

Synthesis and Characterization of Magnesium Zinc Ferrites as Electromagnetic Source

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Abstract: Fabrication of $Mg_{1-x}Zn_xFe_2O_4$ (where $x = 0.2, 0.3, 0.4$ and 0.5) samples using a conventional technique is reported. Oxides of magnesium, zinc and iron with purity of about 99.99% were mixed with distilled water in a milling machine for 12 hours. They were then filtered and pre-sintered at $1250^\circ C$ and sintered at $1350^\circ C$ for 10 h in air. The phase and the crystal structure of the as-prepared samples were identified using X-Ray Diffraction analysis (XRD). Hysteresis graph was evaluated using Vibrating Sample Magnetometer (VSM) to get the saturation magnetization ($emu\ g^{-1}$) and coercivity (G) value. Atomic Force Microscope (AFM) was used to observe the surface morphology. The X-ray diffraction analysis showed major peak at plane (3 1 1) of the cubic structure for all the ferrites. The largest value of density achieved was $4.69\ gcm^{-3}$ which was exhibited by sample with 0.5 mole fraction of zinc content. Highest saturation magnetization value ($3.652\ emu\ g^{-1}$) was obtained for the 0.3 mole fraction of zinc content. A strong correlation between the saturation magnetization and zinc content was observed. We speculate that it is due to the Heisenberg super-exchange interaction of magnetic Fe^{3+} and non-magnetic Zn^{2+} occupying the tetrahedral sites. The sample was used to induce electromagnetic (EM) waves in high operating frequency (5 MHz).

Key words: Tetrahedron, x-ray diffraction, hysteresis, electromagnetic field and super-exchange interaction

INTRODUCTION

Soft magnetic material has been used widely in many electronic devices. The most popular type is the cubic spinel structure which has tetrahedral A site and octahedral B site in the AB_2O_4 crystal structure^[1]. It is because their magnetic properties are easy to control as it depends regularly on the variation of the compositions and cation distribution. Moreover it often has high permeability in the Radio-Frequency (RF) region, high electrical resistivity, mechanical hardness, chemical stability and reasonable cost^[2]. Zinc is known as the main contributor in determining the ferrite properties. It was proven that variations in zinc content is able to tune the resonance frequency^[3]. In fact, the application of ferrite as a magnetic feeder to excite an antenna that can cover wide frequency bandwidth from 200MHz-1Hz with different length 0.3-50m^[4] was reported. Ferrites containing Mg and Zn possess higher resistivity value than Mn-Zn one ($\sim 10^7\ \Omega cm^{-1}$) where

the resistivity of Mg-Zn ferrite is $10^6-10^7\ \Omega cm^{-1}$ and make it more effective in high frequency^[5]. It was found that the sintering temperature has strong effect on the microstructure and as such may affect the hysteresis^[6]. This study premise deals with fabrication and characterization of magnesium zinc ferrite, $Mg_{1-x}Zn_xFe_2O_4$ ($0.2 \leq x \leq 0.5$) samples using conventional solid state reaction. A high saturation magnetization is expected to be seen. The enhancement of the study will be proceeding on the application of the as-prepared sample to be used as an Electro Magnetic (EM) source. This study premise shall be the basis of us using the EM source in hydrocarbon detection for onshore exploration.

Experimental: The raw materials consist of MgO, ZnO and Fe_2O_3 according to their weight ratio were mixed with the distilled water in a milling machine for 24 h. The added distilled water led the mixing process and helped the fluid cooling before the mixture was filtered

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and dried in an oven at 110°C for 24 h. After that the pre-sintering process were done in the carbolite furnace at 1250°C for 10 h. During the pre-sintering process, the mixtures combine by the solid-state reaction where the atoms get into the lattice to form the crystal structure. The pre-sintering process also allows the decomposing of the oxides to avoid formation of unwanted gaseous. It evaporates the unknown masses that remain in the samples. Then the materials are crushed by using porcelain mortar until finer texture is observed and the grinding process will take place in about 6 hours by using Fritsch Planetary Micromill. Next, the mixture was dried in the oven at 110°C for 24 h. The sample was crushed again and added with 1 wt% of Polyvinyl Alcohol (PVA) to act as a binder and 1 wt% of zinc stearate to fill the pores between the particles to give better reaction during sintering. The rate of the grain growth depends on the sintering temperature, the stoichiometry and the oxygen partial pressure^[7]. The sintering process was done at 1350°C for 10 h. in air. The heating permits the mobility of the atoms for the grain growth. Thus at this stage a spinel lattice is expected to be formed. The characterization of the sintered samples was done by Philips X-ray diffractometer to confirm the formation of the single phase cubic spinel crystal structure, Lakeshore-7300 Vibrating Sample Magnetometer (VSM) to evaluate the saturation magnetization and Atomic Force Microscope for surface morphology observation. Small MgZn Ferrite toroid core with 120 turns was used to produce EM field. GW INSPEK GFG-8250A function generator was used to induce a 5 MHz sinusoidal wave. The peak to peak Voltage was detected using a Tektronix digital oscilloscope TDS 3012B.

