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Neodymium ions activated barium ferrite composites for microwave X-band absorber applications: synthesis and characterisations

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Abstract:	Some composites of barium ferrites activated with the neodymium ions (Nd 3+) of composition (20)BaO:(80- x)g-Fe 2 O 3 :(x)Nd 2 O 3 (x = 0, 1 and 2 mol%) were synthesised using the modified mechanical alloying for the first time. The influence of varying Nd 3+ concentrations on the morphologies, microstructures, and magnetic characteristics of these composites were evaluated. In addition, the microwave (MW) reflection loss, complex relative permittivity and permeability of the studied composites in the frequency range of 8.2 – 12.4 GHz were analysed using the Nicholson-Ross-Weir (NRW) method. The inclusion of Nd 3+ in the proposed composites was discerned to influence significantly the permittivity, permeability and reflection loss in the MW X-band region.
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Response to Reviewers:	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.

To
The Editor,
Composites Communications

Dear Sir / Madam,

We are pleased to submit our original manuscript entitled **Neodymium ions activated barium ferrite composites for microwave X-band absorber applications: synthesis and characterisations** authored by Wahyu Widanarto, Ananda Iqbal Ekaputra, Mukhtar Effendi, Wahyu Tri Cahyanto, Sib Krishna Ghoshal, Candra Kurniawan, Erfan Handoko, Mudrik Alaydrus, for the peer-review and publication in **Composites Communications.**

We also declare that the manuscript has not been submitted to any other journal and is not under consideration for publication elsewhere, as well as no conflict of interest exists. If it is accepted, it will not be published elsewhere in the same form, in any language, without the written agreement of the publisher. All Authors have agreed with this submission.

We look forward to hearing from you at your earliest convenience.

Best regards,

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The Editor

Composites Communications

Manuscript Number: COCO-D-20-00008

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.

Editor and Reviewer comments:

Reviewer #1:

In this research, the author synthesized a modified neodymium ion (Nd3+) activated barium iron (20) BaO: (80-x)γ-Fe2O3: (x) Nd2O3 (x = 0, 1, and 2 mol%) using an improved mechanical method Some ferrite composites were alloyed for the first time, and some barium ferrite composites (BFC) doped with Nd3+ were prepared to determine the feasibility of increasing the MW X-band absorption frequency by reducing the Hc and Ms values. The effects of different Nd3+ concentrations on the morphology, microstructure and magnetic properties of these composites were evaluated. In addition, the Nicholson-Ross-Weir (NRW) method was used to analyze the microwave (MW) reflection loss, relative permittivity and permeability (complex number) of the studied composite materials in the frequency range of 8.2-12.4 GHz. I considered it can be published after a major revision in Composites Communications. The reasons are as follows:

1. *The article states that several physical and chemical techniques have been developed to synthesize submicron particles of BHF, but several physical and chemical techniques are not introduced.*
   Response: Thanks for the valuable question and advice. Please note that we have introduced these techniques in the revised MS as suggested.

2. *During the calcination of iron sand (for 3 h at 850 ° C) and BaCO3 powder (for 1 h at 350 ° C), it is not stated whether the atmosphere is air. Crush some particles and bind with resin, no description of what resin is. There is no preparation process flow in the article.*

   Response: Thanks for the useful question and for asking the explanation. Please note that the calcination was conducted in the air atmosphere. The preparation protocols of the samples for the VNA measurement have been added in the revised manuscript.

3. *In the preparation process engineering, there is no introduction to various mechanical methods. Fewer scanned images, no EDS test, and no TEM image.*

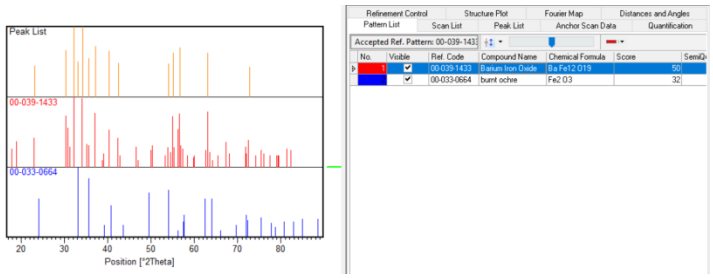
Response: Thanks for the valuable suggestions. Please note that at present, we do not have the EDS and TEM facilities in our university. However, your advice is well taken for future work and we will try to complete them.

4. *the author should add the referees about microwave absorber in the introduction such as "Composites Part B, 2019, 179: 107417., RSC Advances, 2019, 9, 25932-25941.". "Composites Part A, 2020, 128: 105687. Journal of Materials Science: Materials in Electronics, 2019, 30(14): 1347413487.,."*

Response: Thanks for the advices and useful suggestions. Please note that, all the cited references have been added in the revised MS.

5. *No standard PDF card in XRD picture. No explanation was given as to why the real part of the dielectric constant increased. The article did not explain why the X-band test was performed on the sample, which was not convincing.*

Response: Thanks for the question and useful advices. Please note that we have replaces the PDF standard with the symbols that indicated a compound. The standard PDF card is presented hereunder:



The X-band test was carried out using the prepared sample to determine its microwave-absorbing characteristics so that it can be used for anti-radar applications because the radar works at the X band frequency region of 8.2 - 12.4 GHz.

6. *The 3D reflection loss graph is not available, and the absorbing performance of the sample cannot be seen intuitively. No effective bandwidth comparison. When Nd is not doped, the sample has better absorbing performance. What is the significance of this article.*

Response: Thanks for the critical question and advice. Please note that due to the constant thickness of the present sample (0.5 cm) we could not make the 3D reflection loss plot. However, it was shown that the addition of Nd caused a widening of the bandwidth in the 9-10 GHz frequency region and shift the reflection loss peak position of the NdBFCs to higher frequency.

7. *The article said that the mechanical method was used, and there was no comparison with other methods, which could not explain the excellent performance of the samples synthesized by this method.*

Response: Thanks for the valuable question and suggestions. Please note that the efforts are made to obtain some cheap and easily available MW absorbing materials. In fact, so far no reports are available for the comparison.

8. *The entire article is not innovative enough. It simply lists test data, but some necessary tests have not been performed, and the mechanism analysis of microwave absorption is not comprehensive.*

Response: Thanks for the critical observation and remarks. Please note that for the first time we made this effort with several limitations toward a comprehensive study. However, we are trying to perform more tests and characterizations for the future communications.

Reviewer #2:

In this manuscript, neodymium ions activated barium ferrite composites were synthesized by modified mechanical alloying method. The authors have clearly demonstrated the effects of Nb ions on morphologies, crystal structures, magnetic properties, as well as microwave absorption properties. The work is original and creative. However, there are also some deficiencies that need to make some revisions in order to improve your paper before publishing it in Composites Communications.

