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Effect of concentration of barium ferrite nanoparticles on dielectric properties of $BaFe_2O_4$/MWCNT/epoxy nanocomposites

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Abstract : In present work, Co-precipitation method was employed to synthesize BaFe2O4 nanoparticles. The fabrication of BaFe2O4/MWCNT/epoxy nanocomposites with different concentration of BaFe2O4 carried out by mechanical mixing and molding method. The prepared nanocomposites were characterized by X-ray diffraction, UV-Vis spectroscopy, and impedance spectroscopy. The particle size was found to be about 9.457 nm for BaFe2O4 by using Debye Scherrer formula. Impedance spectroscopy measurement of nanocomposites was taken at room temperature and observed that the value of dielectric constant decreases with increasing frequency and dielectric loss increases with increasing frequency. The maximum value of dielectric constant was demonstrated by ECB-5 (40 wt% of BaFe2O4) composite, where the wt% of MWCNT kept constant at 2. The dielectric loss for ECB-5 composite was found to be ~ 0.05 at lower frequency and this value increases with increase in frequency.

Keywords: Nanocomposites; Dielectric constant; Dielectric loss; Co-precipitation; ferrite

Introduction

Nowadays, multifunctional composites are in great demand to meet the requisite of technology. Ferrites are known to be widely applicable ceramic materials in technology due to their interesting magnetic properties; ferrites introduce significant participation in magnetic, electric and dielectric properties due to their spinel structure [1]. Nano-ferrites are synthesized by bottom up approaches such as chemical co-precipitation, sol-gel, micro-emulsion and hydrothermal methods [2]. Several processing parameters effect the quality of materials such as method of formation, sintering time and temperature because size and crystal composition are strongly influenced by these parameters [3]. Chemical co-precipitation technique is economically approachable way to introducing nano-ferrites. Due to high crystal anisotropy, and excellent intrinsic coercivity, barium ferrites are used as permanent magnets in recordings [4]. Due to magnetic properties, physical properties, high corrosion resistance, high resistivity and excellent chemical stability barium ferrite can be used in communication, automobile industries, energy formation, and in biomedical field [5, 6].

Polymers have gained great attention due to their ease of processing, ease of forming, thermal stability and low cost. Polymer/ceramic nanocomposite was studied and proven to be a great combination to crucially obtain the magnetic and dielectric properties, and conductivity by varying wt. % of nanoparticles [7]. Epoxy resin is non-conductive and therefore can be used in electronic industrial field in transistors, integrated, hybrid and printed circuits for protection from dust, moist and short circuit and [8]. Sanida et al.(2018) [9] prepared two series of nanocomposites by combining epoxy and $BaFe_{12}O_{19}$ or $SrFe_{12}O_{19}$ nanoparticles, and dielectric properties were investigated via dielectric spectroscopy and observed enhanced dielectric response. Dielectric values exhibited by $SrFe_{12}O_{19}$ inclusion are higher than $BaFe_{12}O_{19}$ inclusion due to interfacial polarization and re-orientation of dipoles. Wang et al. [10] prepared CNT integrated $BaFe_{12}O_{19}$ composite using ball-milling technique and observed that CNT inclusion ameliorated dielectric loss. Pawar et al. [2] prepared $Co_{1-X}Ba_XFe_2O_4$ (X=0 to 1) nano-ferrites as dielectric and gas sensing material by chemical co-precipitation technique and observed that dielectric value of material enhanced with doping of barium.

In present work, series of nanocomposites were

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fabricated by taking epoxy as matrix and MWCNT and $BaFe_2O_4$ as embedded nanofillers and their dielectric behavior were studied by varying nanofillers wt. % . The performance of fabricated materials was investigated through X-ray diffraction (XRD), while dielectric response was investigated by LCR meter.

Material & Methods

In present work, $BaCl_2.6H_2O$ (barium chloride, 98+% purity), $FeCl_3.6H_2O$ (ferric chloride, 97+% purity), NaOH flakes were used to synthesize ferrite and deionized (DI) water was used as solvent. Commercially available multiwall nanotubes (MWCNTs) and epoxy of Sigma Aldrich Company (china) were used to develop composites.

