Investigation of microstructure and Magnetic properties of composite (soft-hared) ferrites, synthesized via sol-gel method

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ABSTRACT: This work examines the production and characterisation of a nanocomposite powder made of hard barium ferrite and soft nickel ferrite. With three distinct ratios of nickel ferrite to barium ferrite— 100NF/0BF, 0NF/100BF, 50NF/50BF, 25NF/75BF, and 75NF/25BF—nanocomposite is made using the sol-gel process. In a cylindrical stainless steel die, ferrite powder is uniaxially compressed into pellets for 40 seconds at 150 MPa, and then sintered at 1200°C. Different methods, including XRD, are utilized to describe nanocomposite ferrite. The XRD Pattern clearly demonstrates the production of composite phases. The nickel ferrite spinal structure and the peaks of the hexagonal barium ferrite phase are visible. An absorption band between the constituent constituents of the nanocomposite ferrite is visible in the FT-IR. The spherical form of the nickel ferrite powder and the needle-like shape of the barium ferrite were visible in SEM pictures, and the VSM test revealed a difference magnetization. Ms The highest Ms finding in ratio 75NF/25BF is equal to (28.348 emu/g), the highest Hc value in ratio 25NF/75BF is equal to (205.198 Oe), and the greatest Mr value in ratio 75NF/25BF is equal to (2.296 emu/g).

Keywords: nickel ferrite, barium ferrite , nanocomposite ferrite , soft ferrite , hard ferrite , sol-gel method

1. INTRODUCTION

The increased capabilities of nano composite materials that have a precise blend of hard and soft magnetic phases make them candidates for a range of possible applications as permanent magnets [1]. The two phase magnetic nano composites' magnetic behavior is significantly influenced by the exchange and dipolar interactions and even the size, distribution, and shape of the grains. Obtaining high coercivity (Hc) and high saturation of magnetization (Ms) concurrently is a significant difficulty for magnetic materials. As a result, by combining hard and soft phases create composite materials resulted from various attempts to reconcile this issue. [1]. Whereas the phase of hard magnetism introduces strong coercivity, High saturation magnetization is produced by soft phase. Additional research confirmed the extensive homogenous mixing For the creation of exchange spring magnets, The hard and soft ferrite stages have to be balanced [2,3].where a Soft ferrites have high Ms and low Hc are important attributes for soft ferrites. Soft ferrites have small hysteresis loops as a result [4]. The soft ferrites that are most frequently encountered are [5]. NixZn(1-x)Fe_2O_4, commonly known as nickel-zinc ferrite (NiZn), NiZn ferrites are better suited for frequencies over 1 MHz due to their higher resistivity than MnZn ferrites. One of the useful and important softer ferrites for technology is nickel ferrite. [6]. Hard ferrites, on the other hand, which after magnetization have a strong remanence and a high coercivity, are used to create permanent ferrite magnetics. Hard ferrite magnetic made by using iron oxide, barium or strontium carbonate [7] [8]. Ferrite composites composed of hard ferrites and spinel soft are desirable candidates for enhanced permanent magnets because of their higher resistance to corrosion, comparatively high Curie temperature, and high electrical resistivity [9]. Hard and soft magnetic materials may exhibit exchange coupling in specific circumstances. When the high HC from the hard material and the high MS from the soft material are combined into one material (nano composite), the exchange spring magnets, the product (BH)max is significantly increased compared to any one of the nano composite's component phases [10]. The aims of this research to study the effect of combining hard and soft ferrites on the magnetic properties . Modern scientists are interested in useful hard-soft magnetic composites, scientists have developed new methods for integrating hard-soft ferrite materials . such as Rai, et al, in (2014) he is study Synthesis and magnetic properties of hard-soft SrFe10Al2O19/NiZnFe2O4 ferrite nan ocomposites[11]. Neupane, et al, in (2017) study Synthesis and magnetic study of magnetically hard-soft SrFe12-yAlyO19-x Wt.% Ni0. 5Zn0. 5Fe2O4 nano composites[12] and Tavakolinia, et al ,in (2018) study novel hard/soft ferrite composite particles with improved exchange coupling are created and magnetic characteristics[13].

2. MATERIALS

The starting materials used are "Ni $(NO_3)_2.6H_2O$" nickel nitrate hydrate (purity is 99.5 , source of the material is fluka in turkey), Barium nitrate $Ba(NO_3)_2$ (purity is 99.1 , source of the material is fluka in turkey) and hydrate "Fe $(NO_3)_3.9H_2O$" Iron nitrate (purity is 99.1 , source of the material is SDFCL in india) that was dissolved in a specified amount of ethylene glycol $C_2H_4(OH)_2$ solution (purity is 99.0 source of the material is Thomas baker in india).

