Magnetic and Dielectric Properties of Epoxy Composites Reinforced with Hybrid Nanoparticle iron oxide (Fe$_3$O$_4$) and nickel (Ni)

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Abstract. In this research, the effects of hybrid nanoparticles Fe$_3$O$_4$+Ni on the magnetic and dielectric properties of epoxy resin are investigated. Microstructural characterization was performed by Field Emission scanning electron microscopy FESEM, X-ray diffraction spectra XRD, and Fourier-transform infrared spectroscopy (FTIR). The magnetic properties were investigated by vibrating sample magnetometer (VSM) and the dielectric response was investigated by a precision impedance analyzer (Agilent 4294A) LCR meter at room temperature with different frequencies. The study dealing with hybrid nanocomposite (epoxy/Fe$_3$O$_4$+Ni) consisting of epoxy resin as the matrix material reinforcing by magnetite nanoparticles (Fe$_3$O$_4$) with different weight percentages (3wt.%, 6wt.%, 9wt.%, 12wt.%, 15wt. %) and constant weight percentage 2wt% of nickel (Ni) nanoparticles. The samples were prepared using the casting method. The epoxy with the hardener is weighted and mixing in a 2:1 ratio and then add reinforcement materials Fe$_3$O$_4$+Ni into the epoxy. Microstructural analysis showed that a uniform distribution and homogeneously dispersed in the epoxy matrix. The results of this work exhibit that the Increasing additive weight percentages of Fe$_3$O$_4$ nanoparticles with a constant weight of Ni nanoparticles into epoxy resin led to improvement in the magnetic and electric properties of hybrid nanocomposites compared with pure epoxy.

Keywords: hybrid-nanocomposites, Epoxy matrix, Microstructure, Magnetic properties.

1. Introduction
Polymer matrix composites (PMC.s) were first known in 1960 as a new type of materials results to obtain a new composite with improvement in mechanical, electrical, and thermal properties comparing with the properties of polymers. Recently, nanocomposites are interesting materials with a promise multifunctional material with a minimum nanomaterial concentration. Owing to the reinforcement materials in nanometer size made it too dispersed homogeneously in the matrix and then improving
properties of the synthesized nanocomposites. Polymer matrix nanocomposites (PMNCs) are an interest large concern of national research labs and manufacturing owing to their outstanding properties [1]. The advancement in nanocomposite discipline depends extensively on the manufacture of nanomaterials as nanoparticles and nanofibers with different shapes and sizes [2]. Polymer nanocomposites are an important type of hybrid material where the inorganic materials in nanoscale, such as particles, fibers, sheets, or tubes are distributed in a polymer matrix [3]. Between various polymers, epoxy is one of the most important engineered polymers and the most normally used thermosetting epoxide polymer matrix that contains epoxide groups [4]. epoxy can be improved for the incorporation of nanoparticle material as reinforcement, such as metallic oxides [5]. Epoxy matrix has outstanding properties and advantages, such as high dielectric, easy process and shaping, chemical and thermal stability, large strength, mechanical properties like increasing strength, large modulus of elasticity, barrier characteristic and flame retardant, low shrinking, low creep, and great stiffness. It is also widely used in research and industry areas and has a wide range of application fields such as coating materials, adhesives, anti-corrosion, automotive, tissue substitutes, electronics, aerospace, and other industrial fields [5,6]. Generally, epoxy resin has many epoxide groups in a molecule of the structure. The epoxy resin required treatment with a curing hardener or agent to obtain three insoluble and infusible networks [7]. The choice of epoxy resins and their hardener depends on their physical, electrical, mechanical, chemical, and the needed properties of the products and the atmosphere that are used [8]. The many types of research in this day study conductivity, the dielectric, and magnetic response of polymer matrix-based nanocomposites [9]. The goal of this research is to study the effect of (Fe₃O₄, Ni) nanoparticles on the magnetic and dielectric properties of epoxy matrix Nanocomposites manufactured by a casting technique.

