

Optical and Electrical Characterization of ZnO:Ga₂O₃ Thin Films Synthesized by MASP for Oxygen Gas Sensor Applications

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Abstract

This study investigates the effect of oxygen (O₂) exposure on the optical and electrical properties of ZnO:Ga₂O₃ thin films synthesized by the Microwave-Assisted Spray Pyrolysis (MASP) method. A 3% Ga₂O₃ doping level was used as the optimal composition to enhance the film's stability and conductivity. After deposition and annealing at 450 °C, the films were exposed to O₂ gas at pressures ranging from 0 to 400 mTorr to evaluate their oxygen detection capability. Optical properties were characterized using UV-Vis spectroscopy, while electrical measurements were performed with an IV-meter employing the two-point probe method. The results show that the optical band gap increased from 3.19 eV to 3.23 eV with rising O₂ pressure due to the Burstein-Moss effect, indicating a shift of the Fermi level and carrier concentration change upon oxygen adsorption. Simultaneously, the electrical resistance increased from $1.05 \times 10^7 \Omega$ to $1.25 \times 10^7 \Omega$, attributed to the formation of a surface depletion layer that reduces the density of free carriers. In addition, the sensitivity increased from 1.90% at 100 mTorr to 19.05% at 400 mTorr, indicating an increasingly pronounced electrical response with increasing oxygen pressure. The positive correlation between band gap and resistance confirms that oxygen exposure directly affects the film's optoelectronic behavior. With high transparency, structural stability, and linear response to oxygen, the ZnO:Ga₂O₃ thin film demonstrates strong potential for real-time oxygen sensing applications.

Keywords: Optical; Electrical; ZnO:Ga₂O₃; thin film; Microwave-Assisted Spray Pyrolysis; oxygen sensor

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INTRODUCTION

Oxygen gas sensors play a crucial role in a wide range of applications, including life safety systems, medical monitoring, environmental monitoring, and industrial process control [1]. Advances in sensor technology have resulted in the development of various oxygen sensors based on different materials and sensing mechanisms [2]. However, most commercial oxygen sensors still rely on electrochemical or zirconia-based materials that have limitations such as high cost, high operating temperature, and limited lifespan [3]. This has motivated the development of alternative materials that are more efficient, sensitive, and easy to fabricate — one promising approach is

through thin-film metal oxide semiconductors (MOS) such as ZnO.

MOS-based gas sensors have been widely studied due to their high sensitivity, rapid response, and good operational stability [4]. Among these materials, zinc oxide (ZnO) stands out because of its wurtzite crystal structure [5], wide band gap [6], and n-type semiconducting nature [7], which can be tuned through doping and surface treatment. The incorporation of gallium oxide (Ga_2O_3) into ZnO has been reported to enhance its electrical conductivity, optical transparency, and thermal stability, making it a promising material for oxygen (O_2) gas sensing applications [8].

Various deposition techniques have been used to synthesize ZnO-based thin films, including Sol-Gel Spin Coating [9], Chemical Vapor Deposition (CVD) [10], Pulsed Laser Deposition (PLD) [11], and Sputtering [12]. Although these techniques can produce high-quality films, they often require high temperatures, complex vacuum systems, or costly equipment. Therefore, this study employs the Microwave-Assisted Spray Pyrolysis (MASP) method, which offers advantages such as simplicity, low cost, and the ability to produce films with uniform thickness and strong substrate adhesion [13]. The microwave heating mechanism ensures even energy distribution within the aerosol precursors, promoting more homogeneous and efficient crystal growth compared to conventional spray pyrolysis or thermal methods [14, 15].

In a previous study, ZnO: Ga_2O_3 thin films were synthesized using the MASP method with 3% Ga_2O_3 doping, identified as the optimal composition for achieving improved optoelectronic characteristics [14]. After deposition, the films were exposed to oxygen gas (O_2) at pressures ranging from 0 to 400 mTorr to evaluate their capability in detecting the presence of oxygen in the surrounding environment.

The optical properties were characterized using UV-Vis spectroscopy to determine transmittance and band gap energy [16], while the electrical properties were measured using an IV-meter with the two-point probe method to obtain current-voltage (I-V) behavior [17]. The comparison of results before and after O_2 exposure provides insight into the changes in optical and electrical behavior of the films, which reflects their sensitivity and response to oxygen.

