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The Effect of Lanthanum Substitution on Coercivity Field in Oxide Permanent Magnet Based on Ba1-xLaxFe12O19 (x = 0; 0.02; 0.04; and 0.08)

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Keywords

Ba1-xLaxFe12O19;
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Abstract

Synthesis and characterization of $Ba_{1-x}LaxFe_{12}O_{19}$ based permanent magnet samples with variations in composition (x = 0; 0.02; 0.04; and 0.08) has been done. The method used is a solid state reaction. The phase formation of the sample $Ba_{1-x}LaxFe_{12}O_{19}$ was carried out at 1200°C for 2 hours. The results of the X-ray diffraction pattern analysis show that all sample compositions have a single phase $BaFe_{12}O_{19}$. The results of magnetic properties testing using VSM showed that the best coercivity field and remanent magnetization values were obtained at the composition of x = 0.04. The effect of substitution of lanthanum into the barium atom can increase the number of magnetic domains which are indicated by the increase in the coercivity value of the material along with the increasing composition of x. It was concluded that the composition of $Ba_{1-x}LaxFe_{12}O_{19}$ is a permanent magnet with the best product energy.

Contents

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1. Introduction

Ferrites compound-based magnetic materials have still attracted the attention of many researchers and industry players considering their wide application in the electronics and automotive industries. It is known that ferrites-based ingredients still control the world market share of up to 52% only as permanent magnet materials. The permanent magnet material has a basic composition of $BaFe_{12}O_{19}$ (Liangliang Cao, 2017). The advantages of magnetic ferrites include, in addition to its main components, iron oxide (Fe_2O_3) which is abundant in the world and thus can be produced at cheap material costs, this compound also has a relatively high magnetocrystalline anisotropy constant ~0.33 MJ. m^{-3}, Curie temperature is ~450^{o}C so that it can operate at relatively high temperatures, in addition to its total magnetization value of ~ 0.5 T and thus potentially having an energy of ~ 45.26 kJ.m^{-3} (Vinnik *et al.,* 2018). This type of magnetic material has oxide phases which are built on very strong ionic bonds. This shows that in general atmospheric conditions, these two types of material are chemically and physically very stable. The type is ionic, facilitating the process of material synthesis through less difficult methods such as solid-state reaction (Anthony, 2014). But along with the demands of the times and technology, the need for magnets that have strong magnetic properties and can be used for a long time is increasing. Problems like this are what encourage research with the aim of creating magnets that have superior qualities. While other permanent magnets that are widely used in this modern era are permanent magnets based on rare earth metals. This permanent magnet has a much larger energy capacity compared to magnet oxide, which is in the range of 130 kJ/m^3 to 400 kJ/m^3 (Volker, 2013). The role of rare earth metals is very significant in building the anisotropy properties of these magnetic materials. Based on the advantages of rare earth metals, scientific breakthroughs are needed where this rare earth metal is one of the atomic substitutes in order to improve the magnetic properties of ferrite-based magnets (Harta, 2017).

In Indonesia alone, the availability of rare earth metals and iron sand as raw materials for the manufacture of permanent magnets can be said to be abundant. This is shown based on data from the Center for Geological Resources which shows that from 2011 to 2015, the number of iron sand resources in Indonesia increased (Mosleh *et al.,* 2014). Whereas soil metal minerals are rarely found on Bangka Belitung Island in the form of monazite minerals. This monazite mineral processing technology has been successfully carried out by the National Nuclear Energy Agency (BATAN) into rare earth metal oxides. One of the results obtained is cerium oxide and lanthanum oxide which has a relatively high purity above 99%. Thus making Indonesia as one of the countries that have enough potential as a producer of rare earth metals.

In this study, a permanent magnet modification of the ferrite system has been carried out by substituting a barium atom (Ba) with an atom of lanthanum (La). The system built is to synthesize $Ba_{1-x}LaxFe_{12}O_{19}$ based permanent magnet materials using solid-state reaction method, in the hope that lanthanum material can occupy part of the position of the barium atom. Thus the purpose of this study was to determine the effect of lanthanum substitution on the coercivity field on permanent magnets based on Ba1-xLaxFe12O19 with variations in composition (x = 0; 0.02; 0.04; and 0.08), to determine the composition optimal from the structural and magnetic side.