3. Preparation of Composite particles hard –soft ferrites

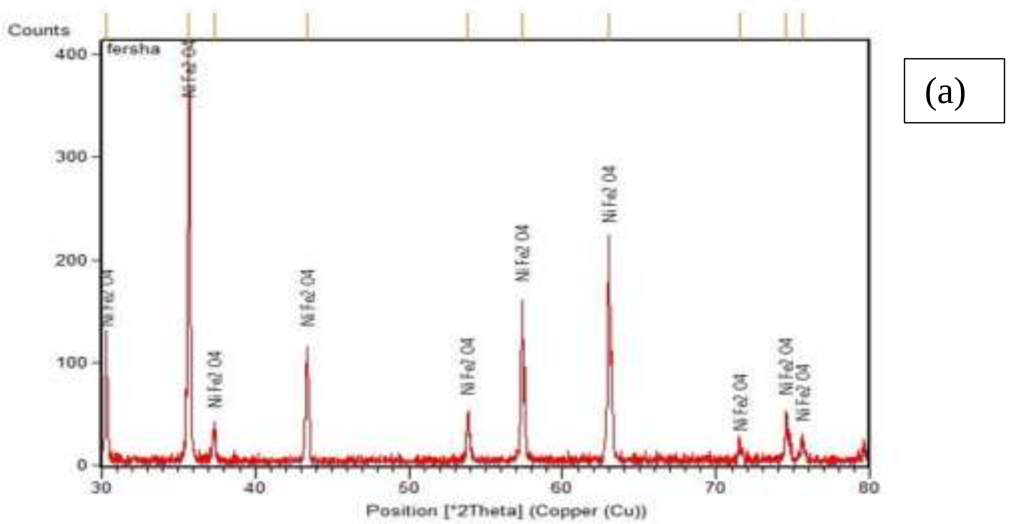
Stoichiometric quantities of Ba(NO3)2 and Fe(NO3)3.9H2O nitrates were dissolved in ethylene glycol. In the same way stoichiometric quantities of Ni(NO3)3•6H2O and Fe(NO3)3•9H2O were combined and dissolved in another beaker in ethylene glycol. The solution was then added to the first beaker containing barium hexaferrite salts, and 120°C was used to heat the combined solution. After the gel formation is caught fire in the microwave at190 °C for 7 hours , creating the dark colored powder. After the calcination process and the formation of the powder, to prepare the pellets as sintered samples for different measurements, the dry powder without binder is pressed uniaxial into pellets with a pressure of 150 MPa for 40 s in a cylindrical stainless steel die. The diameter of the pellets was 10 mm, while the thickness varied from 0.9 to 1.2 mm, according to requirements of the structural and physical properties. After the process of pressed powders for (nickel ferrite) and (barium ferrite), by the sol- gel method will be calcination in an electrical digital furnace(JYMF-1800, High Temperature Muffle Furnace) was used for calcinated all powder the prepared at (800° C to composite powder) with heating rate 7° C/ min. In order to determine the microstructural and magnetic properties of nanocomposite ferrite prepared by sol-gel method and at two burning temperatures, the following tests are performed.

