Effect of Temperature on the Dielectric and Magnetic Properties of $\text{NiFe}_2\text{O}_4@ \text{MgFe}_2\text{O}_4$ and $\text{ZnFe}_2\text{O}_4@ \text{MgFe}_2\text{O}_4$ Core-Shell

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Abstract

The core-shell $\text{NiFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$ ($\text{NiF}@\text{MgF}$) and $\text{ZnFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$ ($\text{ZnF}@\text{MgF}$) are stable nanocomposites. The experimental results showed perfect dielectric and magnetic properties different than their components. The experimental data revealed that the mutual effect between the core and the shell increases the space charge polarization. Also, the samples showed semiconducting-metallic behavior, which varies according to the temperatures and the frequencies. Furthermore, the magnetization $M(T)$ was studied using the Faraday balance method of all samples. The obtained results of $M(T)$ exhibit good magnetic properties of the core-shell samples, particularly the sample $\text{ZnF}@\text{MgF}$, where it possesses magnetization higher than the pure ferrite phase (MgFe_2O_4) and Curie temperature (T_{Cm}) higher than the room temperature, and this is new for Zn-ferrite. Besides, the effective magnetic moment (μ_{eff}) and the Curie-Weiss constant (θ) were obtained from the magnetic susceptibility $\chi(T)$ protocols.

Keywords: $\text{NiFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$, $\text{ZnFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$, Core-Shell, dielectric properties, magnetic susceptibility.

1. Introduction

The nanocomposite possesses many applications especially, in multi-function devices, transducer, actuators, and sensors, so researchers have been interested in developing them. The physical properties of nanocomposite are due to their components, and it depends on the form of accumulation of their components. So, many accumulating geometric shapes of the nanocomposite such as particulate, columnar structures, multilayers, and core-shell

nanostructures were studied in the literature [1–4]. But, the nanocomposite that consists of only nanoferrites has rarely been studied, although it one of the configurations for multisystem and that have useful physical properties by controlling their chemical composition and the size of the structure.

The ferrite family has many compounds such as the NiFe_2O_4 , MgFe_2O_4 , ZnFe_2O_4 , CuFe_2O_4 , MnFe_2O_4 , CoFe_2O_4 , etc., which are magnetic materials that have a cubic spinel structure [5–17]. The familiar of this family is the nickel ferrite, which has significant magnetic properties. Also, the electrical and magnetic properties were studied to a larger extent in the literature [15–21] for the MgFe_2O_4 and ZnFe_2O_4 . Besides, mixed Ni-Mg and Ni-Zn ferrites were studied in the literature [22–25] and obtained results indicate the variation of the electrical and magnetic properties, where electrical and magnetic properties were dependent on the method of preparation and the particle size.

The microstructure, morphological and magnetic hysteresis loops of $\text{NiFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$ and $\text{ZnFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$ core-shell nanoparticles were reported in previous work [4]. In this work, the dependence of the dielectric properties, and the magnetization of the core-shell $\text{NiFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$, and $\text{ZnFe}_2\text{O}_4@\text{MgFe}_2\text{O}_4$ nanoparticles on the temperature and the frequency will be discussed in detail.

2. Experimental and Calculations

The AC measurement was carried out in the frequency range of 100Hz-8MHz and the temperature range of 300–650K using the HIOKI IM3536 LCR meter. A high-quality silver paste coating was applied on the contact surface to make parallel plate capacitor geometry and the ferrite material as a dielectric medium. For temperature control, the sample was mounted in an electric oven. The value of capacitance C , Impedance Z , Impedance phase angle θ , and loss factor $\tan\delta$ measured directly from the LCR meter, and the other parameters will be calculated from the relations::

$$\varepsilon = (C \times t) / (A) \quad (5)$$

and

$$\sigma = t / (Z \times A) \quad (6)$$

where ε is the dielectric constant, and σ is the ac conductivity, C is capacitance (in Farad), t is thickness of the sample (cm), A is the surface area of the sample (cm^2).

