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Structural, FTIR, Optical and Dielectric Properties of $\text{Zn}_{1-x}\text{Al}_x\text{O}$ Ceramics For Advanced Applications

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Abstract

We report here the structural, FTIR, optical and dielectric properties of $\text{Zn}_{1-x}\text{Al}_x\text{O}$ with $x = 00.00 < x \leq 0.20$). The wurtzite structure is conformed to all samples and the lattice constants, crystallite diameter, porosity and average crystalline size are generally decreased. The residual stress is compressive for pure samples, but it is changed to tensile for the doped samples. Interestingly, Debye temperature and elastic modulus are increased as x increases to 0.10, followed by a decrease at $x = 0.20$. Two different energy gaps E_{gh} and E_{gl} are apparent for each sample, corresponding of two transition absorption peaks. Interestingly, the $\Delta E = (E_{gh} - E_{gl}) \sim 0.60$ for all samples. Further, the residual dielectric constant is decreased by increasing x to 0.10, followed by a sharp increase at $x = 0.20$ while the opposite behavior for (N/m^*) . The dielectric constant ϵ' is slightly increased as x increases to 0.025, followed by a sharp increase as x increases to 0.20, as well as the ac conductivity σ' . The conduction is electronic for $x \leq 0.025$ samples, but it is changed to hole with an increase of x to 0.20. The binding energy W_m was decreased as x increases to 0.20, but there is no exact trend against x for the behaviors of minimum hopping distance R_{min} and density of localized states N . In addition, the density of states at Fermi level $N(E_F)$ 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 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 $Z'(g)$ and grain boundaries $Z'(gb)$ are gradually decreased by increasing x 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: Residual stress; Elastic constants; Energy Gap; Ac Conductivity and F-Factor

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1- Introduction

Wurtzite structure of pure n-type ZnO with 3.2 eV and 60 Urbach energy has been used in optoelectronic and solar cell devices [1-4]. The sintering of ZnO and also doping by 3-d elements (TM) exhibits intrinsic defects due to oxygen vacancies and in turn introduces donor states below CB of ZnO to be convenient for such devices [5-13].

The vibration of the lattice in solids in general and in ZnO compounds in particular has been considered early as the subject of many applications, such as optical Kerr shutters (OKS), switching devices, and detectors. Many experimental techniques have been utilised to assess the suitability of ZnO-based compound materials for such applications.

Among those, FTIR analysis significantly helps in approximating the relationship between the spin vacancy and the carrier concentration. Furthermore, it gives an ascertainable indicator of the purity and nature of the metal oxides in the compound. The absorption bands of ZnO arising from interatomic vibrations, stretching and deformation of O-H are assigned to water adsorption on the Zn surface, and some other bands [14-17].

The mechanism of the ZnO conduction band (CB) usually arises when the 4s orbital of Zn is wide enough to allow for effective charge transfer [18-20]. Consequently, the electrons are excited from the valence state due to the absorption of light, and consequently the electron-hole pairs are generated. Above the limit of the ZnO band gap, the recorded signals of photon energy are due to non-radiative and Urbach tail processes. Furthermore, the energy of photons for creating electron-hole pairs can be considered only for materials with a larger Urbach energy like ZnO [21].

The electronic traps of ZnO are mainly localized at the grain boundaries and usually adsorbed oxygen and capture some donor state electrons [22]. Consequently, the Schottky barrier capacitance is found to be frequency dependent due to the finite time constants τ ($\tau = (1/\omega) = RC$) of the deep trap states generated in the depletion layer [23-24]. Ac impedance spectroscopy is able to separate the electric response in different regions of ZnO within the range of τ [25-26]. This response is entirely characterized by the complex dielectric parameters against ac field frequency response [27-29].

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 [30-32]. In contrast, materials that have a low dielectric constant are potential candidates for nonlinear optical and high-frequency devices, which are necessary for such applications [33]. However, some parameters have a great influence on improving ZnO dielectric constants, such as valence state, content, magnetic moment of TM, and also intrinsic defects [34-38].

The dielectric properties of ZnO can be tuned by introducing dopants into the ZnO host matrix with 3-d transition-elements [39-47]. For example, the dielectric constants and ac conductivity of Fe, Ni, Co, and Mn doped into ZnO are significantly decreased. But, ac conductivity increases with an increase in frequency, and complex impedance analysis shows single circular arc type behavior, suggesting the dominance of grain resistance [48-50]. The density of localized states at the Fermi level $N(E_F)$ and the binding energy W_m decrease as the doping content increases. This behavior has been explained in terms of the frequency dependence of the s, p-d exchange interaction between the Zn and dopants in terms of Maxwell-Wagner type interfacial polarization and Koops's phenomenological theory [51-53].