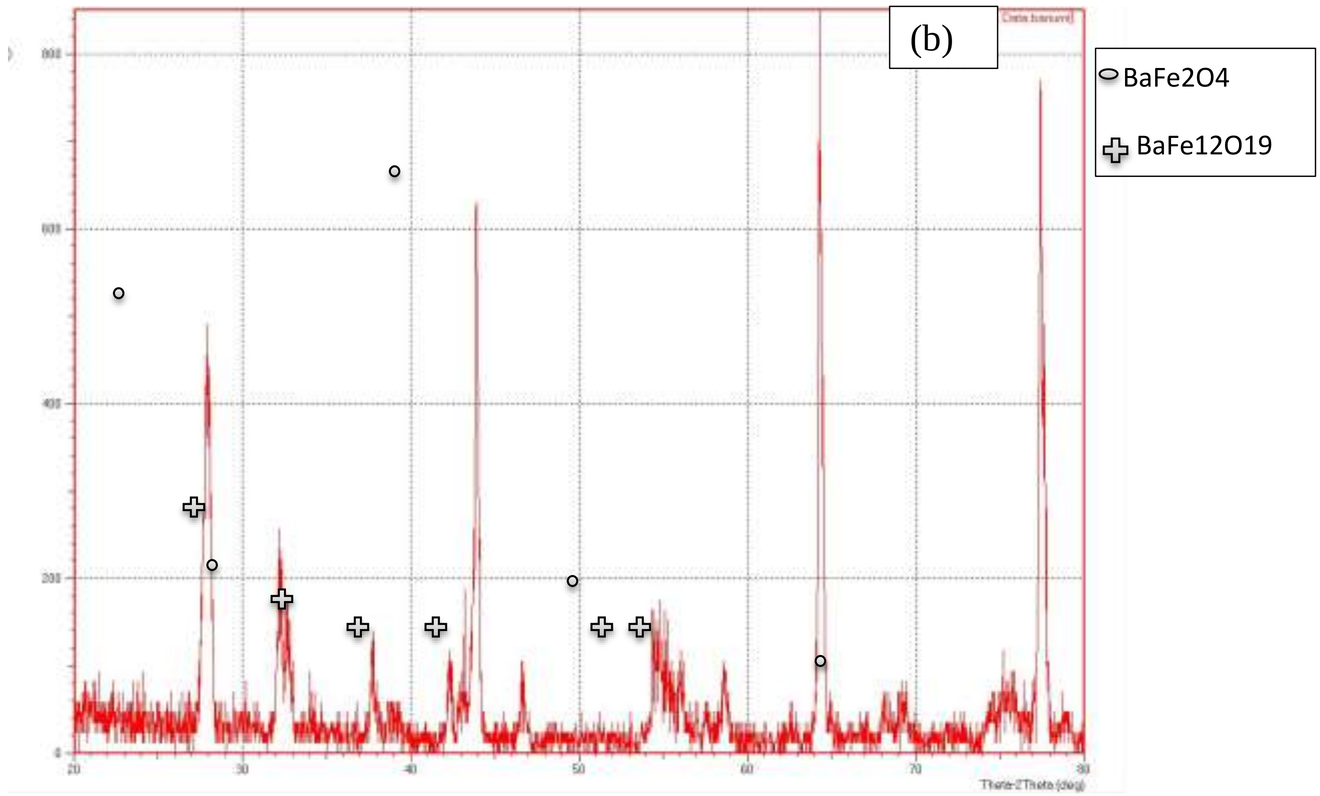
4. RESULTS AND DISCUSSION

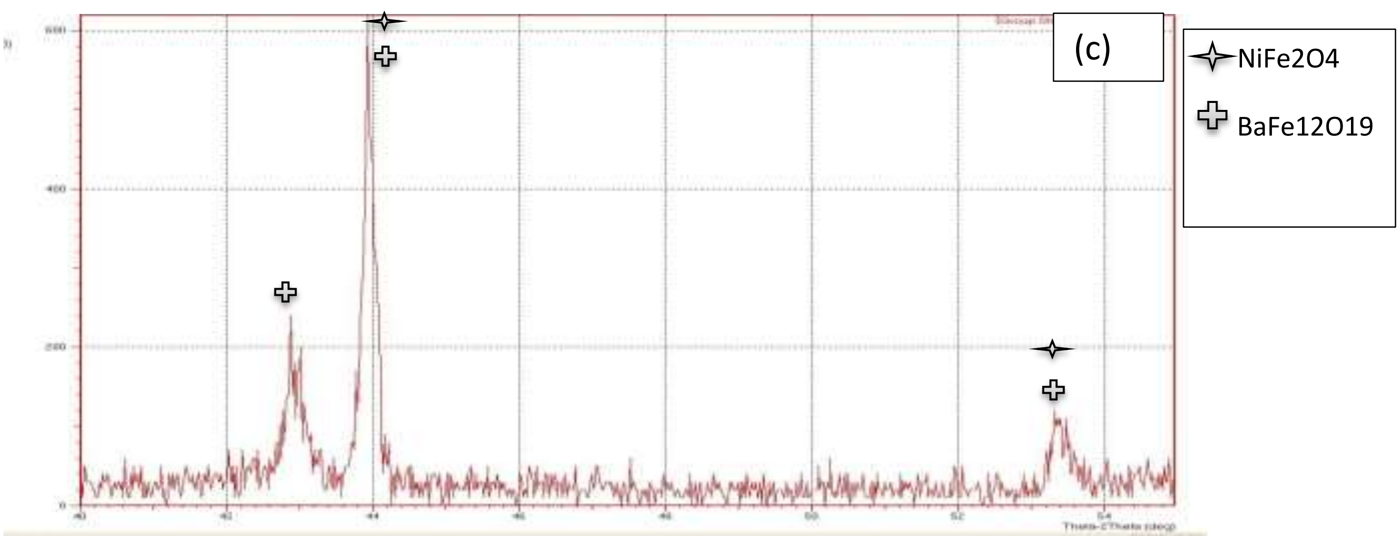
This section provides explanations of the experimental measurements. In order to determine the microstructural and magnetic properties of nanocomposite ferrite prepared by sol-gel method the following tests are performed.

