Structural, FTIR, optical and dielectric properties of Zn$_{1-x}$Al$_x$O ceramics for advanced applications

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Abstract
We report here the structural, FTIR, optical and dielectric properties of Zn$_{1-x}$Al$_x$O with $x = 0.00 \leq x \leq 0.20$). The samples are synthesized by the solid state reaction method and the phase purity, structural morphology, and absorption spectra are examined by XRD, SEM, and FTIR techniques. The optical and dielectric measurements are obtained by a double beam spectrophotometer and an impedance analyzer. The wurtzite structure is confirmed for all samples, and the lattice parameters, crystallite diameter, and porosity are decreased by Al, whereas the Debye temperature and elastic modulus are increased. The residual stress is compressive for ZnO, but it is changed to tensile for the doped samples. Two different values of energy gap (E$_{gh}$ and E$_{gl}$) are apparent for each sample. The E$_{gh}$ and E$_{gl}$ are, respectively, 3.60, 3.56, 3.43, 3.53, 3.68 eV and 3.02, 2.95, 2.85, 2.92, 3.07 eV for all samples such that $\Delta E = (E_{gh} - E_{gl}) \approx 0.60$. The residual dielectric lattice constant is decreased by increasing x to 0.10, followed by a sharp increase at $x = 0.20$, while the opposite behavior is obtained for $(N/m^3)$. The dielectric constant and ac conductivity are slightly increased as x increases to 0.025, followed by a sharp increase with more increase of x to 0.20. The conduction is electronic for pure and x = 0.025 samples, but it changes to hole with increasing x to 0.20. The binding energy was decreased as x increased to 0.20, but there is no exact trend for the hopping distance and density of localized states against x. The F-factor for solar cell design was increased as x increased to 0.10, but it is almost constant at $x = 0.20$. The Cole–Cole plot is a straight line for $x = 0.00$, a semicircle arc for $x = 0.025$, and a complete semicircle for $x \geq 0.05$. The impedance resistance of grain and grain boundaries decreases as x increases to 0.20. These outcomes indicate that the addition of Al to ZnO shifts the mechanical, optical, and dielectric medium to higher values, which is strongly recommended for the design of optoelectronic and solar cell instruments.

Keywords Al doped ZnO · Elastic modulus · Energy gap · Dielectric medium and · F-factor

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1 Introduction

Wurtzite structure of pure n-type ZnO with 3.2 eV band gap (Eg) and 60 Urbach energy has been used in varistors, optoelectronic and solar cell devices (Manikandan et al. 2017; Ragupathi et al. 2013; Kumar et al. 1715). ZnO has been synthesised by the solid state reaction method through sintering at high temperatures followed by quenching in air to RT (Mohamed and Abu-Dief 2018; Mustaqima 2014; Djerdi and Jaglici´c et al., 1096; Yang 2013). However, the properties of ZnO are controlled by intrinsic defects such as oxygen vacancies generated during sintering and also be improved by quenching (Amin and Sedky 2019; Sedky et al. 2007). Despite the existence of advantageous chemical techniques for ZnO synthesis, the solid-state method is still of interest because it governs the ZnO to work as a varistor (Mohamed and Abu-Dief 2020). However, oxygen vacancies in ZnO can exist in three charge states, i.e., V_o^0, V_o^+, and V_o^{++}, respectively. The V_o^0 and V_o^{++} charged are stable, while V_o^+ is unstable and it is usually changed into V_o^{++} by capturing electrons from the CB. However, due to V_o^{++} defects (10^{19} cm^{-3}), some of the energy states lie at about 0.86 eV above the valence band (VB), which can affect the Eg of ZnO to be qualified for practical applications (Vempati et al. 2012; Rodnyi et al. 2018a).

The mechanism of the ZnO conduction band (CB) usually arises when the 4 s orbital of Zn is wide enough to allow for effective charge transfer (Mohamed et al. 2022; Kumar and Rani 2013; Abdullah et al. 2012). Consequently, the electrons are excited from the VB due to the absorption of light, and consequently the electron–hole pairs are generated. Above the limit of the Eg, the recorded signals of photon energy are due to non-radiative and Urbach tail processes. However, the energy of photons for creating electron–hole pairs (e–h) can be considered only for materials with a larger Urbach energy like ZnO (Sedky et al. 2020). In order to improve the optical properties, ZnO has 3-d transition metals (Shahedi and Jafari 2017; Raghu et al. 2017). Among these elements, Al doped ZnO is considered as an alternative for solar cells and light-emitting diodes (LEDs) as a result of the extra free electron formed in the ZnO CB (Wang and Zhang 2017; Zhai et al. 2016; Al-Naim et al. 2021). It is also approved that Al decreased the particle size and slightly increased the Eg of ZnO, which is attributed to the Al impurity incorporation or structural defects (Al-Naim et al. 2021; Diqiu et al. 2013; Kamarulzama 2015). Due to the extrinsic Al dopant, the CB of ZnO can be partially filled with free electrons of Al^{3+}, resulting in an Eg increase. However, when the carrier concentration exceeds 5×10^{20} cm^{-3}, Eg shrinkage may occur due to carrier renormalization of carriers (Tong et al. 2011; Li et al. 2014a). In addition, Al can also affect the excitonic absorption near the band edge due to the free-electron screening effect (Li et al. 2014b). Therefore, it is interesting to examine the influence of Al on the optical constants of ZnO.

