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Complex permittivity and permeability properties analysis of NiCuZn Ferrite-Polymer nanocomposites for EMI suppressor applications

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Abstract. The complex permittivity and permeability studies of NiCuZn ferrite-paraformaldehyde (NCZ-PFD) nanocomposites for electromagnetic interference (EMI) suppressor applications are presented. The NCZ Ferrite and nanocomposites were prepared via microwave hydrothermal and ball-milling, respectively. The nanocomposites were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM) the results indicate that the NCZ-PFD nanocomposites were successfully prepared without impurities. The complex permittivity and permeability were measured over frequency range of 8.2-12.4 GHz and 12.4–18 GHz. The results show that for nanocomposites, the values of the real (ϵ') and imaginary permittivity (ϵ'') and imaginary permeability (μ'') increase, while the value of real permeability (μ') decreases as the polymer content increases. Dielectric relaxations were studied using cole-cole plots of complex permittivity. Magnetic relaxation dispersions were analyzed using cole-cole plots of complex permeability. The possibility to modulate the electromagnetic properties of the composite materials is of a great interest to fabricate microwave absorbing and electromagnetic shielding materials with high performances.

1. Introduction

These The capability of a material to shield electromagnetic energy, whether or not its undesirable energy entering a system or escaping a system and it's far called as its shielding effectiveness. It is along with losses due to absorption, reflection, and multiple reflections [1]. The primary mechanism of EMI shielding is generally reflection and for this kind of the radiation by using the shield, it needs to have mobile charge carriers (electrons or holes) which interact with the electromagnetic fields in the radiation. For this reason, the shield tends to be electrically conducting, without an excessive conductivity. As an instance, a volume resistivity in the order of $1 \Omega\text{cm}$ is commonly used. A secondary mechanism of EMI shielding is generally absorption. For significant absorption of the radiation, the shield must have electric and/or magnetic dipoles which interact with the electromagnetic fields in the radiation.

Metals, along with ferroelectrics and ferrites are typically utilized in EMI suppression and those materials have many risks in terms of their weight, corrosion and physical rigidity [2]. Many scholars have done investigations of the absorption properties of one-component electromagnetic wave absorbers using ferroelectrics, ferrites, conducting polymers and other new absorption materials [3, 4]. However, in comparison with multicomponent materials, the application possibilities of one-component materials are confined due to their poor absorption efficiencies and relatively narrow bandwidth. These properties could be overcome with the development of latest composite materials. Out of all composite materials,



magnetic nanoparticles doped with conducting and insulating polymers are lightweight, flexible options to the micron sized metal components. For this reason, ferrite/polymer nanocomposites are the great selections. Every other significance of magnetic-polymer nanocomposites is that important parameters along with loss tangents and impedance matching, which are important in microwave devices, may be controlled in those materials.

The loss tangent is a measure of the inefficiency of a magnetic system. The loss tangents are primarily determined by magnetic and eddy current losses and which depend upon the resistivity of the material. The resistivity of magnetic nanoparticles will increase with an addition of conducting polymer, thereby decreasing the eddy current losses. Typically, magnetic losses in composite substances are controlled by the material grain structure, domain wall resonances and so forth, these parameters may be manipulated efficiently in nanocomposite substances by way of the size distribution of nanoparticles and their dispersion inside the matrix. EMI suppression over a wide band frequency range requires tunability of the impedance (Z), which relies upon on the tunability of the complex permeability and complex permittivity. The conductivity performs an essential role in a material's capacity to shield electromagnetic energy. therefore, having both conducting and magnetic components in a single system might be used as an EMI shielding material [5].

In the present study, various Paraformaldehyde (PFD) loaded $\text{Ni}_{0.48}\text{Cu}_{0.12}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$ (NCZ) matrix nanocomposites had been prepared using a mechanical milling approach. The NCZ may be used in microwave devices and microwave absorption fields because of their high saturation magnetization, great chemical stability and corrosion resistance. The nanocomposite powders with the effect of the volume fraction of polymer PFD on the frequency dispersion characteristics of the complex permittivity (ϵ' & ϵ'') and permeability (μ' & μ'') were studied and the acquired results had been discussed.