RESULTS AND DISCUSSION

The density of the samples was evaluated by using the Archimedes principle. It shows the densities increased from 3.34 gcm⁻³ to 4.69 gcm⁻³ by increasing the Zn content in sample. The values are in agreement with the literature data that is in between 4.520 gcm⁻³^[8]. Zinc is known to bring about the densification, grain growth and reduce anisotropy^[4].

The X-Ray Diffraction (XRD) measurements were carried out using Cu K_α radiation (λ = 0.54nm) with a Philips diffractometer (PW1830/3210) operated at 35kV and 25mA in the range of 22-80° of the 2θ (°).

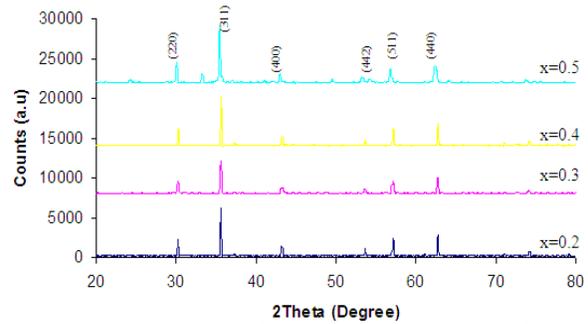


Fig. 1: XRD Pattern After Sintering at 1350°C for Mg_{1-x}Zn_xFe₂O₄ samples

Table 1: Values of density, d-spacing (Å), intensity after presintering and after sintering at plane (3 1 1) for Mg_{1-x}Zn_xFe₂O₄ samples

X (mol)	Density, ρ (gcm ⁻³)	d-spacing (Å)	Intensity After presintering (counts)	Intensity After sintering (counts)
0.2	3.34	2.5195	4322	5418
0.3	3.82	2.5193	4070	6203
0.4	4.68	2.5159	6165	6355
0.5	4.69	2.5338	4767	5308

All of the lines in the pattern were indexed by cell refinement with a cubic structure of space group Fd-3m (SG#227). The XRD patterns of Mg_{1-x}Zn_xFe₂O₄ ferrites after sintering at 1350°C are shown in Fig. 1, indicating a single phase cubic spinel structure (JCPDS card number 00-008-0234) where the diffraction spots at d-spacing values of 2.5195 Å, 2.5193 Å, 2.5159 Å and 2.5338 Å for (3 1 1) plane reflection as the major peaks. A well-crystalline single phase structure were described by the intense sharp peak of the diffraction pattern as well as there were almost no second phases formed in the samples when temperature was increased to 1350°C. The sintering process had cause the atom to move to its lattice completely. In fact it is understood that the Zn²⁺ ions occupy the tetrahedral (A) site, Mg²⁺ ions have a preference for the octahedral (B) site while the Fe³⁺ ions are distributed over both site^[9]. From Table 1, we can observe that the diffracted intensity of all the sintered samples is higher than those of presintering samples.

Hysteresis loops (Fig. 2) are observed for the measurement of saturation magnetization (emu g⁻¹) and coercivity (Gauss) that is a result of several intrinsic and extrinsic properties which depends on some factors such as density, anisotropy, grain size and A-B exchange interactions^[4]. As can be seen from the graph and the table, the Mg-Zn ferrite obtained in this project showed a normal type (S-shape) hysteresis curve with high saturation magnetization and narrow loop that brought the low coercivity values ranging from 0.371-1.023 G.

