

MAGNETIC AND MICROWAVE ABSORPTION PROPERTIES OF NEODYMIUM DOPED NICKEL FERRITE USING MILLING TECHNIQUE

Yunasfi*, Mashadi, Ade Mulyawan

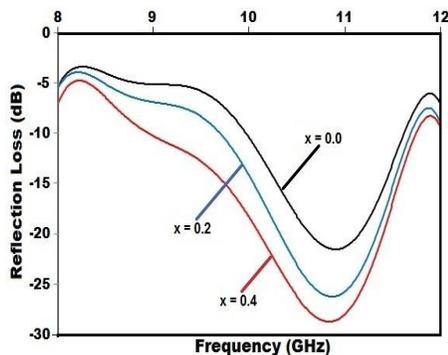
Center for Science and Technology of Advanced Materials
National Nuclear Energy Agency of Indonesia (BATAN), Kawasan
PUSPITEK Serpong, Tangerang – 15314

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*Corresponding author
yunaz@batan.go.id

Graphical abstract



Abstract

Nickel ferrite doped by neodymium in the form of $(\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4)$ with ($x = 0.0 ; 0.2$ and 0.4) have been synthesized using solid state reaction method with milling technique from NiO , Nd_2O_3 and Fe_2O_3 powder. The mixture of those compound materials was milled using High Energy Milling (HEM) machine for 10 hours and then sintered at 1000°C for 5 h. X-ray diffraction patterns showed that a single phase of spinel ferrite has been formed in all of the compositions. The result of morphological observation using Scanning Electron Microscope (SEM) exhibited a homogeneous structure has been formed with particle size about 200 nm. The magnetic measurement using vibrating sample magnetometer (VSM) showed that the sample exhibited a ferromagnetic behavior, where the M_s value decrease (around of 58.4 to 39.40 emu/g) and value of H_c increased (around of 116-170 Oe) along with the addition of the Nd^{3+} ion (x values) content. While the ability of microwaves absorption measured by using Vector Network Analyzer (VNA) indicates that the maximum value of Reflection Loss (RL) obtained at the composition of $x = 0.4$ up to -29 dB at a frequency of 10.81 GHz. It means the $\text{Ni}_{0.6}\text{Nd}_{0.4}\text{Fe}_2\text{O}_4$ sample can absorb microwave up to $\sim 96.5\%$ at a frequency of 10.81 GHz.

Keywords: Nickel ferrite, neodymium, milling technique, microwave absorption, magnetic properties

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1.0 INTRODUCTION

Spinel ferrite-type material with the chemical formula $M\text{Fe}_2\text{O}_4$ ($M = \text{Mn}, \text{Co}, \text{Ni}, \text{Zn}, \text{Mg}, \text{etc}$) with space group $Fd3m$ are known to have excellent magnetic, dielectric, catalytic, and optical properties. Every unit cell of each spinel ferrite are consisted of 56 atom in which 32 oxygen anions are distributed in the closed-cubic structure and 24 cation are located in both position of tetrahedral site (A site) and octahedral site (B site) [1]. Among all the spinel ferrite-type materials, nickel ferrite (NiFe_2O_4) is probably the most highly studied due to its unique physical properties [2].

NiFe_2O_4 nanoparticle exhibits ferrimagnetic behaviour which resulted from the coupling of the anti-parallel momen magnet between Fe^{3+} in the tetrahedral site and $\text{Ni}^{2+}/\text{Fe}^{3+}$ in the octahedral site [2-4]. NiFe_2O_4 is classified as soft magnetic material with low coercivity value but have a good electrical resistivity which makes this type very suitable to be applied in magnetic and magneto-optic application. Due to the a narrow hysteresis curve of NiFe_2O_4 , this material also applicable as a main compound of microwave absorbing material, low and high frequency transformer cores, high density information storage materials, etc. [5].

