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Investigation of Dielectric Response and AC Conductivity of NiO/ZnO Nanocomposites for High-Frequency Dielectric Applications

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Abstract

The rapid development of wireless communication networks and microwave devices has increased the requirement for dielectric materials with improved electrical performance and low dielectric loss at high frequencies. In the current study, NiO nanoparticles, ZnO nanoparticles, and NiO/ZnO nanocomposites were synthesized using a chemical precipitation method. The structural characteristics of the synthesized materials were investigated by X-ray diffraction (XRD), which confirmed the formation of cubic NiO and hexagonal ZnO phases. The crystallite sizes of NiO, ZnO nanoparticles and the NiO/ZnO nanocomposite were found to be 30 nm, 20 nm, and 14.47 nm, respectively. Nitrogen adsorption-desorption showed that NiO/ZnO nanocomposite exhibited a higher specific surface area $41.827 \text{ m}^2 \text{ g}^{-1}$ compared with $32.71 \text{ m}^2 \text{ g}^{-1}$ for NiO nanoparticles, improved surface properties due to the synergistic interaction between NiO and ZnO. High-resolution transmission electron microscopy (HRTEM) confirmed particle sizes of approximately 25 nm for NiO and 18 nm for the NiO/ZnO. Dielectric studies demonstrated that both the dielectric constant and dielectric loss decrease with increasing frequency due to reduced interfacial and space-charge polarization effects. Among all samples, the NiO/ZnO nanocomposite exhibited the lowest dielectric loss (≈ 0.30 at 10^6 Hz) and higher AC conductivity. These enhanced electrical properties arise from strong interfacial interactions between NiO and ZnO, making the nanocomposite a promising candidate for high-frequency dielectric and electronic applications.

Keywords: NiO nanoparticles, ZnO nanoparticles, NiO/ZnO nanocomposites Dielectric properties, Loss tangent, AC conductivity, high frequency dielectric materials

1 Introduction

Advances in communication technologies such as satellite, wireless, radar, and 5G/6G networks have increased the need for dielectric materials that operate reliably at high frequencies [1]. High dielectric constants, dielectric loss, loss tangents, and electrical conductivities can limit the performance of RF and microwave systems. These issues can cause signal loss, impedance mismatch, lower transmission efficiency, and make it harder to miniaturize devices for high-frequency use [2]. To overcome these issues, nanodielectrics and their composites play a major role due to their small particle size and (Maxwell–Wagner) polarization. This characteristic property improves the dielectric constant, dielectric loss, loss tangent, and frequency-dependent electrical conductivity [3]. At low frequencies, nanodielectric shows a high dielectric constant as space-charge polarization occurs, whereas at high frequencies, confined dipole motion stabilises the permittivity and reduces energy loss, which is required for high-frequency dielectric applications [4].

Nickel oxide (NiO) is a P-type semiconductor with a wide bandgap of 3.6 to 4.0 eV. It is affordable and has good chemical, thermal, and dielectric properties. NiO is mainly used in energy storage, energy conversion, sensors, and high-frequency devices [5]. However, it tends to clump together, exhibits low electrical conductivity, and shows high dielectric loss at high frequencies [6]. To figure out the above problems, NiO can be improved with conductive materials like other metal oxides, graphene, or carbon nanotubes (CNTs). Zinc oxide (ZnO) is an n-type semiconductor from the II-VI group, with a direct band gap of 3.37 eV. ZnO is chemically stable and has useful optical and electrical properties, making it suitable for solar cells, gas sensors, photocatalysis, UV protection, and pharmaceuticals [7-8].

Gokul et al. [9] reported that NiO nanoparticles synthesised by the wet chemical method showed a dielectric constant of 35 at low frequencies, due to space charge polarization. Sharma et al. [10] reported enhanced dielectric constant and crystal structure in NiO at higher annealing temperatures. NiO incorporated with other metal oxides shows enhanced dielectric properties compared with pure NiO. Mahajan et al. [11] reported that NiO/ZnO nanocomposites exhibit enhanced dielectric properties through interfacial polarisation between NiO and ZnO, making them suitable for dielectric applications in the MHz frequency range.