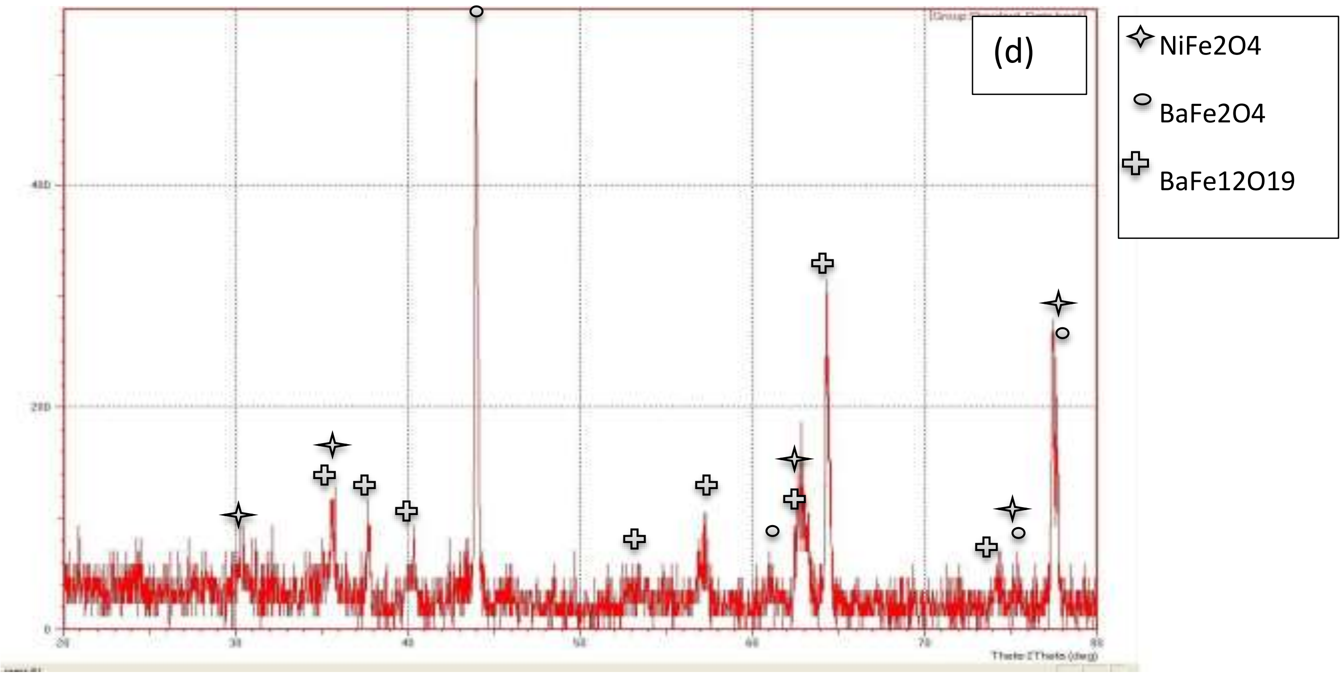
4.1. X-ray-diffraction analysis

The generated Nickel ferrite powder is examined Figure (1 ,a) illustrates X-ray diffraction pattern for NiFe2O4 spinel nanoparticles(100NF/0BF) . It is evident that there is no second phases were formed and the main characteristics peaks are appeared which are 30.580, 35.987, 37.378, 43.678, 53.988, 57.628, and 63.098, respectively, correspond to the single crystal (2 2 0), (3 1 1), (2 2 2), (4 0 0), (4 2 2), (5 1 1). it exhibited that the primary peaks are more sharp with high intensity which indicates to high crystallinity [14]. The diffraction patterns (XRD) for barium ferrite after sintering are shown in Figure (1,b) The identification (creation) of barium ferrite peaks is seen more shaped and clearly at 1200 °C, according to the XRD data. Another phases of barium ferrite is (BaFe12O19) may also be seen in the sample with barium ferrite phase (BaFe2O4) This agree with Ref [15]. At temperature 1200 °C, XRD and a crystalline phase of it shown in figure (1, c), where each of the main peaks of the two materials appeared and were more sharp and clear as a result of sintering at a high temperature, the result of the composite nanopowder when the ratio of nickel ferrite to barium ferrite 75NF/25BF is matched with card number [00-054-0964]. The result of the composite nanopowder at temperature 1200 °C when the ratio of nickel ferrite to barium ferrite 25NF/75BF is matched with card number [01-074-1913], shown in figure (1, b). Where shown the other phases of barium ferrite , appear the two phases of barium there are ($BaFe_2O_4$ and $BaFe_{12}O_{19}$) with a little peaks of nickel ferrites. XRD at temperature 1200 °C revealed nickel and barium ferrite with a ratios 50NF/50BF . The result of composite nano powder matched with card number [01-074-2081] shown in figure (1,c) . shown the main peaks of barium and nickel ferrites with high crystallite and sharp , where shown two phases of barium ($BaFe_2O_4$, $BaFe_{12}O_{19}$). The relative weight ratio of the soft and hard phases affects the diffraction peak intensities of the (barium and nickel) phases. No additional phases were seen within the x-ray diffraction patterns' resolution limit, showing that the two parent phases (soft and hard ferrite) could exist in the two-phase nano composites and that they had high chemical compatibility. This result is contestant with previous study [16].




(b)
BaFe2O4
BaFe12O19
Theta-2Theta (deg)