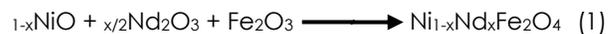
In order to enhance the magnetic and dielectric properties of NiFe_2O_4 , modification process can be done by substituting an appropriate amount of rare earth to the initial spinel structure [6]. In this work, the modification process was done by substituting Nd^{3+} cation into the NiFe_2O_4 structure. The Nd^{3+} cation will influence structural distortion of the initial structure which finally lead into the change of the dielectric transport properties [7]. According to the previous research which have been done by Pervaiz [8], NiFe_2O_4 which have been substituted by different divalent and trivalent cation will also perform different magnetic properties because of the exchange between the two sub-lattice in the modified spinel structure. This enhancement properties either in dielectric or magnetic properties are very important to develop microwave absorbing material which usually indicated by the permittivity (ϵ) and permeability (μ) value [9].

Several unique properties of NiFe_2O_4 are highly depend on their chemical composition and microstructure characteristic, in particular for microstructure characteristic such as the shape and size of the particle can be controlled through the fabrication process [10]. There are several fabrication process which commonly used to synthesize rare earth ions doped NiFe_2O_4 such as sol-gel method [11], co-precipitation method [12], and combustion method [13]. As far as the literature study by authors, there is no previous research which have tried to synthesize rare earth ions doped NiFe_2O_4 using solid state reaction method with milling technique to be applied microwave absorbing materials. Compared to other fabrication route to synthesize nanoparticle [14], milling technique have several advantages such it is much easier to be done and low cost production. In previous research, a single phase NiFe_2O_4 have successfully synthesized by using milling technique using milling technique [15]. In this research, the research is continued to substitute Nd^{3+} cation into the initial structure of NiFe_2O_4 in the form of $(\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4)$ with ($x = 0.0 ; 0.2$ and 0.4) with sintering process at 1000°C for 5 hours. The present of Nd is expected to able to alignment on the grain boundaries, so can limit grain growth however can induct lattice strain that result changing crystallite size and lattice parameter. In addition that the present of Nd can increase intrinsic properties of material as microwave absorbing material. The purpose of this research is to know the effect of the rare earth ion substitution to the magnetic properties and their microwave absorbing properties.

2.0 METHODOLOGY

The following chemical powders were used as a main compound to synthesize of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ ($x = 0; 0.2$ and 0.4) : Fe_2O_3 (Sigma Aldrich, 99% purity), NiO (Sigma Aldrich, 76-77% purity), and Nd_2O_3 (Sigma Aldrich, 99.9% purity). Each composition was synthesized in the

total mass 10 gram with the expected chemical reaction as below;



The process of milling technique was started by pouring all of the chemical powders with the addition of ethanol in stainless vial. All of the compositions were milled for 10 hours which referred to the optimum of the milling time in synthesis of $\text{NiFe}_2\text{O}_4 - \text{NdFe}_2\text{O}_3$ composite [16]. All samples then dried in the oven at the temperature 120°C for 5 hours. The fabrication process was continued to the sintering process at the temperature 1000°C for 5 hours. After sintering process, each composition was grinded by using Agate mortar until fine powder was formed.

X-ray Diffractometer (XRD), Phillips Panalytical PW1710, was used to identify the phase formation in each composition. The morphological observations were carried out by Scanning Electron Microscope (SEM) using HITACHI type SU3500. Magnetic properties of all composition were collected in room temperature by Vibrating Sample Magnetometer (VSM) using OXFORD type with 1 Tesla magnetization. Vector Network Analyzer (VNA) Advatest-R3370 was used to measure Reflection Loss (RL) value from the initial microwave which represents the ability of each composition to absorb the microwave.