Numerous researchers have synthesized NiO and ZnO nanoparticles and investigated their dielectric characteristics for potential applications such as energy storage and communication. NiO has received considerable attention due to its thermal stability and dielectric properties, while ZnO is widely used because of its unique electrical and optical properties. NiO based nanocomposites have been investigated to enhance dielectric characteristics and electrical conductivity. Existing research on NiO and ZnO materials have mainly focused on dielectric characterization and energy storage applications. Very few studies have examined the dielectric behavior of NiO/ZnO nanocomposites at frequency relevant to communication systems. The properties of NiO/ZnO nanocomposites in terms of dielectric loss, AC conductivity, and stability at high frequencies remain inadequately explored. The demand for low dielectric loss and stable electrical characteristics in high frequency and dielectric materials and electronic components indicates the need for further research.

The main objective of this research work is to synthesize NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposites and comparatively investigate their dielectric properties. The microstructural characteristics, dielectric response, loss tangent, and AC conductivity of NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposites were investigated to examine their electrical performance and dielectric behavior. Reduced crystallite size, enhanced specific surface area, lower dielectric loss, and

improved AC conductivity were observed for the NiO/ZnO nanocomposite relative to the individual oxide nanoparticles, indicative of the beneficial role of nanocomposite formation.

2. Experimental procedure

2.1 Materials

For the synthesis of the NiO/ZnO nanocomposite, nickel acetate tetrahydrate ($\text{Ni}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$), zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$), and sodium hydroxide (NaOH) were procured from Sigma-Aldrich and used as received without further purification.

2.2 Synthesis of NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposite

For the synthesis of NiO nanoparticles, 0.2 M nickel acetate solution was prepared in 100 ml of (Deionized) DI water and stirred at 1000 rpm using a magnetic stirrer until homogeneous solution was obtained. Subsequently, a 0.2 M NaOH solution was added dropwise while stirring continuously to induce precipitation. NiO nanoparticles were obtained by filtering and drying the resulting precipitate at 100°C to obtain NiO nanoparticles.

ZnO nanoparticles were synthesized by dissolving 0.2 M Zinc acetate in 100 mL DI water under stirring at 1000 rpm, followed by dropwise addition of 0.2 M NaOH to induce precipitation. The precipitate was filtered, washed, dried at 100 °C to obtain ZnO nanoparticles. The NiO nanoparticles and ZnO nanoparticles should be pressed into pellets and sintered at 450 °C for dielectric measurements.

The NiO/ZnO nanocomposites were prepared by mixing equal volume of (100 ml) 0.2M nickel acetate and zinc acetate solutions under vigorous magnetic stirring. A 0.2 M NaOH solution was added to the mixture to induce precipitation. The precipitate was filtered, dried at 100°C, to obtain the NiO/ZnO nanocomposites, which were subsequently pressed into pellets and sintered at 450 °C for dielectric measurements.

2.3 Characterization

The crystal structure of the synthesized samples was investigated using X-ray diffraction (XRD) performed on a PANalytical diffractometer with $\text{Cu K}\alpha$ radiation ($\lambda = 0.154\text{nm}$). Morphological characterization was carried out by Transmission Electron Microscopy (TEM) employing a TITAN TEM instrument. The specific surface area and pore characteristics were assessed through volumetric nitrogen adsorption-desorption measurements, with data interpreted using the BJH model on an ASAP 2420 V2.09 analyser. Dielectric properties, encompassing dielectric constant, dielectric loss, loss tangent, and AC conductivity, were evaluated using a HIOKI LCR Meter (IM3536) over a frequency range of 10^1 – 10^6 Hz at room temperature.

3 Results and discussion

3.1 Structural analysis

Figure 1 shows the XRD pattern of NiO, ZnO, and NiO/ZnO nanocomposite. The diffraction results confirm that NiO exhibits a cubic crystal structure (JCPDS 78-0643) [12], while ZnO shows a hexagonal structure (JCPDS 89-7102) [13]. The calculated crystallite size for NiO, ZnO, and NiO/ZnO are 30nm, 20nm and 14.47nm, respectively. The XRD patterns of NiO and NiO/ZnO appear similar, indicating that the incorporation of ZnO does not significantly alter the NiO phase. The presence of ZnO reduces agglomeration and leads to a decrease in the crystallite size of NiO in the composite.