Chemical co-precipitation method was used to synthesize $BaFe_2O_4$ (BFO) nanoparticles. Where, to prepare chloride precursors $BaCl_2.6H_2O$ and $FeCl_3.6H_2O$ (1:2) were dissolved in 50 ml DI water in separate beakers measured through measuring flask and stirred for 20 minute each for proper dispersion. Then combined both solutions and stirred for 20 minute. Further, this complex solution was added to 0.8 M of alkaline solution (NaOH) drop by drop through burette and the formation of dark brown precipitation occurred. Then the solution was stirred continuously for 1 more hour after last drop to let it react properly. Then the precipitate was washed off to eliminate all the chloride impurities thoroughly, from the precipitation by washing the precipitation 3-4 times with DI water using centrifugation process. Furthermore, the precipitate was collected and dried in oven at 100 ^{0}C temperature. Then the dried sample was grinded until the fine power was formed and sintered at 200 ^{0}C for 1 hour to get crystalline nanoparticles.

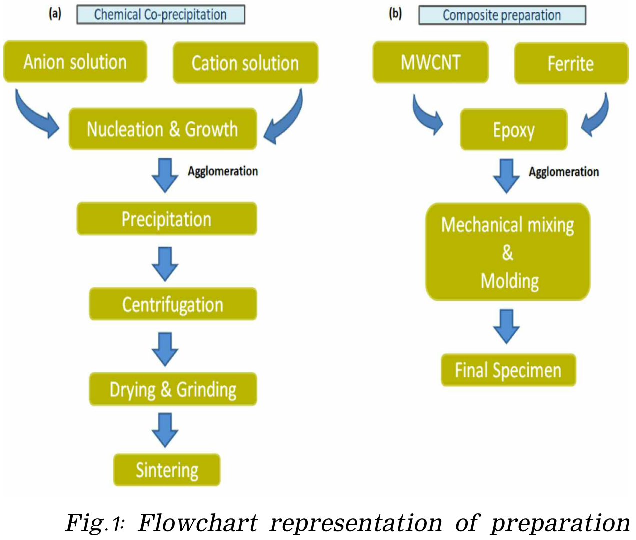

Fig.1: Flowchart representation of preparation methods.

Composite fabrications: To fabricate the composite epoxy and hardener were added in 10:1 ratio and mixed well using a glass rod. Then MWCNT and BFO were added to epoxy solution as fillers and mixed well. Further, the mixed solution was ultrasonicated for better dispersion for 10 minute total. A series of composites were prepared by keeping the MWCNT (2 wt. %) constant and varying the wt. % of BFO from 0, 5, 10, 20, 30, 40, 50 to 60 wt. % shown in table 1.

Composites	MWCNT %	BFO %
E	0	0
EC	2	0
ECC-1	2	5
ECC-2	2	10
ECC-3	2	20
ECC-4	2	30
ECC-5	2	40
ECC-6	2	50
ECC-7	2	60

Table 1: Composite formation of prepared specimen

Characterization

The structural and phase information of BFO nanoparticles were analyzed by X-ray diffractometer by Bruker diffractometer, model D8 Advance by Cu/Kα radiation with 0.5/s scan rate in 34 kV operating and 26 mA operating current of X-ray source in between 20° to 80° (2θ) range. To analyze the optical properties of nanoparticles UV-Vis spectrophotometer (GENESYS) was used in 200 to 1100 nm wavelength. Dielectric properties of prepared composites were measured in 40 Hz to 4 MHz at room temperature (27 ^{0}C) using LCR meter (IM-3536 HIOKI).

Results & Discussions

X-ray diffraction

The XRD pattern of CFO nanoparticles is exhibited in fig. 1. Intensities peaks and the angles of scattered X-ray are used to analyze the structure and phase of particles. Peak positions of BFO is matched with standard JCPD File of BFO (46-0113) which appeared to be in correct positions that confirms pure single-phase orthorhombic structure of BFO nanoparticles. The average sizes of the particles are determined using (311), (212), (220 or 701), (521), (422), (703) miller indices of fig. 1 using Debye Scherer formula.

$$D = \frac{0.89\lambda}{\beta \cos \theta} \qquad (1)$$

Where D is crystalline size of particle, λ is wavelength of X-ray, θ is Bragg's angle, and β is FWHM in radian. The average crystalline size of $BaFe_2O_4$ (BFO) nano-ferrites was obtained around 36.299 nm.

