Solid State Science and Technology, Vol. 23, No 1 & 2 (2015) 26-32
ISSN 0128-7389 | *http://journal.masshp.net*

EFFECT OF EVOLVING MICROSTRUCTURE ON MAGNETIC PROPERTIES OF NOMINAL $MG_{0.45}ZN_{0.5}CU_{0.05}$ FERRITE

Yusnita Yusuf, Mansor Hashim, Ismayadi Ismail

Materials Synthesis and Characterization Laboratory,
Institute of Advanced Technology,
University Putra Malaysia, 43400 Serdang, Malaysia.

Corresponding author: yusnitayusuf@hotmail.com,

ABSTRACT

In this work, we report on the evolution of the microstructure magnetic property relationship in MgZnCu ferrites with composition $Mg_{0.45}Zn_{0.5}Cu_{0.05}$ Fe_2O_4 applicable for producing an industrial ferrite product. The starting powders MgO, ZnO, CuO and Fe_2O_3 were mechanically alloyed for 10 hours in order to obtain nanometer size starting powder and formed into toroidal shape. The MgZnCu ferrite toroidal samples with nanometer size were then sintered from 600°C up to 1100°C with 100°C increments. The phase and surface morphology of the toroidal were characterized by X-ray diffraction (XRD) and field emission scanning electron microscopy (FESEM), respectively. The variation of the complex permeability was measured using an Agilent HP4291A Impedance Analyzer in the range of 1MHz to 1.0GHz. The saturation magnetization was determined from a B-H loop which was obtained via a MATS 2010SD Static Hysteresis graph. The average grain size of a sintered sample was measured over 200 grains by the linear intercept method. It was found that the average grain size increased from 0.014 µm to 1.017 µm. This indicated the microstructure evolution of the samples. The XRD patterns show that every single diffractogram contained multiple-phases due to the effect of copper addition into ferrites in nanostructures. The permeability decreased due to presence of pores and intergranular pores provides the impediment to the domain wall displacement and caused a decrease in permeability. The density, grain size, resistivity and saturation magnetization of all the samples increased with the sintering temperature. Materials can be widely used in electronic circuit in a frequnecy range from 8 to 10 MHz.

Keywords: Magnetic properties; Mechanical alloying; MgZnCu ferrite; Microstructure

INTRODUCTION

The synthesis of nanocrystalline spinel ferrite has been investigated in recent years due to their potential applications in high density magnetic recording, microwave devices and magnetic fluid. MgZn ferrite is considered a candidate material with a spinel structure for high frequency engineering ceramics because of its high electrical resistivity, high curie temperature and low cost [1]. The magnetic cations in the cubic spinel lattice can occupy A-type or B- type crystallographic positions. For example,

Mg^{2+} and Zn^{2+} cations prefer octahedral and tetrahedral sites respectively [2, 3]. MgZn ferrite composition plays a crucial role in lowering the firing temperature. MgCuZn ferrite can be prepared at low sintering temperature with improved density with higher resistivity. Normally MgCuZn ferrites were sintered at temperatures higher than 1000^oC. It is well known that the change in temperature or method of sample preparation can strongly affect the particle size and cation distribution [3]. This paper is focused on the preparation of $Mg_{0.45}Zn_{0.5}Cu_{0.05}$ Fe_2O_4 by the mechanical alloying method. Therefore, MgCuZn ferrites were selected and the detailed study of effect of Cu substitution on the densification characteristics has been carried out [4]. The structure and magnetic properties of the as-synthesized ferrite powders are discussed as dependent on different sintering temperatures.

EXPERIMENTAL

The ferrite composition $Mg_{0.45}Zn_{0.5}Cu_{0.05}$ Fe_2O_4 was synthesized through the mechanical alloying method. The starting powder materials (Alfa Aesar) used were magnesium oxide, zinc oxide, copper oxide and iron oxide. Those powders were mixed up together with polyvinyl alcohol, PVA. The samples were prepared into two samples. The powders mixtures were formed into a toroid by using a hydraulic press. Then, the sintering process was carried out where microstructure of the ferrite changed with the sintering temperature. As the toroids were sintered at different temperatures starting at 600^0C up to 1100^0C, these toroids were characterized with respect to phase identification, grain size and lattice parameter determination using X-Ray Diffraction. The sintered ferrites were investigated for their electrical and magnetic properties such as saturation magnetization, initial permeability, and resistivity.