The electrical properties of Al doped with ZnO are presented by Sedky et.al [54-55]. It is found the addition of Al generally decreases the nonlinear behavior of ZnO and, therefore, the dc electrical conductivity has increased. It is believed that the excess electron density and structural distortion, produced by Al doping in both under and over doped regions, are responsible for the above behavior. With this purpose in mind, structural, FTIR analysis, optical and dielectric behaviors will be examined on the same batch of samples. Interestingly, some important physical

parameters are well estimated, such as Debye temperatures, elastic constants, energy gap, dielectric loss, N/m^* , inter-atomic distance R , density of states at Fermi level $N(E_f)$, F -factor, and ac impedance for solar cell applications. This study may have recommended these samples for optoelectronic and solar cell investigation.

2- Experimental Details

$Zn_{1-x}Al_xO$ samples with $x = 0.00, 0.05, 0.10, 0.20$ were synthesized by using a conventional solid-state reaction method. The powders of ZnO and Al_2O_3 (Aldrich 99.999 purity) are thoroughly mixed in the required proportions and calcined at $900^\circ C$ in air for a period of 16 hours. The resulting powders are ground, mixed, and pressed into disks of 1 cm diameter and 0.3 cm thick. The pellets are then separately sintered at temperatures of $1200^\circ C$ for 12 hours 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 $Cu-K\alpha$ radiation of wave length $1.5418 \text{ \AA}$ at 40 KV and 30 mA settings, and $(20^\circ-70^\circ)$ diffraction angles with a step of 0.06° . FTIR absorption spectra of the samples were carried out in the range of $(400 - 4000 \text{ cm}^{-1})$, 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 grinding time was minimized as possible to avoid structure deformation, ion exchange and water absorption from the atmosphere. The optical properties at room temperature were measured against wavelength $(200-1000 \text{ nm})$ 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} . photometric accuracy of $(\pm 0.002-0.004)$ absorbance and $\pm 0.3\%$ transmittance is motioned. The dielectric properties are measured by broadband dielectric spectroscopy (BDS) utilizing a high resolution Alpha analyzer with an active sample head (Novo control GmbH). 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 . The amplitude of the AC electric signal applied to the samples is about 1 V. The surface of the pellets is well polished to dryness and metalized with a thin layer of silver paste in order to obtain two parallel plates. The pellets are sandwiched between two gold-plated stainless steel electrodes of 20 mm in diameter in parallel plate geometry. Fused silica fibers with a diameter of 50 mm are used as spacer material. 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 Figure 1 (a) that all samples show a wurtzite structure ICSD 01-079-0208 (space group $P6_3mc$) of ZnO . No additional lines could be formed for the samples of $x = 0.00$ and 0.025 samples, 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 Al_2O_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 phase could be formed [56-59]. 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 [60-65]. This is due to the smaller ionic size of Al^{3+} ($0.51 \text{ \AA}$) than that of Zn^{2+} ($0.74 \text{ \AA}$) at the same tetrahedral fold-coordination [66-70]. The porosity calculated by; $PS = [1 - (\rho_{exp}/\rho_{th})]$ is slightly decreased as x increases to 0.10 , followed by an increase at $x = 0.20$. This behavior indicated that Al up to 0.10 helps as possible for decreasing the number of pores of ZnO .

Figure 1(a): XRD patterns for pure and Al doped samples

The crystallite size D_{hkl} given by $D_{hkl} = \frac{K\lambda}{\beta \cos \theta}$ [71] for the most intense peaks listed in Table 1 are decreased by x to 0.20. The vice is versa for dislocation density δ calculated by $(1/D_{hkl}^2)$. The micro-lattice strain $\epsilon_{ms} = \frac{\beta \cos \theta}{4}$ listed in Table 1 is positive and increased by Al. While lattice strain $\epsilon_{ls} = (c-c_0)/c_0$ 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. [72-74]. The SEM micrographs shown in Figure 1(b) indicates that nearly, no second phases are formed at grain boundaries, but the grains are randomly distributed over the matrix structure. As listed in Table 1, Al content up to $x \leq 0.10$ decreased the average grain size of ZnO D_{SEM} , followed by an increase at $x = 0.20$ as obtained from XRD analysis.

