



RESEARCH ARTICLE

Synthesis and magnetic properties of barium hexaferrite/ magnetite composite

M. Sindhuja, R. Justin Joseyphus*

Department of Physics, National Institute of Technology, Tiruchirappalli 620015, Tamil Nadu, India

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Abstract

The exchange spring effect with the combination of hard/soft magnetic composite could be used for permanent magnet applications. The magnetic properties of $\text{BaFe}_{12}\text{O}_{19}$ and Fe_3O_4 hard/soft magnetic ferrites are studied with different ratios of the magnetic particles. The magnetic properties are influenced by the synthesis parameters such as pH annealing temperature and the ratio of the hard-soft phases. High coercivity is obtained with hard/soft fraction upto 1:0.5 whereas it decreases with further increase in soft magnetic fraction.

Keywords

Coercivity

Saturation magnetisation

Exchange coupling

Introduction

The interaction of the permanent intermetallic magnetic particles have been studied for more than two decades though it is not fully explored in ferrites. Hence an attempt has been made to understand the magnetic properties of barium hexaferrite and magnetite composite. Barium hexaferrite is an important class of magnetic material which has high corrosion resistance, high chemical stability and lower cost compared to other rare earth intermetallic permanent magnetic materials [1,2]. The M-Type ferrite with the chemical formula of $\text{BaFe}_{12}\text{O}_{19}$ is cheaper to produce and has high magnetic uniaxial anisotropy along the c-axis compared to the other permanent magnetic materials. Recent developments in nanoscale permanent

magnetic materials suggest applications in areas such as high density recording, magnetic particle hyperthermia and in drug delivery using the particles prepared through chemical. [3-8] Various chemical methods are utilized for the preparation of barium hexaferrite such as co-precipitation [9], micro emulsion [10], hydrothermal [11] and sol-gel method [12-17], reverse micro emulsion [18]. However, the synthesis of single phase barium hexaferrite requires annealing temperature as high as 1000 °C [17], which makes it difficult to prepare them in the asprepared form without the requirement of post annealing. The sol-gel method with the citrate precursor is the potential technique as it is cost effective and less time consuming method to produce size reduced particles on a large scale at a lower annealing temperature. Although nanoparticles could be prepared using the sol-gel auto combustion method with citrate precursor, few drawbacks such as longer Annealing duration exists. Therefore composite nanoparticles could be prepared with annealing, which could be utilized for high energy product applications

*Corresponding author

E-mail: rjustinj@nitt.edu

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Precursor, few drawbacks such as longer annealing duration exists. Therefore composite nanoparticles could be prepared with annealing, which could be utilized for high energy product applications. The exchange spring effect in hard and soft magnetic materials is proposed to enhance the energy product [19]. The exchange coupling with hard-soft ferrites was studied by tailoring the microstructure in barium hexaferrite and nickel zinc ferrite [20]. Barium hexaferrite/magnetite was studied with different ratios and ball milled for 40 h and studied under various annealing temperatures [21]. Though it is studied using top down approach, we have synthesized the barium hexaferrite/magnetite composite with the chemical method and studied the influencing parameters on the magnetic properties.

Experimental

Synthesis

Barium hexaferrite is prepared using the sol-gel autocombustion method. In this method, we have used citrate acid as the fuel for the reaction to take place. The ratio of metal ions to the citric acid is 1:2. The citric acid ratio is the main parameter in the formation of the barium hexaferrite and to reduce the formation of intermediate ferrites, where it chelates the iron cations which are in excess and enhance the formation of barium hexaferrite. The procedure for the synthesis of barium hexaferrite is shown in **Fig 1**. The chemicals, $\text{Ba}(\text{NO}_3)_2$ and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ of analytical purity are added in the molar ratio of 1:12 and dissolved in 100 ml of deionised water.

Citric acid which is then added into the prepared aqueous solution chelates Ba^{2+} and Fe^{3+} in the solution. The solution, while mixing with magnetic stirrer, is maintained at 80°C and the pH is varied to 1, 7 and 10 by adding ammonia. The optimum pH for the synthesis is found to be 7. The solution is stirred for 3h to form a gel and it is evaporated in a heating mantle at 150°C for few minutes till it is completely dried and burnt ash is obtained. The powdered sample obtained is the as prepared sample which is annealed at various temperature for 1h.

The prepared dried gel of barium hexaferrite is made homogeneous in the suspended liquid and then chloride and ferric chloride are added in the ratio of 1:2 with the excess of sodium hydroxide added at 80°C forming magnetite in barium hexaferrite matrix. The black substance formed at the bottom of the vessel

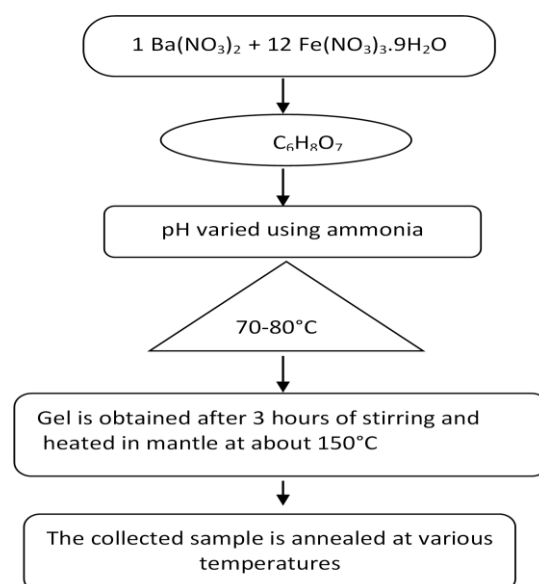


Fig 1 Flow chart showing the procedure for the synthesis of barium hexaferrite using sol-gel autocombustion method.

is separated using magnetic separation and the powders are washed and dried. As the matrix is already barium hexaferrite, the samples are annealed at 950°C for one hour at a heating rate of $10^\circ\text{C}/\text{min}$ with various barium hexaferrite-magnetite ratios.