A Faraday balance is a device for measuring magnetic susceptibility. In this technique, the sample is suspended between electromagnet cores where a magnetic field is applied. Then, the sample is heated gradually using a non-inductive furnace. After measuring the pull of the balance (Δm), many parameters can be calculated such as the molar magnetic susceptibility (χ_m), and the molar magnetization (M_m) can be calculated using the following relations respectively:

$$\chi_m = [(\Delta m \times g \times M_w)] / [m \times H \times (dH/dZ)] \quad (7)$$

$$M_m = \chi_m \times H \quad (8)$$

where: g is the gravity = 980.6 cm/s², M_w is the molecular weight of the sample, m is the mass of the sample, H is the magnetizing field applied, dH/dZ is the magnetic field gradient in the z -direction.

For simplicity, we will name the pure sample MgFe₂O₄ and both core-shell samples NiFe₂O₄@MgFe₂O₄, and ZnFe₂O₄@MgFe₂O₄ by the name MgF, NiF@MgF, and ZnF@MgF respectively.

2. Results and discussion

2.1 Dielectric properties

Figure 1 shows the variation of the dielectric constant (ϵ) in the frequency range (100Hz - 8MHz) and the temperature range (300- 650K) of the pure sample MgF, and both core-shell ZnF@MgF, NiF@MgF. For all samples (Fig 1), the value of the dielectric constant (ϵ) is high at the low-frequency region (Hz region) then decrease at the middle-frequency region (kHz region). So, the dispersion in the frequency Hz-kHz region of the dielectric constant (ϵ) can be explained according to the Maxwell–Wagner relaxation [26, 27], and agreement with Koop's phenomenological theory [28]. The contributions from all types of polarizations like dipolar, interfacial, ionic, and electronic may be the reason for the high value of the dielectric constant in the low-frequency region. But in the high-frequency region (MHz region), the dipoles are unable to follow the applied electric field and the only contribution from the electronic polarization so, a lower value of the dielectric constant is obtained. Besides, the NiF@MgF possesses the highest value of dielectric constant compared to both values of ZnF@MgF and MgF. As mentioned in the literature [29], the microstructure and the phase composition mainly affects the dielectric properties. So, the increase in grain size may be the reason for a significant increase in the dielectric constant of all samples, where the value of the grain size of MgF, ZnF@MgF, and

NiF@MgF samples is 21, 36, and 159 nm, respectively [4]. In other words, "the larger grains possess a higher value of the dielectric constant". Although getting a low dielectric constant value of the core-shell samples NiF@MgF and ZnF@MgF, they possess a higher value of dielectric constant compared to the MgF. Reddy et al. [30] explained the increase in dielectric constant to the presence of impurities (extra phase) around grain boundaries which is responsible for the significant increase. The same condition can be applied to the present case. The core-shell possesses two phases formed around each other boundaries causing their overall resistance to increase. The difference between the charge carriers in the grain interior and grain boundaries produces an interfacial polarization that may be causing the overall dielectric constant to increase [30-32]. In other words, the composite has two heterogeneous substances that may produce an increase in the dielectric constant. The interaction between the two substances (MgF and ZnF – MgF and NiF) leads to a rise in the space charge polarization between them and increasing the dielectric constant. When the electric field is applied on the core-shell NiF@MgF, the space charge created due to the NiF phase accumulates at the interface of the MgF. Therefore, the space charges created due to NiF increases subsequently giving higher values for the dielectric constant. Similar behavior has also been reported in different composites such as the core-shell CuO/CuFe₂O₄ [33], the nanocrystalline CoGd_xFe_{2-x}O₄ [34], and the multiferroic composites BaTiO₃-NiFe₂O₄ [35].

At the octahedral sites of spinel ferrite, an electronic exchange occurs between the Fe²⁺ and Fe³⁺ ions and produces the local displacement of electrons in the direction of the applied electric field, and this results in electrical polarization. So, with increasing the electronic exchange, the polarization and the dielectric constant will be increased. This meaning that if there is more exchange between Fe²⁺ and Fe³⁺ ions, the electrical polarization will be increased. Now the core-shell composite possesses two nanoferrite substances, more exchange between Fe²⁺ and Fe³⁺ ions will occur so, the increase of the electrical polarization is expected. Thus, the mechanism for the electron and hole exchange in the core-shell can be represented as:

where T.E. is the divalent transition element.