2. Materials and Methods

The synthesis of $Ba_{1-x}LaxFe_{12}O_{19}$ was carried out using the solid-state reaction method. The raw materials used in this synthesis are Fe_2O_3, $BaCO_3$, and La_2O_3, with the following stoichiometric calculations:

$$1\text{-}xBaCO_3 + 0{,}5xLa_2O_3 + 6Fe_2O_3 \rightarrow Ba_{1-x}La_xFe_{12}O_{19} + 1\text{-}xCO_2$$

The raw material is weighed according to the stoichiometric calculation, then the raw material will be mixed and placed in a stainless steel based container. The raw material mixture will be added with ethanol and a milling ball with a composition of ethanol volume and raw material of 5: 2 and mass composition of milling balls and raw materials of 2. The raw material mixture will be milled for 5 hours at a speed of 1000 rpm. After passing through the milling process, the raw material is dried and calcined at 800^{o}C for 2 hours. The mixture that is still in powder form will be compressed at 6000 psi and sintered at 1200 oC for 5 hours (Toby, 2001).

To determine the structure and phase formed in the sample, characterization using XRD (X-ray Diffractometer) was carried out by the Phillips Pan Analytical Empyrean brand with an angle of 2θ at 20°-80° using Cu anode with a wavelength of 1.5418740 Å. The resulting X-ray diffraction pattern will be analyzed using the GSAS program (Toby, 2001), and magnetic properties measurements were carried out using Oxford brand VSM (Vibrating Sample Magnetometer) equipment with magnitude 1 Tesla.

3. Results and Discussions

X-ray diffraction pattern of permanent magnet samples $Ba_{1-x}La_xFe_{12}O_{19}$ with variations in composition (x = 0; 0.02; 0.04; and 0.08) synthesized using solid state reaction method is shown in Figure 1. The resulting diffraction pattern along with the data the relative intensity is matched with the standard X-ray diffraction pattern data with the most appropriate peak position and intensity. This phase identification refers to the results of the Obrador study (Obradors *et al.,* 1985), with reference data from the Crystallography Open Database (COD: 1008841).

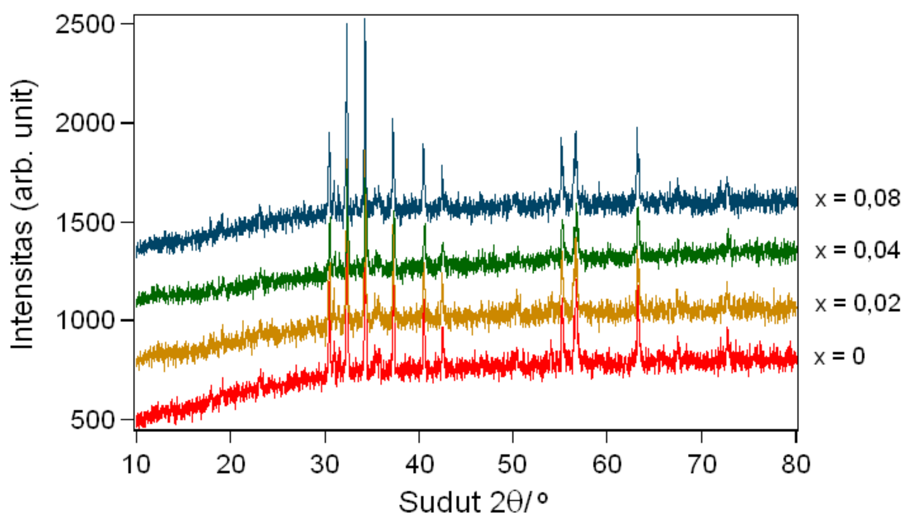
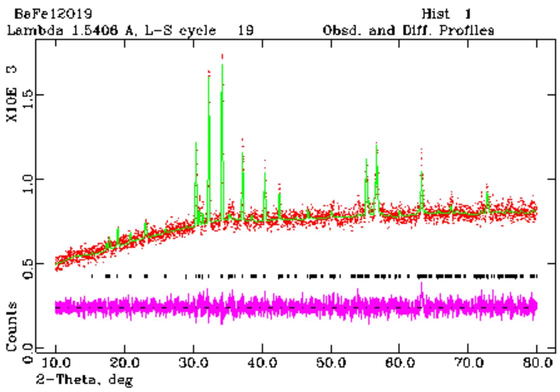


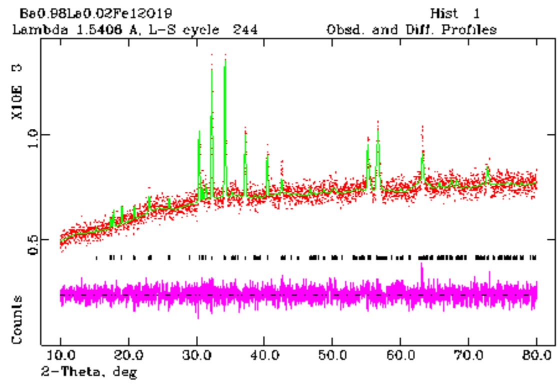
Figure 1. Sample X-ray diffraction patterns of $Ba_{1-x}La_xFe_{12}O_{19}$ (x = 0; 0.02; 0.04; and 0.08)