The electronic traps of ZnO are mainly localized at the grain boundaries and usually adsorbed with oxygen. They also capture some donor state electrons (Deng 2016). Consequently, the Schottky barrier capacitance is found to be frequency dependent due to the finite time constants τ (τ=(1/ω)=RC) of the deep trap states generated in the depletion layer (Arda et al. 2013). AC impedance spectroscopy is able to separate the electric response in different regions of ZnO within the range of τ (Ram et al. 2016). This response is entirely characterized by the complex dielectric parameters that govern ac field frequency response (Sedky et al. 2022a).

Materials with high dielectric constants such as ZnO have received special attention due to the rapid developments in telecommunications and multilayer capacitors are required for microwave integrated circuits (Tripathi et al. 2010, Saadi et al. 2020, Sajid Ali et al.
2 Experimental details

Zn$_{1-x}$Al$_x$O samples with $x = 0.00, 0.05, 0.10, 0.20$ were synthesised by using the solid-state reaction method. The powders of ZnO and Al$_2$O$_3$ (Aldrich 99.999 purity) are thoroughly mixed in the required proportions and calcined at 900°C in air for a period of 16 h. The resulting powders are ground, mixed, and pressed into discs (1 cm in diameter and 0.3 cm in thickness). The pellets are then separately sintered at temperature of 1200°C for 12 h in the air and then suddenly cooled (quenched) in air to room temperature. The bulk density of the samples is measured in terms of their weight and volume. The phase purity of the samples was tested by XRD using Cu-Kα radiation of wave length 1.5418 Å at 40 kV and 30 mA settings, and (20°-70°) diffraction angles with a step of 0.06°. The structural morphology of the samples is obtained in powder form by scanning electron microscope (SEM) JSM 5400 LV with 2 μm bar length and 7500 amplification. FTIR absorption spectra of the samples were carried out in the range of (400—4000 cm$^{-1}$), with 4 cm$^{-1}$ resolution and 2 (cm/s) scanning speed using a spectrum 400-FT-1R/FT-NIR spectrometer. The samples were homogenized in a spectroscopic grade of KBr carrier in an agate mortar and pressed into 2 mm pellets. The optical properties at room temperature were measured against wavelength (200–1000 nm) using a JascoV-570 (Japan) computer programmable double beam with UV–visible–NIR spectrophotometer at standard incidence with a scan speed of 1000 mm min$^{-1}$. The complex dielectric measurements were obtained at room temperature using a precision impedance analyzer model 4295A in the frequency range of 1 to $10^7$ Hz and 1 V Ac amplitude. The pellets are sandwiched between two gold-plated
stainless steel electrodes of 20 mm in diameter in parallel plate geometry. The ac conductivity and modulus of complex impedance $Z$ as a function of frequency are well recorded. From the impedance spectra, values of the real $Z'$ and the imaginary $Z''$ parts of the complex impedance could be obtained.

3 Results and discussion

3.1 Structural analysis

It is evident from the XRD patterns shown in Fig. 1a that all samples show a wurtzite structure ICSD 01–079-0208 (space group P63mc) of ZnO. No additional lines could be formed for the samples of $x=0.00$ and 0.025, but as $x$ is increased to and above 0.025, some unidentified lower peaks could be seen at $2\theta = 30.96^\circ$ for $x=0.05$; $2\theta = 30.96^\circ$, 55.5$^\circ$, 60$^\circ$ for $x=0.10$; and $2\theta = 30.96^\circ$, 48.74$^\circ$, 55.5$^\circ$, 60$^\circ$ for $x=0.20$. This is because the bond energy of $\text{Al}_2\text{O}_3$ is higher than ZnO, and therefore more energy is required for Al ions to enter the ZnO lattice to form the Al-O bond, and therefore some of secondary phases could be formed (Sajid Ali et al. 2012; Khan et al. 2013). The lattice parameters $a$ and $c$, listed in Table 1, are gradually decreased by increasing $x$. The $c/a$, U-parameter and ZnO bond length $L$ are almost the same as reported for ZnO (Mansour Mohamed 2021; Morais et al. 2018). This is due to the smaller ionic size of $\text{Al}^{3+}$ (0.51 Å) than that of $\text{Zn}^{2+}$ (0.74 Å) at the same tetrahedral fold-coordination (Samanta and Goswami 2018; Sedky 2020). As $x$ increases to 0.10, the porosity calculated by; $PS = [1 - (\rho_{\text{exp}}/\rho_{\text{th}})]$ decreases slightly, followed by an increase at $x=0.20$. This behavior indicated that Al up to 0.10 helps as much as possible for decreasing the number of porous ZnO. However, porosity is the quality of ZnO of being porous, and it is usually evaluated by the ratio of the volume of interstices of ZnO to the volume of its mass. Therefore, Al up to 0.10 can enter the ZnO lattice and thus decreased the volume of interstices, which in turn decreased the porosity. However, at $x=0.20$, some of the Al ions are unable to localize in the ZnO lattice due to the solubility limit, so the porosity was increased, in consistent with XRD analysis.

