



ISSN Print: 2394-7500
 ISSN Online: 2394-5869
 Impact Factor: 5.2
 IJAR 2018; 4(12): 478-483
 www.allresearchjournal.com
 Received: 06-10-2018
 Accepted: 10-11-2018

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Structural and dielectric properties magnetic barium cobalt hexaferrite particles-PVA composites with different mass ratio

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Abstract

X-type $\text{Ba}_2\text{Co}_2\text{Fe}_{28}\text{O}_4$ powders were successfully synthesized using a reverse co precipitation technique. $\text{Ba}_2\text{Co}_2\text{Fe}_{28}\text{O}_4$ -PVA composites were prepared with different mass ratio. Structural as well as dielectric properties of $\text{Ba}_2\text{Co}_2\text{Fe}_{28}\text{O}_4$ -PVA composites studied at room temperature.

Keywords: Barium cobalt hexaferrites, PVA, XRD, SEM, dielectric properties

Introduction

Hexaferrites are found to be a technologically important class of magnetic oxides and widely used as permanent magnet material in household products such as refrigerator, magnetic recording media, microwave absorbers because they have high Curie temperature, high saturation magnetization, high electrical resistivity, low eddy current and dielectric loss, excellent chemical stability, corrosion resistance and they are relatively cheap to produce. The crystal structure of X-type hexaferrites (Figure 1) is closely related to M and W-type and constructed as stack of hexagonal R-block and spinel S-block along the hexagonal c-axis with a model as $\text{RSR}^*\text{S}^*\text{S}^*$ (the '*' indicates the rotation of corresponding block by 180° around the c-axis) [1, 2]. The chemical formula of X-type hexaferrite is $2 \text{BaO} \cdot 2 \text{MeO} \cdot 14 \text{Fe}_2\text{O}_3$; (where Me represents 3d-transition metal element). Braun [3] reported the crystal structure of $\text{Ba}_2\text{Fe}_{30}\text{O}_{46}$ hexaferrite. The crystallographic and magnetic characteristics of the metallic sublattices in the X-type $\text{BaMe}_2\text{Fe}_{28}\text{O}_{46}$ hexaferrite is shown in Table 1.

Table 1: The Crystallographic and Magnetic Characteristics of the Metallic Sublattices in the X-type $\text{BaMe}_2\text{Fe}_{28}\text{O}_{46}$ Hexaferrite.

Block	Coordination	Number per block	Expected spin direction
R	Octahedral	2	Down
R	Trigonal bipyramidal	1	Up
R-S	Octahedral	3	Up
S	Octahedral	1	Up
S	Tetrahedral	2	Down
S-S	Octahedral	3	Up

Although there have been many investigations into ferrite powder-polymer composites but less work reported on ferrite-polymer composites where PVA is chosen as matrix [4-6]. Crystallographic x-ray diffraction and magnetic data of polycrystalline samples of X-type hexagonal ferrites reported by Gu *et al.* [7-9]. G. Xiong and Mai [10] reported preparation and magnetic properties of that $\text{Ba}_2\text{Co}_2\text{Fe}_{28}\text{O}_{46}$ (Co_2X) nanocrystals. Pullar and Bhattacharya [11] studied synthesis and characterization of stoichiometric Co_2X fibers were prepared using a sol-gel based process. The preparation and microwave properties of $\text{Ba}_2\text{Zn}_x\text{Co}_{2-x}\text{Fe}_{28}\text{O}_{46}$ hexaferrites reported by Haijun *et al.* [12, 13]. Crystallographic and magnetic properties of Cu_2X , Co_2X , and Ni_2X hexaferrites published by Kenji Kamishima *et al.* [14]. Strontium based X-type ferrite $\text{Sr}_2\text{Fe}_{30}\text{O}_{46}$ is also reported [15].

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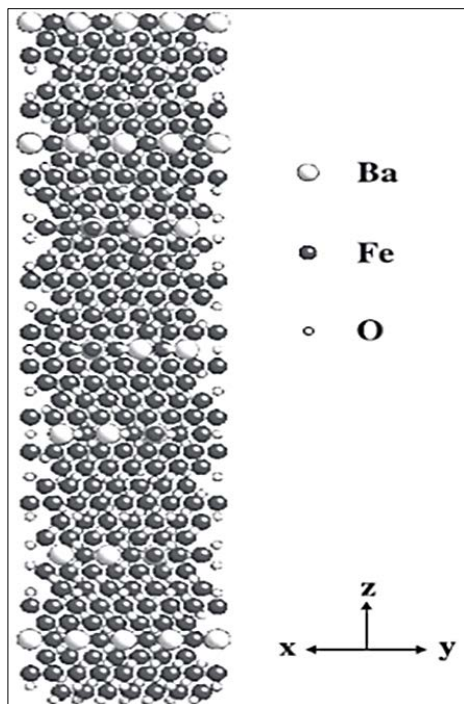


Fig 1: Crystal Structure of X-type Hexaferrite [3].

