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Credit Author statement

Amrin has prepared samples, prepared manuscript, data analysis, plotting graphs, FTIR and dielectric measurements, formal analysis, reply to reviewers, software

Robert C Pullar has checked manuscript fully, corrected, revised and time to time gave critical suggestions to improve the manuscript. English language and grammar are verified, conceptualization, visualization, funding acquisition

Sher Singh Meena has taken XRD, VSM, data analysis, revision and error analysis

Rajshree B. Jotania: supervision, methodology, writing- review and editing, investigation, data curation, administration

Khalid Mujasam Batoo: FE-SEM measurements, modify the draft including references, funding acquisition

All authors have read and agreed to the published version of the manuscript.

Studies of structural, magnetic and dielectric properties of X-type Barium Zinc hexaferrite $Ba_2Zn_2Fe_{28}O_{46}$ powder prepared by combustion treatment method using *ginger* root extract as a green reducing agent

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Abstract

Various quantities of *ginger* (Zingiber officinale) root extract were used to prepare X-type Barium-Zinc hexaferrite with the chemical composition $Ba_2Zn_2Fe_{28}O_{46}$. The powders were prepared using a combustion treatment method, being pre-heated at 550 °C for 4 h. with the *ginger* as a fuel, followed by final heating to 900 °C for 5 h. and natural cooling to room temperature to obtain $Ba_2Zn_2Fe_{28}O_{46}$ hexagonal ferrite powder. The phase composition of heated powder samples was investigated by X-ray diffraction (XRD), indicating the formation of a mixture of X-type and hematite (α-Fe_2O_3). Up to 82.6 wt%, X-ferrite was formed at 900 ºC with 52.5 g of *ginger* root extract. Dielectric analysis of the prepared samples shows the frequency-dependent phenomena. All samples were hard magnets, with coercivity values (H_C) between 262.2-318.3 kA m^{-1}, and squareness ratios > 0.5. The sample prepared with 52.5 g *ginger* root extract possesses the highest value of saturation magnetization (M_S = 33.87 Am^2 kg^{-1}) in comparison with the other prepared samples. Therefore, *ginger* was shown to be a useful natural plant extract as a reducing fuel for the low-temperature synthesis of X-ferrites. The sample prepared with 35 g ginger root extract shows a broad resonance peak between 10 kHz to 100 kHz, while other samples show resonance between 500 kHz to 1 MHz frequency range. At low frequencies (100 Hz to 2 MHz), relative permittivity was constant between 5 and 12 above 800 kHz for all X- ferrites.

Keywords: Green synthesis, X-type hexaferrite, *ginger* root extract, magnetic properties, and dielectric properties.

1. Introduction

Currently, sustainable fresh approaches that use green chemistry to improve and protect our environment are the main concerns in many areas of research. Preparation of novel magnetic materials has become very attractive due to their potential applications in radar absorbing materials (RAM), electronics [1, 2], high-density magnetic recording [3-6], biocompatible magnetic nanoparticles for cancer treatment [7-10] and magnetic resonance imaging (MRI) [11, 12]. The development of biodegradable and cost-efficient synthesis methods of nanomaterial remains a scientific challenge. Nanotechnology is also important to defeat the environmental issues caused by chemical industries such as oils [13], organic dyes [14], mercury, and wastewater. Common nanoparticle synthesis using chemical methods often involves harmful chemicals and solvents which may affect the human body and environment. Hence, biological methods using natural plants or plant extracts for the preparation of nanoparticles can be used as important substitutes for chemical methods [15-18].

There are a large number of methods used for the synthesis of nanoparticles such as co-precipitation [19], sol-gel [20, 21], microemulsion [22] and standard ceramic methods [23, 24], but these methods are often not environmentally friendly as reagents and solvents used in these chemical processes can be flammable or toxic. To overcome this, green synthesis of nanoparticles using eco-friendly materials like plant extracts [25], microorganisms such as fungi, yeast [26] and bacteria [27] is a low cost, and time reducing alternative. Different plants, roots, leaves, fruits, fruit peel, seeds, and their extracts have been used for the preparation of magnetic nanomaterials [25, 28-30].

Ginger (Zingiber officinale) is a flowering plant and its root; rhizome is used as a spice [31]. An active constitute called gingerol in it is responsible for most of the health benefits from ginger [32]. *Ginger* is the world's most cultivated herb from its origin to the present having good historic and medicinal value as a digestive aid, aphrodisiac, etc., over thousands of years [33]. *Ginger* remains an important ingredient of many traditional herbal remedies, as it has a history of multiple uses [34]. Many anti-inflammatory and antioxidant compounds such as capsaicin, beta-carotene, pantothenic acid, curcumin, caffeic acid, and salicylate are present in it. Also, active compounds like

shogaol, zerumbone, terpenoids, flavonoids, paradol, and zingerone present in ginger provide many health benefits [35].

Ginger can be used as a reducing agent for the preparation of nanomaterials as it possesses good antioxidant properties [36, 37, 38]. It has been reported that antioxidant properties are associated with the reducing power of biologically active compounds [39, 40], useful during combustion synthesis.

Antioxidant activity and components vary greatly and depend on the extracting solvent and its concentration [41]. ~~In this work, we used water to produce *ginger* root extract, adding to the sustainability of the process.~~ In comparison with other work, our approach is an appropriate, inexpensive, time reducing and eco-friendly route for the green synthesis of barium-zinc X-type hexaferrite. This procedure is easier compared to earlier reported, being prepared from metal nitrates in water. To the best of our knowledge, it is the first time that barium-zinc hexaferrite (along with some hematite) has been formed at 900 °C by using *ginger* extract as a reducing agent in reactions.

In the present study, we have used different weights of *ginger* root to prepare X-type hexaferrites ($Ba_2Zn_2Fe_{28}O_{46}$) by combustion treatment method and investigated the effect of the different amounts of ginger root on phase formation, as well as on magnetic and dielectric properties.

2. Experimental Procedure

2.1 *Ginger* extract preparation

Fresh *ginger* root (~ 200 g) was taken from the market and cleaned with distilled water. The *ginger root* was first peeled before cutting and then boiled for 45 minutes in 200 ml distilled water to get *ginger* root extract. The resulting extract solution was filtered using a filter paper.