2. Experimental
2.1. Materials
The low viscosity epoxy resin type (Sikadur 52 A) with hardener (Sikadur 52 B) was used as a matrix material were obtained from Sika company placed in Turkey. The reinforcement materials are magnetite (Fe₃O₄) nanoparticles with a particle size of about 30 nm, specific surface area (SSA) 40-60 m²/g, and purity ratio of (Fe₃O₄) is 99.5% provided by (USA) Research Nanomaterial and the other reinforcement material is nickel (Ni) nanoparticles with a particle size (40 nm), specific surface area (SSA) 40-60 m²/g, and purity ratio of (Ni) is 99.9 % obtained from (Hongwu International Group Ltd – China).

2.2. Synthesis of the samples
Many methods can be used to incorporate the nanoparticles into the polymer. However, this work used casting methods to prepare a hybrid nanocomposites (Epoxy/Fe₃O₄+Ni) sample. Firstly, the epoxy resin mixing with a hardener in a 2:1 w/w ratio and was stirred for 5 minutes and then adding different weight percentages of the magnetite nanoparticles (Fe₃O₄) (0wt. %, 3wt. %, 6wt. %, 9wt. %, 12wt. %, 15wt. %) and constant weight (2wt. %) of (Ni) nanoparticles, the mixture was stirred by hand at a slower rate in ultrasonication bath at 50 °C for 15 minutes to get better a homogeneous dispersion of the nanoparticles in an epoxy resin and after that, it is poured into silicone molds to be cured for seven days at room temperature. After polymerization, the samples of the Post-curing took place at 100 °C for 2 hours, and then the samples are cut into suitable sizes according to the required tests [10-12]. Table (1) shows the weight percentages of epoxy resin and nanomaterials using to manufacture the samples and Figure 1. shows the procedure was used to prepare the samples.
Table 1. The weight percentages of epoxy resin and nanomaterials in making specimens.

| No. of Samples | Epoxy + hardener wt.% | Magnetite (Fe₃O₄) wt.% | Nickel (Ni) wt.% |
|----------------|------------------------|-------------------------|-----------------|
| 1 (pure epoxy) | 100                    | 0                       | 0               |
| 2              | 95                     | 3                       | 2               |
| 3              | 92                     | 6                       | 2               |
| 4              | 89                     | 9                       | 2               |
| 5              | 86                     | 12                      | 2               |
| 6              | 83                     | 15                      | 2               |

Figure 1. Shows the Experimental Procedure to prepare the samples.

2.3. Microstructure Examinations

Afterward preparing the samples the microstructure characterizations for all samples of hybrid nanocomposites were examined by using Field Emission Scanning Electron Microscopy (FESEM). The purpose of this examination aimed at finding out the effect of nanoparticles on the microstructure and morphology of the hybrid nanocomposites and to show the agglomeration and diffusion of the nanomaterials in the epoxy. Also, the X-Ray Diffraction analysis was carried out, the objective of this test to find the peaks before and after adding nanomaterials. FTIR examined the chemical structure of the nanocomposite and the bonding nature of nanoparticles within the polymer.
2.4. Magnetic Properties

The magnetic properties measurements of the epoxy/Fe$_3$O$_4$ + Ni Nanocomposites were investigated by vibrating sample magnetometer measurement instrument (VSM) at room temperature. The magnetic properties involve studying the magnetization curve (hysteresis loops) of the produced hybrid nanocomposites using the (VSM). For each sample, saturation magnetization ($M_s$), residual magnetization ($M_r$), and coercivity ($H_c$) values were obtained from the curves of magnetic hysteresis loops. The saturation magnetization ($M_s$) describes the response of the sample to an applied external magnetic field and the coercivity ($H_c$) describes the force that is required when subject to an externally applied magnetic field opposite to the original external applied magnetic field. The ($H_c$) used to completely demagnetize the sample and return applied external magnetic field magnetization of sample to zero, but the residual magnetization ($M_r$) will not be decreased to zero [9, 13].

2.5. Dielectric Properties

The dielectric properties of the hybrid nanocomposites were studied by a precision impedance analyzer (Agilent 4294A) LCR Meter with a frequency range between 50 Hz and 5 MHz. The dielectric properties such as the dielectric constant, imaginary part of dielectric constant, dielectric loss factor ($\tan \delta$), and AC electrical conductivity ($\sigma_{ac}$) was calculated by using a precision impedance analyzer (Agilent 4294A) as shown in Figure 3. The AC electrical conductivity can be calculated by using the following formula [14-16]:

$$\sigma_{ac} = 2 \pi f \tan \delta \varepsilon_0 \varepsilon'$$  \hspace{1cm} (1)

Where $\varepsilon_0$ is the vacuum the permittivity of free space = 8.85 $\times$ 10$^{-12}$ F/m, $\varepsilon'$ is the real part of the dielectric constant and ($\tan \delta$) loss factor can be gained directly from the measurements.