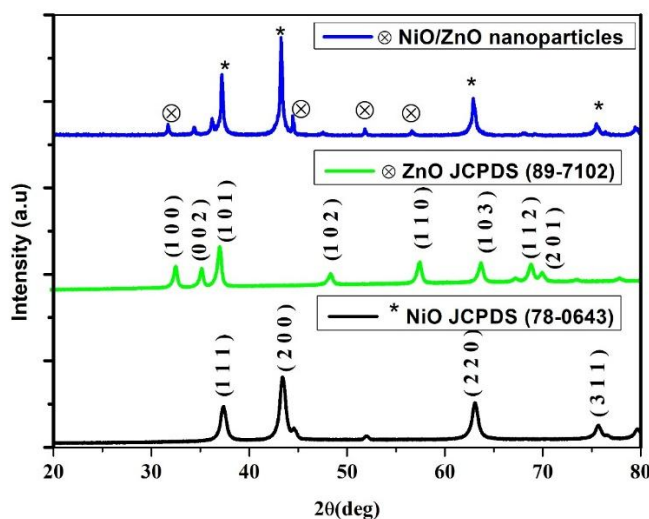


Figure 1. (a) NiO Nanoparticle, (b) ZnO Nanoparticle and (c) NiO/ZnO Nanocomposite

3.2 Morphological analysis

The HRTEM images in Figs. 2(a) and 2(c) reveal the structural morphology of the NiO nanoparticles and the NiO/ZnO nanocomposite, respectively, whereas the corresponding SAED patterns are presented in Figs. 2(b) and 2(d). The NiO nanoparticle show an average particle size of ~25nm in size and tends to agglomerate. In the NiO/ZnO nanocomposite, the particle size is reduced to 18nm and the ZnO plays a role in controlling the growth of NiO. The SAED patterns for both samples exhibit well-defined diffraction rings, which confirm the crystalline nature of the materials.

3.3 Surface-area analysis

The nitrogen adsorption-desorption isotherms of NiO nanoparticles and NiO/ZnO nanocomposite samples are shown in Fig. 3. Both samples exhibit a type IV isotherm, which is characteristic of mesoporous materials. At low relative pressure ($P/P_0 < 0.4$), the gradual increase in adsorbed nitrogen indicates monolayer adsorption on the surface of the materials. At higher relative pressure ($P/P_0 > 0.7$), a sharp increase in adsorption is observed due to capillary condensation in the mesopores.

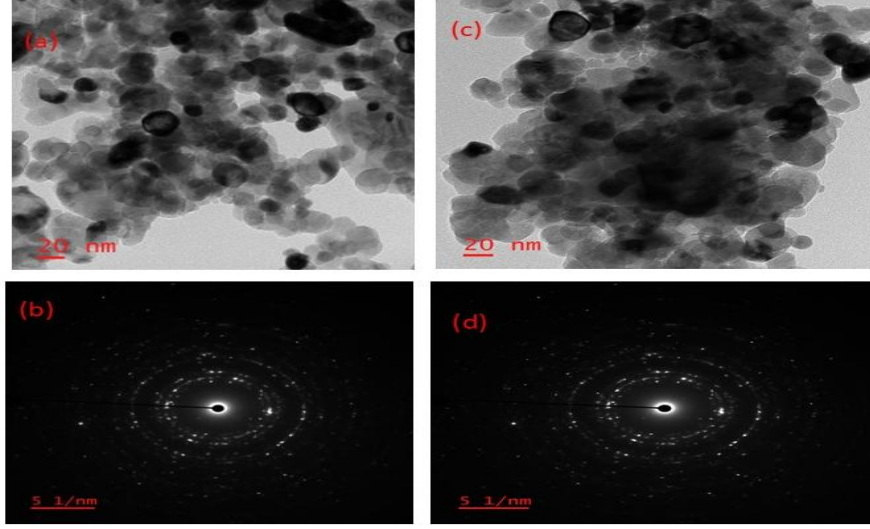


Figure. 2 (a and b) NiO nanoparticles and SAED pattern and (c and d) NiO/ZnO nanocomposite and SAED pattern

The NiO/ZnO nanocomposite exhibits a higher adsorption capacity than NiO nanoparticles, indicating an enhanced specific surface area and pore structure. The BET analysis revealed a surface area of $41.827\text{m}^2\text{g}^{-1}$ for the NiO/ZnO nanocomposite, which is higher than that of NiO nanoparticles ($32.71\text{m}^2\text{g}^{-1}$). The NiO/ZnO nanocomposite average pore diameter and volume were 19.7nm and $0.110\text{cm}^3\text{g}^{-1}$, respectively, which are larger compared to NiO nanoparticles (15.7nm and $0.079\text{cm}^3\text{g}^{-1}$). These results show that incorporating ZnO improves pore size and volume. Therefore, to thus, it provides more accessible surface sites for improved dielectric and high-frequency applications.

