Structural, dielectric, impedance, complex modulus, and optical study of Ni-doped Zn\(_{(1-x)}\)Ni\(_x\)O nanostructures at high temperatures

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Abstract
This experiment addressed the effect of Nickel-doped on the dielectric, ac conductivity, and optical properties of pure and doped Zn\(_{(1-x)}\)Ni\(_x\)O (x = 0, 3 and 6%) nanostructures. The un-doped and Ni-doped ZnO nanostructures were synthesized using co-precipitation. In this paper, the frequency-dependent dielectric and the electrical conductivity of un-doped and Ni-doped Zn\(_{(1-x)}\)Ni\(_x\)O nanostructures were examined at various temperatures ranging from 320 K to 460 K using an LCR meter. For the morphological and optical investigation, the prepared samples were analyzed using field emission-scanning electron microscopy (FE-SEM), and UV visible Spectroscopy was used at room temperature. The dielectric constant (\(\varepsilon'\)), dielectric loss (\(\varepsilon''\)), tangent loss (tan \(\delta\)), the real as well as the imaginary part of the impedance against the frequency ranging from 100 Hz to \(2 \times 10^6\) Hz that declines with increases in frequency at different temperatures ranging from 320–460 K. However, the electrical conductivity (\(\sigma_{ac}\)) increased with the increase in frequency was examined. The ac conductivity (\(\sigma_{ac}\)) follows Jonscher’s power law that the electrical conductivity is enhanced with increasing doping concentration. The optical transmission area also improved due to an increase in Ni-doping concentration in ZnO. The optical bandgap of pure and Ni-doped ZnO nanostructures is in the range lies 3.30–3.12 eV found that to decrease with the increase in Ni doping concentrations.

1. Introduction

Newly, semiconductor nanomaterials have a complete fascination because of their size and shape, relying on electrical, dielectric, and optical properties. ZnO has unique features and potential practical applications in Nanodevices [1]. Frequently, ZnO semiconductor material has main properties like electrical and optical that are expressively changing by their size and shape [2]. Zinc oxide nanomaterials exhibit different shapes and sizes, such as Nanoplates, Nanospheres, nanoflowers, and nanowires [3–5]. ZnO nanostructure is a critical and versatile semiconductor with a direct wide bandgap of 3.37 eV and excitation binding energy (60 meV) at room temperature [6]. ZnO has several benefits associated with a large bandgap containing lower electronic noise, higher breakdown voltages, hydrogen generation, and proficiency in sustaining high electric fields [7]. Due to these properties ZnO, nanostructures are best for practical applications, sensors [8], transducers [9], spintronic [10], optoelectronics [11]. Zn\(^{+2}\) and O\(^{-2}\) atoms form a crystal assembly where Zn\(^{+2}\) cations are tetrahedrally corresponding with four O\(^{-2}\) ions [12]. ZnO semiconductor has a high switching high temperature with transition metals-doped Zinc oxide has developed as a gorgeous candidate from both a theoretical [13] with the investigational point of vision [14]. They were doped with a suitable material in ZnO and fabrication method that altered the electrical, dielectric, and optical properties. However, Nickel-doped ZnO is one of the supreme extensive systems and is highly concerned and very important in spintronic applications [15]. The practical applications in a spintronic variety a request for the capability of a couple of electron charges [16] due to these charge carrier-mediated interchange between transition elements and a ZnO extensive bandgap semiconductor, which show the ferromagnetic assembling at room temperature [17]. Many other 3d transition metals (Co\(^{+2}\), Mn\(^{+2}\), Cr\(^{+2}\), and Cu\(^{+2}\)) substituted in ZnO have been widely studied [18–20].
Nowadays, investigators are giving attention, what way to modify the necessary parameters of doped ZnO semiconductors. Usually, the band structure and the size that shows the key role with these things can be changed with different metals doping that makes secondary energy states within ZnO\[21\]. Many publications described the effect of transitions metals doped on zinc oxides semiconductors\[22–25\]. But Ni is one of the most efficient transition metals dopants, improving dielectric, electrical conductivity, and optical properties\[26\]. The incapacitated Ni$^{2+}$ may trap the electrons with result avoiding electron-hole recombination in ZnO\[27\]. The ionic radius of Ni$^{2+}$ is very near the Zn$^{2+}$, and they both have the same valence, which modifies these properties\[28\].

Furthermore, for high Ni-doping concentrations, has to form nanoclusters of metallic nickel or NiO\[29\] due to the solubility of nickel within the ZnO\[30\]. Different methods were utilized to fabricate ZnO nanostructures, such as co-precipitation\[22\], spin-coating\[31\], spray pyrolysis technique\[32\], sol-gel method\[33\], and solid-state reaction\[34\]. However, the co-precipitation method is a low-cost and simple method to synthesize pure and doped ZnO nanostructures.