3. Results and Discussion

3.1. Microstructural characterization

The morphologies of the prepared samples were examined by (FESEM). FESEM images demonstrate that all hybrid nanocomposites have been successfully manufactured. Through the results of FESEM images, the morphological behavior of the manufactured samples shows that a smooth fracture surface of pure epoxy, while (FESEM) images of (epoxy/Fe$_3$O$_4$ + Ni) nanocomposites revealing that dispersion of nanomaterials in epoxy resin but with an agglomeration of nanomaterial when the additive at high weight percentage. Figure 2. shows the microstructure of pure epoxy matrix and hybrid nanocomposites (epoxy/Fe$_3$O$_4$+Ni) with different weight percentages. FESEM image analyzes are consistent with [17,18].
Figure 2. FESEM images of the examined specimens for epoxy resin with different weight of Fe₃O₄ and constant weight of Ni: (a) pure epoxy, (b) 3wt.% Fe₃O₄ + 2wt.%Ni, (c) 6wt.% Fe₃O₄ + 2wt.%Ni, (d) 9wt.% Fe₃O₄ + 2wt.%Ni, (e) 12wt.% Fe₃O₄ + 2wt.%Ni and (f) 15wt.% Fe₃O₄ + 2wt.%Ni.
3.2. X-Ray Diffraction (XRD) analysis

XRD patterns of (epoxy/Fe₃O₄ + Ni) hybrid nanocomposites are shown in Figure 3. The result of X-ray diffraction obtained by X-Ray diffractometer lab model (XRD-6000) SHIMADZU Europe with CuKα radiation at a wavelength (λ) =1.5405 Å for all the samples of (epoxy/Fe₃O₄ + Ni) hybrid nanocomposites. The main peaks characteristic is the peaks of Fe₃O₄ at room temperature occurred at (2θ) ranging about 30.077°, 35.427°, 37.058°, 43.055°, 53.412°, 56.937°, 62.522°, 70.928° and 74.964° corresponding to (220), (311), (222), (400), (422), (511), (440), (620) and (622) respectively with peak for epoxy matrix at (2θ) equal to 20°. While the peaks of the Ni phase appeared at (2θ) about 44.508°, 51.847° and 76.372° corresponding to (111), (200) and (220). The planes of epoxy/Fe₃O₄+Ni hybrid nanocomposites were defined according to reference code 01-079-419 for the Fe₃O₄ phase and reference code 00-004-0850 for the Ni phase. The XRD analysis exhibited that a uniform dispersion of Fe₃O₄ and Ni nanoparticles in epoxy resin for each sample. These results agree with [17,19].

Figure 3. XRD patterns of (Epoxy/Fe₃O₄ +Ni) hybrid nanocomposites.

Table 2. shows the Peak list for the hybrid nanocomposites.

| No. | 2Theta[deg] | d–spacing [Å] | planes | intensity % |
|-----|-------------|---------------|--------|-------------|
| 1   | 30.077°     | 2.968         | 220    | 29.5        |
| 2   | 35.427°     | 2.531         | 311    | 100         |
| 3   | 37.058°     | 2.423         | 222    | 7.8         |
| 4   | 43.055°     | 2.099         | 400    | 20.1        |
| 5   | 44.508°     | 2.034         | 111    | 100         |
| 6   | 51.847°     | 1.762         | 200    | 42          |
| 7   | 53.412°,    | 1.714         | 422    | 8.5         |
| 8   | 56.937°     | 1.615         | 511    | 27.8        |
| 9   | 62.522°,    | 1.484         | 440    | 36.5        |
| 10  | 70.928°     | 1.327         | 620    | 2.8         |
| 11  | 74.964°     | 1.265         | 622    | 2.9         |
| 12  | 76.372°     | 1.246         | 220    | 21          |
3.3. *Fourier transforms spectroscopy (FTIR)*