Fig. 2 shows the dependence of the dielectric constant (ϵ) on the temperature at certain frequencies of all nanoparticle samples. It is observed that a broadened peak at the low-temperature region due to the contribution of all types of polarizations like dipolar, interfacial, ionic, and electronic of all samples. In other words, the mobility of charge carriers increases with increasing temperatures at low temperatures (semiconducting behavior), leading to an increase in the sample's conductivity and polarization and thereby increasing the dielectric constant. With increasing the temperature, the increase of the dielectric constant is slow indicating that, the polarizations become weak, which means that the contribution of the electric dipoles is the only take place. Also, the intensity of the peak decrease with increasing frequency.

The $\tan \delta$ value indicates the loss of electrical energy in the sample. So, the variation of loss tangent ($\tan \delta$) as a function of frequency and temperature is shown in Fig. 3 and 4, respectively. It is observed from Fig. 3 the value of $\tan \delta$ decreases rapidly in the low-frequency region (Hz), while in the middle region (kHz) the rate of decrease is slow and then becomes almost zero in the high region (MHz) i.e., the loss tangent ($\tan \delta$) is independent on the frequency in the high-frequency region. In the low-frequency region, the value of $\tan \delta$ is high this means that, the loss of electrical energy value is high. Such behavior due to the low conductivity of grain boundaries, so more energy is required for an electron to exchange between Fe^{2+} and Fe^{3+} ions. In the high-frequency region, the situation will be reversed, where the conductivity of grains will take place in the high-frequency region [34, 36] so, small energy is required to transfer an electron between the Fe^{2+} and Fe^{3+} ions at the octahedral site. The value of $\tan \delta$ decreases with the frequency because of the decrease of the conductivity for the core-shell. In other words, the dielectric loss decreases for the core-shell due to the presence of two substances that have different grain structures. Fig. 3c shows the relation between the $\tan \delta$ and the frequency of NiF@MgF , as exhibits it is an abnormal shape.

It is observed from Fig. 3 that, all samples exhibit a loss peak in the MHz region following the Debye type of relaxation. This peak is strong for MgF and ZnF@MgF and weak for NiF@MgF . This peak is observed in ferrite materials when there is a resonance between the frequency of the electrons or the holes of ions and the frequency of the applied field, so the jumping/hopping will occur. In other words, this peak is resonance a phenomenon, i.e., maximum electric energy is transferred to the electrons and the loss shoots up at resonance. The

electric dipoles cannot follow the alternating field instantaneously; some time is needed for the alignment of dipoles with the field [37]. The maximum peak in the curve of the dielectric loss ($\tan\delta$) vs frequency is obeyed to the following expression [38]:

$$\omega\tau = 1 \quad (12)$$

where $\omega = 2\pi f_{\max}$, τ is the relaxation time, and $f_{\max}$ is the maximum frequency. The relaxation time (τ) and the jumping probability per unit time is related as:

$$\tau = 1/2p \quad (13)$$

Thus, the maxima frequency is the hopping frequency of electrons and equal to the applied frequency alternating electric field when the electrons are hopping as in Eqs. (9-11); $f_{\max} \propto p$.

The temperature dependence of the loss tangent ($\tan\delta$) in the frequency range from 100 Hz to 8 MHz is shown in Fig. 4. The value of the dielectric loss was lower at lower temperatures and increased with an increase in temperature, similar to the behavior of the dielectric constant. Dielectric loss peak was observed, particularly for the samples MgF, and NiF@MgF. This peak is not shifting towards higher temperature as the frequency was increased, which indicates not a thermally activated relaxation mechanism is occurred. In other words, the relaxation is thermally activated, when the frequency of the maximum of the relaxation peak is shifted towards a higher frequency with the temperature of the measurement increased. The behavior of $\tan\delta$ with temperature is similar to other samples reported by other researchers [36, 39]. Moreover, the second peak at 620K for NiF@MgF may be corroborated with ferroelectric-paraelectric phase change, i.e., the transition temperature of NiF@MgF.

