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Dielectric and Magnetic Study of CNTs-Fe_3O_4 Composites

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A comparative study of dielectric and magnetic properties of Fe_3O_4 and CNTs-Fe_3O_4 composite has been reported in paper. In this, CNTs-Fe_3O_4 composites have been prepared by dispersing the CNTs in ionic (Fe^{+2}/Fe^{+3}, in 1:2) solution via chemical co-precipitation route. The dielectric properties of composites have been investigated in 10^1-10^6 Hz frequency range. The saturation magnetization (M_s) of CNTs-Fe_3O_4 composite is reduced. The dielectric permittivity of composites with increasing weight fraction of CNTs is also reduced which may be due to the space polarization of charges accumulated via conducting channels of CNTs.

Keyword: Composite materials, Chemical vapour deposition (CVD), Magnetic materials, Magnetic properties, Dielectric properties.

1. INTRODUCTION

In advanced phase of research in nanotechnology, carbon nanotubes have become a charming material due to their prominent electrical, mechanical and chemical properties [1,2]. Besides weak magnetic nature of CNTs, the highly magnetic nano-crystalline material like Fe_3O_4, has achieved various technological applications in biomedical research like cell labelling, magnetic resonance imaging (MRI), targeted drug delivery and magnetic ferrofluids [3,4]. As such, nano-materials became failure in fulfilment of all required properties, so the nano-crystalline composite materials came in interest [5]. The CNTs based magnetic composites have also obtained various potential applications as in microwave absorption [6,7,8] and targeted drug delivery [9]. The earlier research on CNTs based magnetic composites have explored about the synthesis and coating of Fe_3O_4 within the CNTs, by filling of ferrofluids, by decomposition of ferrocene in CNTs at different temperatures, and by co-precipitation of ionic (Fe^{2+}&Fe^{3+}) solution in presence of CNTs [10-15]. Thus in this work, CNTs-Fe_3O_4 composites of varying weight fraction of CNTs have been fabricated by chemical co-precipitation route. The magnetic and dielectric study of composites has also been done to investigate.

2. EXPERIMENTAL

2.1. Synthesis of CNTs, Fe_3O_4 & CNTs-Fe_3O_4

CNTs have been synthesized by chemical vapour deposition in which toluene as hydrocarbon source and ferrocene as iron catalyst burnt in argon atmosphere at 750°-760°C [16]. Then amorphous carbons have been removed by oxidising the above produced CNTs in air at 350^oC for 1hr. The CNTs were treated with acids to enhance their chemical reactivity.

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The synthesis of Fe_3O_4 particles was through chemical co-precipitation method. In this method, the transparent aqueous solution of Fe^{+2} and Fe^{+3} ions was stirred at temperature 70°C for half an hour and complexes of ferrous hydroxide-ferric hydroxide $[Fe(OH)_2.Fe(OH)_3]$ were precipitated out on adding NH_4OH solution maintaining the pH~10. The precipitate get washed properly with deionised water (Millipore, Direct-Q5, 18.2MΩcm) and finally dried at 80°C to get Fe_3O_4.

The CNTs-Fe_3O_4 composites have been synthesized on dispersion of CNTs in ionic (Fe^{+2}/Fe^{+3}, in 1:2) aqueous solution via chemical co-precipitation route. The composites prepared have 0.20g and 0.25g of CNTs respectively. The black powder of Fe_3O_4 and composites: CNTs-Fe_3O_4 (0.20g) and CNTs-Fe_3O_4 (0.25g) are indicated by *a*, *b* and *c* respectively.

2.4. Characterization

The phase formation and crystalline nature of samples were analysed using Rigaku make powder X-ray diffractometer (XRD) equipped with CuKα radiation at 0.02°/s scanning rate in 2θ range 20-70° at 40 kV, 30 mA with automatic divergence slit. The morphological study of composites has been analyzed by scanning electron microscope (Model-LEO 440 with EDS attachment Model-OXFORD-LINK ISIS-300) operated at 30-40 kV. The magnetic measurements of samples were done by search coil method. For this study, a polytronic power supply (Model-BCS-1000), electromagnet (Type Hem-100) and flux meter (Model-FM109) were utilized. This instrument was calibrated with Ni. For dielectric study, the Novo control alpha-A high performance frequency analyzer was used in 10^1-10^6 Hz frequency at 1volt AC and 1volt DC feedback biasing.

3. RESULTS AND DISCUSSION

3.1. SEM

The scanning electron micrograph of CNTs-Fe_3O_4 (0.25g) composite is shown in Figure 1. It was done to see the attachment of Fe_3O_4 particles with CNTs and thus exploring the composite prepared.