1. *In the Fig.1, is there one phase or more than one phase that we can see in the SEM images? If there is more than one phase, you'd better mark different phases. In addition, you said "The average size of the NdBFC grains was decreased and the porosity was increased with the increase in the Nd3+ levels." It is not clear to observe the decreasing grain size and increasing porosity. I think the authors should mark the crystal size in the Fig.1. If it is not obvious in SEM, I advise you do a TEM testing.*

Response: Thanks for the valuable suggestions. Please note that at present, we do not have the TEM facilities in our laboratory. However, your advice is well taken for future work.

2. *In the Introduction, is it the "H_c and M_s" or "H_c and M_s" in the last sentence of the first paragraph? Please pay attention to the consistency of the signs.*

Response: Thanks for the valuable suggestions. Please note that we have revised the MS and added as advised.

3. *In the Fig. 4, (a) and (b) are different sizes, please revise it.*

Response: Thanks for the valuable suggestions. Please note that we have added it in the revised MS.

4. *Compare the NdBFC1 with NdBFC2, the permeability is a little different no matter for μ' or μ'', but the permittivity is almost the same for ε' and ε''. Can you explain it?*

Response: Thanks for the question. Please note that the NdBFC1 and NdBFC2 consisted of two phases which were almost the same both in terms of the structure and constituent

elements, only the composition of $BaFe_9O_{12}$ and $Ba_3Fe_{32}O_{51}$ made the difference. This made the permeability of both sample almost similar.

5. *For the Fig.5, you said "The observed shift in the peak frequency (toward the higher side) with the increase in the Nd^{3+} doping level was mainly due to the enhanced porosity and emergence of a new barium hexaferrite phase ($Ba_3Fe_{32}O_{51}$) in the composite structure." If $Ba_3Fe_{32}O_{51}$ (a new phase) is a non-negligible factor according to what you said in the paper, why can I hardly see the significant peak shift between the NdBFC1 and NdBFC2 samples?*

   Response: Thanks for the question. Presently, we do not know the reason for such insignificant peak shift. The new phase might have contributed to the other properties of the composite. However, it is worth to identify the mechanism for such disclosure.

6. *The authors clearly demonstrated the highlights and I also agree with the novelty of your paper. However, can you list some microwave absorption property in other works about barium ferrite or doped barium ferrite materials? If so, it is helpful to compare your materials with others' materials to prove superiority of your NdBFCs.*

   Response: Thanks for the encouragements and questions. Please note that we have added some properties in the revised MS for better comparison.

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

- Neodymium ions activated barium ferrite composites (NdBFCs) through the modified mechanical alloying
- Changing porous microstructure, crystal structure and saturation magnetization of the NdBFCs due to adding neodymium ions
- Shifting complex relative permittivity, permeability and reflection loss position of the NdBFCs by controlling the concentration of neodymium ions

Neodymium ions activated barium ferrite composites for microwave X-band absorber applications: synthesis and characterisations

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ABSTRACT

Some composites of barium ferrites activated with the neodymium ions (Nd^{3+}) of composition (20)BaO:(80-*x*)γ-Fe_2O_3:(*x*)Nd_2O_3 (*x* = 0, 1 and 2 mol%) were synthesised using the modified mechanical alloying for the first time. The influence of varying Nd^{3+} concentrations on the morphologies, microstructures, and magnetic characteristics of these composites were evaluated. In addition, the microwave (MW) reflection loss, complex relative permittivity and permeability of the studied composites in the frequency range of 8.2 – 12.4 GHz were analysed using the Nicholson-Ross-Weir (NRW) method. The inclusion of Nd^{3+} in the proposed composites was discerned to influence significantly the permittivity, permeability and reflection loss in the MW X-band region.

Keywords: neodymium, magnetic properties, permittivity, permeability, reflection loss

1. Introduction

Recent studies revealed that the inclusion (as dopant) of various divalent or trivalent rare-earth ions (REIs) into the crystal structure of the barium hexaferrites (BHFs) can change their magnetic properties (such as coercivity (H_c), remanent magnetisation (M_r) and saturation magnetisation (M_s), natural resonant frequency, complex permeability ($\mu' + i\,\mu''$) and permittivity ($\varepsilon' + i\,\varepsilon''$) as well as the magnetic field anisotropy [1,2]. Consequently, the influence of electromagnetic interference on such ferrites can be strengthened. Additionally, the excellent compatibility related to the hexagonal lattice structure and extraordinary relaxational attributes of the REIs (act as dopant or activator also called surrogate ions) has also been exploited over the decades by activating them inside various crystals, glasses and glass-ceramics. It has been established that by doping these REIs inside the BHFs their magnetic and MW absorption properties can remarkably be altered and customised [3–7]. In addition, the REIs-doped BHFs that possess weak H_c and low M_s are beneficial for enhancing the MW absorption capacity [8–11]. Based on these factors, we prepared some barium ferrite composites (BFCs) doped with Nd^{3+} to determine the feasibility of enhancing the MW X-band absorption frequency by reducing the values of H_c and M_s.

Of late, several physical and chemical techniques have been developed to synthesise submicron particles of BHFs such as the solid-state reaction, mechanical alloying, co-precipitation, electrostatic self-assembly, sol-gel process and combustion methods [12–19]. These methods play a significant role to control the morphologies (sizes and shapes), structures, and magnetic characteristics of the produced BHFs. In addition, dedicated efforts have been made to modify the structures, morphologies and magnetic properties of various functional materials to obtain the best MW absorbing attributes [19–22]. Amongst these synthesis techniques, the mechanical alloying being a low-cost and simple approach can produce BHF powder with distinct attributes such as the wide specific area of the grain boundary and high atomic volume fraction at the boundary [8]. Despite some efforts, an accurate method to synthesise the Nd^{3+}-doped BHF composites with controlled magnetic and MW absorption characteristics has been deficient.

Considering the diverse applied interests of REIs-doped BHFs we prepared some new type of Nd^{3+}-doped BF composites (hereinafter named as NdBFCs) using the modified mechanical alloying and characterised them. The role of different Nd^{3+} doping levels on the structural, morphological, magnetic and MW absorption traits of these NdBFC was determined. Besides, the MW reflection loss (R_L) of the proposed NdBFCs in the frequency (f) range of 8.2-12.4 GHz was evaluated to determine their feasibility in favour of MW X-band absorber applications.

