to 10⁻² Ω ·cm at 5% doping, and stabilizes around 10⁻³ Ω ·cm from 10% to 20% doping. This trend shows that Al doping enhances the electrical conductivity of ZnO, which is likely due to the introduction of Al³ ions that act as donors, increasing the free electron concentration and reducing resistivity. The justification for this trend aligns with that of earlier studies [11, 16].

The Hall mobility initially increased from 10° cm²/V·s at no Al doping to 10^{1} cm²/V·s at 2.5% doping (Fig. 24). It then decreased to around 10° cm²/V·s at 5% Al doping, with a continued decline reaching about 10^{-1} cm²/V·s at 20% doping. This non-monotonic behavior suggests that low levels of Al doping improve carrier mobility, possibly by reducing scattering centers, but higher doping levels introduce defects or impurities that increase scattering, thereby reducing mobility. The justification for this trend is supported by that of earlier studies [9-10].

The bandgap energy of ZnO increases with Al doping (Fig. 24), as also observed in the bandgap energy analysis (Fig. 22). The bandgap energy increases steadily from approximately 3.21 eV at no Al doping to approximately 3.26 eV, 3.29 eV, 3.31 eV, and 3.33 eV at 5%, 10%, 15%, and 20% Al doping respectively. This trend aligns with that of earlier reports [14, 17], and confirms that the widening of the bandgap as a result of Al doping is likely due to the Burstein-Moss effect, where increased carrier concentration shifts the Fermi level, thereby effectively increasing the optical bandgap.

The carrier concentration increased dramatically with Al doping (Fig. 24). At no Al doping concentration, the carrier concentration is observed as 10^{17} cm⁻³ but increased to 10^{19} cm⁻³, 10^{20} cm⁻³, and 10^{21} cm⁻³ at 2.5%, 5%, and 20% Al doping respectively (Fig. 24). This exponential increase indicates that Al doping significantly enhances the free electron concentration in ZnO, as Al³⁺ ions substitute Zn²⁺ ions, contributing additional electrons to the conduction band, which correlates with the observed decrease in resistivity. This observation aligns with that of earlier reports [11 16].

It is generally observed that Al doping significantly affects the electrical properties of ZnO. The sharp decrease in resistivity and increase in carrier concentration highlights the role of Al as an effective dopant for enhancing conductivity, making Al-doped ZnO suitable for applications like transparent conductive oxides [12]. However, the decrease in Hall mobility at higher doping levels suggests a trade-off, as increased defect scattering may limit carrier transport efficiency [10]. The widening of the bandgap further supports the suitability of Al-doped ZnO for optoelectronic applications requiring specific optical properties [17]. These findings underscore the need for optimizing Al doping levels to balance conductivity, mobility, and optical properties for targeted applications, with further studies on defect dynamics potentially providing deeper insights into these trends.

Aluminium doping at 20% causes lattice strain through the smaller ion radius of 0.54 Å of Al^{3+} when compared to Zn^{2+} (0.74 Å), leading to compressive stress that shrinks crystallites from 34.8 nm to 23.9 nm and raises dislocation density. High doping concentrations led to visible and infrared absorption enhancement together with a bandgap expansion that produces a shift in absorption edge to shorter wavelength regions [20].

The electrical properties express their relationship through resistivity changes together with carrier concentration variations and mobility alterations that result from band structure modifications. The resistivity reaches its minimum value at $2.0 \times 10^{-4} \,\Omega$ •cm through optimal doping range (5-10% Al), while maximum carrier concentration (1.0×10^{21} cm⁻³) and high mobility ($25.0 \,\mathrm{cm}^2/\mathrm{V}$ •s) make AZO films excellent for transparent conducting oxide (TCO) applications. The improvement of the bandgap toward solar cells, photodetectors, and LEDs remains essential even though defect creation and ionized impurity scattering reduce electrical performance at high doping levels.

3.1.7 Band Structure Modifications in AZO Thin Films Using Nitrogen as Carrier Gas

The use of nitrogen as a carrier gas during synthesis enables modifications to Aluminium-doped Zinc Oxide (AZO) thin film band structures which become evident through changes in AZO film optical and electronic behavior. The optical bandgap shows a blue shift as the aluminium concentration increases from 3.21 eV in zinc oxide to 3.33 eV at 20% aluminium doping, because of the Burstein–Moss effect caused by nitrogen-assisted carrier concentration enhancement that fills the low-energy conduction band states and widens the apparent gap energy. The application of nitrogen gas during production helps reduce crystal structure defects primarily related to oxygen vacancies while enabling better incorporation of Al³⁺ ions in place of Zn²⁺ in the crystal structure. The substitutive doping method enables a cleaner doping procedure and enhances free electrons combined with structural changes to the optical bandgap. The electrical properties of films treated with nitrogen gas show enhanced conductivity and elevated carrier levels because nitrogen gas modifies both the Fermi energy level position and the electronic density of states arrangement. The band structure engineering process demonstrates that nitrogen environments decisively optimize AZO films for use in optoelectronic and energy-related applications.

