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Impact of Argon Flow Rate on Electrical and Optical Properties of Al-doped ZnO Thin Films with Insight into Ti/Al Ohmic Behaviors to n-ZnO

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ABSTRACT

This study explores the impact of Ar flow rate on the electrical and optical properties of Al-doped ZnO thin films and the performance of Ti/Al Ohmic contacts to n-ZnO films. Al-doped ZnO thin films were deposited on glass substrates using a tri-axis RF sputtering system under varying Ar flow rates (30–80 sccm). Electrical properties, including resistivity, carrier concentration, and mobility, were evaluated using Hall measurements, while optical transmittance was analyzed through UV-visible spectroscopy. The resistivity decreased with increasing Ar flow rate, achieving a minimum of 9.74×10^{-4} Ω·cm at 80 sccm, alongside improved mobility and carrier concentration due to enhanced film morphology and crystallinity. We have obtained that the optical transmittance values are between 75% and 80% and we know this as a blue shift phenomenon that occurs at the UV absorption edge due to the Burstein-Moss effect. It is seen that there is an almost ideal behavior in the Ti/Al Ohmic contact, with a specific contact resistivity as low as 1.8×10−5 Ω·cm2 in 15 nm Ti layers. We can conclude the importance of optimizing sputtering parameters on film properties for solar cell and optoelectronic device applications. Thus, we conclude that there is an interaction between deposition conditions and material performance to improve electrical conductivity and high transparency in Al-doped ZnO films.

Keywords: *Al-doped ZnO thin films, Argon flow rate, Electrical properties, Optical transmittance, Ohmic contact*

1. INTRODUCTION

In recent years, sputtered Al-doped ZnO/n-Si thin films have been potential candidates for solar cells and optoelectronic device applications because of their excellent electrical and optical properties. Owing to the large bandgap of 3.3 eV and exciton binding energy of 60 meV at environmental temperature, these are highly suited for such devices [1], [2]. High-quality Zinc Oxide (ZnO) films require much knowledge of how the growing conditions influence material properties, a prominent area of research even today [3]. Especially for high-performance solar cells, precise control over resistivity, transparency, and electrical contact resistance is necessary.

There has been a great deal of work focused on lowering ZnO's resistivity, with aluminum doping being among the best techniques. Al doping enhances free carriers, enhancing electrical conductivity without losing transparency [4]. Numerous studies have explored Al doping's impact on resistivity, mobility, carrier concentration, transmittance, bandgap, structure, and surface topography [5]. The Al^{3+} impurity increases carrier concentration by substituting Zn in the lattice and acting as a donor, with a direct impact on the electrical features of the thin film [6], [7]. There are still issues confronting films with high conductivity and transparency [8].

ZnO is superior in terms of performance compared to other semiconductors due to its wide bandgap (~3.3 eV), high excitonic binding energy (~60 meV), and strong piezoelectricity, which makes ZnO suited for use in optoelectronic and energy-harvesting applications [9]–[12]. ZnO differs from silicon (Si), gallium nitride (GaN), and indium tin oxide (ITO) semiconductors due to its wide bandgap (3.37 eV), high light transparency, low costs, and environmental compatibility. Compared with light-absorbing Si, ZnO is transparent for visible light with high electron mobility and doping flexibility [13]. ZnO is superior to GaN and ITO with increased abundance and ease of processing within low-cost sputtering methods [14], which permits tight control on thin-film features [10]. Sputtering poses plasma-induced damage, yet advantages such as flat deposition, nanostructure flexibility, and lower processing temperatures outweigh disadvantages. ZnO is non-toxic, unlike GaN, and avoids the limitation of being as rare as indium-based ITO. These qualities make ZnO a superior choice for optoelectronic, transparent electronic, and high-frequency semiconductor applications [11].