2. Material and Methods

Three NdBFCs of chemical composition (20)BaO:(80-x)γ-Fe_2O_3:(x)Nd_2O_3, where x = 0, 1 and 2 mol% were prepared by the mechanical alloying strategy. Analytical grade chemical reagents (all in powder form) of barium carbonate [$BaCO_3$, Merck purity of 99%], neodymium oxide [Nd_2O_3, Sigma Aldrich of purity 99%] and natural gamma phase iron oxide [γ-Fe_2O_3, Sigma Aldrich of purity 99%] were utilised as the starting raw materials to prepare the proposed composites. Firstly, the iron sand and $BaCO_3$ powder were calcinated in an air atmosphere at 850 °C for 3 hours and 350 °C for an hour, respectively to acquire the natural γ-Fe_2O_3 by removing the existed carbon component. Then, the constituent materials with the nominal compositions were placed into a vial before being blended and milled using a Shaker Mill PPF-UG system for three hours in the on (5 min) and off (10 min) modes to achieve the homogeneous mixture of composite powders. Later, the fine powders were compacted to get pellets of 1 mm thick and 10 mm diameter [9]. The obtained pellets were strengthened using an electrical tube furnace (operated at 800 °C for an hour) and then sintered at 1100 °C for 5 hours with the heating rate of 10 °C/min in an air atmosphere before being cooled down normally to the

ambient temperature. Depending on the Nd^{3+} doing contents of 0, 1, and 2 mol% these pellets were coded as NdBFC0, NdBFC1, and NdBFC2. For the additional analyses, some of the pellets were crushed and bound using the epoxy resin to impart a rectangular shape consistent with the WR90 sample holder (dimension of 2.3 cm × 1.0 cm × 0.5 cm).

The morphology and microstructural analyses of the as-prepared NdBFCs were performed using the Hitachi (SU 3500) scanning electron microscopy (SEM) [9,11,23]. The crystal structures and phases of the NdBFCs were recorded on a SmartLab (3 kW) X-ray diffractometer equipped with Cu–Kα line of $\lambda \approx 0.1541874$ nm. An Oxford (1.2H) vibrating sample magnetometer (VSM) was used to measure the magnetic properties of the NdBFCs. A Keysight (PNA-L N5232A) vector network analyser (VNA) was used (in the range of 8–13 GHz) to measure the scattering parameters (S) of the prepared NdBFCs. The MW absorption measurement was carried out to yield the components of S (values of S_{11}, S_{12}, S_{21}, and S_{22}) wherein the values of S_{11} and S_{21} signified the coefficient of reflection (Γ) and transmission (T), respectively. The measured values of S_{22} and S_{12} were disregarded due to their equivalence to S_{11} and S_{21}, respectively. The Nicholson-Ross-Weir (NRW) method was followed to obtain the values of relative complex permeability (μ_r) and permittivity (ε_r).

3. Results and Discussion

Fig. 1 shows the SEM micrographs of the studied NdBFCs, which consisted of roughened porous microstructures. The substitution of Nd^{3+} into the crystal lattice of the NdBFCs was found to affect significantly the surface morphologies (grain sizes and shapes) and distributions of the grains. The average size of the NdBFC grains was decreased and the porosity was increased with the increase in the Nd^{3+} levels. It was argued that the enhanced porous structures are advantageous to achieve a longer propagation path for the electromagnetic waves, thereby effective for stronger reflections and scattering [24]. In addition, the magnetic characteristics and loss of MW reflection can be improved by increasing the porosity of the NdBFCs [11].

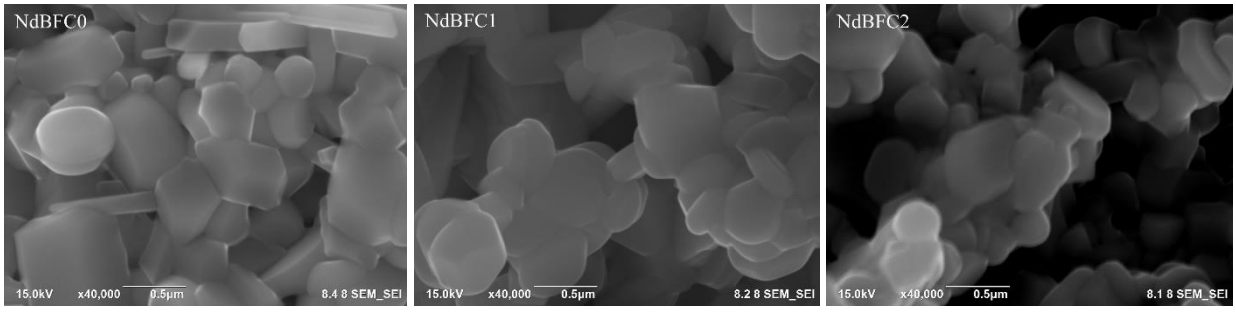

Fig. 1. The SEM images of the obtained NdBFCs

Fig. 2 shows the XRD pattern of the prepared NdBFCs, which comprised of several sharp peaks characteristics of different crystalline lattice planes. The observed peaks for the NdBFC0 sample (without Nd^{3+} doping) were allocated to the major hexagonal crystal structure of $BaFe_{12}O_{19}$ that matched to the ICDD number 00-039-1433 with lattice parameters $a = b =$ 0.5894 nm, c = 2.3215 nm, $\alpha = \beta = 90°$ and $\gamma = 120°$. Conversely, the α-Fe_2O_3 rhombohedral crystal structure (ICDD number 00-033-0664) appeared at 33.336°. The crystal structure of the studied NdBFCs was significantly altered with the inclusion of Nd_2O_3 of 1 mol% as seen from the XRD pattern of NdBFC1. The appearance of six new peaks in the NdBFC1 sample was consistent with the orthorhombic crystal structure of $BaNd_2O_4$ (ICDD number 00-042-1499). Finally, the NdBFC2 sample containing Nd_2O_3 of 2 mol% disclosed a phase transformation from the hexagonal $BaFe_{12}O_{19}$ to hexagonal $Ba_3Fe_{32}O_{51}$ (ICDD number 00-041-0846). It was affirmed that substitution of Nd_2O_3 into the crystal structures of the BFC indeed produced a new primary hexagonal crystalline phase from $Ba_3Fe_{32}O_{51}$. The observed broadening in the diffraction peaks associated with all the prepared NdBFCs was due to the emergence of the quantum size effects (nanoscale particles) and lattice strain [25,26].

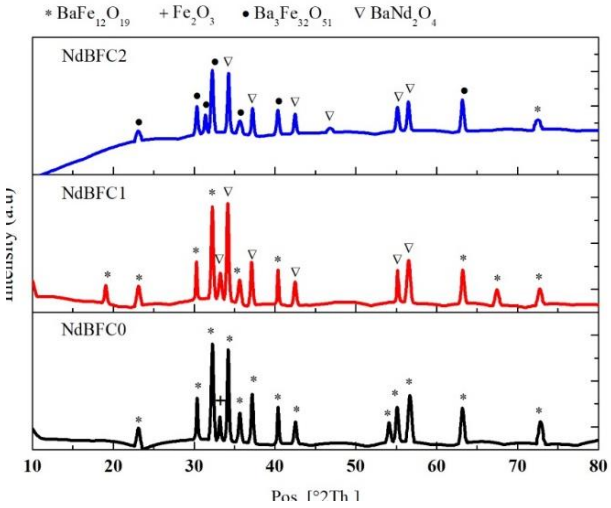

Fig. 2. The XRD patterns of the prepared NdBFCs.