3.4 Frequency-dependent dielectric constant

Dielectric properties of materials are characterised by the complex dielectric constant (ϵ), which is represented by:

$$\epsilon = \epsilon' - j\epsilon'' \quad (1)$$

where a real component, ϵ' , describes the stored energy and an imaginary component, ϵ'' related to energy dissipated with the material. The influence of frequency on the real and imaginary parts of the dielectric constants for NiO nanoparticles, ZnO nanoparticles, NiO/ZnO nanocomposite as shown in Figures 4 and 5. The dielectric constants ϵ' and ϵ'' of the materials have been computed by the relation

$$\epsilon' = \frac{C_p t}{A} \epsilon_0 \quad (2)$$

where C_p is the measured capacitance of the sample, t is the thickness of the pellet, A is the surface area of the electrode, and ϵ_0 is the permittivity of free space.

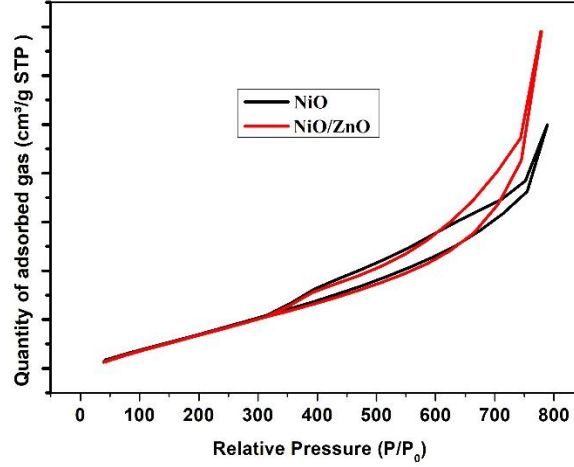


Figure. 3 BET nitrogen adsorption–desorption isotherms of NiO and NiO/ZnO.

The variation of dielectric constant with frequency reveals that both ϵ' and ϵ'' decrease as frequency increases for all samples. The decrease in ϵ'' is more rapid compared to ϵ' , and both parameters tend to approach nearly constant values at higher frequencies. In the lower-frequency region, comparatively high dielectric constant values. This behavior is due to the dominant contribution of space-charge and interfacial polarizations. At this frequency range, charge carriers collect at grain boundaries and interfaces between the material. The accumulation of charges improves the polarization method and dielectric constant values. The dielectric constant progressively decreases as polarization reduces, an effect of dipoles and charge carriers being unable to respond effectively to the rapidly alternating electric field at higher frequencies. This dielectric dispersion behavior can be explained by the Maxwell–Wagner interfacial polarisation mechanism together with Koop’s phenomenological theory. The dielectric behaviour of material can be explained with Koop’s theory, which represent the materials as a heterogeneous structure consisting of conducting grains and resistive grain boundaries. The heterogeneous structure contributes distinctly to the overall permittivity and dielectric loss response. The dielectric response at low frequencies is mainly controlled by grain boundary contributions, which facilitate large polarization and which improves higher dielectric constant values in the material [15].

Among the studied nanoparticles, NiO shows the highest dielectric constant of approximately $\epsilon' \approx 5.5$ at low frequencies ($\sim 10^1$ Hz), attributed to improved polarization contributions, whereas ZnO nanoparticles yield a comparatively lower value of $\epsilon' \approx 1.2$. The NiO/ZnO nanocomposite displays comparatively lower dielectric constant values of approximately $\epsilon' \approx 0.28$. The frequency increases to 10^3 Hz, the dielectric constant decreases for all the samples with NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposite values of $\epsilon' \approx 0.45, 0.09$ and 0.03 . At higher frequencies ($\sim 10^6$ Hz), the dielectric constant becomes nearly stable, with values of approximately $\epsilon' \approx 0.045$ for NiO, $\epsilon' \approx 0.011$ for ZnO, and $\epsilon' \approx 0.007$ for the NiO/ZnO nanocomposite. NiO nanoparticles shows the highest dielectric constant across the complete frequency range among all investigated samples, a conduct that may be attributed to the greater number of polarization centers and charge carriers inherently present in the material. At higher frequencies, the NiO/ZnO nanocomposite exhibits a lower and stable dielectric constant. This may be due to the reduced response of polarization and charge carriers to the rapidly varying electric field. The enhanced dielectric behavior is formed interfacial interactions between NiO and ZnO phases. Such stable dielectric behavior is advantageous for high-frequency electronic and RF communication applications, where consistent dielectric properties are required.