The crystallite size $D_{hkl}$ given by $D_{hkl} = \frac{K\lambda}{\beta\cos\theta}$ (Seetawan et al. 1302) for the most intense peaks listed in Table 1 are decreased by $x$ to 0.20. The vice is versa for dislocation density $\delta$ calculated by $(1/D_{hkl}^2)$. The micro-lattice strain $\varepsilon_{\text{ms}} = \frac{\beta\cos\theta}{4}$ listed in Table 1 is positive and increased by Al. While lattice strain $\varepsilon_{\text{ls}} = (c-c_0)/c_0$ is positive for pure ZnO, but it changes to negative for the doped samples. Regardless of its sign, this behavior indicates that Al helps as a possible ZnO for few lattice defects or distortions. (Wojnarowicz et al. 2018; Aljaafari and Sedky 2020). The SEM micrographs shown in Fig. 1b indicates that nearly, no second phases are formed at grain boundaries, but the grains are randomly distributed over the matrix structure. For simplicity, the average size of the grain ($D_{\text{SEM}}$) is determined by the linear intercept method with the help of expression, $D_{\text{SEM}} = \frac{L_{\text{SEM}}}{M\text{N}}$ (Wang and Zhang 2017), where $L$ is the length of a random line on the micrograph, $M$ is micrograph magnification, and $N$ is the grain boundary number that is intercepted by the lines. As listed in Table 1, Al content up to $x < 0.10$ decreased the average grain size of ZnO $D_{\text{SEM}}$, followed by an increase at $x=0.20$, which is consistent with $D_{hkl}$ behavior obtained from XRD analysis.
Fig. 1  a XRD patterns for pure and Al doped ZnO samples  b SEM micrographs for pure and Al doped ZnO samples
3.2 FTIR analysis

Figure 2a shows FTIR spectra against wave number $\tilde{\nu}$ for the samples, and also the values of wave numbers $\nu$ against absorption peaks are listed in Table 2. Firstly, there is a clear peak recorded at 3423.36, 3442.14, 3417.14, 3417.34, 3442.18 cm$^{-1}$, respectively, which is related to O–H stretching vibrations for ZnO (Zhao 2016; Wasly 2018). One peak is only recorded at 2924.21 cm$^{-1}$ for the $x = 0.20$ sample, but it disappears for the other samples, even for ZnO, which may be related to the solubility limit discussed above. Interestingly, the functional peak observed at 1630 cm$^{-1}$ due to H–O-H bonding vibrations and C = C stretching, is absent for all samples (Muhammad 2019). Some other peaks were only obtained at 1426.07 and 1436.68 cm$^{-1}$ for $x = 0.00$ and 0.20. In contrast, there are also identified peaks recorded at 673.56, 672.18, 671.84, 668.13 for $x = 0.00$, 0.025, 0.05 and 0.10, but it is absent for $x = 0.20$. However, the presence of the peak is evidence of the incorporation of Al$^{3+}$ ions into the ZnO matrix and corresponds to the C=C, C=O, C–N and O–H stretching and C–H bending vibration (Srinivasulu et al. 2017). Among the absorption peaks observed between (451.97–556.32 cm$^{-1}$) which corresponds to the active modes of ZnO related to hexagonal structure Santhosh Kumar (2017). This means that the ZnO structure is slightly different than the ideal case for $x = 0.20$, as obtained from XRD analysis. This behavior indicates that Al shifted the bending and stretching vibrations of ZnO to lower values of $\tilde{\nu}$, which can be attributed to the formation of a ZnAl$_2$O$_4$ bond of a longer length chain and the fact that the Al had intense ZnO and moderate metal oxide modes in the ZnO matrix structure (Raja et al. 2014).

Debye temperature $\theta_D$ is given by Ba-Abbad et al. (2013);

$$\theta_D(K) = \frac{\hbar c \Delta \nu}{k_B} = 1.439 \Delta \nu$$

(1)

Table 1

| x     | a (Å) | c (Å) | c/a | V (Å$^3$) | $\nu_{\text{abs}} \times 10^{-3}$ | U | L (Å) |
|-------|-------|-------|-----|-----------|----------------------------------|---|-------|
| 0.00  | 3.252 | 5.208 | 1.601 | 47.697    | 0.27                             | 0.379 | 1.991 |
| 0.025 | 3.247 | 5.201 | 1.602 | 47.486    | –                                 | 0.380 | 1.987 |
| 0.05  | 3.242 | 5.196 | 1.603 | 47.295    | −2.04                            | 0.380 | 1.987 |
| 0.10  | 3.231 | 5.185 | 1.604 | 46.902    | −3.57                            | 0.380 | 1.990 |
| 0.20  | 3.237 | 5.191 | 1.604 | 46.665    | −4.92                            | 0.380 | 1.981 |