Fig. 3 illustrates the magnetic field (H) dependent magnetisation (M) response (hysteresis loop) of the NdBFCs. The values of M_s, M_r, and H_c for the pristine sample (NdBFC0) were 33.54 emu/g, 11.56 emu/g, and 726.06 Oe, respectively. The value M_s of the composites changes significantly because of expansion concentration of Nd_2O_3. The value of M_s was lowered with the addition of 1 mol% Nd_2O_3 into the composite (NdBFC1) which was due to the formation of anti-magnetic material ($BaNd_2O_4$). Furthermore, the value of M_s was increased due to the incorporation of 2 mol% of Nd_2O_3 into the composite (NdBFC2) and became higher than

NdBFC0, which was ascribed to the occurrence of a new phase of barium hexaferrite ($Ba_3Fe_{32}O_{51}$), increased porosity and higher surface roughness. It was argued that such enhanced porosity with extremely roughened granular surfaces could alienate the magnetic domain walls, enabling regular polarisations of the unpaired spin magnetic moments when a magnetic field is applied externally.

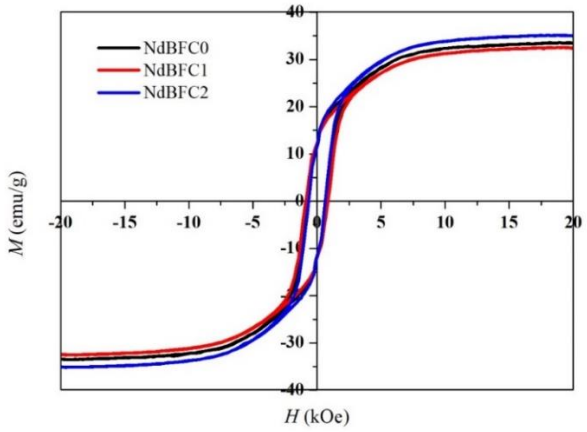

Fig. 3. The hysteresis loops of the as-prepared NdBFCs.

Fig. 4(a) depicts the frequency (ranged from 8.2–12.4 GHz) dependence of the permeability (real, μ' and imaginary, μ'' parts) of the NdBFCs. The value μ' of all the NdBFCs was increased significantly in lower MW frequency region and then decreased gradually in the higher frequency region. The onset of the decreasing permeability was approximately 8.5 GHz for the undoped sample (NdBFC0) and approximately 9.3 GHz for the doped composites (NdBFC and NdBFC2). The magnetic energy storing ability of the proposed NdBFCs was primarily ascribed to the natural spin magnetic moments' polarisations. Moreover, the value of μ'' (signifies the magnetic loss factor) was decreased rapidly to zero at approximately 8.5 GHz for the pristine sample and 9.3 GHz for the doped ferrites, confirming the absence of magnetic loss in the higher region. It is important to note that the imaginary permeability plays an essential role in the microwave absorption traits of hexaferrite [27] via the relaxation of the domain wall resonance [28].

Fig. 4(b) shows the frequency (ranged from 8.2–12.4 GHz) dependent relative complex permittivity (real, ε' and imaginary, ε'' components) of the NdBFCs. The intrinsic electric dipole polarisations in the MW frequency domain of the NdBFCs were responsible for the emergence of ε' and ε'' [29,30]. The real permittivity signified the energy stored in the material from an external electric field [31]. For the undoped or pristine sample (NdBFC0), the real permittivity

was first increased up to 3 with the increase in the frequency and then decreased to nearly -1 above 12 GHz. Conversely, the real permittivity for the doped samples (NdBFC1 and NdBFC2) was first dropped in the low-frequency region and then increased up to 2 with the rise in the frequency. The ε'' of the undoped specimen (NdBFC0) revealed a shrinking tendency while for the doped samples (NdBFC1 and NdBFC2) it dropped rapidly below 8.2 GHz before being reached nearly to zero in the high-frequency region. The permittivity of the NdBFC1 and NdBFC2 sample was almost similar because they consisted of two phases, which were almost the same in terms of both structure and constituent elements, only the composition of $BaFe_9O_{12}$ and $Ba_3Fe_{32}O_{51}$ made the difference. In addition, a significant increase in the permittivity is caused by the dipolar polarisation originated from the bond formation between O^{2-} with Ba^{2+} and Nd^{3+} in the non-magnetic $BaNd_2O_4$ phase.

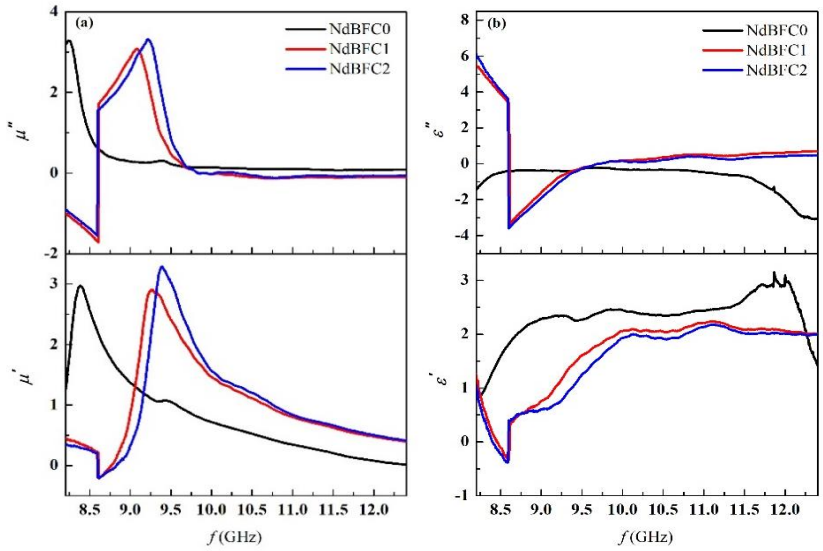

Fig. 4. Frequency-dependent (a) permeability and (b) permittivity of the NdBFCs with the thickness of 0.5 cm.