RESULTS AND DISCUSSION

The XRD patterns of $Mg_{0.45}Zn_{0.5}Cu_{0.05}Fe_2O_4$ before sintering and the sintered samples at different temperatures starting from 600°C to 1100°C are shown in Figure 1. As can be observed from the XRD spectra, the trend of peak changes suggests that MgO diffused into Fe_2O_3 and the Mg- Fe_2O_3 phase was formed. ZnO diffused into Fe_2O_3 and formed Zn- ferrites. The broadening effect be attributed to a very small size of particle produced by mechanical alloying [8]. At 600°C sintering temperature, there were a few traces of Zn-ferrite. The presence of Cu-ferrite, Mg-ferrite, and Zn-ferrite at sintering temperature 700°C phases suggested that the thermal energy supplied with the materials during the sintering process was not enough to complete the reaction. For a further sintering temperature at 900°C, the major peaks were still as sintered before. MgZnCu alloy were formed. This may be due to increase of the grain size and crystallinity of the materials. At further sintering to 1100°C, Mg-ferrite, Cu-ferrite and MgZnCu were obtained. The expected ferrite of composition MgZnCu ferrite was not formed instead 3 different phases were obtained. Thus, the effect of adding Cu in nano size starting powder did not produce a single phase ferrite as obtained using micron size powder.

