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Enhancement in Magnetic and Dielectric Properties of Magnesium Ferrite by Lithium Substitution Applicable for High Frequency Shielding Material

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Abstract—*Magnesium ferrite is a soft ferromagnetic, highly resistive, low electric loss and high Curie temperature material. It is generally useful for microwave devices. In order to further improve its magnetic and dielectric properties lithium is substituted at magnesium site. Composition series $Mg_{1-x}Li_xFe_2O_4$ ($0 \leq x \geq 0.5$) has been synthesized by solid state reaction method. Single phase spinel structure has been confirmed by X-ray diffraction pattern up to x=0.4 lithium substitution. Room temperature magnetization of composition increase from 23 emu/g for x=0.0 to 37 emu/g for x=0.5 lithium substitution. Dielectric constant and loss (tan δ) has been performed for the composition series in the frequency range 20 Hz to 50 MHz. The value of loss tangent decreases with increasing frequency for all compositions. Dielectric constant has been found to increase with Li substitution and is maximum for (x=0.3) composition at low frequency region. Improved value of magnetic and dielectric constant by substitution of Li in magnesium ferrite is of much interest for the application in microwave devices such as isolators, circulators, gyrators and phase shifter.*

Keywords*: $MgFe_2O_4$, XRD, Dielectric, M-H curve.*

1. INTRODUCTION

In the recent years ferrite materials has attracted the interest of researchers due to their interesting magnetic and dielectric properties. Ferrite comprises a large application area including physics, material science and biomedicine. Among all the ferrites magnesium ferrite $MgFe_2O_4$ is versatile material due to its high resistivity and high permeability [1]. Magnesium ferrite covers a large application area including spintronics, microwave absorbers, sensors and electrical devices [2]. Due to low cost high saturation magnetization, high Curie temperature, high resistivity and low eddy current loses they are quite useful for microwave applications [3]. In spinel ferrite, oxygen forms face center cubic (FCC) lattice with divalent cations at tetrahedral (A) and/or octahedral (B) sites. Magnesium ferrite ($MgFe_2O_4$) has an inverse spinel structure with the preference of Mg^{2+} cations mainly on octahedral sites [4].

In the present work we prepared pure $MgFe_2O_4$ and $Mg_{1-x}Li_xFe_2O_4$ ($0 \leq x \geq 0.5$) composition series samples. The purpose of Li substitution is to enhancement in magnetic and dielectric properties for developing microwave application and as a shielding material. For this purpose the composition series was examined under different characterization techniques.

2. EXPERIMENTAL

Sample series of composition $Mg_{1-x}Li_xFe_2O_4$ ($0 \leq x \geq 0.5$) were prepared using solid state reaction method of analytical grade reagents $MgCO_3$, Li_2CO_3 and Fe_2O_3. Stoichiometric ratios of these compounds were wet ground in acetone using an agate mortar with a pastel for 30-40 minutes. The grinded and homogenized powder was then taken into quartz crucible and kept in a box furnace for pre sintering at temperature 800˚C for 8 hours. Pre-sintered powder then pelletized in rectangular shape of dimension 12mm × 7mm × 2mm using a hydraulic press (10 K Ton). The pellets formed were placed for final sintering at a temperature 1100˚C for 5 hours.

Crystal phase identification and particle size were analyzed by taking X-ray diffraction pattern using Bruker A_x diffractometer of the sample series. Magnetic properties of the sample series were determined using "lake Shore Model 662" Vibrating Sample Magnetometer (VSM) at room temperature.

The dielectric measurement for all the samples were carried out at room temperature using a 4 probe setup on "WAYNE KERR 6500 B precision impedance analyzer" in frequency range 20 Hz to 50 MHz.

3. RESULTS AND DISCUSSION

3.1 X-Ray Diffraction (XRD)

The X-ray diffraction pattern for the composition of $Mg_{1-x}Li_xFe_2O_4$ (0.0, 0.2, 0.5) has been taken for crystal structure and phase identification as shown in Fig. 1.

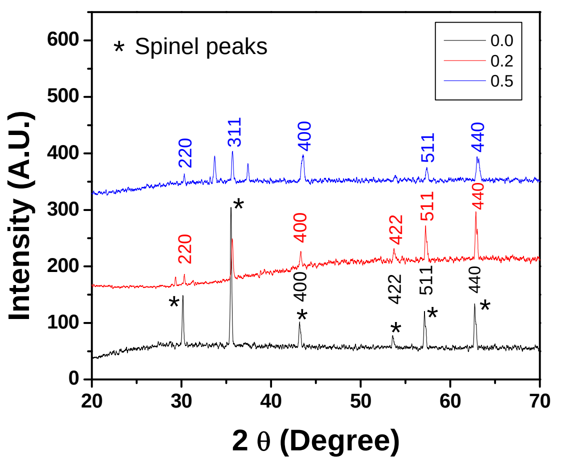

Fig. 1: X-ray diffraction pattern for the composition $Mg_{1-x}Li_xFe_2O_4$ (0 ≤x ≥0.5).