From the above works, we are inspired to study the nickel-doped ZnO nanostructures with different doping concentrations at $x = 0, 0.03, \text{and } 0.06$ to check dielectric, electric conductivity, and optical properties. Zinc chloride ($\text{ZnCl}_2$) and nickel (II) acetate 4-hydrate Ni(OCOCH$_3$)$_2.4\text{H}_2\text{O}$ were chosen as a host and doping precursor.

2. Experimental details

2.1. Nanopowders synthesis

The co-precipitation method was used to prepare Ni-doped ZnO nanostructures with ($x = 0, 3, \text{and } 6\%)$. The initial materials are Zinc chloride ($\text{ZnCl}_2$) and nickel (II) acetate 4-hydrate Ni(OCOCH$_3$)$_2.4\text{H}_2\text{O}$ were taken as the host and doping material. These materials are mixed according to a stoichiometric ratio. A suitable amount of Zinc chloride ($\text{ZnCl}_2$) was dissolved in distilled water. 6.814 g of ZnCl$_2$ dissolved in a beaker, which confined 250 ml of distilled water at room temperature, magnetic stirring for 2 h. Then a proper quantity of sodium hydroxide ($\text{NaOH}$) solution was mixed slowly and dropwise to obtain the pH value of 12. The obtained prepared solution was stirred for 50 min. Then the final solution was washed with distilled almost 08–10 times using filter paper to eliminate the crusts. Finally, it shifted cautiously for drying in a microwave oven at a temperature of 80 °C for 24 h for removing water fragments. After that, the final product was annealed at 550 °C for two and a half hours. A similar process was used for the fabrication of Ni-doped ZnO nanostructures ($x = 3, \text{and } 6\%)$. A cylindrical pellets of 10 mm diameter, 2.1 mm thickness, and 0.7 g mass were arranged by uniaxial pressure in a hydraulic press at a 3-ton load for five min. These were pellets annealed at the same temperature for strengthened for two and a half hours at the same temperature.

2.2. Characterization methods

X-ray diffraction (XRD) was analyzed at room temperature with a Bruker D8 with CuK$\alpha$ radiation ($\lambda = 1.5406$ Å) for $2\theta$ changing from 20° to 80°. The ZnO nanostructures of pure and doped ZnO ($x = 0, 3, \text{and } 6\%)$ samples were examined by the Field emission-scanning electron microscopy (FE-SEM) to calculate the average crystalline size. The dielectric and electrical conductivity properties were analyzed by determining the complex impedance by the (Agilent E980A) LCR meter at different temperatures ranging from 320 K to 460 K with

![Figure 1. X-ray diffraction patterns of pure and Nickel-doped Zn$_{1-x}$Ni$_x$O nanostructures.](image-url)
frequency ranging from 100 Hz to 2 MHz. The temperature-controlled with the help of the Quiet Chuck controller MDC (490 B). Finally, the Optical transmittance measurements were executed in the wavelength range 300 to 600 nm with the help of a UV–vis Spectrometer.

3. Results and discussions

3.1. X-ray diffraction analysis

XRD patterns of pure and doped ZnO nanostructures shown in figure 1 were indexed using the JCPDS data card No. 36–1451. All samples are in hexagonal wurtzite structure, and the preferred orientation along the (101) plane with additional impurity peak at 6% doping due to NiO (2 0 0) corresponds to JCPDS card no. 897130 shows that nickel ions have been incorporated into the Zn$^{2+}$ lattice site, prompting cluster formation with a metallic Ni or NiO. The lattice constants of pure and Ni-doped ZnO nanoparticles were calculated using the following formula [35].

$$\frac{1}{d^2} = \frac{4}{3} \left( \frac{h^2 + hk + k^2}{a^2} \right) + \frac{l^2}{c^2}$$  (1)

Where a and c are lattice constants and h, k and l are miller indices.

