

AN INVESTIGATION OF STRUCTURE AND COMPLEX IMPEDANCE BEHAVIOR OF COMPOSITE $(1-x)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / x\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$

V. Vekky R. Repi^{1, 2, a} and Azwar Manaf¹

¹ Postgraduate Program of Materials Science, Faculty of Mathematics and Natural Science, Universitas Indonesia Jalan Salemba Raya 4, Jakarta 10430, INDONESIA

² Department of Engineering Physics, Universitas Nasional, Jakarta 12520

^a vekky_repi@yahoo.com

Keywords : Composite material, complex impedance, barium hexaferrite, mechanical alloying, electrical charge transport, electromagnetic wave absorber.

ABSTRACT

We have investigated of composite $(1-x)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / x\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ by conventional ceramic method. The structure of composite and complex impedance are carried out by XRD, SEM and Impedance Spectroscopy. With changing the weight ratio from $x=0$; 0.3; 0.5 and $x=1$ the phase exhibit the composite behavior. Very interestingly, for the case of the case of weight ratio $x=0.5$ and 0.7 showed the complex impedance with xemicircle pattern in frequency range from 1 KHz to 1 MHz.

INTRODUCTION

Recently, the type ferrite composite materials have been greatly interesting since its have potential for electromagnetic wave absorption application such as magnetic sensor, wireless communication, and microwave absorber devices [1-3]. Understanding the ferrite composite materials is very essential for electromagnetic wave devices realization especially below the microwave range. Several investigation have been reported about the phenomenon intrinsic complex impedance below the microwave frequencies from 1 kHz to 1 MHz in ferrite and ferroelectric materials for sensor application [4,5]. The intrinsic complex impedance spectrum of polycrystalline ceramics is affected by the distribution of grain boundary [4]. The temperature has also changed the intrinsic complex impedance in solid-state reaction processing [6]. However, few studies have been addressed about intrinsic complex impedance in $(1-x)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / (x)\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ composite with various weight ratios below microwave regime.

The composite materials are prepared by conventional ceramics method. The structures of composite materials are investigated by X-ray diffraction analysis (XRD) and Scanning Electron Microscopy (SEM). We also carried out the impedance using Impedance spectroscopy. Very interestingly, the composite materials for the weight ratio of $x=0.5$ and 0.7 exhibit the semicircle pattern in complex impedance.

MATERIAL AND METHOD

The $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / \text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ samples ($x = 0 - 1$) were prepared as following steps. Firstly, the polycrystalline $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19}$ and $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ was prepared by conventional ceramic reaction method. The stoichiometric amount of BaCO_3 , SrCO_3 , MnCO_3 , TiO_2 , Fe_2O_3 , La_2O_3 with the purity is higher than 99% were ground completely to achieve the homogeneity of the mixed powder for 20 h with dry milling by using planetary ball milling and then sintered at 1100°C for 10 h. Secondly, the prepared $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19}$ and $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ was mixed for 10 h to put out homogeneity composite as to $(1-x)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / x\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$. Finally, the composite were ground to powder, pressed into pellet form and sintered at 1100°C for 4 h to get compact sample. Phase structure of the

composite material was analyzed using X-ray diffraction (XRD) Phillip PW 7310 with a source of Co-k α radiation at room temperature. Surface morphology of the samples studied using SEM JEOL JSM-5310 LV. The complex impedance of the samples was measured at room temperature using a HIOKI LCR Hi-Tester 3532-50 with a frequency range of 1 Hz to 1 MHz at ac electric signal 1 volt.

RESULTS AND DISCUSSION

XRD pattern of $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19}$ (A) and $\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ (B) composite at room temperature are shown in Fig. 1. Hexagonal and orthorhombic phase can be found in the XRD patterns. The patterns also reveal that sample A and B presents types of peaks corresponding to each other. With the observation of the XRD peaks, intensity of B phase gradually increases with increasing x , and no additional peaks of other phase are indicate.

To confirm the coexistence of both phases in the composites, the A / B samples with different composition of x were observed by SEM micrographs.

As seen in Figure 1, the phase changes to the morphology of the composite material can be proven from the SEM image in figure 2. The morphology of the surface of the material looks much grain particles with connections to a less robust (Fig. 2a, 2b, 2c). Nevertheless, from the observation seems consistent with XRD diffraction patterns.