The results of sample identification show that for all compositions x = 0; 0.02; 0.04; and 0.08 is thought to have a single phase barium hexaferrite ($BaFe_{12}O_{19}$). It is shown that the main peaks formed have diffraction angle positions and intensities that correspond to the BaFe12O19 phase. Based on the phase identification it is known that the $BaFe_{12}O_{19}$ phase has the highest relative intensity at a lattice distance of about 2.62 Å which corresponds to the hexagonal crystallographic plane (114). Then it is followed by other peaks which are characteristic of the BaFe12O19 phase that is the top of the plane (107) at a lattice distance of about 2.77 Å, (203) at a lattice distance of about 2.42 Å, (110) at a lattice distance of around 2.95 Å, (2011) at a lattice distance of about 1.62 Å, (205) at a lattice distance of about 2.23 Å, (166) at a lattice distance of about 2.17 Å, (304) at a lattice distance of about 1.63 Å. Thus Ba1-xLaxFe12O19 with variations in composition (x = 0; 0.02; 0.04; and 0.08) shows indications of the formation of a single phase with a barium hexaferrite structure which has a hexagonal crystal structure with space group P 63/mmc (194) with lattice parameters are a = b = 5.8920 Å and c = 23.1830 Å.

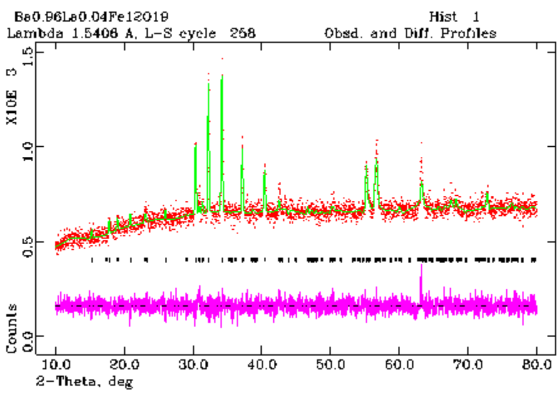
system with variations in composition (x = 0; 0.02; 0, 04; and 0.08) the results of synthesis by the solid-state reaction method as shown in Figure 2.



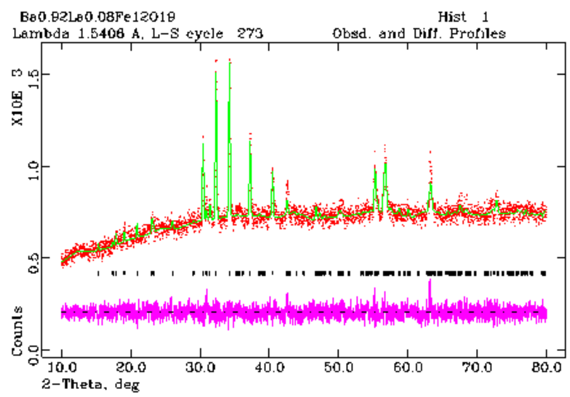
(a) x = 0



(b) x = 0,02



(c) x = 0,04



(d) x = 0,08

Figure 2. Refinement of the X-ray diffraction pattern of samples Ba1-xLaxFe12O19 with variations in composition (x = 0; 0.02; 0.04; and 0.08)

Figure 2 (a-d) shows a sample with the composition x = 0; 0.02; 0.04; and 0.08 that the refinement results can be seen that a single phase Ba1-xLaxFe12O19 has been formed. The results of quantitative analysis based on refinement above refer to Crystallography Open Database with number 1008841 in the form of BaFe12O19 phase.

Structural parameter data results from refinement of XRD data from samples Ba1-xLaxFe12O19 with variations in composition (x = 0; 0.02; 0.04; and 0.08) can be seen in Table 1. Based on Figure 2 and Table 1 can be seen that the results of the XRD data refinement show the compatibility between the sample data and the existing database. This is illustrated by the value of Rwp and chi2. Rwp is the difference in mass ratio between calculation data and experimental data (Rwp <10%), while chi2 is a comparison between the experimental XRD pattern and the expected XRD pattern (1 <chi2 <1.3).