The volume of the unit cell can be estimated using the equation [36].

$$V = \frac{\sqrt{3}}{2} a^2 c$$  (2)

Average crystallite sizes were calculated using Scherer’s formula at the most intense peaks [36].

$$D_{hkl} = \frac{0.9 \lambda}{\beta \cos \theta}$$  (3)

The calculation from figure 1 gives the lattice constants $a = 3.245 \text{ Å}$ and $c = 5.20 \text{ Å}$ with a unit cell volume $V = 47.419 \text{ Å}^3$ for undoped ZnO nanostructures. The unit cell volume of Ni-doped ZnO decrease to 47.027 Å$^3$ with the increase of Ni-doped with x = 3%, due to the decline in lattice parameters $a = 3.235 \text{ Å}$ and $c = 5.189 \text{ Å}$ [37, 38].
Further, Ni was doped into the ZnO lattice, decrease in the unit cell volume, \( V = 46.960 \, \text{Å}^3 \), was noticed having lattice constants \( a = 3.234 \, \text{Å} \) and \( c = 5.185 \, \text{Å} \) with compared to pure ZnO as shown in figures 2(a)–(b). The reduction in lattice constants with doping is due to lesser ionic radii of Ni\(^{2+}\) (0.69 Å) is matched to Zn\(^{2+}\) (0.74 Å). The peak positions moved to higher angles for the Ni-doped ZnO nanoparticles, which are consistent with this Ni\(^{2+}\) ions substitute the Zn\(^{2+}\) ions in the ZnO lattice [39]. The crystallite size of undoped, Ni-doped ZnO nanoparticles was calculated from the XRD at most intense (101) peaks by the Scherer formula as shown in figure 1(c). The rise in crystallite size with increasing Ni-doping is recognized as the grain growth inhibition of Ni-doped ZnO. The Average grain growth comprises an increase in the crystalline size of the grains with a parallel reduction in the number of grains. This procedure certainly involves vanishing some grains, commonly the smaller ones, which has led to grain growth being described as an inhibitor process. Secondary recrystallization consists of the evolution of very few grains in a relatively unchanged fine-grained matrix, leading to the development of sizeable crystalline size [40]. The increasing concentrations also led to segregation on the grain boundary and increased grain size [41]. Figure 2(c) shows that the mean crystalline size increased with increasing Ni concentration; this might be due to increased Ni concentration at the ZnO matrix, which will escalate the particles’ nucleation and increase the grain growth of Ni-doped ZnO NPs. This result is in good agreement with those of previous studies [42, 43].

### 3.2. Morphology analysis

The surface morphology of pure and Ni-doped ZnO nanostructures is shown in figures 3(a)–(c). The surface morphology of ZnO nanostructures was studied with the field emission scanning-electron microscopy (FE-SEM) technique. Figure 3(a) shows pure ZnO nanostructures that are distributed randomly have different crystallite sizes. Figures 3(a)–(c) indicates that the particles are slightly changing from spherical to rod types, increasing Ni concentrations ranging from \( x = 0\% \), 3\%, and 6\%. Figures (b) and (c) with 3 and 6\% Ni-doped ZnO nanostructures observed two types of spherical nanostructures and some other rod types. But ZnO nanostructures at 3\% and 6\% doping, the diameter lies between 40 to 80 nm, and the length varies between
~130–360 nm. Crystallite size mentions the grain boundary of any particle. Thus in any particle, so many grain boundaries are present. If the particle contains only a one–grain boundary, then the size is equal to the crystallite size. But it is a very rare case, and always crystallite size is less the size of the crystallite size obtained from SEM image; secondly, dimension changes from spherical to rad types. Therefore, there is a difference in the size calculated from these two techniques [44].

3.3. Dielectric constant
In oxide materials, the dielectric constant is calculated by the equation.

\[ \varepsilon = \varepsilon' - j\varepsilon'' \]  \hspace{1cm} (4)

with \( \varepsilon' \) is the real part called energy storage and \( \varepsilon'' \) is the imaginary part associated with the energy dissipation of materials.

Figures 4(a)–(c) shows the distinction of the dielectric constant of ZnO with \( x = 0, 3 \) and 6\% nanostructures versus frequency ranging from 100 Hz to 2 MHz with temperatures 320 K to 460 K. The dielectric constant can be measured at different temperatures using the following equation.

\[ \varepsilon' = \frac{Ct}{A\varepsilon_0} \]  \hspace{1cm} (5)