2.2 X-type ferrite synthesis

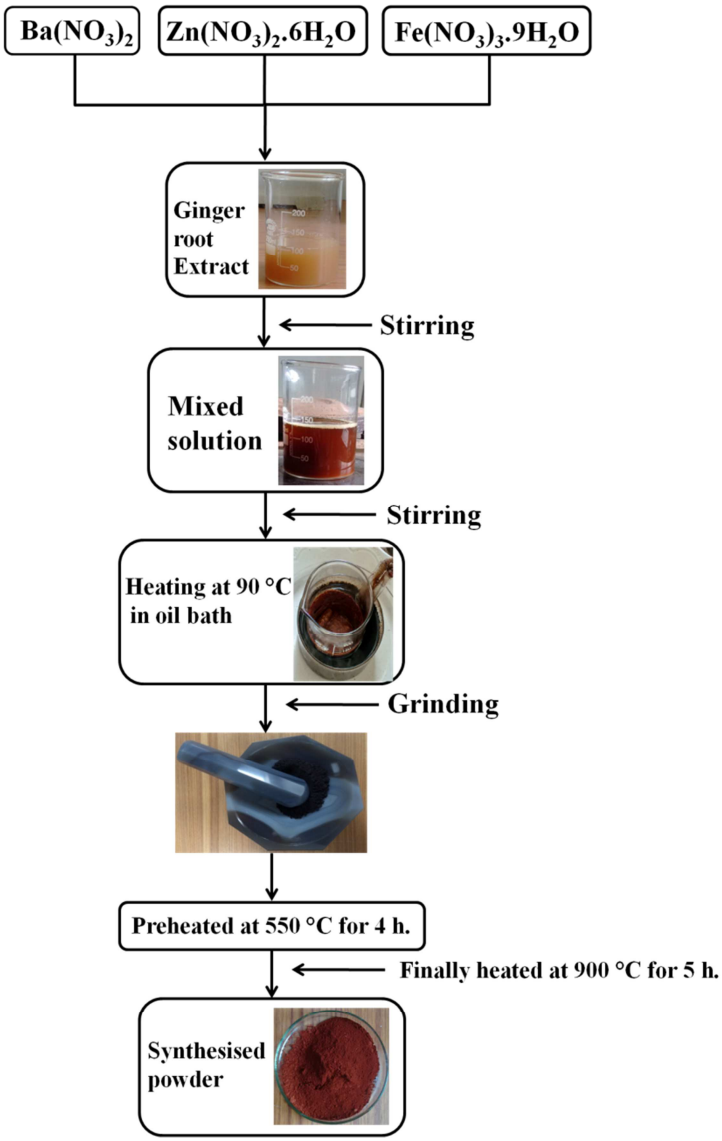

Fig. 1. Flowchart for the preparation of X-type hexaferrite $Ba_2Zn_2Fe_{28}O_{46}$ powder.

Stoichiometric quantities of barium nitrate $Ba(NO_3)_2$ (≥ 99.0% pure, Sigma–Aldrich), zinc nitrate $Zn(NO_3)_2.6H_2O$ (≥ 98.0% pure, Sigma–Aldrich) and ferric nitrate $Fe(NO_3)_3.9H_2O$ (≥ 98.0%, pure, Sigma–Aldrich) were dissolved one by one in a total solution of 100 ml of *ginger* extract. Precursor solutions were made with 0.0, 17.5, 35.0, 52.5, and 70.0 g of *ginger* root and in all cases a total of 51.79 g of metal salts were added. During this process, the solution was continuously stirred using a magnetic stirrer. The prepared mixture was heated at 90 °C in an oil bath to obtain a gel. The obtained gel was then dried in a muffle furnace at 100 °C for 20 h., to remove water. The resulting dried powder was ground in a mortar and pestle and then preheated at 550 °C for 4 h. The preheated powder was finally heated to 900 °C for 5 h., in a muffle furnace and allowed to cool naturally to room temperature to obtain

the $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite powder. Fig.1 shows the flowchart for the preparation of the X-type hexaferrite $Ba_2Zn_2Fe_{28}O_{46}$ powder. Heated samples were coded as Sample (A), Sample (B), Sample (C), Sample (D) and Sample (E) for the $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite prepared with 17.5, 35.0, 52.5, 70.0 and 0.0 g of *ginger* root extract, respectively.

2.3 Characterisation

A FTIR spectrometer (Bruker Tensor 27 Model) was used to record infrared (IR) spectra of all synthesised samples over the range of 4000–400 cm^{-1} at room temperature. KBr pellet method was used to record IR spectra. X-ray diffraction (XRD) technique was used to identify the crystalline phase formation, using a Rigaku X-ray diffractometer with Cu - K_α radiation (λ = 1.5406 Å), and the 2θ scanning range was 20°- 80° with equal steps of 0.02°. The morphology of all samples was investigated using a Nano Nova 450 Field Emission Gun Scanning Electron Microscope (FEG-SEM). The magnetic hysteresis loops were recorded at room temperature under an applied field of ±1.0 T using a vibrating sample magnetometer (EG & G Princeton Applied Research instrument, Model 4500) The low-frequency dielectric measurements were carried out at room temperature over a frequency range of 100 Hz to 2 MHz using a Precision LCR meter (Agilent E4980A). The pellets of sintered ferrite powder were made using a hydraulic press for low-frequency dielectric measurements and density measurements.

3. Results and discussions

3.1 FTIR Analysis

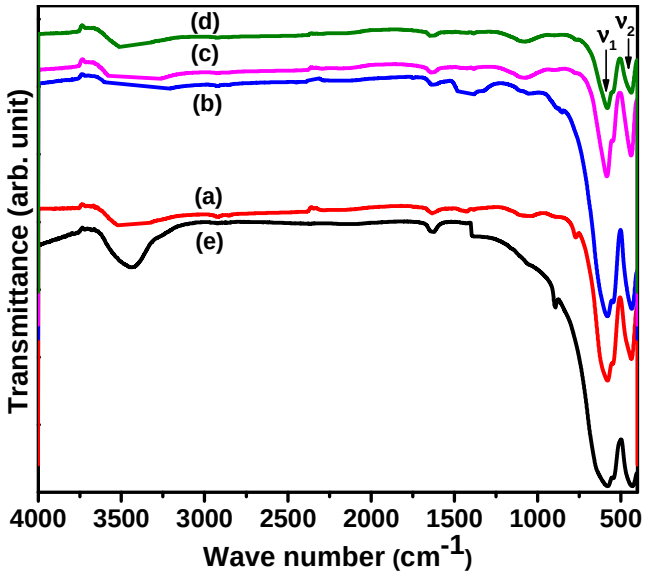

Fig. 2. FTIR spectra of X-type $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with and without the presence of different weight ratios of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D), and (e) Sample (E).

Fig. 2 shows the FTIR spectra of $Ba_2Zn_2Fe_{28}O_{46}$ powder synthesised with and without the presence of *ginger* root extract heated at 900 °C for 5 h (Sample (A-E)). The absorption bands in the powder at 584 cm^{-1} and 434 cm^{-1} are assigned to the tetrahedral (ν_1) and octahedral (ν_2) metal-oxygen bond vibrations of the crystal lattice, and are characteristic of hexagonal ferrites [42]. Absorption bands in the range 434-439cm^{-1} (ν_2) are assigned to the Fe-O stretching of Fe-O_6, while, the bands in the range 580-585cm^{-1} (ν_1) are assigned to the Fe-O stretching of Fe-O_4 [43]. All heated samples show a small absorption band at ~1632 cm^{-1}, indicating the O-H stretching vibrations which may be due to the polyol [44]. However, the sample prepared without ginger root extract shows a broad (strong) absorption peak between 3300-3600 cm^{-1}, which is due to the stretching vibration of an O-H bond [45, 46], and is typical of water. The fact that this is absent in the samples with *ginger* suggests that the sample without *ginger* (Sample (E), Fig. 2e) is much more hygroscopic and has absorbed atmospheric moisture much more rapidly after cooling.