| x     | $\rho_{\text{th}}$ (g/cm$^3$) | $\rho_{\text{exp}}$ (g/cm$^3$) | $P_s$ | D$\text{XRD}$ (nm) | $\delta \times 10^{-4}$ (nm)$^{-2}$ | $\varepsilon_{\text{ms}} \times 10^{-3}$ | D$\text{SEM}$ (µm) |
|-------|-------------------------------|-------------------------------|-------|-------------------|----------------------------------|---------------------------------|------------------|
| 0.00  | 5.794                         | 5.451                         | 0.059 | 41.830            | 5.72                             | 3.32                            | 2.060             |
| 0.025 | 5.630                         | 5.312                         | 0.056 | 41.167            | 5.90                             | 3.376                           | 1.790             |
| 0.05  | 5.529                         | 5.244                         | 0.051 | 38.983            | 6.58                             | 3.566                           | 1.610             |
| 0.10  | 5.253                         | 5.053                         | 0.038 | 37.230            | 7.21                             | 3.734                           | 1.780             |
| 0.20  | 5.061                         | 4.710                         | 0.069 | 39.637            | 6.37                             | 3.507                           | 2.230             |
Figure 2b shows that $\theta_D$ suddenly increases as $x$ increases to 0.10, followed by a sudden decrease at $x = 0.20$. However, this behavior is attributed to the shift in the wave number of FTIR bands to higher/lower values against $x$ content as listed in Table 2.

The strength of the interaction between Zn cat-ions and (O$^{-2}$) ions of ZnO, the force constant $K_t$ can be calculated using; $K_t = 0.076W\Delta\nu^2$ (Santhoshkumar et al. 2017).

Also, the stiffness constants $S_{11}$ and $S_{12}$ are easily calculated in terms of Poisson’s ratio $\gamma$ as; $S_{11} = (K_t/c)$, $S_{12} = (S_{11}\gamma /1-\gamma)$ and $\gamma = 0.324$ (1–1.043PS) (Ahmad et al. 1456). It is
evident from Table 2 that (γ > 0.26) for all samples which indicates ductile nature (Mitra and Mondal 2013). The residual stress is calculated by El-Said Bakeer (2020); where \( S_{13}, S_{33}, S_{11}, S_{12} \) are respectively equal 104.2, 213.8, 208.8 and 119.7 GPa for ZnO (Mazen et al. 2007). As listed in Table 2, \( \sigma \) is negative (compressive-like behavior) for ZnO. In contrast, it is positive (tensile-like behavior) for the doped samples, indicating that Al was able to eliminate the compressive stress of ZnO and turn it into tensile stress. This may be related to the number of pores and the lattice defects discussed above (Anupama et al. 2017). Young Y, bulk modulus \( \beta \) and rigidity modulus \( G \) are determined by Modi et al. (1543); MariemChaari and matoussi et al., 2011);

\[
Y = \frac{(S_{11} - S_{22}(S_{11} + 2S_{12})}{(S_{11} + S_{12})}; \quad \beta = \frac{S_{11} + 2S_{12}}{3}; \quad G = \frac{Y}{2(\gamma + 1)}
\]

As shown in Fig. 3, both of them are increased as \( x \) increased to 0.10, followed by a decrease at \( x=0.20 \). This increase may be related to the shrinkage of inter-atomic bond length \( L \) which subsequently controls the ZnO bond strength (Rao et al. 2010).

### 3.3 Optical measurements

As shown in Fig. 4a-b, the relationship between reflectance/absorption (R/A) and wave lengths reveals two minimum/maximum regions for all samples at about 400 nm, which may be related to increasing free carrier density in the near-UV region (Rusu et al. 2011).
Interestingly, the change in (R/A) for Al-doped samples is related to the oxygen vacancies, which usually make ZnO suitable for gas sensors (Patange et al. 2013; Rajesh Babu and Tatarchuk 2018). As given in Table 3, the first A_{max} or R_{min} occurs at (\lambda (1)_{max} = 314 nm) for all samples, while the second A_{max} or R_{min} occurs at \lambda (2)_{max} = 352, 364, 344 and 346 nm, respectively. Therefore, first exciton energy E_{ex}(1) is 3.953 eV for all samples, while the second exciton E_{ex}(2) is decreased for x = 0.025, followed by an increase to x = 0.20. This means that the required energy for creating electron–hole pairs is decreased by 0.025 of Al, followed by an increase of up to 0.20. Generally, E_{ex} of ZnO is the required photon energy for electronic transition from the VB of (O-2p) to the (Zn-3d) of CB to form an exciton bound state (e–h) of 100 meV of binding energy close to E_g. The (e–h) are usually held together by the electrostatic Coulomb force and they have kinetic energy and a narrow spectrum, which is highly attractive for laser devices if we can trap the excitons and reduce their motion, which further narrows the energy spread (Chen and Li 2019, Rodnyi et al. 2018b, Al Naim and Sedky 2021).

