Effect of Sintering Temperatures on Structural, Magnetic and Microwave Properties of Barium Ferrites/Epoxy Composites

Nurshahiera Rosdi, Raba'ah Syahidah Azis, Ismayadi Ismail, Muhammad Syazwan Mustaffa, Nurhidayaty Mokhtar

Abstract: ***This research highlights the structural magnetic and microwave properties of nanoparticles of M-type hexagonal barium ferrites ($BaFe_{12}O_{19}$). The samples were sintered at varied sintering temperatures (800, 900, and 1000 ℃). The effect of temperatures on the structural, magnetic and microwave properties was highlighted. Barium ferrites are well-known materials used for radar absorbing materials (RAM). RAM materials with good absorbing performance should have high permeability, small permittivity and high magnetic or dielectric loss at microwave frequency. High microwave absorption can be created effectively in magnetic materials, as well as wideband absorption. The structural, microstructural and microwave properties were analyzed via an X-ray Diffractometer (XRD), a Field Emission Scanning Electron Microscope (FESEM) and a Vector Network Analyzer (VNA), respectively. The XRD results showed a full phase hexagonal structure was formed in the samples sintered at 900 and 1000 ℃. $BaFe_{12}O_{19}$ composite with a thickness of 3 mm showed a minimum Reflection Loss (RL) at −9.01 dB at a frequency of 9.16 GHz at temperature 1000 ℃.***

Keywords: Barium hexaferrite, High energy ball milling (HEBM), Structural and microstructural properties, Electromagnetic and Microwave properties.

I. INTRODUCTION

Hexagonal structure ferrites have excellent magnetic properties for use in electromagnetic wave absorber of RAM [1]. Among all types of ferrites, M-type hexaferrite, in particular $BaFe_{12}O_{19}$, has been observed to reveal the largest saturation magnetization, suitable permeability values, and planar anisotropic conduct in microwave properties. [2]. In recent years, the development of technology based on electromagnetic (EM) radiation phenomena has become crucial owing to their use in modern life and as important elements of a stealth defense system for military reasons.

Revised Manuscript Received on July 22, 2019.

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EM wave absorbers can efficiently decrease the reflection of these waves that have become more appealing to distinct apps, and thus generating concern in the field between many researchers [3]. Microwave absorbers are usually split into two classifications of dielectric and magnetic absorbent materials that communicate with the occurrence of electromagnetic waves at a particular frequency of electromagnetic waves with ferromagnetic resonance, leading in peak energy absorption from incident waves. [4]. In this study, barium hexagonal ferrites were studied on X-band (8-12 GHz) frequencies. The effect of sintering temperatures on it's structural, microstructural and microwave properties were investigated.

In this works, $BaFe_{12}O_{19}$ was successfully prepared by using the simplest top down method which is HEBM. The nanocomposites then sintered with three different temperatures and were characterized by using XRD, FESEM, and VNA. With the good characteristics that presented, the nanocomposites would expect to have higher potential in absorbing materials applications.

II. EXPERIMENTAL

The raw materials which is hematite (Fe_2O_3) and barium carbonate ($BaCO_3$) were weighed according to the stoichiometric ratio and ground using an agate mortar for 1 h.

The raw ground powders were subjected to high energy ball milling using a SPEX800D HEBM machine for 6 h with the ball to powder (BPR) ratio 10:1 for high energy ball milling (HEBM). A Carbolite furnace was used to sinter all samples at 800, 900 and 1000 ° C for 6 h at a rate of 3 ° C / min. At a concentration of 70:30 by weight, ground samples of hexaferrite $BaFe_{12}O_{19}$ were then combined with epoxy resin (Araldite 506 epoxy resin, Sigma Aldrich) and hardener as filler. The sample blend was then dried in a metal back with a 1, 2 and 3 mm rectangular sample holder for 48 h. X-ray diffraction, XRD (Philips X'pert Diffractometer model 7602 EA Almelo) studied the phase structure of the composite samples. The measurement was performed out at room temperature, with a diffraction angle (2θ) between of 20° to 80°. Using X'pert HighScore software, the information was assessed and contrasted with an electronic PDF2 library Using field emission scanning electron microscopy, FESEM (FEI Nova NanoSEM 230), the microstructure of $BaFe_{12}O_{19}$ composite samples was detected.

