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Manuscript Draft

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Title: Bio-silica incorporated barium ferrite composites: Evaluation of structure, morphology, magnetic and microwave absorption traits

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Keywords: bio-silica; barium ferrite; magnetic properties; permittivity; permeability; reflection loss

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Abstract: A series of bio-silica ion incorporated barium-ferrite-composites with the composition of (x)Bio-SiO2:(80-x)Fe2O3:(20)BaO, where x = 0, 1, 2, and 3 wt% were prepared using the modified solid-state reaction method. The influence of different bio-silica (extricated from sintered rice husk) contents on the surface morphologies, structures, and magnetic characteristics of these composites were assessed. The relative complex permittivity and permeability were resolved using the Nicholson-Ross-Weir strategy in the frequency range of 8□13 GHz. Meanwhile, the reflection loss was estimated through the transmission/reflection line theory to assess the MW absorption properties of the composites. Incorporation of the bio-silica in the barium ferrite composites generated a new hexagonal phase (Ba3Fe32O51) and a tetragonal phase (BaFeSi4O10) which led to a decrease in the saturation magnetization and significant shift in the MW frequency absorption peak positions.

Suggested Reviewers:

Ethical Statement for Current Applied Physics

Hereby, I (Wahyu Widanarto, Mukhtar Effendi, Sib Krishna Ghoshal, Candra Kurniawan, Erfan Handoko, Mudrik Alaydrus) consciously assure that for the manuscript **Bio-silica incorporated barium ferrite composites: Evaluation of structure, morphology, magnetic and microwave absorption traits** the following is fulfilled:

1) This material is the authors' original work, which has not been previously published elsewhere.

2) The paper is not currently being considered for publication elsewhere.

3) The paper reflects the authors' own research and analysis in a truthful and complete manner.

4) The paper properly credits the meaningful contributions of co-authors and co-researchers.

5) The results are appropriately placed in the context of prior and existing research.

6) All sources used are properly disclosed (correct citation). Literally copying of text must be indicated as such by using quotation marks and giving a proper reference.

7) All authors have been personally and actively involved in substantial work leading to the paper and will take public responsibility for its content.

The violation of the Ethical Statement rules may result in severe consequences.

I agree with the above statements and declare that this submission follows the policies of Current Applied Physics as outlined in the Guide for Authors and in the Ethical Statement.

Date: 27/10/2019

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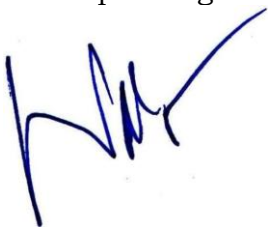


The Editor

Current Applied Physics

Manuscript Number: CAP-D-19-01273

Dear Sir,

Thanks to the reviewers for critical reading of the manuscript, providing invaluable comments and suggestions to modify it substantially. Please find attached herewith the responses to the reviewer's comments point by point on the paper. The authors gratefully acknowledge the invaluable comments and advise by the reviewer. The reviewer comments are presented in *italics (BLACK)* and the corresponding replies as standard text in BLUE below each comment. The newly added texts are highlighted in YELLOW in the revised manuscript.

Reviewer's Comments:

Reviewer #1

In this manuscript, authors prepared Bio-SiO2 incorporated barium ferrite composites by solid-state reaction method. Subsequently, they have studied structure, morphology, magnetic and microwave absorption properties. However, there are many confusions needs to be resolved in detailed explanations to accept this manuscript.

1. *What is the motivation of Bio-silica doping? Whether authors wanted to prepare Bio-SiO2/barium ferrite composites or Bio-SiO2 incorporated barium ferrite and its composites/secondary phases formed as a consequence of the annealing process?. The formation of Bio-SiO2 incorporated barium ferrite composites is solely due to the annealing or they desired?*

   Response: Thanks for the valuable and significant question. Please note that the motivation for the addition of the silica in the barium ferrite is to replace the rare earths (expensive and scarce) and shift the microwave absorption frequency in the new material. In addition, the efforts are made to obtain some cheap and easily available MW absorbing materials. Yes, these Bio-SiO_2 incorporated barium ferrite and its composites/secondary phases are formed as a consequence of the annealing process in the solid-state reaction.

2. *Categorically, silica is a famous semiconducting material that can be obtained either commercially or from plentiful natural resources. Interestingly, rice husk after complete combustion can be a great source of bio-silica. Such bio-silica derived from rice husk (as abundant raw material) has several advantages compared to silica minerals, including the existence of fine grain, high reactivity, low cost and can function as a heavy metal binder. Encouraged by these notable benefits of rice husk extracted bio-silica, we prepared some biosilica integrated barium ferrite composites (hereafter called SiBFCs). What is the point of interest of silica, when there is no silica in your final product of SiO2 incorporated barium ferrite composites?*

Response: Thanks for the critical remark and question. Please note that the incorporated bio-silica in the proposed SiBFCs enabled the formation of some new tetragonal ($BaFeSi_4O_{10}$) and hexagonal crystalline phase which in turn affected the surface morphologies, structures, magnetic and MW absorption properties of these composites. The relative complex permittivity and permeability in the MW frequency range of 8–13 GHz showed improved spectral profile. In short, the presence of bio-silica and subsequent formation of a new phase in the composites could affect the ability of the material to absorb microwaves and shift the minimum reflection loss peak position. For the first time we showed a possibility of customising the MW absorption characteristics of the proposed comosites by adjusting the bio-silica contents.

3. *notable benefits of rice husk extracted bio-silica, we prepared some biosilica integrated barium ferrite composites (hereafter called SiBFCs) to reduce the values of the coercive field (Hc) and saturation magnetization (Ms), in that way enhancement of the selective M absorption capacity of the achieved SiBFCs. The resultant magnetic properties may decrease as we incorporate the nonmagnetic matrix in magnetic samples. But by reducing the magnetic properties how can MW absorption properties can be enhanced. What are the basic requirements to improve the MW absorption properties? Has Bio-SiO2 incorporated barium ferrite and its composites just prepared to reduce the magnetic properties?*

   Response: Thanks for the useful question. Please note that the main idea was to improve the MW absorption, reflection loss and magnetic properties of the BFCs by incorporating some alternative materials (such as bio-silica which is plentiful, cheap and eco-friendly) other than the conventional rare earths (costly and sparse). Interestinly, the incorporated bio-silica contributed to the generation of some new crystalline phase, thereby causing a significant reduction of the saturation magnetisation (*Ms*). This in turn led to a change in the zero field ferromagnetic resonance frequency (*f*), magnetic loss (μ″) and dielectric loss, indicating a significant effect on the reflection loss useful for device applications.

4. *SiBFCs with the composition of (x)Bio-SiO2:(80-x)Fe2O3:(20)BaO, (x = 0, 1, 2 and 3 in wt%) were synthesized via the modified solid-state reaction. How did the authors calculate the wt% of silica, is it with respect to Fe2O3 or barium ferrite?. If silica is incorporated/doping why they considered wt%, why not stoichiometry? Like Fe2-xSixO3*
5.

   Response: Thanks for the important question. Please note that the wt% of silica is chosen with respect to the total weight of the sample. We used the wt% in the solid-state reactions because the process is easy and fast to get the absorbent material. Besides, the industrial world wants a process that is faster, easier and inexpensive compared to the exisitng art-of-the methods. This was the reason of using wt% in the composition.

6. *we crushed and glued some pellets with resin to get a rectangular-shaped sample of dimension (2.3 cm × 1.0 cm × 0.5 cm) using a WR90 sample holder. Please explain in detail, the fabrication process.*

   Response: Thanks for asking the explanation. Please note that the fabrication process has been added in the revised manuscript.

7. *In Fig. 1 XRD patterns show that undoped barium ferrite, SiBF0 samples exhibit hexagonal phase BaFe12O19 whereas, silica incorporated samples lead to a new hexagonal phase of* $Ba_3Fe_{32}O_{51}$ *along with secondary phases of Si compounds, are* $BaFeSi_4O_{10}$, $Ba_3Si_5O_{13}$ *and* $BaFeSi_4O_{10}$, *respectively, for 1 wt%, 2wt% and 3wt% of silica. However,* Si^{4+} *replacing* Fe^{3+} *is not remaining in the hexagonal phase indeed forming the byproducts and is not consistent with all the wt%. Therefore, how the properties will be correlated with silica wt%?*

   Response: Thanks for the question. Please note that in the solid reaction process, it is very difficult to control the parameters such as the concentration, temperature, time of the sintering process to obtain single phase. Therefore, we are looking for the optimum concentration where the added silica can penetrate the barium ferrite structure and modify the properties of the composite. In fact, the presence of silica with different wt% is observed to produce a new phase after the sintering process, indicating the active participation of silica and formation of a new phase (composite) due to an imperfect reaction mechanism, influencing the reflection loss of the composites.