The diffraction pattern shown for the pure $MgFe_2O_4$ (x=0.0) peaks identified as (220), (311), (400), (422), (511), (440) planes corresponds to spinel phase plane matched with JCPDS Card number 73-2211. With Lithium substitution x=0.2 intensity of spinel plane decreased. On x=0.5 lithium substitution at Mg site small peaks of lithium ferrite JCPDS card no. 82-1436 start appearing. Crystallite size of the composition has been calculated with Li substitution using Scherer's eq. 1.

$$D = \frac{0.89\lambda}{\beta_{1/2}\cos\theta} \quad \text{.......... (1)}$$

Where D is average crystallite size (nm) of the particle, λ is the X-ray wavelength (1.54Å), β is the full width at half maximum (FWHM) of maximum intensity (311) peak and θ is the corresponding angle. . The crystallite size has been calculated as 6.6 nm for x=0.0, 8nm for x=0.2 and 7.3 nm for x=0.5 composition. The lattice constant has also been calculated for the sample x=0.0, 0.2and 0.5 using the formula for cubic crystal system as follows:

$$\frac{1}{d^2} = \frac{h^2 + k^2 + l^2}{a^2}$$

And the d spacing was found using Bragg's Law

$$\lambda = 2d_{hkl}sin_{hkl}\Theta$$

The value of lattice constant calculated using the above formula was 8.39 for x=0.0, 8.33 for x=0.2 and 8.33 for x=0.5.

3.2 Magnetic Measurement

The magnetic properties of the samples were analyzed by taking hysteresis loop for the series $Mg_{1-x}Li_xFe_2O_4$ (0 ≤ x ≥ 0.5) is shown in the Fig. 2.

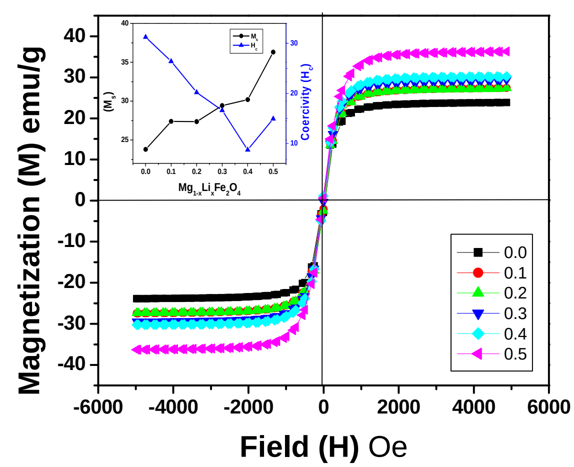

Fig. 2: M-H curves for the composition series $Mg_{1-x}Li_xFe_2O_4$ (0 ≤ x ≥0.5).

For the pure $MgFe_2O_4$ sample the saturation magnetization (M_s) and coercivity (H_c) value is found to be 23.87 emu/g and 31.24 respectively. For the other compositions saturation magnetization M_s values are considerably increased by Li substitution as M_s value increases from 23 emu/g for x=0.0 to 36.30 emu/g for x=0.5 lithium substitution. The increase in M_s value due to substitution of Li suggests the distribution of Fe^{2+} & Fe^3 among octahedral and tetrahedral lattice sites. The coercivity (H_c) value is found to be decrease with increasing Li substitution from 31 Oe (x=0.0) to 14.9 Oe for x=0.5. It may be due to impeding of domain wall motion by Li substitution at Mg site.

In spinel lattice there is parallel alignment of spins in tetrahedral site which is ferromagnetic and spins are align antiparallel at octahedral site shows antiparallel behavior. The combined effect of two sub lattice is ferromagnetic behavior.

3.3 Dielectric Measurements

The dielectric measurement for the samples series were taken at room temperature in frequency range of 20 Hz to 50 MHz as shown in Fig. 3. Silver paste was applied on the sample pellets for making ohmic contacts. The response of capacitance, loss tangent (tan δ) and dielectric constants have been analyzed.

Fig. 3 shows that the value of tan δ is high at low frequency region (100Hz - 1000Hz) and found to decrease exponentially with increasing in frequency.

