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Structural, Electrical and Dielectric Properties of Ball-Milled $Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$/ SWCNT Nanocomposites

***S.E.Naina Vinodini, **P.Raju**

**Department of Physics, Government City College, (A), Nayapul, Osmania University, Hyderabad, Telangana 500002, India*

***Department of Physics, Osmania University, Hyderabad, Telanana-500007, India*

ABSTRACT

Nanocomposites of $Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$ with 1–5 wt.% SWCNTs were prepared by means of the high energy ball-milling method combined with the solid-state mixing method. Structural and electrical characteristics of the prepared nanocomposites were studied via X-ray diffraction and impedance spectroscopy methods. As revealed via XRD study, only single-phase cubic spinel ferrite phase was observed without any secondary phases; thus, it can be concluded that incorporation of SWCNTs into the ferrite matrix does not affect crystal structure of ferrite. Electrical conductivity increases in the samples with increase in content of SWCNTs due to the formation of conductive channels in the composites. Dielectric constant decreases with increasing frequency, while higher content of SWCNTs contributes to the increase in dielectric characteristics due to the presence of interfacial polarization. Enhanced electrical and dielectric properties reveal the contribution of SWCNTs into improvement of functional properties of NiCuZn ferrites.

Key Words: *NiCuZn ferrite; Single-walled carbon nanotubes (SWCNTs); Spinel ferrite; Ball milling; Nanocomposites; X-ray diffraction; Dielectric properties; AC conductivity.*

INTRODUCTION

Ni-Cu-Zn spinel ferrites are an important category of magnetic ceramics due to their high electrical resistivity, absence of eddy current losses, high chemical stability, and good magnetic properties that make them appropriate for usage in high-frequency electronics [1,2]. Among the spinel ferrites, Ni-Cu-Zn ferrites possess high initial permeability, low coercivity, and magnetic losses; hence, they find extensive application in transformer cores, multilayer chip inductors, antennas, microwave components, and EMI shielding materials [3-5]. Physical characteristics of Ni-Cu-Zn ferrites depend on their chemical composition, synthesis method, crystallite size, and microstructure [6,7]. Decreasing the particle size to the nanometer range increases the surface area and affects electrical and magnetic characteristics of ferrites, which is why nanocrystalline ferrites are highly desirable in modern electronic devices [5,8].

In addition to remarkable mechanical strength and thermal stability, carbon nanotubes possess exceptionally high electrical conductivity and high aspect ratio [9,10]. For this reason, CNTs have been widely used as conductive additives in ceramics and ferrites to improve the charge transport properties, dielectric response, and electromagnetic characteristics [11,12]. In relatively small amounts, single-walled CNTs (SWCNTs) can form the conductive paths in the ferrite matrix; thus, the electrical conductivity is enhanced while keeping the crystal structure of the material unchanged [12,13].

Several methods such as sol-gel, co-precipitation, hydrothermal synthesis, and high-energy ball milling have been used for production of ferrite nanoparticles [6,14]. Among those methods, the high-energy ball milling appears to be very promising due to its simplicity, economy, scalability, and capability to produce nanocrystalline powders with homogeneous composition [14,15]. Despite numerous studies of Ni-Zn and Ni-Cu-Zn ferrites, not many studies deal with $Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$/SWCNT nanocomposites prepared by the ball milling process. Hence, the goal of this research work is to synthesize $Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$/SWCNT nanocomposites with different concentrations of SWCNT and analyze the effect of SWCNT content on the crystal structure, AC conductivity, dielectric constant, and dielectric loss.

The selected material for the matrix was nickel-copper-zinc ferrite ($Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$), which was chosen considering its high electrical resistivity, low magnetic losses, chemical stability, and high initial permeability, making it ideal for use in high frequency electronic devices and EMI shielding applications [1-3]. The specific composition of this material provides a balance between its magnetic and electrical characteristics by partially substituting nickel with copper and zinc, which will lead to reducing eddy current losses [5]. Single walled carbon nanotubes (SWCNTs) were used as a filler because of its electrical conductivity, high aspect ratio, and mechanical properties. Addition of SWCNTs will help increase charge and dielectric transport and electrical conductivity through providing a conductive path through the ferrite matrix without disturbing the crystal structure of spinels [7-10].

MATERIAL AND METHODS

Analytical purity of nickel oxide (NiO), copper oxide (CuO), zinc oxide (ZnO), and ferric oxide (Fe_2O_3) were used as starting materials in the preparation of $Ni_{0.48}Cu_{0.12}Zn_{0.40}Fe_2O_4$ ferrite. Single walled carbon nanotubes (SWCNTs) were used as conductive additives.