Fig. 5 displays the f against R_L plot of the synthesised NdBFCs. The undoped specimen (NdBFC0) revealed three prominent absorption bands centred at approximately 8.4, 10, and 11 GHz with the corresponding R_L values of -37, -17, and -16 dB. Meanwhile, the graph for both the doped samples NdBFC1 and NdBFC2 showed a similar spectral pattern, which consisted of 4 durable absorption bands with values less than -10 dB accompanied by a significant shift. The observed shift in the peak frequency (toward the higher side) with the increase in the Nd^{3+} doping level was mainly due to the enhanced porosity and emergence of a new barium hexaferrite phase ($Ba_3Fe_{32}O_{51}$) as well as $BaNd_2O_4$ in the composite structure. Consequently, the values of H_c, M_s, M_r, permeability, and permittivity of the prepared NdBFCs were modified. This in turn, led to an alteration in the natural resonance frequency and impedance [32–34],

thereby a shift in the *peak* R_L value towards higher frequency. It was also shown that the addition of Nd^{3+} caused a widening of the bandwidth in the 9-10 GHz frequency region and the R_L signals were more reversible to the MW frequencies compared to the previous findings [8,35,36]

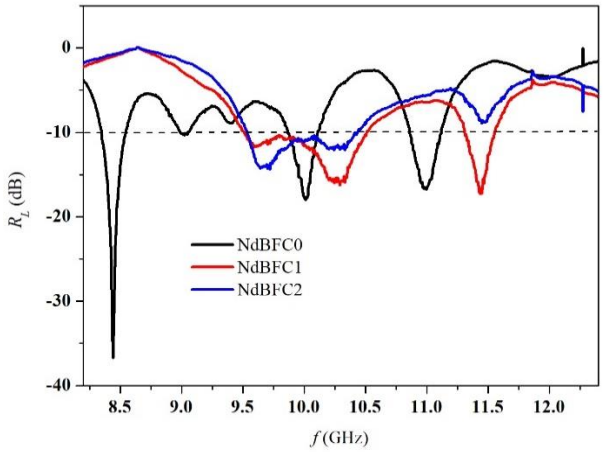

Fig. 5. The MW reflection loss as a function of the frequency for the studied NdBFCs of 0.5 cm thick.

4. Conclusions

Following the modified mechanical alloying, a new type of Nd^{3+} activated BFCs was synthesised for the first time and characterised. The incorporation of Nd^{3+} into the hexagonal crystal lattice of BFs was found to influence considerably the surface morphologies, microstructures, crystal structures and phases, magnetic properties, complex permittivity and permeability as well as the reflection loss in the MW frequency domain of the resultant NdBFCs. The obtained NdBFCs consisted of highly porous and roughened microstructures with a homogeneous distribution of grains. Undoped sample revealed the presence of $BaFe_{12}O_{19}$ (hexagonal lattice) and α-Fe_2O_3 (rhombohedral lattice) crystal phases. The doped specimens (NdBFC1 and NdBFC2) showed the new hexagonal crystalline phase of $Ba_3Fe_{32}O_{51}$ and $BaNd_2O_4$. The achieved minimum reflection loss with the absorption of several MW frequencies (in the range 8.2-12.4 GHz) shown by the proposed NdBFCs are established to be beneficial for the MW X-band absorber applications.

Acknowledgements

The authors are grateful to the Universitas Jenderal Soedirman, Kemenristekdikti Indonesia and UTM Malaysia (MOHE, FRGS/KPT 5F050 & UTMFR 20H65) for financial supports.

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optical, dielectric and magnetic properties of $Mn_{1-x}Cu_xFe_{1.85}La_{0.15}O_4$ spinel nanoferrites, Ceram. Int. 43 (2017) 15868–15879. doi:10.1016/j.ceramint.2017.08.160.

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Composites Communications

Neodymium ions activated barium ferrite composites for microwave X-band absorber applications: synthesis and characterizations

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Abstract:	Some composites of barium ferrites activated with the neodymium ions (Nd3+) of composition (20)BaO:(80-x)γ-Fe2O3:(x)Nd2O3 (x = 0, 1 and 2 mol%) were synthesized using the modified mechanical alloying for the first time. The influence of varying Nd3+ concentrations on the morphologies, microstructures, and magnetic characteristics of these composites were evaluated. In addition, the microwave (MW) reflection loss, complex relative permittivity, and permeability of the studied composites in the frequency range of 8.2 – 12.4 GHz were analysed using the Nicholson-Ross-Weir (NRW) method. The inclusion of Nd3+ in the proposed composites was discerned to influence the permittivity, permeability and reflection loss significantly in the MW X-band region.
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To
The Editor,
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Dear Sir / Madam,

We are pleased to submit our original manuscript entitled **Neodymium ions activated barium ferrite composites for microwave X-band absorber applications: synthesis and characterisations** authored by Wahyu Widanarto, Ananda Iqbal Ekaputra, Mukhtar Effendi, Wahyu Tri Cahyanto, Sib Krishna Ghoshal, Candra Kurniawan, Erfan Handoko, Mudrik Alaydrus, for the peer-review and publication in **Composites Communications.**

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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.

Editor and Reviewer comments:

Reviewer #1: The author has well replied all my questions and revised their manuscript according to my suggestions. I accept publish this manuscript.

Reviewer #2: The authors have revised the manuscript and answered the most of questions. However, there were also some questions about which the author did not give the explanation. After answering these questions, it can be considered to be published in Composites Communications.

1. *In terms of Q1, the author did not answer the question that 'In the SEM images of Fig.1, is there one phase or more than one phase? If there is more than one phase, you'd better mark different phases in the SEM.*
   Response: Thanks for the the question. Please note that from the SEM images it is difficult to determine and distinguish the specific phases. Moreover, it is expected that both phases are present and intermixed which can be seen clearly from the varied shapes and crystalline grain orientations. The SEM micrographs are used to evaluate the surface morphology of the sample. However, XRD characterization revealed that each sample consists of two constituent phases.

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   Response: Thanks for the question. Please note that the observed little difference in the permeability between NdBFC1 and NdBFC2 may be due to the existence of different amounts of the $BaFe_9O_{12}$ and $Ba_3Fe_{32}O_5$ phases with different magnetic properties. Thus, both phases played an important role in the permeability changes associated with the magnetic properties. However, the almost similar nature of the permittivity for both NdBFC1 and NdBFC2 is due to the existence of the non-magnetic phase of $BaNd_2O_4$ in the samples, resulting in the intrinsic electric dipole polarizations.

3. *In terms of Q5, if the author was not sure about the mechanism of new phase to properties, you can explain it with possible words but cannot write in the paper with certainty. For example, you said 'the observed shift in the peak frequency (toward the higher side) with the increase in the Nd3+ doping level was mainly due to the enhanced porosity and emergence of a new barium hexaferrite phase (Ba3Fe32O51) as well as BaNd2O4 in the composite structure' in the manuscript. I think it is not precise, which easily mislead others who read this paper. You should change the words with 'maybe, may, possibly, probably', and so on. In addition, please check the whole manuscript and revise it.*
   Response: Thanks for the valuable suggestions. Please note that we have revised it.