PVA; $[-CH_2-CHOH-]_n$) is a white, odorless, water-soluble, semi-crystalline synthetic polymer. PVA has a two-dimensional hydrogen-bonded network sheet structure and widely used in diverse applications such as adhesives for paper, wood, textiles, leather, other water-absorbent, surgical devices, sutures, hybrid islet transplantation, implantation, blend membrane and in synthetic cartilage in reconstructive joint surgery [16-19]. The X-type hexaferrite with chemical composition $Ba_2Co_2Fe_{28}O_{46}$ chosen for this study.

The $Ba_2Co_2Fe_{28}O_{46}$ hexaferrite powder was prepared using a reverse co-precipitation technique. The $Ba_2Co_2Fe_{28}O_{46}$ hexaferrite powder (filler) -PVA (matrix) composite samples were prepared by mixing ferrite powder to gather with PVA in different mass ratio 9:1, 8:2, 7:3, 6:4 and 5:5. The mixed powder pressed in a steel mould under 55 MPa for 300s. In present paper we report structural and dielectric properties of $Ba_2Co_2Fe_{28}O_{46}$ hexaferrite (filler)-PVA (matrix) composites.

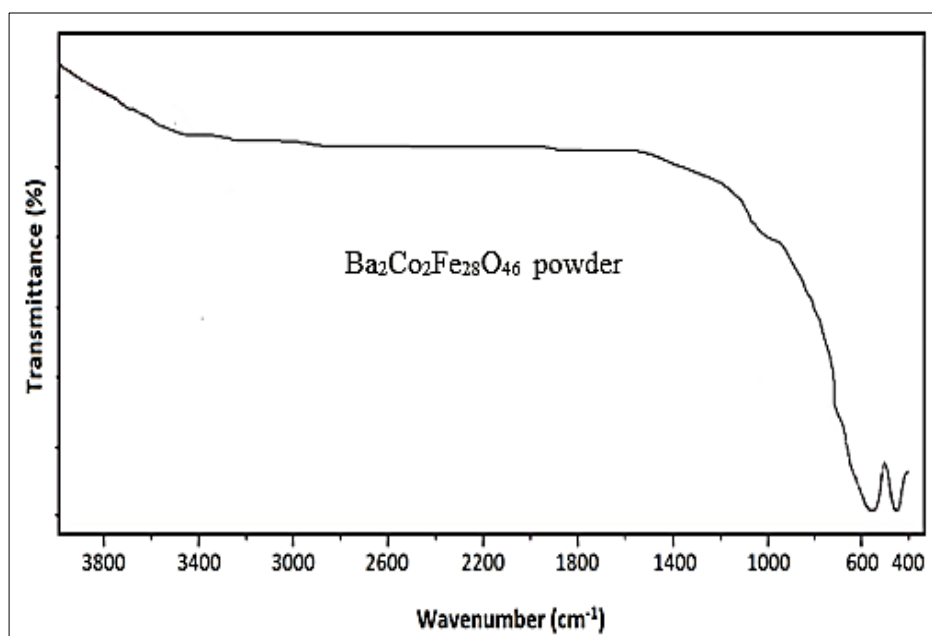
Materials and Methods

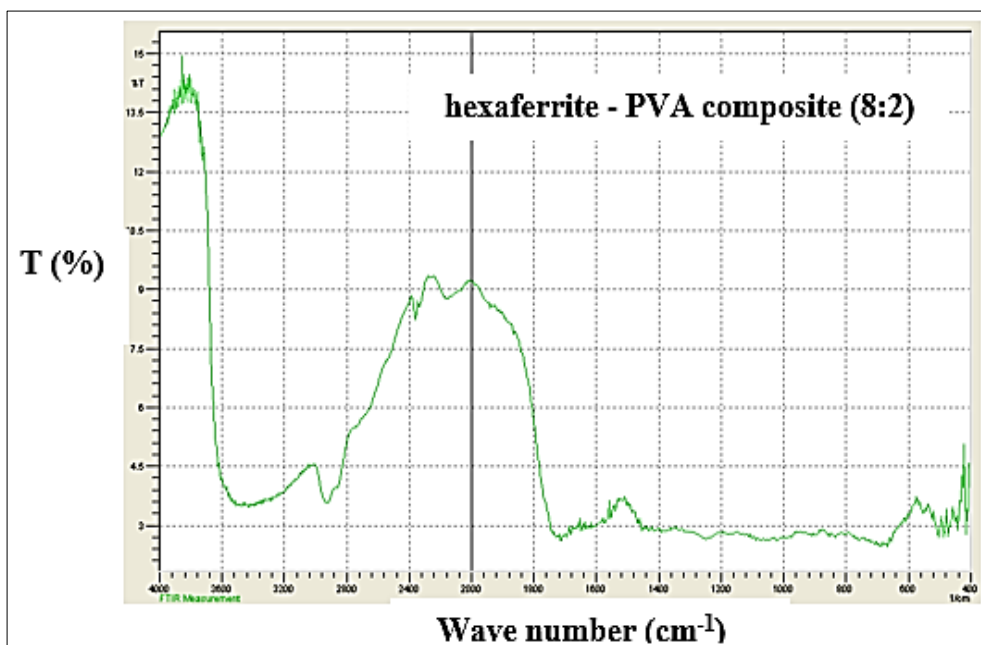
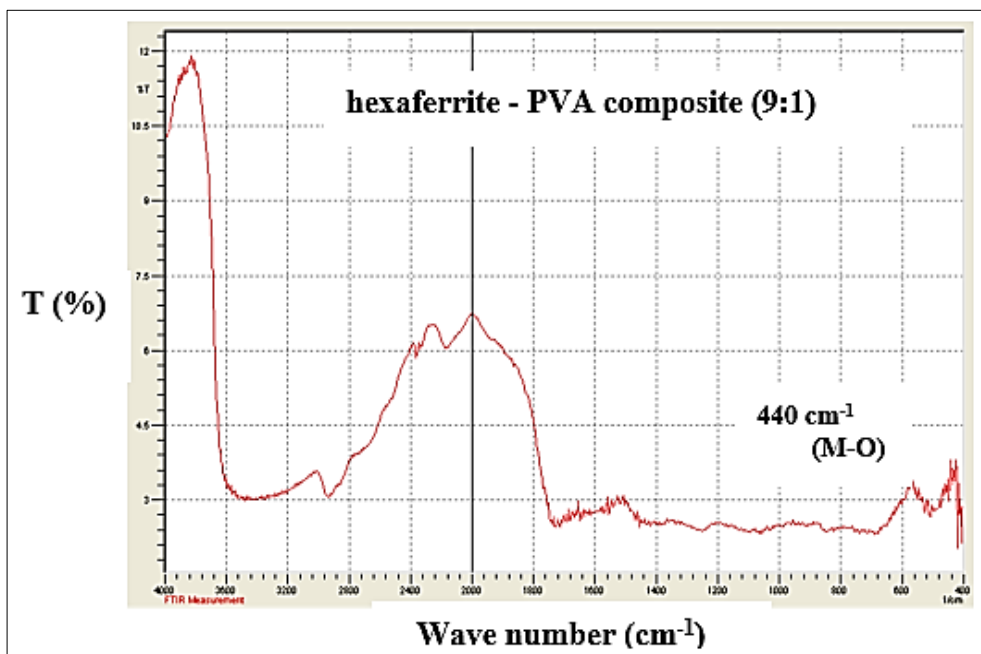
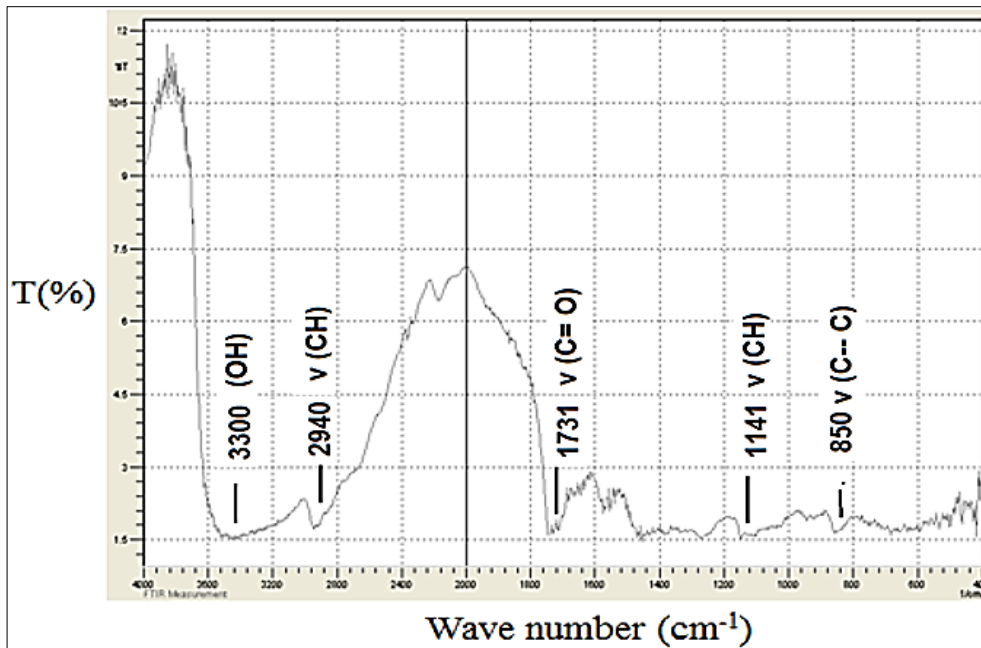
The reverse co-precipitation technique is used to X-type $Ba_2Co_2Fe_{28}O_{46}$ hexaferrite powder. The $Ba(NO_3)_2$, $Co(NO_3)_2$, $Fe(NO_3)_3$ dissolved in distilled water according to weight formula, then sodium hydroxide was added drop until the precipitates form (pH~7). The obtained solution kept 24 hours for aging and then filtered. The radish precursors recollected and kept at 100 °C for 24 h then grind well. The dried powder preheated at 450 °C for 4 h in a muffle furnace and finally material is sintered at 950 °C for 4h to obtain barium-cobalt hexaferrite powder. The $Ba_2Co_2Fe_{28}O_{46}$ hexaferrite mixed well with PVA (poly vinyl alcohol) in different mass ratio 9:1, 8:2, 7:3, 6:4 and 5:5 to prepare $Ba_2Co_2Fe_{28}O_{46}$ -PVA composites. The structural properties of the composites were characterized by Infrared spectroscopy (FTIR). The effect of the concentration of $Ba_2Co_2Fe_{28}O_{46}$ hexaferrites on structural and dielectric properties of magnetic-PVA composites was investigated.