Therefore, this study aims to investigate the effect of oxygen exposure on the optical and electrical properties of ZnO: Ga_2O_3 thin films synthesized by the MASP method, as a preliminary step toward evaluating their potential as O_2 gas sensor materials.

METHOD

The precursors, dopant, solvent, and stabilizer used in this study were zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$), gallium oxide (Ga_2O_3), isopropanol ($\text{CH}_3\text{CHOHCH}_3$), and ethanolamine ($\text{C}_2\text{H}_7\text{NO}$). The precursor solution was prepared by dissolving 2.195 g of zinc acetate dihydrate in 20 mL of isopropanol and stirring with a magnetic stirrer at room temperature at 1000 rpm until completely dissolved. Subsequently, gallium oxide (Ga_2O_3) was added to the ZnO solution at a fixed concentration of 3 wt% and stirred for 5 minutes until homogeneous. The temperature was then raised to 70 °C, and the solution was stirred continuously for 1 hour while 1.452 mL of ethanolamine was added dropwise until a clear and stable solution was obtained. The prepared solution was then aged for 24 hours at room temperature to ensure chemical and viscosity stability. Glass substrates (1 × 1 cm) were sequentially cleaned using acetone, methanol, and distilled water in an ultrasonic bath for 15 minutes, then dried with nitrogen gas to remove residual solvents and fine particles. The deposition process was carried out using the Microwave-Assisted Spray Pyrolysis (MASP) method, in which the preheated substrate (250 °C for 10 minutes) was placed

inside a microwave operating at a medium-high power level ($\sim 200^\circ\text{C}$). The $\text{ZnO}:\text{Ga}_2\text{O}_3$ solution was evenly sprayed using a spray gun mounted at the top of the microwave, allowing the generated microwave plasma to assist precursor decomposition and promote the formation of a uniform thin-film layer on the substrate [14]. After spraying, the film was kept inside the microwave for 20 minutes to ensure complete drying, followed by annealing in a furnace at 450°C for 1 hour to improve crystallinity and structural stability [18]. After cooling to room temperature, the resulting $\text{ZnO}:\text{Ga}_2\text{O}_3$ thin films were exposed to O_2 gas at varying pressures of 0, 100, 200, 300, and 400 mTorr to evaluate their oxygen detection capability. The optical properties were analyzed using UV-Vis spectroscopy to determine transmittance and band gap energy, while electrical characteristics were measured using an IV-meter with the two-point probe method to obtain current-voltage (I-V) profiles before and after oxygen exposure.

RESULTS AND DISCUSSION

The optical and electrical properties of $\text{ZnO}:\text{Ga}_2\text{O}_3$ thin films were investigated to evaluate their sensitivity toward oxygen (O_2) exposure at various pressures ranging from 0 to 400 mTorr. The UV-Vis spectral analysis revealed that the optical behavior of the film was highly dependent on oxygen interaction, particularly in terms of transmittance and band gap energy variation [19]. As shown in **Figure 1**, the transmittance spectrum exhibited a sharp increase from below 10% at around 370 nm to approximately 97% above 500 nm, remaining stable up to 800 nm. The high transparency of the unexposed sample (0 mTorr) indicates a smooth, homogeneous surface with minimal light scattering [20].

The transmittance spectra indicate that the thin film exhibit high transparency in the visible region. However, a gradual decrease in transmittance was observed, from 97% (0 mTorr) to 93% (200 mTorr), and approximately 90% (300-400 mTorr), as shown in **Figure 1**. this reduction in transmittance suggests an increase in the density of defect centers, which serve as active adsorption sites for oxygen molecules. Such behavior reflects a balance between crystallinity and the formation of functional defects, both of which play a critical role in enhancing the sensitivity of photoconductive oxide-based sensors [23].

