Microwave absorbing properties of Nd$_2$Fe$_{14}$B/αFe nanocomposite magnet

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Abstract. We have investigated microwave absorbing properties of Nd$_2$Fe$_{14}$B/αFe nanocomposite prepared by melt spinning method. Structures, magnetic characteristics and microwave absorbing properties were characterized using XRD, PERMAGRAPH and Vector Network Analyzer (VNA), respectively. The complex permeability and permittivity, as well as reflection loss (RL) values of the Nd$_2$Fe$_{14}$B/αFe nanocomposites were calculated in the range of 8.2–12.4 GHz. Magnetic hysteresis loop of Nd$_2$Fe$_{14}$B/αFe nanocomposite with a weight ratio of 52.2% of Nd$_2$Fe$_{14}$B and 47.8% of αFe resulted the magnetic saturation (Ms) 0.8 Tesla and coercivity (Hc) 300 kA/m. The RL value of Nd$_2$Fe$_{14}$B/αFe is less than -10 dB and tends to decrease up to -20 dB at 11.5 GHz. As a consequence, it has absorbed microwave minimum 70% in 8.2 - 12.4 GHz.

1. Introduction
Nowadays, the development of the use of communication and electronic equipment continues to increase [1]. It is also followed by the use of high frequencies of electromagnetic waves on the gigahertz scale. Instead of the advantages, the application in high frequency tools also causes many problems, for example the occurrence of wave interference, electromagnetic irradiation and electromagnetic (EM) pollution [2, 3]. The basic requirement for electromagnetic absorption materials are lightweight, thin, wide wave band and have strong absorptions ability [3–5]. To overcome the problems, various kinds of microwave absorption (MA) materials have been synthesized such as carbon black (CB), nano size silicon carbide (SiC) [6], conductive polymers [7], and magnetic materials [8–11]. Among of them, Nd$_2$Fe$_{14}$B/αFe nanocomposite as a hard and soft magnetic phase and a permanent magnet with high saturation (Ms), coercive force (Hc) and remanent (Mr) magnetization is considered to be a promising material for electromagnetic wave absorber [12–17]. Many efforts to modify composition, structure, and method of manufacture of the Nd$_2$Fe$_{14}$B/αFe nanocomposite to obtain optimum magnetic properties have been reported [18–20]. To improve the value of remanent and high magnetic saturation, Nd$_2$Fe$_{14}$B/αFe as nano-composite has been developed using Nd$_2$Fe$_{14}$B as hard magnets and αFe as soft magnets [21–23]. Due to the uniqueness of magnetic
properties (higher \(M_S\), larger \(H_C\) and \(M_I\)) of the conventional ferrites, large resistivity, and high permeability value, so \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nano-composite is a potential candidate as a microwave absorbing material in the high frequency range [17].

In this paper, we have studied a crystal structure and magnetic properties of the \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nano-composite which was produced by melt spinning method. The microwave absorbing properties of \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nano-composite were investigated in the 8–13 GHz ranges.

2. Experimental method

\(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nano-composite was prepared in the form of small ribbons by melt spinning method from Magnet Quench Ltd. A fine composite magnetic powder was obtained by using a mortar in toluene to avoid oxidation. Then, a powder was characterized using x-ray diffraction (XRD) (Phillips Co-K\(\alpha\) with 40 kV and 30 mA) to analyze crystal structure. The magnetic properties of \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nano-composite were obtained from the measurements on the maximum external field 1 Tesla with PERMAGRAPH type Electromagnet EP 3 from magnet physik Dr. Steingroever GmbH. \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nanocomposite magnet powder mixed with polyvinyl alcohol as adhesive is prepared in a 5 mm thick sample holder and then measured using vector network analyzer (VNA) Rohde-Schwarz ZVL13 to analyze microwave absorbing properties in 8–13 GHz frequency range. The values of complex permeability (\(\mu\)) and permittivity (\(\varepsilon\)) were determined from the reflected signal data S11 and transmission (S21) which calculated using Nicholson Ross Weir (NRW) formula [24] and then the reflection loss (RL) value of \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nanocomposite can be obtained based on the measured complex permittivity and permeability using the transmit line theory [25–27].