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

- Neodymium ions activated barium ferrite composites (NdBFCs) through the modified mechanical alloying
- Changing porous microstructure, crystal structure and saturation magnetization of the NdBFCs due to adding neodymium ions
- Shifting complex relative permittivity, permeability and reflection loss position of the NdBFCs by controlling the concentration of neodymium ions

Neodymium ions activated barium ferrite composites for microwave X-band absorber applications: synthesis and characterizations

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ABSTRACT

Some composites of barium ferrites activated with the neodymium ions (Nd^{3+}) of composition (20)BaO:(80-*x*)γ-Fe_2O_3:(*x*)Nd_2O_3 (*x* = 0, 1 and 2 mol%) were synthesized using the modified mechanical alloying for the first time. The influence of varying Nd^{3+} concentrations on the morphologies, microstructures, and magnetic characteristics of these composites were evaluated. In addition, the microwave (MW) reflection loss, complex relative permittivity, and permeability of the studied composites in the frequency range of 8.2 – 12.4 GHz were analysed using the Nicholson-Ross-Weir (NRW) method. The inclusion of Nd^{3+} in the proposed composites was discerned to influence the permittivity, permeability and reflection loss significantly in the MW X-band region.

Keywords: neodymium, magnetic properties, permittivity, permeability, reflection loss

1. Introduction

Recent studies revealed that the inclusion (as dopant) of various divalent or trivalent rare-earth ions (REIs) into the crystal structure of the barium hexaferrites (BHFs) can change their magnetic properties (such as coercivity (H_c), remanent magnetization (M_r) and saturation magnetization (M_s), natural resonant frequency, complex relative permeability ($\mu' + i\mu''$) and permittivity ($\varepsilon' + i\varepsilon''$) as well as the magnetic field anisotropy [1,2]. Consequently, the influence of electromagnetic interference on such ferrites can be strengthened. Additionally, the excellent compatibility related to the hexagonal lattice structure and extraordinary relaxational attributes of the REIs (act as dopant or activator also called surrogate ions) has also been exploited over the decades by activating them inside various crystals, glasses, and glass-ceramics. It has been established that by doping these REIs inside the BHFs their magnetic and MW absorption properties can remarkably be altered and customized [3–7]. In addition, the REIs-doped BHFs that possess weak H_c and low M_s are beneficial for enhancing the MW absorption capacity [8–11]. Based on these factors, we prepared some barium ferrite composites (BFCs) doped with Nd^{3+} to determine the feasibility of enhancing the MW X-band absorption frequency by reducing the values of H_c and M_s.

Of late, several physical and chemical techniques have been developed to synthesize submicron particles of BHFs such as the solid-state reaction, mechanical alloying, co-precipitation, electrostatic self-assembly, sol-gel process, and combustion methods [12–19]. These methods play a significant role in controlling the morphologies (sizes and shapes), structures, and magnetic characteristics of the produced BHFs. In addition, dedicated efforts have been made to modify the structures, morphologies and magnetic properties of various functional materials to obtain the best MW absorbing attributes [19–22]. Amongst these synthesis techniques, the mechanical alloying being a low-cost and simple approach can produce BHF powder with distinct attributes such as the wide specific area of the grain boundary and high atomic volume fraction at the boundary [8]. Despite some efforts, an accurate method to synthesize the Nd^{3+}-doped BHF composites with controlled magnetic and MW absorption characteristics has been deficient.

Considering the diverse applied interests of REIs-doped BHFs we prepared some new types of Nd^{3+}-doped BF composites (hereinafter named as NdBFCs) using the modified mechanical alloying and characterized them. The role of different Nd^{3+} doping levels on the structural, morphological, magnetic and MW absorption traits of these NdBFC was determined. Besides, the MW reflection loss (R_L) of the proposed NdBFCs in the frequency (f) range of 8.2-12.4 GHz was evaluated to determine their feasibility in favor of MW X-band absorber applications.

2. Material and Methods

Three NdBFCs of chemical composition (20)BaO:(80-x)γ-Fe_2O_3:(x)Nd_2O_3, where x = 0, 1 and 2 mol% were prepared by the mechanical alloying strategy. Analytical grade chemical reagents (all in powder form) of barium carbonate [$BaCO_3$, Merck purity of 99%], neodymium oxide [Nd_2O_3, Sigma Aldrich of purity 99%] and natural gamma phase iron oxide [γ-Fe_2O_3, Sigma Aldrich of purity 99%] were utilized as the starting raw materials to prepare the proposed composites. Firstly, the iron sand was calcinated in an air atmosphere at 850 °C for 3 hours to acquire the natural γ-Fe_2O_3. Meanwhile, the $BaCO_3$ powder was calcinated in an air atmosphere at 350 °C for an hour to achieve the BaO powder by removing the existing carbon component. Then, the constituent materials with the nominal compositions were placed into a vial before being blended and milled using a Shaker Mill PPF-UG system for three hours in the on (5 min) and off (10 min) modes to achieve the homogeneous mixture of composite powders. Later, the fine powders were compacted to get pellets of 1 mm thick and 10 mm diameter [9]. The obtained pellets were strengthened using an electrical tube furnace (operated at 800 °C for an

hour) and then sintered at 1100 °C for 5 hours with the heating rate of 10 °C/min in an air atmosphere before being cooled down normally to the ambient temperature. Depending on the Nd^{3+} doing contents of 0, 1, and 2 mol% these pellets were coded as NdBFC0, NdBFC1, and NdBFC2. For the additional analyses, some of the pellets were crushed and bound using the epoxy resin to impart a rectangular shape consistent with the WR90 sample holder (dimension of 2.3 cm × 1.0 cm × 0.5 cm).

The morphology and microstructural analyses of the as-prepared NdBFCs were performed using the Hitachi (SU 3500) scanning electron microscopy (SEM) [9,11,23]. The crystal structures and phases of the NdBFCs were recorded on a SmartLab (3 kW) X-ray diffractometer equipped with Cu–Kα line of $\lambda \approx 0.1541874$ nm. An Oxford (1.2H) vibrating sample magnetometer (VSM) was used to measure the magnetic properties of the NdBFCs. A Keysight (PNA-L N5232A) vector network analyzer (VNA) was used (in the range of 8–13 GHz) to measure the scattering parameters (*S*) of the prepared NdBFCs. The MW absorption measurement was carried out to yield the components of *S* (values of S_{11}, S_{12}, S_{21}, and S_{22}) wherein the values of S_{11} and S_{21} signified the coefficient of reflection (Γ) and transmission (*T*), respectively. The measured values of S_{22} and S_{12} were disregarded due to their equivalence to S_{11} and S_{21}, respectively. The Nicholson-Ross-Weir (NRW) method was followed to obtain the values of relative complex permeability (μ_r) and permittivity (ε_r).

3. Results and Discussion

Fig. 1 shows the SEM micrographs of the studied NdBFCs, which consisted of roughened porous microstructures. The substitution of Nd^{3+} into the crystal lattice of the NdBFCs was found to affect significantly the surface morphologies (grain sizes and shapes) and distributions of the grains. The average size of the NdBFC grains was decreased and the porosity was increased with the increase in the Nd^{3+} levels. It was argued that the enhanced porous structures are advantageous to achieve a longer propagation path for the electromagnetic waves, thereby effective for stronger reflections and scattering [24]. In addition, the magnetic characteristics and loss of MW reflection can be improved by increasing the porosity of the NdBFCs [11].