3.2 XRD analysis

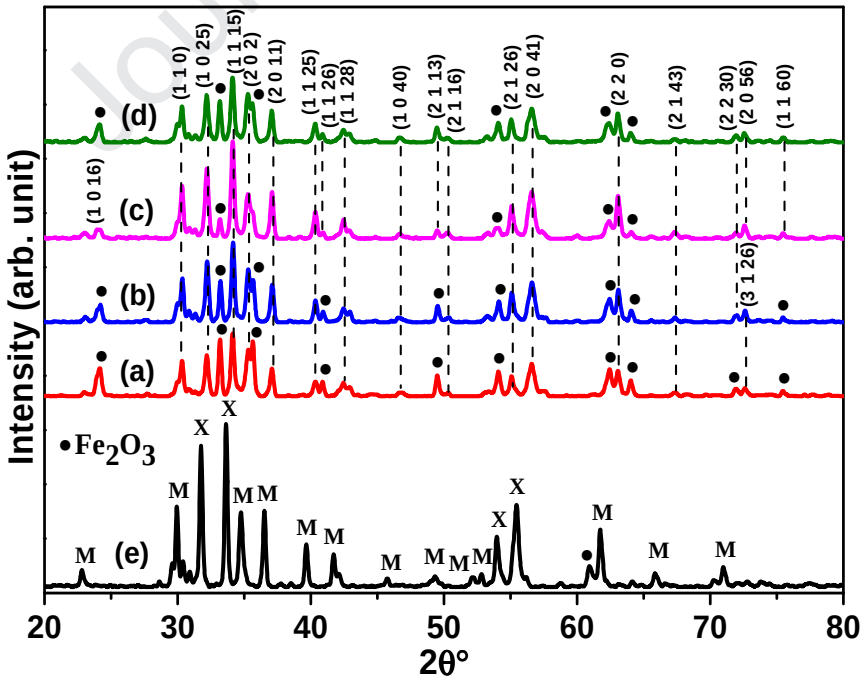

Fig. 3. XRD patterns of X-type $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with and without the presence of different weight ratios of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D), and (e) Sample (E).

XRD patterns of X-type $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples were indexed using the standard patterns for X-type ($Ba_2Me_2Fe_{28}O_{46}$) hexagonal crystals (JCPDS # 01-073-2034), and are shown in Fig. 3. XRD analysis of the samples, prepared in the presence of different weight ratios of *ginger* root extract and heated at 900 °C for 5 h. revealed the formation of X-type ferrite and hematite (α-Fe_2O_3) (JCPDS # 04-015-9569) as a minor secondary phase. However, the sample prepared without *ginger* root extract (Sample (E)) shows the formation of a mixture of phases: 72.2% of M-type ferrite ($BaFe_{12}O_{19}$) (JCPDS # 84-0757), only 22.2% X-type, and 5.6% hematite. Therefore, the addition of the *ginger* extract has enabled the formation of a single hexaferrite phase (X-type) at a low temperature of only 900 ºC, with non-magnetic α-Fe_2O_3 as a minor secondary phase. The phase composition values are given in Table 1.

It can be seen from Fig. 3 that the sample with 52.5 g weight of *ginger* root extract (Sample (C), Fig. 3c) contained the lowest amount of hematite compared to the others, with 82.61 % X ferrite, while the quantity of hematite increased again when the weight ratio of *ginger* root extract increased to 70 g (Sample (D), Fig. 3d), yielding 75.0 % X ferrite. Therefore, the XRD patterns confirmed that 52.5 g weight of *ginger* root extract (Sample (C), Fig. 3c) was the optimum for the synthesis of X-type ($Ba_2Zn_2Fe_{28}O_{46}$) hexaferrites.

The variation of lattice constants and unit cell volume with different weight ratios of *ginger* root extract is shown in Fig. 4 and Table 1. The average crystallite size was investigated from the most intense peak using the following Scherrer formula [47].

$$D_{(xrd)} = \frac{K\lambda}{\beta cos\theta} \quad (1)$$

Where K is a dimensionless shape factor and has a value about of 0.9 which varies with the actual shape of the crystallite; λ is the X-ray wavelength (1.5406 Å); θ is the angle of Bragg diffraction, and β is the full width at half the maximum intensity. The obtained structural parameters are listed in Table 1. The crystallite size of all samples, heated at 900 °C for 5 h. varies in the range of 27-37 nm.

The unit cell volume was calculated using the following equation.

$$V = a^2 c \sin 120° \quad (2)$$

Where '*V*' is the volume of the unit cell and '*a*' and '*c*' are the lattice constants.

The crystallite size was ~37 nm without *ginger* extract, and ~30 nm in all cases with *ginger* extract, suggesting that the reducing agent had the effect of slightly lowering crystallite size in the ferrites produced. Similarly, the lattice parameters and cell volume were larger, and similar, for all samples produced with *ginger* extract compared to that without.

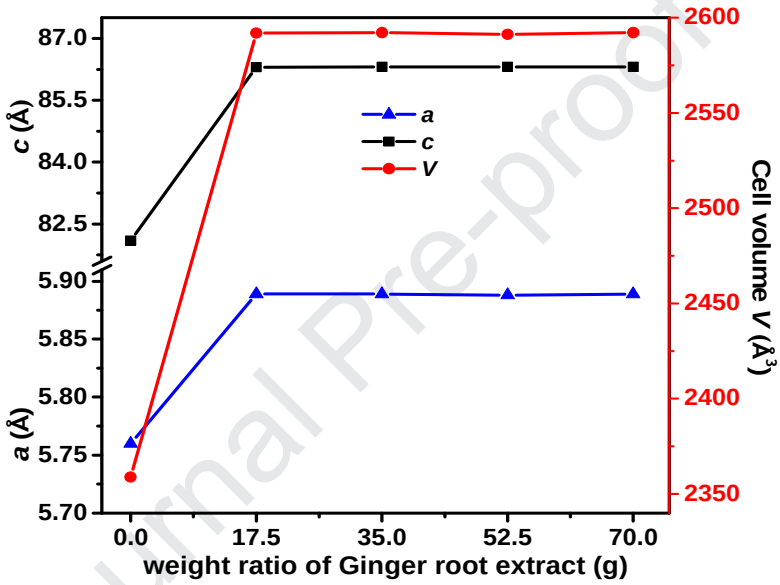

Fig. 4. Variation of Lattice constants (*a*, *c*) and unit cell volume (*V*) of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples prepared without *ginger* root extract and with different weight ratios of *ginger* root extract heated at 900 °C for 5 h. (Sample (A-D))

Table 1. Structural parameters of X-type $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples prepared without and with the different weight ratio of *ginger* root extract and heated at 900 °C for 5 h.

Code	Lattice parameters		Cell volume *V* (Å^3) (±5)	Crystallites size D_{xrd} (nm) (±0.2)	FWHM (deg.)	% of X-phase (±0.1)	% of α-Fe_2O_3 (±0.1)	% of M-phase (±0.1)
	a (Å) (±0.02)	*c* (Å) (±0.2)						
Sample (A)	5.889	86.303	2592.03	29.30	0.2964	58.33	41.67	-
Sample (B)	5.889	86.308	2592.18	31.34	0.2771	62.50	37.50	-
Sample (C)	5.888	86.310	2591.35	30.15	0.2880	82.61	17.39	-
Sample (D)	5.889	86.310	2592.24	27.91	0.3111	75.00	25.00	-
Sample (E)	5.760	82.10	2358.88	36.95	0.2350	22.22	5.56	72.22

The values of Bulk density (d_B) and X-ray density (D_x) were calculated using the following equations [48].

$$d_B = \frac{m}{\pi r^2 h} \quad (3)$$

$$D_x = \frac{3M}{Na^2c} \quad (4)$$

Where m is mass of pellet, r is a radius of pellet, h is the height (thickness) of pellet, M is molecular weight, a and c are lattice constants, and N is Avogadro's number (6.02 x 10^{23}/mole).