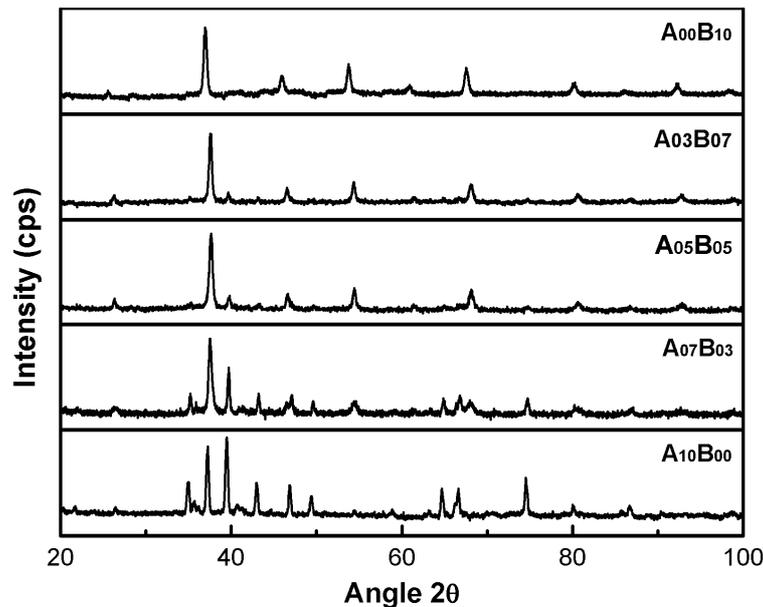


Figure 1: XRD pattern of composite material as synthesized and heat treated at 1100°C for 4 h.

Table 1: Summary of composite used in this study

Notation in this paper	Chemical formula (1-x)A / xB
$A_{10}B_{00}$	$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19}$
$A_{07}B_{03}$	$(0.7)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / (0.3)\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$
$A_{05}B_{05}$	$(0.5)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / (0.5)\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$
$A_{03}B_{07}$	$(0.3)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / (0.7)\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$
$A_{00}B_{10}$	$\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$

In Figure 2, the morphology of the samples shown for each composite materials. As seen in figure 2(a) is the surface morphology of barium hexaferrite material having a hexagonal crystal structure and space group P 63/mmc, while in Figure 2(b), (c), (d) are the surface morphology of composite materials with a variety of composite x . Figure 2(e) is the surface morphology of material LaMnO_3 which has a crystal structure orthorhombic with space group Pbnm.