(c)
NiFe2O4
BaFe12O19



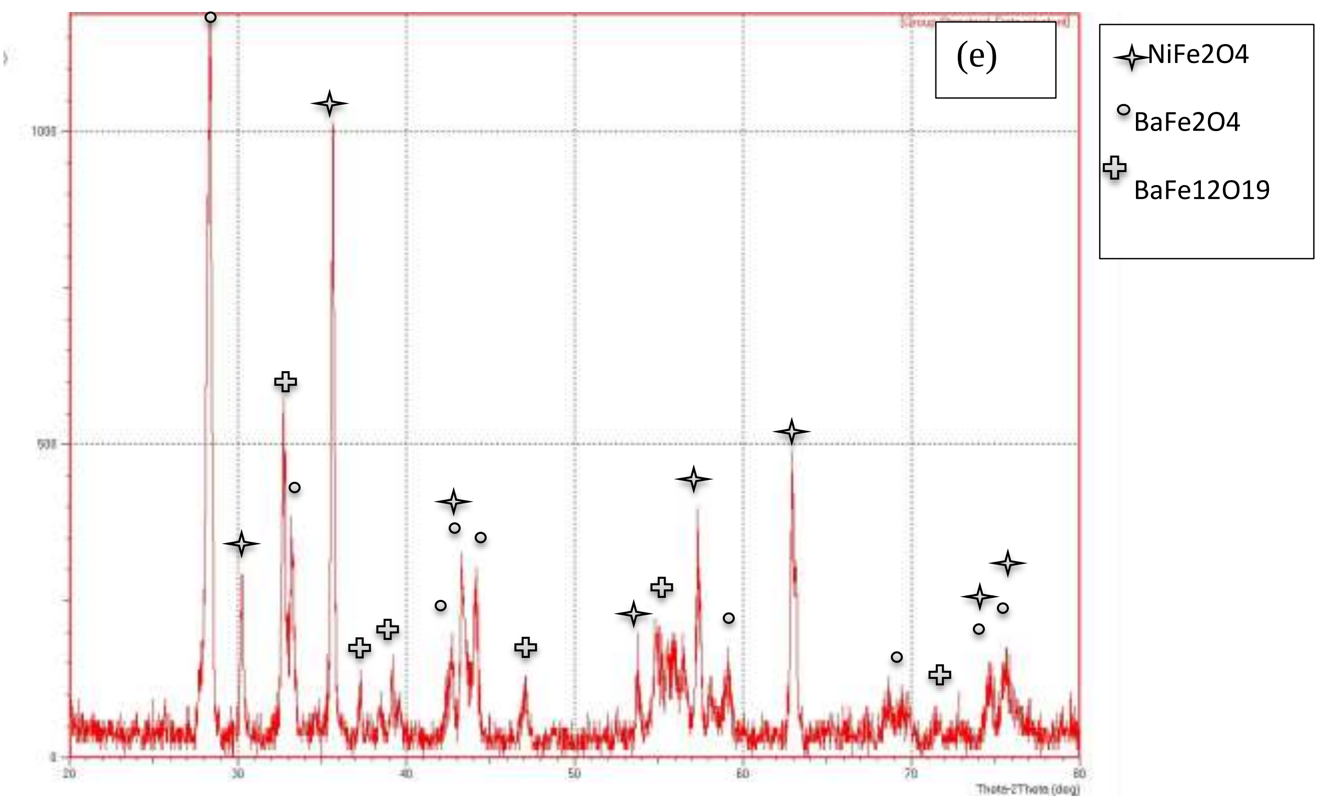
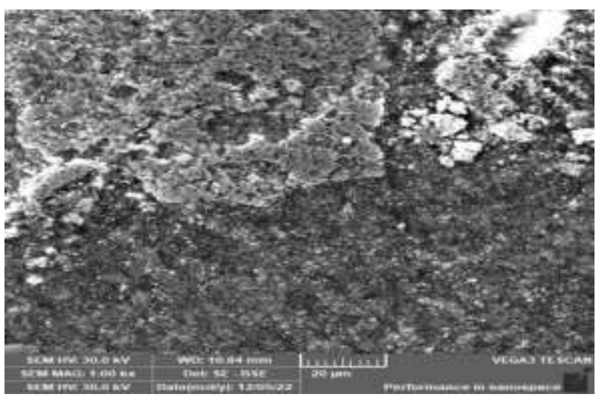


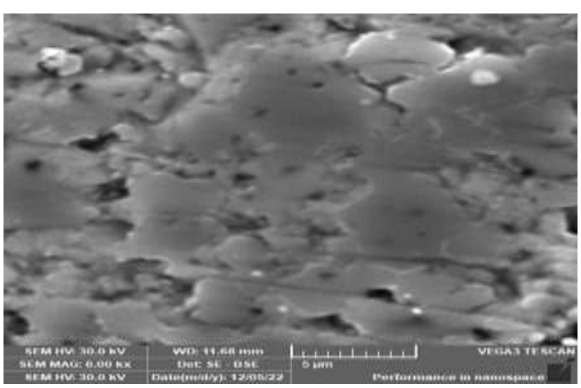
Figure 1: a- XRD of nickel to barium ferrites 100:0 at 1200, b- XRD of nickel to barium ferrites 0:100 at 1200,c- XRD of nickel to barium ferrites 75:25 at 1200 , d- XRD of nickel to barium ferrites 25:75 at 1200, e- XRD of nickel to barium ferrites 50:50 ,at 1200

4.2 Scanning Electron Microscopy (SEM) analysis

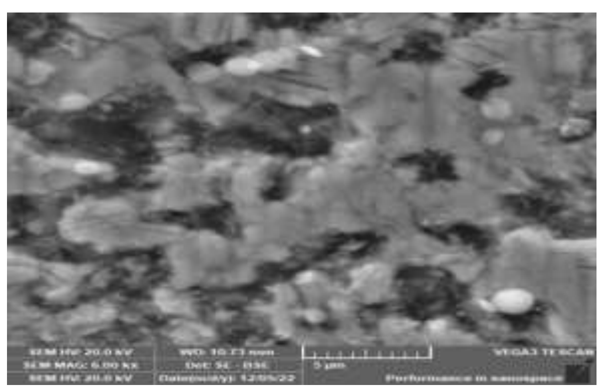
The produced samples (nickel ferrite/barium ferrite samples) are used to provide information on the morphology using the powerful technology known as SEM. In the figure (2, a) shows the SEM images of the sample in which the ratio of nickel ferrite to barium ferrite is 75NF/25BF, where the images appear after sintering and pressing processes. The shape of the spherical granules that represent the nickel ferrite is more widespread than the hexagonal shape of the barium ferrite granules. The same is the case with the shape(.2 ,b) where the percentage of nickel ferrite to barium ferrite is 25NF/75BF, and notice the spread of the hexagonal shape and its clarity represented by barium ferrite, as well as the presence of Pores spread between the two phases after the sintering process. the SEM images of the composite material with a ratio of 50NF/50BF of nickel ferrite and barium ferrite shown in figure (2 c) Typical SEM images after sinteringg at 1200 °C as show all pictures show two distinct phases, Most barium ferrite grains have shape of a hexagonal granule, and the value after the sintering process is the growth of the granules, as well as for the granules of nickel ferrite, and we note that there is no cracking in the sample after the sintering and pressing process.