FTIR spectrum is a very useful technique to deduce structural investigation of chemical compounds. Figure 4 exposed the FTIR spectra of (epoxy/Fe3O4+Ni) hybrid nanocomposites. The FTIR transmittance spectrum was observed in the range of 400–4000 cm\(^{-1}\). The bands of the absorption peaks at a wavelength of about (450–480 cm\(^{-1}\)) and (500–590 cm\(^{-1}\)) are related to the stretching vibration modes of Fe–O functional group bonds. IR bands are observed in the range of (700–4000 cm\(^{-1}\)), which are originated from the vibrations of the following chemical bonds/groups [22], the IR absorptions at (1029.99–1035 cm\(^{-1}\)) and (1234.44–1292.31 cm\(^{-1}\)) Correspond to asymmetrical aromatic C–O stretching vibration, (1111–1180.44 cm\(^{-1}\)) Corresponds to asymmetrical aliphatic C–O stretching. The C–C stretching, vibration at (1450–1850 cm\(^{-1}\)), and the C–H stretching bands were observed at around (2850.79–2920.23 cm\(^{-1}\)). The aliphatic and aromatic C–H bond stretching at (2330.01–2366.87 cm\(^{-1}\)). Finally, the peaks at around (3738.04–3741.90 cm\(^{-1}\)) are assigned to the stretching vibration of the hydroxyl group (OH). The result agrees with [20,21].
3.4. Magnetic Characterization

Magnetic hysteresis loops (M-H) for the pure epoxy and hybrid nanocomposites (epoxy/Fe\textsubscript{3}O\textsubscript{4} + Ni) at room temperature are shown in Figure 5. The magnetic behavior of each specimen is characterized by saturation magnetization (Ms), residual magnetization (Mr), and coercivity field (Hc) values were taken from hysteresis loops curves. The results show that increasing the magnetite (Fe\textsubscript{3}O\textsubscript{4}) nanoparticle content, led to improving the values of saturation magnetization, residual magnetization, and coercivity field in the hybrid nanocomposites. Magnetite nanoparticles (Fe\textsubscript{3}O\textsubscript{4}) induce magnetic properties to the epoxy matrix. The maximum values of (Ms) and (Mr) have increased from zero in pure epoxy to 17.2 emu/g and 1.81 emu/g, respectively at (15wt. % Fe\textsubscript{3}O\textsubscript{4} + 2wt. % Ni) incorporated in an epoxy matrix. While the maximum value of coercive field (Hc) is 99.09 Oe. Table 2. listed the magnetic properties of the hybrid nanocomposites. The result agrees with [22-24].

Figure 4. FTIR spectra of (Epoxy/Fe\textsubscript{3}O\textsubscript{4} + Ni) nanocomposites, (a) 3wt.% Fe\textsubscript{3}O\textsubscript{4} + 2wt.%Ni, (b) 6wt.% Fe\textsubscript{3}O\textsubscript{4} + 2wt.%Ni, (c) 9 wt.% Fe\textsubscript{3}O\textsubscript{4} + 2wt.%Ni, (d) 12 wt.% Fe\textsubscript{3}O\textsubscript{4} + 2wt.%Ni, (e) 15 wt.% Fe\textsubscript{3}O\textsubscript{4} + 2wt.%Ni.
Figure 5. Magnetic hysteresis loops for (epoxy/Fe$_3$O$_4$+Ni) nanocomposites.

Table 3. Magnetic properties of (epoxy/Fe$_3$O$_4$+Ni) samples carried out at room temperature.

| Sample no. | Fe$_3$O$_4$ wt.% | Ni wt.% | Saturation Magnetization $M_s$ (emu/g) | Residual Magnetization $M_r$ (emu/g) | Coercivity Field $H_c$ (Oe) | Remanence Ratio $M_r/M_s$ |
|------------|------------------|---------|--------------------------------------|-------------------------------------|--------------------------|--------------------------|
| 1          | 0                | 0       | 0                                    | 0                                   | 0                        | 0                        |
| 2          | 3                | 2       | 4.22                                 | 0.40                                | 94.18                    | 0.095                    |
| 3          | 6                | 2       | 7.69                                 | 0.79                                | 95.3                     | 0.099                    |
| 4          | 9                | 2       | 9.72                                 | 1.02                                | 98.05                    | 0.105                    |
| 5          | 12               | 2       | 11.42                                | 1.23                                | 98.7                     | 0.108                    |
| 6          | 12               | 2       | 17.19                                | 1.81                                | 99.09                    | 0.104                    |