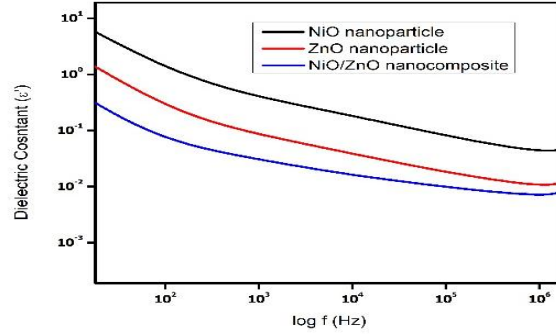


Figure 4. Frequency-dependent dielectric constant of NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposite.

3.5 Frequency-dependent dielectric loss

The variation of dielectric loss with frequency reveals that ϵ'' decreases as frequency increases for all samples, and this decrease is more rapid compared to ϵ' , with ϵ'' also tending to approach a nearly constant value at higher frequencies. In the lower-frequency region, the samples exhibit comparatively high dielectric loss values, which is likewise attributed to the dominant contribution of space-charge and interfacial polarization. At this frequency range, charge carriers collect at grain boundaries and interfaces between the materials, and the accumulation of these charges increases energy dissipation and, consequently, the dielectric loss values. As frequency increases, the dipoles and charge carriers are unable to respond effectively to the rapidly alternating electric field, causing the dielectric loss to fall sharply before stabilizing.

This behavior can similarly be explained using the Maxwell–Wagner interfacial polarization mechanism together with Koop's phenomenological theory, where the conducting grains and resistive grain boundaries of the heterogeneous structure contribute distinctly to the overall dielectric loss response. The loss response at low frequencies is mainly controlled by grain boundary contributions, which facilitate large polarization and correspondingly higher energy dissipation; as frequency increases, this grain-boundary contribution diminishes rapidly, causing ϵ'' to decrease faster than ϵ' and eventually stabilize at high frequencies.

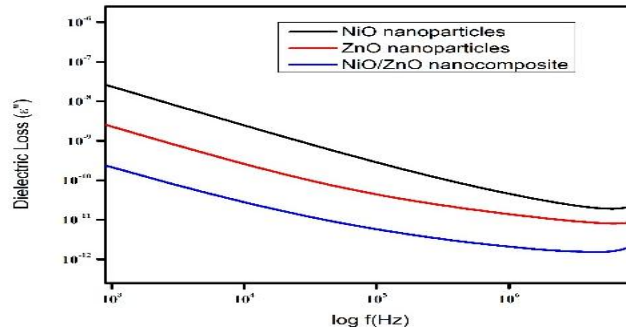


Figure 5. Frequency-dependent dielectric Loss of NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposite

The Figure 5 shows the NiO nanoparticles have the highest dielectric loss of approximately $\epsilon'' \approx 3 \times 10^{-7}$ at low frequencies ($\sim 10^2$ Hz), attributed to greater charge-carrier density and enhanced interfacial polarization, whereas ZnO nanoparticles yield a comparatively lower value of $\epsilon'' \approx 3 \times 10^{-8}$. The NiO/ZnO nanocomposite displays the lowest dielectric loss of approximately $\epsilon'' \approx 2 \times 10^{-9}$ at this frequency. As the frequency increases to $\sim 10^4$ Hz, the dielectric loss decreases sharply for all samples, with NiO nanoparticles, ZnO nanoparticles, and the NiO/ZnO nanocomposite showing values of $\epsilon'' \approx 4 \times 10^{-9}$, 8×10^{-10} , and 6×10^{-11} , respectively. At higher frequencies ($\sim 10^6$ Hz), the dielectric loss becomes nearly stable, with values of approximately $\epsilon'' \approx 2 \times 10^{-11}$ for NiO, $\epsilon'' \approx 8 \times 10^{-12}$ for ZnO, and $\epsilon'' \approx 2 \times 10^{-12}$ for the NiO/ZnO nanocomposite.