3.0 RESULTS AND DISCUSSION

The X-ray diffraction patterns of all $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ ($x = 0; 0.2$ and 0.4) compositions are shown in Figure 1. Referring to the Crystallographic Open Database (COD) number 96-100-6117, all of the compositions exhibit a single phase form of spinel ferrite which has a cubic structure ($a = b = c = 0.58836$ nm) and space group Fd3m. The most intense peak in the position of 2θ around 35° is a main characteristic for all spinel ferrite-type material which resemble to the crystal plane of (311). The single formation of spinel ferrite in all compositions is also confirmed by the presence of several diffraction patterns in the position around $18^\circ, 30^\circ, 37^\circ, 43^\circ, 53^\circ, 57^\circ, 63^\circ$, which can be indexed to the crystal plane of (111), (220), (222), (400), (422), (511), and (440) respectively. In this study, there is no additional phase along with the increasing substitution of Nd^{3+} to the initial structure of NiFe_2O_4 . The results of the phase identification for all $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ composition were similar with Joshi *et al.* [17] who studied the synthesis of nickel ferrite by co-precipitation method.

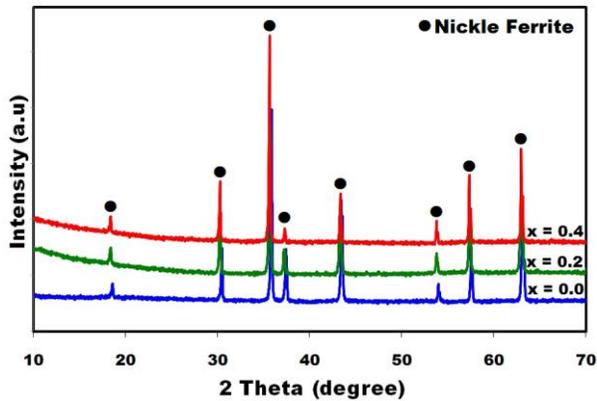


Figure 1 X-ray diffraction pattern of all $Ni_{1-x}Nd_xFe_2O_4$ compositions with ($x = 0.0; 0.2$ and 0.4) after milling for 10 h

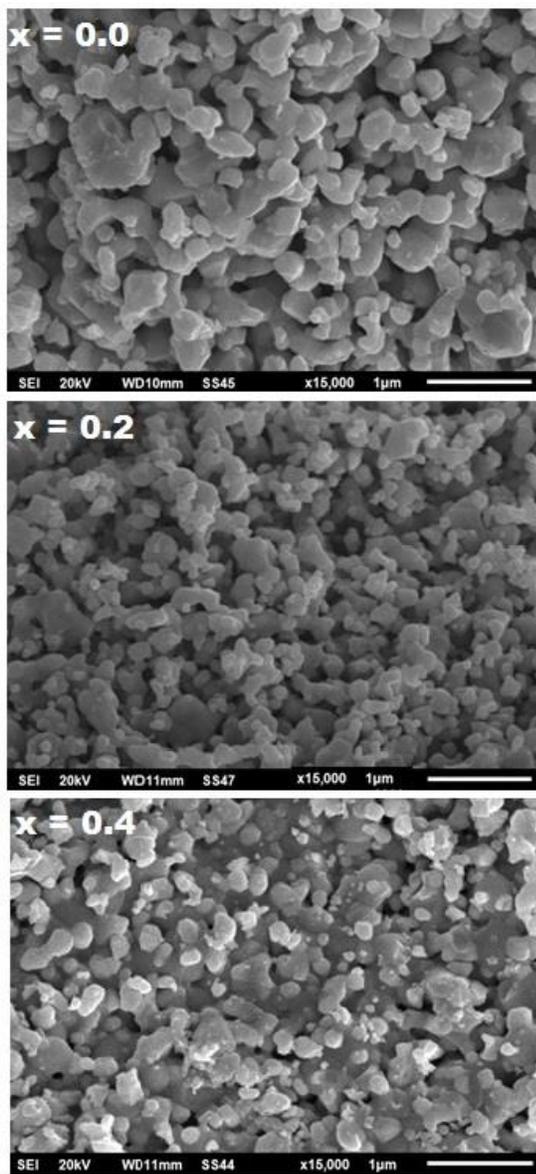


Figure 2 Morphological structure of all $Ni_{1-x}Nd_xFe_2O_4$ compositions with ($x = 0.0; 0.2$ and 0.4) compositions with 15.000 times magnification