Using a N5227A PNA network analyzer with using coaxial transmission / reflection technology, the complex permittivity, complex permeability and microwave absorbing characteristics of the prepared composite were performed (Fig. 1).

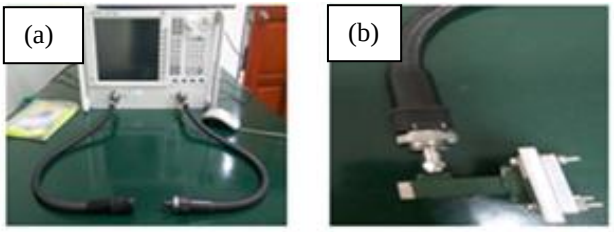


Fig. 1.Vector network analyzer (a), and (b) Sample holder with an adapter.

III. RESULT AND DISCUSSION

Fig. 2.indicates the XRD pattern of associated nanoparticles of barium hexaferrite; confirms the presence of well-coherent peaks. At 800 °C, secondary phase which is hematite (Fe_2O_3) was existed. The composition of a single phase $BaFe_{12}O_{19}$ without impurity started at the sintering temperatures of 900 and 1000 ° C. The Fe_2O_3 phase has been diminished as the sintering temperatures were increased.

Table- I: Crystallite size of sintered $BaFe_{12}O_{19}$.

Temperatures, (°C)	Crystallite size, *D* (nm)
800	42.6
900	50.9
1000	49.8

Table I shows the crystallite size of sintered $BaFe_{12}O_{19}$. Crystallite size (*D*) was calculated using the Scherrer Eq. 1[5]:

$$D = \frac{k\lambda}{\beta \cos\theta} \qquad (1)$$

The diffraction angle is larger for the sample sintered at 800 °C compared to those sintered at 900 and 1000 °C. This is reflected by the decrease in the crystallite size of the samples (Eq. 1). Fig. 4.shows the variation relation parameters of the *d*-spacing, the full width of half maximum (FWHM), the crystallite size and height intensity of the highest peak at (114). The *d*-spacing for sample sintered at 800 °C is larger compared to those sintered at 900 and 1000 °C. These shifts in the respective peak parameters show the rise in particle size as well as the increase in sintering temperatures. The X-ray patterns also reveal that the structural deformation of the samples increases as the temperature rises. This is obvious, the rising in height of the significant deforming peak at (114) (Fig. 3). The peak is heights between 1983 and 2248, with temperatures rising from 800 to 1000 °C. This indicates that the crystallinity of the samples rises with temperature increase. The similar trend also reported in previous reports [6]- [9]. The FWHM was decreased as the sintering temperature increased owing to the increase in the crystal size of the samples.

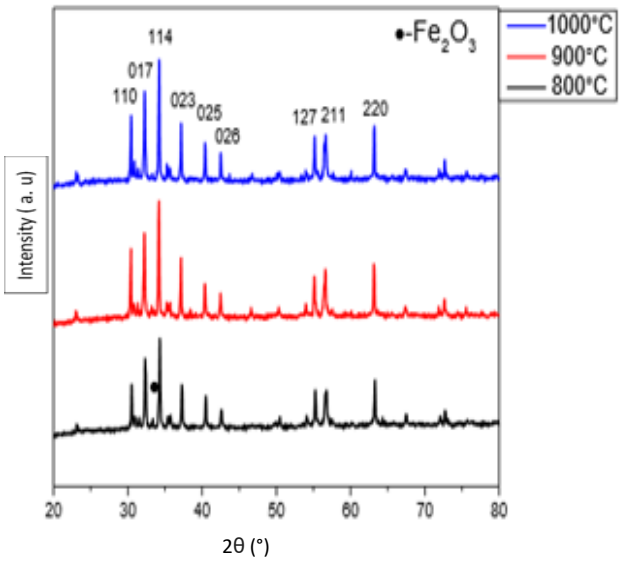


Fig. 2.X-ray diffraction pattern of $BaFe_{12}O_{19}$ at 800, 900 and 1000 °C.