The effects of argon (Ar) flow rates during sputtering have been extensively studied to optimize film properties. Higher Ar flow rates improve crystallinity and reduce grain boundary scattering, leading to lower resistivity and enhanced carrier mobility [4]. We know that in the visible spectrum and the shift of the UV absorption edge, there is a Burstein-Moss effect phenomenon due to the effect of the Ar flow rate [11]. This effect is due to the presence of highly doped semiconductors. As a result, the optical band gap shifts as a result of increasing the concentration of free carriers, so that the consequence is that the conduction band will be filled with excess electrons, which can generate electrons from the valence band, so that the optical band gap becomes wider. The presence of a high concentration of Al as a carrier in ZnO thin films resulted from increasing the Ar flow rate. As a result, there is a clear blue shift in the absorption edge, which we know as the Burstein-Moss effect, so we know that the Ar flow rate is an important parameter for adjusting the electrical and optical characteristics of Al-doped ZnO thin films.

In this study, we will study the optimization of Ar flow rate on the electrical and optical properties of Al-doped ZnO thin films. Furthermore, we will also study its resistivity, charge carrier concentration, and optical transmittance. In addition, the device performance will be studied through the presence of Ti/Al ohmic contacts on n-ZnO thin films. Thus, the aim is to provide insight into the importance of Ar flow rate optimization on high-quality ZnO thin films with excellent electrical conductivity and transparency for solar cell and optoelectronic applications [15].

2. THEORETICAL BACKGROUND

We know that because it has excellent electrical conductivity and high optical transparency in thin films of aluminum-doped zinc oxide (AZO) that it is categorized as an optoelectronic and semiconductor material. This is because it is significantly affected by different deposition parameters, especially by the flow rate of Ar during the sputtering process. It has been shown that the optimal Ar flow rate can increase the carrier concentration, decreasing AZO film's resistivity with high transparency [21]. However, we know that excessive Ar flow results in defects and distortion of the structure, which can then affect the increase of scattering and decrease the conductivity [22]. Therefore, achieving the balance of electrical and optical characteristics of AZO film is very important through Ar flow optimization.

The metal contact interface is one of the factors directly responsible for AZO films' electrical conductivity. It is known that Titanium (Ti) and aluminum (Al) metals are widely used to make ohmic contacts with n-type ZnO due to their suitable work functions and high electrical conductivity [23]. In addition, it is known that the deposition conditions and post-deposition annealing treatments can significantly affect the behavior of these metal contacts. Several studies show Ti/Al contacts on the ZnO surface have low ohmic resistance values for optimized conditions. This is highly recommended as an electronic device [24]. On the other hand, the influence of improper annealing can result in unwanted interfacial reactions, which have an impact on electrical properties.

In addition, the contact characteristics of Ti/Al-AZO films can be changed through annealing temperature parameters. The increase in ohmic behavior can be achieved by reducing the contact electrical resistance and increasing the carrier injection efficiency [25]. However, the electrical properties will be damaged as a high temperature effect due to the presence of interface compounds such as NiZnO [26]. It is also known that there is a change from ohmic behavior to rectifier behavior due to the role of the polarity of the ZnO surface. Therefore, strategic interface engineering is needed for the electrical contact to be stable and highly efficient.

For optoelectronic device applications, including solar cells and transparent conductive electrodes, the optical properties of AZO thin films play an important role. This is because it is known that controlled Ar flow rate during deposition can reduce the defect state and consequently increase the optical transmittance without sacrificing the electrical resistance [21]. It is also reported that high Ar flow during sputtering due to high-energy ion bombardment is evidence of defects and reduces the transparency of the film [24].

In AZO thin films, the goal is to optimize their use in several applications. The effect of Ar flow rate on AZO thin films' electrical and optical characteristics is significant. Deposition conditions, especially Ar flow rate and post-annealing, are important determinants of AZO film quality and compatibility with Ti/Al ohmic contacts. Understanding their relationship allows researchers and engineers to fabricate high-quality AZO-based optoelectronic devices with high conductivity and transparency. To optimize their applications, more work is required to determine the long-term behavior of such materials exposed to various environmental conditions.

3. METHODOLOGY

In this study, Al-doped ZnO thin films were fabricated on glass substrates using a Tri-Axis Semicore RF sputtering system (Figure 1) [26]. The sputtering process utilized a 3-inch diameter, 99.99% pure Al-doped (5%) ZnO target sourced from Semiconductor Wafer Inc., which was mounted on one of the RF cathodes. The substrate was positioned approximately 10 cm from the target surface and maintained at a deposition temperature of 400°C. The sputtering power was fixed at 50 W, and the Ar flow rates during deposition varied at 30, 50, 60, 70, and 80 sccm [27]. A base pressure of 10-6 mbar was maintained to ensure an ultra-clean environment. Following the ZnO deposition, Ti/Al metal electrodes were deposited on the n-type ZnO thin films using a Leybold electron beam evaporator with 99.999% pure metal sources [30].