2.2 AC Conductivity

To go further into dielectric results analysis, the ac conductivity vs. frequency plots for all samples at temperature range 290-650K are presented in Fig. 5. As shown in Fig. 5 it is easy to notice that all samples are frequency and temperature-dependent, and the conductivity value of the MgF and ZnF@MgF are lower than NiF@MgF. This behavior of the ac conductivity with the frequency can also be elucidated following Koop's phenomenological model which supposes that the material behaves as a multilayer capacitor. So, the conductivity is due to the grain boundaries and grains, at the low and high frequencies respectively. Also, according to the literature [33], the behavior of the conductivity with frequency can base on the polaron type of electrical conduction. From Fig. 5, it was observed that there are different values of the

conductivity between the low-frequency region and high-frequency region at all temperatures for all samples. In this context, it can say that the movement of charge carriers decreased by grain boundaries. So, the hopping between cations decreases, and hence the conductivity remains low at low frequencies. When the frequency increases to MHz, the grains become more dominant, so the number of hopping between cations will be increased, which leads to high conductivity values at high frequencies.

Figure 5 shows the relation between the conductivity and the frequency, which is linear relation, and obey the dynamical ac power law [40]:

$$\sigma(\omega) = A \omega^s \quad (14)$$

where $\omega = 2\pi f$ is the angular frequency, A is a constant and has units of σ , and (s) is dimensionless. The value of (s) can be determined from the slope of $\ln \sigma$ versus $\ln \omega$ plots at room temperature. The value of (s) lies between 0 and 1 as shown in Fig. 6. The conductivity mechanism for any material could be explained by the temperature-dependent behavior of (s) [41–42]. In other words, the relation between the temperature and the value of (s) gives a good knowledge about the charge transport mechanism in dielectric substances. There are three theoretical models (according to the frequency and the temperature dependence of the exponent (s)) to understand the electrical conduction mechanism in the materials;

- When the exponent (s) is temperature independent and frequency-dependent, the simple quantum mechanical tunneling (QMT) model can be proposed.
- There is another mechanism for polaron tunneling, named by the overlapping large polaron tunneling (OLPT) model [43], the exponent (s) is dependent on both frequency and temperature, and its value less than unity with an increase in temperature. Sometimes the large polarons, (s) continue to decrease with increasing temperature tending to its value predicted by the QMT model. In opposite, when the exponent (s) is temperature-dependent and increases with increasing temperature, the model will be the non-overlapping small polaron tunneling (NSPT) [44]. Sometimes the small polarons (SP), (s) exhibits a minimum at a certain temperature and subsequently increases with an increase in temperature in this case named by overlapping small polaron tunneling (OSPT).
- In the correlated barrier hopping (CBH) model [42], a temperature-dependence of exponent (s) and decreases with the increase in temperature.

Figure 6 shows the relation between the (s) value and the temperature. It is observed that there is a variation of this relation, indicating different conducting mechanisms dependent on the temperature and the frequency. Here, there are two different values of (s) for each sample, i.e., in the frequency range 100Hz -1MHz, the value (s_L) and in the high-frequency range 1-8MHz the other value (s_H) . For the samples MgF and ZnF@MgF, it clear that the exponent (s_L) has variation behavior with temperature, it is decreased in the low temperature and then increase and then decreased again, so the samples signified the coexistence of two SP and CBH models. But for the NiF@MgF the exponent (s_L) is slowly increased with temperature, so it tends to propose the SP model of conduction. But for the exponent (s_H) the behavior is different, i.e., the CBH model for the samples MgF at high temperature, but at low temperature obeys to the model QMT and the NiF@MgF the model SP is starting then QMT, whereas the ZnF@MgF tends to propose the QMT model of conduction. This relation between the (s) value and the temperature is similar to the relation of BaTiO₃/NiZnFe₂O₄ [45]. Besides, as the exponent value (s) of the NiF@MgF composites reaches 0.6 with increasing temperature, the hopping of the charges is becoming faster [33].