8. *The XRD patterns of 2 wt% and 3 wt% of silica the intensity of secondary phases (Ba3Si5O13 and BaFeSi4O10) appears much higher than the hexagonal phase of samples why? The authors should provide experimental evidence of elemental wt% of the compounds from the EDX analysis.*

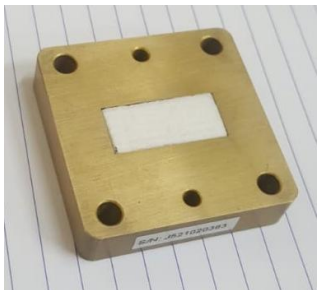
   Response: Thanks for the valuable suggestions. Please note that at present we do not have the EDX facility in our laboratory and thus we are unable to provide the EDX analysis of the SiBFCs at this moment. However, your advice is well taken for the future work.

9. *Fig. 3 shows the SEM micrographs of the prepared SiBFCs. The surface morphologies of the composites consisted of rough microstructures with high porosity. The composite particles within the samples were bonded tightly to each other and formed some intergranular pores due to the occupation of Si4+ ions in the preferred lattice sites. Consequently, more porous microstructures are formed on the surface. These pores, in turn, can favor the entrapment of the MW and its subsequent scattering in all directions by the surface of the particle present in the pores. In all the synthesized SiBFCs, these particles disclose a typical hexagonal morphology with a particle size of about 0.5 m. The morphology of the samples clearly exhibits bulk and porous is not consistent in samples as authors discussed. The whole sentence should be rewritten.*

   Response: Thanks for the valuable advice and suggestion. Please note that the whole paragraph is rewritten with better explanation.

10. *Fig. 6 The value of RL is evaluated at what thickness?*

    Response: Thanks for the question. The thickness of the sample was 0.5 cm according to the WR sample holder



11. *Why there are multiple peaks in RL spectra and what is the origin?*

Response: Thanks for the question. Please note that the multiple peaks in the RL spectra originated from the atomic bonds vibration in the barium ferrite due to MW frequencies, where several atomic bonds (Ba-Fe, Fe-O, Ba-O) are present. The peaks in the black spectrum are the origin.

12. *Authors should compare the merits of their results (RL maximum) with literature? How are results are best compared to the others?using different analytical tools to determine the influence of Nd3+ ions on the microstructures, surface morphologies, magnetic characteristics, and MW reflection loss in the X band. Achieved $Ba_6Nd_2Fe_4O_{15}$ and $BaFe_2O_4$ phases of the proposed BFCs were shown to be beneficial for sundry applications. I recommend this manuscript to be published after a minor revision by addressing the following concerns.*

Response: Thanks for the question and remark. Please note that the international standard for R_L is less than - 10 dB. If the R_L of a material is less than -10 dB, then the material can be nominated as the microwave absorber candidate.

Reviewer #2:

Manuscript CAP-D-19-01273, entitled "Bio-silica incorporated barium ferrite composites: Evaluation of structure, morphology, magnetic and microwave absorption traits" Comments: This manuscript describes a series of bio-silica ion incorporated barium-ferrite-composites with the composition of (x)Bio-SiO2:(80-x)Fe2O3:(20)BaO prepared by the modified solid-state reaction. The impact of different bio-silica substances on the surface morphologies, structures, and magnetic characteristics of these composites were assessed.

The work is placed in a proper context and the title clearly identifies the subject matter. The technical content of the paper merits publication in Current Applied Physics, and I would suggest accepting it after addressing my comments.

1. *The background and highlights of this manuscript need to be further untangled. The innovation and scientific significance need to be further lighted.*

Response: Thanks for the valuable comments, encouragements and inspiring words. Please note that the background and highlights of this manuscript have been revised and added in the manuscript as advised.

2. *The abstract and conclusion need further refinement, such as eliminating unnecessary modifications to make it more logical.*

   Reponse: It has been done

3. *Has the XRD data been refined? If not, please refine the XRD data as this is the needed process that should undergone for ceramic sample data.*

   Response: Thanks for the question. Yes, the XRD data has been refined

4. *Some highly related papers should be cited to discuss about the microwave materials and absorption, give a broader view of those materials and performance, such as some related literatures should be cited, such as: (1) Ceramics International. 2018, 44: 6953-6958; (2) ACS Applied Materials & Interfaces. 2017, 9, 11711-11720; (3) Nanoscale. 2017, 9, 8591-8599; (4) Appl. Phys. Lett. 111, 242902 (2017).*

   Response: Thanks for the question and advices. Please note that, we cannot access (1) ACS Applied Materials & Interfaces. 2017, 9, 11711-11720, (2) Nanoscale. 2017, 9, 8591-8599; (3) Appl. Phys. Lett. 111, 242902 (2017).

5. *Please polish the whole manuscript before it is accepted for publication.*

   Response: Thanks for the advices. We have polished the manuscript

The manuscript has been resubmitted for your kind perusal. We look forward for your positive response and acceptance of the revised manuscript.

Sincerely,

Wahyu Widanarto

Highlights (for review)

- Bio-silica integrated barium ferrite composites (SiBFCs) were prepared.
- Structure, morphology, magnetic and MW absorption traits of such SiBFCs were assessed.
- The SiBFCs revealed a new hexagonal ($Ba_3Fe_{32}O_{51}$) and tetragonal ($BaFeSi_4O_{10}$) phase.
- Achieved SiBFCs were shown to be useful for the MW absorption devices applications.
- Minimum reflection loss was tailored by controlling the bio-silica contents in SiBFCs.

Bio-silica incorporated barium ferrite composites: Evaluation of structure, morphology, magnetic and microwave absorption traits

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ABSTRACT

~~The wireless communications and electronic devices based on the microwave (MW) frequency are useful due to several implications. In this view,~~ A series of bio-silica ion incorporated barium-ferrite-composites with the composition of (*x*)Bio-SiO_2:(80-*x*)Fe_2O_3:(20)BaO, where *x* = 0, 1, 2, and 3 wt% were prepared using the modified solid-state reaction method. The influence of different bio-silica (extricated from sintered rice husk) contents on the surface morphologies, structures, and magnetic characteristics of these composites were assessed. The relative complex permittivity and permeability were resolved using the Nicholson-Ross-Weir strategy in the frequency range of 8–13 GHz. Meanwhile, the reflection loss was estimated through the transmission/reflection line theory to assess the MW absorption properties of the composites. Incorporation of the bio-silica in the barium ferrite composites generated a new hexagonal phase ($Ba_3Fe_{32}O_{51}$) and a tetragonal phase ($BaFeSi_4O_{10}$) which led to a decrease in the saturation magnetization and significant shift in the MW frequency absorption peak positions.

Keywords: bio-silica; barium ferrite; magnetic properties; permittivity; permeability; reflection loss

1. Introduction

In recent times, microwave (MW) absorbing materials became one of the key high-tech candidates for the development of the high-performance device in the field of anti-radar technology, wireless communications, and shielding of electromagnetic wave interferences [1–3]. The coating of the targets with MW absorbing materials emerged as a strategy to reduce the intensity of the reflected electromagnetic waves. This technique utilises the absorption or dispersion of electromagnetic energy in the material medium between the electromagnetic wave source and the protected target. For such purposes, materials must be fit for weakening and dispersing the overabundance measure of the electromagnetic radiation in the form of heat through the mechanisms of magnetic and dielectric loss [4]. Likewise, innovative magnetic

materials, particularly ferrites based, are uncovered to be suitable for absorbing the MW and have frequently been investigated [5–13].

It has been verified that a barium ferrite system (a magnetic material) with a wide crystalline anisotropic magnetic field can potentially be used in the GHz frequency range compared to other ferrites with spinel and garnet structures [2,14]. Thus, the barium hexaferrite (BHF) owing to their unusual strong uniaxial anisotropic magnetic field was prepared as MW absorbing material [6–8,15,16]. The literature reports revealed that the replacement of Fe^{3+} by trivalent lanthanide ions (used as doping agent) is an effective way to shift the resonant frequency (f_r) and change the anisotropy field (H_A), influencing the MW absorption capacity of the magnetic material [2,9,17–21]. However, the scarcity and high prices of rare earth materials limit their widespread usages. Thus, the exploration of the doping agent alternative to the rare-earth ions (acts as an activator) became mandatory to fulfil such need.

