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MICROWAVE ABSORBING PROPERTIES OF $La_{0.67}Ba_{0.33}Mn_{1-X}Ni_XO_3$

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ABSTRACT

MICROWAVE ABSORBING PROPERTIES OF $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$. The doped lanthanum manganites have unusual magnetic and transport properties, which makes it possible for this material to be used for microwave absorbing. In this study, $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04, and 0.06) were prepared by solid state reaction method as microwave absorption material. The crystal structure and magnetic properties were characterized by X-Ray Diffractometer (XRD) and Vibrating Sample Magnetometer (VSM), respectively. Refinement results of X-Ray diffraction pattern using High Score Plus Software showed that the samples with various x = 0 to x = 0.04 had a single phase with monoclinic crystal structure, while the sample with x = 0.06 had two phases with monoclinic and hexagonal structures. Hysteresis loops showed that the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04, and 0.06) samples are soft magnetic materials. Microwave absorption properties were investigated in the frequency range of 8-12 GHz using Vector Network Analyzer (VNA). An optimal reflection loss of -8.85 dB is reached at 11.58 GHz for x = 0.04 with sampel thickness of 2 mm.

Keywords: Magnetic, Lanthanum, Manganites, Microwave absorbing, $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$

ABSTRAK

SIFAT PENYERAP GELOMBANG MIKRO DARI $La_{0,67}Ba_{0,33}Mn_{1-x}Ni_xO_3$. Bahan lantanum manganat memiliki sifat magnetik dan sifat transport yang unik, sehingga bahan ini kemungkinan dapat diaplikasikan sebagai bahan penyerap gelombang mikro. Pada penelitian ini bahan penyerap gelombang mikro $La_{0,67}Ba_{0,33}Mn_{1-x}Ni_xO_3$ (x = 0, 0,02, 0,04 dan 0,06) disintesis dengan metode reaksi padatan. Struktur kristal dan sifat magnet bahan dikarakterisasi dengan menggunakan *X-Ray Diffractometer* (*XRD*) dan *Vibrating Sample Magnetometer* (*VSM*). Hasil *XRD* kemudian diolah dengan perangkat lunak *High Score Plus*. Diperoleh hasil bahwa sampel $La_{0,67}Ba_{0,33}Mn_{1-x}Ni_xO_3$ untuk x = 0 sampai x = 0,04 adalah satu fasa dengan struktur kristal monoklinik, sedangkan untuk x = 0,06 terdapat dua fasa dengan struktur kristal monoklinik dan heksagonal. Sifat magnetik bahan $La_{0,67}Ba_{0,33}Mn_{1-x}Ni_xO_3$ dikarakterisasi dengan menggunakan VSM. Hasil kurva histeresis menunjukkan bahwa sampel $La_{0,67}Ba_{0,33}Mn_{1-x}Ni_xO_3$ termasuk bahan magnet lunak. Sifat penyerapan gelombang mikro diteliti dengan menggunakan VNA pada rentang frekuensi 8-12 GHz. Hasilnya harga *reflection loss* yang paling optimum diperoleh pada frekuensi 11,58 GHz sebesar -8,85 dB untuk sampel $La_{0,67}Ba_{0,33}Mn_{0,96}Ni_{0,04}O_3$ dengan ketebalan 2 mm

Kata kunci: Magnetik, Lantanum, Manganat, Penyerap gelombang mikro, $La_{0,67}Ba_{0,33}Mn_{1-x}Ni_xO_3$

INTRODUCTION

Microwave in the range of 1-20 GHz are increasingly used in the field of wireless communication, local area network, and so on. However, the ElectroMagnetic Interference (EMI) limits their applications and hence much attention has been paid to find suitable microwave absorption materials. Conventional spinel-type ferrites do not function well in the GHz range due to a drop in the complex permeability

μ_r as given by Snoek's limit [1-2]. In recent years, the doped rare earth-transition metal oxides, especially doped $LaMnO_3$, with their colossal magneto-resistance (CMR) effect, have shown high potential applications in the field of magnetic electronics functional materials. Besides the CMR effect, the particular electronic structure and unusual electromagnetic characteristics of the oxide indicate that it can be a potential candidate for microwave absorption materials at high frequency over gigahertz [3-6].

There have been a lot of researches on the electromagnetic and microwave absorption properties of doped $LaMnO_3$. For example, Zhou et al. [7] reported that $La_{0.8}Ba_{0.2}MnO_3$ prepared by traditional sol-gel method had excellent property. The microwave absorbing peak was 13 dB at 6.7 GHz for the sample with the thickness of 2.6 mm. Cheng et al. [8] prepared carbonyl iron/$La_{0.6}Sr_{0.4}MnO_3$ composites with the optimal reflection loss of -12.4 dB is reached at 10.5 GHz with the thickness of 0.8 mm.