Figure 7 shows the curves of $\log(\sigma)$ versus $1000/T$ which could be divided into three regions, denoted as I, II, and III. As is seen in the first region (I), the σ_{ac} values increase up to a certain temperature denoted as T_s i.e., the σ_{ac} increases with increasing temperature which is semiconducting behavior, then start to (the second II) decrease up to another certain temperature T_m i.e., metallic behavior, and finally (the third III) increases with increasing temperature (semiconducting behavior) which may be the ferrielectric-paraelectric transition or the Curie temperature (T_c) of the all samples. The T_s and T_m temperatures correspond to the transition temperatures from semiconducting to metallic and metallic to semiconducting regions, respectively. The value of the T_s temperature is different for each sample MgF, ZnF@MgF, and NiF@MgF 343, 333, and 338 K respectively, while the value of T_m is the same for the pure Mg ferrite and the ZnF@MgF core-shell ($T_m = 433$ K), whereas the core-shell NiF@MgF has $T_m = 443$ K. Also, the Curie temperature T_c (as presented in Table 1) occurs for pure Mg ferrite, and the ZnF@MgF core-shell is the same temperature (503 K) but for NiF@MgF = 653 K.

The phenomenon of semiconducting and metallic conduction of all samples is similar to many cases of ferrites [33, 46]. The origin of this phenomenon is the d electrons of the transition metal ions, which are responsible for electrical and magnetic properties. There are electrostatic

interactions between anion and cation electrons that cause two splittings of $3d$ -level [47], the first is more stable triply degenerate t_{2g} level, and the other is less stable doubly degenerate e_g level. In other words, there are two interactions in ferrites, cation-anion-cation and cation-cation interactions [48-49]. Both interactions can be simultaneously presented, in the case of the cation-anion-cation interactions are dominant, so the materials have semiconductor or insulator behavior. Otherwise, if the transition metal has different valences, the ferrites may have a metallic type σ - T character below a transition temperature [49]. In the case of strong cation-cation interaction, these ferrites have a metallic behavior and may become semiconductors at low temperatures. The composite under investigation is content two nanoferrite materials with the spinel structure. Their conduction properties are arising due to the charge transfer between octahedral cations by the hopping of localized d-electrons. This hopping mechanism is restricted to the valence of cations that occupy the oxygen octahedral site.

Another explanation of the semiconducting and metallic conduction behavior, in the composite which made by two different materials that have different properties: one of these materials may be affected on the second by an enhancement or non-enhancement to yield new properties of the new compound [50]. Therefore, the σ - T curves can be explained based on the type interactions (enhancement or non-enhancement) between the NF and MF in NF@MF and between ZF and MF in ZF@MF. As shown in Fig. 7, the semiconducting-metallic behavior of MgF and NiF@MgF is stronger than ZnF@MgF. Also, the transition temperature of NiF@MgF nanocomposite is lower than that of NF ($T_c = 850$ K) [46] and higher than that of MgF ($T_c = 503$ K). As for ZnF@MgF, the situation is different where the transition temperature is the same as MgF = 503 K. This indicates that the effect of MgF on both NiF and ZnF is positive, because the net conduction of NiF@MgF will be increased and decreased the transition temperature, and the net conduction and the transition temperature of ZnF@MgF are increased concerning ZnF. This is in agreement with the magnetic behavior of NiF@MgF and ZnF@MgF core-shell [4].

As mentioned above, the relaxation is thermally activated when the frequency of the maximum of the relaxation peak is shifted towards a higher frequency with the temperature of the measurement increased. So the ac conductivity has a thermally activated process dependent on the temperature as shown in Fig. 7. According to Arrhenius relation:

$$\sigma = A \exp^{E_a/kT} \quad (15)$$

where A is constant, E_a is the activation energy, which is the energy needed to jump an electron from an ion to a neighboring ion, so giving rise to the electrical conductivity, k is Boltzmann's constant, and T is the absolute temperature. The activation energy can be obtained by plotting $\log \sigma$ versus $1000/T$, the slope of the linear parts will be equal to (E_a/k) . The activation energies for these processes are calculated and presented in Table 1. It is observed that the value of the activation energies is varying with temperature due to the varieties of the conduction mechanism.

