EFFECT OF RARE-EARTH CO-DOPING ON THE MICROSTRUCTURAL AND MAGNETIC PROPERTIES OF BaFe$_{12}$O$_{19}$

ABSTRACT

$B_{0.5}(La,Y)_{0.15}Fe_{12}O_{19}$ hexaferrite magnets were produced using the powder metallurgy method. The phase analysis of the ferrite magnets was carried out by X-ray diffraction (XRD) technique. A single hexaferrite phase was present in both samples as revealed by XRD patterns. The microstructural evolution in the hexaferrite samples was examined using Scanning Electron Microscopy (SEM) equipped with Energy Dispersive X-Ray Spectroscopy (EDS). The grain morphology altered with the sintering temperature. Room temperature ferrimagnetic hysteresis curves were obtained by Vibrating Sample Magnetometer (VSM). The crystallite size and the lattice parameters ($a,c$) were also calculated after sintering at 1150$^\circ$C and 1250$^\circ$C. Saturation magnetizations, $M_s$ were determined to be 48.60 emu/g and 52.95 emu/g for the samples sintered at 1150$^\circ$C and 1250$^\circ$C, respectively whereas the remanent magnetizations, $M_r$ were 29.26 emu/g and 31.17 emu/g. The coercivity, $H_c$ decreased from 3.95 kOe to the value of 2.44 kOe with the sintering temperature due to the increase of the crystallite size. The squareness ratios ($M_r/M_s$) of the ferrimagnetic samples were different because the uniaxial anisotropies altered after sintering at 1150$^\circ$C and 1250$^\circ$C. The maximum energy product, $(BH)_{\text{max}}$ dropped from 35.81 kJ/m$^3$ to 27.38 kJ/m$^3$ when the sintering temperature increased. This result can be attributed to a combination of higher magnetization and the lower coercivity.

Keywords: Hexaferrites; powder metallurgy; co-doping; ferrimagnetic

INTRODUCTION

Hexaferrites with the basic $MFe_{12}O_{19}$ ($M = \text{Ba, Sr}$) formula are doped and they are frequently used as ferrimagnetic compounds. Hexagonal ferrites are magnetically hard and they have been investigated by many researchers. They exhibit high Curie temperature, high magnetic anisotropy and coercivity. These materials have been utilized as the permanent magnets, magnetic recording media and microwave absorbers due to their superior properties [1-3]. That is why, the attempts to improve the magnetic properties of the hexaferrites are still among the recent research topics. In order to achieve an improvement in the magnetic properties, the $M$ element is replaced with a couple of rare-earth ions and this method is
The replacement of Ba and/or Sr crystallographic sites in the hexaferrite lattice by the rare earth cations results in changing the valence state of Fe\(^{3+}\) to Fe\(^{2+}\) which is located on the 2a site. The presence of Fe\(^{2+}\) cation provides the hexaferrite with a considerable rise in the magnetocristalline anisotropy property. Thus these deliberately doped hexaferrites have been reported to possess an improved coercivity (Hc), saturation magnetization (Ms) and other related magnetic properties [6-8].

Most of the recent hexaferrite researches are focused on Sr-hexaferrites. Single doping for hexaferrites has been carried out in some researches where M-type hexagonal ferrites have been doped by Sm\(^{3+}\) and Sr-hexaferrites has been doped by Nd\(^{3+}\) cations [9,10]. In a couple of recent researches, Gd\(^{3+}\) and Tb\(^{3+}\) have been utilized as dopants for Sr-hexaferrites [11,12].

Nano-hexaferrites have been prepared using La-Nd, Mn-Y, Zr-Nd and Sm-Er co-dopants [13-16]. The co-doping for the magnetization enhancement of Sr-hexaferrites has been performed by Al-Ce, Nb-Y, Ce-Zn, Ce-Mn, La-Co and Dy-Al and in these studies sol-gel, co-precipitation and low temperature sintering methods have been carried out [17-23].

On the other hand, single La\(^{3+}\) and Nd\(^{3+}\) doping has been done for Ba-hexaferrites. In addition, co-doped Tm-Tb, Eu-Nd, Sm-Co Ba-hexaferrites have also been investigated and have been shown that co-doping of the rare-earth ions provides hexaferrites with a rise in the magnetization and/or coercivity [24-27]. Among other magnetic properties, some attempts have also been made to improve the maximum energy product, (BH)max for Sr-hexaferrites and Ba-hexaferrites using La-Sm [28,29].Researchers have different manufacturing methods to produce hexaferrites such as traditional ceramic methods [30-34] or several chemical methods [35-41].