The morphological structures of all $Ni_{1-x}Nd_xFe_2O_4$ compositions with 15.000 times magnification are shown in Figure 2. This figure revealed that the particles of all $Ni_{1-x}Nd_xFe_2O_4$ compositions are distributed homogenously with low degree agglomeration in the surface area with particle size around 200 nm.

The magnetic properties of all $Ni_{1-x}Nd_xFe_2O_4$ compositions with ($x = 0.0; 0.2$ and 0.4) were collected by using Vibrating Sample Magnetometer (VSM) with 1 Tesla magnetization are shown in Figure 3.

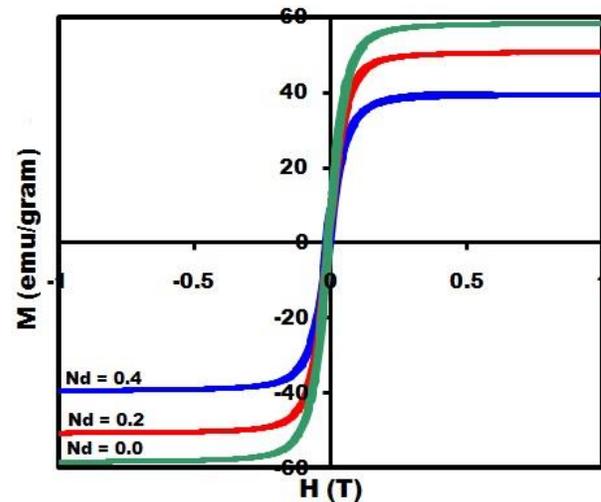


Figure 3 Hysteresis curve of $Ni_{1-x}Nd_xFe_2O_4$ sampel with $x = 0.0; 0.2$ and 0.4) after milling for 10 h

The results of the magnetic properties and detailed extracted value such as magnetization saturation (M_s), magnetization remanence (M_r), and coercivity value (H_c) of all compositions are shown in Figure 3 and Table 1. Referring to shape of the M-H hysteresis curve in Figure 3, all of the compositions exhibit ferromagnetic behavior with low coercivity value. The low coercivity value can be known by converting the external magnetization scale from Tesla became Oersted (Oe). According to the extracted results from M-H hysteresis curve in Table 1, it can be noticed that as the substitution of Nd^{3+} cation made the magnetization saturation was gradually decreased but the the coercivity value was increased. Similar result was reported by Sabikoglu *et al.* [2015] who studied the effect Nd^{3+} substituted nickel ferrite [18] and Rathod *et al.* [2015] for La^{3+} substituted Ni nickel ferrite [19]. This results showed that the increasing substitution of Nd^{3+} affected the magnetic moments in the initial structure became harder to be magnetized by external magnetic field and tend to have a bigger coercivity value in demagnetization process

Table 1 Magnetic properties data of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ with ($x = 0.0; 0.2$ and 0.4) after milling for 10 h

| Composition (x) | Sample | Magnetic parameter | | |
|-----------------|---|--------------------|------------|---------|
| | | Ms (emu/g) | Mr (emu/g) | Hc (Oe) |
| 0.0 | $\text{Ni Fe}_2\text{O}_4$ | 58.4 | 13.6 | 116 |
| 0.2 | $\text{Ni}_{0.8}\text{Nd}_{0.2}\text{Fe}_2\text{O}_4$ | 50.7 | 11.8 | 123 |
| 0.4 | $\text{Ni}_{0.6}\text{Nd}_{0.4}\text{Fe}_2\text{O}_4$ | 39.4 | 10.5 | 170 |