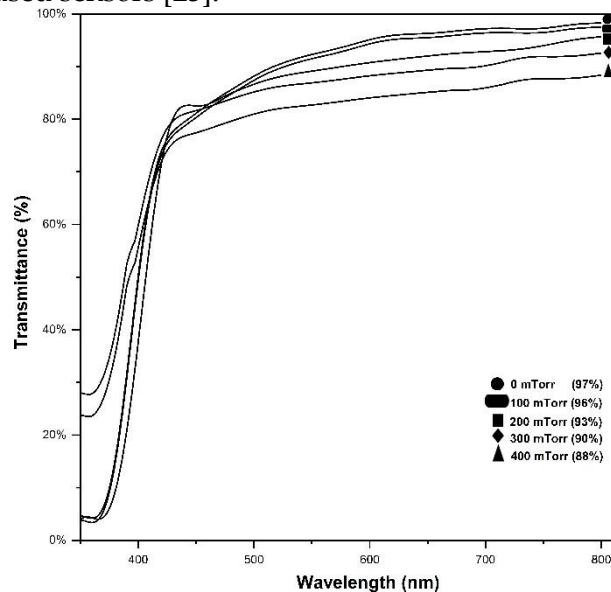


Figure 1. Transmittance Spectrum of $\text{ZnO}:\text{Ga}_2\text{O}_3$ Thin Film

Furthermore, a sharp optical absorption edge was observed in the wavelength range of 370-390 nm, indicating direct electronic transition from the valence band to the conduction band. To evaluate the optical absorption properties of the films, the transmittance data were converted into the absorption coefficient (α) using the following equation:

$$\alpha = \frac{1}{t} \ln \left(\frac{1}{T} \right) \quad (1)$$

where T is transmittance and t is the film thickness. And relationship indicates that a decrease transmittance results in an increase in the absorption coefficient, reflecting a stronger interaction between incident photons and the materials. As the density of oxygen-induced active defect increases, the light absorption behavior near the absorption edge is consequently modified.

Table 1. Band Gap of ZnO:Ga₂O₃ Thin Film

<i>Exposure O₂ (mTorr)</i>	<i>Band gap (eV)</i>
0	3.19
100	3.19
200	3.21
300	3.21
400	3.23

The optical bandgap energy (E_g) was subsequently determined using the Tauc plot method based on the calculated absorption coefficient. Since ZnO is a direct bandgap semiconductor, the direct allowed transition model was adopted. Accordingly, the optical bandgap was estimated from the linear extrapolation of the $(\alpha h\nu)^2$ versus $h\nu$ plot according to the Tauc relation:

$$(\alpha h\nu)^2 = A(h\nu - E_g) \quad (2)$$

where h is Planck's constant, ν is the photon frequency, A is a material dependent constant, and E_g is the optical bandgap energy. The corresponding Tauc plots are represented in **Figure 2**.