3.5. Dielectric properties.
The epoxy matrix can be used in the electronic device owing to its outstanding dielectric properties with a low and stable dielectric constant depending on a wide range of frequencies [13]. So the effect of magnetite (Fe$_3$O$_4$) and nickel (Ni) nanoparticles on the dielectric properties of the epoxy matrix were studied here. The real part of the dielectric Permittivity ($\varepsilon'$) as a function of the frequency (50 to 5 MHz), at room temperature for the pure epoxy and hybrid nanocomposites, demonstrated in Figure (6). The relative permittivity of epoxy hybrid nanocomposites decreases with the increasing frequency and then keeps nearly constant in the high-frequency range (above $2 \times 10^6$ Hz). The decrease in relative permittivity $\varepsilon'$ at higher frequencies is due to the dropping out of interfacial polarization by the interaction between the nanoparticles and epoxy matrix would cause the improvement of real permittivity [9,13]. The relative permittivity $\varepsilon'$ increases with increasing the adding weight ratio of nanoparticles.
The AC conductivity behavior shows that the incorporation of nanoparticles into the epoxy matrix led to an increase in conductivity. Also, it is found that the (AC) electrical conductivity for (epoxy/Fe$_3$O$_4$+Ni) hybrid nanocomposites increases with an increase in frequency. This behavior of AC conductivity (σac) increase with an increase in frequency is a common response for semi-conductor, polymer, and dielectric material. The increase in conductivity can be owing to the formidable increase in the mobility of charge carriers in the nanocomposites system that occurs by applying the electrical field and the increase in filler concentration decreases the mutual distance of the average interparticle leading to the probability of continuous conducting path formation. The content of the conductive phase (Fe$_3$O$_4$) is the main responsible for all conductivity of the epoxy nanocomposites. The result agrees with [22,25].

**Figure 6.** Shows the real part of dielectric permittivity as a function of frequency, for pure epoxy and hybrid nanocomposite.
**Figure 7.** Shows the AC conductivity as a function of frequency, for pure epoxy and hybrid nanocomposites.

### 4. Conclusions

In this work, a series of hybrid nanocomposites systems consisting of epoxy resin and different types of nanoparticles (Fe$_3$O$_4$, Ni) were successfully synthesized using the casting and sonication methods. It was found from FESEM, XRD, and FTIR results exhibit that uniform dispersion or distribution of nanoparticles in an epoxy matrix with an aggregation of nanomaterial when the additive at high weight percentages, also the results show that the interfacial adhesion between the hybrid nanoparticles (Fe$_3$O$_4$+Ni) with an epoxy matrix is strong. From the experimental data, it appears that the embedding of the nanoparticles improves significantly the dielectric and magnetic properties. Dielectric permittivity or dielectric constant ($\varepsilon'$) in the frequency range of 50 Hz to 50 MHz increasing with an increase in Fe$_3$O$_4$+Ni content. Dielectric constant ($\varepsilon'$) values decrease rapidly with an increase in frequency. Maximum AC conductivity ($\sigma_{ac}$) of ($10^{-4}$ S/m) was obtained at a frequency of 50 MHz for 15wt.% (Epoxy/Fe$_3$O$_4$+Ni) hybrid nanocomposite. Generally, the dielectric constant and AC conductivity increases with the increase of the nanomaterial added to the epoxy matrix. The magnetic measurements show that improving the values of magnetic properties including saturation magnetization (Ms), residual magnetization (Mr), and Coercivity field (Hc) of the hybrid nanocomposite. The induced magnetic properties increase with the incorporation of magnetite (Fe$_3$O$_4$) nanoparticles. The maximum values of (Ms), (Mr), and (Hc) have increased from zero in pure epoxy to (17.2 emu/g), (1.81 emu/g), and (99.09 Oe) for 15wt. % (Epoxy/Fe$_3$O$_4$ + Ni) hybrid nanocomposite.
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