The results of the microwave absorbing properties of all $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ compositions ($x = 0.0; 0.2$ and 0.4) are shown in the Reflection Loss (RL) curve in Figure 4. The results were carried out by using Vector Network Area (VNA) in the frequency range 8-12 GHz as the function of frequency and RL value. The reflection loss phenomenon shows the magnetic spin resonance mechanism between the initial electromagnetic wave or microwave with the magnetic spin of the material [21]. As shown in Figure 4, each composition of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ ($x = 0.0; 0.2$ and 0.4) shows 3 different RL peak positions which indicated that this composition is able to absorb electromagnetic wave in a broad frequency position. Increased reflection losses associated with the value of coercivity (Hc) of material. The greater the value of Hc the absorption of microwaves is also getting bigger. The composition with the Nd^{3+} substitution of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ ($x=0.4$) showed the deepest RL value among all compositions with the reflection loss value of -29 dB which mean the material is able to absorb about 96.5% from the initial electromagnetic wave in the frequency of 10.81 GHz.

Referring to the Figure 4, the results of the microwave absorbing properties of Nd doped Nickel Ferrite in the form of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ ($x=0.2$ and 0.4) exhibited a better performance to be applied as Radar absorbing materials compared to the other research which have been done by Li *et al.*, [2014] who studied spinel ferrite as microwave absorbing material [21].

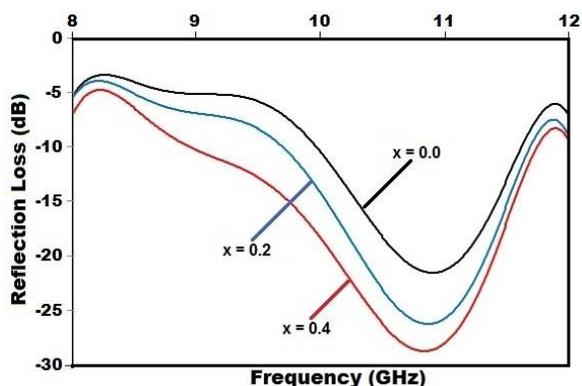


Figure 4 Reflection loss curva of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ sample with ($x = 0.0; 0.2$ and 0.4) after milling for 10 h

Electromagnetic wave is consisted of two components which are magnetic field and electrical field. This components make the initial electromagnetic wave can interact with magnetic materials and finally lead into the emergence of electromagnetic or microwave absorbing phenomenon. According to the transmission line theory, the ability of magnetic materials to absorb the electromagnetic is highly depend on the impedance matching between the material and the electromagnetic wave through resonance frequency mechanism. This impedance matching is also highly influenced by the permittivity (ϵ_r) and permeability (μ_r) value of the magnetic material. A magnetic material as the candidate of electromagnetic absorbing material is highly required to have an easily moving magnetic spin so that the resonance of the electromagnetic wave could be maintained. It means if the magnetic material exhibited hard magnetic properties which has a large anisotropy field then it should be minimized. Whereas if the magnetic material is a soft magnetic that has a very small anisotropy field then it should be increased.

4.0 CONCLUSION

The synthesis of Nickel ferrite doped by neodymium as a candidate of electromagnetic absorbing material through solid state reaction method by using milling technique have been conducted by resulting a single phase of nickel ferrite in all compositions. According to the hysteresis curve as the results of the magnetic properties measurement, it is known that all compositions exhibit ferromagnetic behavior. The increasing substitution of Nd^{3+} causes a decrease in the magnetization saturation but increases the coercivity value. The deepest RL value was reached in the composition of $\text{Ni}_{1-x}\text{Nd}_x\text{Fe}_2\text{O}_4$ ($x=0.4$) with the reflection loss value of -29 dB which mean the material is able to absorb about 96.5% from the initial electromagnetic wave in the frequency of 10.81 GHz.

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