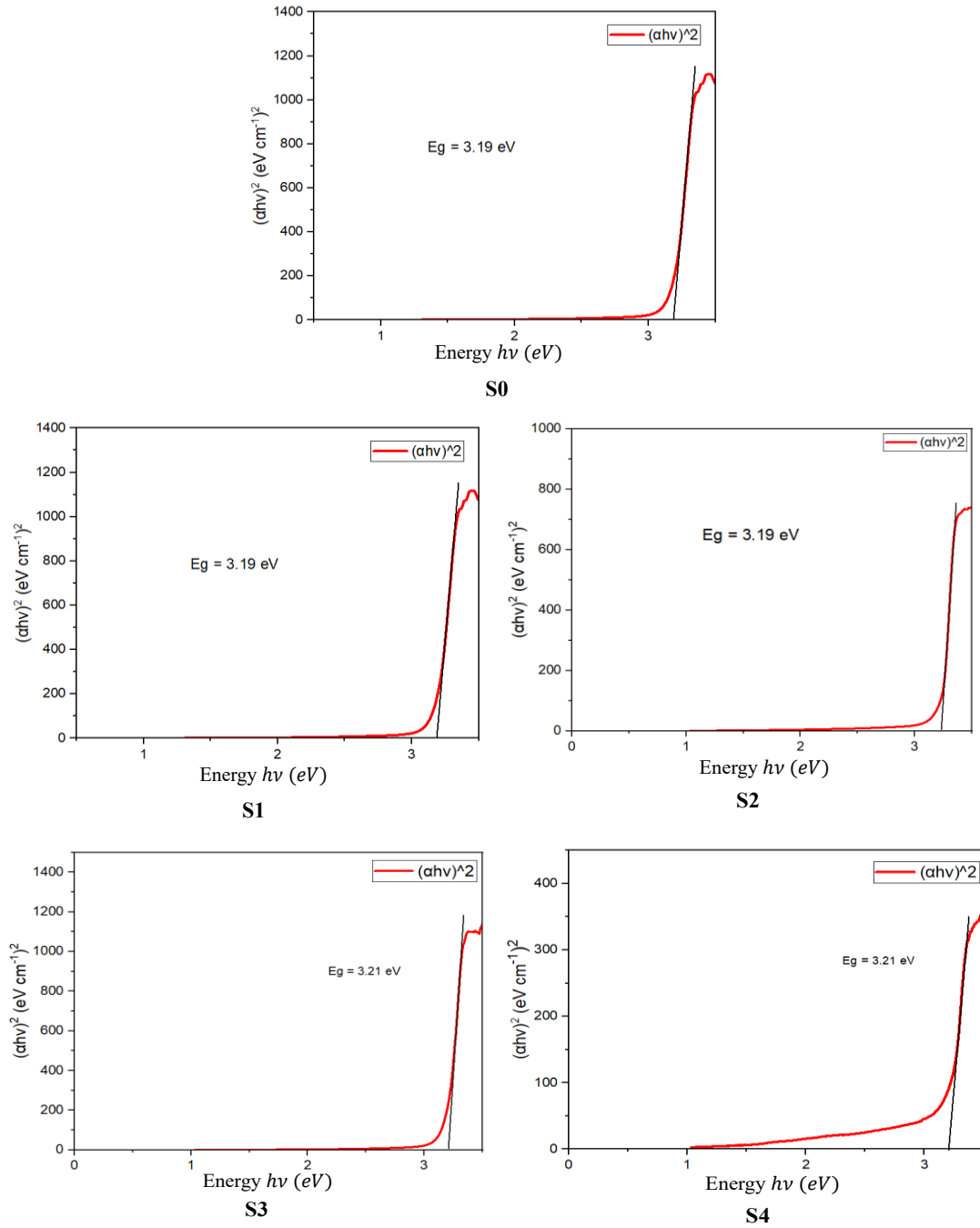


Figure 2. Tauc Plot of ZnO:Ga₂O₃ Thin Film

The optical bandgap of the ZnO:Ga₂O₃ thin films increased with increasing O₂ exposure. As summarized in **Table 1**, the E_g values increased from 3.19 eV for the unexposed sample (0 mTorr) to 3.19 eV at 100 mTorr, 3.21 eV at 200–300 mTorr, and 3.23 eV at 400 mTorr. The gradual widening of the optical band gap indicates that oxygen exposure influences the electronic characteristics of the ZnO:Ga₂O₃ thin films. Since the increase in band gap occurs concurrently with an increase in electrical resistance, the observed behavior cannot be adequately explained by the Burstein–Moss effect, which is typically associated with an increase in free carrier concentration. Instead, the band gap widening is more likely related to oxygen-induced modifications of surface electronic states

and defect-related states that affect the optical absorption behavior of the films [21].

The increase in band gap energy suggests that oxygen exposure alters the electronic structure of the ZnO:Ga₂O₃ thin films without causing significant deterioration of their optical properties. Oxygen adsorption can influence the distribution of electronic states near the band edges, leading to changes in the optical transition energy and the measured band gap. This interpretation is consistent with the resistance measurements, which indicate a reduction in free charge carrier concentration upon oxygen exposure. Therefore, the observed increase in band gap is more appropriately associated with modifications of surface and defect-related electronic states rather than an increase in carrier concentration [22].