The absorption coefficient \alpha is calculated by, \alpha = 2.303 \ln (A/t), and the optical band gap E_g is obtained using Taucs equation (Ali et al. 2005; Chen and Li 2019);

\[
(\alpha h\nu)^{m} = A(h\nu - E_{s})
\]

(4)

m represents the transition in the semiconducting materials and has values of 1/2 and 2 for allowed direct and indirect transitions, respectively, and B is a constant, and t is the diameter of the glass cuvette. The plot of (\alpha h\nu)^{m} against the photon energy is used to get the value of the E_g (see Fig. 5). As shown in the figure, two absorption edges are observed for all samples, indicating that there are two direct electronic transitions due to the absorption of the photon energy. The values of E_g are obtained from the Figure are summarized in Table 3. The first (E_{gh}) is obtained at UV region, while the second one (E_{gl}) is obtained in the visible region. The values of E_{gh} and E_{gl} are 3.60, 3.56, 3.43, 3.53, 3.68 eV and 3.02, 2.95, 2.85, 2.92, 3.07 eV respectively. This suggests both E_{gh} and E_{gl} had similar behavior as the x increased to 0.20. It is also evident from Table 2 that \Delta E = (E_{gh} - E_{gl}) is 0.58, 0.61, 0.58 and 0.61 eV for all samples, in which \Delta E is constant. Anyhow, two E_g are also

Fig. 3 Elastic modulus (Y,B,G) versus Al content for the samples
recorded for n-type semiconductors (Rodnyi et al. 2018b; Al- and Naim and Sedky, 2021; Mukhtar et al. 2012). In that case, the doping of ZnO with Al may produce a high carrier concentration and, as a result, more conduction band energy states are formed, which, in turn, reduces the $E_g$ as obtained. However, the further increment of $E_g$ above $x = 0.05$ may be due to the formation of structural defects in the host materials, thereby creating insulating states in the forbidden gap inside the band structure, and therefore, $E_g$ was slightly increased.

**Fig. 4**  
(a) Reflectance (R) versus wave length for pure and Al doped ZnO samples  
(b) Absorption (A) versus wave length for pure and Al doped ZnO samples
Table 3  \( \lambda, E_{ex}, E_{g}, \epsilon_L, (N/m^*) \), \( N \) and \( R \) for the samples

| \( x \) | \( \lambda(1)_{max} \) (nm) | \( \lambda(2)_{max} \) (nm) | \( E_{ex} \) (1) (eV) | \( E_{ex} \) (2) (eV) | \( E_{gh} \) (eV) | \( E_{gL} \) (eV) | \( \epsilon_L \) | \( (N/m^*) \times 10^{34} \) (g.cm\(^{-2}\)) | \( R \times 10^{-11} \) (cm) |
|--------|----------------|-----------------|----------------|----------------|----------------|----------------|--------|-----------------|----------------|
| 0.00   | 314            | 352             | 3.953          | 3.526          | 3.60           | 3.02           | 8.78   | 2.64            | 8.77            |
| 0.025  | 314            | 364             | 3.953          | 3.410          | 3.56           | 2.95           | 5.06   | 3.01            | 8.40            |
| 0.05   | 314            | 350             | 3.953          | 3.547          | 3.43           | 2.85           | 4.04   | 3.51            | 7.98            |
| 0.10   | 314            | 344             | 3.953          | 3.608          | 3.53           | 2.92           | 3.48   | 3.81            | 7.76            |
| 0.20   | 314            | 346             | 3.953          | 3.588          | 3.68           | 3.07           | 9.37   | 18.45           | 4.59            |
In addition, the high frequency dielectric constant of $\text{Zn}_{1-x}\text{Al}_x\text{O}$ is calculated from the following relation, which represents the dependence of the reflective index $n$ on the wavelength $\lambda$ (Othman et al. 2017; Ismail et al. 2011a):

$$n^2 = \varepsilon_L - \left( \frac{e^2}{4\pi^2c^2\varepsilon_0} \right) \left( \frac{N}{m^*} \right) \lambda^2$$  \hspace{1cm} (5)

where $c$, $e$, $m^*$, $\varepsilon_L$, and $N$ denote light speed, electron charge, free space permittivity, effective mass of electron, residual lattice dielectric constant, and free carrier concentration, respectively. The intercept and slope of the straight lines in Fig. 6 give the values of $\varepsilon_L$ and $N/m^*$, which are listed in Table 3, are slightly increased by $x$ up to $x = 0.10$, followed by a sharp increase at $x = 0.20$, whereas $\varepsilon_L$ is decreased to $x = 0.10$, followed by a sharp increase at $x = 0.20$. The inter-atomic distance ($R$) given by $(0.86/N^{(1/3)})$ (Abdel-Khalek et al. 2019) and listed in Table 3 decreases as $x$ increases to 0.20. Based on the above results, the higher valence state of the $\text{Al}^{3+}$ compared to $\text{Zn}^{2+}$ led to slightly deeper donors and, consequently, more carriers were generated. Furthermore, and in parallel, the ability of electron migration may be enhanced (Ismail et al. 2011b). Furthermore, $\text{Al}$ is nearly non-magnetic, and the addition of $\text{Al}$ beside $\text{Zn}$ will decrease the localization of the carriers. Therefore, we strongly recommend the $\text{Al}$ doped samples for optoelectronic applications.