Table 1: I_{avg} , a , c , c/a , V , U , L , δ , ρ , P_s , ϵ_{ms} , ϵ_{ls} , σ and n for the samples

x	a (Å)	c (Å)	c/a	V (Å) ³	ϵ_{ls} $\times 10^{-3}$	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	-1.08	0.380	1.987
0.05	3.242	5.196	1.603	47.295	-2.04	0.380	1.987
0.10	3.231	5.188	1.606	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	ρ_{th} (g/cm ³)	ρ_{exp} (g/cm ³)	P_s	D (nm)	$\delta \times 10^{-4}$ (nm) ⁻²	ϵ_{ms} $\times 10^{-3}$	D_{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 1(b): SEM micrographs for pure and Al doped samples

3.2- FTIR Analysis

Figure 2 (a) shows FTIR spectra against wave number $\bar{\nu}$ for the samples and also the values of wave numbers $\bar{\nu}$ 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 [75-76]. One peak is only recorded at 2924.21 cm⁻¹ for the $x = 0.20$ sample, but it disappeared 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 [77-78]. Some other peaks were only obtained at 1426.07 cm⁻¹ 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 an evidence for

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 [79-80]. Among the absorption peaks observed between ($451.97\text{-}556.32\text{ cm}^{-1}$) which corresponds to the active modes of ZnO related to hexagonal structure [81-82]. 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 $\bar{\nu}$. This is attributed to formation of ZnAl_2O_4 bond of more length chain and really that the Al had intense ZnO and moderate metal oxide modes in the ZnO matrix structure [83-84].

Figure 2 (a): FTIR spectra of for pure and Al doped samples

Table 2: FTIR absorption peaks, residual stress, Poisson's ratio , and elastic constants for the samples

$x= 0.00$ $\nu\text{ (cm)}^{-1}$	$x= 0.025$ $\nu\text{ (cm)}^{-1}$	$x= 0.05$ $\nu\text{ (cm)}^{-1}$	$x= 0.10$ $\nu\text{ (cm)}^{-1}$	$x= 0.20$ $\nu\text{ (cm)}^{-1}$
3423.36	3442.14	3417.14	34 7.34	3442.18
2330	-----	-----	-----	2340.21
1590	-----	1610	-----	1623
1426.07	-----	-----	1428.8	1436.68
673.56	672.18	671.84	668.13	-----
-----	-----	-----	556.32	-----
-----	-----	-----	502.43	-----
452.31	452.11	451.97	-----	467.76
		γ		
0.304	0.305	0.307	0.311	0.301
		$\sigma(\text{Gpa})$		
-0.063	0.250	0.474	0.832	1.145
		$S_{11}(\text{D/cm}^2)$		
2.23E+11	2.23E+11	2.23E+11	2.76E+11	2.39E+11
		$S_{12}(\text{D/cm}^2)$		
9.73E+10	9.78E+10	9.86E+10	1.25E+11	1.03E+11

Debye temperature θ_D at about 500 cm^{-1} is given by [85-87];

$$\theta_D (K) = \frac{hc \bar{\Delta\nu}}{K_B} = 1.439 \bar{\Delta\nu} \quad (1)$$

Figure 2 (b) shows that θ_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.

Figure 2 (b): Debye temperature (θ_D) versus Al content for the samples

The strength of the interaction between Zn cat-ions and (O^{-2}) ions of ZnO, the force constant K_t can be calculated using; $K_t = 0.076W\Delta\nu^2$ [88-89]. Also, the stiffness constants S_{11} and S_{12} are easily calculated in terms of Poisson's ratio γ as; $S_{11} = (K_t/c)$, $S_{12} = (S_{11}\gamma / 1 - \gamma)$ and $\gamma = 0.324$ (1-1.043PS) [90]. It is evident from Table 2 that ($\gamma > 0.26$) for all samples which indicates ductile nature [91]. The residual stress σ is calculated by [92-93];

$$\sigma = \frac{[2S_{13}^2 - S_{33}(S_{11} + S_{12})]\epsilon_{Ls}}{2S_{13}} = -232.8\epsilon_{Ls} \quad (2)$$

Where S_{13} , S_{33} , S_{11} , S_{12} are respectively equal 104.2, 213.8, 208.8 and 119.7 GPa for ZnO [94-95]. As listed in **Table 2**, σ 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 [96-97]. Young Y , bulk modulus β and rigidity modulus G are determined by [98-103];

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

As shown in Figure 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 [104-105].