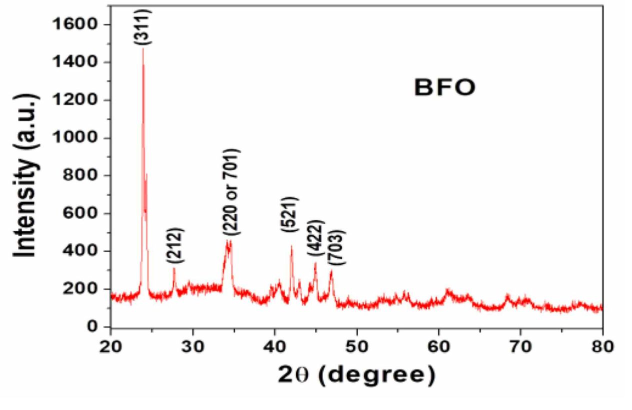

Fig. 2 : XRD pattern of synthesized $BaFe_2O_4$ (BFO) nano-ferrites

UV-Visible spectroscopy

The optical properties of BFO nano-ferrite are illustrated in Fig. 2. Prepared nano-ferrite was exposed in optical range of UV-Visible range (200-700 nm). Absorption edge of BFO nano-ferrite were observed to be around 236 ± 2 nm using given absorption formula in eq.(2)

$$\alpha = \frac{A(h\nu - Eg)^{1/2}}{h\nu} \qquad (2)$$

Where, α is absorption coefficient, A is constant, hν is photon energy, Eg is energy band gap. The band gap energy (Eg) for BFO was estimated from an extrapolation of the linear region of plot of $(\alpha h\nu)2$ vs. hν (energy) estimated in fig. 2(b). The energy band gap (Eg) was found to be ~4.04 eV for BFO nano-ferrite.

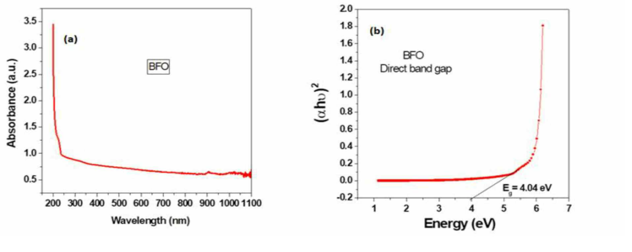

Fig. 2: (a) Energy absorption spectrum of BFO nano-ferrite. (b) Energy band gap of BFO nano ferrite

Dielectric analysis

The dielectric behavior for $BaFe_2O_4$/ MWCNT/ epoxy composites with 0, 5, 10, 20, 30, 40, 50, 60 wt. % of BFO were studied by LCR meter in frequency range from 40 Hz to 4 MHz at room temperature.

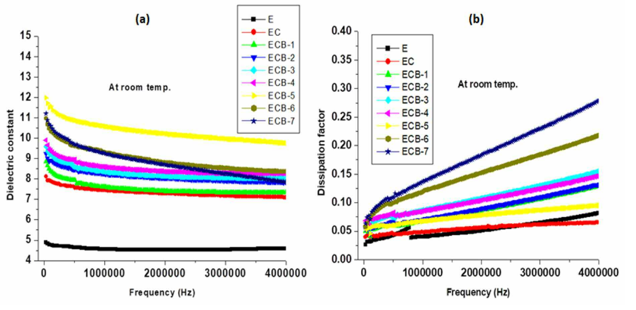

Fig. 3: (a) Dielectric constant and (b) Dissipation factor of composites within 40 Hz to 4 MHz at room temperature (27 ℃)

$$\varepsilon_r = \frac{Cd}{\varepsilon_0 A} \qquad (3)$$

Where, ε_r is the dielectric constant/permittivity, C is capacitance, d is thickness of sample, A is cross-section area of sample and ε_0 is absolute permittivity and ε_0=8.85×10—12 F/m.

From Fig. 3(a) and (b), high value of dielectric constant was obtained at initial frequency and it started to fall on increasing frequency and become constant after 500kHz due to saturation of polarization. Dielectric constant increased with filler wt. % (from 0 to 40 wt. %) attributed to augmentation of charges at interface responsible for interfacial polarization. Further, incorporation of filler (>40 wt. %) exhibited lower dielectric constant compare to composite with 40 wt. % led by less number of bound charges. Thus, among all the prepared nanocomposites highest dielectric constant value of 11.9 was found for ECB-5. However, the dissipation factor tends to increase with increasing frequency due to increased oscillation of charges. However, the dielectric loss were found maximum for building block having 60% doping of BFO and the lowest loss were found 0.055 for ECB-5 compare to other. Hence, we can say that the 40% doping of BFO in CNT along with epoxy shows the better dielectric properties.

Conclusion

It was concluded that $BaFe_2O_4$/MWCNT/epoxy nanocomposites (BFO wt. % 0, 5, 10, 20, 30, 40, 50, 60) were successfully prepared by chemical co-precipitation, mechanical mixing and molding technique. From the

XRD data, successful formation of pure BFO nanoparticles was confirmed. It was estimated that the absorption edge was found in UV (236 ± 2 nm) range. The calculated band gap energy was 4.04 eV. The highest dielectric constant and lowest dissipation factor were found for ECB-5 nanocomposites.

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