3. Results and discussion

3.1. Crystal structure

The powder was characterized using XRD with 2\(\theta\) interval 20°-105° for analyzing its structure. Figure 1(a) shows XRD patterns of \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) magnet powder after matching results with the Powder Diffraction File (PDF) reference patterns number 00-039-0473. Based on the qualitative analysis of the observed peaks, the XRD pattern indicated that the magnetic bands have two phases of \(\text{Nd}_3\text{Fe}_{14}\text{B} \) and \(\alpha\text{Fe} \). Each phase has formed a polycrystalline of magnetic composite with a weight ratio of 52.2% of \(\text{Nd}_3\text{Fe}_{14}\text{B} \) and \(\alpha\text{Fe} \) of 47.8% by weight. This indicates that the composite magnetic material synthesized by the melt spinning method has formed a crystalline state. FWHM and crystallites size from the XRD data are shown in table 1. By using Scherrer formula [8], the crystallite size was measured and shown in figure 1(b). The crystallites size of \(\text{Nd}_3\text{Fe}_{14}\text{B} \) phase in the range of 30–40 nm and \(\alpha\text{Fe} \) phase at 40–50 nm. This result confirmed that \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) ribbon is a magnetic nano-composite. This sample also indicated the change of lattice parameter values from the standard which refer to PDF number 00-039-0473 (\(a = b = 8.8020\) Å and \(c = 12.1790\) Å) to \(a = b = 8.82139\) Å and \(c = 12.19574\) Å for \(\text{Nd}_3\text{Fe}_{14}\text{B} \) ribbon and \(a = b = c = 2.86842\) Å for \(\alpha\text{Fe} \) phase (ICSD ref no. 98-018-0969, \(a = b = c = 2.8690\) Å).

![Figure 1](image-url)  

Figure 1. (a) XRD pattern and (b) distribution of crystallite size of \(\text{Nd}_3\text{Fe}_{14}\text{B}/\alpha\text{Fe} \) nanocomposite magnet.
Table 1. Peak position and crystallite size of Nd$_2$Fe$_{14}$B/$\alpha$Fe nanocomposite magnet.

| Position $2\theta$ (deg) | FWHM $2\theta$ (deg) | h  | k  | l  | Crystallite Size (nm) | Phase       |
|-------------------------|---------------------|----|----|----|-----------------------|-------------|
| 38.4793                 | 0.2714              | 1  | 3  | 1  | 33.51                 | Nd$_2$Fe$_{14}$B |
| 43.5989                 | 0.2754              | 1  | 2  | 4  | 35.42                 | Nd$_2$Fe$_{14}$B |
| 44.7982                 | 0.2764              | 0  | 1  | 5  | 35.39                 | Nd$_2$Fe$_{14}$B |
| 45.8148                 | 0.2773              | 1  | 3  | 3  | 37.02                 | Nd$_2$Fe$_{14}$B |
| 48.5154                 | 0.2798              | 2  | 2  | 4  | 36.29                 | Nd$_2$Fe$_{14}$B |
| 49.4984                 | 0.2807              | 1  | 4  | 0  | 35.04                 | Nd$_2$Fe$_{14}$B |
| 50.3051                 | 0.2815              | 1  | 4  | 1  | 37.35                 | Nd$_2$Fe$_{14}$B |
| 51.0322                 | 0.2823              | 3  | 3  | 0  | 36.43                 | Nd$_2$Fe$_{14}$B |
| 51.5928                 | 0.2829              | 1  | 3  | 4  | 39.32                 | Nd$_2$Fe$_{14}$B |
| 52.4078                 | 0.2285              | 0  | 1  | 1  | 48.61                 | $\alpha$Fe   |
| 77.2428                 | 0.2562              | 0  | 0  | 2  | 53.36                 | $\alpha$Fe   |
| 99.9777                 | 0.3036              | 1  | 1  | 2  | 52.17                 | $\alpha$Fe   |