To the best of our knowledge, there are no recent studies which have reported the simultaneous effect of the sintering temperature and La\(^{3+}\)-Y\(^{3+}\) dopants on the properties of barium hexaferrites. That is why, we report the improvement of the microstructural and the ferrimagnetic properties of BaFe\(_{12}\)O\(_{19}\) by rare earth co-doping. A number of samples with Ba\(_1\)\(_x\)(La,Y)\(_{3+}\)Fe\(_{12}\)O\(_{19}\) (x=0.15) composition have been produced by the conventional ceramic method.

**EXPERIMENTAL PROCEDURE**

**Preparation of Hexagonal Ferrites**

Ba\(_{1-x}\)(La,Y)\(_{3+}\)Fe\(_{12}\)O\(_{19}\) (x=0.15) hexagonal ferrites were produced using the conventional ceramic method. The starting powders of BaCO\(_3\) (Refsan, 99.21%), Fe\(_2\)O\(_3\) (Refsan, 95%), La\(_{2}\)O\(_3\) (99.99%, Sigma-Aldrich) and Y\(_2\)O\(_3\) (99.99%, Sigma-Aldrich) were mixed for 30 min. at a rotation of 300 rpm. using the grinding balls of 0.75 mm. diameters in Fritsch Pulverisette ball mill. In order to perform a particle size analysis, a sample was taken from the mixed powder and then placed in the analysis container together with ethanol. The particle size measurement of the mixed powder was carried out using Hydro 2000G particle size analyzer.
and the measurement was monitored using Mastersizer 2000 programme. Figure 1 gives the particle size distribution of powder mixtures.

![Particle size distribution](image)

**Fig. 1.** The particle size distribution of mixed powders

After the grinding, the mixed powder was placed in a drying oven to remove the ethanol. Prior to dry pressing, the powder mixture was wetted and the powders were shaped into the pellets of 4.5 g under 50 MPa pressure. The green pellets were sintered at 1150°C and 1250°C for 1 h. in air.

**Characterization of Hexaferrite Samples**

The phase analysis of hexaferrite samples were carried out by a Rigaku D-Max 2200 PC X-ray Diffractometer in a standard measurement using Cu Kα radiation with the wavelength, \( \lambda \) of 0.154 nm. Using Scherrer formula, the average crystallite sizes of hexaferrite samples were calculated. In the Scherrer formula, the single peaks between Bragg angles of 30-50° were selected in order to obtain more precise results. The first five peaks with the highest intensity were used in the average crystallite size calculation as shown in Equation (1):

\[
D_p = \frac{0.94 \times \lambda}{\beta \times \cos \Theta}
\]  

(1)

where, \( D_p \) average crystallite size (nm), \( \beta \) line broadening (rad), \( \Theta \) Bragg angle, \( \lambda \) X-ray wavelength (nm). \( K \) is the Scherrer constant with a common value of 0.94 [42].

Based on Bragg’s Law, the plane spacing of hexagonal crystal, \( d \) is related to the lattice parameters (\( a \) and \( c \)) as shown in Equation (2) [43]:

\[
\frac{1}{d^2} = \frac{4}{3} \cdot \frac{h^2 + h \cdot k + k^2}{a^2} + \frac{l^2}{c^2}
\]

(2)

The lattice constants of \( a \) and \( c \) were calculated from the values of \( d_{hkl} \) corresponding to (107) peaks and (114) peaks. Diffraction planes of \( d_{107} \) and \( d_{114} \) are 2.7692 nm and 2.6181 nm for 1150°C; \( d_{107} \) and \( d_{114} \) are 2.7644 nm and 2.6124 nm for 1250°C, respectively.
The microstructural features of the hexaferrite magnets were examined by Scanning Electron Microscopy (SEM) using JEOL-JSM 6060-2003. Ferrimagnetic hysteresis and related properties were measured by Vibrating Sample Magnetometer (VSM) at room temperature under the magnetic field of 10 kOe.