SYNTHESIS OF $Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$ NANOPARTICLES AND CHARACTERIZATION

Nanoparticles of $Ni_{0.48}Cu_{0.12}Zn_{0.40}Fe_2O_4$ were obtained using high energy ball milling. The amount of NiO, CuO, ZnO, and Fe_2O_3 powders that were stoichiometrically accurate was carefully weighed and then blended well. The blend was subjected to ball milling, where a uniform precursor with minimized particle size was obtained. The ball milled blend was subjected to calcination at the right temperature to obtain the spinel ferrite structure. After cooling to room temperature, the calcinated product was then grounded to a fine powder.

The nanocomposites with 1, 2, 3, 4, and 5 wt.% of single-walled carbon nanotubes (SWCNTs) were produced by means of solid-state mixing technique. The required amount of SWCNTs was added to the synthesized ferrite powder and homogeneously dispersed by means of ball milling for the uniform dispersion into the ferrite host material. As a result, fine nanocomposite powders labeled as NC1, NC2, NC3, NC4, and NC5 were obtained depending on the increased SWCNT concentration.

X-ray diffraction (XRD) analysis was carried out in order to determine the crystal structure of the synthesized nanocomposites. The electrical and dielectric behavior of the nanocomposites was studied using impedance spectroscopy. Impedance spectra were used for the calculation of AC electrical conductivity, dielectric constant (ε'), and dielectric loss (tan δ).

RESULTS AND DISCUSSION

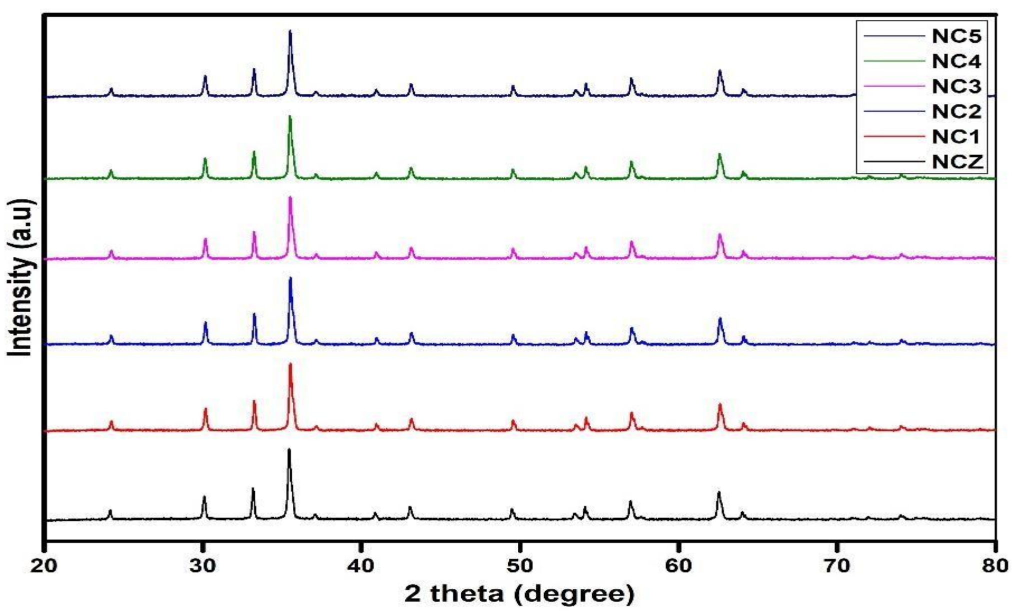
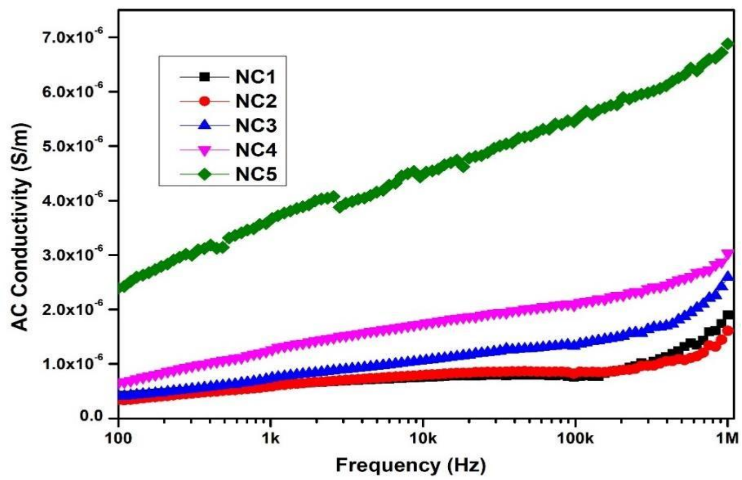