(a)



(b)



(c)

Figure 2: images SEM : a- nano composite with ratio 50/50 at 1200 °C b – nano composite with ratio 75/25 nickel ferrite / barium ferrite at 1200 °C c - nano composite with ratio 75/25 nickel ferrite / barium ferrite at 1200 °C

4.3 FT-IR Analysis

The FT-IR spectrum is shown in Figure (3) of composite nickel/barium ferrites at 1200 °C ,in the range of 400 cm-1 to 4000 cm-1 for composites , This figure supports the structural details of the creation of composites that was previously described using XRD patterns. It may be said that barium ferrite/nickel ferrite nanocomposites were successfully made as a result. The composites' spectra exhibit absorption bands at wavelengths of 440 cm-1, 541 cm-1, and 595 cm-1. The stretching vibration of the M-O bonds form at the tetrahedral location is cause of high-frequency band between 570 and 610 cm-1. Additionally, the bands between 390 and 440 cm-1 have a connection to the vibration of metal and oxygen at the octahedral site. The presence of bands in samples supports the ferrites structure's creation in produced nanocomposites. These bands clearly alter somewhat when the hard/soft content ratio rises, which may be a result of hard and soft phases coupling in nanocomposites. This agrees with Ref. [17].

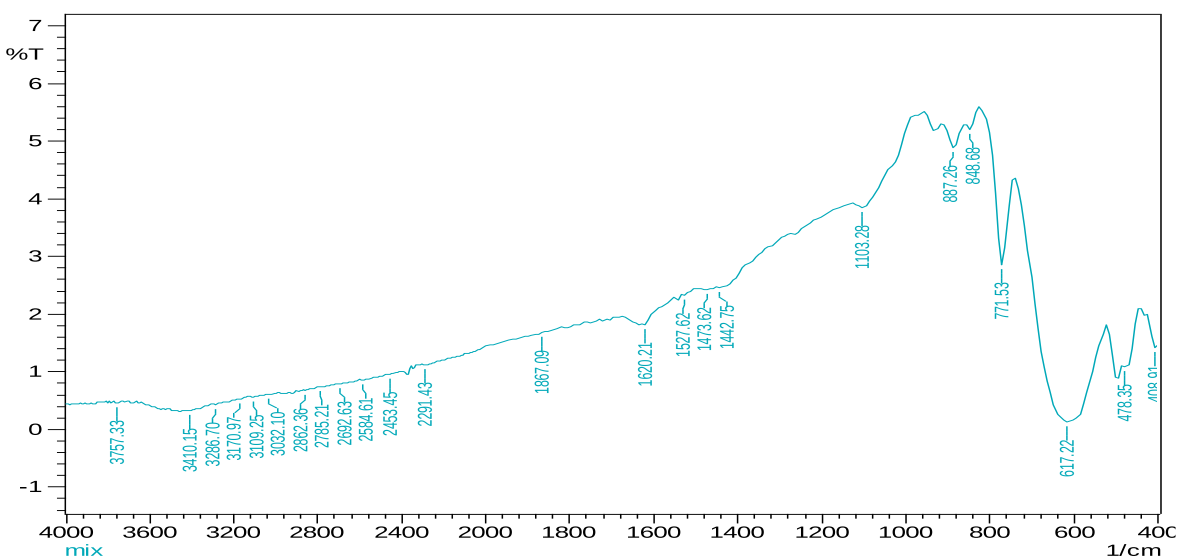


Figure 3: FT -IR spectrum composite nickel/barium with ratio 50/50 ferrites at 1200 °C

4.4 Vibrating Samples Magnometer (VSM) measurement

To investigate the behavior of the produced nanocomposites' exchange spring, room-temperature hysteresis loops were obtained, Figure (4,a) shows hysteresis loop of the soft /hard with ratio

75NF/25BF nickel ferrite /barium ferrite sample sintering at 1200°C. This sample's single smooth hysteresis loop, which significant exchange coupling between the hard and soft phases is indicated, can be observed clearly. However, Along with the dipolar interaction, the exchange interaction also plays a significant role in the coercivity of the soft phase, since it dropped as the weight ratio of the soft phase increased. Barium hexagonal ferrite has a substantially coercivity than Nickel spinel ferrite [18]. To raise both the alignment and the magnetization of magnetic systems, both hard and soft, exchange interaction really aids in aligning , magnetic moments of hard and soft phases are parallel, can be shown in figure (4,b). Figure (4,c) demonstrates the hysteresis loops of the soft/hard nanocomposite powders ,ratio 25NF/75BF nickel ferrite / barium ferrite at 1200°C. This sample's single hysteresis loop, which indicates between the hard and soft phases, there is a significant exchange coupling, can be observed clearly. The corictivity increased as the amount of hard magnetics phase increased [18].In table (1) show magnetic properties of Ferrites samples.