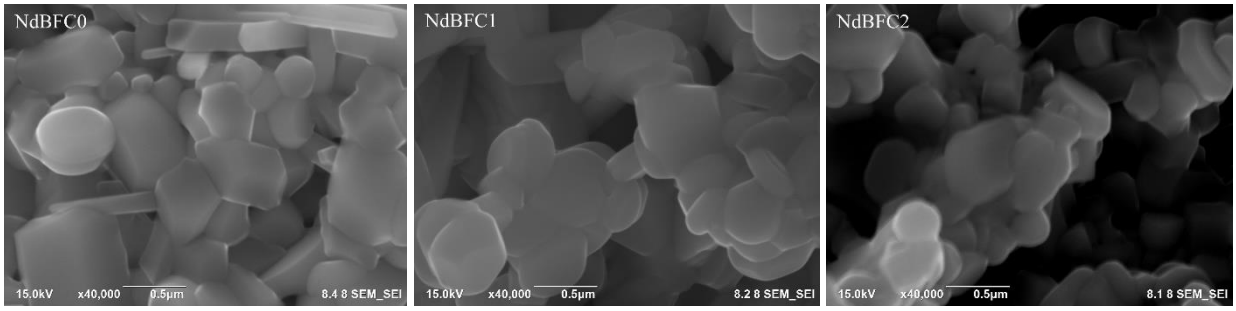

Fig. 1. The SEM images of the obtained NdBFCs

Fig. 2 shows the XRD pattern of the prepared NdBFCs, which comprised of several sharp peaks characteristics of different crystalline lattice planes. The observed peaks for the NdBFC0 sample (without Nd^{3+} doping) were allocated to the major hexagonal crystal structure of $BaFe_{12}O_{19}$ that matched to the ICDD number 00-039-1433 with lattice parameters $a = b =$ 0.5894 nm, c = 2.3215 nm, $\alpha = \beta = 90°$ and $\gamma = 120°$. Conversely, the α-Fe_2O_3 rhombohedral crystal structure (ICDD number 00-033-0664) appeared at 33.336°. The crystal structure of the studied NdBFCs was significantly altered with the inclusion of Nd_2O_3 of 1 mol% as seen from the XRD pattern of NdBFC1. The appearance of six new peaks in the NdBFC1 sample was consistent with the orthorhombic crystal structure of $BaNd_2O_4$ (ICDD number 00-042-1499). Finally, the NdBFC2 sample containing Nd_2O_3 of 2 mol% disclosed a phase transformation from the hexagonal $BaFe_{12}O_{19}$ to hexagonal $Ba_3Fe_{32}O_{51}$ (ICDD number 00-041-0846). It was affirmed that substitution of Nd_2O_3 into the crystal structures of the BFC indeed produced a new primary hexagonal crystalline phase from $Ba_3Fe_{32}O_{51}$. The observed broadening in the diffraction peaks associated with all the prepared NdBFCs was due to the emergence of the quantum size effects (nanoscale particles) and lattice strain [25,26].

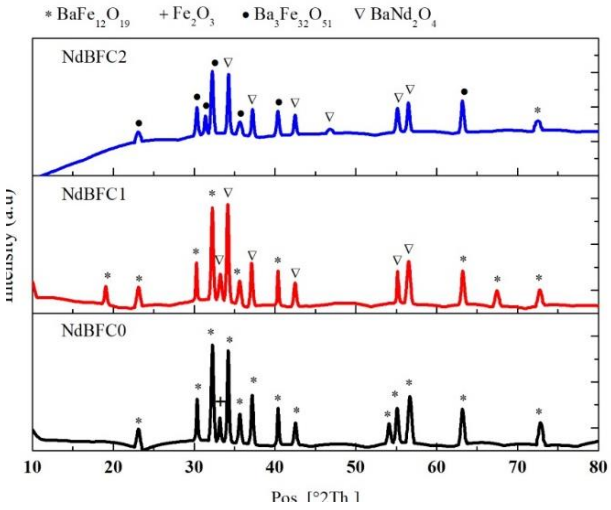

Fig. 2. The XRD patterns of the prepared NdBFCs.

Fig. 3 illustrates the magnetic field (H) dependent magnetization (M) response (hysteresis loop) of the NdBFCs. The values of M_s, M_r, and H_c for the pristine sample (NdBFC0) were 33.54 emu/g, 11.56 emu/g, and 726.06 Oe, respectively. The value M_s of the composites changes significantly because of the expansion concentration of Nd_2O_3. The value of M_s was lowered with the addition of 1 mol% of Nd_2O_3 into the composite (NdBFC1) which was possibly due to the formation of anti-magnetic material ($BaNd_2O_4$). Furthermore, the value of M_s was increased due to the incorporation of 2 mol% of Nd_2O_3 into the composite (NdBFC2) and became higher than NdBFC0, which may be ascribed to the occurrence of a new phase of barium hexaferrite ($Ba_3Fe_{32}O_{51}$), increased porosity and higher surface roughness. It was argued that such enhanced porosity with extremely roughened granular surfaces might have alienated the magnetic domain walls, enabling regular polarizations of the unpaired spin magnetic moments when a magnetic field is applied externally.

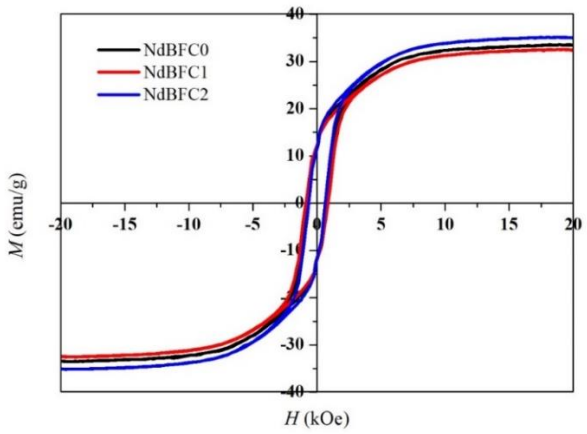

Fig. 3. The hysteresis loops of the as-prepared NdBFCs.

Fig. 4(a) depicts the frequency (ranged from 8.2–12.4 GHz) dependence of complex relative permeability (real, μ' and imaginary, μ'' parts) of the NdBFCs. The value μ' of all the NdBFCs was increased significantly in the lower MW frequency region and then decreased gradually in the higher frequency region. The onset of the decreasing permeability was approximately 8.5 GHz for the undoped sample (NdBFC0) and approximately 9.3 GHz for the doped composites (NdBFC and NdBFC2). The magnetic energy storing ability of the proposed NdBFCs was primarily ascribed to the natural spin magnetic moments' polarizations. Moreover, the value of μ'' (signifies the magnetic loss factor) was decreased rapidly to zero at approximately 8.5 GHz for the pristine sample and 9.3 GHz for the doped ferrites, confirming the absence of magnetic loss in the higher region. It is important to note that the imaginary

permeability plays an essential role in the microwave absorption traits of hexaferrite [27] via the relaxation of the domain wall resonance [28].