RESULTS AND DISCUSSION

Phase analysis

Figure 2 shows X-ray diffraction (XRD) patterns of hexaferrites co-doped with La and Y with a total mole of 0.15. After XRD phase analysis, both patterns of the samples indicated a barium hexaferrite phase with a magnetoplumbite crystal type. The standard patterns of both samples were found to match with JCPDS file peaks no. 79-0007 and the space group P63/mmc. The integrated intensities of the two patterns were observed to be different where the sample sintered at 1250°C had a higher intensity in comparison to that of 1150°C. The difference in the integrated intensities can be attributed to the amount of the magnetic hexaferrite phase, which is higher in the sample with a sintering temperature of 1250°C. The complete formation of a single hexaferrite phase in both of the two patterns indicated that sintering time of 1h. was sufficient for this particular composition, \( \text{Ba}_{1-x}(\text{La},\text{Y})_x\text{Fe}_{12}\text{O}_{19} \) \( (x=0.15) \). The results of XRD are in good agreement with that reported by Sardjono et al. [44].

![Fig. 2. X-ray diffraction patterns of La-Y co-doped hexaferrites](image)

Considering the highest intensities, the strongest peak was found to be (107) and the second highest peak was (114). Utilizing these particular peaks, average crystallite sizes, \( D_p \) of La-Y co-doped hexaferrite samples were calculated according to Scherrer formula. Table 1 gives the average crystallite sizes of the two hexaferrite samples. When the sintering temperature rised from 1150°C to 1250°C, the average crystallite size, \( D_p \) increased from 41.15 nm to 41.15 nm The lattice parameter \( a \), decreased from 0.5875 nm to 0.5856 nm and \( c \), decreased from 2.311 nm. to 2.308 nm after sintering at 1150°C and 1250°C, respectively. In the study of Sepelak et al. [45], the lattice parameter, \( a \) is given as 0.589 nm and the lattice parameter, \( c \) is given as 2.317 nm, respectively.
In the present study, average crystallites had precise values of 41.15 nm and 45.25 nm after sintering at 1150°C and 1250°C, respectively. These crystallite values are well below 460 nm that is critical domain size for a hexaferrite [51]. The ionic radii of O\(^2\), Fe\(^{3+}\) and Ba\(^{2+}\) are 1.32 Å, 0.64 Å and 1.34 Å, respectively. The substitution of Ba\(^{2+}\) partially by La\(^{3+}\) (1.016 Å) and Y\(^{3+}\) (0.89 Å) by co-doping in the same mole percentage resulted in a decrease in the ionic radii. Thus the lattice parameter of \(\text{Ba}_{0.85}(\text{La},\text{Y})_{0.15}\text{Fe}_{12}\text{O}_{19}\) was lower than that of pure \(\text{BaFe}_{12}\text{O}_{19}\) due to the different ionic radii [52].

**Microstructural examination**

Figure 3-6 indicate SEM images and EDS analysis of \(\text{Ba}_{0.85}(\text{La},\text{Y})_{0.15}\text{Fe}_{12}\text{O}_{19}\) samples sintered at 1150°C and 1250°C, respectively. It was observed from Figure 3 that the grains were hexagonal platelets the distribution was somewhat homogeneous. Similarly, platelet-shaped hexagonal grains were observed in doped barium hexaferrites [51]. Also, for pure and Ce-doped hexaferrites, SEM images showed that there are grains with a platelet morphology [36].
Fig. 4. Corresponding EDS analysis of La-Y co-doped hexaferrite sintered at 1150°C

Fig. 5. SEM images of Ba0.85(La,Y)0.15Fe12O19 sintered at 1250°C for a.1000x, b.5000x and c.10000x

Fig. 6. Corresponding EDS analysis of La-Y co-doped hexaferrite sintered at 1250°C
The grain boundaries were evident in Figure 3.b and the average grain size was determined to be 2.6 μm. However, the grain morphology altered with the sintering temperature. Figure 5 shows that the grains are prone to agglomeration to each other after sintering at 1250°C. Due to the agglomeration, it was not easy to determine the average grain size. According to EDX analysis in Figure 4 and Figure 6, the percentage of the elements was closer to the estimated theoretical stoichiometry in M-type hexaferrites [1]. For a comparison, in a recent study the grain size was measured to be 0.4 mm for a doped barium hexaferrite [51]. In another study, a pure hexaferrite sample had coarser grains with a size of 0.5 mm, which then refined to reach a size of 0.35 mm after doping [36].