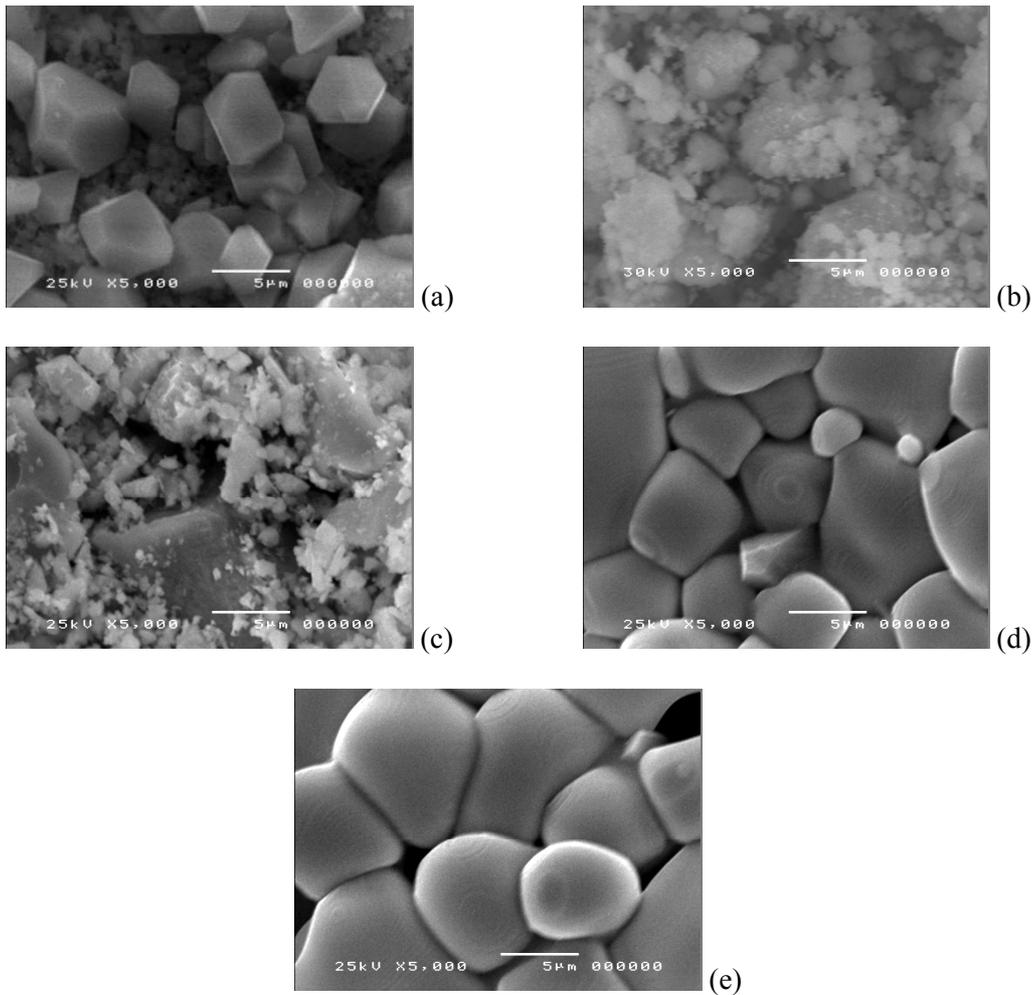


Figure 2: SEM Micrograph showing of $(1-x)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15}\text{O}_{19} / x(0.3)\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ composite. a) $x = 0$, b) $x = 0.3$, c) $x = 0.5$ d) $x = 0.7$, e) $x = 1$.

Figure 3 shows the cole-cole plot imaginary part of the complex impedance Z'' vs. real part of complex impedance Z' . By fitting data using semicircle, we can know grain boundary resistance from intercept of the fitted curve on the Z' axis at lower frequency and higher frequency side. The measurement of complex impedance which are to know the electrical charge transport is controlled by the highly conductivity grains and poor conducting grain boundary regions of the material [6]. In figure 3 (a), (b), (e) shows that there is no pattern semicircle, in the picture occurs grouping data into these two points due to absence of electrical effects on sample material causes electrical charge transport does not occur. This might be due to the insulating behavior of the PZT–NZFO ceramic composites [7]. In figure 3 (c) and (d) single semicircle pattern seen that the grain boundary resistivity on the composite composition $x = 0.5$ and $x = 0.7$.

The coincidence of the impedance Z values at higher frequencies at all the temperatures indicates a possible release of space charge. Real part of impedance with frequency is suitable for evaluation of the relaxation frequency of most resistive component. This exhibits the occurrence of relaxation in the system. The relaxation frequency is obtained either from the plot of Z versus frequency or semicircles (from the Nyquist plot). The peak broadening (due to increase in temperature) suggests the presence of temperature-dependent relaxation processes in the compound. The imaginary part of impedance decreases with rise in frequency [8].