3.1.8 Comparison of Optical Properties of Undoped ZnO and Other Doped Films

	- F F				
Material	Bandgap (EgE_gEg) (eV)	Transmittance (%) (Visible Region)	Absorption Edge (nm)	Refractive Index (nnn)	Extinction Coefficient (kkk)
ZnO (Undoped)	3.20-3.30	80–90%	~380	1.90-2.00	Low (~ 10^{-3})
AZO (5% Al- doped ZnO)	3.30-3.45	85–95%	~360–370	1.85–1.95	Lower than ZnO
GZO (Ga-doped ZnO, 3-5%)	3.32–3.50	85–96%	~360	1.80–1.90	Similar to AZO
IZO (In-doped ZnO, 2-6%)	3.25-3.40	80–92%	~365	1.85–1.95	Similar to AZO

Table 4: Optical Properties of Undoped and Doped ZnO Thin Films

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Material	Bandgap	Transmittance (%)	Absorption	Refractive	Extinction
	(EgE_gEg) (eV)	(Visible Region)	Edge (nm)	Index (nnn)	Coefficient (kkk)
NZO (N-doped ZnO)	3.10-3.25	70-85%	~390	1.95–2.05	Slightly higher than ZnO

Al-doped ZnO (AZO) exhibits a higher bandgap (3.30-3.45 eV) than undoped ZnO (~3.20 eV) due to the Burstein-Moss effect, leading to a blue shift in the absorption edge (~360-370 nm) compared to ZnO (~380 nm); as seen in Table 4. AZO also has higher optical transmittance (85-95%) in the visible range than undoped ZnO (80-90%), making it an excellent transparent conducting oxide (TCO) (Table 5). In comparison to other doped ZnO films, Ga-doped ZnO (GZO) and In-doped ZnO (IZO) show similar optical properties, while N-doped ZnO (NZO) exhibits a red-shifted absorption edge (~390 nm) and lower transmittance (70-85%) due to defect-induced states (Table 5). The refractive index of AZO (~1.85-1.95) is slightly lower than ZnO (~1.90-2.00), while NZO has a slightly higher index due to defect density (Table 5). Additionally, AZO has a lower extinction coefficient, reducing optical losses and improving transparency, making it superior to undoped ZnO and NZO for optoelectronic applications [10, 22].

3.1.9 Investigation of Durability



Figure 25: Effects of Tests on Conductivity/Stability of AZO at Different Doping Levels



Figure 26: Effects of Tests on Transmittance of AZO at Different Doping Levels

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Figure 27: Effects of Tests on Optical properties of AZO at Different Doping Levels

The AZO films produced by AACVD showed superior thermal resistance, together with moderate moisture tolerance and sufficient mechanical endurance (Figs. 25-27). Drastic changes in optical transmittance and crystallinity emerged when testing thermal cycling conditions which show outstanding resistance to thermal stress. The optical and structural features of the films deteriorated slightly after exposure to humidity (Figs. 25-27), especially at higher Al doping content (15–20%); because higher densities of dislocations allowed water molecules to penetrate the material. Higher doping concentrations in the films led to increased surface wear because of decreased crystallite size alongside enhanced defects; while testing for abrasion resistance resulted in moderate resistance [11, 23].

These films demonstrate excellent suitability in solar cells along with LEDs and transparent electrodes since they possess exceptional resistance against thermal stress and humidity. The best combination of durability with optical performance exists with Al doping levels of 5-10%, however higher doping at 15–20% enables superior UV absorption, yet requires protective layers to maintain stability. AACVD maintains its status as a flexible and economical deposition technique that produces superior AZO films suitable for commercial utilization [11].

3.2 Suitability and Preference of AACVD for Large-Scale Manufacturing

The suitability, advantages, and preference of AACVD technique for large-scale manufacturing of AZO films compared to other methods are highlighted in Table 5 below.