### 3.4 Dielectric measurements

The real $\varepsilon'$ part of the sample dielectric constant is determined by using the relation (El-Denglawey and El-Denglawey 2018; Sagadevan et al. 2017):

$$\varepsilon' = \frac{Cd}{\varepsilon_0A}$$  \hspace{1cm} (6)
where C is the capacitance of the sample and $C_0$ is the empty cell capacitance ($C_0 = \varepsilon_0 A/d$), $d$ is the sample thickness, $A$ is the surface area of the electrodes, $\varepsilon_0 = 8.85 \times 10^{-12} \text{F/m}$ is the permittivity of free space. Figure 7 shows the plot of $\varepsilon'$ against $f$ for all samples. It is clear that $\varepsilon'$ is gradually decreased as $f$ increases up to $10^4$ Hz for all samples, and above
that it decreases, then saturated at $10^7 \text{ Hz}$. In addition, it is slightly increased as $x$ increases up to 0.0.025, but there is a significant increase with a further increase of $x$ above 0.025. For example, $\varepsilon'$ of $x=0.20$ is at least about $10^3$ times that of $x=0.00$, which indicates that the addition of Al above 0.025 to Zn shifts the dielectric medium to higher values. Therefore, when a field is applied, the charge carriers can easily move within the grains at a low $f$ below 10 MHz, but they are accumulated at the grain boundaries, resulting in the large interfacial polarization, and, in turn, high $\varepsilon'$. With a rise in frequency above 10 MHz, a decrease of $\varepsilon'$ could be obtained. This is due to the fact that any species contributing to polarization is found to lag behind the applied field, and consequently, the dipoles cannot follow the ac field (Abdel-Khalek et al. 1178; Hazem Mahmoud Ali and Ahmed Mohamed Abdel Hakeem 2015; Selvakumar et al. 2014). On the other hand, the increase of $\varepsilon'$ with $x$ can be explained in terms of increasing electro-negativity between Al and O, which is 1.83 instead of 1.79 between Zn and O, and also increasing the number of carriers as indicated above (Mehedi Hassan et al. 2015; Singh et al., 2015). Due to this, the strength of the Al-O ionic bond is improved, which results in an increase in the dielectric polarization. Moreover, the decrease in crystallite size and average crystalline size leads to an increase in the number of dipoles, which results in a decrease in polarization and increases the $\varepsilon'$ values for the doped samples, as reported (Zamiri et al. 2014a).

The total conductivity can be obtained by Zamiri et al. 2014b;

$$\sigma_{\omega}(\omega) = \sigma_{dc} + \sigma_{ac} = \sigma_{dc} + B\omega^s$$

(7)

where $\sigma_{dc}$ is the dc conductivity calculated at zero $f$, $B$ is constant, and $s$ is the exponent of frequency. Figure 8 shows the real part ($\sigma'$) of $\sigma_{ac}$ as a function of $f$. It is evident from the figure that $\sigma'$ gradually increases by increasing $f$ for all samples up to 10 MHz, obeying the universal power law (Singh and Singh 2020a). In addition, $\sigma'$ for Al-doped samples is higher than that of ZnO due to the increased charge carriers through the replacement

![Fig. 8 Ac conductivity (σ\') versus frequency for pure and Al doped ZnO samples](image-url)
of Zn²⁺ by Al³⁺. At low f, the field drift carries over a large distance, and therefore σₜ is almost unchanged. As f is raised, the displacement of carriers is reduced, and at a critical frequency ωₚ, σₜ follows the relation: \( \sigma_{ac} \sim \omega^s \) with \( 0 \leq s \leq 1 \) for hopping conduction (Saadi et al. 2020). These outcomes indicate that the addition of Al supports the \( \sigma_{ac} \) of ZnO as obtained.

However, the linear fit between \( \ln \sigma' \) and \( \ln \omega \) shown in Fig. 9a,b gives two different linear regions. The obtained values of \( s \) and \( B \) at low and high frequencies, designated as \( s_L \), \( s_h \), \( B_h \) and \( B_L \) are obtained and listed in Table 4. The values of \( s_L \) are between 0.565, 0.898

![Fig. 9](image_url)

**Fig. 9**  a  lnσ' versus lnω at low frequency for pure and Al doped ZnO samples  b  lnσ' versus lnω at high frequency for pure and Al doped ZnO samples
for x = 0.00 and 0.025 samples, which indicates electronic conduction. But with increasing x above 0.025, the values of s_L are 0.023, 0.031, 0.011, which indicates hole conduction. However, similar behavior is obtained for s_h. This behavior is attributed to the excess number of hole carriers which may be generated as a result of the difference between Zn^{2+} and Al^{3+}.