2. Experimental procedure

As synthesized nanopowder of NCZ the use of microwave hydrothermal (M- H) approach and bought PFD had been combined at different weight percentage to obtain the nanocomposites of $(1-x)\text{NCZ} + x\text{PFD}$, ($x=0, 0.1, 0.3, 0.5, 0.7, 0.9$ and 1) the samples have been named as NCZ, NF1, NF2, NF3, NF4, NF5, and PFD respectively. The mixed powders were mechanical milled the use of a Retsch Co high strength planetary ball mill through using the hardened tungsten carbide (WC) vial collectively with ten 12 mm WC balls for 20 hr. A ball to powder mass charge ratio of 14:1 was selected. The speed of the mill turned into set at 300 rpm with an interval of 40 mins. The 20-hrs milled powders were subjected to anneal at 1100C/30 min in an air atmosphere. The prepared samples have been characterized through X-ray diffraction (XRD) and scanning electron microscopy (SEM).

The samples of NCZ-PFD nanocomposites were formed to fit precisely into rectangular X-band waveguide (WR90, $h=10.16$ mm, $w=22.86$ mm and $t=2.1$ mm) and Ku-band waveguide (WR62, $h=7.8994$ mm $w=15.7988$ mm and $t=2.1$ mm) for measurements. The complex scattering parameters that correspond to the reflection (S_{11} or S_{22}) and transmission (S_{21} or S_{12}) within the composite samples were measured the use of an Agilent 8722ES vector network analyzer. Full -port calibrations have been initially carried out on the test setup so that you can eliminate errors because of the directivity, source match, load match, isolation, and so forth., in both the ahead and opposite path. The complex permittivity (ϵ' & ϵ'') and permeability (μ' & μ'') had been then determined from the measured scattering parameters using Agilent software module 85071, primarily based on the process given in HP product note.

3. Result and discussions

XRD spectra of NCZ, PFD, and NCZ-PFD nanocomposites were done at room temperature within the range from $2\theta = 200$ -800 are given in Figure 1. All samples did not show other peaks for impurities. The X-ray diffraction patterns of the nanocomposites contain phases, that are the crystalline and amorphous phases. It may be seen that the crystallinity of the nanocomposites expanded with increasing of NCZ. This becomes because of the added crystalline NCZ phase that migrates into the amorphous phase of PFD Polymer, for that reason, decreasing the amorphous domains of the sample. The crystallite size was estimated using Debye-Scherrer formula $D_m = K\lambda/\beta\cos\theta$ where K is a constant, β is the full width half maxima, λ is the wavelength of x-rays used and θ is the diffraction angle and provided in Table 1. It is able to be found from the table that the crystallite size of the samples is in nano range and decreases with an

increase of polymer content. It could be because of the NCZ particles are bonded to polymer chains and additionally because of the pressure triggered through the ferrite particles. The lattice parameters of composites had been calculated with the assist of (3 1 1) peak using equation $d_{hkl} = \sqrt{h^2 + k^2 + l^2}$ and are presented in table 1. The diffractogram additionally suggests that the structure of NCZ inside the nanocomposites is maintained. The bulk density of the composites is measured through Archimedic's principle the bulk density of the nanocomposites reduced with polymer content material because the molecular weight of ferrite is more than that of the polymer (Table 1).