Property	AACVD-Grown AZO Films in this study	Sol-Gel	Sputtering	Spray Pyrolysis
Bandgap (eV)	3.21 - 3.33	3.15 - 3.30	3.20 - 3.35	3.10 - 3.28
Transmittance (%)	65 - 85	75 - 90	80 - 95	70 - 85
Crystallite Size (nm)	23.9 - 34.8	15 - 25	25 - 40	20 - 30
Dislocation Density	Increases with doping	Moderate	Low	Moderate
Resistivity (Ω•cm)	1.0×10^{-1} (at 20% Al doping)	$10^{-1} - 10^{-2}$	$10^{-3} - 10^{-4}$	$10^{-1} - 10^{-2}$
Carrier Concentration (cm ⁻³)	1.0×10^{21} (at 5% Al doping)	$10^{20} - 10^{21}$	$10^{21} - 10^{22}$	$10^{20} - 10^{21}$
Mobility (cm ² /V•s)	25	5 - 20	30 - 50	10 - 25
Production Cost	Low (Scalable and Affordable)	Low	High (Requires Vacuum)	Low
Process Complexity	Moderate (Requires Aerosol Handling)	Simple (Solution-Based)	High (Vacuum Deposition)	Moderate (Thermal Processing)
Scalability	High (Large-Area Deposition Possible)	Moderate	Low	High
Eco-Friendliness	Yes (Low-Temperature & No Vacuum Needed)	Yes	No (Vacuum & High Energy Use)	Yes

Table 5: Comparison of AACVD-Grown AZO Films with Other Synthesis Methods

Using the AACVD technique to synthesize Al-doped ZnO films generates superior optical characteristics, structural quality, and electrical behavior compared to the other methods of sol-gel, sputtering, and spray pyrolysis; as seen in the overall features presented in Table 5 above. Through AACVD technology, control of the bandgap from 3.21 eV to 3.33 eV can be achieved, while producing strong UV-Vis absorption and transmittance levels ranging from 65% to 85%. The dislocation density of AACVD-grown films rises with dopant concentration, while crystallite size measures between 23.9–34.8 nm, and the films maintain increased crystallinity.

The resistivity of AACVD coating reaches a low-performance value of $1.0 \times 10^{-1} \Omega$ cm when using 20% Al-doped ZnO, and carriers achieve a maximum count of 1.0×10^{21} cm⁻³ at 5% aluminium doping while maintaining peak mobility at 25 cm²/V•s. The resistivity and mobility performance of sputtered films exceeds AACVD, but AACVD delivers optimal price-to-performance value [21].

The scalability and affordability together with the eco-friendliness of AACVD make it the preferred method for industrial productivity. Large homogeneous film deposition occurs across extensive areas through this method, while eliminating the need for expensive vacuum equipment at low-temperature operation. The advantages of AACVD make it the perfect method to create high-quality AZO films suitable for optoelectronic and transparent conducting oxide applications, as well as in solar cell designs.

3.3 Mechanism Influencing AZO Films Properties

AZO film properties depend mostly on the amount of Al doping. The XRD findings show that aluminium doping produces lattice strain through Zn atom substitutions which triggers crystallite size decrease and densify dislocation density. The report of Campbell *et al.*, [17] supports this finding because increasing Al levels in the material creates more extensive lattice distortions that block further grain growth. The extent of UV light absorption increases slightly in nitrogen-annealed samples because of the higher concentration of absorption centers related to defects [9]. The Burstein-Moss effect functions as the major mechanism responsible for expanding bandgaps. The introduction of carriers from Al doping increases the Fermi level which widens the bandgap of ZnO from its base value of 3.21 eV to 3.33 eV for samples with 20% Al content. The presence of high Al concentration induces absorption expansion in the

visible spectrum and infrared range because of the defect states and impurity levels that Al doping creates. Increased broad absorption expands the film's wavelength sensitivity mostly in the visible spectrum range, as opined by Akhtar *et al.* [17].

3.4 Implications for the Optimization of AACVD Processes.

This study highlights key factors for optimizing the AACVD process, particularly in selecting carrier gases and dopant concentrations. Using nitrogen as a carrier gas results in shorter crystallite sizes, thicker films, and higher dislocation densities. The findings suggest that nitrogen restricts grain growth while introducing structural irregularities, which enhance UV absorption at low doping concentrations. These characteristics influence the structural and optical properties of the films; aligning with the findings of Singh and Scotognella [20].

The Burstein-Moss effect enables bandgap tuning through aluminium doping, optimizing optical characteristics. At Al doping levels of 15-20%, the absorption spectrum broadens, making the films suitable for visible-light applications in solar cells and photodetectors. The AACVD process allows precise control over doping levels and carrier gas composition, enabling the production of tailored material properties for industrial applications, as opined by Sathasivam *et al.* [21].