Table 1
Phase tables, lattice parameters, volume, density, mass fraction of samples Ba1-xLaxFe12O19 with variations in composition (x = 0; 0.02; 0.04; and 0.08)

Sample (x)	Phase	lattice parameters (Å) a	b	c	V (Å^3)	ρ (g/cm^3)	Fraction (%)	Rwp	Chi
0	$BaFe_{12}O_{19}$	5.8865	5.8865	23.1868	695.81	11,570	100	3,98	1,194
0,02	$BaFe_{12}O_{19}$	5.8789	5.8789	23.1512	692.95	11,015	100	4,01	1,146
0,04	$BaFe_{12}O_{19}$	5.8745	5.8745	23.1306	691.29	10,870	100	4,18	1,253
0,08	$BaFe_{12}O_{19}$	5.8720	5.8720	23.1208	690.40	10,547	100	4,34	1,244

In Table 1 it can be seen that the phase formed in the sample $Ba_{1-x}La_xFe_{12}O_{19}$ with variations in composition (x = 0; 0.02; 0.04; and 0.08) has a single phase in all compositions. Figure 3, it appears that the curve forms a closed-loop hysteresis pattern, in other words, that the relationship between M and H is not linear. When the intensity of the magnetic field H is increased to reach H = 1 Tesla, the value of M experiences a saturation point called saturation magnetization, then when the H magnetic field is lowered, the curve trajectory does not return past the original curve. At this price of H = 0, magnetization M has a certain value. This condition is called Mr remanent magnetization or material remanence. Furthermore, the price of the intensity of the H magnetic field is lowered continuously (negative value), the M curve will cut the axis on the magnetic field H which is denoted as Hc. This Hc intensity is needed to make the flux density B = 0 or remove flux in the material. The intensity of the Hc magnet is called material coercivity or coercivity field.

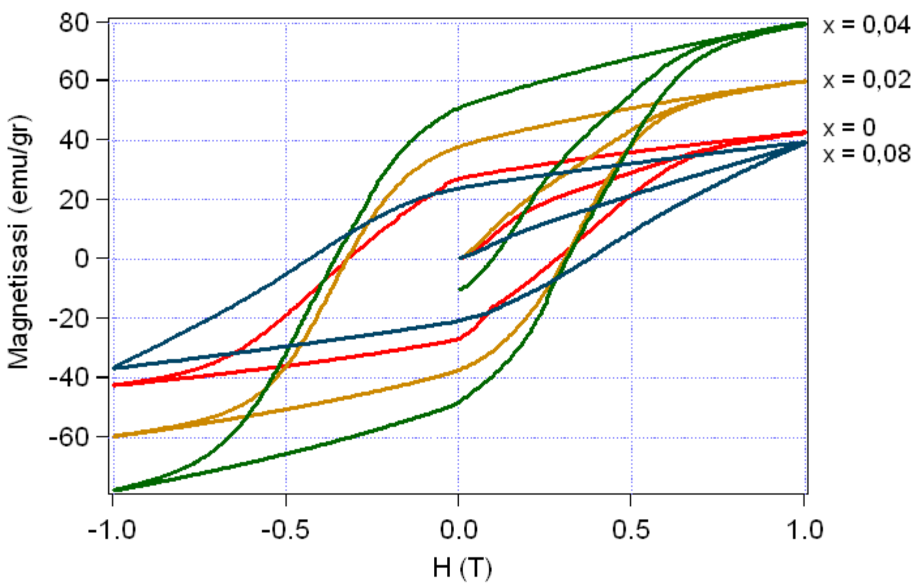


Figure 3. Sample hysteresis curve $Ba_{1-x}La_xFe_{12}O_{19}$ with variations in composition (x = 0; 0.02; 0.04; and 0.08)

This shows that the $Ba_{1-x}La_xFe_{12}O_{19}$ sample for all of these compositions has ferromagnetic properties and belongs to the hard magnet. It is indicated that all samples have a very large energy anisotropy which is indicated by the increasing coercivity field value of the material along with the increase in lanthanum content. This means that lanthanum has succeeded in making this material's magnetic domain become more numerous so that it is thought to have an impact on the increasing number of the hard axis which is one of the characteristics of anisotropic energy increase from this material. But that does not mean that the greater the value of this coercivity field makes the permanent magnet better. There are two main factors that influence if the permanent magnet is said to be good, ie the material must have the greatest coercivity field and the largest remanent magnetization or often called the energy product, namely the multiplication between the coefficient field and the remanent induction field. Based on the results obtained from this study that the best product energy is obtained at the composition of x = 0.04. So in this study, it can be concluded that the composition of $Ba_{0,96}La_{0,04}Fe_{12}O_{19}$ is a permanent magnet with the best product energy.

4. Conclusion

Permanent magnets based on $Ba_{1-x}LaxFe_{12}O_{19}$ with variations in composition (x = 0; 0.02; 0.04; and 0.08) have been understood. Based on the results of the XRD measurements showed that the substitution of the lanthanum atom into the barium atom was successfully carried out. The effect of substitution of lanthanum into the barium atom can increase the number of magnetic domains which are indicated by the increase in the coercivity value of the material along with the increasing composition of x. Based on the results obtained from this study that the best product energy is obtained at the composition of x = 0.04. So in this study, it can be concluded that the composition of $Ba_{0,96}La_{0,04}Fe_{12}O_{19}$ is a permanent magnet with the best product energy.

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