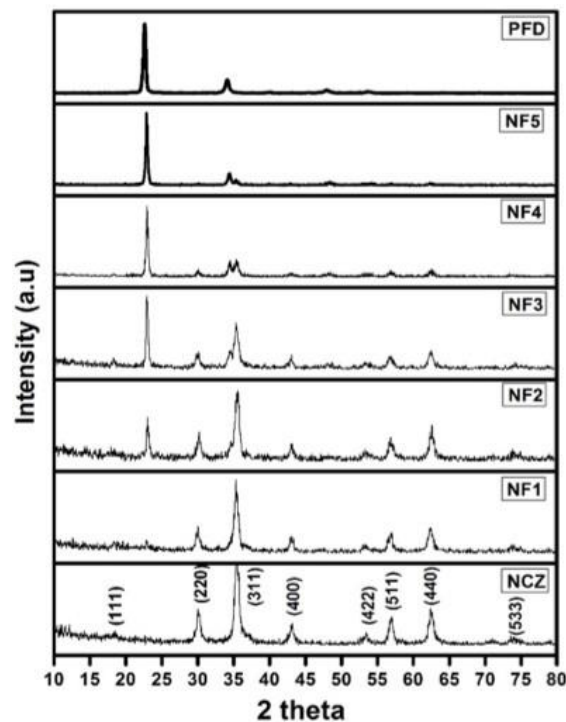


Figure 1: XRD patterns of NCZ-PFD nanocomposites

Figure 2 gives SEM photographs for the NCZ-PFD nanocomposites. It may be visible from the SEM micrographs that the connectivity of NCZ grains is discontinued due to the presence of polymer, which results in the variant in the dielectric and magnetic properties of the composites. As ferrite content became increased the magnetic grains are agglomerated because of the strong interaction among the ferrite particles and this led to the uneven distribution of PFD inside the NCZ matrix.

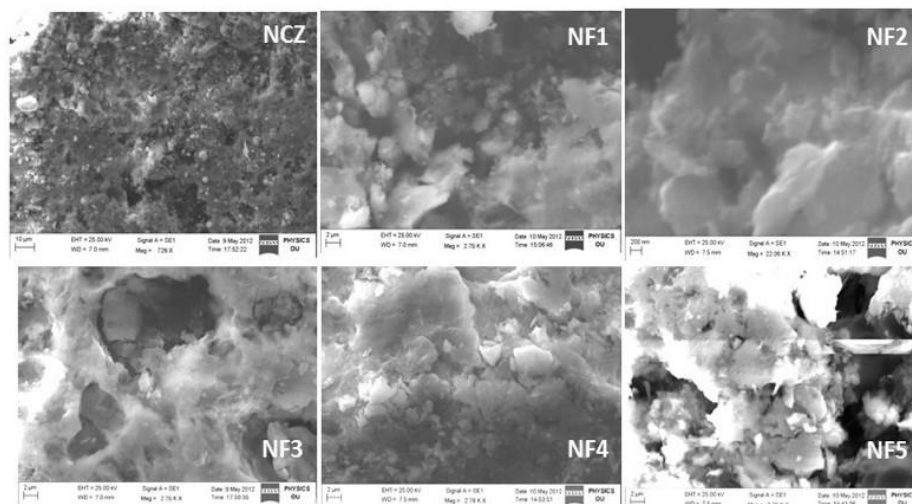


Figure 2: SEM images of NCZ-PFD nanocomposites

The variations within the complex permittivity (ϵ' & ϵ'') for NCZ-PFD nanocomposites over the frequency range of 8.2-18 GHz are shown in Figure 3. It was discovered that the real part (ϵ') of permittivity is increased with polymer up to NF3 after which reduced and the imaginary part (ϵ'') of permittivity of the nanocomposite increased with the increasing weight percentage of the polymer. When the load fraction of PFD increased, greater intrinsic polarization and interfacial polarization were generated, so the real a part of permittivity improved. The metal ions and defects at the ferrite and the interface between ferrite and polymer result in interfacial polarization. Further, greater PFD generates more interfacial and intrinsic electric dipole polarization, growing the dielectric constant and the dielectric loss [6]. The decrease in dielectric constant for the samples NF4 and NF5 can be due to much less contribution of interfacial polarization of ferrite particles. The acquired values are tabulated in Table 2.

The Debye dipolar relaxation may be utilized to recognize absorbing mechanisms of the dielectric loss materials. a couple of superimposed Cole–Cole semicircles (Figure4) are observed for the NCZ-PFD nanocomposite samples, which might also advise that there are more than one dielectric relaxation processes. The ensuing multiple relaxations can end result from the interface polarization, which is due to the heterogeneous interfaces of magnetic nanoparticles and PFD. Interfacial polarization happens whenever there's a build-up of a charge at a boundary among regions or materials [7]. Consequently, the interfacial polarization and related relaxation should contribute to the improved dielectric loss and microwave absorption performance.