Table 1: Electric and magnetic susceptibility parameters.

Sample	T_c (K)	E_a I (eV)	E_a II (eV)	E_a III (eV)	T_x (K)	θ (K)	C	μ_{eff} B.M.
MgF	503	1.3	0.9	0.13	350	349	0.57	2.1
NiF@MgF	503	0.1	0.13	0.12	426.5	424	9.2	8.56
ZnF@MgF	653	0.29	0.47	0.27	483	483	1.8	3.7

2.3 The magnetic susceptibility

The measurement of the magnetic susceptibility is an interesting method for determining phase boundaries in magnetic systems as there is a distinct variation in magnetic properties that occurred during a phase transition. The temperature dependence of the magnetization $M(T)$ for all samples is shown in Fig. 8. The temperature plots are obtained in a magnetic field of 0.8mT in the range of 300–500K. The data reveals the normal trend of ferrimagnetic materials; $M(T)$ decreased with the temperature reaching the paramagnetic region where the magnetocrystalline anisotropy ceased, and the magnetization drops at the well-known Curie point. In the paramagnetic region, the thermal energy increased the entropy and randomized the spin orientations where the forces overcome the magnetocrystalline energy. Also notice that, the magnetization of the core-shell ZnF@MgF higher than that of the pure MgF nanoferrite, which confirmed the previous work [4]. The increase in the magnetization for nanocomposites ZnF@MgF at the expense of MgF can be explained by two factors:

- The canting angle between the moments in the B-site may be decreased, which leads to an increase in the magnetization [51].

- The increasing of the value of the total magnetic moments in the octahedral site contributes to an increase in magnetization.

The two factors appear to interact with one another and cause magnetization to increase for ZnF@MgF at the expense of MgF. Therefore, the sample ZnF@MgF is a ferrimagnetic substance.

From the experimental data $M(T)$ as shown in Fig. 8, with increasing the temperature, a smoothly drop-in the magnetization occurred. The temperature at which magnetization drops is considered as the Curie temperature (at the Curie temperature(T_{Cm}), the material will be transfer from ferrimagnetic-paramagnetic). In other words, the smooth transition at the Curie temperature T_{Cm} can be used as an indicator of the degree of compositional homogeneity in all samples [52]. The Curie temperature (T_{Cm}) is an important magnetic property and is a compositional dependent parameter. The Curie temperature (T_{Cm}) was calculated from the plot of the first derivative of magnetization (dM/dT) versus temperature (T) as shown inset of Fig. 8 and listed in Table 1. The transition temperature T_{Cm} for MgF, ZnF@MgF, and NiF@MgF is 350K, 426.5K, and 483K respectively, which is lower than the NiF (865K), and MgF (648K) [53]. This may be due to the influence of the interaction between the core (MgF) and the shell (NiF or ZnF) on the magnetic transition temperature (T_{Cm}). In other words, the mutual effect between MgF and NiF or ZnF leads to a lower magnetic transition temperature. The same behavior has also been observed in many compounds of nanoparticles [52, 54-55]. Moreover, it was observed that the T_{Cm} and T_C values were different for all samples. From this work, it can be comprehended that due to the magnetic cations and electric charges for both the core and the shell, the magnetic and dielectric properties will be greatly varied. Similar variations of the T_{Cm} and T_C values were noticed in the literature [41, 56-57].