NiO nanoparticles exhibit the highest dielectric loss across the entire frequency range among the investigated samples, consistent with their higher dielectric constant and attributed to the greater number of charge carriers and polarization centers inherently present in the material. In comparison, the NiO/ZnO nanocomposite exhibits the lowest dielectric loss values and enhanced stability at higher frequencies, indicating suppressed charge-carrier contributions and reduced active polarization mechanisms formed through interfacial interactions between the NiO and ZnO phases. This low and stable dielectric loss behavior is advantageous for high-frequency electronic and RF communication applications, where minimal energy dissipation is required.

3.6 Loss Tangent

The loss tangent ($\tan \delta$) describes the energy dissipation within a dielectric material and is directly referred to the imaginary component of the dielectric constant (ϵ''). It shows the amount of electrical energy converted into heat during polarization processes in the material. The loss tangent is expressed as

$$\tan \delta = \epsilon''/\epsilon' \quad (3)$$

Figure 6 shows the variation of dielectric loss tangent with frequency for NiO nanoparticles, ZnO nanoparticles, and the NiO/ZnO nanocomposite. The loss tangent decreases slowly with increasing frequency across NiO, ZnO nanoparticles and NiO/ZnO nanocomposites. At lower frequencies (around 10^3 Hz), the dielectric loss. For NiO, ZnO nanoparticles and NiO/ZnO nanocomposite. the loss tangent values are ~ 35 , 11 and 6 respectively. Because charge carriers and dipoles are able to follow the slowly changing electric field, there is a greater loss at low frequencies. This leads to increased interfacial and dipolar polarisation and higher dielectric loss.

As the frequency increases, the response of charge carriers and dipoles becomes slower compared with the rapidly alternating electric field. Due to this limitation, the polarization mechanisms gradually weaken, resulting in a reduction in dielectric loss. At around 10^4 Hz, the loss tangent decreases to nearly 12 for NiO, about 3.5 for ZnO, and approximately 1.5 for the NiO/ZnO nanocomposite. When the frequency further increases to 10^5 Hz, the loss values reduce to roughly 4 for NiO, 1.2 for ZnO, and about 0.6 for the NiO/ZnO nanocomposite.

At higher frequencies 10^6 Hz, the dielectric loss decreases significantly. The dielectric loss decreases of NiO, ZnO nanoparticles and NiO/ZnO nanocomposites 0.9 , 0.45 and 0.30 , respectively. At higher frequencies, dielectric loss tends to be lower, as the charge carriers and dipoles respond less effectively to the rapidly changing electric field. NiO nanoparticles shows the high dielectric loss during the entire frequency range while ZnO nanoparticles show comparatively lower loss values. By NiO and ZnO nanoparticles the

NiO/ZnO nanocomposite shows the reduced loss tangent at higher frequencies. The lower dielectric loss in the composite can be related to improve interfacial interaction between the NiO and ZnO which control accumulation and reduce conduction-related losses. The lower dielectric loss in the NiO/ZnO nanocomposite (≈ 0.30 at 10^6 Hz) shows that the material is suitable for high-frequency dielectric applications, where minimal energy dissipation is required.

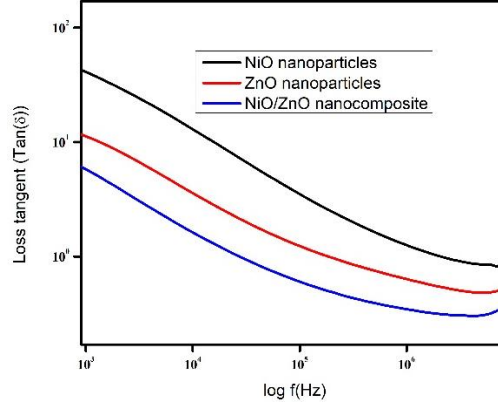


Figure 6. Loss tangent of NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposite.

3.7 AC Conductivity

The AC conductivity, σ_{AC} is measured using the following relation:

$$\sigma_{AC} = \epsilon' \epsilon_0 \omega \tan \delta \quad (\Omega^{-1} \text{ cm}^{-1}) \quad (4)$$

Figure 7 shows the AC conductivity of NiO, ZnO nanoparticles and the NiO/ZnO nanocomposite at various frequencies. It can be seen that the conductivity increases gradually with increasing frequency for all the samples, because, at higher frequencies, charge carriers can respond more quickly to the alternating electric field. As a result, electrons can move more easily between nearby sites, increasing conductivity.

At lower frequencies, the conductivity remains relatively small since the movement of charge carriers is limited by grain boundaries and structural defects present in the material. As the frequency increases, electrons gain enough energy to move between localized states more easily, which improves the electrical conduction.