Zhang et al. [9] also confirmed that the incorporation of transition metal (TM = Fe, Co, or Ni) could influence the electromagnetic properties of $La_{0.7}Sr_{0.3}MnO_{3\pm\delta}$. It is found that the electromagnetic loss has been enhanced after TM doping, and the microwave absorption properties have been significantly improved.

In this paper, with the aim finding a material suitable for uses as a microwave absorber in GHz band range, $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04 and 0.06) powders were prepared by the solid state reaction method. Our experimental results demon strate that $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ is promising for the application of broadband and effective micro-wave absorbers.

EXPERIMENTAL

$La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04, 0.06) ceramic samples were synthesized following the solid-state reaction route by mixing La_2O_3, $MnCO_3$, $BaCO_3$, and NiO (Sigma Aldrich 99.99% purity) in their stoichiometric ratios, with Planetary Ball Milling for 25 hours. Thermo gravimetric analyzer (TGA) TA Instruments was used to determine the crystallization temperature. The calcinations performed at 800 °C for 10 h. The calcined powders were then pressed into pellets and finally sintered at 1200 °C for 2 h.

The crystal structure of the samples were determined by X-ray Diffractometer (XRD) Shimadzu-7000 using CuKα radiation (λ = 1.54 Å) operated at 35 kV and 25 mA. The magnetic properties of the samples were observed by Vibrating Sample Magnetometer (VSM) Oxford VSM1.2H. The reflection loss (RL) of the samples was measured between 8 and 12 GHz using Vector Network Analyzer (VNA) Advantest type R3770.

RESULT AND DISCUSSION

Figure 1 presents the XRD patterns of the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04, 0.06) samples. Crystal structure was further refined by High Score Plus software from PANanalytical. Refinement results of the samples can be seen in Table 1. The refinement results provide information that $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples with x = 0-0.04 have formed a single phase with monoclinic crystal structure and space group I 1 2/c 1. For the sample with x = 0.06, there are two phases namely $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ phase with monoclinic crystal structure and hexagonal $BaMnO_3$ phase exist within the sample.

To study the magnetic properties of the samples, the magnetic hysteresis loops of the samples at room temperature were measured. It is shown that the samples are ferromagnetic at room temperature. Figure 2 shows the image of magnetic hysteresis loops, it exhibits the soft magnetic property of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04, 0.06) samples. It indicates that the total magnetization was decreased with increasing nickel doping level. This suggests that the decrease of magnetization with increasing Ni content is due to the weakening of exchange coupling between

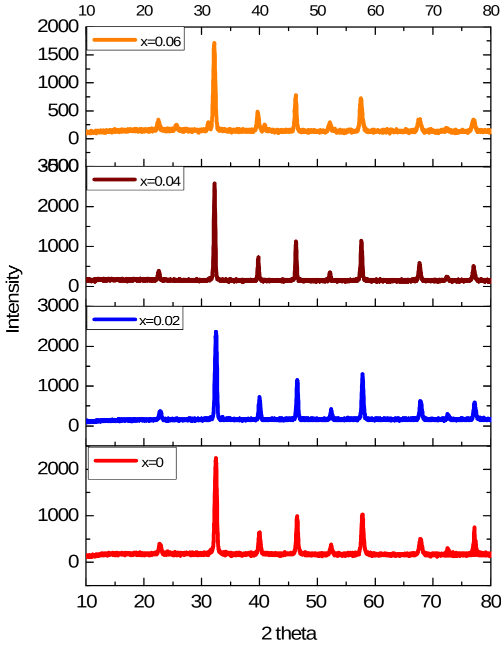

Figure 1. XRD pattern of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples

Tabel 1. Refinement results of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples

x	a (Å)	b(Å)	c(Å)	β	γ	χ2
0	9.56	5.52	5.55	125.28	90	1.17
0.02	9.57	5.52	5.54	125.28	90	1.15
0.04	9.55	5.51	5.53	125.28	90	1.24
0.06 (phase 1)	9.58	5.51	5.54	125.28	90	1.32
0.06 (phase 2)	5.69	5.69	4.82	90	120	

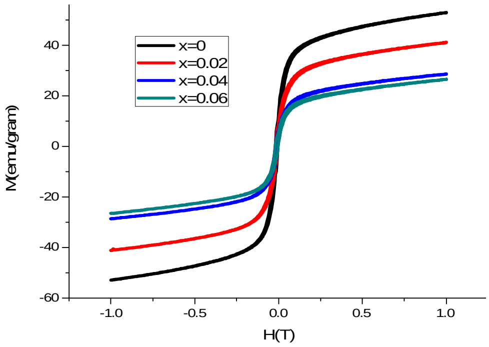

Figure 2. Magnetic Hysteresis loops of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples

Mn^{3+} - O - Mn^{4+} [10,11]. The total magnetization for x = 0.06 composition is the lowest among all samples. It could be explained from the fact that the sample is not single phase due to the presence of additional phases as reported from XRD. Thus, less magnetic phase in this sample.