The optical properties of the Al-doped ZnO thin films were analyzed using a Shimadzu Solidspec 3700 DUV spectrophotometer. Measurements were conducted over a wavelength range of 200 to 1500 nm to determine the transmittance characteristics of the films [29]. Hall effect measurements were performed to evaluate the electrical properties using the Lakeshore 7704A Hall measurement system, which provided data on carrier concentration, mobility, and resistivity under various Ar flow rates. These measurements were essential to correlate the deposition conditions with the electrical performance of the films [30]. Additionally, the current-voltage (I-V) characteristics of the Ti/Al contacts were acquired at room temperature using a Keithley 617 Picoammeter. These measurements provided insights into the Ohmic behavior of the contacts and their specific contact resistivity [31]. By systematically studying the impact of Ar flow rates and deposition parameters, this work offers valuable data on optimizing the sputtering process for achieving high-quality Al-doped ZnO thin films with desirable electrical and optical properties for advanced device applications.

Figure 1. Tri-Axis Semicore sputtering system.

4. DATA COLLECTION AND ANALYSIS

The data collection process for studying the impact of argon (Ar) flow rate on the electrical and optical properties of aluminum-doped zinc oxide (AZO) thin films involves several key methodologies. The AZO films are typically deposited using RF magnetron sputtering, where the Ar flow rate is systematically varied to observe its influence on film characteristics. Parameters such as substrate temperature, deposition time, and target power are carefully controlled to ensure consistency during deposition. Post-deposition, the film thickness and X-ray diffraction (XRD) were used to understand the microstructural changes induced by different Ar flow rates. For electrical characterization, Hall effect measurements are conducted to determine carrier concentration, mobility, and resistivity of the AZO films. Additionally, four-point probe measurements are used to assess the sheet resistance of the films, providing insights into their conductivity. The contact behavior of Ti/Al with AZO films is examined by fabricating metal-semiconductor contacts and measuring current-voltage (I-V) characteristics using a Keithley source meter. The influence of annealing temperature on the ohmic behavior of Ti/Al contacts is also analyzed to determine optimal processing conditions. For optical characterization, UV-Vis spectrophotometry is used to measure the optical transmittance and bandgap of the films, ensuring that the deposition conditions do not compromise transparency [3].

Next, we conducted data analysis through correlation studies between electrical and optical data with deposition parameters. In addition, we conducted studies of defects or grain boundary effects that contribute to resistivity variations through microstructural observations from XRD with electrical measurements. These results Analysis can provide a deeper understanding of the interaction between Ar flow rate, film microstructure, and Ti/Al contact behavior, thus helping optimize AZO thin films as high-performance optoelectronics.

5. DISCUSSION AND RESULTS

The resistivity, carrier concentration, and mobility properties of AZO thin films have been studied systematically as a function of Ar flow rate. The results of the study (Figure 2) show that the resistivity decreases significantly with an increase in the Ar flow rate of 80 sccm. The results of XRD analysis indicate that the decrease in resistivity is due to the increase in crystallinity of the film and larger grain size. The grain size of ZnO thin films can be estimated using the Scherrer equation [10], where a decrease in the full width at half maximum (β) of the diffraction peak with increasing Ar flow rate indicates grain growth. This correlation between improved crystallinity and reduced resistivity suggests that optimizing the Ar flow rate enhances the films' structural and electrical properties. XRD analysis is crucial for quantitatively validating this claim, ensuring that the observed improvements in electrical performance are directly linked to crystallinity enhancement. The reduction in resistivity is primarily attributed to larger grain sizes, which minimize grain boundary scattering and enhance carrier transport. Similar trends have been observed in previous studies, where controlled sputtering conditions led to optimized electrical properties of ZnO thin films [28]. The enhanced crystallinity and structural homogeneity at higher Ar flow rates facilitate efficient charge carrier movement, significantly lowering resistivity values. These findings highlight the importance of fine-tuning sputtering parameters to achieve high-performance ZnO thin films with superior electrical characteristics [33].