Categorically, silica is a famous semiconducting material that can be obtained either commercially or from plentiful natural resources. Interestingly, rice husk after complete combustion can be a great source of bio-silica. Such bio-silica derived from rice husk (as abundant raw material) has several advantages compared to silica minerals, including the existence of fine grain, high reactivity, low cost and can function as a heavy metal binder. Encouraged by these notable benefits of rice husk extracted bio-silica, we prepared some bio-silica integrated barium ferrite composites (hereafter called SiBFCs) to reduce the values of coercive field (H_c) and saturation magnetisation (M_s), in that way enhancement of the selective MW absorption capacity of the achieved SiBFCs.

This paper reports the synthesis and characterisations of the newly prepared SiBFCs, wherein the bio-silica at various concentrations were merged in pure barium hexaferrite. The undoped and doped composites were prepared via solid–state reaction method. These as-prepared SiBFCs were characterised using diverse analytical apparatuses to assess the effects of various bio-silica contents on the microstructure, morphology, magnetic properties, and MW reflection losses in the frequency range of GHz. The obtained $BaFeSi_4O_{10}$ and $Ba_3Fe_{32}O_{51}$ composites were given away to be valuable for several applications.

2. Experimental Procedures

Four samples of SiBFCs with the composition of (x)Bio-SiO_2:(80-x)Fe_2O_3:(20)BaO, (x = 0, 1, 2 and 3 in wt%) were synthesised via the modified solid-state reaction method. Highly pure powders of $BaCO_3$ (from Merck with the purity of 99%), bio-silica, and γ-Fe_2O_3 as primary raw constituents were utilised to prepare these SiBFCs. The $BaCO_3$ powder was calcined at 350 °C for 15 minutes in the air atmosphere to eliminate the presence of the carbon component. The bio-silica was obtained by sintering the rice husk ash ~~in the furnace~~ at a temperature of 1000 °C for three hours in the air atmosphere. Meanwhile, the Fe_3O_4 was extracted from the iron sand [22] and sintered at 850 °C in the air atmosphere for three hours to obtain γ-Fe_2O_3. Afterwards, the bio-silica powder was mixed gradually with the γ-Fe_2O_3 and BaO powder. The mixed constituent powder was compressed to yield the pellet of 1 mm thick and 10 mm diameter [19]. Additionally, all the pellets were strengthened and sintered at 800 °C (for one hour) and 1100 °C (for 5 hours) an air atmosphere before they were cooled down to the room temperature naturally. The acquired pellets were named as SiBF0, SiBF1, SiBF2, and SiBF3, depending on the matching bio-silica content of 0, 1, 2, and 3 wt%. For further characterizations, some pellets were crushed. Finally, the crushed pellets were mixed with epoxy resin at a mass ratio of 7:3 to get a rectangular-shaped sample of dimension (2.3 cm × 1.0 cm × 0.5 cm) using a WR90 sample holder.

The surface morphology and microstructure of the prepared SiBFCs were characterised using the scanning electron microscope (SEM, Hitachi SU 3500). The crystalline nature of the prepared SiBFCs was confirmed using the X-ray diffraction (XRD, SmartLab 3 kW) furnished with Cu–Kα radiation of wavelength (λ) ≈ 0.1541874 nm. The vibrating sample magnetometer (VSM, Oxford 1.2H) was used to scrutinise the magnetic traits of the proposed samples. The scattering parameters (S) of the samples were recorded on a vector network analyser (VNA, Keysight PNA-L N5232A) operated in the frequency range of 8–13 GHz. The MW absorption measurement was conducted to yield the scattering parameters such as S_{11}, S_{12}, S_{21}, and S_{22} (S-parameters). The value of S_{11} (= S_{22}) signified the reflection coefficient (Γ) and S_{21} (= S_{12}) represented the transmission coefficient (T). The values of relative complex permeability (μ_r) and permittivity (ε_r) were obtained following the Nicholson-Ross-Weir (NRW) relations given by:

$$\mu_r = \frac{1+\Gamma}{\Lambda(1-\Gamma)\sqrt{\frac{1}{\lambda_0^2}-\frac{1}{\lambda_c^2}}} \quad (1)$$

$$\frac{1}{\Lambda^2} = -\left[\frac{1}{2\pi d}\ln\left(\frac{1}{T}\right)\right]^2 \quad (2)$$

$$\varepsilon_r = \frac{\lambda_0^2}{\mu_r}\left(\frac{1}{\lambda_c^2}-\left[\frac{1}{2\pi d}\ln\left(\frac{1}{T}\right)\right]^2\right) \quad (3)$$

where λ_0 is the wavelength in vacuum, λ_c is the cut-off wavelength, *c* is the speed of the light, and *d* is the thickness of the sample.

3. Results and Discussions

Fig. 1 depicts the XRD pattern of the synthesised bio-silica, confirming all the diffraction peaks due to the tetragonal crystal system of SiO_2. Fig. 2 displays the XRD diffractograms of the produced SiBFCs that consisted of many characteristic peaks assigned to altering crystalline lattices. All the observed peaks in the pristine sample (SiBF0 without bio-silica incorporation) were due to the main hexagonal crystalline lattice of $BaFe_{12}O_{19}$ and tallied to the ICDD card number 00-039-1433 with crystal configurations of $a = b = 0.5894$ nm, $c = 2.3215$ nm, $\alpha = \beta = 90°$ and $\gamma = 120°$. Nevertheless, the appeared peak at 27.52° was due to the monoclinic crystalline phase of $Ba_2Fe_2O_5$ that corresponded to the ICDD card number 00-043-0256. The XRD pattern of SiBF1 sample revealed that the replacement of Fe^{3+} in the barium ferrite lattice by Si^{4+} ions at 1 wt% of SiO_2 did not cause any significant changes of the crystal structures. The appearance of two dominant peaks in SiBF1 confirmed the formation of the major phase of $Ba_3Fe_{32}O_{51}$ and minor phase of barium iron silicate $BaFeSi_4O_{10}$. The occurrences of sharp XRD peaks were due to the hexagonal crystal lattice of $Ba_3Fe_{32}O_{51}$ that corresponded to the ICDD card number 00-041-0846 with the crystal configurations of $a = b = 0.5892$ nm, $c = 2.3198$ nm, $\alpha = \beta = 90°$ and $\gamma = 120°$. Moreover, the peaks centred at 26.52° and 35.48° were assigned to the tetragonal crystal lattice planer orientation of $BaFeSi_4O_{10}$ (tallied to the ICDD card number 00-003-0402). The XRD pattern of the composite prepared with two wt% of bio-silica revealed a

new phase of barium silicate $Ba_3Si_5O_{13}$ with the monoclinic crystalline lattice structure (agreed to the ICDD number 00-026-0179). The composite containing three wt% of bio-silica exhibited the dominant barium iron silicate $BaFeSi_4O_{10}$ ordered phase. It was claimed that the replacement of Fe^{3+} ions by Si^{4+} in the produced BFCs can create another significant hexagonal phase of $Ba_3Fe_{32}O_{51}$ and a tetragonal phase of $BaFeSi_4O_{10}$. The observed broadening in the XRD peaks for all the studied SiBFCs is attributed to the insolvent of lattice strain and nano-crystallites size confinement effects [23,24].