Complex permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) and permeability ($\mu = \mu' - j\mu''$) in the range of 8-12 GHz were measured using vector network analyzer. They are commonly used to express the dielectric and magnetic response of the materials in an applied microwave field. The real parts of permittivity and permeability (ε' and μ') imply the capability to store microwave energy, and the imaginary parts measure the ability to dissipate the stored energy into heat. Figures 3 and 4 show the permittivity and permeability of the single phase samples, respectively.

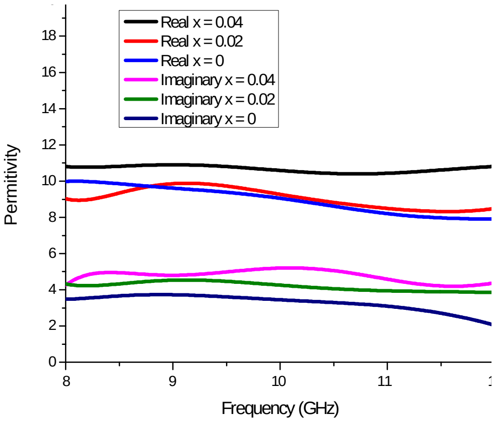

Figure 3. Permittivity of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples

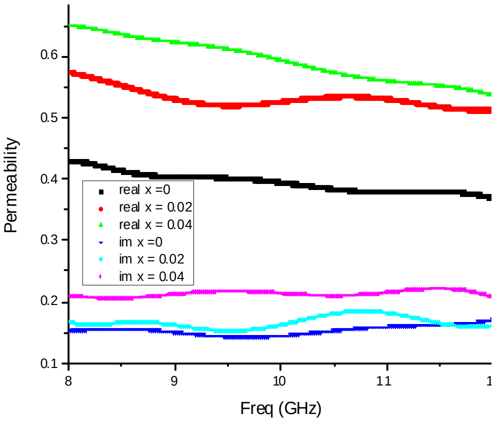

Figure 4. Permeability of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples

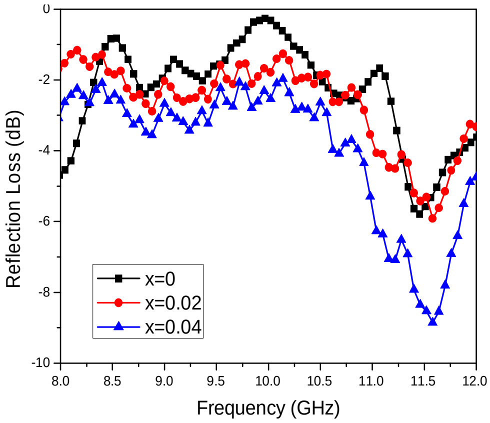

Figure 5. Microwave absorption performance of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$

Table 2. Microwave absorption properties of $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ samples

Sample	Frequency (GHz)	Reflection Loss (dB)
$La_{0,67}Ba_{0,33}MnO_3$	11.45	-5.79
$La_{0,67}Ba_{0,33}Mn_{0,98}Ni_{0,02}O_3$	11.58	-5.90
$La_{0,67}Ba_{0,33}Mn_{0,96}Ni_{0,04}O_3$	11.58	-8.85

Figure 3 shows the frequency dependence of real and imaginary parts of relative complex permittivity for the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, and 0.04) samples. In the range of 8-12 GHz $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0.02 and 0.04) samples had relatively larger ε' and ε'' than that of $La_{0.67}Ba_{0.33}MnO_3$. It indicates that the ability to store energy and the dielectric oss of $La_{0.67}Ba_{0.33}MnO_3$ has been enhanced in this waveband.

Figure 4 reveals the real (μ') and imaginary (μ'') parts of the relative complex permeability for $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, and 0.04) samples. Replacing permeability with magnetic susceptibility (χ) based on $\mu' = 1 + \chi$, it is clear that χ is negative, implying its anti-ferromagnetism. Meanwhile, it is clear that the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, and 0.04) samples possess weaker ferromagnetism (Figure 2).

Microwave absorption performance of the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02 and 0.04) samples were investigated with absorber's thickness of 2 mm. Figure 5 shows the reflection loss spectra of the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02 and 0.04) single phase samples in the frequency range of 8 – 12 GHz. Table 2 reveals the microwave absorption properties of the $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, and 0.04) samples. The $La_{0.67}Ba_{0.33}Mn_{0.96}Ni_{0.04}O_3$ sample had the best microwave absorption performance. The maximum reflection loss was -8.85 dB at matching frequency 11.58 GHz.

CONCLUSION

In summary, the transition metal doped $La_{0.67}Ba_{0.33}Mn_{1-x}Ni_xO_3$ (x = 0, 0.02, 0.04, and 0.06) powders have been fabricated successfully by the conventional solid state reaction method. The electromagnetic loss has been enhanced after Ni doping. The $La_{0.67}Ba_{0.33}Mn_{0.96}Ni_{0.04}O_3$ sample had the best microwave absorption performance. The maximum reflection loss was -8.85 dB at matching frequency 11.58 GHz.

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