**Magnetic Characterization**

The effect of sintering temperature on the saturation magnetization, \( M_s \) and the remanent magnetization, \( M_r \) of hexaferrite magnets is shown in Figure 7. From Figure 7, it is obvious that the saturation magnetization, \( M_s \) increased with the sintering temperature. The saturation magnetizations, \( M_s \) of \( \text{Ba}_{0.85}\text{(La,Y)}_{0.15}\text{Fe}_{12}\text{O}_{19} \) sintered at 1150°C and 1250°C were 48.60 emu/g and 52.95 emu/g whereas the remanent magnetizations, \( M_r \) were 29.26 emu/g and 31.17 emu/g, respectively. Due to the increase of the magnetic phase content, both of the magnetizations increased with the sintering temperature.

![Fig. 7. Effect of sintering temperature on the hysteresis curves of \( \text{Ba}_{0.85}\text{(La,Y)}_{0.15}\text{Fe}_{12}\text{O}_{19} \) samples for a. 1150°C and b. 1250°C](image)

On the contrary, the coercivity, \( H_c \) decreased from 3.95 kOe to the value of 2.44 kOe with the sintering temperature. In Table 2, the coercivities of various hexaferrite magnets were compared. The literature data is in accordance with our coercivity results. A rise in the sintering temperature of a barium-strontium hexaferrite sample resulted in a decline of the coercivity. Also the drop in the coercivity due to the rise of the sintering temperature has been observed in the La-doped strontium hexaferrite as well. This effect has been attributed to the increase of the crystallite size due to a higher sintering temperature [53]. This result is in good agreement with microstructural images with a drop of coercivity to 2.44 kOe because of the grain growth. The coercivities that were obtained in this study are higher than that in undoped
barium hexaferrite, which is 1.62kOe [28]. The low coercivities and also high values of saturation magnetization shown in Table 2 can be utilized in the field of high-density magnetic recording [51].

Saturation and remanent magnetization values in our study are in good agreement with the literature data as shown in Table 2. The squareness ratios ($Mr/Ms$) of the ferrimagnetic samples in this study are 0.60 and 0.59 which is attributed to the fact that the uniaxial anisotropies after sintering at 1150°C and 1250°C are different [1]. When these squareness ratios were compared to that in undoped barium hexaferrite, which is 0.66; it is noted that these values are below the undoped value [28].

Figure 8 presents the demagnetizing curves of the hexaferrite magnets measured after sintering at 1150°C and 1250°C. By making use of the demagnetizing curves, the maximum energy products, $(BH)_{max}$ of the ferrimagnetic samples were calculated. Several trials were made to find the maximum area in the second quadrant of the hysteresis loop as in Figure 8, then the results were shown in Table 3.

**Table 2. Magnetization and coercivity of La-Y co-doped hexaferrites**

| Sample            | Magnetization (emu/g) | Coercivity (kOe) | Reference     |
|-------------------|-----------------------|------------------|---------------|
|                   | $Ms$                  | $Mr$             |               |
| BHF               | 54.02                 | 24.51            | 3.10 [45]     |
| SBHF              | 80.30                 | 50.00            | 3.64 [54]     |
| BHF composites    | 55.00                 | 35.00            | 8.73 [55]     |
| BHF               | 44.94                 | 27.82            | 3.58 [56]     |
| SHF               | 51.81                 | 34.55            | 1.73 [28]     |
| L-Y BHF/15        | 52.95                 | 29.26            | 3.95 Present Study |
| L-Y BHF/25        |                       |                  | 2.44 Present Study |

**Fig. 8.** Effect of sintering temperature on the demagnetization curves of Ba$_{0.85}$(La,Y)$_{0.15}$Fe$_{12}$O$_{19}$ samples for a.1150°C and b.1250°C
Table 3. Maximum energy product calculations of hexaferrite magnets sintered at a. 1150°C and b. 1250°C

|          | BHmax (kJ/m³) |          | BHmax (kJ/m³) |
|----------|---------------|----------|---------------|
|          | Magnetic field (T) | Magnetization (emu/g) | Magnetic field (T) | Magnetization (emu/g) |
| a.       | 0.0261 0.0555 0.0959 0.1447 0.1943 | 30.05 28.86 27.04 23.66 17.69 | b.       | 0.0369 0.0991 0.1443 0.1948 0.2446 | 27.38 | 35.81 |