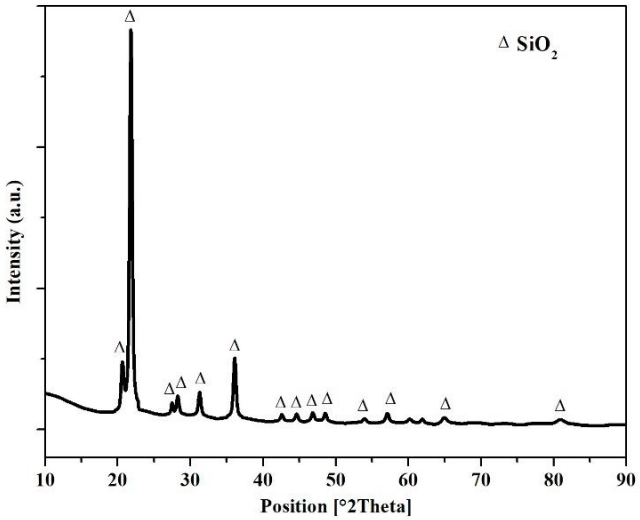

Fig. 1. The XRD pattern of the bio-silica (SiO_2) at 1000 °C

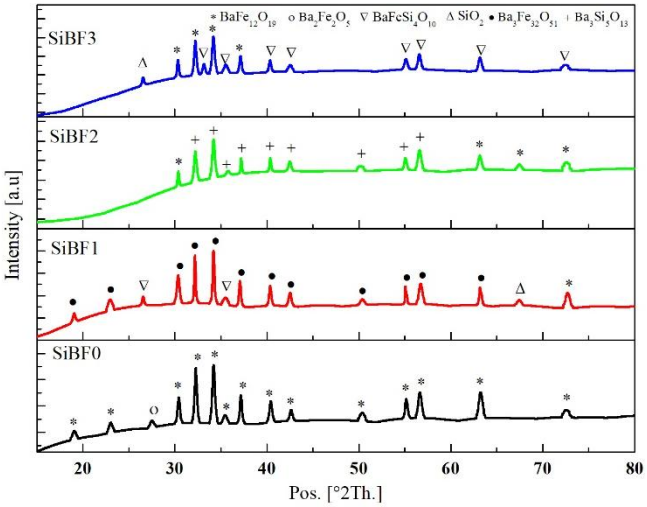

Fig. 2. The XRD patterns of the Si^{4+} undoped and doped SiBFCs

Fig. 3 shows the SEM micrographs of the prepared SiBFCs, which consisted of hexagonal particles morphology (particle size of about 0.5 μm) with irregular microstructures packing. These porous composite particles were attached to each other and formed some intergranular pores. It can be argued that the substitution of the Fe^{3+} by Si^{4+} (from the bio-silica) in the produced BFCs that created a new tetragonal phase of $BaFeSi_4O_{10}$ were closely packed to the hexagonal phase of $Ba_3Fe_{32}O_{51}$. In the intergranular pores of these composite particles phase, the Si^{4+} were preferentially occupied in the lattice sites, imparting more porosity to the microstructures. These pores in turn favored the entrapment of the MW wherein the finer particles present in the pores could scatter the MW randomly in all directions, thereby modifying the MW absorption characteristics of the composites. In all the prepared SiBFCs, these distinct types of the surface morphologies and particles distribution were responsible for the changes in the magnetic and reflection loss properties.

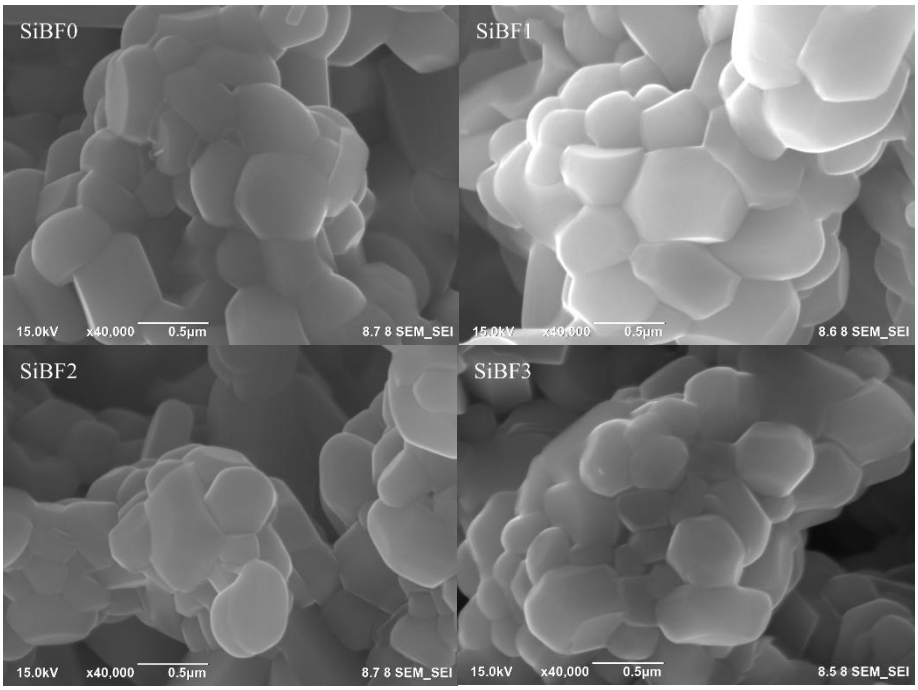

Fig. 3. The SEM images of the SiBFCs

Fig. 4 represents the room temperature hysteresis loops (*M-H* curves) of the as-synthesised SiBFCs. The values of saturation magnetisation of the studied SiBFCs are decreased (from 39.5 to 30.4 emu/g) significantly with the addition of bio-silica (from 1 to 3 wt%) into the

composites. However, the coercive field values (altered from 775.9 – 811.0 Oe) are not affected appreciably due to the increase in bio-silica contents. The maximum reduction in the saturation magnetisation displayed by the SiBF3 sample is ascribed to the presence of dominant tetragonal barium iron silicate ($BaFeSi_4O_{10}$) crystalline phase in the composite. The coercive field values of the composites are observed to increase slightly due to the rise in bio-silica contents. With the increase in bio-silica content from 0 to 2 wt%, the area of the hysteresis loop of the SiBFCs enlarges from 15.9 to 31.0 kOe.emu/g, respectively. Furthermore, the hysteresis loop area of SiBFC shrinks at three wt% of bio-silica. The observed change in the magnetic permeability of SiBFCs with the addition of bio-silica into the composites is majorly attributed to the alteration in the saturation magnetisation and coercive field values.

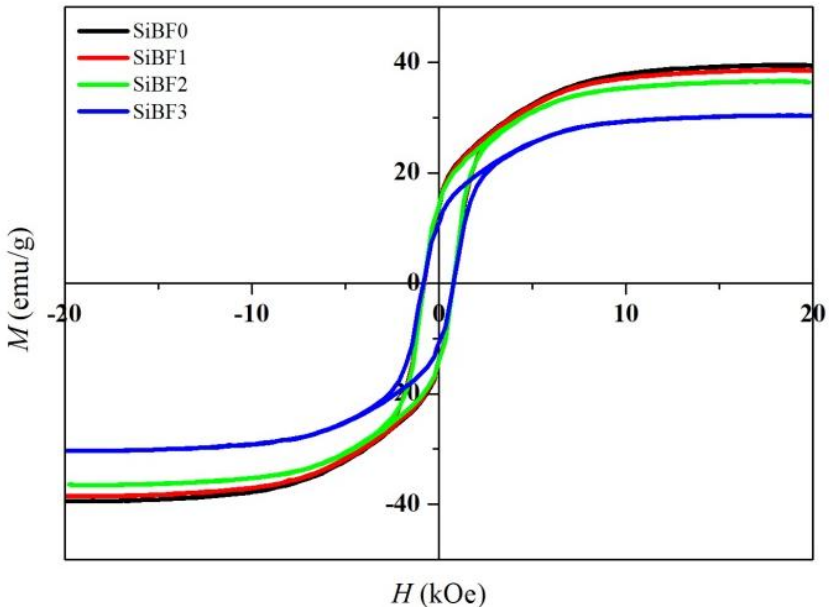

Fig. 4. The room temperature hysteresis loops (*M-H*) of the SiBFCs

Fig. 5(a) describes the frequency dependence of the relative permeability ($\mu_r = \mu' + \mu''$) of all the SiBFCs. The real part of the permeability (μ') specifies a gradual decrease in their magnetic energy storing capacity due to polarization of magnetic dipole in higher MW frequency. The μ' value of the composites drops from about 2.5 (for SiBF0 and SiBF2) and 0.9 (for SiBF1 and SiBF3) to near-zero with the increase in bio-silica content from 0 to 3 wt%. Meanwhile, the observed magnetic loss is revealed by the imaginary part of the permeability (μ''

). The μ'' value of SiBF0 and SiBF2 declines rapidly from 0.9 to near-zero and almost remains constant near-zero for SiBF1 and SiBF3 due to the relaxation process, which generates dissipation energy of MV as thermal energy.

Fig. 5(b) displays the frequency-dependent relative complex permittivity ($\varepsilon_r = \varepsilon' + \varepsilon''$) of all SiBFCs. The ε' and ε'' are the real and imaginary parts of the permittivity, respectively. The ε' value indicated the lack of energy absorption by the composite from an externally applied electric field [25]. The ε' values of the composite without bio-silica doping (SiBF0) were dropped sharply. Conversely, the ε' values of the composites containing bio-silica (1 to 3 wt%) were increased significantly above 12 GHz due to the electrical dipole polarization. The value of ε'' (called the dielectric loss factor) denotes the electrical energy dissipation ability of the SiBFCs. The ε'' values of all SiBFCs were decreased gradually with the rise in the frequency.