Among the three samples, the NiO/ZnO nanocomposite shows the highest conductivity throughout the frequency range, while ZnO nanoparticles show moderate conductivity and NiO nanoparticles exhibit the lowest values. The improved conductivity in the NiO/ZnO nanocomposite is mainly due to the interaction between NiO and ZnO phases. The interface between these two materials provides additional pathways for electron transport, allowing charge carriers to move more easily through the composite than through the individual oxides.

Therefore, the enhanced conductivity observed in the NiO/ZnO nanocomposite indicates that combining NiO with ZnO improves the electrical transport behaviour, which can be beneficial for high-frequency electronic and communication applications.

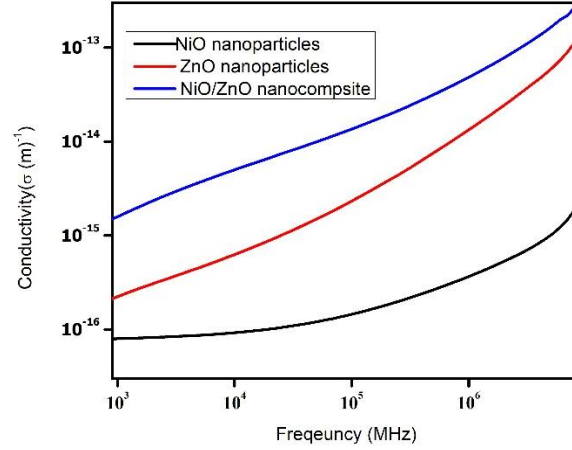


Figure 7. AC Conductivity of NiO nanoparticles, ZnO nanoparticles and NiO/ZnO nanocomposite.

4 Conclusions

In this study, NiO nanoparticles, ZnO nanoparticles, and NiO/ZnO nanocomposites were successfully synthesised using a chemical precipitation method. The X-ray diffraction analysis confirmed the formation of cubic NiO and hexagonal ZnO phases, while the crystallite size of the NiO/ZnO nanocomposite (14.47 nm) was smaller than that of the individual oxides. BET analysis revealed that the NiO/ZnO nanocomposite possesses a higher specific surface area ($41.827\text{m}^2\text{g}^{-1}$) compared with NiO nanoparticles ($32.71\text{m}^2\text{g}^{-1}$), indicating improved microstructural characteristics. HRTEM observations further confirmed nanoscale particle size: NiO nanoparticles were about 25 nm, while the NiO/ZnO nanocomposite showed smaller particles of about 18 nm. The reduced loss tangent of the NiO/ZnO nanocomposite (≈ 0.30 at 10^6 Hz) in comparison to NiO (≈ 0.90) and ZnO (≈ 0.45) nanoparticles is attributed to the synergistic effect between NiO and ZnO. At low frequencies ($\sim 10^1$ Hz), NiO nanoparticles exhibited the highest dielectric constant of $\epsilon' \approx 5.5$, while ZnO nanoparticles and the NiO/ZnO nanocomposite comparatively lower values of $\epsilon' \approx 1.2$ and $\epsilon' \approx 0.28$, respectively. At higher frequencies ($\sim 10^6$ Hz), the dielectric constant stabilized at approximately $\epsilon' \approx 0.045$, $\epsilon' \approx 0.011$, and $\epsilon' \approx 0.007$ for NiO, ZnO, and NiO/ZnO nanocomposite, respectively. The heterointerfaces established at the NiO–ZnO phase boundaries generate potential barriers that restrict excessive charge accumulation and suppress interfacial polarization losses, thereby minimizing energy dissipation within the composite structure. The reduced crystallite size of the nanocomposite results in a higher density of grain boundaries, which collectively impede charge carrier mobility and minimize energy dissipation during each polarization cycle. Collectively, this synergistic effect is responsible for the superior dielectric performance of the NiO/ZnO nanocomposite.

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Author contributions

Author Contributions: Hari Sivaram: Writing – original draft, Formal analysis, Data curation, Conceptualization. S. Sriram: Methodology, Investigation, Formal analysis, Data curation, Conceptualization. Duraisamy Selvakumar: Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. Hitesh Borkar: Writing – review & editing, Validation, Supervision, Project administration, Conceptualization. : Maxim Shevtsov Writing – review & editing, Validation, Supervision, Project administration. All authors have read and agreed to the published version of the manuscript.

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