It can be seen from Figure 9 that (BH)max decreased from 35.81 kJ/m³ to 27.38 kJ/m³ when the sintering temperature was raised from 1150°C to 1250°C. The higher the maximum energy product, (BH)max is, the better the ferrimagnet is. This relationship between the rare-earth doping and the energy product, (BH)max was in good agreement with literature data where barium substitution with a rare earth and strontium improved the energy product by about 14% [53]. In our study, a rise in the sintering temperature resulted in a drop in the energy product, (BH)max because of a combination of higher magnetization and the lower coercivity.

CONCLUSIONS

In the present study, Ba₀.₈₅(La,Y)₀.₁₅Fe₁₂O₁₉ hexaferrite magnets were produced using the powder metallurgy method. The result of the phase analysis indicated a single hexaferrite phase in both samples. As can be observed from SEM images, the grain morphology in the hexaferrite samples altered with the sintering temperature. The crystallite size and the lattice parameters (a, c) were calculated using XRD pattern data by Scherrer formula and Bragg’s Law, respectively. From the room temperature ferrimagnetic hysteresis curves, the saturation and the remanent magnetizations were determined to be 48.60 emu/g and 29.26 emu/g after sintering at 1150°C; they were 52.95 emu/g and 31.17 emu/g at 1250°C. Since the crystallite size increased due to a rise in the sintering temperature, the coercivity, Hc decreased from 3.95 kOe to the value of 2.44 kOe. A change in the uniaxial anisotropies altered the squareness ratios (M_r/M_s) of the ferrimagnetic samples. The maximum energy product, (BH)max dropped from 35.81 kJ/m³ to 27.38 kJ/m³ when the sintering temperature increased. This result can be attributed to a combination of higher magnetization and the lower coercivity.

ACKNOWLEDGMENT

We acknowledge the analysis support from Middle East Technical University (ODTU), Central Laboratory for VSM measurements. Also, we are indebted to Mr. Haluk Gürses from Dokuz Eylül University (DEU), Industrial Service for providing the facility of SEM/EDS and XRD analysis.
REFERENCES

1. Mahmood S.H., Abu-Aljarayesh I.: Hexaferrite Permanent Magnetic Materials. Materials Research Foundations, vol.4, 2016.
2. Singh C.: Engineering Magnetic, Dielectric and Microwave Properties of Ceramics and Alloy. Materials Research Foundations LLC, vol.57, 2019.
3. Krishnan K.M.: Fundamentals and Applications of Magnetic Materials. Oxford University Press, 1st Ed., 2016.
4. Bruck E.: Handbook of Magnetic Materials. North Holland 1st Ed., vol.26, 2017.
5. Castro W.S., Correa R.R., Paulim Filho P.I., Rivas Mercury J.M., Cabral A.A.: Dielectric and magnetic characterization of barium hexaferrite ceramics. Ceramics International, (2015), 41, 241-246.
6. Zhukov A.: Novel Functional Magnetic Materials Fundamentals and Applications. Springer International Publishing, vol.231, Switzerland, 2016.
7. Vinnik D.A., Zherebtsov D.A., Mashkovtseva L.S., Nemrava S., Yakushechkina A.K., Semisalova A.S., Gudkova S.A., Anikeev A.N., Perov N.S., Isaenko L.I., Niewa R.: Tungsten substituted BaFe12O19 single crystal growth and characterization. Materials Chemistry and Physics, (2015), 155, 99-103.
8. Zhang S., Zhao D.: Advances in Magnetic Materials: Processing, Properties, and Performance. Advances in Materials Science and Engineering CRC Press, 2017.
9. Yasmin N., Mirza M., Muhammad S., Zahid M., Muhammad A.: Influence of samarium substitution on the structural and magnetic properties of M-type hexagonal ferrites. Journal of Magnetism and Magnetic Materials, (2018), 44615, 276-281.
10. Perez-Juache T.J., Guerrero A.L., Cabal A.A., Sertkol M., Ercan I.: Nd3+ substituted strontium hexaferrites: Structural, magnetic and optical investigation and cation distribution. Journal of Rare Earths, (2020), 82015, 153-180.
11. Hu J., Liu C., Kan X., Liu X., Ur Rehman K.M.: Structure and magnetic performance of Gd substituted Sr-based hexaferrites. Journal of Alloys and Compounds, (2020), 82015, 402-410.
12. Thakur A., Barman P.B., Singh R.R.: Effects of La3+-Nd3+ ions and pre-calcination on the growth of hexaferrite nanoparticles prepared by gel to crystallization technique: Non-isothermal crystallization kinetics analysis. Materials Chemistry and Physics, (2015), 15615, 29-37.
13. Almessiere M.A., Slimani Y., Gungunes H., Sertkol M., Ercan I.: Nd3+ substituted strontium hexaferrites: Structural, magnetic and optical investigation and cation distribution. Journal of Rare Earths, (2020), 38(4), 732-740.
14. Luo J., Xu Y., Mao H.: Magnetic and microwave absorption properties of rare earth ions (Sm3+,Er3+) doped strontium ferrite and its nanocomposites with polypyrrole. Journal of Magnetism and Magnetic Materials, (2015), 3811, 365-371.
17. Venkatesh G., Subramanian R., Satish Kumar T., Abuthakir J., Sethupathi K.: Investigation on structural and magnetic properties of Al$^{3+}$ and Ce$^{3+}$ doped hexaferrites. Materials Today: Proceedings, 2019.