Figure 3: Elastic modulus (Y,B,G) versus Al content for the samples

3.3-Optical Measurements

Reflectance (R/A) versus wave lengths, as shown in Figure 4(a-b), 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 [106]. Interestingly, the change in (R/A) for Al-doped samples is related to the oxygen vacancies, which usually make ZnO suitable for gas sensors [107-108]. As given in Table 3, the first $A_{\max}$ or $R_{\min}$ occurs at $(\lambda (1)_{\max} = 314 \text{ nm})$ for all samples, while the second $A_{\max}$ or $R_{\min}$ occurs at $\lambda (2)_{\max} = 352, 364, 350, 344$ and 346 nm , respectively. Therefore, first exciton energies $E_{\text{ex}}(1)$ is 3.953 eV for all samples, while the second $E_{\text{ex}}(2)$ 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.

Figure 4 (a): Reflectance (R) versus wave length for pure and Al doped samples

Figure 4 (b): Absorption (A) versus wave length for pure and Al doped samples

Figure 5: $(\alpha h\nu)^2$ versus photon energy for pure and Al doped samples

The absorption coefficient α is calculated by, $\alpha = 2.303 \ln (A/t)$, and the optical band gap E_g is obtained using Taucs equation [109-110];

$$(\alpha h\nu)^{\frac{1}{m}} = A(h\nu - E_g) \quad (4)$$

where m represents 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 that indicates the disorder state in the materials as well as the probability of the electronic transition mechanism. α is the absorption coefficient which is given by ; $\alpha = 2.303 \frac{A}{d}$ where A is the absorbance and d is the diameter of glass cuvette. The plot of $(\alpha h\nu)^2$ against the photon energy is used to get the value of the optical direct band gap (see Figure 5).

As shown in the figure, two absorption edges are observed for all samples, indicating that there are two direct electronic transitions due to 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 at 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, 0.61 and 0.61 eV for all samples, in which ΔE is constant. Anyhow, two E_g are also recorded for n-type semiconductors [113-116]. In that case, the doping of ZnO with Al may produce high carrier concentration and, as a result, more conduction band states are blocked, which causes an increase, which, in turn, causes an increment in E_g as obtained. However, the increment of E_g above $x = 0.05$ may be due to the formation of some defects in the host materials, thereby creating insulating states in the forbidden gap inside the band structure.

Table 3: λ , E_{ex} , E_g , ϵ_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)	ϵ_L	(N/m^*) $\times 10^{24}$ ($g \cdot cm^{-3}$)	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 $Zn_{1-x}Al_xO$ is calculated from the following relation, which represents the dependence of the reflective index n on the wavelength λ [117-120]:

$$n^2 = \epsilon_L - \left(\frac{e^2}{4\pi^2 c^2 \epsilon_0} \right) \left(\frac{N}{m^*} \right) \lambda^2 \quad (5)$$

where c , e , m^* , ϵ_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 Figure 6 give the values of ϵ_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 ϵ_L is decreased to $x = 0.10$, followed by a sharp increase at $x = 0.20$. The inter-atomic distance (R) given by $(0.86/N^{1/3})$ [121-122] and listed in Table 3 is decreases as x increases to 0.20. Based on the above results, the higher valence state of the Al^{3+} compared to Zn^{2+} led to slightly deeper donors and, consequently, more carriers are generated. Further, and in parallel, the ability of electron migration may be enhanced [123-124]. Furthermore, Al is nearly non-magnetic, and the addition of Al beside Zn will decrease the localization of the carriers, and therefore, we strongly recommend the Al doped samples for optoelectronic applications.

Figure 6: Real part of optical dielectric constant ($n^2 - k^2$) versus wave length for pure and Al doped samples

3.4- Dielectric Measurements

The real ϵ' part of the sample dielectric constant is determined by using the relation [125-127];

$$\epsilon' = \frac{Cd}{\epsilon_0 A} \quad (6)$$

Where C is the capacitance of the sample and C_0 is the empty cell capacitance ($C_0 = \epsilon_0 A/d$), d is the sample thickness, A is the surface area of the electrodes, $\epsilon_0 = 8.85 \times 10^{-12}$ (F/m) is the permittivity of free space. Figure 7 shows the plot of ϵ' against f for all samples. It is clear that ϵ' is gradually decreased as f increases up to 10^4 Hz for all samples, and above that it decreases, and then saturated at 10^7 Hz. In addition, it is slightly increased as x increases up to 0.025, but there is a significant increase with further increase of x above 0.025. For example, ϵ' of $x = 0.20$ is at least about 10^3 times 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 ZnO large interfacial polarization, and in turn, high ϵ' . With a rise in frequency above 10 MHz, a decrease of ϵ' 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 [128-130]. On the other hand, the increase of ϵ' 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 indicted above [131-133]. Due to this, the strength of the Al-O ionic bond is improved, which results in increasing the dielectric polarization. Moreover, the decrease of 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 ϵ' values for the doped samples as reported [134-135].