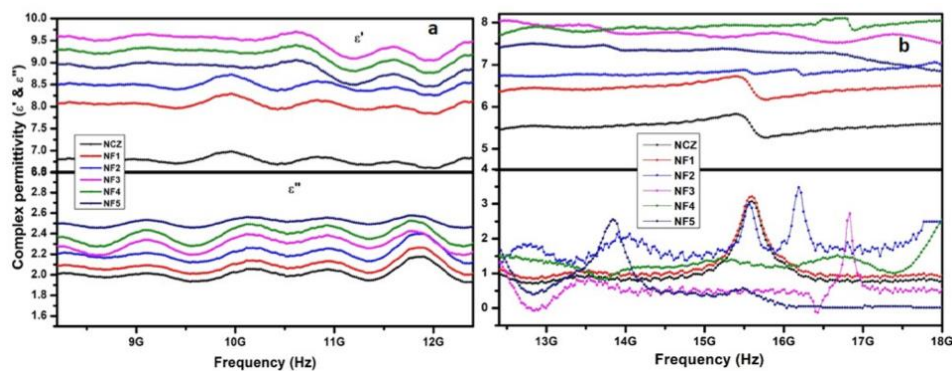


Figure 3: Complex permittivity of x NCZ+ (1-x) PFD nanocomposites a) in X-band b) in Ku-Band region.

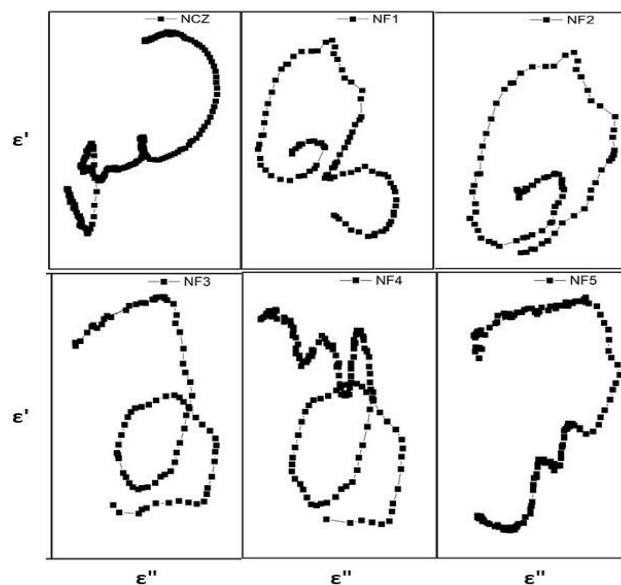


Figure 4: Typical Cole–Cole semicircles (ϵ' vs ϵ'') for NCZ-PFD nanocomposites in the frequency range of 8.2–18 GHz.

The real (μ') and imaginary (μ'') parts of the complex permeability of (NCZ- PFD) nanocomposites samples measured within the frequency range from 8.2 to 18 GHz are shown in Figure 5. The real part (μ') of the permeability for the sample NCZ decreases with increase in frequency, while the imaginary part (μ'') reveals resonance peaks at different frequencies. The real part of the permeability μ' comes from the cooperative effect of the domain wall resonance and spin rotational resonance of ferrite nanoparticles. It was observed that the μ' value of the composites reduced with the increasing weight percent of the polymer. The decreased permeability of magnetic materials is often found once they were coated or embedded through nonmagnetic material, which can be attributed to the reduction of saturation magnetization after mixture with nonmagnetic substances [8].

The magnetic loss (μ'') is a result of eddy current consequences, natural resonances, and anisotropy energy present inside the composites [9]. In the microwave ranges, the presence of nanoferrite particles inside the composite is the principal purpose of eddy current. The natural resonances within the X-band can be attributed to the small size of ferrite in the composites. Anisotropy energy of the small size materials, [10] specifically inside the nanoscale, would be higher due to the surface anisotropic field because of the small size effect [11]. Better anisotropy energy also contributes to the enhancement of microwave absorption. For μ'' there is considerable resonance peak at around 10 GHz for the nanocomposite samples. Based on the Aharoni's concept, [12] the resonance peaks at high frequency within the curve of μ'' are associated with the exchange resonance, and the peaks located at lower frequency are related to the natural ferromagnetic resonance [13]. With polymer content, the magnetic loss μ'' improved it could be due to the magnetic loss inside the composites and pure ferrite are due to the sum of domain wall resonance and natural resonance at high frequencies. From the graph, it is able to additionally be seen that the resonance peaks of both μ' and μ'' of complex permeability are shifted to a higher frequency with increasing addition of polymer. Even though the sample with 30 and 10 wt% ferrite loading has no peak at the frequency range of 12–18 GHz, it can be forecasted that its peak will seem at a frequency higher than 18 GHz in step with the trend found from the graphs. The resonance peak seems at 14.5–17 GHz with composites can be ascribed to spin rotational resonance, that is sensitive to composition and structure [14, 15]. The received complex permeability values of nanocomposites were tabulated in Table 2.

