

EFFECT OF SINTERING TEMPERATURE ON PHYSICAL AND ELECTROMAGNETIC PROPERTIES OF $MgFe_2O_4$ FERRITES VIA SOLUTION COMBUSTION METHOD

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Polycrystalline $MgFe_2O_4$ ferrite was prepared by solution combustion method using metal nitrates and citric acid. Enhancement of the density of the samples has been observed with the increasing in sintering temperature and has great influence on the electromagnetic properties of $MgFe_2O_4$. The XRD analysis of the as-synthesized powder confirms the formation of the single phase $MgFe_2O_4$ spinel structure. Decrease in resistivity with increase in sintering temperature confirms the semiconducting behavior of the prepared ferrites. It was found that magnetic properties can be tuned by different sintering conditions. Also the increased value of dielectric constant (ϵ') has been observed with the increase in sintering temperature. There was no maximum dielectric loss within the measured frequency range until 1 GHz due to excellent compositional control in this method.

Keywords: ferrite/ resistivity/ dielectric loss/ dielectric constant/ magnetic

INTRODUCTION

The study of spinel ferrite is of great importance from both the fundamental and the applied research point of view. High resistivity, low dielectric losses, and chemical stability make them very promising candidates for high frequency applications [1]. The ferrite is not completely defined by its chemistry and crystal structure. So, in order to reduce the losses many parameters such as density, grain size, porosity and their intra and intergranular distribution must be controlled [2]. Optimum sintering temperature is of great importance as sintering influences both densification and grain growth and many magnetic and electrical properties benefit from both a high relative density and a small grain size. This opens the doors for tailoring given properties by careful synthesis of the building blocks (atoms and molecules) and their assembly to fabricate functional materials with improved properties. In this study, the effects of sintering temperature on physical and electromagnetic properties of the $MgFe_2O_4$ are investigated.

METHOD AND MATERIALS

Nanocrystalline $MgFe_2O_4$ ferrite powders were prepared following a procedure and schematized in form of a flow diagram in Fig.1.

$Fe(NO_3)_2 \cdot 9H_2O$ (Aldrich, 99.8%), $Mg(NO_3)_2 \cdot 6H_2O$ (Sigma-Aldrich 99%), Citric acid (Sigma-Aldrich 99.5%) were used respectively as sources of metal ions and chelating-fuel agent. Their aqueous solutions were mixed together into a Pyrex beaker with 1:1 molar ratio of metal to citric acid. The clear orange brown solution thus obtained had a pH < 1. The dark brown solution was allowed to evaporate on a hot plate maintaining the solution temperature at 80 °C. Upon the formation of a dense sticky gel, the temperature was increased to 120 °C for the condensation process. The temperature was then increased rapidly and when it reached to ~180 °C, large amount of gases (CO_2 , H_2O , N_2) were liberated and a dark brown ferrite powder was produced through the combustion process.

The as burnt ash was calcined at 500 °C/5h for better crystallization and homogeneous cation distribution in the spinel. As the auto combustion is so rapid that the cations may not be well distributed in ferrite lattice of as-burnt ashes. The calcined ferrite powder was granulated using agate motor and was uniaxially pressed at a pressure of 100 MPa to form pellet specimens. The specimens were sintered at 1100 to 1250 °C in air atmosphere. The calcined powders and the sintered pellets were characterized with respect to phase identification, crystallite size and lattice parameter determination using X-ray diffraction (Bruker, D8) with CuK_{α} radiation. The bulk density and porosity of sintered ferrites were

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measured by Archimedes' method. The scanning electron microscopy (SEM) photographs were recorded using Zeiss Supra 35VP scanning electron microscope. Impedance/material analyser (HP/Agilent 4291B RF) was used to measure the dielectric constant and dielectric loss. Saturation magnetization measurement was carried out at room temperature using vibrating sample magnetometer (VSM) with a maximum magnetic field of 10kOe.