Fig. 4(b) shows the frequency (ranged from 8.2–12.4 GHz) dependent complex relative permittivity (real, ε' and imaginary, ε'' components) of the NdBFCs. The intrinsic electric dipole polarizations in the MW frequency domain of the NdBFCs were responsible for the Type equation here.emergence of ε' and ε'' [29,30]. The real permittivity signified the energy stored in the material from an external electric field [31]. For the undoped or pristine sample (NdBFC0), the real permittivity was first increased up to 3 with the increase in the frequency and then decreased to nearly -1 above 12 GHz. Conversely, the real permittivity for the doped samples (NdBFC1 and NdBFC2) was first dropped in the low-frequency region and then increased up to 2 with the rise in the frequency. The ε'' of the undoped specimen (NdBFC0) revealed a shrinking tendency while for the doped samples (NdBFC1 and NdBFC2) it dropped rapidly below 8.2 GHz before being reached nearly to zero in the high-frequency region. The observed almost similar nature of the permittivity for both NdBFC1 and NdBFC2 is due to the presence of the non-magnetic phase of $BaNd_2O_4$ in the samples, resulting in the intrinsic electric dipole polarizations. However, the little difference in the permeability between NdBFC1 and NdBFC2 may be due to the existence of different amounts of the $BaFe_9O_{12}$ and $Ba_3Fe_{32}O_5$ phases with different magnetic properties. Thus, both phases played an important role in the permeability changes associated with the magnetic properties. In addition, a significant increase in the permittivity is possibly triggered by the dipolar polarization originated from the bond formation between O^{2-} with Ba^{2+} and Nd^{3+} in the non-magnetic $BaNd_2O_4$ phase.

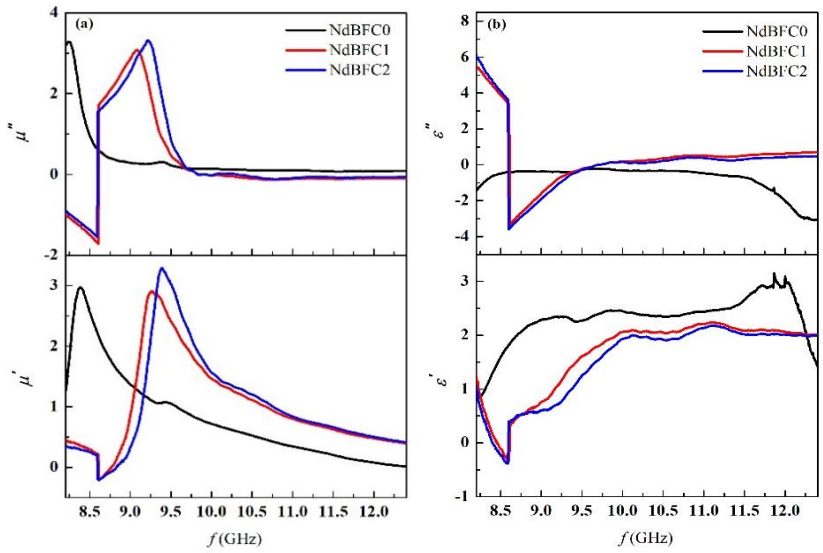

Fig. 4. Frequency-dependent (a) permeability and (b) permittivity of the NdBFCs with a thickness of 0.5 cm

Fig. 5 displays the *f* against R_L plot of the synthesized NdBFCs. The undoped specimen (NdBFC0) revealed three prominent absorption bands centered at approximately 8.4, 10, and 11 GHz with the corresponding R_L values of -37, -17, and -16 dB. Meanwhile, the graph for both the doped samples NdBFC1 and NdBFC2 showed a similar spectral pattern, which consisted of 4 durable absorption bands with values less than -10 dB accompanied by a significant shift. The observed shift in the peak frequency (toward the higher value) with the increase in the Nd^{3+} doping level was probably due to the enhanced porosity and emergence of a new barium hexaferrite phase ($Ba_3Fe_{32}O_{51}$) as well as $BaNd_2O_4$ in the composite structure. Consequently, the values of H_c, M_s, M_r, permeability, and permittivity of the prepared NdBFCs were modified. This, in turn, led to an alteration in the natural resonance frequency and impedance [32–34], thereby a shift in the *peak* R_L value towards higher frequency. It was also shown that the addition of Nd^{3+} caused a widening of the bandwidth in the 9-10 GHz frequency region and the R_L signals were more reversible to the MW frequencies compared to the previous findings [8,35,36].

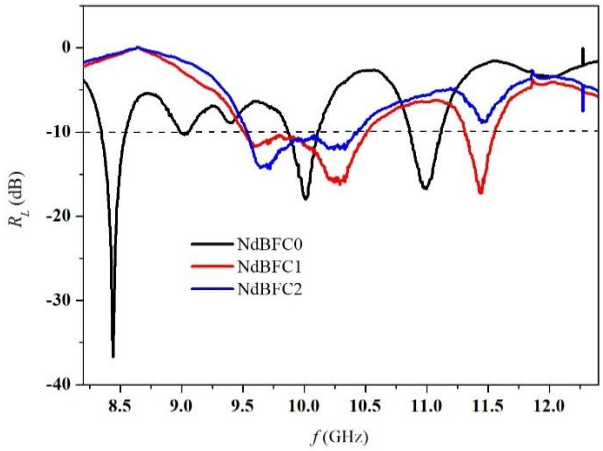

Fig. 5. The MW reflection loss as a function of the frequency for the studied NdBFCs of 0.5 cm thick

4. Conclusions

Following the modified mechanical alloying, a new type of Nd^{3+} activated BFCs was synthesized for the first time and characterized. The incorporation of Nd^{3+} into the hexagonal crystal lattice of BFs was found to influence considerably the surface morphologies, microstructures, crystal structures and phases, magnetic properties, complex permittivity, and permeability as well as the reflection loss in the MW frequency domain of the resultant NdBFCs. The obtained NdBFCs consisted of highly porous and roughened microstructures with a homogeneous distribution of grains. The undoped sample revealed the presence of $BaFe_{12}O_{19}$ (hexagonal lattice) and α-Fe_2O_3 (rhombohedral lattice) crystal phases. The doped specimens (NdBFC1 and NdBFC2) showed the new hexagonal crystalline phase of $Ba_3Fe_{32}O_{51}$ and

$BaNd_2O_4$. The achieved minimum reflection loss with the absorption of several MW frequencies (in the range 8.2-12.4 GHz) shown by the proposed NdBFCs is established to be beneficial for the MW X-band absorber applications.

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