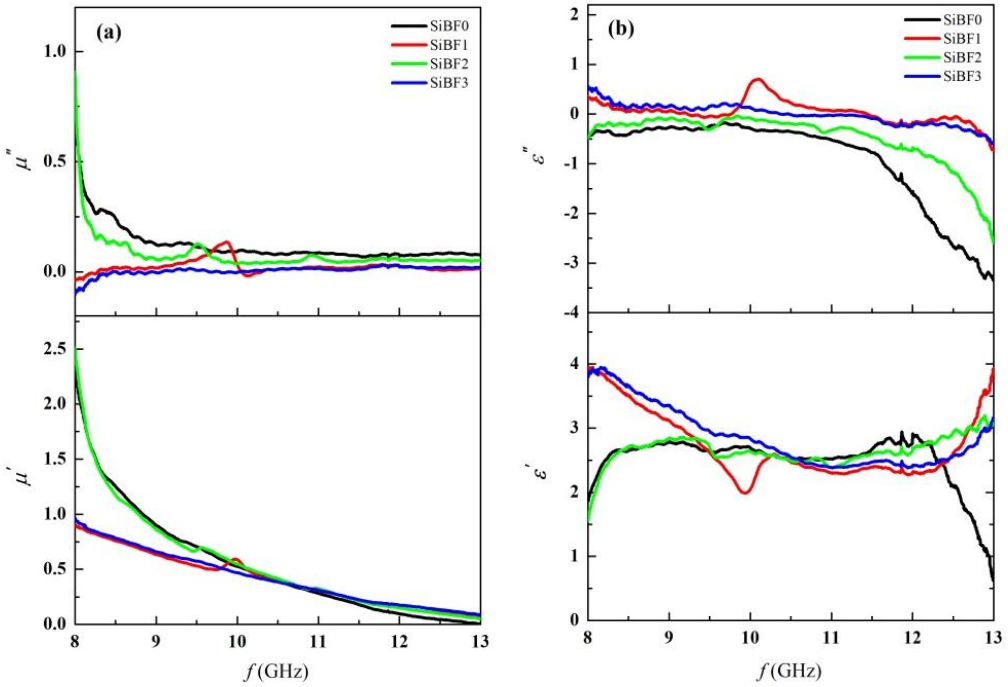

Fig. 5. Frequency-dependent (a) permeability and (b) permittivity of the SiBFCs of thickness 0.5 cm

The MW absorption properties of SiBFCs were evaluated by calculating the values of reflection loss (R_L) based on the transmission/reflection line theory [26–28]:

$$R_L = -20\log\left|\frac{Z_{in} - Z_0}{Z_{in} + Z_0}\right| \quad (4)$$

$$Z_{in} = Z_0\sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left[j\frac{2\pi fd}{c}\sqrt{\mu_r\varepsilon_r}\right] \quad (5)$$

where Z_0 and Z_{in} are the corresponding intrinsic impedance in a vacuum and the input impedance in the material, f represents the MW frequency, d denotes the sample thickness, and c is the light speed in the vacuum.

Fig. 6 illustrates the frequency-dependent changes in the R_L values of the prepared SiBFCs. The MW absorption ability of materials is determined by two crucial factors i.e. R_L and bandwidth. The value of R_L for SiBF0 was valued less than -10 dB at the frequency of 8.35, 9.37, and 10.60 GHz. The band positions (peak frequencies) of the composites disclose a significant shift with the change in bio-silica contents with the bandwidth of less than 1 GHz. The sample prepared with one wt% of bio-silica (SiBF1) reveals the lowest R_L value of -28.56 dB at the frequency of 12.18 GHz. This disclosure affirms that the MW absorption capacity of the proposed SiBFCs can be essentially customised by controlling the impurity or dopant (bio-silica) concentration, fulfilling the application demand in the X band. Some strategies were employed to modify the dielectric and magnetic parameters including the single trivalent cation substitution and different combinations of cations to minimise the MW reflection loss [27,29]. Compared to the earlier materials, present bio-silica based composites are cheaper and easy to prepare.

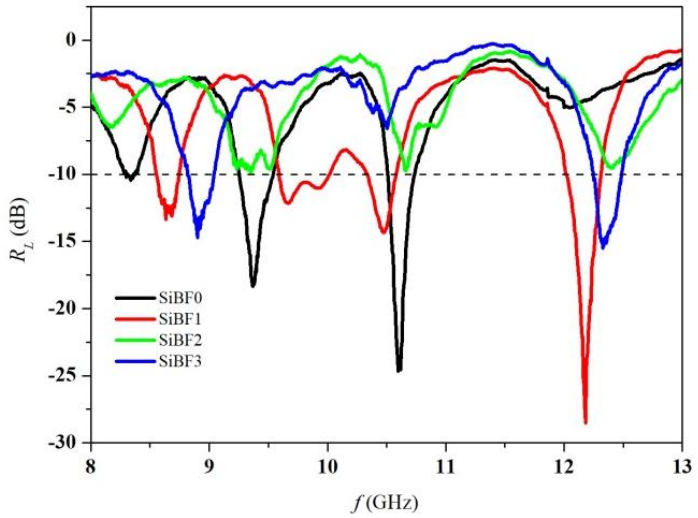

Fig. 6. Reflection loss as a function of MW frequency of the SiBFCs of 0.5 cm thick.

4. Conclusions

Considering the MW absorption potency of barium ferrites ~~and its usefulness in wireless communications~~, we synthesised four SiBFCs (without and with bio-silica activation) via the modified solid-state reaction approach. Rice husk derived abundant and cheap bio-silica was doped into the barium ferrites to improve their magnetic and MW absorption attributes. The structure of the studied composites revealed the presence of various crystalline phases. The surface of SiBFCs disclosed hexagonal morphology with a particle size of about 0.5 μm. The surface morphology, structure, MW reflection loss, permittivity, permeability, coercivity, saturation magnetisation, and MW absorption of the proposed SiBFCs were found to be sensitive to the substitution of Fe^{3+} by Si^{4+} ions into the composite matrix. It was established that the MW absorption ability of the newly composed SiBFCs can be tailored by adjusting the bio-silica content. Present knowledge may contribute towards the development of the SiBFCs based MW absorption devices.

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Keywords: bio-silica; barium ferrite; magnetic properties; permittivity; permeability; reflection loss

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Abstract: A series of bio-silica ion incorporated barium-ferrite-composites with the composition of (x)Bio-SiO2:(80-x)Fe2O3:(20)BaO, where x = 0, 1, 2, and 3 wt% were prepared using the modified solid-state reaction method. The influence of different bio-silica (extricated from sintered rice husk) contents on the surface morphologies, structures, and magnetic characteristics of these composites were assessed. The relative complex permittivity and permeability were resolved using the Nicholson-Ross-Weir strategy in the frequency range of 8-13 GHz. Meanwhile, the reflection loss was estimated through the transmission/reflection line theory to assess the MW absorption properties of the composites. Incorporation of the bio-silica in the barium ferrite composites generated a new hexagonal phase (Ba3Fe32O51) and a tetragonal phase (BaFeSi4O10) which led to a decrease in the saturation magnetization and significant shift in the MW frequency absorption peak positions.

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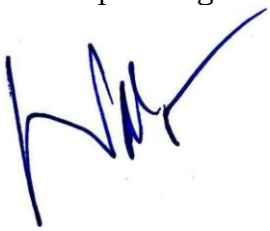

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   Fig. 5(a) describes the frequency dependence of the relative permeability ($\mu = \mu' + \mu''$) of all the SiBFCs.
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Highlights (for review)

- Bio-silica integrated barium ferrite composites (SiBFCs) were prepared.
- Structure, morphology, magnetic and MW absorption traits of such SiBFCs were assessed.
- The SiBFCs revealed a new hexagonal ($Ba_3Fe_{32}O_{51}$) and tetragonal ($BaFeSi_4O_{10}$) phase.
- Achieved SiBFCs were shown to be useful for the MW absorption devices applications.
- Minimum reflection loss was tailored by controlling the bio-silica contents in SiBFCs.

Bio-silica incorporated barium ferrite composites: Evaluation of structure, morphology, magnetic and microwave absorption traits

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ABSTRACT

A series of bio-silica incorporated barium-ferrite-composites with the composition of (*x*)Bio-SiO_2:(80-*x*)Fe_2O_3:(20)BaO, where *x* = 0, 1, 2, and 3 wt% were prepared using the modified solid-state reaction method. The influence of different bio-silica (extricated from sintered rice husk) contents on the surface morphologies, structures, and magnetic characteristics of these composites were assessed. The relative complex permittivity and permeability were resolved using the Nicholson-Ross-Weir strategy in the frequency range of 8–13 GHz. Meanwhile, the reflection loss was estimated through the transmission/reflection line theory to assess the MW absorption properties of the composites. Incorporation of the bio-silica in the barium ferrite composites generated a new hexagonal phase ($Ba_3Fe_{32}O_{51}$) and a tetragonal phase ($BaFeSi_4O_{10}$) which led to a decrease in the saturation magnetization and significant shift in the MW frequency absorption peak positions.