RESULTS AND DISCUSSION

X-ray analysis

The as-burnt and calcined ferrites were characterized by XRD and it was shown in Figure 1. The powders were in crystalline state and identification revealed spinel ferrite phases similar to JCPDS card number 01-073-2410. There was no metal oxide phase in the as burnt powder. This observation revealed that the nano-crystalline MgFe₂O₄ ferrite powders could be synthesized directly by the auto-combustion of nitrate-citrate gels. The crystallite size was calculated by using Scherer equation. It was found that the crystallite size of as-burnt powders was 8.16 nm. The as burnt powders were calcined at 500 °C to get more crystalline homogeneous spinel phase. The crystallite size of calcined powder was 12.77 nm.

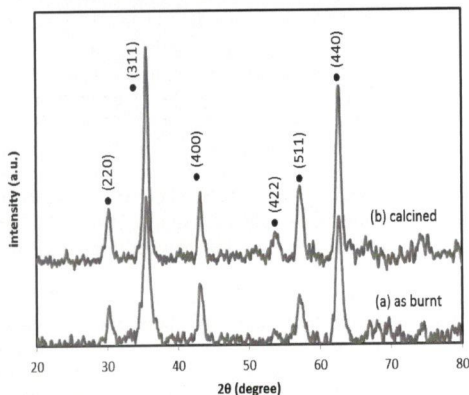


Fig.1. XRD spectra of (a) as-burnt powders (b) calcined powder (500 °C)

Fig. 2 illustrates the XRD patterns for different sintered ferrites. As can be observed from the figure, the sintered specimens had formed a stable single phase of MgFe₂O₄ ferrites with cubic spinel structure.

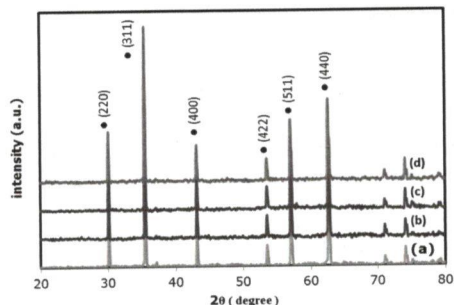


Fig.2. XRD spectra of MgFe₂O₄ ferrite sintered at (a) 1100 °C (b) 1150 °C, (c) 1200 °C, (d) 1250 °C prepared by SCM.

Table 1 presents the crystallite size, lattice parameter and X-ray densities of sintered ferrites at different temperatures. Lattice parameters were calculated from the XRD analysis with Eva software and X-ray density was determined using equation 1. The crystallite size of the specimen was dependent on the sintering temperature. By increasing the temperature from 1100 to 1250 °C, the crystallite sizes were found to increase from 65 to 75 nm. Increasing the sintering temperature increase the crystallite size. However, there was no noticeable change in lattice parameters as well as X-ray densities. This slightly reduction in lattice parameter is attributed to the arrangement of cations in the nanosized MgFe₂O₄ crystallite and also O₂ release from the samples leading to the lattice shrinkage [3].

$$\rho = \frac{8 M_W}{N a^3} \quad (1)$$

where, *M_w* is the molecular weight, *N* is Avogadro's number and 'a' is the lattice parameter of unit cell.

Table 1 Crystallite size, Lattice parameter and X-ray density of MgFe₂O₄ ferrite by SCM.

Temperature (°C)	Crystallite size (nm)	Lattice Parameter (a) °Å	X-ray density (g/cm ³)
1100	65.55	8.3863	4.5144
1150	72.31	8.3863	4.5044
1200	73.76	8.3853	4.5044
1250	74.17	8.3861	4.5018

TEM analysis

Fig.3 shows the TEM image of $MgFe_2O_4$ ferrite powder calcined at 500 °C by SCM. It was found that the shape of the particle is irregular and it exhibits a soft agglomerated monophase polycrystalline and estimated average particle size is ~7 to 10 nm, which is fairly consistent with the crystallite size determined by using XRD method. Hence, this observation again supported that nanocrystallite $MgFe_2O_4$ ferrite powders can be produced by SCM.