The value of porosity for all samples is calculated using the following equation.

$$P = (1 - \frac{d_B}{D_x}) \times 100\ \% \quad (5)$$

The calculated values of X-ray, bulk density and porosity of all the samples heated at 900 °C for 5 h. are shown in Table 2. The value of bulk density varies in the range of 1.90 - 3.30 g/cm^3 (Table 2). The sample prepared with 52.5 g *ginger* root extract (Fig. 3c), that contained the most X-ferrite phase, also had the minimum value of porosity and maximum value of bulk density. However, as these pellets were only heated to 900 ºC, hence they are very poorly sintered and are not close to the maximum density possible (5.313 g/cm^3). The calculated X-ray densities were higher, at ~ 4.50 g/cm^3 (see Table 2).

Table 2. The bulk density, X-ray density, and the porosity of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h.

Code	Bulk density d_B (g/cm^3) (± 0.01)	X-ray density D_x (g/cm^3) (± 0.02)	Porosity P (%) (± 0.1)
Sample (A)	1.92	4.502	57.40
Sample (B)	3.16	4.502	29.79
Sample (C)	3.26	4.504	27.60
Sample (D)	2.87	4.502	36.25

Fig. 5 shows the variation in bulk density (d_B), X-ray density (D_x) and porosity (P) with the different weight ratios of *ginger* root extract for the samples heated at 900 °C for 5 h.

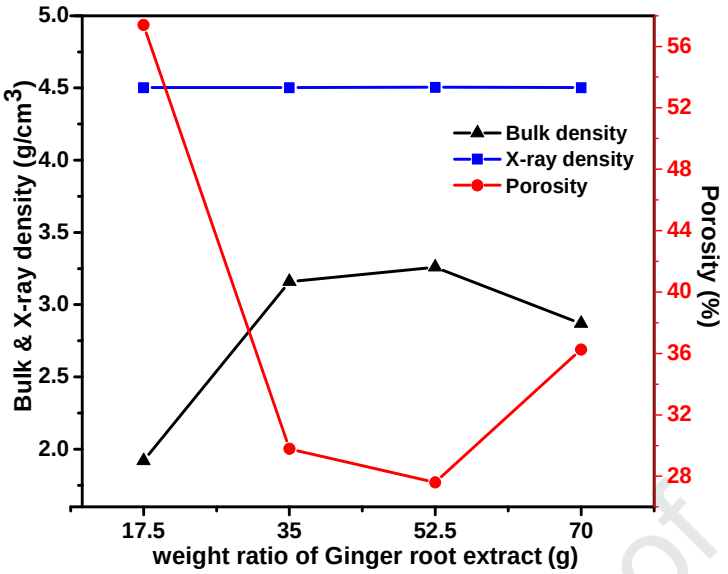

Fig. 5. Variation of the Bulk density, X-ray density, and porosity of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples prepared with the different weight ratios of *ginger* root extract heated at 900 °C for 5 h. (Sample (A-D))

3.3 Surface Morphology

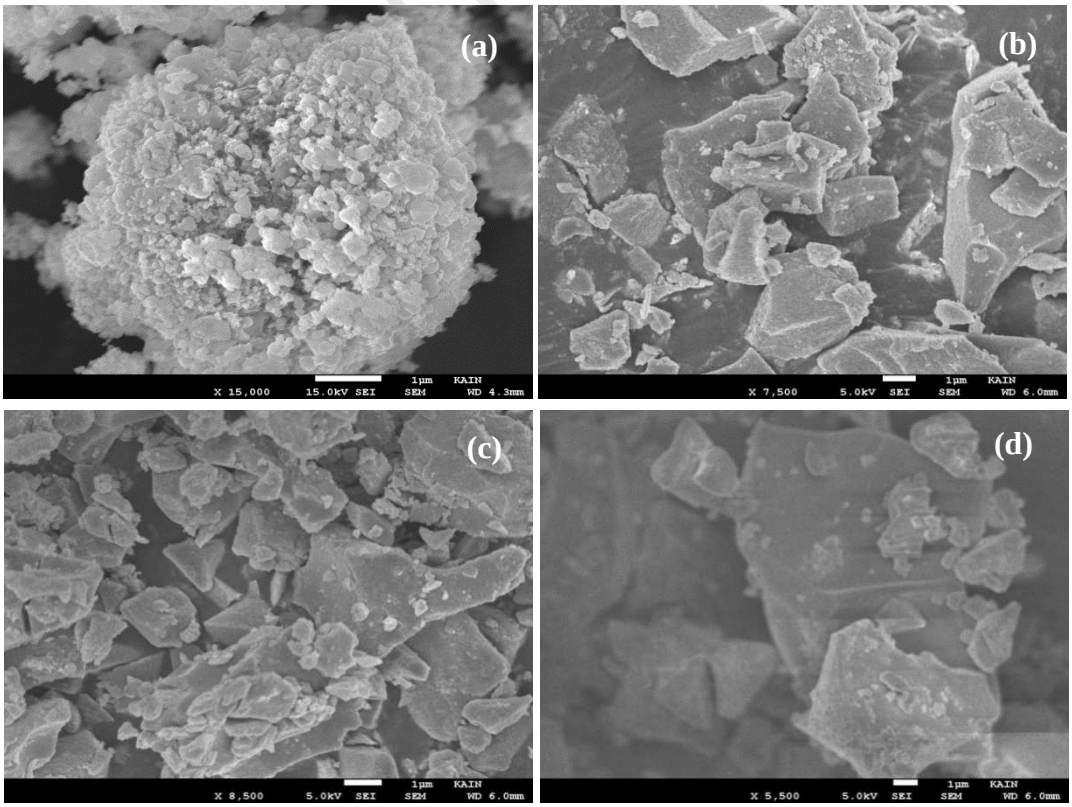

Fig. 6. SEM images of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the presence of different weight ratios of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

Fig.6 shows the SEM micrographs for all the samples (Sample (A-D)) showing the grain morphology of prepared hexaferrites. It is observed from micrographs that the grains are agglomerated and are irregular in shape. As the weight ratio of *ginger* root extract increases, porosity decreases, and agglomeration increases. The sample prepared in presence of 17.5 g *ginger* root extract (Sample (A), Fig. 6a) possesses spongy structure and it also possesses a high value of porosity (57.40 %) and low value of bulk density (1.92 g/cm^3) compared to all other samples.

3.4 Magnetic properties

The hysteresis loops of $Ba_2Zn_2Fe_{28}O_{46}$ samples prepared in the presence of *ginger* root extract, heated at 900 °C were recorded on a VSM under an applied field of ± 1.0 T and shown in Fig. 7. The different magnetic parameters such as saturation magnetisation (M_S), remanent magnetisation (M_r), coercivity (H_C), and squareness ratio (M_r/M_S) of prepared hexaferrites are calculated from the M-H loops and their values are listed in Table 3.