3.2. Magnetic properties

Figure 2 shows a magnetic hysteresis loop of Nd$_2$Fe$_{14}$B/$\alpha$Fe nanocomposite with the magnetic saturation ($M_S$) and coercivity ($H_C$) of 0.8 tesla and 300 kA/m, respectively. However, the $H_C$ value confirmed the sample as hard magnet eventhough it is lower than $H_C$ of Nd$_2$Fe$_{14}$B nanocrystalline, where the $M_S$ value is greater than 1 tesla thus increasing the remanent magnetization ($M_r$) value. The decrease of the values of saturation magnetization ($M_S$) and coercivity ($H_C$) can be caused by grinding process. Ribbon sizes after grinding and non-solid samples also reduced magnetic saturation ($M_S$) and remanent ($M_r$) values. On the other hand, a fine loop shape also shows that there has been an interaction between the hard magnet of Nd$_2$Fe$_{14}$B and the soft magnet $\alpha$Fe and that also caused in the decrease of the coercivity ($H_C$) value. It can be explained by the exchange interaction between hard and soft magnets [15, 21].

3.3. Microwave absorbing properties

Based on the measured signal of $S_{11}$ and $S_{21}$ parameters of Nd$_2$Fe$_{14}$B/$\alpha$Fe nanocomposite magnet, the complex permeability ($\mu$) and permittivity ($\varepsilon$) were calculated by using NRW formula. As we can see in figure 3, the real ($\mu'$) and imaginary part ($\mu''$) of the permeability tend to decrease in the range of 8–13 GHz. Similar conditions is also observed in the real ($\varepsilon'$) and imaginary part ($\varepsilon''$) of the permittivity. Meanwhile, the complex relative permittivity tends to constant.
Figure 3. (a) Complex relative permeability and (b) permittivity of Nd$_2$Fe$_{14}$B/αFe nanocomposite magnet.

Figure 4 shows the results reflection loss (RL) of Nd$_2$Fe$_{14}$B/αFe nanocomposite magnet at 8–13 GHz in frequencies range is calculated based on the measured complex permeability ($\mu = \mu' - j\mu''$) and permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) (figure 3(a) and 3(b)) according to the transmittance line theory, which can be described by the following equations:

\[
RL \ (dB) = -20 \log \left| \frac{(\varepsilon - 1)}{(\varepsilon + 1)} \right|
\]  

\[
Z = \sqrt{\frac{\mu}{\varepsilon}} \tanh \left[ \left(-j2\pi f d / c\right)\sqrt{\mu / \varepsilon} \right]
\]

where $\mu = \mu' - j\mu''$ and $\varepsilon = \varepsilon' - j\varepsilon''$, $f$ is frequency and $d$ is the thickness of the sample.

The RL of Nd$_2$Fe$_{14}$B/αFe nano-composite magnet in the range of 8–13 GHz is less than -10 dB and tends to decrease to -20 dB at 11.5 GHz. However, Nd$_2$Fe$_{14}$B/αFe nano-composite magnet has absorbed minimum 70% in 8 – 13 GHz.

Figure 4. Absorption characteristics of Nd$_2$Fe$_{14}$B/αFe nanocomposite magnet.

4. Conclusions
Nd$_2$Fe$_{14}$B/αFe nanocomposite magnet ribbon that was prepared by melt spinning method has two phases and polycrystalline form. Their crystallite sizes are in the order of nanometer resulted a nanocomposite magnet with a weight ratio of 52.2% Nd$_2$Fe$_{14}$B and αFe 47.8%. Magnetic hysteresis loop of Nd$_2$Fe$_{14}$B/αFe nanocomposite resulted from the saturation magnetization ($M_s$) 0.8 Tesla and coercivity ($H_c$) 300 kA/m. The complex permeability tends to decrease in range 8–13 GHz and the
complex permittivity tends to be constant. The reflection loss values (RL) of Nd$_2$Fe$_{14}$B/$\alpha$Fe nanocomposite magnet is less than -10 dB and tends to decrease to -20 dB at 11.5 GHz.

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