where \( C \) is the capacitance in Farad, \( A \) is the cross-sectional area of pellets, \( t \) is the thickness and \( \varepsilon_0 \) is constant which is called the permittivity of free space. The decrease of the real part of the dielectric constants is rapidly at the initial frequency and slowly at an upper frequency and becomes almost constant at higher frequency regions. The polarization happens in oxide between Ni\(^{2+}\) and Zn\(^{2+}\) ions by a conduction mechanism method. At the low frequency at all temperatures range, the dielectric constant values are very high that can be recognized based on the space charge polarization owed to the uniformities of dielectric material. For all the sample’s the dielectric values are saturated at a higher frequency can be expressed in the Maxwell Wagner model [45]. According to polarization, well-conducting grains are separated by weakly conducting grain boundaries in a given material. After a particular frequency, the polarization tendency decrease and approach constant values at a higher.
frequency; an electron exchange between Ni$^{2+}$ and Zn$^{2+}$ cannot follow the alternating field. The reductions in dielectric constant through two phases, frequency, and temperature dependence at different compositions, the increasing frequency ranging from 100 Hz to 2 MHz, the dielectric constant of all concentrations indicate a common tendency having the highest values in lower-frequency areas. The dielectric constant values increase the increase of Nickel doping concentrations of the Zn site up to 6% given in table1 at 2 MHz at temperature 460 K. The dielectric constant is higher with Ni-doping due to the presence of N$^{2+}$ ions, including other defects such as grain boundaries, dislocations, and oxygen vacancy defects. The dielectric constant values increase with the increase of N-doping concentrations of the Zn site, which is valid for charge storage devices applications. The increase in the dielectric constant with Ni-doping is due to extra and more defects (positive, negative, or neutral) or oxygen vacancies that are arbitrarily created (at the surface), and lastly, a disorder in the system. As a consequence of recombination, the number of dipoles reduces, which clues to a decrease in the orientational polarization [47]. The reduction in orientational polarization increases a dielectric consonant. Second, frequent oxygen vacancies and zinc interstitials in the prepared samples are considered possible sources for the orientation polarization. The O$^{2-}$ vacancies and the Zn$^{2+}$ ions in the neighbourhood exchange their positions by a single jump and effort to align with the applied field direction [48]. Another reason for higher dielectric constant because of Ni-doping at lower frequency is the numerousness of the effect, such as grain boundary defects in the presence of oxygen vacancies [49, 50].

3.4. Dielectric loss ($\varepsilon''$)

The dielectric loss can be calculated using the following formula.

$$\varepsilon'' = \varepsilon' \tan \delta$$

(6)

Where $\varepsilon'$ is dielectric constant and $\tan \delta$ is called tangent loss.

![Figure 5](https://example.com/fig5.png)

**Figure 5.** Dielectric loss versus frequency at various temperatures of ZnO nanostructures, with (a) 0%, (b) 3% and (c) 6%.

| Composition(x) | $x = 0\%$ | $x = 3\%$ | $x = 6\%$ |
|----------------|-----------|-----------|-----------|
| $\varepsilon'$ | 7.391     | 11.594    | 14.202    |
| $\tan \delta$ | 0.165     | 0.478     | 0.590     |
| $\sigma_{ac}$ ($s \,cm^{-1}$) | $1.356 \times 10^{-4}$ | $6.162 \times 10^{-4}$ | $9.310 \times 10^{-4}$ |

*Table 1.* Dielectric constant $\varepsilon'$, Tangent loss ($\tan \delta$) and Conductivity ($\sigma_{ac}$) at temperature 460 K with frequency 2(MHz).
The \( \tan \delta = \frac{\text{Imaginary part of the dielectric constant}}{\text{real part of dielectric constant}} \). So, the imaginary part of the dielectric constant \( \epsilon'' \) the real part of the dielectric constant \( \epsilon' \). The frequency-dependent behaviour of the dielectric loss is shown in figures 5(a)–(c), ranging from 100 Hz to 2 MHz with temperatures ranging from 320 to 460 K. The configurations achieved by the dielectric loss \( \epsilon'' \) show that closeness behaviour with the real part of the dielectric constant has a high value in low-frequency areas and becomes constant after a specific frequency. With Koop's model, the low-frequency region is associated with highly resistive grain boundaries parameters, which signifies the high energy that is necessary for the electrons to hop among the nickel and Zn ions. The value of dielectric loss for undoped ZnO at 2 MHz was found at 7.391 temperature 460 K. But for Ni-doped ZnO \( (x = 3 \text{ and } 6\%) \), the values that are going to increase 11.597 and 14.202 versus frequency at temperature 460 K are listed in table 1. Moreover, the applied electric field is less than the hopping frequency of electrons between nickel and zinc in the lower-frequency areas. The hopping charge carrier follows the applied electric field via more energy utilized for the hopping charge carrier that gets stuck by polarization due to impurities, flaws, and porosity. These outcomes show that the dielectric loss decreased with the increasing frequency at different temperatures ranging from 320 to 460 K in the low-frequency region. Inescapable electrode polarization and conduction losses are more than the undoped ZnO \[51\].