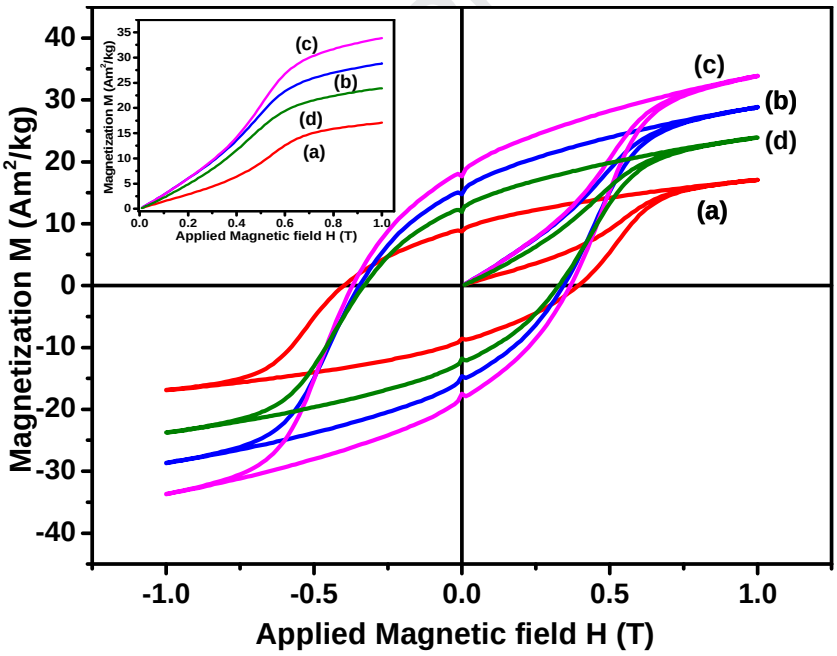

Fig.7. M-H loops and initial magnetization curves (inset) of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

Table 3. Magnetic parameters of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrites prepared with the different weight ratios of *ginger* root extract heated at 900 °C for 5 h.

The values of saturation magnetisation (M_S) and remanent magnetisation (M_r)

Sr. No.	Code	Remanence Magnetization M_r (Am^2 kg^{-1})	Saturation Magnetization M_S (Am^2 kg^{-1})	Squareness ratio M_r/M_S	Coercivity H_C (T)	H_a (kA m^{-1})	H_a (Oe)
1	Sample (A)	8.75	17.08	0.5122	0.40	149.71	1881.25
2	Sample (B)	14.75	28.86	0.5110	0.35	156.73	1969.47
3	Sample (C)	17.66	33.87	0.5214	0.38	147.17	1849.34
4	Sample (D)	11.92	20.92	0.5698	0.33	108.10	1358.38

increase as the weight ratio of *ginger* root extract increases except for the sample prepared with 70.0 g *ginger* root extract (Sample (D)), that had a lower X-ferrite content than that with 52.5 g, as shown in Table 3. The value of coercivity lies in the range of 262.6-318.3 kA m^{-1} (0.33-0.40 T = 3300-4000 Oe) in all cases, which shows that the prepared samples belong to the family of hard ferrites [49], as would be expected for $Ba_2Zn_2Fe_{28}O_{46}$. The variation of saturation magnetisation (M_S), remanent magnetisation (M_r) and coercivity (H_c) with the different weight ratios of *ginger* root extract are shown in Fig. 8 (a).

In the present study, the sample prepared with 52.5 g *ginger* root extract (Sample (C)) possesses the highest value of saturation magnetisation (M_S = 33.87 Am^2 kg^{-1} or emu/g) in comparison with the remaining prepared samples, that is because this sample exhibits the maximum amount of X-phase (79.16 %). The variation in M_S is matching with X-phase presented in the sample as shown in Fig. 8 (b). This M_S value (33.87 Am^2 kg^{-1} or emu/g) is particularly low in comparison with the reported maximum value of saturation magnetisation (M_S = 73.1 Am^2 kg^{-1} or emu/g) [50]. The low value of M_S is partially attributed to the presence of the antiferromagnetic phase of hematite (17.39 % of α- Fe_2O_3) [51, 52], as confirmed by XRD analysis (Fig. 3), as a sample containing only 82.61 wt % X ferrite would be expected to have a M_S value of 57.75 Am^2 kg^{-1}. The fact that the value here is still only around 2/3 of this value could be explained by the low synthesis temperature used here – magnetisation is known to increase as synthesis/sintering temperature increases, and the low crystallite size seen in these samples is well below the typical maximum magnetic domain sizes reported for hexaferrites of between 0.5-1 μm [2]. The squareness ratio (M_r/M_S) is also calculated from the values of M_r and M_S and their values are tabulated in Table 3. Previously, it has been reported that the squareness ratio of hexaferrite samples at or above 0.5 shows the material possesses a single magnetic domain and below 0.5 shows the formation of a multi-

domain structure. In the present study, the squareness ratio of all the samples is in the range of 0.510 - 0.570 (Table 3) showing that all the samples possess a single domain structure.

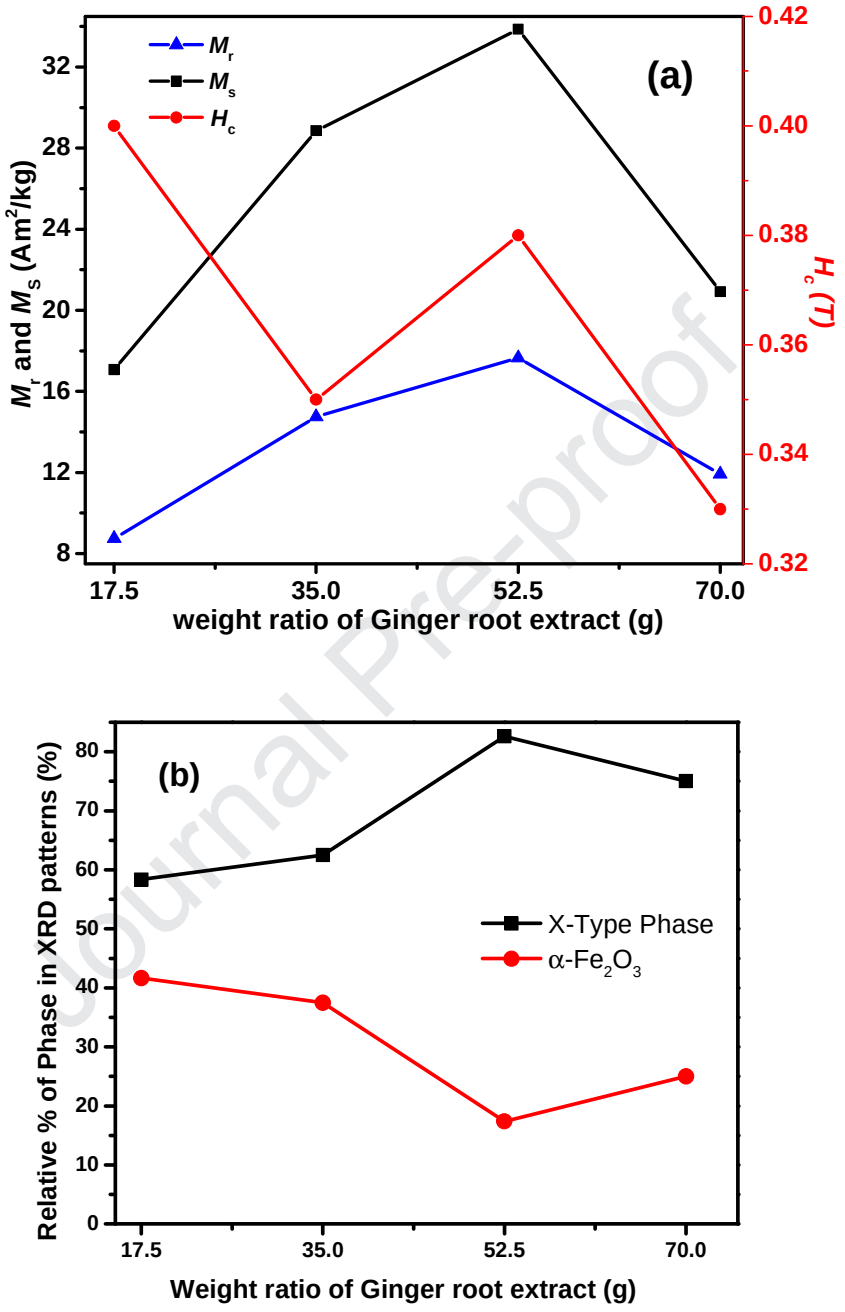

Fig. 8. (a) Variation in saturation magnetization (M_S), remanent magnetization (M_r), (H_c), and, **(b)** Relative percentage of two phases of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples prepared with the different weight ratios of *ginger* root extract heated at 900 °C for 5 h. (Sample (A-D)).