Figure 6 is the Cole-Cole plots for the curves of complex permeability of NCZ-PFD nanocomposites. The Cole-Cole plots of μ' versus μ'' show some semi-circle formed curves, this means that the relaxation dispersion. But there are a few variations within the shape of curves comparing to the normalized curves of complex permeability. The spinel ferrites in applications display relaxation phenomena as increasing the frequency because of the damping within the magnetization process [16]. This gives some other technique of demonstration to identify the relaxation phenomena for various ferrites.

The magnetic loss of NCZ-PFD nanocomposites, originating from the magnetic properties of the NCZ particles, plays an essential role in improving the adsorption. The magnetic losses for magnetic materials together with ferrite are especially derived from domain wall resonance, hysteresis loss, eddy current loss, and natural resonance. [17, 18] Normally, the contributions to magnetic loss, inclusive of magnetic hysteresis, domain wall displacement can be excluded in NCZ ferrite. Reasoning about the eddy current effect and natural resonance mechanism, the eddy-current loss is associated with the diameter of nanoparticle (D_m) and electric conductivity (σ), which may be expressed through the equation $C_0 = \mu''(\mu')^2 f^{-1}$. If the magnetic loss outcomes from eddy-current loss, the value of C_0 need to be constant when the frequency varies, which is called the skin-effect criterion [19]. It can be visible from figure 7 that the value of C_0 changes significantly as a function of frequency in the frequency range of 8.0–18.0 GHz. But, as can be seen, for NCZ sample, C_0 remains about constant. therefore, one can conclude that the magnetic loss at 10.0 GHz is resulting from the natural resonance. Meanwhile, the alternative two minor peaks at 13.0 and 16.0 GHz are ascribed to eddy current effect, which may be added about by the relatively large diameter particles coming from the agglomeration of small nanocomposites.

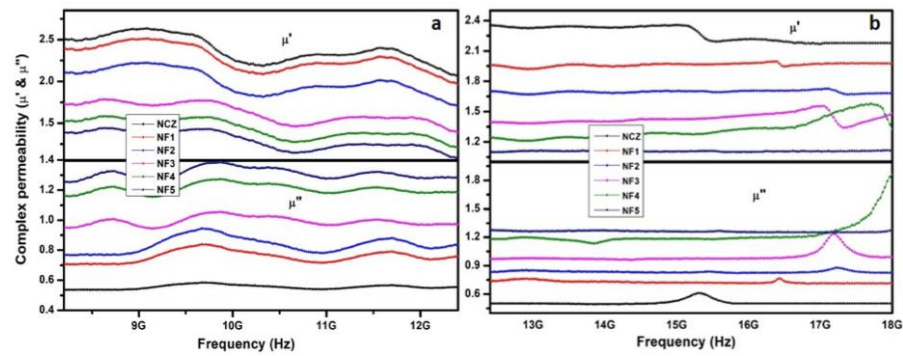


Figure 5: Complex permeability of x NCZ+ (1-x) PFD nanocomposites a) in X-band b) in Ku-Band region