Keywords: bio-silica; barium ferrite; magnetic properties; permittivity; permeability; reflection loss

1. Introduction

In recent times, microwave (MW) absorbing materials became one of the key high-tech candidates for the development of the high-performance device in the field of anti-radar technology, wireless communications, and shielding of electromagnetic wave interferences [1–3]. The coating of the targets with MW absorbing materials emerged as a strategy to reduce the intensity of the reflected electromagnetic waves. This technique utilizes the absorption or dispersion of electromagnetic energy in the material medium between the electromagnetic wave source and the protected target. For such purposes, materials must be fit for weakening and dispersing the overabundance measure of the electromagnetic radiation in the form of heat through the mechanisms of magnetic and dielectric loss [4]. Likewise, innovative magnetic materials, particularly ferrites based, are uncovered to be suitable for absorbing the MW and have frequently been investigated [5–13].

It has been verified that a barium ferrite system (a magnetic material) with a wide crystalline anisotropic magnetic field can potentially be used in the GHz frequency range compared to other ferrites with spinel and garnet structures [2,14]. Thus, the barium hexaferrite (BHF) owing to their unusual strong uniaxial anisotropic magnetic field was prepared as MW absorbing material [6–8,15,16]. The literature reports revealed that the replacement of Fe^{3+} by trivalent lanthanide ions (used as doping agent) is an effective way to shift the resonant frequency (f_r) and change the anisotropy field (H_A), influencing the MW absorption capacity of the magnetic material [2,9,17–21]. However, the scarcity and high prices of rare earth materials limit their widespread usages. Thus, the exploration of the doping agent alternative to the rare-earth ions (acts as an activator) became mandatory to fulfill such need.

Categorically, silica is a famous semiconducting material that can be obtained either commercially or from plentiful natural resources. Interestingly, rice husk after complete combustion can be a great source of bio-silica. Such bio-silica derived from rice husk (as abundant raw material) has several advantages compared to silica minerals, including the existence of fine grain, high reactivity, low cost and can function as a heavy metal binder. Encouraged by these notable benefits of rice husk extracted bio-silica, we prepared some bio-silica integrated barium ferrite composites (hereafter called SiBFCs) to reduce the values of coercive field (H_c) and saturation magnetization (M_s), in that way enhancement of the selective MW absorption capacity of the achieved SiBFCs.

This paper reports the synthesis and characterizations of the newly prepared SiBFCs, wherein the bio-silica at various concentrations were merged in pure barium hexaferrite. The undoped and doped composites were prepared via a solid−state reaction method. These as-prepared SiBFCs were characterized using diverse analytical apparatuses to assess the effects of various bio-silica contents on the microstructure, morphology, magnetic properties, and MW reflection losses in the frequency range of GHz. The obtained $BaFeSi_4O_{10}$ and $Ba_3Fe_{32}O_{51}$ composites were given away to be valuable for several applications.

2. Experimental Procedures

Four samples of SiBFCs with the composition of (x)Bio-SiO_2:(80-x)Fe_2O_3:(20)BaO, (x = 0, 1, 2 and 3 in wt%) were synthesised via the modified solid-state reaction method. Highly pure powders of $BaCO_3$ (from Merck with the purity of 99%), bio-silica, and γ-Fe_2O_3 as primary raw

constituents were utilized to prepare these SiBFCs. The $BaCO_3$ powder was calcined at 350 °C for 15 minutes in the air atmosphere to eliminate the presence of the carbon component. The bio-silica was obtained by sintering the rice husk ash at a temperature of 1000 °C for three hours in the air atmosphere. Meanwhile, the Fe_3O_4 was extracted from the iron sand [22] and sintered at 850 °C in the air atmosphere for three hours to obtain γ-Fe_2O_3. Afterward, the bio-silica powder was mixed gradually with the γ-Fe_2O_3 and BaO powder. The mixed constituent powder was compressed to yield the pellet of 1 mm thick and 10 mm diameter [19]. Additionally, all the pellets were strengthened and sintered at 800 °C (for one hour) and 1100 °C (for 5 hours) an air atmosphere before they were cooled down to the room temperature naturally. The acquired pellets were named as SiBF0, SiBF1, SiBF2, and SiBF3, depending on the matching bio-silica content of 0, 1, 2, and 3 wt%. For further characterizations, some pellets were crushed. Finally, the crushed pellets were mixed with epoxy resin at a mass ratio of 7:3 to get a rectangular-shaped sample of dimension (2.3 cm × 1.0 cm × 0.5 cm) using a WR90 sample holder.

The surface morphology and microstructure of the prepared SiBFCs were characterized using the scanning electron microscope (SEM, Hitachi SU 3500). The crystalline nature of the prepared SiBFCs was confirmed using the X-ray diffraction (XRD, SmartLab 3 kW) furnished with Cu–Kα radiation of wavelength (λ) ≈ 0.1541874 nm. The vibrating sample magnetometer (VSM, Oxford 1.2H) was used to scrutinize the magnetic traits of the proposed samples. The scattering parameters (*S*) of the samples were recorded on a vector network analyzer (VNA, Keysight PNA-L N5232A) operated in the frequency range of 8–13 GHz. The MW absorption measurement was conducted to yield the scattering parameters such as S_{11}, S_{12}, S_{21}, and S_{22} (*S*-parameters). The value of S_{11} (= S_{22}) signified the reflection coefficient (Γ) and S_{21} (= S_{12}) represented the transmission coefficient (*T*). The values of complex relative permeability (μ_r) and permittivity (ε_r) were obtained following the Nicholson-Ross-Weir (NRW) relations given by:

$$\mu_r = \frac{1+\Gamma}{\Lambda(1-\Gamma)\sqrt{\frac{1}{\lambda_0^2}-\frac{1}{\lambda_c^2}}} \tag{1}$$

$$\frac{1}{\Lambda^2} = -\left[\frac{1}{2\pi d}\ln\left(\frac{1}{T}\right)\right]^2 \quad (2)$$

$$\varepsilon_r = \frac{\lambda_0^2}{\mu_r}\left(\frac{1}{\lambda_c^2} - \left[\frac{1}{2\pi d}\ln\left(\frac{1}{T}\right)\right]^2\right) \quad (3)$$

where λ_0 is the wavelength in vacuum, λ_c is the cut-off wavelength, c is the speed of the light, and d is the thickness of the sample.

3. Results and Discussions

Fig. 1 depicts the XRD pattern of the synthesized bio-silica, confirming all the diffraction peaks due to the tetragonal crystal system of SiO_2. Fig. 2 displays the XRD diffractograms of the produced SiBFCs that consisted of many characteristic peaks assigned to altering crystalline lattices. All the observed peaks in the pristine sample (SiBF0 without bio-silica incorporation) were due to the main hexagonal crystalline lattice of $BaFe_{12}O_{19}$ and tallied to the ICDD card number 00-039-1433 with crystal configurations of $a = b = 0.5894$ nm, $c = 2.3215$ nm, $\alpha = \beta = 90°$ and $\gamma = 120°$. Nevertheless, the appeared peak at 27.52° was due to the monoclinic crystalline phase of $Ba_2Fe_2O_5$ that corresponded to the ICDD card number 00-043-0256. The XRD pattern of SiBF1 sample revealed that the replacement of Fe^{3+} in the barium ferrite lattice by Si^{4+} ions at one wt% of SiO_2 did not cause any significant changes of the crystal structures. The appearance of two dominant peaks in SiBF1 confirmed the formation of the major phase of $Ba_3Fe_{32}O_{51}$ and minor phase of barium iron silicate $BaFeSi_4O_{10}$. The occurrences of sharp XRD peaks were due to the hexagonal crystal lattice of $Ba_3Fe_{32}O_{51}$ that corresponded to the ICDD card number 00-041-0846 with the crystal configurations of $a = b = 0.5892$ nm, $c = 2.3198$ nm, $\alpha = \beta = 90°$ and $\gamma = 120°$. Moreover, the peaks centered at 26.52° and 35.48° were assigned to the tetragonal crystal lattice planer orientation of $BaFeSi_4O_{10}$ (tallied to the ICDD card number 00-003-0402). The XRD pattern of the composite prepared with two wt% of bio-silica revealed a new phase of barium silicate $Ba_3Si_5O_{13}$ with the monoclinic crystalline lattice structure (agreed to the ICDD number 00-026-0179). The composite containing three wt% of bio-silica exhibited the dominant barium iron silicate $BaFeSi_4O_{10}$ ordered phase. It was claimed that the replacement of Fe^{3+} ions by Si^{4+} in the produced BFCs could create another significant hexagonal phase of

$Ba_3Fe_{32}O_{51}$ and a tetragonal phase of $BaFeSi_4O_{10}$. The observed broadening in the XRD peaks for all the studied SiBFCs is attributed to the insolvent of lattice strain and nano-crystallites size confinement effects [23,24].