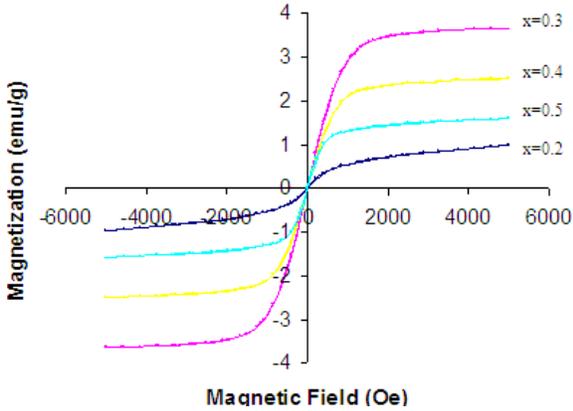


Fig. 2: Hysteresis for $Mg_{1-x}Zn_xFe_2O_4$ samples

Table 2: Value of Saturation Magnetization, Coercivity and Average Particle Size of $Mg_xZn_{1-x}Fe_2O_4$ samples

X (mole fraction)	Saturation Magnetization, M_s (emu g^{-1})	Coercivity (Gauss)	Average Particle Size, μm
0.2	0.974	1.023	1.94
0.3	3.652	0.889	1.48
0.4	2.508	0.548	1.40
0.5	1.598	0.371	1.64

The saturation magnetization increases from 0.974-3.652 $emu\ g^{-1}$ and then decreasing to 2.508 and 1.598 $emu\ g^{-1}$. The saturation magnetization value supposed to increase by substituting the zinc content. Instead, M_s shows a decreasing trend when $x = 0.4$. This phenomenon can be explained further. $MgZnFe_2O_4$ has inverted spinel crystal structure^[1]. It is known that Zn^{2+} ion prefer tetrahedral (A-sites), Mg^{2+} ion prefer octahedral (B-sites) while Fe^{3+} ion distributed over both sites. The preference of the non-magnetic Zn^{2+} ion in the A-site will encourage the migration of Fe^{3+} ion into B-site which gives rise to antiparallel spin coupling and spin canting resulting in weakening of the A-B exchange interaction thus increase the saturation magnetization^[3]. However, with more and more Zn^{2+} ion, the ($x > 0.4-0.5$) the ferrite become normal spinel. The A-B super-exchange interaction weakens and this will result stronger B-B interaction. Due to this, magnetic moments of a part of Fe^{3+} ions (which are now only in B-position) becomes reversibly oriented which leads to the decrease of the total magnetic moment in ferrites^[1].

Microstructure analysis: As can be seen from the Fig. 3, the surface morphology of the samples is rough. It is nonhomogeneous microstructure because the average particle size for every sample is different. Each particle usually contained a large number of atoms. From

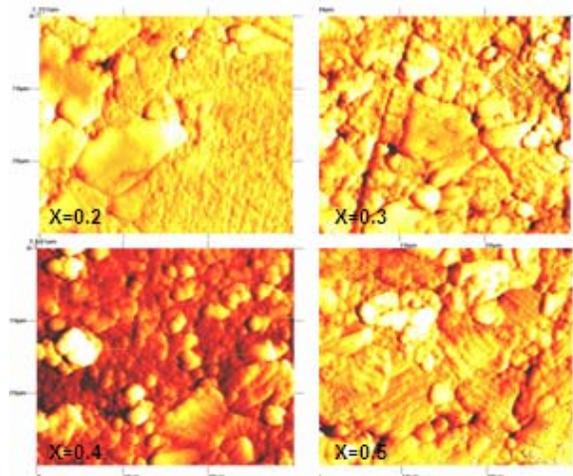


Fig. 3: The morphology of $Mg_{1-x}Zn_xFe_2O_4$ samples where $X = 0.2, 0.3, 0.4$ and 0.5 after sintering at $1350^\circ C$.

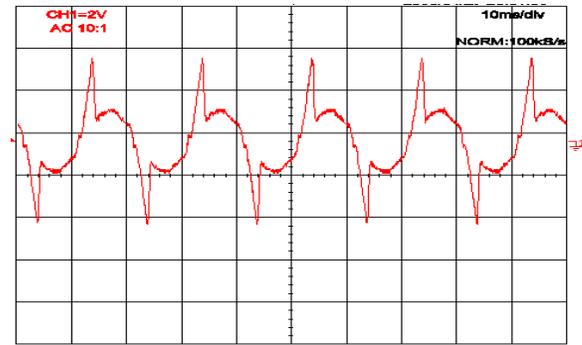


Fig. 4: Voltage versus time of the MgZnFerrite based EM source

Table 2, we can see that 0.2 mole fraction zinc content has the largest average particle size.

The ferrite sample was used to induce electromagnetic field at 5 MHz. The voltage (peak to peak) obtained was about 12.8 V (Fig. 4). The toroidal shape EM source were able to transmit EM wave to more than 1 meter. It should be noted that an induction coil EM detector was used to detect the EM waves. Traces of the hysteresis behavior can be seen on the graph plot. More work on the EM source system shall be carried out in our lab.

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