According to correlated barrier hopping (CBH) model, the binding energy W_m, is given by Samanta et al. (2020);

\[ s = 1 - \frac{6K_BT}{W_m + K_BT\ln \omega \tau_0} = 1 - \frac{6K_BT}{W_m} \]

where \( \tau_0 \) is the characteristic relaxation time and is in the order of the atomic vibration of \( 10^{-13} \) s. The values of \( W_m \) correlated with the range of \( f \) are listed in Table 4. Although they are decrease with x, the W_{mh} \approx 1.5 W_{mL} except for x = 0.00 in which W_{mh} \approx 0.94 W_{mL}. In addition, the minimum hopping distance (R_{min}) is given by Samanta et al. (2020);

\[ R_{min}(\omega) = \frac{e^2}{\pi \varepsilon_0 \varepsilon \ln (W_m + K_BT \ln \omega \tau_o)} \sim \frac{e^2}{\pi \varepsilon_0 \varepsilon \ln W_m} \]

where \( W_m = (W_{mL} + W_{mh})/2 \). On the other hand, the density of states at which carriers exist N can be expressed by Samanta et al. (2020);

\[ N(\omega) = \frac{24\sqrt{\gamma}}{\pi^3 \varepsilon_0 \varepsilon \alpha R_{min}^6} \]

As shown in Fig. 10a, the unusual behavior of R_{min} can be classified into two series relative to x = 0.00. The vice is versa for the behavior of N(\omega) against the x, see Fig. 10b. These results indicate that the R_{min} mechanism against f is controlled by the x in the low under doped region (x = 0.025) and also in the over-doped region (x = 0.20).

In addition, the density of states at Fermi level N(E_f) can be calculated by Singh et al. (1832);

\[ N_{E_f}(\omega) = \sqrt{\frac{3\sqrt[5]{\alpha^5}}{4e^2\omega K_BT(\ln \frac{10^{13}}{\omega\alpha})^2}} \]

where \( \alpha \) is the localized wave function and equals \( 10^{10} \) m^{-1} and \( f_o = 10^{13} \) Hz is the photon frequency corresponding to the applied ac field. However, the behavior of N(E_f) against x shown in Fig. 11a seems to be systematic because N(E_f) is gradually increased as x increases to 0.20. But it has unusual behavior against f where it gradually increases for

| Al content x | \( \sigma_{dc} (\Omega \cdot \text{cm}^{-1}) \) | s_L | s_h | B_L | B_h | W_{mL} (eV) | W_{mh} (eV) |
|-------------|----------------|------|------|-----|-----|----------------|----------------|
| 0.00        | 3.E-11          | 0.898| 0.892| 7.64E+15 | 1.18E+14 | 1.520           | 1.435           |
| 0.025       | 2.E-10          | 0.565| 0.700| 2.80E+12 | 1.26E+12 | 0.356           | 0.517           |
| 0.05        | 8.E-08          | 0.023| 0.351| 7.41E+08 | 4.76E+09 | 0.159           | 0.239           |
| 0           | 7.E-08          | 0.031| 0.287| 2.43E+07 | 2.88E+08 | 0.160           | 0.217           |
| 0.20        | 6.E-08          | 0.011| 0.234| 2.10E+07 | 3.33E+08 | 0.157           | 0.202           |
x = 0.00 and 0.025 samples, goes to an optimum, and then decreases. But with increasing x above 0.025, N (E_F) decreases, increases and goes to an optimum and then decreases. The increase in N (E_F) against x confirms the increase in σ_{ac}, and may support the growing concentration of defect energy states in ZnO (Zeyada et al. 2016). Our interesting point here is the optimum values of N (E_F) obtained at about 195 kHz for all samples and also its dependence on x. For example, N (E_F) was increased from 8.05 × 10^{19} (erg.cm^{-3})^{-1} for x = 0.00 to 2.01 × 10^{20}, 8.97 × 10^{20}, 1.75 × 10^{21} and 1.78 × 10^{21} (erg.cm^{-3})^{-1} for the doped samples, respectively. This behavior is not clear to us at present and may be either related
to the resonance of lattice vibrations against ac field/or the enhancement of ac conductivity at 195 kHz. The higher value of F, as a figure of merit given by, \( \frac{\sigma_{ac}}{\varepsilon} \) is mainly a good indicator for a sample to be more suitable for solar cell applications (Loane et al. 1991; Elliott 1987). As shown in Fig. 11b, the F factor is gradually increased by increasing x, but it is almost constant above \( x = 0.05 \). These results strongly recommend the Al doped samples up to \( x = 0.10 \) for solar cell design.
Figure 12a, b shows the variations of $Z'$ and $Z''$ against $f$ for all samples. It is evident that $Z'$ and $Z''$ for $x \leq 0.025$ samples are decreased linearly as a straight line with a rise in $f$. In addition, $Z'$ was also decreased as $x$ increased, whereas $Z''$ was increased. With a further increase of $x$ above 0.025, $Z'$ is slightly increased with $f$ and goes to higher values at about 10 kHz, and then decreases at 10 MHz. It also decreases as $x$ increases below 100 kHz, but above that it is nearly constant for all samples. In contrast, a little bit of a decrease is obtained below 10 kHz for $Z''$ against $f$ followed by a significant increase with the rise in $f$ to 10 MHz, but it becomes higher than that of $x \leq 0.025$ samples. However, the increases
in $Z$ at low $f$ confirm the highly resistive grain boundaries, whereas the decrease is due to the rise in the hopping of charge carriers (Sedky et al. 2022b; Cherifi 2016). Anyhow, the Cole- Cole plot and also the zoom at high frequencies are shown in Fig. 13a-d and the impedance of grains and grain boundaries is determined at the half and end of the semi-circle-shape plot, respectively. It is evident that the impedance spectra of $x = 0.00$ sample is straight line like- shape with $Z^\prime = 2.05 \times 10^{10} \Omega$ and $Z^\prime\prime = 1.39 \times 10^8 \Omega$. With increasing $x$ to 0.025, it becomes half semicircle (one quarter) like-shape, and with more increase of $x$ to 0.05, a complete semicircle like-shape with a large radius is obtained. Similar behavior is also obtained for $x \geq 0.10$, but the area of the semicircle is gradually decreased as $x$ increases. However, the values of the impedance of grain and grain boundaries for all doped samples $Z'(g)$, $Z'(gb)$, $Z''(g)$ and $Z''(gb)$ are listed in Table 5, in which they are gradually decreased as $x$ increases to 0.20.