18. Güner S., Almessiere M.A., Slimani Y., Baykal A., Ercan I.: Microstructure, magnetic and optical properties of Nb$^{5+}$ and Y$^{3+}$ ions co-substituted Sr hexaferrites. Ceramics International, (2020), 46(4), 4610-4618.

19. Yasmin N., Iqbal M.Z., Zahid M., Gillani S.F., Mirza M.: Structural and magnetic studies of Ce-Zn doped M-type SrFe$_{12}$O$_{19}$ hexagonal ferrite synthesized by sol-gel auto-combustion method. Ceramics International, (2019), 45(1), 462-467.

20. Neupane D., Wang L., Adhikari H., Alam J., Mishra S.R.: Synthesis and characterization of co-doped SrFe$_{12-x}$(DyAl)$_x$O$_{19}$ hexaferrites. Journal of Alloys and Compounds, (2017), 70115, 138-146.

21. Yasmin N., Abdulsatar S., Hashim M., Zahid M., Mirza M.: Structural and magnetic studies of Ce-Mn doped M-type SrFe$_{12}$O$_{19}$ hexagonal ferrites by sol-gel auto-combustion method. Journal of Magnetism and Magnetic Materials, (2019), 4731, 464-469.

22. Huang T., Peng L., Li L., Wang R., Tu X.: Low temperature sintering behavior of La-Co substituted M-type strontium hexaferrites for use in microwave LTCC technology. Journal of Rare Earths, (2016), 34(2), 148-151.

23. Serletis C., Litsardakis G., Pavlidou E., Efthimiadis K.G.: Magnetic properties of co-precipitated hexaferrite powders with Sm-Co substitutions optimized with the molten flux method. Physica B: Condensed Matter, (2017), 52515, 78-83.

24. Satyapal H.K., Singh R.K., Kumar N., Sharma S.: Low temperature synthesis and influence of rare earth Nd$^{3+}$ substitution on the structural, magnetic behaviour of M-type barium hexaferrite nanomaterials. Materials Today: Proceedings, 2020.

25. Verma S., Pandey O.P., Paesano A., Sharma P.: Comparison of structural and magnetic properties of La$^{3+}$ substituted BaFe$_{12}$O$_{19}$ prepared by different substitution methods. Physica B: Condensed Matter, (2014), 4481, 57-59.

26. Almessiere M.A., Slimani Y., Guner S., Aldakhl S., Baykal A.: Ultrasonic synthesis, magnetic and optical characterization of Tm$^{3+}$ and Tb$^{3+}$ ions co-doped barium nanohexaferrites. Journal of Solid State Chemistry, (2020), 286, 121310.

27. Almessiere M.A., Slimani Y.A., Korkmaz D., Baykal A., Ercan I.: A study on the spectral, microstructural, and magnetic properties of Eu-Nd double-substituted Ba$_{0.5}$Sr$_{0.5}$Fe$_{12}$O$_{19}$ hexaferrites synthesized by an ultrasonic-assisted approach. Ultrasonics Sonochemistry, (2020), 62, 104847.