Results and Discussion

FTIR analysis

FTIR spectra of ferrite, PVA and composite samples were obtained within the range between $4,000\text{ cm}^{-1}$ and 400 cm^{-1} using FTIR spectrometer (Bruker Tensor 27 Model).





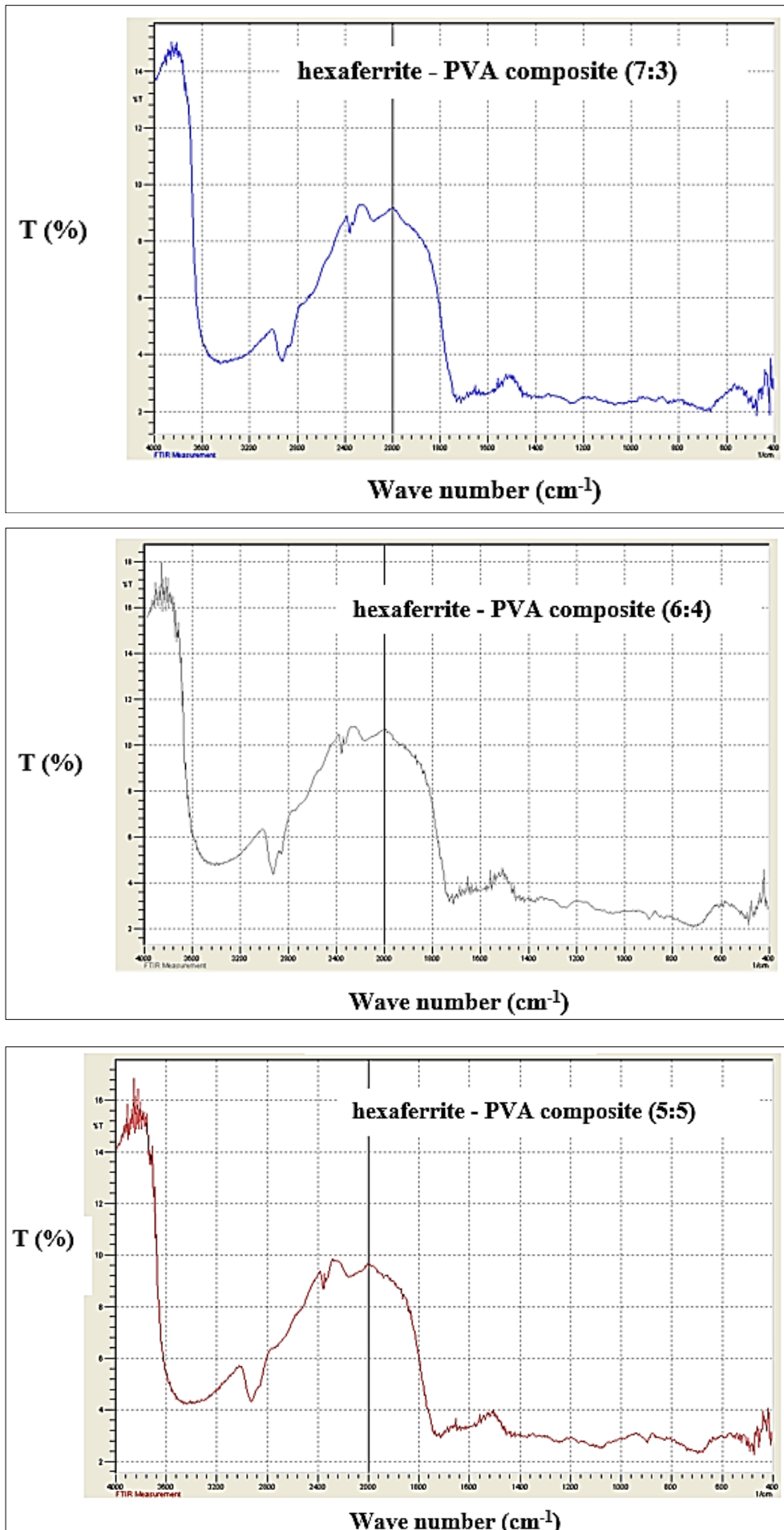


Fig 2: FTIR spectra of Ba₂Co₂Fe₂₈O₄₆ hexaferrite powder, PVA and composite samples with different mass ratio of ferrite to PVA.

Figure 2 shows FTIR spectra of calcined Ba₂Co₂Fe₂₈O₄₆ powder, PVA and composites with different mass ratio of ferrite to PVA (9:1, 8:2, 7:3, 6:4 and 5:5). The broad band observed in PVA sample at 3,300 cm⁻¹ may be assigned to O–H stretching due the strong hydrogen bond, the absorption bands around 2940 cm⁻¹, 1141 cm⁻¹ are the C–H alkyl stretching bands. The absorption band at approximately 1,731cm⁻¹ may be attributed to the stretching vibration of νC=O and the band observed at 850cm⁻¹ may be assigned to C–C stretching [20-22]. The existence of weak absorption bands in Ba₂Co₂Fe₂₈O₄₆ powder sample between 400 cm⁻¹ to 700 cm⁻¹ are due to stretching of M-O, which attributed to the formation of the ferrite phase [23, 24]. Composite samples show all absorption bands of PVA as well as weak band of ferrite at 440 cm⁻¹.

Dielectric properties

The dielectric measurements were carried out over the frequency range of 200 Hz to 2 MHz at room temperature using an Agilent Precision LCR meter (Model No. E4980A). Dielectric loss tangent (tan δ) of all the samples was calculated using equation (1)

$$\text{Tan } \delta = \frac{\epsilon''}{\epsilon'}$$
(1)

Where ε' is a real dielectric constant and ε'' is a complex dielectric constant. The variation of dielectric constant (real ε') and dielectric loss tangent (tan δ) as a function of frequency for composite samples with different mass ratio of PVA (from 9:1, 8:2, 7:3, 6:4 and 5:5) are shown in Figures 5(a) and (b), respectively.