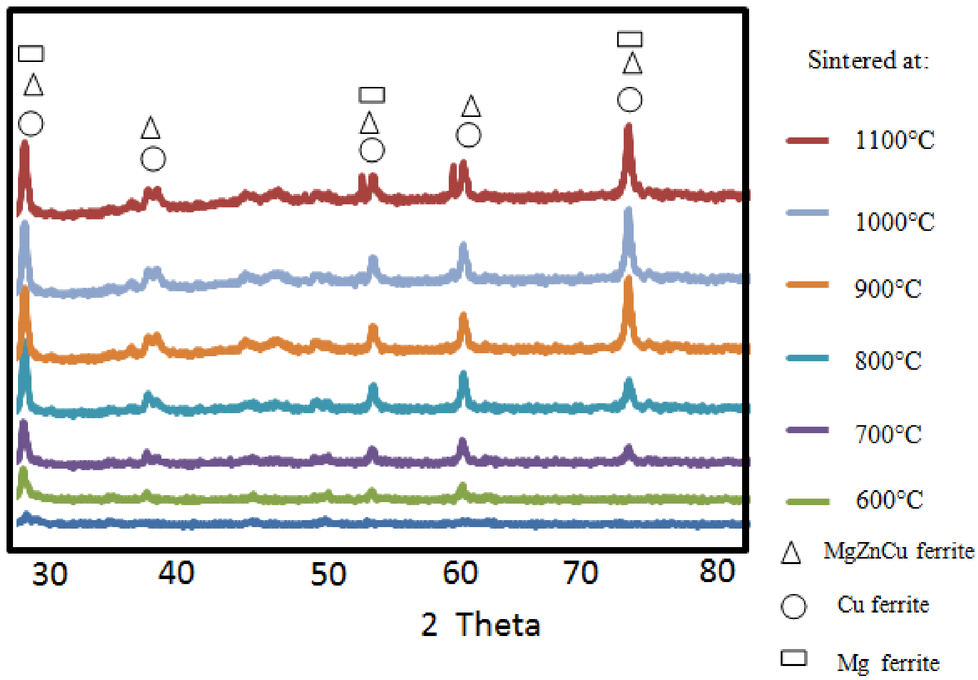

Figure 1: XRD graphs of $Mg_{0.45}Zn_{0.5}Cu_{0.05}Fe_2O_4$

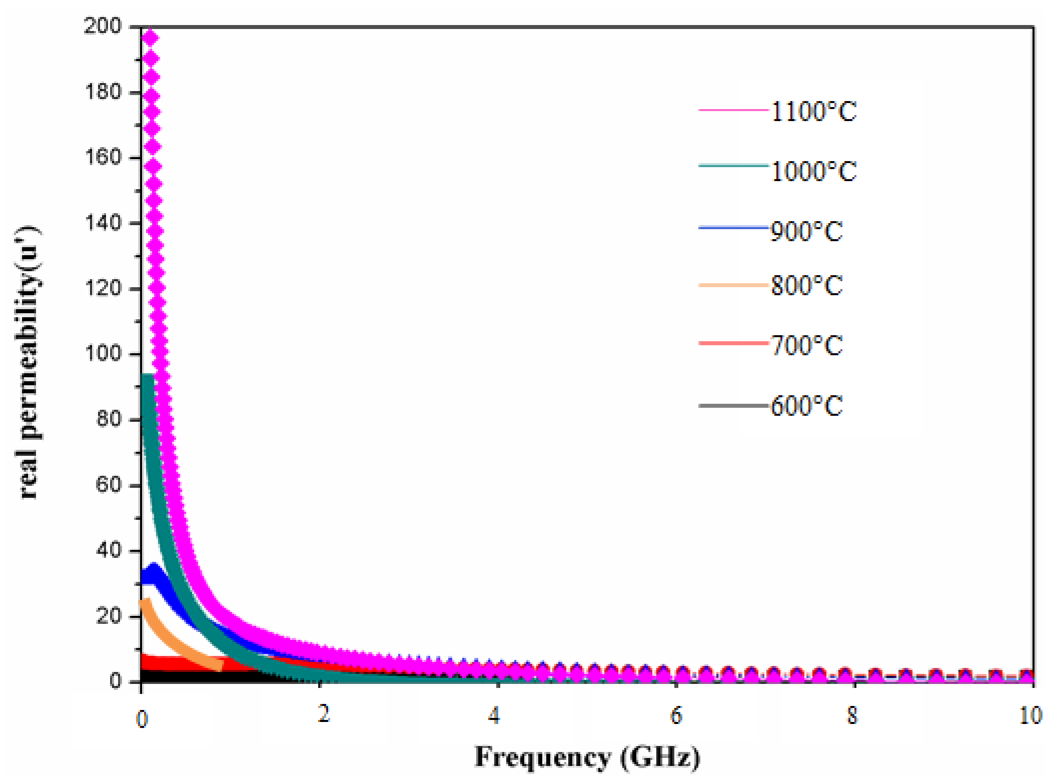

Figure 2: A graph of initial permeability against frequency

Solid State Science and Technology, Vol. 23, No 1 & 2 (2015) 26-32
ISSN 0128-7389 | *http://journal.masshp.net*

Based on the results shown in Figure 2, permeability versus frequency shows a decreasing trend. There is a linear relation between initial permeability and grain size. The domain wall motion is greatly affected by the grain size. However, larger grains tend to contain a greater number of domain walls and a large initial permeability is a result of the easy reversal of domain wall displacement [8]. The greater the number of domain walls, the higher the initial permeability. The permeability decreases due to presence of pores and intergranular pores provides the impediment to the domain wall displacement and causes decrease in permeability [9].

Figure 3 shows the B-H loops of $Mg_{0.45}Zn_{0.5}Cu_{0.05}$ ferrite sintered at different temperatures from 600^{o}C up to 1100^{o}C. At a lower temperature which is 600^{o}C, the magnetic flux density is linearly proportional with magnetic field, H. Further sintering up to 900^{o}C produced a noticeable increase of B in the hysteresis loop indicating an increase of ordered magnetism in the sample. At this stage, ferromagnetism was becoming dominant compared to the paramagnetism behaviour. For sintering at 1000^{o}C and 1100^{o}C, the hysteresis loops show very obvious formation of ordered magnetism. The magnetic properties show a well established hysteresis loop of a normal soft ferrite sample. At this stage, ferromagnetic behaviour was dominant with some traces of paramagnetism and supermagnetism being insignificant relative to ferromagnetism in the samples.