Figure 9 shows the relation between the reciprocal magnetic susceptibility (χ_M^{-1}) and the temperature (T) for all investigated samples. The relation obeyed the well-known Curie-Weiss law in the paramagnetic region. So the experimental effective magnetic moments μ_{eff} and the Curie Weiss constant (Θ) values can be calculated from the plot of χ_M^{-1} versus T using the relation:

$$\mu_{eff} = 2.82787\sqrt{C} \quad (16)$$

where C is the Curie constant equal to the slope of the line in the paramagnetic region and presented in Table 1. The variation of μ_{eff} with the substituted ions depends on its grain size. In general, can say that the decrease in grain size leads to minimizing the magnetization [58]. Also, the net magnetic moment is strongly dependent on the Fe^{3+} ions on the (B) sites. This means that the magnetic interaction between the MgF and the ZnF is an enhancement, where the net magnetic moment of ZF@MF will be increased. In other words, the mutual effect between MgF and ZnF leads to an increase in the net magnetic moment of ZF@MF.

The Curie Weiss constant (Θ) values equal to the cross of the slope with the x-axis in Fig. 9 and listed in Table 1. The Curie-Weiss constant (Θ) was found to possess large +ve values pointing to perfect ferrimagnetic character. The rise in Θ values with NiF@MgF assures the enhanced ferrimagnetic order of the core-shell from the increase in the ordered of the (MgF) at the expense of the (NiF). This indicates all samples have magnetic ferrimagnetic ordering in the temperature range 300-500K. Also, the values of the Curie Weiss constant (Θ) for all samples are closed to the values of the Curie temperature of the magnetization (T_{cm}).

Conclusion

The mutual effect between ZnF and MgF or NiF and MgF improved the physical properties of MgF, NiF, and ZnF. When studying the dielectric constant under the influence of temperature (300-650K) and frequency (100Hz - 8MHz), it was found that the mutual effect increases the space charge polarization, which contributes to the dielectric constant. Also from the previous work [4] and the present work, can conclude that the sample that possesses a large grain size, will be has a higher value of dielectric constant. Besides, the value of the loss of electrical energy ($\tan\delta$) decreases with the core-shell composites. And also, the conduction mechanism varies between SP, CBH, and QMT models according to the temperature and the frequency, so the conduction mechanism of all samples is semiconductor-metallic behavior. The experimental results of the ac electrical conductivity found that the transition temperature of a ferrielectric-paraelectric is lower than the transition temperature of the alone components of the core-shell. The mutual effect in the magnetic properties is more strong, where all samples have magnetic ordering in the temperature range 300-500K. The novelty in this work is an unexpected behavior of ZnF@MgF which possesses magnetization higher than the pure ferrite phase

(MgFe_2O_4), and Curie temperature (T_{Cm}) higher than the room temperature. Therefore, the sample ZnF@MgF is a ferrimagnetic substance.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability Statements

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Figure captions

Figure 1: Frequency dependence of ϵ at different temperatures for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 2: Temperature dependence of ϵ at different frequencies for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 3: Frequency dependence of $\tan\delta$ at different temperatures for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 4: Temperature dependence of $\tan\delta$ at different frequencies for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 5: Frequency dependence of $\text{Log}(\sigma)$ at different temperatures for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 6: the exponent (s) versus T (K).

Figure 7: The Conductivity $\log(\sigma)$ versus $1000/T$ for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 8: Temperature dependence of the magnetization for a) MgF, b) ZnF@MgF and c) NiF@MgF.

Figure 9: Temperature dependence of the χ_m^{-1} for a) MgF, b) ZnF@MgF and c) NiF@MgF.

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Figures

Figure 1

Frequency dependence of ϵ'' at different temperatures for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 2

Temperature dependence of ϵ at different frequencies for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 3

Frequency dependence of $\tan \delta$ at different temperatures for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 4

Temperature dependence of $\tan \delta$ at different frequencies for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 5

Frequency dependence of $\text{Log}(\sigma)$ at different temperatures for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 6

the exponent (s) versus T (K).

Figure 7

The Conductivity $\log(\sigma)$ versus $1000/T$ for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 8

Temperature dependence of the magnetization for a) MgF , b) ZnF@MgF and c) NiF@MgF .

Figure 9

Temperature dependence of the χ_m^{-1} for a) MgF , b) ZnF@MgF and c) NiF@MgF .