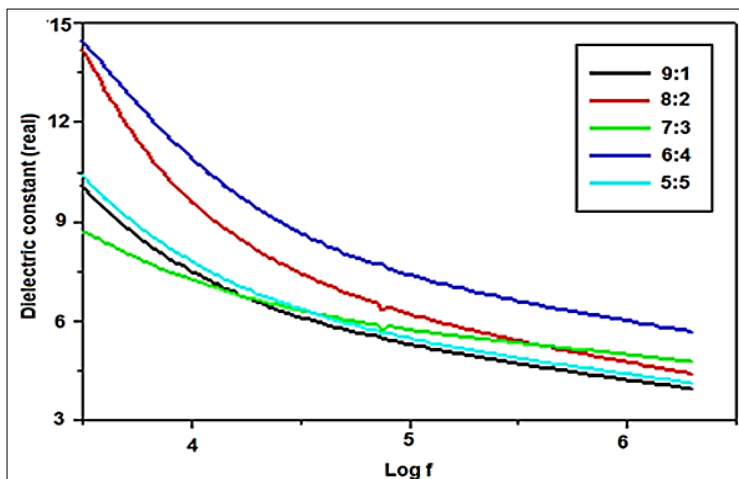


Fig 5a: The Variation of Dielectric Constant (real) with Log Frequency of Ba₂Co₂Fe₂₈O₄₆-PVA Composite Samples with Different Mass Ratio.

It is clear from Figures 4a, b that the value of dielectric constant (as well as dielectric loss tangent) decreases with increasing of frequency for all the samples. The variation of dielectric constant with frequency reveals the dispersion due to Maxwell-Wagner [26, 27] type interfacial polarization in agreement with Koop's phenomenological theory [28]. The dielectric properties of polycrystalline ferrite-polymer composites are depending upon two factors: the interfacial polarization and intrinsic electric dipole polarization. Interfacial polarization results from the heterogeneous structure of ferrites comprising low-conductivity grains

separated by higher resistivity grain boundaries [28]. These grain boundary layers may be attributed to the superficial reduction or oxidation of the crystals in the porous material as a result of their direct contact with the firing atmosphere. The higher value ε', for composite with mass ratio 6:4 may be due to the significant contribution of Co²⁺ ions in addition to Fe³⁺ and Fe²⁺ ions to interfacial polarization. The dielectric loss tangent represents the phase lag of the dipole oscillations with respect to the applied electric field, depends on the number and nature of ions present.

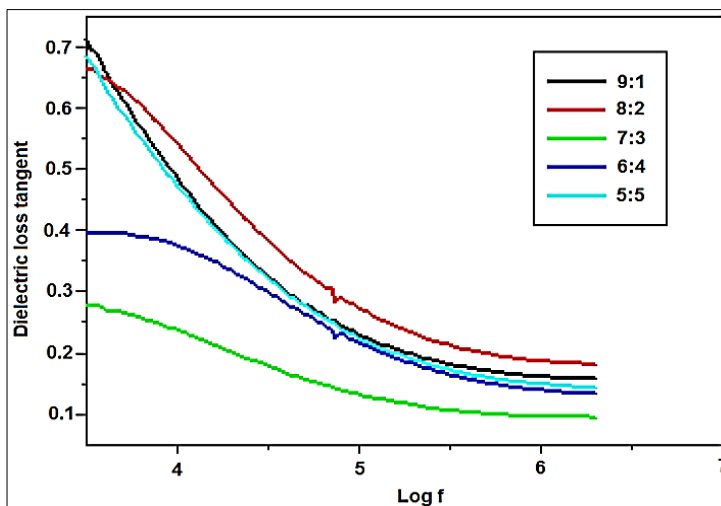


Fig 5b: The Variation of Dielectric Loss Tangent (tan δ) with Log Frequency of Ba₂Co₂Fe₂₈O₄₆-PVA Composite Samples for Different Mass Ratio.

Conclusion

Magnetic barium cobalt hexaferrite particles-PVA composites with of different mass ratio: 9:1, 8:2, 7:3, 6:4 and 5:5 were prepared. Structural properties of the composites were characterized by Infrared spectroscopy (FTIR), X-ray diffraction (XRD) and scanning electron microscope (SEM). The effect of the concentration of Ba₂Co₂Fe₂₈O₄₆ hexaferrites on structural and dielectric properties of magnetic-PVA composites were investigated. FTIR result revealed that composite samples show absorption peaks of both ferrites and PVA. The X-ray diffraction investigations confirm the formation of mono phase of hexaferrite particles. Scanning electron microscopy images reveal that the shape and size of barium cobalt hexaferrite powder particles is irregular. The addition of PVA not much changed the shape and size of particles (ranging from 200-300 nm) but porosity found to decrease.

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