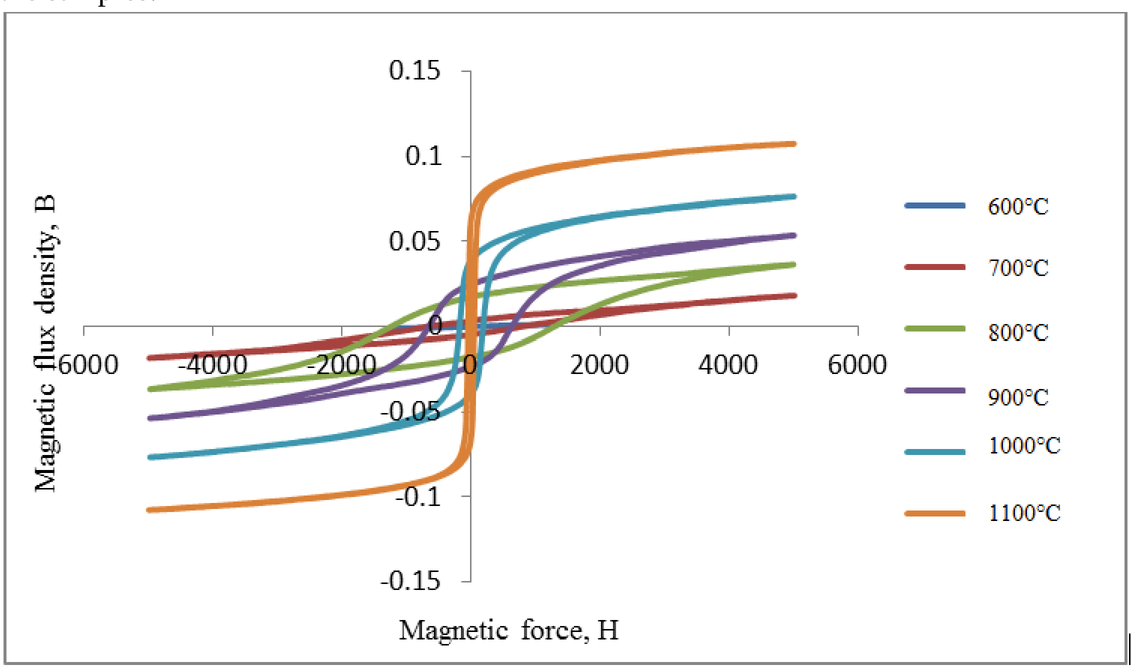

Figure 3: Magnetic flux density versus magnetic field

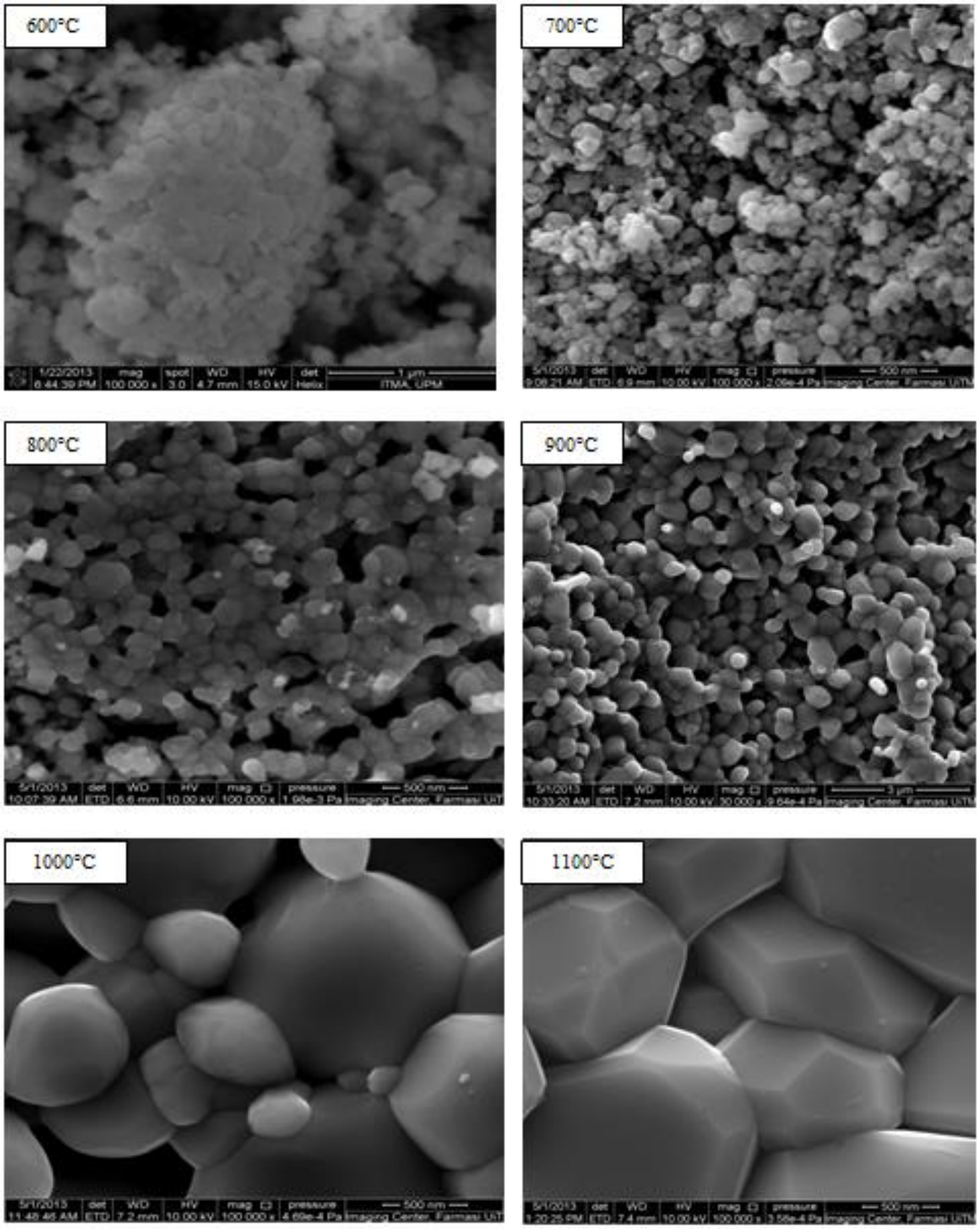

Figure 4: Microstructures of MgZnCu ferrites at various temperatures

The single-sample sintering at various temperatures showed a microstructural evolution with the increase of sintering temperatures. The average grain size (Figure 4) of a sintered sample was obtained over 200 grains by the linear intercept method [10]. It was found that the average grain size increased from 0.014 µm to 1.017 µm (figure 5). This indicated the microstructure evolution of the samples. The increased grain size shows a sign of the development of grain growth. The grains growth to larger grain size indicated more development of domain walls and exhibits a threshold of the evolution of bulk ferromagnetic properties. For sintering temperatures at 1000^{o}C up to 1100^{o}C, the grains were large enough to accommodate domain wall motion in the material. The increasing number of grains contributed to increasing magnetization. Addition of copper in the material helps the grain sizes to grow fast with increasing sintering temperature.