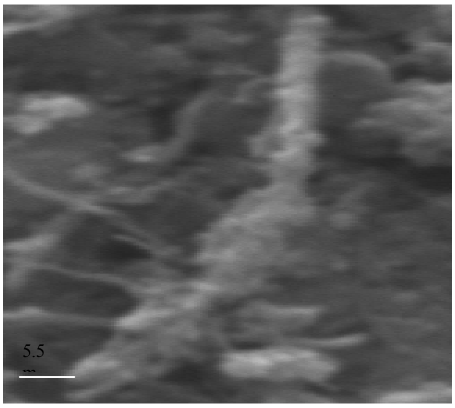

Fig. 1: Scanning electron micrograph of composite CNTs- Fe_3O_4 (0.2g).

3.2. XRD

The X-ray diffraction patterns of samples are shown in Figure 2. The characteristic diffraction peaks at 35^{o} (311) and 26° (002) are due to Fe_3O_4 and CNTs respectively, confirming the phase formation. The crystallite size of Fe_3O_4 corresponds to (311) peaks have been calculated by using Scherrer formula, D=kλ/βcosθ. The average crystallite size is varied in range of 13-15 nm as given in Table 1.

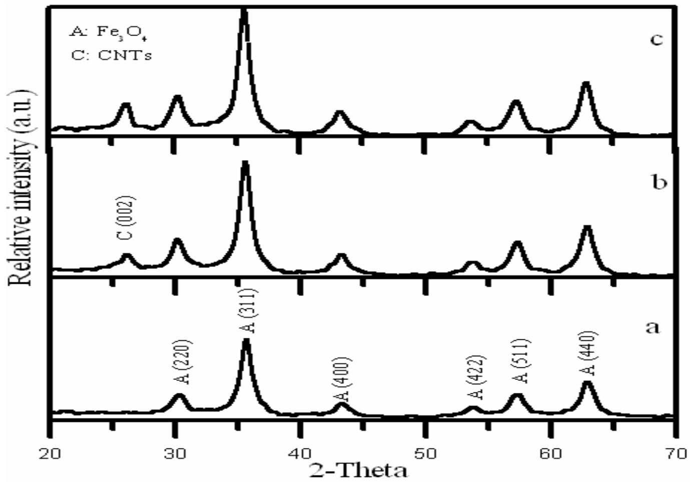

Fig. 2: X-ray diffraction pattern of a: Fe_3O_4 and composites *b* and *c*.

Table 1: Parameters of different samples.

Samples	**Crystallite size (nm)**	**σ_s (emu/g)**	**Coercivity (G) $H_c = (H_{c1}+H_{c2})/2$**	**Retentivity (emu/g)**
Fe_3O_4	13.12	85.96	131.5	13.42
Composite *b*	14.61	72.99	124.54	10.65
Composite *c*	15.26	67.85	133.5	9.35

3.3. Magnetic measurements

Magnetic properties of samples have been investigated at room temperature. The magnetization curves are shown in Figure 3. All composites have exhibited the typical ferromagnetic behaviour. The saturation magnetizations (σs) of composites have been reduced with increasing CNTs weight fraction. The saturation magnetization (σ_s), retentivity (M_r) and coercivity (H_c) of composites are given in Table 1. It reveals that CNTs in magnetic composites cause the significant changes in magnetic properties.

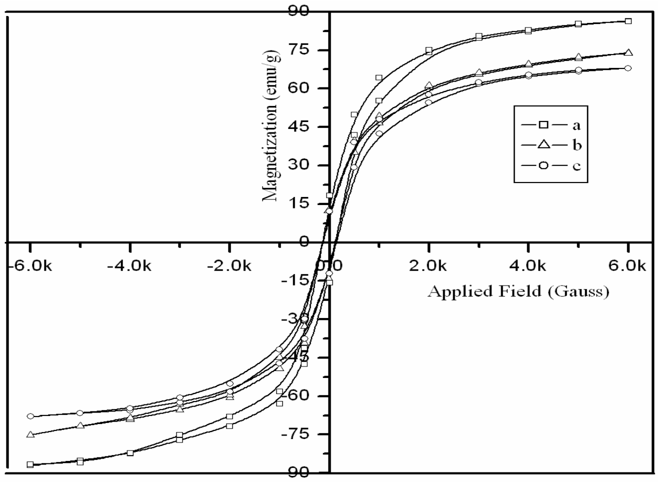

Fig. 3: Magnetization curves of a: Fe_3O_4, composites *b* and *c*.

3.4. Dielectric study

The dielectric properties of Fe_3O_4 and CNTs-Fe_3O_4 composites like dielectric permitivity (ε'), dielectric loss (ε'') and tangent loss (δ) have been investigated in 10^1–10^6 Hz at room temperature and is shown in Figure 4 (a, b and c). In Figure 4(a), dielectric permittivity of composites decreased sharply below 10^2 Hz only and while in Fe_3O_4, it is regularly decreased in whole frequency range.