3.5 Dielectric properties

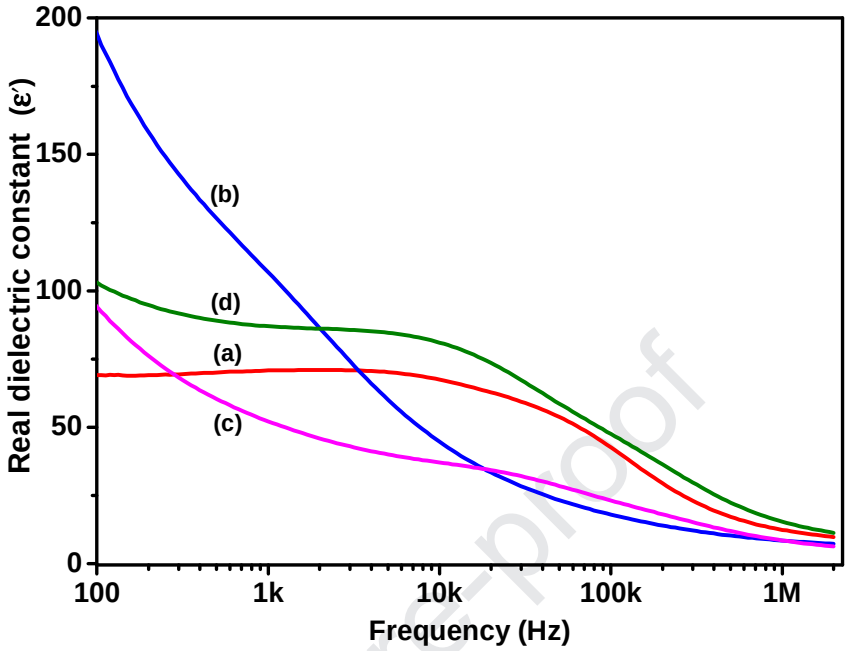

Fig. 9. Variation in real dielectric constant (ε′) of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

It is observed from Fig. 9 that at low-frequency, the dielectric constant of the samples prepared with 35 g *ginger* root extract (Sample (B)) decreases rapidly and approaches a constant above 1 MHz, while the other samples (Sample (A), (C) and (D)) show lower initial permittivity, and resonance peaks at around 10-50 kHz. This decrease in dielectric constant with frequency is a normal behaviour of most ferri-magnetic materials and has also been observed by several other researchers [53-55]. This behaviour of dielectric dispersion can be explained based on Koop's phenomenological theory, which is based on the Maxwell-Wagner interfacial polarisation model [56]. According to this model, the dielectric medium of ferrite consists of two layers. The first layer of fairly well- conducting boundaries is separated by the second layer of relatively poor conducting boundaries. The first layer is strongly effective at higher frequencies, while the second layer is dominant at lower frequencies. The free electrons reach the grain boundaries through hopping [57] and 'pile-up' at the grain boundaries if the grain boundaries' resistance is high enough and, hence, produce induced polarisation. In ferrites, the polarisation is a similar process to that of the conduction process. The free electrons in a dielectric medium need a finite time to line up their axes in the

direction of the applied field. If the frequency of the applied field is increased, free electrons cannot align with the applied field at a certain point and hence cannot follow the changes in the applied field over a certain frequency range [58]. As a result, the probability of free electrons reaching grain boundaries is deceased and the free electron polarisation virtually does not contribute to the polarisation that decreases the dielectric constant [56, 59]. At 1 MHz and above, all of the X ferrites have similar permittivity values of ~10-12.

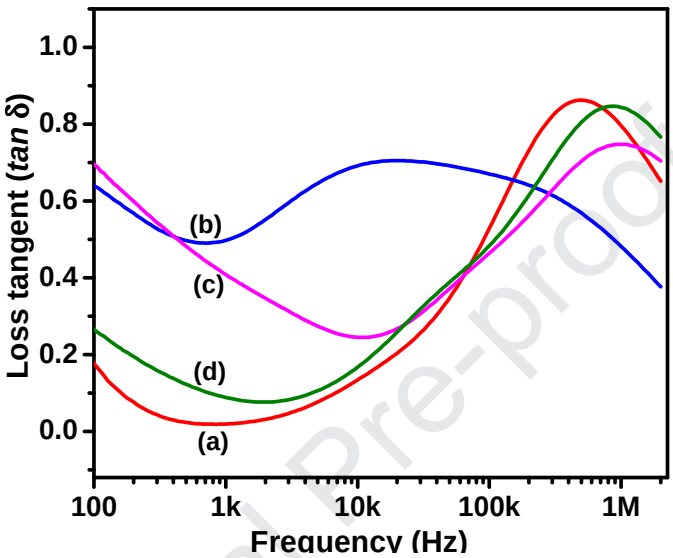

Fig. 10. Variation in loss tangent (*tan δ*) as a function of frequency for $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

Fig.10 shows the variation of loss tangent (*tan* δ) of all the prepared samples (Sample (A-D)) with frequency at room temperature. The value of loss tangent depends on different factors such as Fe^{+2} content, structural homogeneity, stoichiometry ratio, preparation time and heating temperature [60]. The value of loss tangent decreases with increasing frequency, that is because beyond a certain frequency limit hopping frequency of charge between Fe^{3+} and Fe^{2+} cannot follow the frequency of the applied electric field. The sample prepared with 35 g ginger root extract (Sample (B)) shows a broad resonance peak between 10 kHz to 100 kHz, while other samples show resonance between 500 kHz to 1 MHz frequency range.