### 3.5. Tangent loss (\( \tan \delta \))

The tangent loss can be determined using the following relationship.

\[
\tan \delta = \frac{\epsilon''}{\epsilon'}
\]

Figures 6(a)–(c) shows the variation of tangent loss (\( \tan \delta \)) versus frequency ranging from 100 Hz to 2 MHz and temperatures ranging from 320 to 460 K. The Tangent loss is defined as the real part of the dielectric constant and dielectric loss ratio. The tangent loss of the pure ZnO shown in figure 6(a) plot displays a relaxation peak that occurs whenever the frequency of the localized charged carriers becomes nearly equivalent to the applied electric field. Tangent loss happens when the polarization, due to the applied electric field, with defects such as impurities and grain boundaries. The tangent loss for ZnO with \( (x = 0, 3, \text{and } 6\%) \) nanostructures at lower frequency regions are higher, then decreases with the increase of frequency. These losses values are high due to the resistivity of the grain boundaries and other defects. The values of undoped ZnO at 2 MHz were found at 0.165 at temperature 460 K. But for Ni-doped ZnO \( (x = 3 \text{ and } 6\%) \), values of tangent loss rise with the increase of Ni-doping concentrations are listed in table 1. In the higher frequency areas, the obstruction resistance of grains falls, so fewer electrons are hopping with small energy feasting that indicates the decrease of tangent loss at high frequency. For Pure ZnO, a broad tangent loss peak shows a relaxation time division rather than a single relaxation time. Such types of behaviour can be observed only in an oxide material when \( \omega r = 1 \) whereas \( r \) is relaxation time and \( \omega \) is angular frequency \[52\]. At temperature 460 K, the Tangent loss values
increase with the increase of doping concentrations due to the substitution of Ni$^{2+}$. The peaks shifted towards the higher frequency regions for pure ZnO due to purities and lower defects for un-doped ZnO nanostructure. So we concluded that this Ni-doped ZnO is useful for photovoltaics and high-frequency device applications [53].

3.6. Complex real electric modulus analysis

The electric modulus is utilized in the analysis of dielectric and electrical conductivity behaviour. The complex electric modulus emphasizes the grain boundary conduction mechanism and is useful for conductivity relaxation times. Using the following equation, the complex electric modulus is imitative from the dielectric constant and dielectric loss [54].

$$M' = \frac{\varepsilon'}{\varepsilon''^2 + (\varepsilon'')^2}$$

Figures 7(a)–(c) shows the real complex electric modulus ($M'$) versus frequency ranging from 100 Hz to 2 MHz at various temperatures ranging from 320 to 460 K for the pure and Ni-doped ZnO nanostructures with $x$ (0, 3 and 6%). It observed that the complex electric modulus was imminent to zero at low frequencies regions at all samples. Such clarifications with observations correlated to the lack of restoring force dominate the movement of charge carriers due to the applied electric field that suggests the long-range distant mobility of charge carriers because of polaron hoping [55]. In lower frequency regions, the complex electric modulus approaches zero due to the influence of the electrode effect in total impedance. But the magnitude of complex electric modulus instantly increases with frequency at different temperatures for un-doped and Ni-doped ZnO nanostructures. Figures 7(a)–(c) shows that electric modulus increased frequency and gathered at constant values at higher frequency areas. It is noticed that the positions of the complex electric modulus peaks swing toward the higher frequencies with the increased temperatures at all compositions. The electric modulus low values in low-frequency areas that value increases with increasing frequency and temperatures but, reaching constant values after a specific frequency, shows the conduction mechanism due to the short-range movement of charge carriers in all samples. With that, the relaxation frequency that is the maximum frequency corresponding to the complex

![Figure 7. Complex electric Modulus versus frequency with different temperatures of ZnO nanostructures (a) 0%, (b) 3% and (c) 6%.]
electric modulus provided the shift from long-range to short-range movement with frequency and temperatures.

3.7. Nyquist plot analysis
Figures 8(a)–(c) shows the Nyquist plots of ZnO with (x = 0, 3, and 6%) versus frequency and temperatures ranging from 100 Hz to 2 MHz and 320 to 460 K. This procedure is a suitable examination technique because
grains’ impedance can be separated from the base sources known as electrode effects and grain boundaries. Figures 8(a)–(c) shows impedance’s imaginary part versus impedance’s real part at different temperatures ranging from 320 K to 460 K. These figures are categorized as the appearance of semicircular arcs. Figures 8(a)–(c) shows the single semicircle at all temperatures at all doping compositions samples that means that procedures follow a single relaxation appliance. These semicircles recorded depressed and showed a relaxation time of non-Debye in all doping concentrations. The diameter of all doping compositions of ZnO with ($x = 0, 3,$ and $6\%$) decreases with increasing temperatures that mean that the conductivity is rising. The decaying of the $Z'$ part of the impedance correlated to the conduction mechanism of the semiconductor material. So the real part of impedance $Z'$ steadily decays at a high-temperature area. However, all kinds of polarization happen in low-frequency regions, but in the high-frequency range, only electronic polarization occurs [56]. For all doping concentrations, the semicircle diameter declines with the increase of temperature, illuminating the reduction in the semiconductor materials’ bulk resistance. The Nyquist plots of Zn$_{(1-x)}$Ni$_x$O with the frequency ranging from 100 Hz to 2 MHz for $x = 0.03$, $x = 0.06$ exhibit depressed semicircles equivalent to grain boundary and grain influence. With that, the grain boundary influence in these materials looks to be the mobility of the small Ni ions in the host Zn site [57]. At a high-frequency area with temperatures indicating the increased ac conductivity of all Ni-doping concentrations, also in high-frequency-region, the plateau-like environment of real ($Z'$) suggests which may occurrence of space charge polarization due to the applied electric field. The grain boundaries resistance decrease with the increasing temperatures indicating a semiconducting behaviour for undoped and Ni-doped ZnO. It discovered that the outcome of grain boundaries supports lower the blockade to the mobility of charger carriers, causing increased electrical transport with an increase in temperatures ranging from 320 K to 460 at all composition samples [58].