28. Shekhawat D., Singh A.K., Roy P.K.: Structural and electro-magnetic properties of high (BH)max La-Sm substituted Sr-hexaferrite for brushless DC electric motors application. Journal of Molecular Structure, (2019), 11795, 787-794.

29. Mohseni F., Pullar R.C., Vieira J.M., Amaral J.S.: Enhancement of maximum energy product in exchange-coupled BaFe$_{12}$O$_{19}$/Fe$_3$O$_4$ core-shell-like nanocomposites. Journal of Alloys and Compounds, (2019), 80625, 120-126.

30. Mahmood S., Aloqaily A., Maswadeh Y., Awadallah A., Bsoul I., Juwhari H.: Structural and magnetic properties of mo-zn substituted (BaFe$_{12-3x}$Mo$_{x}$Zn$_{3x}$O$_{19}$) M-type hexaferrites. Material Science Research India, (2014), 11, 09-20.

31. Awadallah A., Mahmood S., Maswadeh Y., Bsoul I., Awawdeh Q., Mohaidat H., Juwhari H.: Structural, magnetic, and Mossbauer spectroscopy of Cu substituted M-type hexaferrites. Materials Research Bulletin, (2016), 74, 192-201.
32. Kang Y.M., Kwon Y.H., Kim M.H., Lee D.Y.: Enhancement of magnetic properties in Mn-Zn substituted M-type Sr-hexaferrites. Journal of Magnetism and Magnetic Materials, (2015), 382, 10-14.

33. Joshi R., Singh C., Kaur D., Zaki H., Ghimire M.: Structural and magnetic properties of Co²⁺-W⁴⁺ ions doped M-type Ba-Sr hexaferrites synthesized by a ceramic method. Journal of Alloys and Compounds, (2017), 695, 909-914.

34. Singh J., Singh C., Kaur D., Zaki H., Meena S.S.: Elucidation of phase evolution, microstructural, Mössbauer and magnetic properties of Co²⁺-Al³⁺ doped M-type BaSr hexaferrites synthesized by a ceramic method. Journal of Alloys and Compounds, (2017), 695, 1112-1121.

35. Din M.F., Ahmad I., Ahmad M., Farid M.T., Iqbal M.A., Murtaza G., Khan M.A.: Influence of Cd substitution on structural, electrical and magnetic properties of M-type barium hexaferrites co-precipitated nanomaterials. Journal of Alloys and Compounds, (2014), 584, 646-651.

36. Mosleh Z., Kameli P., Poorbaferani A., Ranjbar, Salamati H.: Structural, magnetic and microwave absorption properties of Ce-doped barium hexaferrite. Journal of Magnetism and Magnetic Materials, (2016), 397, 101-107.

37. Jamalian M.: An investigation of structural, magnetic and microwave properties of strontium hexaferrite nanoparticles prepared by a sol-gel process with doping Sn and Tb. Journal of Magnetism and Magnetic Materials, (2015), 378, 217-220.

38. Kaur P., Chawla S.K., Meena S.S., Yusuf S.M., Bindra Narang S.: Synthesis of Co-Zr doped nanocrystalline strontium hexaferrites by sol-gel auto-combustion route using sucrose as fuel and study of their structural, magnetic and electrical properties. Ceramics International, (2016), 42(13), 14475-14489.

39. Kaur P., Chawla S.K., Bindra Narang S., Pubby K.: Structural, magnetic and microwave absorption behavior of Co-Zr substituted strontium hexaferrites prepared using tartaric acid fuel for electromagnetic interference suppression. Journal of Magnetism and Magnetic Materials, (2017), 422, 304-314.

40. Kaur P., Chawla S.K., Meena S.S., Yusuf S.M., Bindra Narang S.: Modulation of physico-chemical, magnetic, microwave and electromagnetic properties of nanocrystalline strontium hexaferrite by Co-Zr doping synthesized using citrate precursor sol-gel method. Ceramics International, (2017), 43(1), 590-598.

41. Mudsainiyan R.K., Gupta M., Chawla S.K.: Physico-chemical and magnetic properties of Co-Zr doped Ba-hexaferrites using self-combustion and urea assisted method-A comparative study. Materials Today: Proceedings, (2016), 3(2), 507-512.