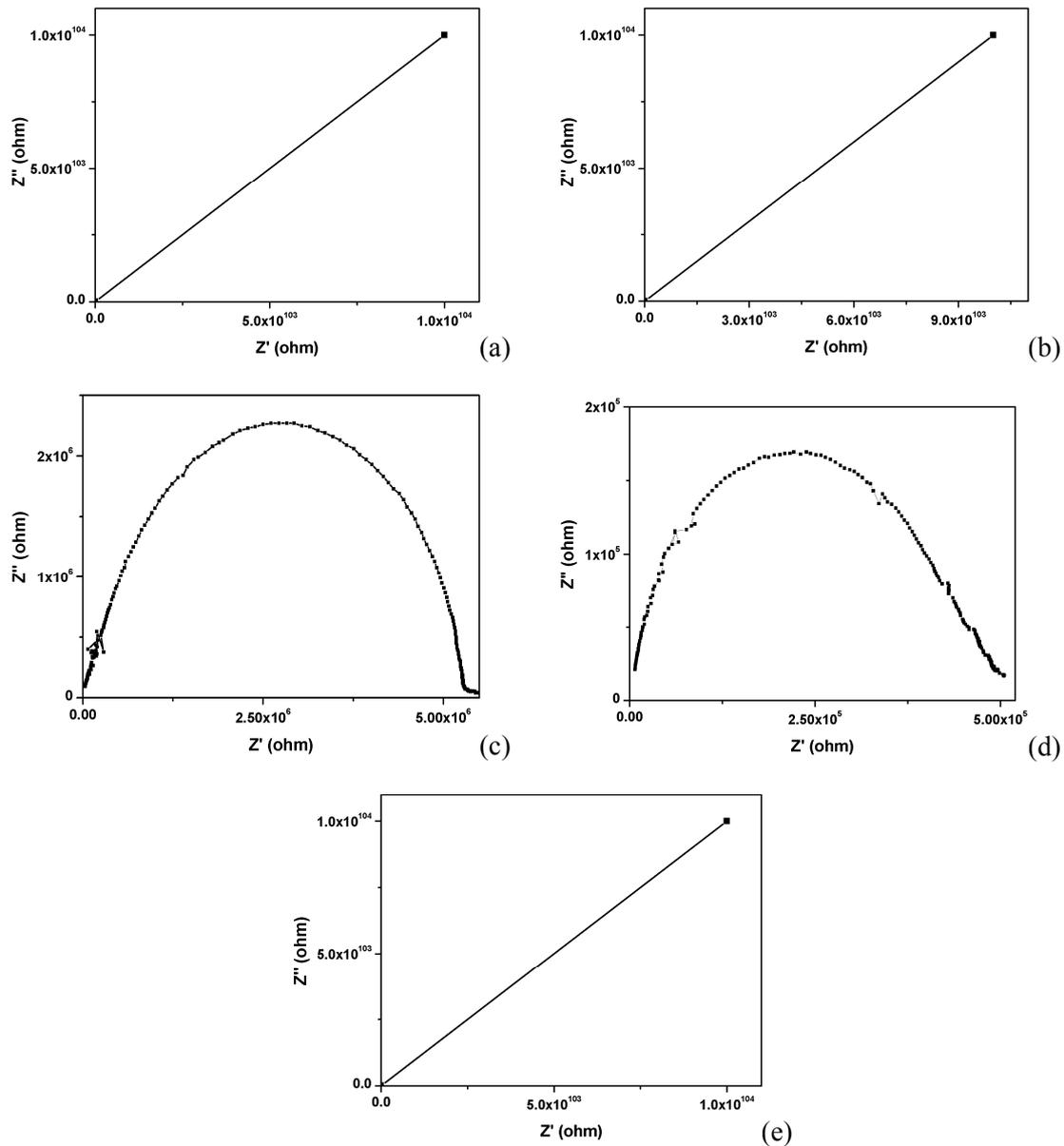


Figure 3: Plot the real and imaginary parts of impedance of composite material : a) $x = 0$, b) $x = 0.3$, c) $x = 0.5$, d) $x = 0.7$, e) $x = 1$

CONCLUSION

The composite of $(1-x)\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_{11.7}\text{Mn}_{0.15}\text{Ti}_{0.15} / x\text{La}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ ($x = 0; 0.3; 0.5; 0.7; 1$) has been synthesized by mechanical alloying. The composite with composition $x = 0.5$ and 0.7 provide the semicircle for frequency in range MHz.

ACKNOWLEDGMENT

This work was carried out under a research grant no. 2026/H2.R12.3/PPM.00 Penelitian/2011 and grant no. 014/K3.KU/2012 from which the authors are very grateful.

REFERENCES

- [1] Wei Chen, Ji Zheng, Yan Li, "Synthesis and electromagnetic characteristics of BaFe₁₂O₁₉/ZnO composite material," *Journal of Alloys and Compounds*, 513 (2012) 420– 424, doi:10.1016/j.jallcom.2011.10.060
- [2] Sachin Tyagi, Himanshu B. Baskey, Ramesh Chandra Agarwala, Vijaya Agarwala, Trilok Chand Shami, "Synthesis and Characterization of Microwave Absorbing SrFe₁₂O₁₉/ZnFe₂O₄ Nanocomposite," *Trans Indian Institute of Metal*, DOI 10.1007/s12666-011-0068-7, Dec 8, 2011.
- [3] L.A. Ramajo *, A.A. Cristóbal, P.M. Botta, J.M. Porto López, M.M. Reboredo, M.S. Castro, "Dielectric and magnetic response of Fe₃O₄/epoxy composites," *Composites: Part A* 40 (2009) 388–393.
- [4] K.K. Patankar, S.A. Kanade, D.S. Padalkar, B.K. Chougule, "Complex impedance analyses and magnetoelectric effect in ferrite–ferroelectric composite ceramics," *Physics Letters A* 361 (2007) 472–477
- [5] V.Vekky R. Repi, Azwar Manaf, "Substitution effect of (Mn,Ti) to the dielectric properties of barium-strontium hexaferrite for absorbing electromagnetic wave," *AIP Conf. Proc.* 1454, 282 (2012).
- [6] Yong Jun Seo, Geun Woo Kim, Chang Hoon Sung, Chan Gyu Lee and Bon Heun Koo, "Electrical Transport Properties and Magnetoresistance of (1-x)La_{0.7}Sr_{0.3}MnO₃/xZnFe₂O₄ Composites," *Kor. J. Mater. Res.* Vol. 20, No. 3 (2010), DOI: 10.3740/MRSK.2010.20.3.137
- [7] Hongfang Zhang, Chee-Leung Mak, "Impedance spectroscopic characterization of fine-grained magnetoelectric Pb(Zr_{0.53}Ti_{0.47})O₃–(Ni_{0.5}Zn_{0.5})Fe₂O₄ ceramic composites," *Journal of Alloys and Compounds*, 513 (2012) 165– 171.
- [8] Praveen Khatri, Banarji Behera, V. Srinivas, and R. N. P. Choudhary, "Complex Impedance Spectroscopic Properties of Ba₃V₂O₈ Ceramics," *Research Letters in Materials Science*, Volume 2008, Article ID 746256, 5 pages, doi:10.1155/2008/746256