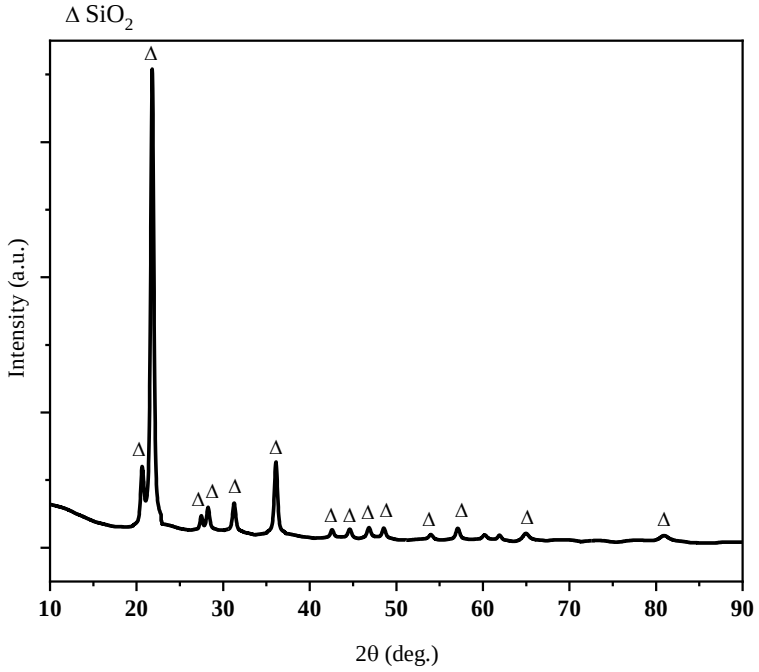

Fig. 1. The XRD pattern of the bio-silica (SiO_2) at 1000 °C

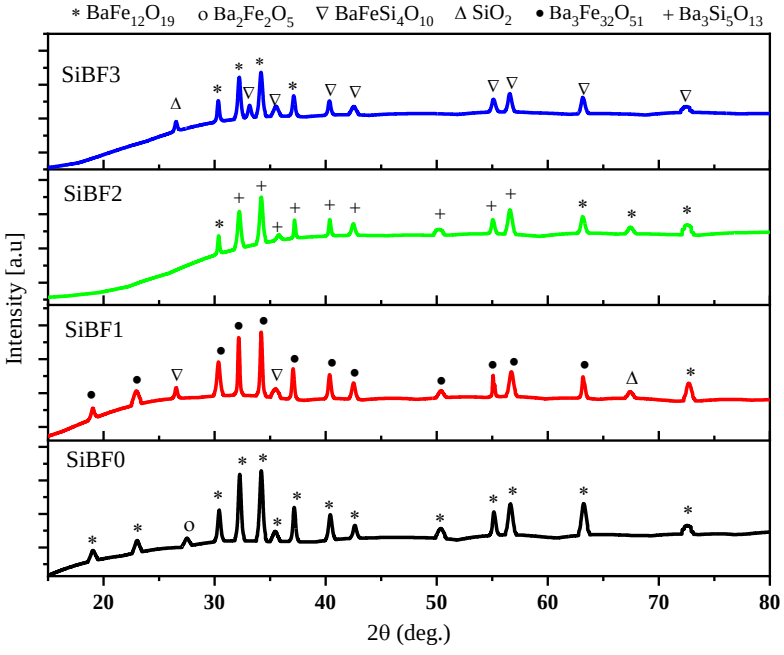

Fig. 2. The XRD patterns of the Si^{4+} undoped and doped SiBFCs

Fig. 3 shows the SEM micrographs of the prepared SiBFCs, which consisted of hexagonal particle morphology (particle size of about 0.5 μm) with irregular microstructures packing. These porous composite particles were attached to each other and formed some intergranular pores. It can be argued that the substitution of the Fe^{3+} by Si^{4+} (from the bio-silica) in the produced BFCs that created a new tetragonal phase of $BaFeSi_4O_{10}$ were closely packed to the hexagonal phase of $Ba_3Fe_{32}O_{51}$. In the intergranular pores of these composite particles phase, the Si^{4+} were preferentially occupied in the lattice sites, imparting more porosity to the microstructures. These pores, in turn, favored the entrapment of the MW, wherein the finer particles present in the pores could scatter the MW randomly in all directions, thereby modifying the MW absorption characteristics of the composites. In all the prepared SiBFCs, these distinct types of surface morphologies and particle distribution were responsible for the changes in the magnetic and reflection loss properties.

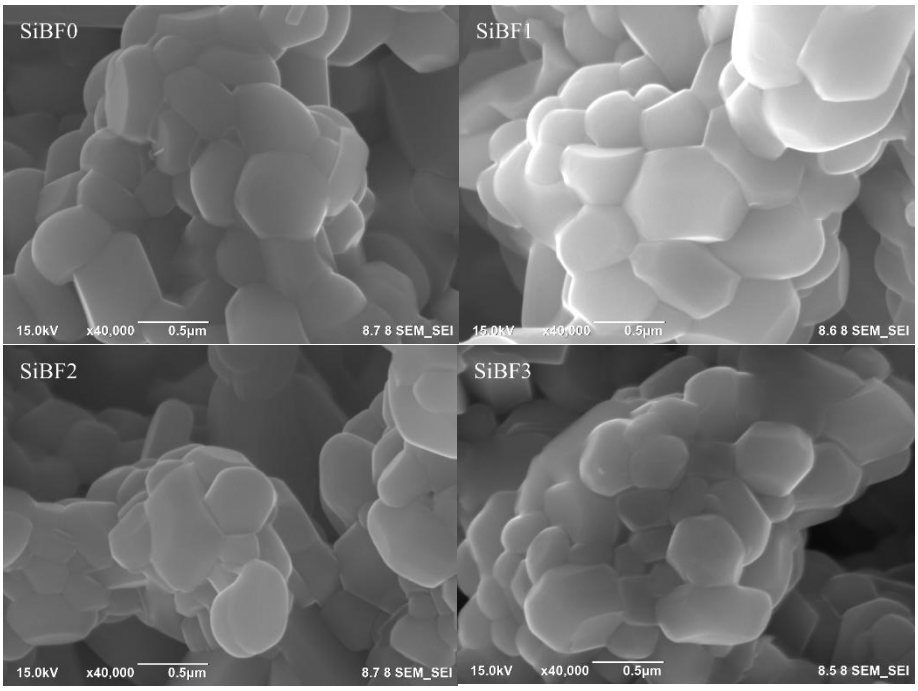

Fig. 3. The SEM images of the SiBFCs

Fig. 4 represents the room temperature hysteresis loops (*M-H* curves) of the as-synthesized SiBFCs. The values of saturation magnetization of the studied SiBFCs are decreased (from 39.5 to 30.4 emu/g) significantly with the addition of bio-silica (from 1 to 3 wt%) into the composites. However, the coercive field values (altered from 775.9 – 811.0 Oe) are not affected appreciably due to the increase in bio-silica contents. The maximum reduction in the saturation

magnetization displayed by the SiBF3 sample is ascribed to the presence of dominant tetragonal barium iron silicate ($BaFeSi_4O_{10}$) crystalline phase in the composite. The coercive field values of the composites are observed to increase slightly due to the rise in bio-silica contents. With the increase in bio-silica content from 0 to 2 wt%, the area of the hysteresis loop of the SiBFCs enlarges from 15.9 to 31.0 kOe.emu/g, respectively. Furthermore, the hysteresis loop area of SiBFC shrinks at three wt% of bio-silica. The observed change in the magnetic permeability of SiBFCs with the addition of bio-silica into the composites is majorly attributed to the alteration in the saturation magnetization and coercive field values.