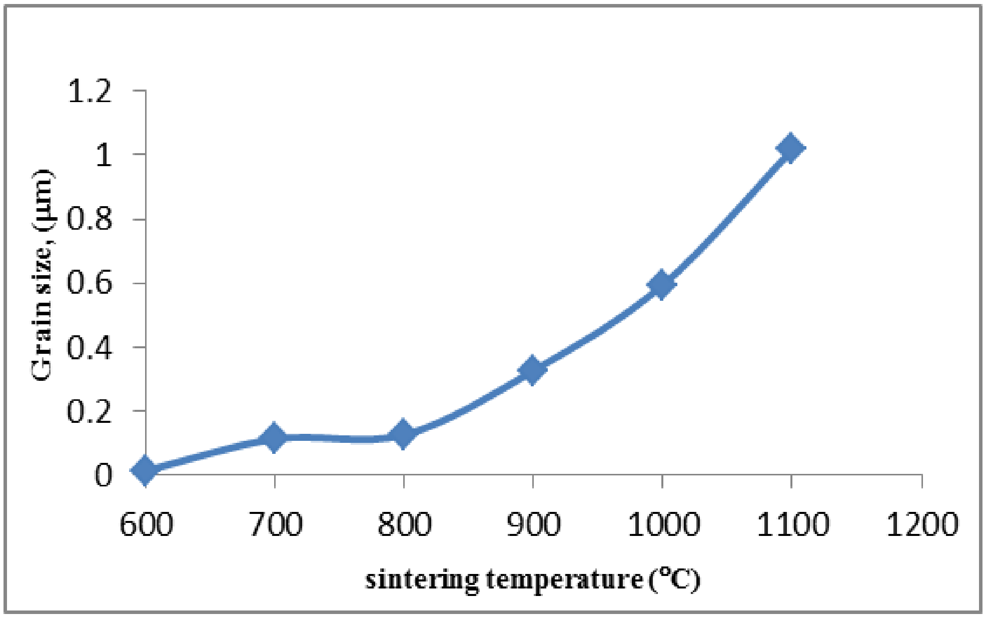

Figure5: Average grain size versus sintering temperatures

CONCLUSION

To conclude, the synthesis of $Mg_{0.45}Zn_{0.5}Cu_{0.05}Fe_2O_4$ has been attempted using the mechanical alloying method with ball to powder weight ratio (BPR). The effects on the magnetic properties of the materials were studied by using XRD, FESEM and impedance analyser with the frequency range of 1MHz to 1.8GHz. The increasing trend of average grain size was observed after 600°C. Adding copper oxide affects the evolving of microstructures at low sintering temperatures. However, based on the XRD patterns, the substitution of copper oxide in nominal composition $Mg_{0.45}Zn_{0.5}Cu_{0.05}Fe_2O_4$ produced a multi-phase sample. This is a new finding for a nanostructured ferrite. Thus, the effect of adding Cu in nano size starting powder did not produce a single phase ferrite as obtained using micron size powder.

ACKNOWLEDGEMENT

The author are grateful to the Universiti Putra Malaysia for kindly providing the instrumental measurements in completing this research.

REFERENCES

[1]. Penchal Reddy, *Superlattices and Microstructures*, **56** 99-106 (2013)
[2]. Shenoy, S.D., P.A. Joy, and M.R. Anantharaman, *Journal of Magnetism and Magnetic Materials,* **269** (2) 217-226 (2004)
[3]. Mukherjee, K. and S.B. Majumder, *International Journal of Hydrogen Energy*,**39** (2*)* 1185-1191 (2014)
[4]. Rezlescu, *Journal of Magnetism and Magnetic Materials,* **185** 199-206 (1998)
[5]. Toniolo, J.C., *Materials Research Bulletin*, **40** (3) 561-571 (2005)
[6]. Sharifi, I., *Journal of Magnetism and Magnetic Materials,* **324** 1854-1861 2012.
[7]. Vasanthi, V., *Journal of Magnetism and Magnetic Materials,* **324** (13) 2100-2107 (2012)
[8]. Ismail. I., *Journal of Magnetism and Magnetic Materials*, **323** 1470-1476 (2011)
[9]. Barati, M.R., Journal of Alloys and Compounds, **478** (1-2) 375-380 (2009)
[10]. Porter, D.L., G.K. Bansal, and A.H. Heuer: *Journal of the American Ceramic Society* 59 (3-4) 179-182 (1976)