AC conductivity (σ_{ac}) of the samples was calculated by the formula [61];

$$\sigma_{ac} = 2\pi f \varepsilon_0 \varepsilon'' \quad (6)$$

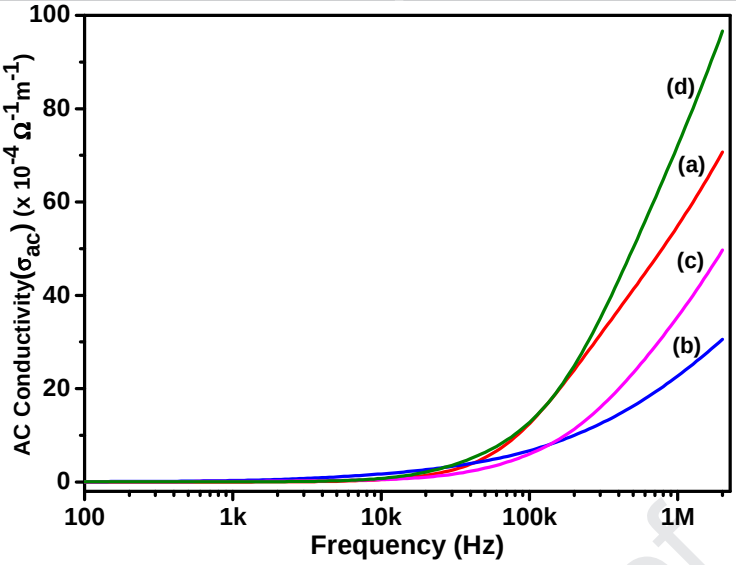

Fig. 11. AC conductivity of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

The variation of ac conductivity of all the hexaferrite samples (Sample (A-D)) with frequency (100 Hz - 2 MHz) at room temperature is shown in Fig. 11. It can be observed that ac conductivity increases with the increase of frequency, which is because increment in frequency increases the hopping frequency of the charge carriers between Fe^{2+} and Fe^{3+}.

3.5.1 Frequency-dependent impedance

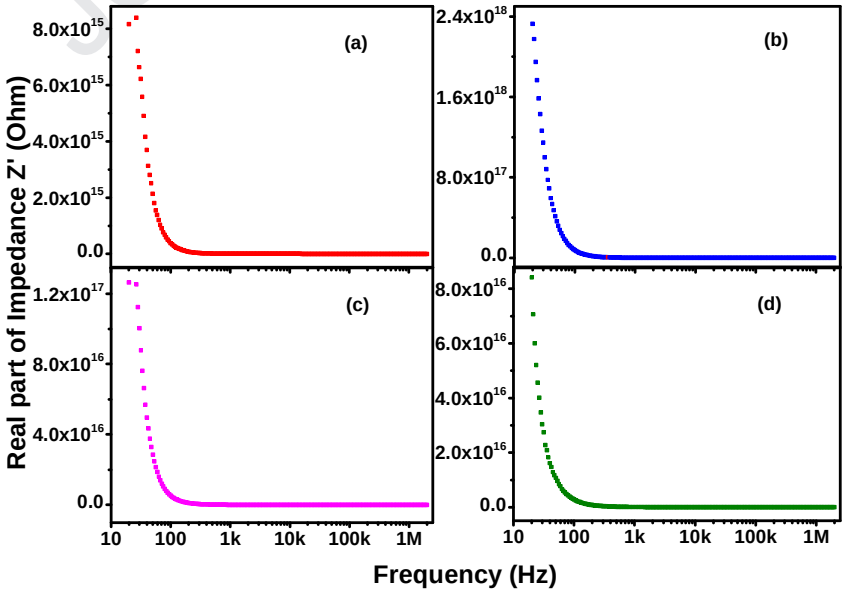

Fig. 12. Variation of the real part of impedance (Z') with frequency for X-type $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

The real part of impedance (Z') and complex part of impedance (Z") as a function of applied field frequency in the range of 10 Hz to 2 MHz for X-type, $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h. measured at room temperature are shown in Fig. 12 and Fig. 13 respectively.

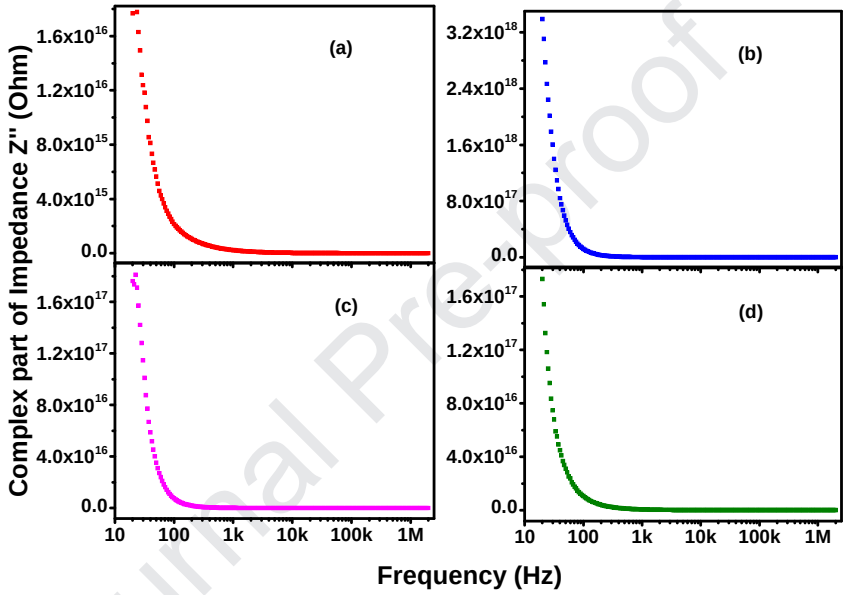

Fig. 13. Variation of the complex part of impedance (Z") with frequency for X-type, $Ba_2Zn_2Fe_{28}O_{46}$ samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

It can be seen from Fig. 12 and Fig. 13 that the real part of impedance (Z') and complex part of impedance (Z") for all different weight ratios of *ginger* root extract decreases fast as frequency increases. Their values become almost constant beyond 300 Hz frequency and approach to nearly zero, that shows no dependency of Z' and Z" on frequency.

3.5.2 Electric Modulus analysis

Analysis of electric modulus is important to understand the various parameters of electrical transport performance such as the rate of ion hopping and conductivity relaxation time contribution to the conductivity of the prepared hexaferrite material [62].

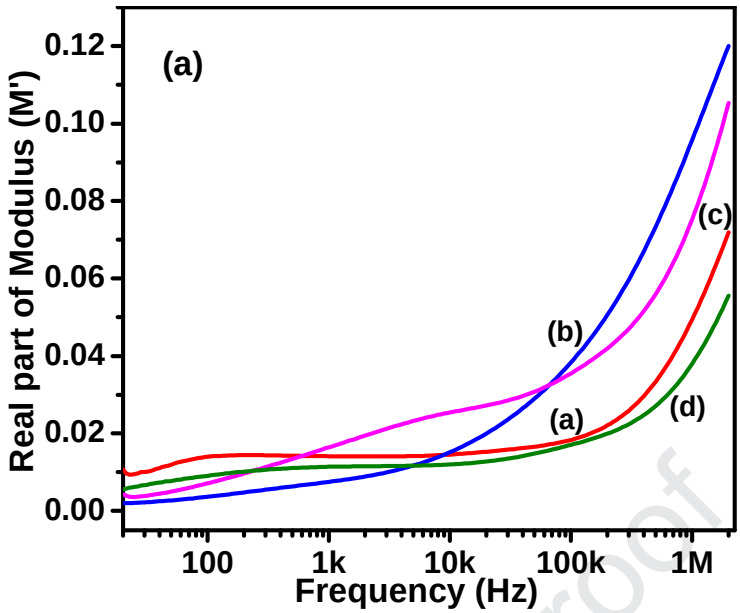

Fig. 14 (a). Variation in the real part of electric modulus as a function of frequency for $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