Figure 2 illustrates the effect of Ar flow rate on the electrical properties of Al-doped ZnO thin films. Also, trends in resistivity, carrier concentration, and mobility as a result of the increased rate of Ar flow are seen. As the Ar flow rate increases from 20 sccm to 60 sccm, resistivity decreases significantly due to improved crystallinity and larger grain sizes, which reduce grain boundary scattering and enhance carrier transport. Simultaneously, carrier concentration increases steadily, reaching approximately 8.0×10^{20} cm^3 at 80 sccm, indicating enhanced Al dopant incorporation. Electron mobility follows a similar trend, peaking at around 8 $cm^2/V \cdot s$ at 60 sccm due to reduced defect scattering and improved structural homogeneity; however, beyond 60 sccm.

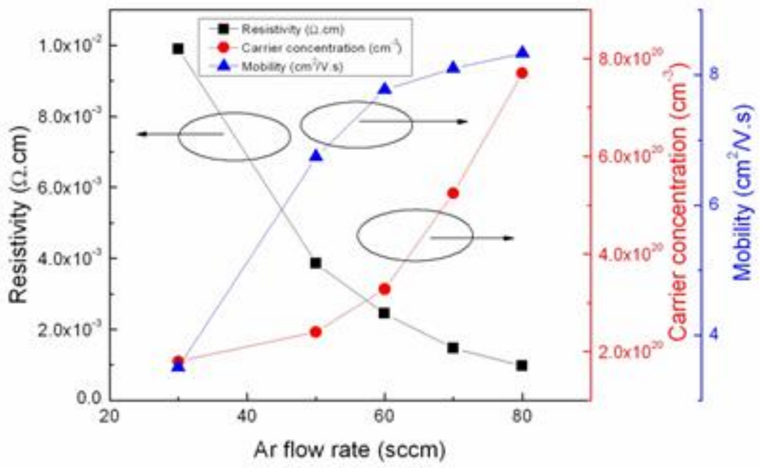

Figure 2. Ar flow rate effect on the electrical properties of Al-doped ZnO film on glass substrate at P=150 W, Tsub = 400° C, and d = ~ 100 nm.

The carrier concentration notably increased with higher Ar flow rates, as seen in Figure 2. This increase can be linked to the higher energy provided during deposition, which enhances Al incorporation into the ZnO lattice as a donor dopant. The resulting increase in free electron density significantly boosts the carrier concentration, consistent with earlier observations that doping and sputtering conditions play a vital role in determining carrier density [37], [38]. This behavior aligns with the Burstein-Moss effect, where increased carrier concentration shifts the Fermi level into the conduction band, further influencing the optical and electrical performance of the films [35].

In addition, it is seen that there are effects between crystallinity and doping due to the influence of thin film mobility through a higher Ar flow rate. It is also seen that the increase in carrier concentration and electron mobility reduces resistivity. This indicates superior electrical properties as a result of deposition at an Ar flow rate of 80 sccm. These results strengthen the theory that deposition parameters, such as gas flow rate and power, can affect the electrical behavior of ZnO-based thin films [31]. Meanwhile, increased mobility can be related to reduced carrier scattering at grain boundaries and layer morphology for high-performance optoelectronic applications.

As a result of increasing the Ar flow rate, at the UV absorption edge, a blue shift phenomenon known as the Burstein-Moss effect is seen as a result of higher carrier concentration, causing the Fermi level to move to the conduction band, which increases the optical band gap. It can also be observed that the concentration and mobility of carriers have been reported previously in this study, as the Pauli exclusion principle, where there is a higher energy filling by additional carriers [34]. Thus, there is an interaction between electrical and optical properties as a result of increasing doping and carrier mobility, simultaneously increasing transparency and changing the characteristics of the band structure.