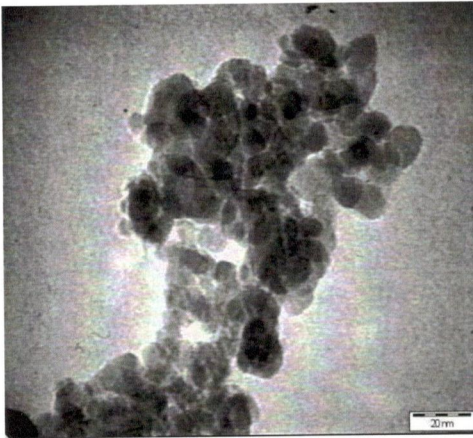


Fig 3. TEM micrograph of $MgFe_2O_4$ ferrite powder calcined at 500 °C by SCM

SEM analysis

The microstructures of the sintered pellets were shown in Figure 4. The average grain size was calculated using image J software. It was found that the average grain sizes were increased from ~0.2 (1100 °C) to ~2 μm (1250 °C) and the associated porosity decreased slightly with the increase in sintering temperature. It was also observed that the brittle nature of failure of the ferrite with the smooth surface and the similar behavior was observed in the microstructure of $NiFe_2O_4$ ferrite synthesized by sol gel auto combustion [4].

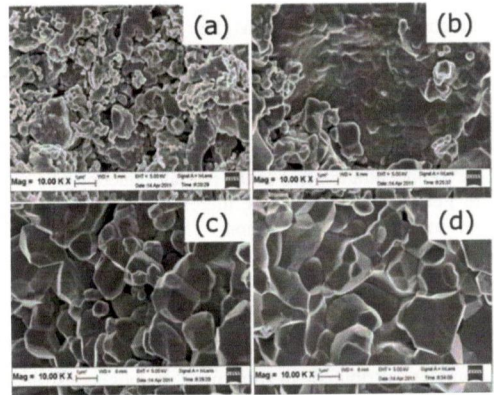


Fig 4. SEM micrographs of $MgFe_2O_4$ ferrites sintered at (a) 1100 °C, (b) 1150 °C, (c) 1200 °C, (d) 1250 °C prepared by SCM

Electrical properties

The variation of dielectric constant (ϵ'), dielectric loss ($\tan \delta$) and resistivity (ρ) were studied in the frequency range 1MHz to 1GHz at room temperature to investigate the electrical properties of $MgFe_2O_4$ ferrites prepared by solution combustion method.

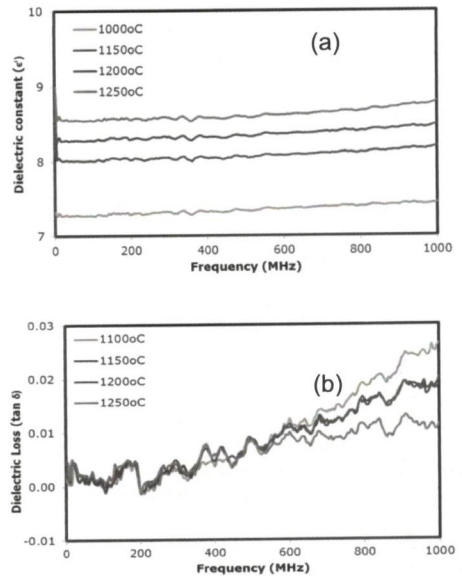


Fig 5. Variation of (a) dielectric constant and (b) dielectric loss with frequency for $MgFe_2O_4$ sintered at different temperatures

Dielectric constant and dielectric loss as a function of frequency for $MgFe_2O_4$ ferrites sintered at different temperatures are shown in Fig.5. The values of dielectric constant increase with increase in sintering temperature and is in the range of 7- 9, from a frequency of 1 MHz to 1 GHz. The increase of sintering temperature would bring to increase in the Fe^{2+} concentration due to the partial reduction of Fe^{3+} to Fe^{2+} takes place and is, therefore, responsible for an increase in polarization. Thus, high value of dielectric constant was observed for the sample sintered at 1250 °C.

The dielectric loss of all the samples in SCM is within 10^{-2} and there is no significant variations. The low dielectric losses obtained for the present samples attributed to be more homogenous and structurally perfect ferrites prepared by SCM. This finding is similar with the study of dielectric properties of Al substituted Co ferrite nanoparticles prepared by sol-gel method [5,6]. In table 2, the resistivity is decreased with increased in sintering temperature. This may be attributed to the increase in grain size. It is known that larger grains result in less number of grain boundaries, which act as a scattering center for the flow of electrons and therefore decrease of resistivity [7].