The complex impedances data were fitted using impedance analysis Z VIEW computer software with an equivalent circuit containing two series resistance with a parallel CPE element for the grain and grain boundary response. From fitted data of grain resistance, $R_g$ is greatly lesser than grain boundary resistance $R_{gb}$. The grain and grain boundary resistance obtained from the fitted data plotted in the Arrhenius format are shown in figures 9(a)–(c). These plots follow the Arrhenius law presentation shown in figures 9(a)–(c) with activation energies 0.42 eV, 0.18 eV and 0.14 eV (grain) and 0.76 eV, 0.39 eV and 0.16 eV (grain boundary) were calculated from the figure 9. It could be concluded that the activation energies decline with the increase of ac conductivity with Ni-doping in ZnO as compared to un-doped ZnO [54].

Figure 10. The real part of the impedance versus frequency of Zn$_{(1-x)}$Ni$_x$O at different temperatures (a) 0%, (b) 3% and (c) 6%.
3.8. Real and imaginary impedance versus frequency

Figures 10(a)–(c) shows the discrepancy of the real part of impedance $Z'$ versus frequency (100 Hz to 2 MHz) of ZnO with ($x = 0, 3,$ and $6\%$) at different temperatures ranging from 320 to 460 K. The impedance values are greater at the lower temperatures in the low-frequency regions and then began to decrease slowly with the increasing frequency with temperatures, which indicates the improvement of ac conductivity with the increase of frequency and temperatures ranging from 320 to 460 K. It noticed that real impedance $Z'$ values combine and become uniform at the high-frequency side due to a promising release of space charge at the specific frequency. Regions where the release of space charge ensues also depend upon the Ni concentrations [59]. The hopping between Ni$^{1+2}$ and Zn$^{2+}$ is the least in the low-frequency region, indicating strong impedance values. The decreasing particle size with increasing Ni concentrations with decreasing Zn ions and increasing Ni$^{1+2}$ ions are accountable for high impedance for all doping concentrations obtained by the previous result [60]. However, in our case, with the increase of particle size, the real part of the impedance with Ni concentration in ZnO at 460 K for all Ni-doped ZnO NPs decreases, which increases ac conductivity values are given in table 1 in good agreement with the other results [61]. The decreasing value of impedance with the temperature rise is due to a relaxation process in the material. Further, with the increasing temperature, the value of $Z'$ decreases at low frequency, showing negative temperature coefficient of resistance (NTCR) type of behaviour [62].

Figures 11(a)–(c) shows the imaginary part of the impedance versus frequency ranging from 100 Hz to 2 MHz with temperatures ranging from 320 to 460 K respectively of ZnO with ($x = 0, 3,$ and $6\%$). The imaginary part of impedance decays with the increase of frequency and Ni-doping at different temperatures, for $x = 0, 3,$ and $6\%$. This development is due to charge carriers, imperfections, defects, and oxygen vacancies at higher temperatures [63]. Figures 11(a)–(c) indicates the imaginary part of impedance as a function of frequency at different temperatures. A broad peak for 320 K and the increasing temperature gradually decreases because the impedance values reduce with the temperature rise for all samples. These peaks frequency, termed as relaxation frequency, move towards the higher frequency side as the temperature increases, implying that the hopping rate of localized charge carriers increases [64]. With the rise of Ni concentrations and temperatures ranging from...
320 K to 460 K, the magnitude of the imaginary part of the impedance declines, and all the peaks pure and doped samples shift towards the higher frequency side and again decrease with the increase of frequency and become uniform at high-frequency regions [65]. Moreover, relaxation peaks show broad nature suggestions for non-Debye type behavior with relaxation time spread [66].
3.9. a.c conductivity

The values of ac electrical conductivity of the semiconductor materials were calculated using the equation

$$\sigma_{ac} = \left( \frac{Z'}{Z'^2 + Z''^2} \right) \frac{t}{A} \quad (9)$$

Whereas $A$ is the cross-sectional area, $t$ is the thickness of pellets, $Z'$ is real and $Z''$ is an imaginary part of impedance [67].