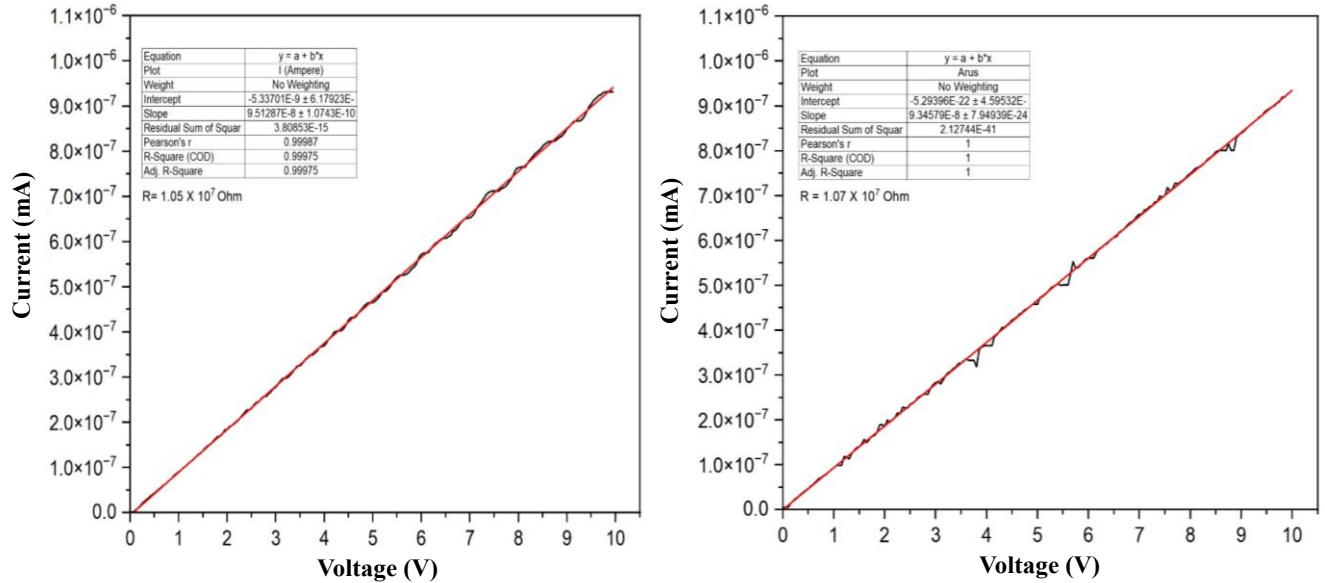


Figure 3. I–V Curve of ZnO:Ga₂O₃ Thin Film Before (a) and After (b) O₂ Exposure

Electrical measurements were carried out using the two-point probe method to evaluate the current–voltage (I–V) characteristics of the films. The I–V curves (**Figure 3**) exhibited linear behavior for all oxygen exposure levels, indicating an ohmic conduction mechanism and good electrode–film contact. The resistance values summarized in **Table 2** increased gradually from $1.05 \times 10^7 \text{ } \Omega$ (0 mTorr) to $1.07 \times 10^7 \text{ } \Omega$ (100 mTorr), $1.19 \times 10^7 \text{ } \Omega$ (200 mTorr), $1.20 \times 10^7 \text{ } \Omega$ (300 mTorr), and $1.25 \times 10^7 \text{ } \Omega$ (400 mTorr). This consistent increase in resistance corresponds to the adsorption of oxygen molecules on the film surface, which capture free electrons from the conduction band to form O₂⁻ ions [24]. As a result, the electron density decreases, and a surface depletion layer is formed, reducing the overall conductivity of the material.

Table 2. Resistance of ZnO:Ga₂O₃ Thin Film

Exposure O ₂ (mTorr)	Resistance ($\times 10^7$ Ω)	Characteristics
0	1.05	Ohmic
100	1.07	Ohmic
200	1.19	Ohmic

<i>Exposure O₂</i> (mTorr)	Resistance ($\times 10^7$ Ω)	Characteristics
300	1.20	<i>Ohmic</i>
400	1.25	<i>Ohmic</i>

The strong correlation between optical and electrical responses confirms that oxygen exposure influences both the electronic band structure and the carrier dynamics of ZnO:Ga₂O₃ films [25]. The simultaneous increase in band gap and resistance implies that the reduction of free carrier density due to oxygen adsorption directly affects the film's optoelectronic characteristics [26].

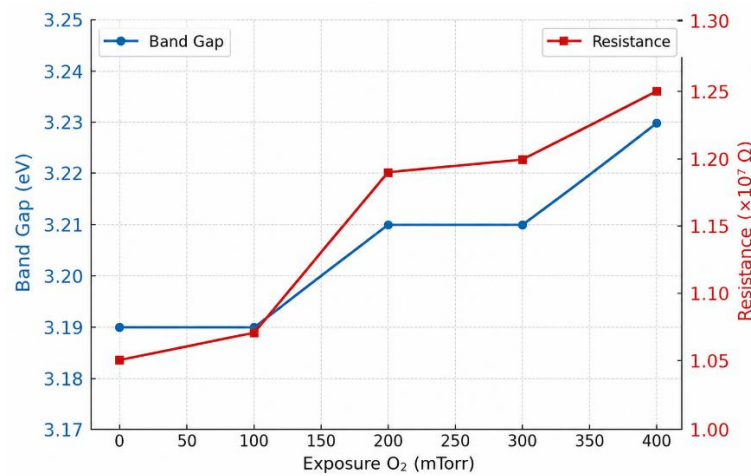


Figure 4. Correlation between Band Gap and Resistance as a Function of O₂ Pressure

Figure 4 show the correlation between the optical band gap and electrical resistance of ZnO:Ga₂O₃ thin films as a function of O₂ pressure ranging from 0 to 400 mTorr. Both parameters exhibit a gradual increase with higher oxygen pressure, indicating a direct relationship between the film's optical and electrical properties. The optical band gap increased from 3.19 eV to 3.23 eV, while the electrical resistance increased from $1.05 \times 10^7 \Omega$ to $1.25 \times 10^7 \Omega$. This strong correlation between the optical and electrical responses suggests that oxygen exposure significantly influences the electronic characteristics of the ZnO:Ga₂O₃ thin films.