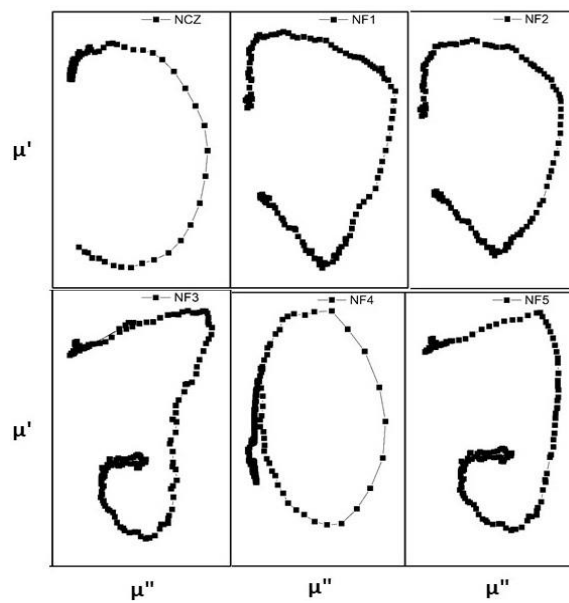


Figure 6: cole-cole plot of complex permeability of NCZ-PFD nanocomposite

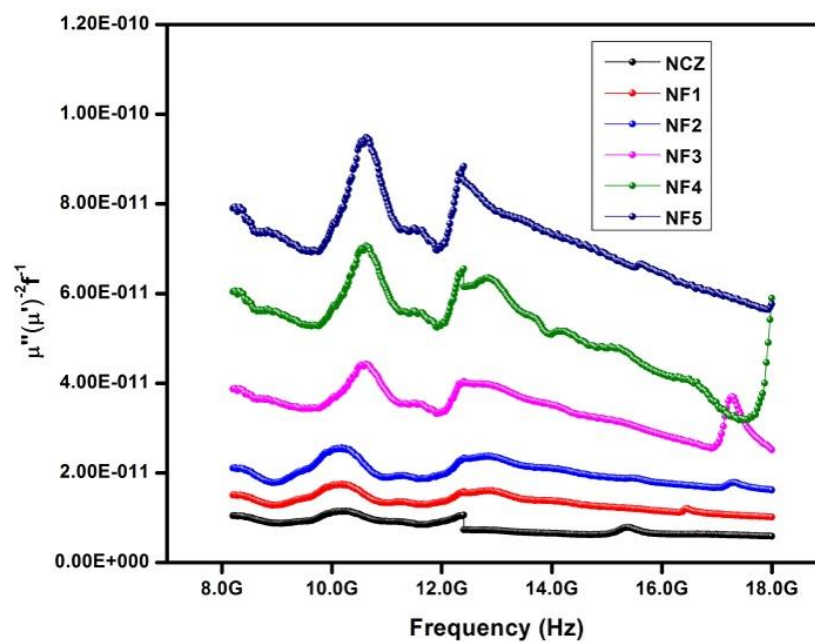


Figure 7: Plot of $\mu''(\mu')^{-2} f^{-1}$ vs frequency for the NCZ-PFD nanocomposite

Table 1: Complex permittivity and permeability properties of NCZ-PFD nanocomposites.

Sample	Crystallite size (nm)	Lattice parameter	at 8.2 GHz (X-band)				at 12.4 GHz (X-band)			
			ϵ'	ϵ''	μ'	μ''	ϵ'	ϵ''	μ'	μ''
NCZ	35	8.439	6.8	2.0	2.5	0.504	5.4	1.0	2.3	0.5
NF1	31	8.423	8.0	2.0	2.3	0.704	6.3	1.1	1.9	0.7
NF2	28	8.422	8.5	2.2	2.1	0.765	6.7	1.2	1.6	0.8
NF3	23	8.412	9.6	2.2	1.7	0.948	8.0	1.3	1.3	0.9
NF4	21	8.404	9.3	2.3	1.5	1.155	7.7	1.5	1.2	1.1
NF5	21	8.371	8.9	2.5	1.3	1.252	7.4	1.7	1.1	1.2

4. Conclusions

In summary, the NCZ-PFD nanocomposites were efficiently prepared by the use of mechanical milling approach. The structural characteristics of nanocomposites were investigated through XRD and SEM. Electrical and magnetic studies of nanocomposites had been investigated. The measurements of cole-cole properties found out that with the addition of PFD each dielectric and magnetic losses had been increased, multiple relaxations mechanism have been discovered. Therefore, the NCZ-PFD nanocomposites might be an attractive candidate for microwave adsorption materials.

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