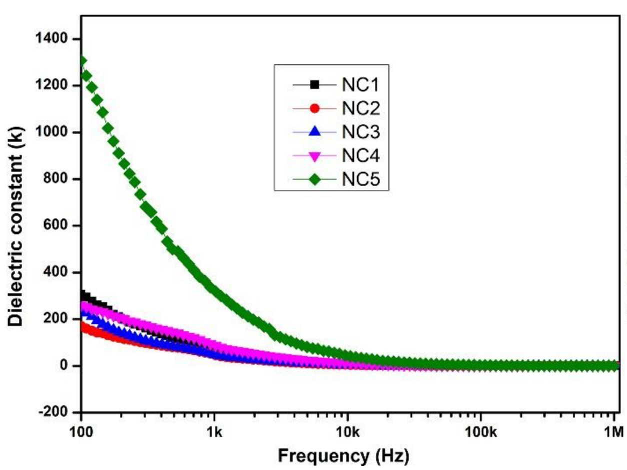
Figure 1 displays the XRD patterns of NCZ (pure Ni0.48Cu0.12Zn0.40Fe2O4) and SWCNT-incorporated nanocomposites (NC1-NC5). All the diffracted peaks match the cubic spinel ferrite phase, confirming that the ferrite phase has been formed successfully without any secondary phase formation. Addition of SWCNTs in the ferrite matrix has not shown any significant change in the crystal structure. This indicates that SWCNTs are uniformly distributed in the ferrite matrix without affecting the spinel structure. Decrease in peak intensity along with the widening of diffracted peaks with increase in the content of SWCNTs suggests decreased crystalline nature and crystal size due to the addition of carbon nanotubes.

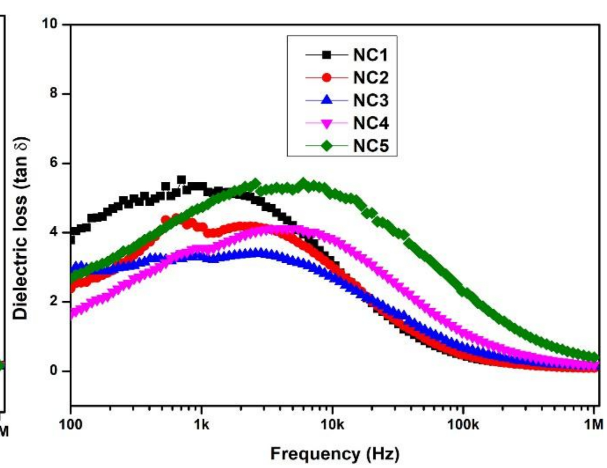
AC CONDUCTIVITY



Frequency dependence variation of the AC conductivity of the prepared nanocomposites is shown in Fig. 2. With all the nanocomposites, conductivity increased with increase in frequency, a phenomenon observed with ferrites. Addition of SWCNTs resulted in a large increase in electrical conductivity, with NC5 having the highest value. It is attributed to the formation of conductive path due to interlinked network of SWCNTs that enhances charge carriers' movement through the ferrite structure.

DIELECTRIC PROPERTIES





The frequency-dependent variation of the dielectric constant and dielectric loss is shown in Figures 3 and 4, respectively. The increase in frequency leads to the decrease in the dielectric constant in all compositions which is consistent with the Maxwell–Wagner interfacial polarization and Koops' theory. Interfacial polarization increases the dielectric properties at low frequencies, but at higher frequencies the dipoles do not have enough time to follow the changes in the oscillating electric field, and thus a decline in the value of the dielectric constant is observed.

The effect of frequency on dielectric loss was also studied. Compositions with a higher concentration of SWCNTs exhibit relatively larger dielectric loss due to increased charge-carrier hopping and interfacial polarization. However, an increase in frequency causes a decrease in the value of dielectric loss.

To conclude, the addition of SWCNTs improves the electrical conductivity and dielectric properties of $Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$ nanocomposites without changing the spinel crystal structure. These characteristics show the possibility of using such materials in high-frequency electronics and EMI shielding.

CONCLUSION

$Ni_{0.48}Cu_{0.12}Zn_{0.4}Fe_2O_4$/SWCNT nanocomposites have been prepared using high-energy ball milling and then solid-state mixing process. It is well established that the addition of single walled carbon nanotubes (SWCNTs) greatly increases the electrical and dielectric characteristics of the ferrite system without any degradation in its structural stability. From the increase in performance with respect to increase in the concentration of SWCNTs, it can be said that the incorporation of carbon nanotubes improves the behavior of NiCuZn ferrites. Based on structural stability and improved electrical performance of the nanocomposites, they can be used in high frequency electronics, EMI shielding, microwave absorption, and sensor applications. Further studies on their magnetic and electromagnetic performance can provide more information about their applicability in multifunctional devices.

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