Characterization

The X-ray diffraction (XRD) patterns were recorded using a Rigaku Ultima 3 X-Ray diffractometer equipped with $\text{Cu K}\alpha$. The magnetic characterization was carried out at room temperature with vibrating sample magnetometer (VSM) (Model 7404, Lakeshore USA).

Results and discussion

X-Ray Diffraction analysis

The hard and soft magnetic composites synthesized with barium hexaferrite to magnetite ratios of 1:1, 1:0.5, 1:0.25 are analysed using the XRD as shown in the **Fig 2**. When the ratio of ferrite is much reduced, the dominance of hard magnetic phase is seen along with $\beta\text{-Fe}_2\text{O}_3$, which is formed only in the smallest ratio. Small quantities of hematite, with the rhombohedral crystal structure is seen. Hence the peaks of most of the oxide compounds are observed when the ratio of barium Hexaferrite to magnetite is 1:0.25. When the ratio of soft ferrite is decreased, Fe_2O_3 and $\alpha\text{-Fe}_2\text{O}_3$ are also present at a higher value with barium hexaferrite. With equal ratio of hard and soft magnetic sample, we have obtained the highest intensity peak with magnetite and lower barium hexaferrite and $\alpha\text{-Fe}_2\text{O}_3$.

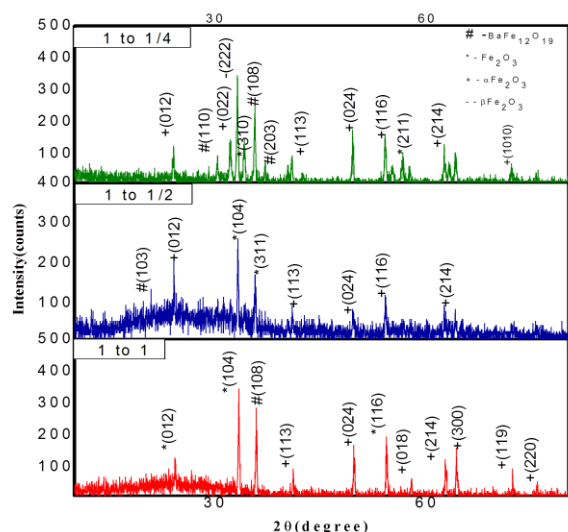


Fig 2 XRD pattern of prepared samples with the ratio of hard to soft phase fraction 1:1, 1:0.5 and 1:0.25.

Magnetic characterization

The hysteresis loops of prepared samples with the ratio of hard to soft fractions 1:1, 1:0.5 and 1:0.25 are shown in **Fig 3**. The presence of β - Fe_2O_3 has been noted for the sample annealed at 950°C . This is the paramagnetic form and hence there is reduction in the saturation magnetization. The coercivity has decreased from 5302 Oe to 4896 Oe and the saturation magnetization changes from 23 emu/g to 17 emu/g when the hard soft ratio is changed from 1:0.25 to 1:0.5, respectively. As the magnetic material barium hexaferrite is prevalent, the coercivity is higher. When the sample is then prepared with the equal ratio of hard and soft magnetic materials tremendous amount of loss in coercivity and saturation magnetization to 2257 Oe and 3 emu/g, respectively because of the dominance of soft ferrite. We have obtained better magnetic properties than in [21] where the reported coercivity is very less when the fraction of hard soft phases are 1:0.5 and 1:0.25. In the composites higher coercivity and lower saturation magnetisation is noticed because of the single domain nature and the pinning of domains by various Fe oxide phases, where the domain rotation is hindered.

Conclusions

We have synthesized soft/hard ferrite composite at different ratios of barium hexaferrite/magnetite such as 1:1, 1:0.5 and 1:0.25 through chemical route. The experiments conclude that high coercivity barium hexaferrite is formed at the neutral pH. Magnetic analysis shows that the magnetic properties depend on the pH and the annealing temperature. A comparatively high coercivity is obtained in the hard soft composite due

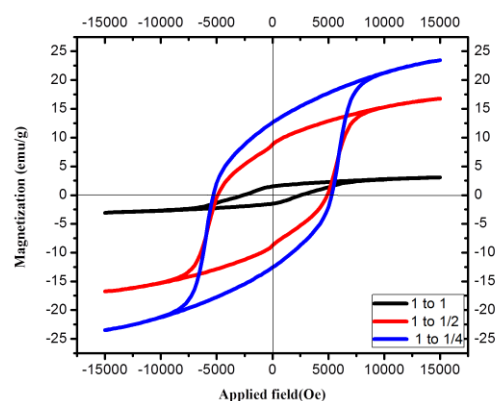


Fig 3. The hysteresis loops of prepared samples with the ratio of hard to soft phases 1:1, 1:0.5 and 1:0.25.

to the pinning of magnetic moments by the distributed phases. The coercivity is not reduced significantly with a hard/soft fraction up to 1:0.5.

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