At longer wavelengths (above 1200 nm), the transmittance decreases with higher Ar flow rates due to free carrier absorption. This phenomenon arises as increased carrier concentration leads to enhanced interaction between free carriers and incident photons, causing a reduction in transmission. Similar trends have been reported in previous studies where higher doping levels or carrier densities were associated with increased free carrier absorption and reduced transparency in the infrared region [34]. These results highlight the trade-off between achieving high carrier concentration for better electrical conductivity and maintaining optical transparency, particularly in the near-infrared range. Optimizing Ar flow rates, therefore, becomes crucial for balancing these properties in Al-doped ZnO films for advanced optoelectronic applications.

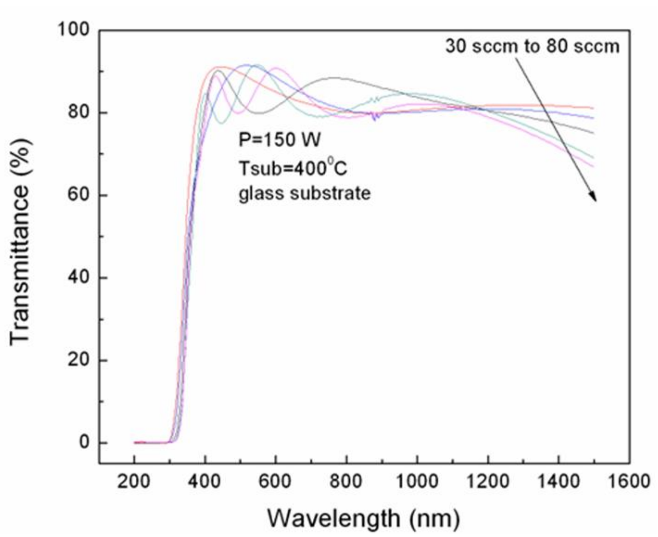

Figure 3. Transmittance of Al-doped ZnO film on glass substrate at various Ar flow rates between 30 sccm and 80 sccm.

The optical transmittance of Al-doped ZnO thin films, as shown in Figure 3, reveals significant insights into the effect of Ar flow rate on transparency. The transmittance in the visible range (400–700 nm) lies between 75% and 80%, indicating the high optical quality of the films deposited at various Ar flow rates. These values are consistent with the requirements for transparent conducting oxides in optoelectronic applications, such as solar cells and displays [33]. The high transparency suggests that the films possess minimal surface roughness and defect density, which is critical for maintaining optical performance [34]. Additionally, the films demonstrate a sharp absorption edge in the ultraviolet region, highlighting their suitability for UV-blocking applications.

To explore Al-doped ZnO thin films for various device applications, we follow up with our highest electrical properties in the previous section. Thus, we fabricated and characterized Ti/Al Ohmic contacts to Al-doped n-type ZnO thin films. Meanwhile, controlling metal thickness is one of the most promising methods for improving the performance of Ohmic contacts. For instance, Wang et al. reported that at various metal thicknesses, the Al/ZnO exhibited nearly ohmic behavior [20]. However, the physical mechanism of metal thickness on the electrical properties of the contacts is not well known. In this work, the Ti metal contact pairs were evaporated on the Al/n-ZnO/SiO_2/n-Si layer.

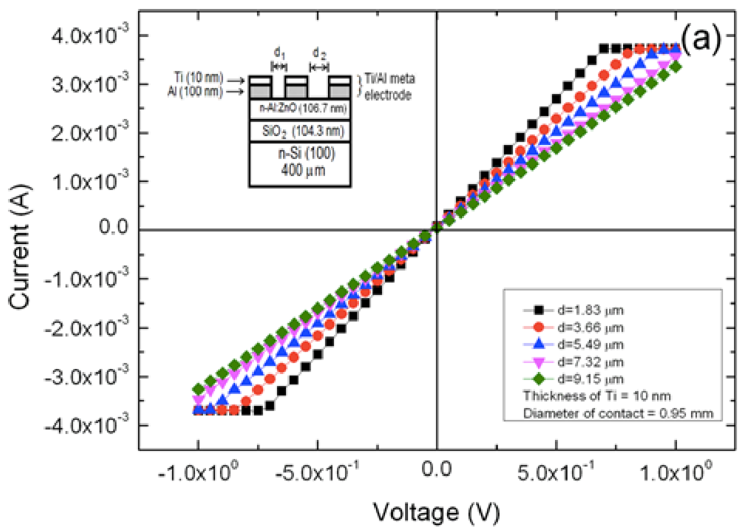

Figure 4. Linear I–V characteristics of the Ti/Al Ohmic contact to Al-doped ZnO thin films at various thicknesses of Ti: (a). Ti = 10 nm, (b). Ti = 15 nm. (The inset shows a cross-sectional schematic of the device Ti/Al in contact with n-Si/SiO_2/n-ZnO).