The increase in electrical resistance indicates a reduction in the concentration of free charge carriers participating in the electrical conduction process [27]. Simultaneously, the observed widening of the optical band gap cannot be adequately explained by the Burstein-Moss effect, since that mechanism is typically associated with an increase in carrier concentration accompanied by a decrease in electrical resistance. In contrast, the present results demonstrate that the band gap enlargement occurs concurrently with an increase in resistance.

Therefore, the observed band gap widening is more likely associated with oxygen-induced modifications of the surface electronic states. The interaction between oxygen species and the film surface may alter the distribution of electronic states near the band edges, thereby affecting the optical absorption characteristics of the material. These surface-related modifications may also

influence charge transport processes, which is reflected in the increase in electrical resistance [28]. Consequently, the simultaneous increase in band gap and resistance can be regarded as complementary manifestations of oxygen-induced changes in the electronic characteristics of the ZnO:Ga₂O₃ thin films. This parallel behavior confirms that oxygen exposure not only affects the optical response of the films but also modulates their electrical conductivity, further demonstrating the strong coupling between the optical and electrical properties of the material [29].

Table 3. Sensitivity of ZnO:Ga₂O₃ Thin Film

<i>Exposure</i> O ₂ (mTorr)	R_Before O ₂ Exposure ($\times 10^7 \Omega$)	R_After O ₂ Exposure ($\times 10^7 \Omega$)	Sensitivity (%)
0	1.05	1.05	0
100	1.05	1.07	1.90
200	1.05	1.19	13.33
300	1.05	1.20	14.29
400	1.05	1.25	19.05

This interpretation is further supported by the sensitivity results summarized in **Table 3**, which were calculated from the resistance changes before and after oxygen exposure. As shown in **Table 3**, the sensitivity increased from 1.90% at 100 mTorr to 13.33% at 200 mTorr, followed by 14.29% and 19.05% at 300 and 400 mTorr, respectively. The continuous increase in sensitivity indicates that the resistance variation induced by oxygen exposure becomes more pronounced at higher oxygen pressures. These findings demonstrate that the ZnO:Ga₂O₃ thin films exhibit a measurable electrical response to variations in the oxygen environment. The correlation among band gap widening, resistance increase, and sensitivity enhancement suggests that oxygen exposure simultaneously affects both the optical and electrical properties of the films.

Overall, the results demonstrate that ZnO:Ga₂O₃ thin films synthesized via the MASP method exhibit significant changes in both optical and electrical properties under O₂ exposure up to 400 mTorr. The gradual increase in optical band gap, electrical resistance, and sensitivity with increasing oxygen pressure confirms the responsiveness of the films to variations in the surrounding oxygen environment. Combined with their high optical transparency and linear I–V characteristics, these findings highlight the promising potential of ZnO:Ga₂O₃ thin films as active materials for oxygen gas sensing applications [30]. Therefore, the synthesized films represent a promising candidate for future oxygen monitoring technologies and semiconductor-based gas sensor systems.

CONCLUSION

It can be concluded that ZnO:Ga₂O₃ thin films synthesized by the Microwave-Assisted Spray Pyrolysis (MASP) method with 3% Ga₂O₃ doping exhibit measurable optoelectronic changes under oxygen exposure in the pressure range of 0–400 mTorr. The optical band gap increased from 3.19

eV to 3.23 eV, while the electrical resistance increased from $1.05 \times 10^7 \Omega$ to $1.25 \times 10^7 \Omega$ with increasing oxygen pressure. In addition, the sensitivity increased from 1.90% at 100 mTorr to 19.05% at 400 mTorr, indicating an enhanced electrical response at higher oxygen pressures. The positive correlation among band gap, resistance, and sensitivity confirms that oxygen exposure affects the optoelectronic characteristics of the films. These findings provide preliminary evidence that ZnO₂O₃ thin films possess promising potential for oxygen sensing applications.

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