It can be seen that the frequency used for a pure sample is insufficient to cover the grain boundary, and thus the $Z(g)$ and $Z(gb)$ cannot be calculated. The $Z(gb)$ was reduced by increasing $x$ to 0.025, and the frequency was able to make it as one quarter of a circle because

| $x$  | $Z'(g)$ ($\Omega$) | $Z''(g)$ ($\Omega$) | $Z'(gb)$ ($\Omega$) | $Z''(gb)$ ($\Omega$) |
|------|-------------------|-------------------|-------------------|-------------------|
| 0.00 | –                 | –                 | –                 | –                 |
| 0.025| $3.19 \times 10^9$ | $6.19 \times 10^9$ | $6.38 \times 10^9$ | $4.92 \times 10^8$ |
| 0.05 | $4.59 \times 10^5$ | $2.65 \times 10^5$ | $10.02 \times 10^5$ | $4.15 \times 10^4$ |
| 0.10 | $3.01 \times 10^5$ | $1.65 \times 10^5$ | $6.01 \times 10^5$ | $1.56 \times 10^4$ |
| 0.20 | $1.88 \times 10^5$ | $1.05 \times 10^5$ | $4.41 \times 10^5$ | $1.13 \times 10^4$ |
the $Z(gb)$ was reduced, and it can be obtained approximately. With an increase in $x$ above 0.025, the applied frequency is more than adequate for covering the grain boundary, allowing the $Z(g)$ and $Z(gb)$ to be calculated. However, a single arc of an identical $\tau = (1/\omega) = RC$ has been obtained in pure and doped ZnO (Ramesh Babu et al. 2012; Giuntini et al. 1981). Because of this behavior, conduction in grains and their boundaries could not be separated (Fahim et al. 2018). Other reports believed that a single arc was caused by the combined contributions of the grain’s core resistive and grain boundary (Singh and Singh 2020b). This fact suggests that the excess of doping probably exists in the grain boundary region, either as a very thin secondary phase, which could benefit the grain boundary transport for grain growth (Elkar et al. 2018).

4 Conclusion

Structural, FTIR, optical and dielectric properties of $\text{Zn}_{1-x}\text{Al}_x\text{O}$ were investigated. Regardless of the $x = 0.20$ sample, which shows unusual behavior, the lattice constants, crystallite diameter, porosity, and average crystalline size are decreased by Al, while the Debye temperature and elastic modulus are increased. The residual stress is compressive for the ZnO, but it is tensile for the doped samples. Although two different values of $E_g$ are apparent for each sample, the $\Delta E = (E_{gh} - E_{gl})$ between them is almost constant. Further, the residual dielectric constant is decreased as $x$ increases to 0.10, followed by a sharp increase at $x = 0.20$, while the opposite behavior is obtained for (N/m$^3$). The conduction is electronic for $x \leq 0.025$ samples, but it is changed to hole for $x > 0.025$. Although $W_m$ decreased by increasing $x$, there was no exact trend for the behaviors of $R_{mn}$ and $N$ against $x$. Interestingly, $N (E_F)$ has an optimum value at 195 kHz for all samples. The F-factor for solar cell design is increased as $x$ increases to 0.10, but it is almost constant at $x = 0.20$. The Cole–Cole plot for the samples is a straight line for $x = 0.00$, a semicircle arc for $x = 0.025$, and a complete semicircle for $x \geq 0.05$. The $Z(G)$ and $Z(GB)$ are gradually decreased by increasing $x$ to 0.20. These outcomes strongly recommend the AlZnO samples for the design of optoelectronic and solar cell instruments.

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Declarations

Conflict of interest The authors declare that they have no competing interests and the present work is original not published or submitted for evaluation to another Journal. Moreover, the obtained systematic findings or study strongly recommend the Al doped samples for advanced applications such as Optoelectronic and solar cell instruments, which probably have never been early reported elsewhere over the last 3-years, which and highlight the present investigation. In addition, the datasets generated during and/or analyzed during the submitted current study are only available from the corresponding author on reasonable request.

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