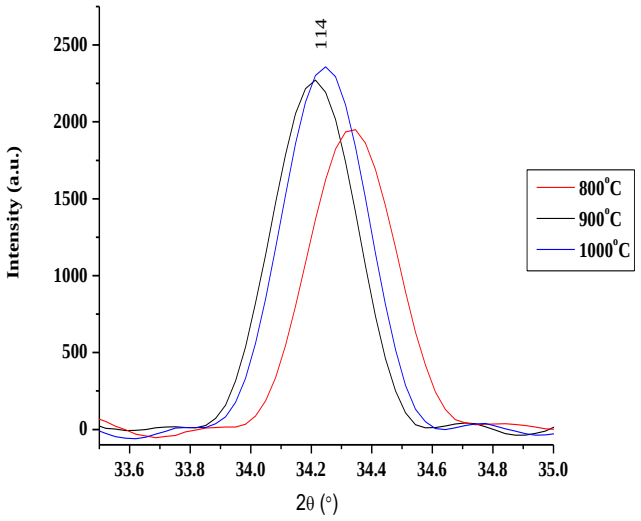
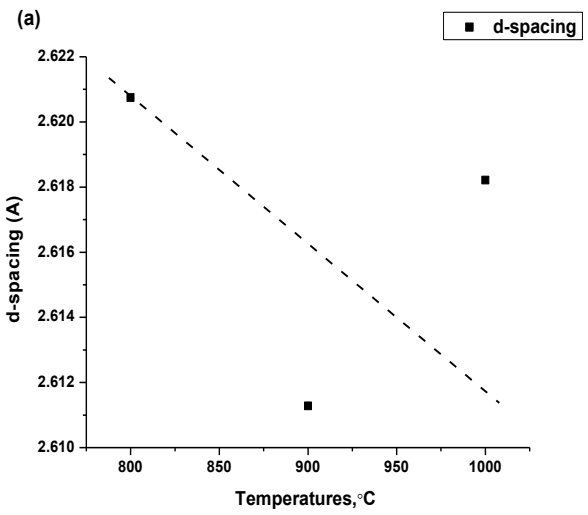
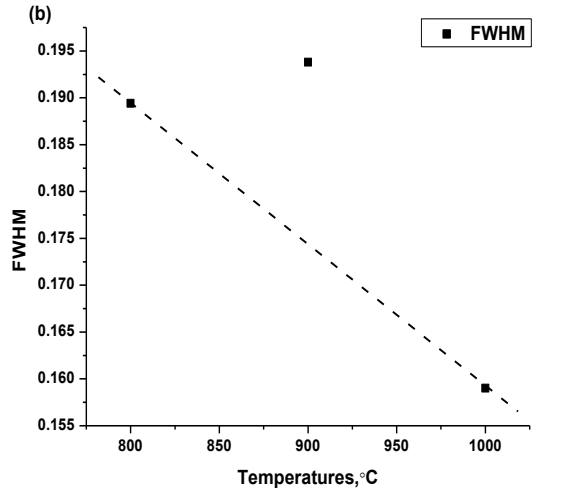
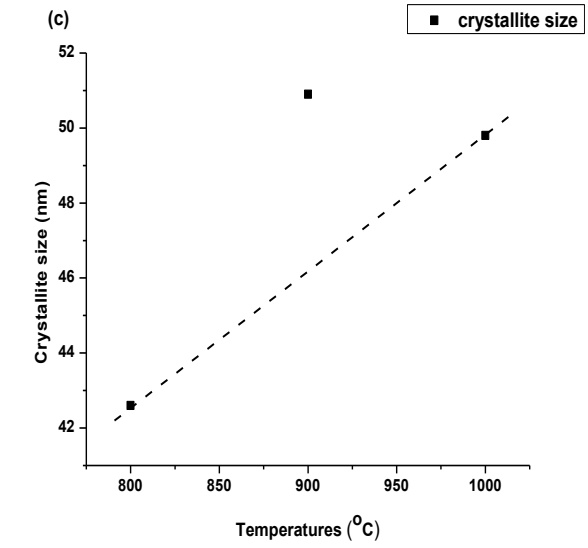


Fig. 3.XRD spectra at 2θ at the highest intensity (114).