The diameter and thickness of the Ti metal electrode were maintained as constant at ~0.95 mm and ~10–15 nm, respectively. The thickness of Al metal, Al-doped ZnO thin films, and the insulator layer SiO_2 were kept at 100 nm, 106.7 nm, and 104.3 nm, respectively (see inset Figure 4(a) and 4(b)). The I-V measurements of the contact resistance were made in the voltage range between -1.0 and 1.0 V at room temperature.

Figure 4 shows the forward and reverse bias I–V characteristic curves of Ti/Al/n-ZnO/SiO_2/Si structures. This figure indicates that Ti/Al metal contacts have nearly Ohmic behavior with Al-doped ZnO thin films either in forward or reverse bias. It can be seen that the contacts with 15 nm-thick Ti exhibited better Ohmic behavior than those of 10 nm-thick Ti. Using the transfer length transmission line model (TLM), the specific contact resistivity ρ_c given by [29]:

$$\rho_c = R_{sheet} \times L_T^2 \tag{1}$$

where R_{sheet} is the semiconductor sheet resistance (in Ω/sqr), and L_T is the transfer length (in cm), the total resistance was plotted as a function of the inter-contact distance for all the samples. The important device parameters can be extracted from the resistance as a function of inter-contact distance curves. The intercept of the plot at d = 0 gives the contact resistance, and the intercept at d = 0 gives -d = $2L_T$ [35]. The inter-contact distance was varied between 1.83 and 9.15 m. In Figure 4, the resistance is shown as a function of inter-contact distance at various thicknesses of Ti. For the sample evaporated at 10 nm-thick Ti, the total resistance and transfer length are 175.5 Ω and 0.38 μm, respectively. The specific contact resistivity ρ_c of 3.6 × 10^{-5} Ω. cm^2 was then calculated. On the other hand, the specific contact resistivity ρ_c of 1.8 × 10^{-5} Ω. cm^2 was obtained at 15 nm-thick Ti. However, a deeper understanding of these phenomena may be expressed by [36]:

$$\rho_c \propto \exp\left(\frac{\phi_B}{E_{00}}\right) \tag{2}$$

where ϕ_B is the barrier height and E_{00} is the tunneling energy parameter defined by

$$E_{00} = \frac{qh}{4\pi}\sqrt{\frac{N}{m^*\varepsilon_0\varepsilon_s}} \tag{3}$$

For $\frac{E_{00}}{kT} \gg 1$ i.e. high doping densities, field emission is dominant, and the contact becomes Ohmic. Our metal contact indicates the carrier tunnels through the full barrier width. Based on our result above, the values of the specific contact resistivity to Al-doped ZnO thin films are low enough for exploring efficient electronic and optoelectronic devices.

4. CONCLUSIONS

We have examined the effect of the Ar flow rate on the electrical and optical characteristics of Al-doped ZnO thin films on a glass substrate by RF sputtering. The present research illustrates that the electrical and optical characteristics of Al-doped ZnO thin films, namely, the resistivity, carrier concentration, mobility, and transmittance, are susceptible to the Ar flow rate. The process permits control over the quality of films for solar cells and transparent conducting oxides for display purposes. In addition, the metal thickness and physical mechanism for the Ohmic contact Ti/Al to n-ZnO thin films were fabricated and analyzed. Electrical performance is improved with increasing metal thickness up to that value, since it favors interface homogeneity and minimizes contact resistance. For greater metal deposition beyond this value, however, greater surface roughness and probable interfacial defects occur, deteriorating the qualities of contacts. Our findings are important for attaining high electrical values for highly transparent ZnO thin film-based solar cells.

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