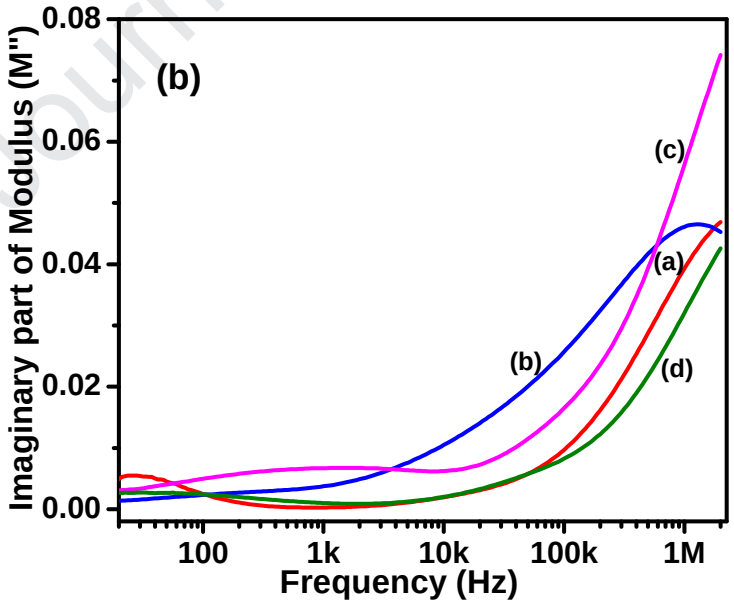

Fig. 14 (b). Variation in the imaginary part of electric modulus as a function of frequency for $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

Fig. 14 (a) shows the variation of the real part of the electric modulus (M') of samples heated at 900 °C as a function of frequency. It can be observed from Fig. 14 (a) that at low frequency, the value of real electric modulus (M') of all samples prepared in presence of ginger root extract (Sample (A-D)) is low and at the high-frequency region, it increases very fast. When the electric field is applied, it decreases the restoring force between the charge carriers, which is responsible for the high value of M' at the high-frequency region [63]. The maximum value of real modulus (M'~0.12 at 2 MHz) is observed in the sample prepared with 35.0 g *ginger* extract.

The imaginary part of the electric modulus (M") for all samples heated at 900 °C (Sample (A-D)) is shown in Fig. 14 (b). The present graph explains the brief information about the charge transport mechanism, such as ion dynamics, conductivity relaxation and electrical transport mechanism as a function of the frequency [62]. The maximum value (M" ~0.075 at 2 MHz) is observed in the sample prepared with 52.5 g ginger root extract (Sample (C)). The relaxation peak is observed around 1 MHz in the sample prepared with 35.0 g *ginger* root extract (Sample (B)) due to the mobility of charge carriers. Below the relaxation peak, the charge carriers are mobile and above the relaxation peak, the charge carriers are immobile, which confined to a potential well [64].

Cole-Cole type plots provide information about the grains, grains boundary effects; are plotted and shown in Fig. 15. The Cole-Cole plot type (Nyquist plot) of electric modulus analysis is more helpful than impedance analysis (Z" versus Z'). This analysis accurately separates the relaxation effects from grain and grain boundary in ferrite material. The imaginary part of impedance (Z") gives the relaxation dynamics from largest resistance of the material, while, the imaginary part of the modulus (M") gives the smallest capacitance (i.e. the highly conductive part) of the material [65]. It can be observed from Fig. 15 that all samples (Sample (A-D)) show semicircle arc in the low-frequency region attributed to the grain resistance [66]. The sample prepared with 52.5 g ginger root extract shows semicircle at the low-frequency region, that indicates the relaxation phenomena with different relaxation time (τ), associated with each relaxation. The capacitance values can be calculated at the maximum frequency (f_{max}) using the following relation [67]:

$$M'' = (\varepsilon_o / 2C) \quad (7)$$

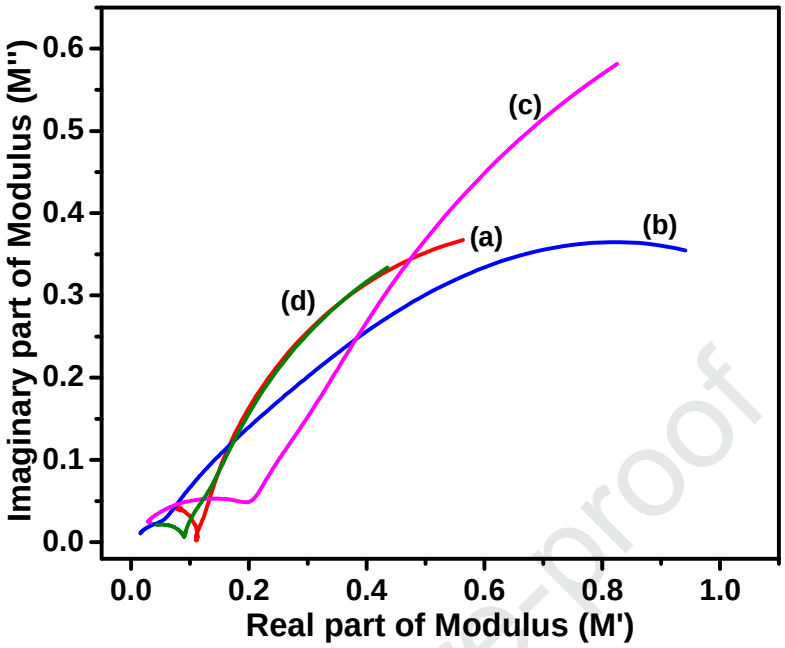

Fig. 15. Cole-Cole type plots of $Ba_2Zn_2Fe_{28}O_{46}$ hexaferrite samples heated at 900 °C for 5 h., prepared with the different weight ratio of *ginger* root extract: : (a) Sample (A), (b) Sample (B), (c) Sample (C), (d) Sample (D).

Conclusions

X-type Barium zinc hexaferrite $Ba_2Zn_2Fe_{28}O_{46}$ powder has been successfully synthesized using the combustion treatment method at 900 °C for 5 h. in presence of different weight ratios of *ginger* root. From XRD analysis it has been found that the sample with 52.5 g *ginger* root, heated at 900 °C contained less impurity of hematite peak (17.39 %) compared to other samples heated at 900 °C. The sample prepared with 52.5 g *ginger* root extract possesses the highest value of saturation magnetization (M_S = 33.87 $Am^2 kg^{-1}$) in comparison with the other prepared samples, and all were hard ferrites. At low frequencies (>2 MHz), relative permittivity was constant between 5 and 12 above 800 kHz.

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Highlights of the work

- The effect of the different amounts of ginger root extract on phase formation, as well as on magnetic and dielectric properties of $Ba_2Zn_2Fe_{28}O_{46}$ has been investigated.
- Barium-zinc hexaferrite (along with some hematite) has been formed at 900 °C by using *ginger* extract as a reducing agent in reactions.
- Prepared X-ferrites were hard magnets, with coercivity values (H_c) between 108-157 kA m^{-1}, and squareness ratios > 0.5.
- 52.5 g *ginger* root gave the highest saturation magnetisation (33.87 Am^2 kg^{-1}), showing it to be a useful natural plant extract as a reducing fuel for the low temperature synthesis of X-ferrites.
- 35 g ginger gave a broad resonance peak at 10 kHz - 100 kHz, while others showed resonance at 500 kHz - 1 MHz.
- Low frequency relative permittivity was between 5-12 at 800 kHz – 2 MHz for all X ferrites.

Declaration of interest

The authors declare that they have not known to competing financial interest or personal relationship that could appeared to influence to work reported to this paper.

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