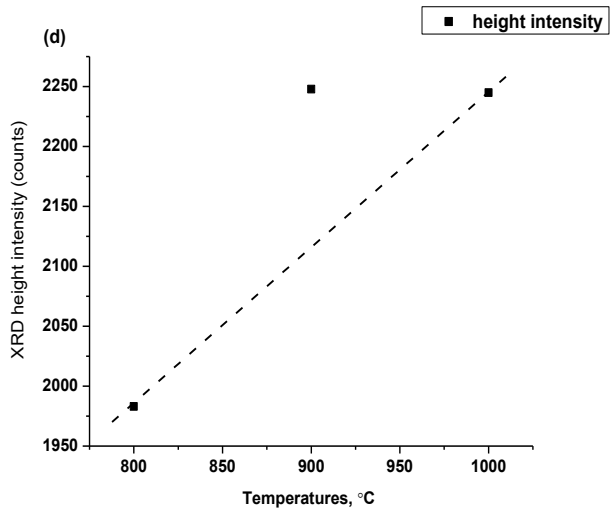


Fig. 4.Variation of *d*-spacing (a), full width of half maximum (FWHM) (b), the crystallite size (c), XRD height intensity of highest peak at (114) (d) of $BaFe_{12}O_{19}$ at various temperatures

Fig. 5.shows the morphology of $BaFe_{12}O_{19}$ nanoparticles powder sintered at 1000 °C, examined using FESEM. The average grain size observed is ~55 nm and is usually spherical in form and agglomerated owing to its magnetic nature.

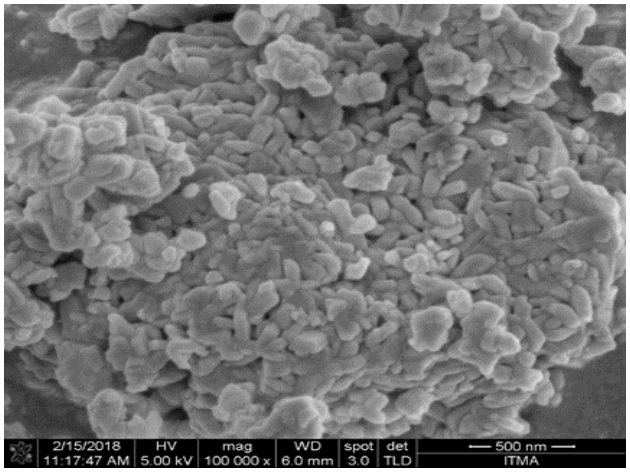


Fig. 5.FESEM image of $BaFe_{12}O_{19}$ at 1000 °C.

Fig. 6.indicates the minimum reflection loss, RL approximately −9.01 dB at a corresponding frequency of 9.16 GHz at 1000 ºC sintered sample (Table II) with a thickness of 3 mm. It is clearly shown that the RL was enhanced as the sintering temperature rises owing to improved stoichiometry and crystallinity [10]- [12]. The much higher absorption is expected to occur at a much higher frequency due to a higher anisotropy field of $BaFe_{12}O_{19}$. At X-band (8-12 GHz) frequency, the barium ferrite shows the dielectric and magnetic permeability properties with low attenuation ability [13].

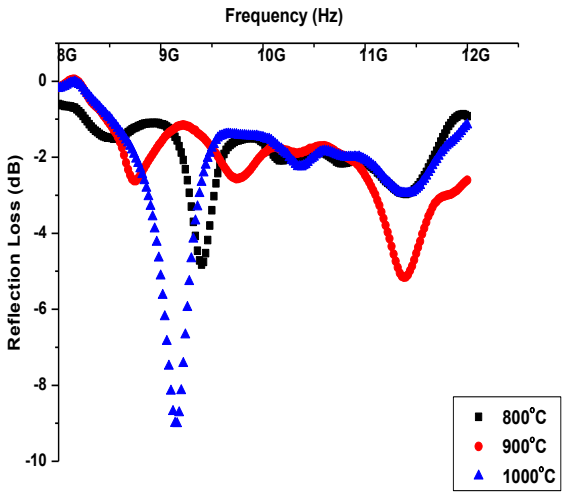
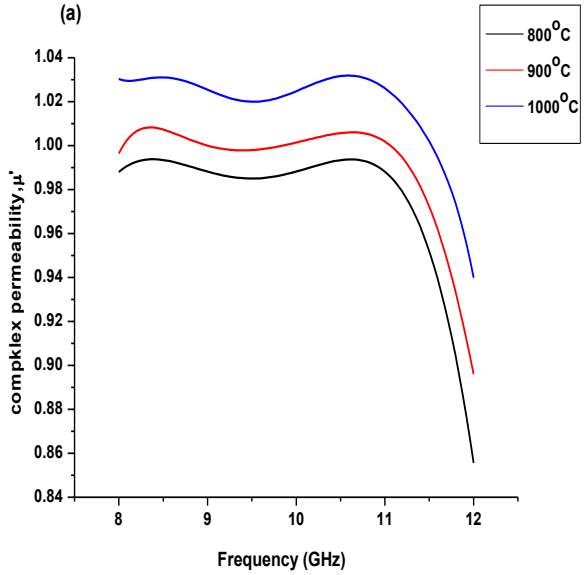