42. Speakman S.A.: Estimating Crystalite Size Using XRD. The MIT Materials Research Science and Engineering Center (MRSEC), 2019.

43. Rahimi F., Rahmati A., Mardani S.: Determination and analysis of structural and optical properties for thermally evaporated ZnO thin films. Soft Nanoscience Letters, (2014), 4(01), 1-5.

44. Sardjono P., Suprapedi S., Muljadi M., Djauhari N.R.: Microstructure, physical properties, and magnetic flux density analysis of permanent magnet BaFe₁₂O₁₉ using milling and sintering preparation methods. Journal of Physics: Conference Series, (2016), 739(1), 1-6.

45. Sepelak V., Myndyk M., Witte R., Röder J., Menzel D., Schuster R.H., Hahn H., Heitjans P., Becker K.D.: The mechanically induced structural disorder in barium hexaferrite, BaFe₁₂O₁₉, and its impact on magnetism. Faraday Discussions, (2014), 170, 121-135.

46. Davarpanah A.M., Rahdar A., Azizi Dastnæ M., Zeybek O., Beyzaee H.: (1-x)BaFe₁₂O₁₉xCoFe₂O₄ hard/soft magnetic nanocomposites: Synthesis, physical characterization and antibacterial activities study. Journal of Molecular Structure, (2019), 1175, 445-449.
47. Molaei M.J., Rahimipour M.R.: Microwave reflection loss of magnetic/dielectric nanocomposites of BaFe\textsubscript{12}O\textsubscript{19}/TiO\textsubscript{2}. Materials Chemistry and Physics, (2015), 167, 145-151.

48. Al Dairy A.R., Al-Hmoud L.A., Khatatbeh H.A.: Magnetic and structural properties of barium hexaferrite nanoparticles doped with titanium. Symmetry, (2019), 11, 732.

49. Manawan M., Saragi T., Sukandi A., Fachrudin B., Kurniawan A., Manaf E.P., Boedijono R.: Crystallite size determination of barium hexaferrite nanoparticles using WH-plot and WPPM. IOP Conf. Series: Journal of Physics: Conf. Series, (218), 1080, 012008.

50. Choi M., Cho S., Song Y., Baek S., Kim H., Jung J., Lee H., Park C., Park S., Kim Y.: Synthesis and characterization of hollow BaFe\textsubscript{12}O\textsubscript{19} submicron spheres for advance functional magnetic materials. Current Applied Physics, (2014), 14(9), 1208-1211.

51. Awadallah A., Mahmood S.H., Maswadeh Y., Bsoul I., Aloqaily A.: Structural and magnetic properties of vanadium doped M-type barium hexaferrite (BaFe\textsubscript{12-x}V\textsubscript{x}O\textsubscript{19}). IOP Conf. Series: Materials Science and Engineering, (2015), 92, 012006.

52. Uzun H., Fındık F., Salman S.: Malzeme Biliminin Temelleri. Değişim Publications, 2008.

53. Mahmood S.H., Abu-Aljarayesh I.: Hexaferrite Permanent Magnetic Materials. Materials Research Forum LLC, 2016.

54. Rusianto T., Waziz Wildan M., Abraha K., Kusmono K.: The magnetic and mechanical properties of sintered ceramic magnets Sr\textsubscript{x}Ba\textsubscript{1-x}Fe\textsubscript{12}O\textsubscript{19}. Ijet-Ijens, (2015), 15(05), 41-45.

55. Zafar A., Rahman A., Shahzada S., Anwar S., Khan M., Nisar A., Ahmad M., Karim S.: Electrical and magnetic properties of nano-sized Eu doped barium hexaferrites. Journal of Alloys and Compounds, (2017), 727, 683-690.

56. Li L., Zhang Z., Xie Y., Zhao J.: Preparation, characterization and magnetic properties of the BaFe\textsubscript{12}O\textsubscript{19}-chitosan composites. Solid State Sciences, (2016), 57, 44-48.

57. Almessiere M.A., Slimani Y., Gungunes H., Manikandan A., Baykal A.: Investigation of the effects of Tm\textsuperscript{3+} on the structural, microstructural, optical, and magnetic properties of Sr hexaferrites. Results in Physics, (2019), 13, 102-166.