Figure 7: Real part of dielectric constant (ϵ') versus frequency for pure and Al doped samples

The total conductivity can be obtained by [136];

$$\sigma_t(\omega) = \sigma_{dc} + \sigma_{ac} = \sigma_{dc} + B\omega^s \quad (7)$$

Where σ_{dc} is the dc conductivity calculated at 0f, B is constant and s is the exponent of frequency. Figure 8 shows the real part (σ') of σ_{ac} as a function of f. It is evident from the figure that σ' gradually increases by increasing f for all samples up to 10 MHz, obeying the universal power law [137]. In addition, σ' for Al-doped samples is higher than that of ZnO due to the increased charge carriers through the replacement of Zn^{2+} by Al^{3+} [55-57]. At low f, the field drift carries over a large distance, and therefore σ' is almost unchanged. As f is raised, the displacement of the carriers is reduced, and at a critical frequency ω_p , the σ' follows the law ($\sigma_{ac} \sim \omega^s$) with ($0 \leq s \leq 1$) characterizing hopping conduction [138-139]. These outcomes indicate that the addition of Al supports the σ_{ac} of ZnO as obtained.

However, the linear fit between $\ln\sigma'$ and $\ln\omega$ shown in Figure 9 (a,b) gives two different linear regions. The obtained values of s and B at low and high frequencies named 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 and 0.898 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 and 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+} .

Figure 8: Real part of conductivity (σ') versus frequency for pure and Al doped samples

Figure 9 (a): $\ln \sigma'$ versus $\ln \omega$ at low frequency for pure and Al doped samples

Figure 9(b): $\ln \sigma'$ versus $\ln \omega$ at high frequency for pure and Al doped samples

Table 4: S, B and W_m for pure and doped samples

Al content x	σ_{dc} ($\Omega.cm$) ⁻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
0.10	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

According to correlated barrier hopping (CBH) model, the binding energy W_m , is given by [140];

$$s = 1 - \frac{6K_B T}{W_m + K_B T \ln \omega \tau_0} = 1 - \frac{6K_B T}{W_m} \quad (8)$$

Where τ_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 [141];

$$R_{min}(\omega) = \frac{e^2}{\pi \epsilon_0 \epsilon' (W_m + K_B T \ln \omega \tau_0)} \sim \frac{e^2}{\pi \epsilon_0 \epsilon' \bar{W}_m} \quad (9)$$

Where $\bar{W}_m = [(W_{mL} + W_{mh})/2]$. On the other hand, the density of localized states at which carriers exist N can be expressed by [142];

$$N(\omega) = \frac{24 \sigma'}{\pi^3 \epsilon_0 \epsilon' \omega R_{min}^6} \quad (10)$$

As shown in Figure 10 (a), 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 Figure 10 (b). 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 ($x = 0.20$).

Figure 10 (a): Hopping distance (R_{min}) versus frequency for pure and Al doped samples

Figure 10 (b): Density of localized states (N) versus frequency for pure and Al doped samples

In addition, the density of states at Fermi level $N(E_f)$ can be calculated by [143];

$$N_{E_f}(\omega) = \sqrt{\frac{3\sigma \alpha^5}{4e^2 \omega K_B T (\ln \frac{10^{13}}{\omega})^2}} \quad (11)$$

Where α is the localized wave function and equals 10^{10} m^{-1} and $f_0=10^{13} \text{ Hz}$ is the photon frequency corresponding to the applied ac field. However, the behavior of $N(E_f)$ against x shown in Figure 11(a) 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 with f for $x = 0.00$ and 0.025 samples, goes to 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 [144]. 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 $8.05 \times 10^{19} \text{ (erg.cm}^3)^{-1}$ for $x = 0.00$ to 2.01×10^{20} , 8.97×10^{20} , 1.75×10^{21} and $1.78 \times 10^{21} \text{ (erg.cm}^3)^{-1}$ 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, (σ_{ac}/ϵ) is mainly good indicator for a sample to be more suitable for solar cell applications [145-146]. As shown in Figure 11(b), 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 11(a): Density of states at Fermi level $N(E_f)$ versus frequency for the samples