Table 2 Resistivity of $MgFe_2O_4$ for the different sintering temperature prepared by SCM.

Temperature (°C)	Resistivity (* $10^6 \Omega cm$) at 1 MHz
1100	24.93
1150	23.78
1200	18.82
1250	18.73

Magnetic properties

The magnetic hysteresis curves for $MgFe_2O_4$ ferrites sintered at different temperatures are displayed in Fig.6. The magnetic moments per unit cell and saturation magnetization of the ferrites were found to increase with the increase in sintering temperature while the coercivity and remanent magnetization decreased with the increase in sintering temperature. The calculated values of some magnetic properties are presented in Table 3.

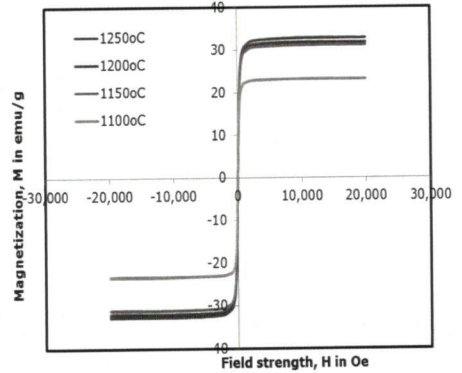


Fig.6. The magnetization curves of $MgFe_2O_4$ ferrites with different sintered temperature measured by VSM $\pm 10KOe$ at room temperature prepared by SCM.

Table 3 The value of M_s , M_r , H_c and n_B for the Mg ferrites with different sintered temperature prepared by SCM.

Sintered Temperature(°C)	M_s (emu/g)	M_r (emu/g)	H_c (Oe)	n_B (μB)
1100	23.41	8.09	10.40	0.81
1150	31.43	6.61	6.53	0.89
1200	32.38	6.06	8.00	1.04
1250	33.05	0.67	6.65	1.18

CONCLUSIONS

The influences of sintering temperature on the physical and electromagnetic properties of $MgFe_2O_4$ ferrites were investigated. Enhancement of relative density has been observed with the increase in sintering temperature. A decrease in resistivity with an increase in sintering temperature may be attributed to the microstructural factors such as grain size, porosity and grain boundary area. Increased value of dielectric constant has been observed because with an increase in sintering temperature conversion of trivalent Fe^{3+} ions to the divalent Fe^{2+} ions took place and thereby dielectric constant increased. The magnetic properties can be tuned by different sintering temperature.

REFERENCES

- [1] Lal, M., Sharma. D.K. and Singh, M. Effect of processing and polarizer on the electrical properties of Mn-Zn ferrites, (2005). *In. J. Pure & Apply Phys.* Vol 42, pp. 291.
- [2] Ishini, K and Naruyima, Y. Development of magnetic ferrites: Control and application of losses, (1987). *Am. Ceram. Soc. Bull.*, Vol 66, pp. 1469.
- [3] Verma, A and Chatterjee, R. Effect of Zn concentration on the structural, electrical and magnetic properties of mixed Mn-Zn and Ni-Zn ferrites synthesized by the citrate precursor technique, (2006). *J. Magn. Magn. Mater.*, 306, pp. 313-320
- [4] Pradeep. A., Priyadharsini, P and Chandrasekaran, G. Production of single phase nano sized $NiFe_2O_4$ particles using sol-gel auto combustion route by optimizing the preparation conditions, (2008). *Mater. Chem. Phys.* Vol 112. pp. 572-576
- [5] Waje, S.B., Hashim. M., Yusoff W. Dau.W. and Abbas. Z. Sintering temperature dependence of room temperature magnetic and dielectric properties of $Co_{0.5}Zn_{0.5}Fe_2O_4$ prepared using mechanically alloyed nanoparticles, (2010). *J. Magn.Magn.Mater.* Vol 322. pp 686-691
- [6] Raghavende, A.T and Jadhav, K. M. Dielectric properties of Al substituted Co ferrite nanoparticles, (2009). *Bull. Mater. Sci.*, Vol 32, pp 575-578
- [7] Verma, A. and Dube, D.C. Processing of nickel-zinc ferrites via citrate precursor route for high frequency applications, (2005). *J.Am.Ceram. Soc.*, Vol 88, pp 519-523.