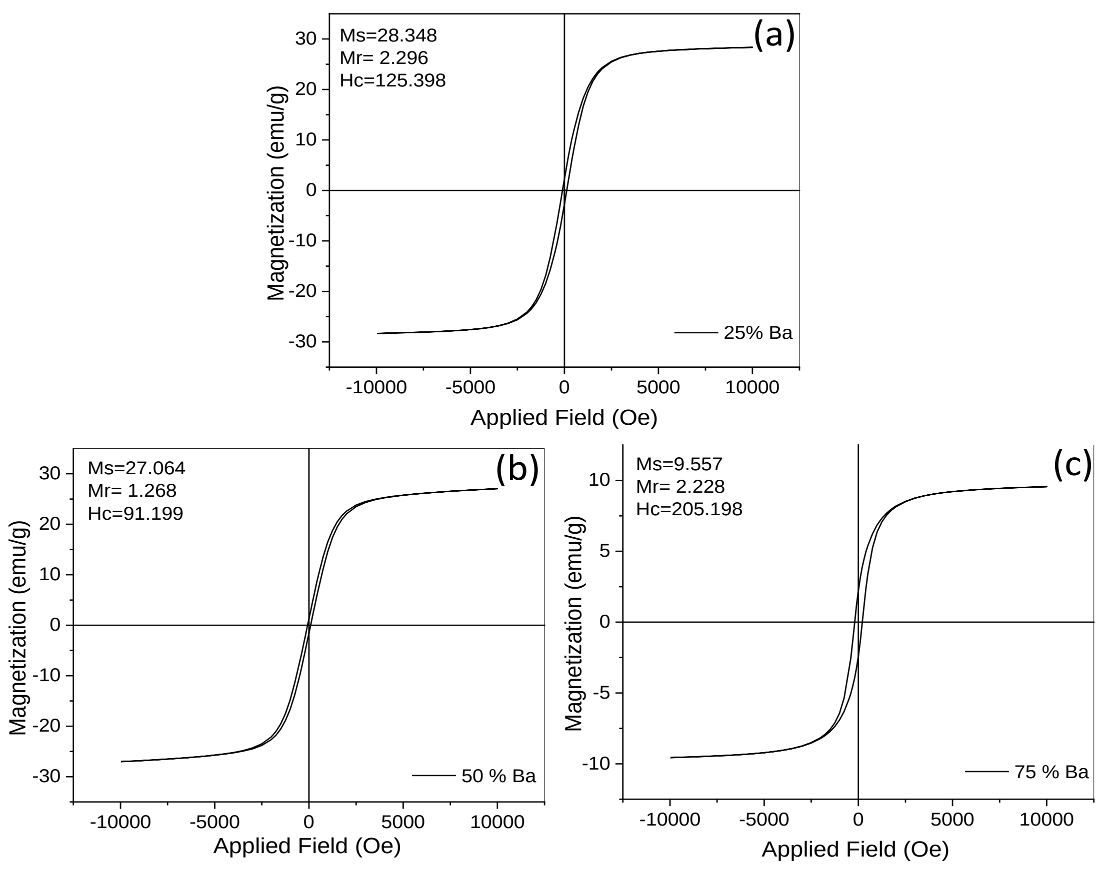


Figure 4: a- hysteresis loops soft(nickel) /hard(barium) with 25% Ba sintering at 1200°C b- hysteresis loops soft(nickel) /hard(barium) with 50% Ba sintering at 1200°C c- hysteresis loops soft(nickel) /hard(barium) with 75% Ba sintering at 1200°C

Table 1: Magnetic properties of Ferrites Samples

Ferrites	Calcination temp.(°C)	Ms (emu/g)	Mr (emu/g)	Hc(Oe)
Composite ferrite with a ratio 25BF/75NF	1200°C	28.348	2.296	125.398
Composite ferrite with ratio 50NF/50BF	1200°C	27.064	1.268	91.199
Composite ferrite with a ratio 25NF/75BF	1200°C	9.557	2.228	205.198

CONCLUSION

1. The possibility of synthesized nanocomposite with different ratio by using sol-gel method
2. XRD were shown the tow different peaks of barium and nickel ferrite depend on the ratio of nickel ferrite and barium ferrite, the two different phases of barium ferrite (BaFe2O4 , BaFe12O19) appear with peaks of nickel ferrite (NiFe2O4) after sintering at 1200 °C .
3. In the FT-IR for nanocomposite ferrite after sintering at 1200 °C show absorption bands at 440 cm-1,541cm-1-,and 595cm-1,are visible in the wavenumber region of 1000-400 cm-1.
4. The SEM photos of the composite material for a pressed sample after sintering at 1200°C incressing in a certain size showed A typical distribution of both magnetic phases is supported by the finding of both nanoplatelets with large hexagonal phase and quasispherical nanoparticles of spinel phase.
5. The hysteresis loops of the room temp ferromagnetism RTFM have different magnetization Ms , different hysteresis loops, single smooth hysteresis loop for a nanocomposite . The highest Ms finding in ratio 75NF/25BF is equal to (28.348 emu/g), the highest Hc value in ratio 25NF/75BF is equal to (205.198 Oe), and the greatest Mr value in ratio 75NF/25BF is equal to (2.296 emu/g).