Figure 11(b): F-factor versus frequency for the samples

Figure 12 (a,b) shows the variations of $Z^{\parallel}$ and $Z^{\perp}$ against f for all samples. It is evident that $Z^{\parallel}$ and $Z^{\perp}$ for $x \leq 0.025$ samples are decreased linearly as a straight line with a rise in f . In addition, $Z^{\parallel}$ was also decreased as x increases, whereas $Z^{\perp}$ was increased. With further increase of x above 0.025, $Z^{\parallel}$ is slightly increased with f and goes to higher values at about 10 KHz, and then decreases at 10 MHz. It is also decreases as x increases below 100 KHz, but above that it is nearly constant for all samples. In contrast, a little bit decrease is obtained below 10 KHz for $Z^{\perp}$ 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 of Z at low f confirm the highly resistive grain boundaries, whereas the decrease is due to the rise in the hopping of charge carriers.[147-150]. Anyhow, the Cole- Cole plot is shown in Figure 13 (a, b) and the impedance of grains and grain boundaries is determined at the half and the end of semicircle-shape plot, respectively. It is evident that the impedance spectra of $x= 0.00$ sample is straight line like- shape with $Z^{\parallel} = 2.05 \times 10^{10} \Omega$ and $Z^{\perp} = 1.39 \times 10^8 \Omega$. 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 large radius is obtained. Similar behavior is also obtained for $x \geq 0.10$, but the area of 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.

Table 5: Z' and Z'' for grain and grain boundaries for pure and Al doped samples

x	$Z'(g)$ (Ω)	$Z'(g)$ (Ω)	$Z'(gb)$ (Ω)	$Z'(gb)$ (Ω)
0.00	-----	-----	-----	-----
0.025	3.19×10^9	6.19×10^9	6.38×10^9	4.92×10^8
0.05	4.59×10^5	2.65×10^5	10.02×10^5	4.15×10^4
0.10	3.01×10^5	1.65×10^5	6.01×10^5	1.56×10^4
0.20	1.88×10^5	1.05×10^5	4.41×10^5	1.13×10^4

It is seen that the applied frequency for a pure sample is not sufficient to cover the grain boundary, and therefore the $Z'(g)$ and $Z'(gb)$ cannot be evaluated. By increasing x to 0.025, the $Z'(gb)$ was reduced, and the frequency was able to make it as one quarter of a circle because the $Z'(gb)$ was reduced, and it can be obtained approximately. With an increase of x above 0.025, the applied frequency is more than sufficient for covering the grain boundary, and therefore the $Z'(g)$ and $Z'(gb)$ could be evaluated. However, a single arc of an identical $\tau = (1/\omega) = RC$ has been obtained in pure and doped ZnO [151-156]. Because of this behavior, conduction in grains and their boundaries could not be separated [157]. Other reports believed that a single arc was caused by the combined contributions of the grain's core resistive and grain boundary [158]. 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 [159-161].

Figure 12 (a): Real part of impedance (Z') versus frequency for pure and Al doped samples

Figure 12 (b): Imaginary part of impedance (Z'') versus frequency for pure and Al doped samples

Figure 13 (a): The Cole- Cole plot for $x = 0.00$ and 0.025 samples

Figure 13 (b): The Cole- Cole plot for $x = 0.05$, 0.10 and 0.20 samples

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 show unusual behavior, the lattice constants, crystallite diameter, porosity, average crystalline size Debye temperature and elastic modulus are increased for the doped samples. The residual stress is compressive for the pure sample, but it is tensile for the doped samples. Although two different values of E_g are apparent for each sample, the $\Delta E = (E_{gh} - E_{gl})$ is almost constant. Further, the ϵ_L decreased while (N/m^*) was increased as x increases to 0.20. On the other hand, the values of ϵ' and σ' are generally increased as x increases to 0.20. The conduction is electronic for $x \leq 0.025$ samples, but it is changed to hole for $x \geq 0.20$. Although W_m decreased by increasing x , there was no exact trend for the behaviors of R_{min} 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. It is approved that the addition of Al to ZnO improves the mechanical, optical, and dielectric properties of ZnO, and strongly recommended for the design of optoelectronic and solar cell instruments..

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Funding and/or Conflicts of interests/Competing interests

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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