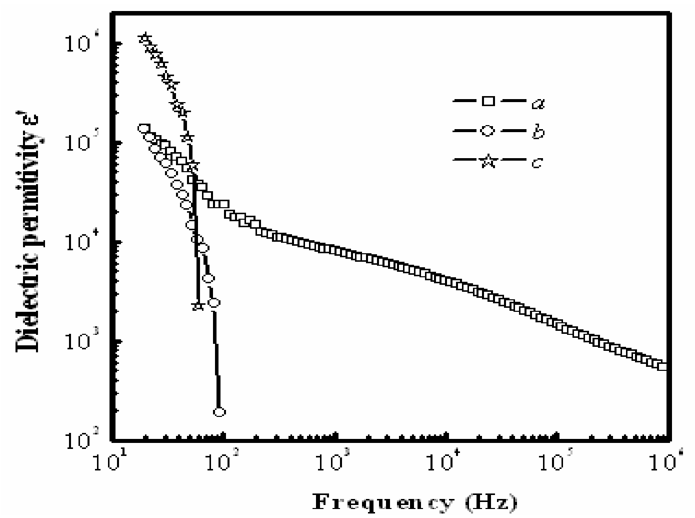

Fig. 4(a): Dielectric permitivity variation with applied frequency: Fe_3O_4, sample *a*, *b* and *c*.

The tangent loss (δ) factor of samples has shown in Figure 4(b), in composites have larger loss factor than Fe_3O_4, which decreases linearly above 10^2 Hz. Thus CNTs-Fe_3O_4 composites have larger dielectric permittivity and tangent loss than Fe_3O_4. The variation of AC conductivity of Fe_3O_4 and CNTs-Fe_3O_4 composites with applied frequency is shown in Figure 4(c).The conductivity can be expressed as $\sigma(\omega) = \omega\varepsilon_0\varepsilon''$, where σ is the real part of the conductivity and ε'' is the imaginary part of dielectric constant. The conductivity of Fe_3O_4 seems independent to frequency below 10^3Hz and above it, the conductivity increased linearly. While in composite *b* and *c*, conductivity is higher below 10^2 Hz. The calculated values of AC conductivity of Fe_3O_4 and composites *b* and *c* have 84.31×10^{-3}, 10.15×10^1 and 58.11×10^1 S/cm. This reveals that conducting network of CNTs has leading the polarization of space charges in Fe_3O_4 which caused of increasing conductivity of composite with applied frequency and thus affecting the dielectric properties of Fe_3O_4 and composites.

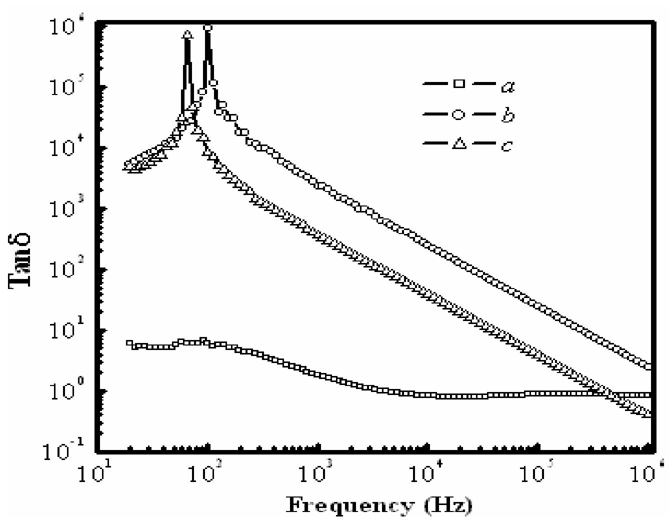

Fig. 4(b): Dielectric loss variation with applied frequency: Fe_3O_4, sample *a*, *b* and *c*.

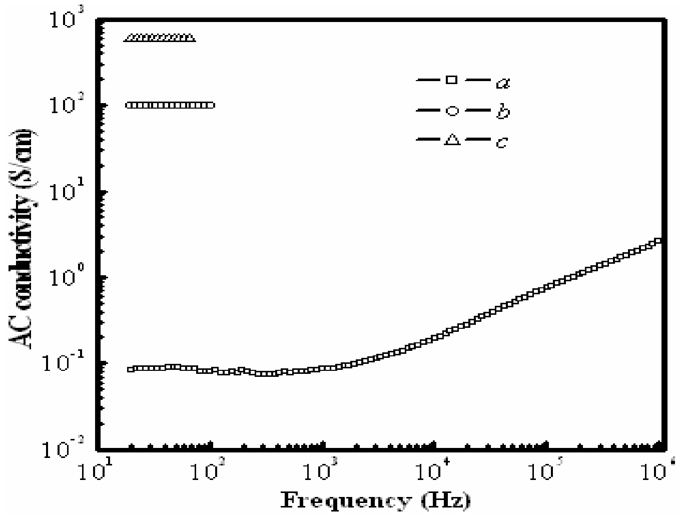

Fig. 4(c): Tan δ variation with applied frequency: Fe_3O_4, composites *a*, *b* and *c*.

4. CONCLUSIONS

The CNTs-Fe_3O_4 magnetic composites were prepared by chemical co-precipitation method. The crystalline natures, magnetic and dielectric properties of Fe_3O_4 and composites have been investigated. These composites can have good application in magnetic drug delivery and absorption. These composites can also have good application in magnetoelectric materials.

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