Figures 12(a)–(c) shows the variation in ac conductivity of ZnO with ($x = 0.00, 0.03, 0.06$) with the frequency ranging from 100 Hz to 2 MHz with temperature ranging start from 320 K to 460 K. The ac conductivity steadily increases for all Ni-doping concentrations with the increase of temperature and frequency. Commonly the total conductivity is calculated by the equation $\sigma_T = \sigma_{dc}(T) + A \omega^3$ [68]. In this equation, where the first part is dc conductivity because of band conduction, the second part is the ac conductivity due to the migration of electric charge. In contrast, $A$ is a material-specific constant that depends on temperature, $\omega$ is the angular frequency, and $s$ ($0 \leq s \leq 1$) temperature. The fitted values $\sigma_{dc}$ and $s$ are given in table 2. The values of $\sigma_{dc}$ increase with the increase of frequency and temperature because of thermally activated drift mobility of ions. It calculated that ac conductivity of all Ni-doping concentrations was found to steadily increase at the selected

### Table 2. Values of $\sigma_{dc}$ (s cm$^{-1}$) and $s$ and at temperature range 320–460 K for Pure, 0.03 and 0.06 Nickel doped Zn$_{1-x}$Ni$_x$O Nanostructures.

| Composition ($x$) | T (K) | $\sigma_{dc}$ (s cm$^{-1}$) | s | $\sigma_{dc}$ (s cm$^{-1}$) | s | $\sigma_{dc}$ (s cm$^{-1}$) | s |
|-------------------|-------|-----------------|---|-----------------|---|-----------------|---|
|                   | 320   | $4.952 \times 10^{-17}$ | 1.018 | $2.595 \times 10^{-18}$ | 0.909 | $6.390 \times 10^{-18}$ | 0.718 |
|                   | 340   | $6.087 \times 10^{-17}$ | 0.974 | $1.253 \times 10^{-17}$ | 0.812 | $8.445 \times 10^{-18}$ | 0.680 |
|                   | 360   | $9.487 \times 10^{-17}$ | 0.927 | $3.156 \times 10^{-17}$ | 0.733 | $1.364 \times 10^{-17}$ | 0.653 |
|                   | 380   | $2.299 \times 10^{-16}$ | 0.874 | $6.168 \times 10^{-17}$ | 0.655 | $2.737 \times 10^{-17}$ | 0.601 |
|                   | 400   | $7.579 \times 10^{-16}$ | 0.770 | $1.062 \times 10^{-16}$ | 0.488 | $7.701 \times 10^{-17}$ | 0.580 |
|                   | 420   | $2.287 \times 10^{-15}$ | 0.655 | $1.655 \times 10^{-16}$ | 0.375 | $1.652 \times 10^{-16}$ | 0.472 |
|                   | 440   | $1.416 \times 10^{-14}$ | 0.652 | $2.902 \times 10^{-16}$ | 0.231 | $3.981 \times 10^{-16}$ | 0.443 |
|                   | 460   | $3.633 \times 10^{-14}$ | 0.451 | $5.471 \times 10^{-16}$ | 0.133 | $1.535 \times 10^{-15}$ | 0.418 |

### Table 3. Calculated activation energies from dc conductivity ($\sigma_{dc}$) grain and grain boundary resistance.

| Composition ($x$) | X = 0% | X = 3% | X = 6% |
|-------------------|--------|--------|--------|
| $\sigma_{dc}$ E$_a$ (eV) | 0.918  | 0.492  | 0.478  |
| R$_g$, E$_a$ (eV)  | 0.758  | 0.394  | 0.163  |
| R$_g$, E$_a$ (eV)  | 0.423  | 0.184  | 0.143  |