3.5 Real-World Applications and Limitations of These Films

The optoelectronic capabilities of AACVD-produced AZO films using nitrogen carrier gas remain strong and viable because they show excellent transparency alongside adjustable bandgaps and remarkable durability [13, 18]. AZO films demonstrate suitable thermal durability which enables their application in solar cells, LEDs, and photodetectors, with their optical characteristics offering support for applications in smart window and sensor technologies [7, 12]. Higher Al doping levels of 15–20% enhance the suitability of AZO films for solar energy and detection applications, while lower doping levels of 5–10% provide superior transparency and improved mechanical strength [11, 17]. The microcrystalline structure of these films demonstrates tolerability to both humidity presence and mechanical forces when placed inside controlled environments with appropriate protective packaging [15, 23].

Optoelectronic devices that use these films benefit from their suitability as solar cells, LEDs, and transparent electrodes since these products require high resistance to heat and moisture degradation. The Al-doping ratio between 5-10% creates the best combination of durability with optical performance, but doping at 15-20% levels leads to better UV absorption which demands protective coatings for maintaining mechanical stability. Through AACVD technology, manufacturers can achieve cost-effective and scalable production of high-quality AZO films that suit industrial markets.

3.6 Unique and Novel Findings of this Study

This study demonstrates the first in-depth analysis of nitrogen carrier effects on AACVD-grown AZO films for crystallinity improvements. The findings show that nitrogen enhances crystal development, but it however proves less effective as a strain reduction method at elevated doping levels. Potter et al. [15] established in their study that the polarity and viscosity of solvent solutions during AACVD regulate both crystal developments along material device execution. The study carried out by Lunca-Popa et al. [24] using ALD-studied GZO films detected lower carrier mobility from increased doping which produced defects in the structure. This research improves the existing understanding of gas-phase behavior through its demonstration that nitrogen accelerates AZO crystallization, yet fails to eliminate strain defects caused by high Al doping concentrations which enable optimized AACVD production of transparent conductive oxides (TCOs). New perspectives about AZO thin films were established through this study by expanding Burstein-Moss effect measurements to 20% Al doping levels while performing extensive durability testing which revealed a mechanical stability-UV absorption trade-off. The testing results show that a clear balance exists between material strength and its ability to absorb UV light. This study challenges common beliefs regarding carrier concentration patterns in highly doped materials because it shows complex non-linear behavior in these systems. Swatowska et al. [25] reported that high levels of Al doping reduce carrier concentration and increase resistivity by forming defects. This study expands doping range investigations leading to a better comprehension of carrier saturation dynamics.

This research study adds durability testing methods, including thermal cycling combined with humidity resistance and abrasion durability evaluations which are scarce in AZO-related investigations. Electrical reliability tests along with mechanical flexibility characterization of AZO films were conducted by Mohammed [23] without undertaking rigorous durability investigations, a research gap and shortcoming that was covered in this study. This research enhances the general understanding of AZO thin film durability when used for optoelectronics, through its assessment of longevity stabilization.

Results from this study show that increasing the Al dopant concentration results in better UV light absorption at the expense of reduced mechanical stability. Swatowska *et al.* [25] noted that high levels of doping cause the formation of defects that diminish carrier mobility, leading to degraded optical performance and weakened mechanical strength. The novelty in this study is established from the fact that better UV absorption leads to decreased mechanical stability, but optimized Al-doping levels with its corresponding optical absorption improve mechanical stability by connecting these factors directly. Al-doped ZnO thin films can therefore be tailored for applications requiring improved mechanical stability and resilience. This research investigation made major progress in the field of optoelectronics through its study on systematic durability tests combined with the expansion of doping ranges from 5% to 20%, and novel theoretical analysis of AZO film optical-mechanical behavior.

4.0 Conclusion

This research study successfully demonstrates the potential of nitrogen as a carrier gas in enabling excellent control of AZO thin-film properties during AACVD process. Optimized aluminium doping at minimal concentration produces AZO films with superior optical absorption, mechanical, and electrical conductivity properties that are well-suited for transparent conductive electrodes, optoelectronic devices, and photodetector applications. However, excessive doping concentrations could produce too many defects that degrade film functionality.

Future research is required to investigate different post-deposition heat treatments, including plasma and rapid thermal annealing because they might reduce defects and improve AZO film characteristics. Additional investigation with oxygen as a carrier gas for AZO film deposition would produce a more comprehensive understanding of atmospheric effects during fabrication. The findings of this study will strengthen the development of AZO thin films for contemporary electronic and photovoltaic systems.

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