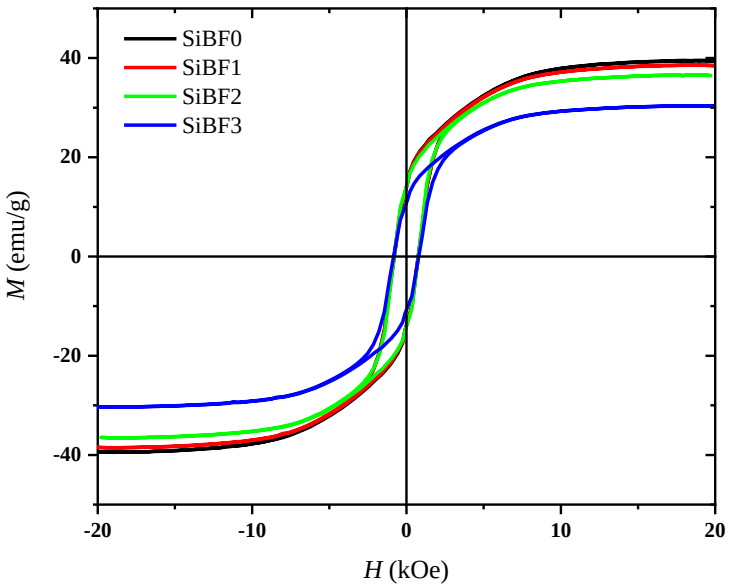

Fig. 4. The room temperature hysteresis loops (*M-H*) of the SiBFCs

Fig. 5(a) illustrates the frequency-dependent complex relative permeability ($\mu_r = \mu' + \mu''$) of all the SiBFCs. The real part of the permeability (μ') specifies a gradual decrease in their magnetic energy storing capacity due to polarization of magnetic dipole in higher MW frequency. The μ' value of the composites drops from about 2.5 (for SiBF0 and SiBF2) and 0.9 (for SiBF1 and SiBF3) to near-zero with the increase in bio-silica content from 0 to 3 wt%. Meanwhile, the observed magnetic loss is revealed by the imaginary part of the permeability (μ''). The μ'' value of SiBF0 and SiBF2 declines rapidly from 0.9 to near-zero and almost remains constant near-zero for SiBF1 and SiBF3 due to the relaxation process, which generates dissipation energy of MV as thermal energy.

Fig. 5(b) displays the complex relative permittivity ($\varepsilon_r = \varepsilon' + \varepsilon''$) of all the SiBFCs in the frequency range of 8-13 GHz. The ε' and ε'' are the real and imaginary parts of the permittivity, respectively. The ε' value indicated the lack of energy absorption by the composite from an externally applied electric field [25]. The ε' values of the composite without bio-silica doping (SiBF0) were dropped sharply. Conversely, the ε' values of the composites containing bio-silica (1 to 3 wt%) were increased significantly above 12 GHz due to the electrical dipole polarization. The value of ε'' (called the dielectric loss factor) denotes the electrical energy dissipation ability of the SiBFCs. The ε'' values of all SiBFCs were decreased gradually with the rise in the frequency.

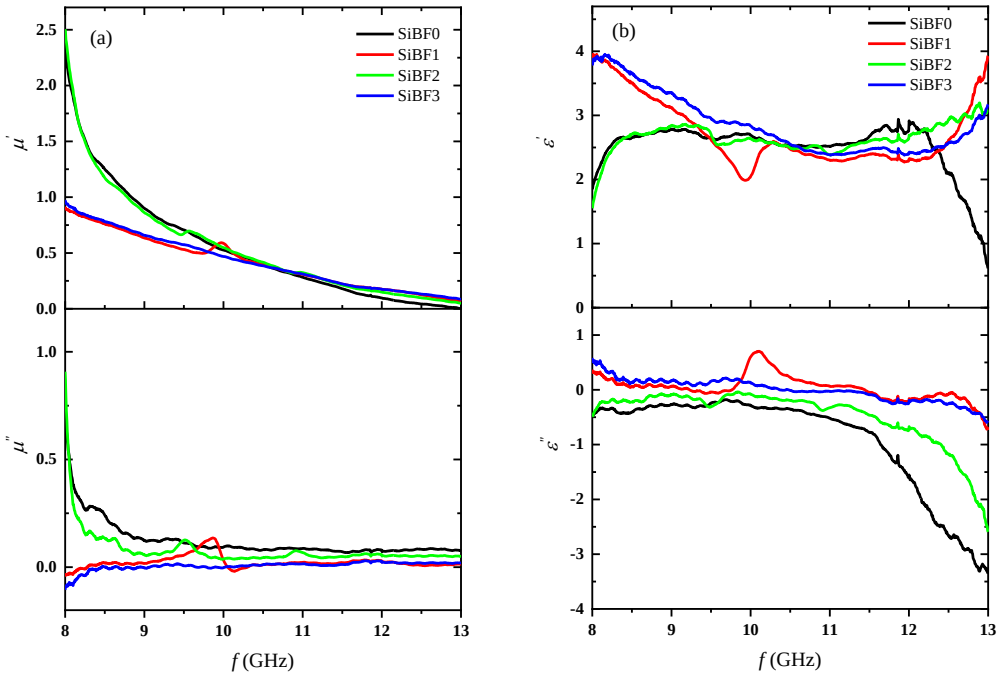

Fig. 5. Frequency-dependent complex relative (a) permeability and (b) permittivity of the SiBFCs with a thickness of 0.5 cm

The MW absorption properties of SiBFCs were evaluated by calculating the values of reflection loss (R_L) based on the transmission/reflection line theory [26–28]:

$$R_L = -20\log\left|\frac{Z_{in} - Z_0}{Z_{in} + Z_0}\right| \quad (4)$$

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\varepsilon_r}} \tanh\left[j \frac{2\pi f d}{c} \sqrt{\mu_r \varepsilon_r} \right] \quad (5)$$

where Z_0 and Z_{in} are the corresponding intrinsic impedance in a vacuum and the input impedance in the material, f represents the MW frequency, d denotes the sample thickness, and c is the light speed in the vacuum.

Fig. 6 illustrates the frequency-dependent changes in the R_L values of the prepared SiBFCs. The MW absorption ability of materials is determined by two crucial factors i.e. R_L and bandwidth. The value of R_L for SiBF0 was valued less than -10 dB at the frequency of 8.35, 9.37, and 10.60 GHz. The band positions (peak frequencies) of the composites disclose a significant shift with the change in bio-silica contents with the bandwidth of less than 1 GHz. The sample prepared with one wt% of bio-silica (SiBF1) reveals the lowest R_L value of -28.56 dB at the frequency of 12.18 GHz. This disclosure affirms that the MW absorption capacity of the proposed SiBFCs can be essentially customized by controlling the impurity or dopant (bio-silica) concentration, fulfilling the application demand in the X band. Some strategies were employed to modify the dielectric and magnetic parameters including the single trivalent cation substitution and different combinations of cation to minimize the MW reflection loss [27,29]. Compared to the earlier materials, present bio-silica based composites are cheaper and easy to prepare.

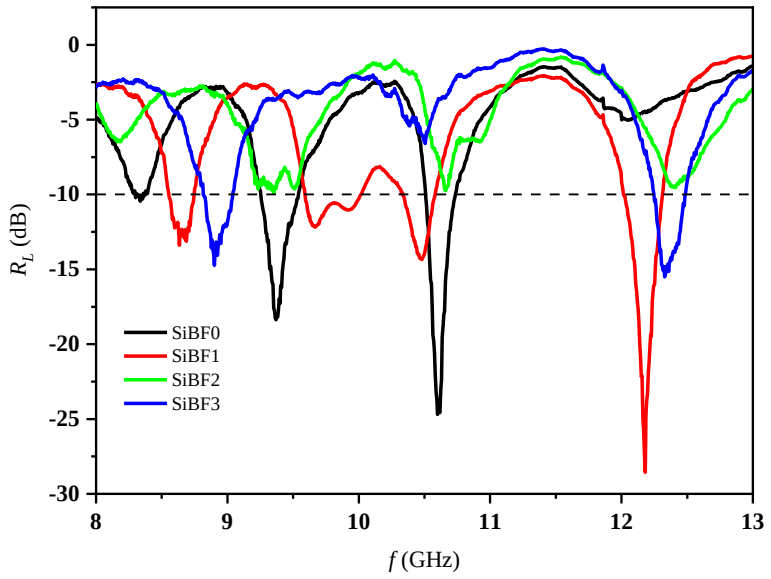

Fig. 6. Reflection loss as a function of the MW frequency for the studied SiBFCs of 0.5 cm thick

4. Conclusions

Considering the MW absorption potency of barium ferrites, we synthesized four SiBFCs (without and with bio-silica activation) via the modified solid-state reaction approach. Rice husk derived abundant and cheap bio-silica was doped into the barium ferrites to improve their magnetic and MW absorption attributes. The structure of the studied composites revealed the presence of various crystalline phases. The surface of SiBFCs disclosed hexagonal morphology with a particle size of about 0.5 μm. The surface morphology, structure, MW reflection loss, permittivity, permeability, coercivity, saturation magnetization, and MW absorption of the proposed SiBFCs were found to be sensitive to the substitution of Fe^{3+} by Si^{4+} ions into the composite matrix. It was established that the MW absorption ability of the newly composed SiBFCs can be tailored by adjusting the bio-silica content. Present knowledge may contribute towards the development of the SiBFCs based MW absorption devices.

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