Figure 14. The variations of parameter $s$ as a function of temperatures for Zn$_{1-x}$Ni$_x$ nanostructure.
temperature at a high-frequency region at 2 MHz (460 K). It is clear from table 1, the ac conductivity of pure ZnO is $1.356 \times 10^{-4}$ S cm$^{-1}$. It increases with the increase of Ni-doping concentration, which recognized that the dopant of Ni hosts the minimum defects in the ZnO structure. These minimum flaws incline to segregation at the grain due to the diffusion procedure resulting from sintering. The electrical conductivity of ZnO with ($x = 0, 3$ and $6\%$) at low frequencies regions the dc conductivity part is independent of frequency at low-frequency regions. With the increase of frequency and temperatures, the slope of conductivity changed where the slope is changing is called hopping frequency. The value of $\sigma_{dc}$ increasing with the of temperatures as well as frequency because of thermally activated drift mobility of charge. The $\sigma_{dc}$ data is represented by Arrhenius format $\sigma_{dc} \propto \exp \left( \frac{E_a}{k_B T} \right)$ whereas $\sigma_{dc}$ is called the dc conductivity part, $\sigma_0$ is the exponential factor, $T$ is the temperature in kelvin, $E_a$ is the dc conductivity activation energy and $k_B$ is the Boltzmann constant. Figures 13(a)–(c) demonstrate the variation of dc conductivity ($\sigma_{dc}$) with the inverse of $K_B T$ of ZnO. The activation energy was calculated from a linear fit plot of $\ln(\sigma_{dc})$ versus $1/K_B T$ shown in figures 13(a)–(c). The activation energies of ZnO with ($x = 0, 3$, and $6\%$) were calculated to be $0.92$ eV, $0.49$ eV, and $0.48$ eV, respectively listed in table 3, which are nearly similar behaviour to the activation energies obtained from the grain and grain boundary resistance plots. Therefore, with the increase of conductivity, compared to pure ZnO, the activation energy declines with Ni-doping.

4. Optical properties

Absorbance spectra of pure and Ni-doped Zn$_{(1-x)}$Ni$_x$O nanostructures are shown in figure 15. The significance of nickel on the energy bandgap doped ZnO with ($x = 0, 3$, and $6\%$) exchanged of Ni$^{2+}$ ions in tetrahedral sites of ZnO. The UV-Visible optical spectroscopy was recorded in the range of 200–700 nm. The optical absorbance of doped ZnO nanostructures decreases with the increase in Ni content. In 3 and $6\%$, the Ni-doped ZnO samples’ absorption edge move towards the upper wavelength side. The change in the absorption peaks positions with the Ni-doping in ZnO samples showed the tune in band structures. The optical bandgap energy values of un-doped and Ni-doped samples were calculated using the Tauc equation $(\alpha h)^{\frac{1}{2}} = A(hv - E_g)$ [68]. From the absorbance, the constant $\alpha$ is calculated from the absorption spectra using
the relation \( \alpha = \frac{1}{d}A \) where \( A \) is called the absorbance constant and \( d \) is known as the thickness of the samples cell [68].

The energy bandgap \( E_g \) is calculated using the intercept of the linear part of curve \((\alpha h\nu)^2\) along \( Y = 0 \). The plots of \((\alpha h\nu)^2\) as a function of energy are shown in figure 16. We calculated the energy bandgap of Zn\(_{(1-x)}\)Ni\(_x\)O with \( x = 0.00, 0.03, \) and \( 0.06 \) nanostructures decreasing from 3.2 to 3.0 eV with increasing of Ni concentrations. The reduction in bandgap energy is clearly shown in figures 16(a)–(c). The sp-d spin alteration interface might be among the band of electrons and d localized electrons of Ni ions doping in Zn sites [71]. Optical energy bandgap decreases with increasing Ni concentrations due to an increase in average crystallite size. The experimental project indicates the change in a redshift in the bandgap also observed, which shows that the redshift of the bandgap approves the doping of Ni ions in Zn\(_{(1-x)}\)Ni\(_x\)O nanostructures. The increase of impurities and defect amounts under doping of Ni influence may be another reason for decreasing bandgap energy [72].

4.1. Conclusion
Zn\(_{(1-x)}\)Ni\(_x\)O nanostructures have been prepared by the co-precipitation method. All samples show the formation of nanostructures; with the increase of Ni, the shape of the particle changes from spherical to rod types with larger crystallite sizes. It observed that doping in ZnO plays a vital part in enlightening its dielectric, electrical and optical properties. The dielectric constant, dielectric loss and tangent loss tendencies decrease with the increase in frequency but increase with the doping of Ni in ZnO. Dielectric constants show dispersion type of behaviour in a low-frequency area. Based on Koop’s theory of space-charged polarization, the ac conductivity increases with increasing frequency at different temperatures and the doping concentrations. The parameters in the power-law show the predominate of the correlated barrier hopping (CBH) model for un-doped and Ni-doping. Nyquist plots indicated that a decrease in impedance values was observed with the increase in Ni content. All un-doped and doped samples have a negative temperature coefficient of resistance (NTCR) behaviour. Moreover, the energy bandgap of ZnO decreased with increasing Ni doping.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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