REFERENCES

1.Shahab Torkian, Ali Ghasemi , Reza Shoja Razavi, Magnetic properties of hardsoft SrFe10Al2O19/Co0.8Ni0.2Fe2O4ferrite, synthesized by one-pot sol–gel autocombustion,mJ. Magn. Magn. Mater. 416 (2016) 408–416.

2. S. Tyagi, P. Verma, H. B. Baskey, R. C. Agarwala, V. Agarwala, T. C. Shami, Microwave absorption study of carbon nano tubes dispersed hard/soft ferrite nanocomposite, Ceram. Int. 34 (2012) 4561–4571.

3. X. Q. Shen, F. Z. Song, J. Xiang, M. Q. Liu, Y. W. Zhu, Y. D. Wang, ShapemAnisotropy, Exchange-Coupling Interaction and Microwave Absorption of Hard/Soft Nanocomposite Ferrite, J. Am. Ceram. Soc. 95 (2012) 3863–3870.

4. Okamoto, A. (2009). "The Invention of Ferrites and Their Contribution to the Miniaturization of Radios". 2009 IEEE Globecom Workshops. pp. 1–42. doi:10.1109/GLOCOMW.2009.5360693. ISBN 978-1-4244-5626-0. S2CID 44319879.

5. Ullah, Zaka; Atiq, Shahid; Naseem, Shahzad (2013). "Influence of Pb doping on structural, electrical and magnetic properties of Sr-hexaferrites". Journal of Alloys and Compounds. 555: 263–267. doi:10.1016/j.jallcom.2012.12.061

6.Gunjakar JL, More AM, Gurav KV, Lokhande CD. Chemical synthesis of spinel nickel ferrite (NiFe2O4) nano-sheets. Appl Surf Sci. 2008;254(18):5844–5848. doi: 10.1016/j.apsusc.2008.03.065. [CrossRef] [Google Scholar]

7. "Ferrite Permanent Magnets". Arnold Magnetic Technologies. Archived from the original on 14 May 2012. Retrieved 18 January 2014. .

8. Barium Carbonate". Chemical Products Corporation. Archived from the original on 1 February 2014. Retrieved 18 January 2014

9. S. Hazra, N.N. Ghosh, “Preparation of nanoferrites andtheir applications”, J. Nanosci. Nanotechnol.,14 [2](2014) 1983–2000

10.Coldebella, E. H., Chagas, E. F., Albuquerque, A. P., Prado, R. J., Alzamora, M., & Baggio-Saitovitch, E. (2021). Study of soft/hard bimagnetic CoFe2/CoFe2O4 nanocomposite. Journal of Nanoscience and Nanotechnology, 21(10), 5181-5187

11. Rai, B. K., Wang, L., Mishra, S. R., Nguyen, V. V., & Liu, J. P. (2014). Synthesis and magnetic properties of hard-soft SrFe10Al2O19/NiZnFe2O4 ferrite nanocomposites. Journal of Nanoscience and Nanotechnology, 14(7), 5272-5277

12. Neupane, D., Ghimire, M., Adhikari, H., Lisfi, A., & Mishra, S. R. (2017). Synthesis and magnetic study of magnetically hard-soft SrFe12-yAlyO19-x Wt.% Ni0. 5Zn0. 5Fe2O4 nanocomposites. AIP Advances, 7(5), 055602

13. Tavakolinia, F., Yousefi, M., Afghahi, S. S. S., Baghshahi, S., & Samadi, S. (2018). Synthesis of novel hard/soft ferrite composites particles with improved magnetic properties and exchange coupling. Processing and Application of Ceramics, 12(3), 248-256

14. Hassan, M. A., Saqib, M., Shaikh, H., Ahmad, N. M., & Elaissari, A. (2013). Magnetically engineered smart thin films: toward lab-on-chip ultra-sensitive molecular imaging. Journal of biomedical nanotechnology, 9(3), 467-474

15. Rashad, M. M., & Ibrahim, I. A. (2011). A novel approach for synthesis of M-type hexaferrites nanopowders via the co-precipitation method. Journal of Materials Science: Materials in Electronics, 22, 1796-1803

16. Nikmanesh, H., Moradi, M., Kameli, P., & Bordbar, G. H. (2017). Effects of annealing temperature on exchange spring behavior of barium hexaferrite/nickel zinc ferrite nanocomposites. Journal of Electronic Materials, 46(10), 5933-5941

17. Nikmanesh, H., Moradi, M., Kameli, P., & Bordbar, G. H. (2017). Effects of annealing temperature on exchange spring behavior of barium hexaferrite/nickel zinc ferrite nanocomposites. Journal of Electronic Materials, 46(10), 5933-5941

18. Nikmanesh, H., Moradi, M., Kameli, P., & Bordbar, G. H. (2017). Effects of annealing temperature on exchange spring behavior of barium hexaferrite/nickel zinc ferrite nanocomposites. Journal of Electronic Materials, 46(10), 5933-5941