Fig. 6.Frequency dependency of reflection loss value of $BaFe_{12}O_{19}$ composite at 3 mm thickness with different sintering temperature.

Table- II: Electromagnetic wave absorption properties of $BaFe_{12}O_{19}$.

Temperatures (°C)	Peak value f_m (GHz)	Minimum *RL value* (dB)
800	9.40	−4.82
900	11.40	−5.15
1000	9.16	−9.01

Fig. 7.(a) and (b) showed the real and imaginary permeability (μ' and μ'') for 800, 900 and 1000 °C sintered samples at thickness 3 mm at 8-12 GHz respectively. There was shown the dependence of sintering to μ' and μ'' values in which the value of μ' of permeability for all temperature are varied as increase the sintering temperatures. As for μ", losses throughout the measured frequency stayed considerably continuous. The resonance peaks, however, happened between 8.5-11.5 GHz ranges. The variation of permeability with respect to the frequency can be attributed to the rotation of spins or the displacement of the domain wall. Large μ" are expected as a microwave absorber that allows for excellent absorption of incident radiation. Highlights of the microwave behavior of magnetic materials are precession, resonance and relaxation.



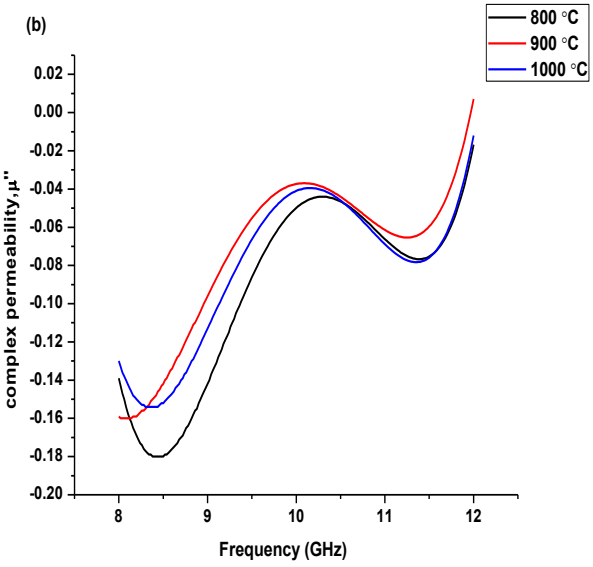


Fig. 7.Real (a) and imaginary (b) permeability of $BaFe_{12}O_{19}$/epoxy composites.

IV. CONCLUSION

In summary, $BaFe_{12}O_{19}$ was effectively prepared using a method of high-energy ball milling followed by sintering at different temperatures of 800, 900 and 1000 °C. The XRD analysis revealed the crystal structure of the hexagonal M-type phase, $BaFe_{12}O_{19}$, with the space group $P6_3/mmc$. The reflection loss peak showed the characteristic of maximum energy absorption of more than 50% (−9.01 dB) at a frequency 9.16 GHz when the sample thickness was 3 mm at a temperature of 1000 °C.

ACKNOWLEDGMENT

The authors are thankful for grants from Universiti Putra Malaysia (UPM), Inisiatif Putra Berkumpulan (IPB) GPB/2017/9541600, GP/2018/ 9628400 and IPS Putra research grant, UPM/ 800-3/3/1/(GP– IPS/2017/9533300, GP-IPS/2017/9539100. The authors also thanks to the Fundamental Research Grant Scheme (FRGS) and Long-term Research Grant Scheme [LRGS/ B-U/ 2013/ UPNM/ Defence & Security-P2], Malaysian Ministry of Higher Education (MOHE).

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