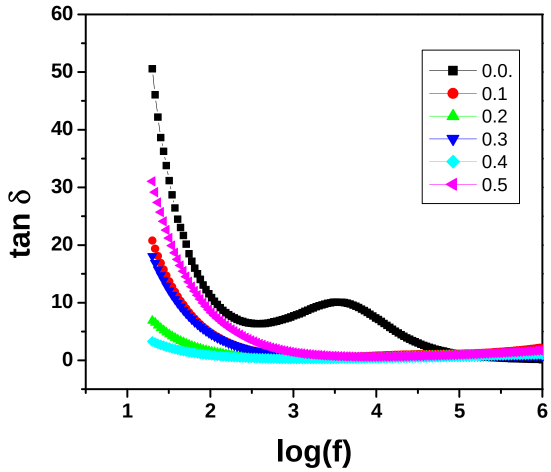

Fig. 3: Variation of loss tangent (tan δ) of $Mg_{1-x}Li_xFe_2O_4$ (0 ≤x ≥0.5) with log(f).

At low frequency grain boundary resistivity plays a major role [5]. It suggests by Li substitution at Mg site reducing the space charge at grain boundary region. Since lithium carbonate has low melting point it acts as liquid phase sintering agent thus activates the boundaries movement [6]. It has been also observed by x-ray diffraction pattern the crystallite size increased with Li substitution. At higher frequency region (≤ 1 KHz) it becomes almost constant with increase in frequency. This behavior of tan δ is similar for all the composition of $Mg_{1-x}Li_xFe_2O_4$ ($0 \le x \ge 0.5$). The value of tan δ is found to be maximum in pure $MgFe_2O_4$ and decreases considerably with increase in Li substitution. The tan δ is minimum for x=0.4 composition. At high frequency hopping between Fe^{2+} and Fe^{3+} at octahedral site is dominant. In addition to this losses in ferrites arises due to other factors like domain wall pinning, hysteresis loss and eddy current loses.

The variation of dielectric constant of $Mg_{1-x}Li_xFe_2O_4$ with frequency at room temperature is shown in Fig. 4. The values of dielectric constant K has been calculated in frequency range 20 Hz to 50 MHz from the following relation:

$$K = \frac{(C \times d)}{\varepsilon_0 \times A}$$

Where C is the capacitance in farad, $\epsilon_0 = 8.854 \times 10^{-12}$ Fm^{-1} is the permittivity of free space, d is the thickness of pellet in meter and A is the cross-sectional area of flat surface of the pellet.

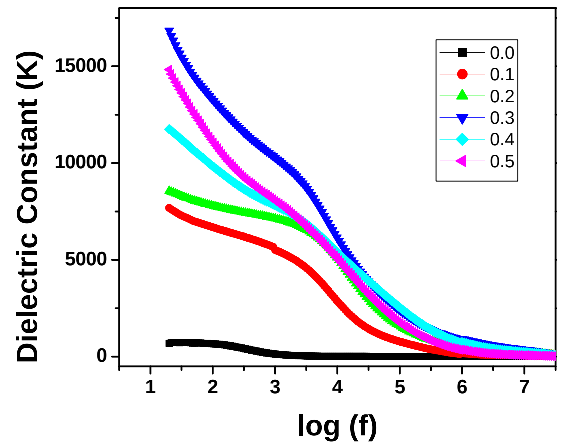

Fig. 4: Dielectric constant K of $Mg_{1-x}Li_xFe_2O_4$ (0 ≤x ≥0.5) vs. log(f) at room temperature.

Dielectric constant of the composition series has been found to increase with increasing Li substitution. At low frequency region dielectric constant is higher due to space charge polarization at electrode/material and grain boundary interface. Diffusivity of small monovalent Li ion is fast and promotes fast nucleation [6]. At high frequency Li substituted magnesium ferrite shows high dielectric constant then pure sample. Lithium prefer to occupy octahedral site thus effect the distribution of Fe^{2+}/Fe^{3+} ratio at octahedral site and improves the hopping of electron. Thus dielectric constant slightly improved with Li substitution at high frequency. The redistribution of Fe^{3+} ions towards tetrahedral site by Li substitution results into increase in ferromagnetism which has been confirmed by M-H loops in Fig. 2.

4. CONCLUSION

A sample series of pure and Li substituted magnesium ferrites has been synthesized by solid state reaction method and studied for structural, magnetic and dielectric properties. Spinel phase formation has been confirmed by XRD. Saturation magnetization of magnesium ferrite has been found to increase with lithium substitution due to redistribution of Fe^{3+} ions in tetrahedral site and improved Fe^{2+} ions at octahedral site. The dielectric constant value is found to be quite high at low frequency regions for the Li substituted compositions which have been due to grain boundary space charge polarization. The loss tangent is high for pure magnesium ferrite which has been decreased by low loss tangent, improved dielectric and saturation magnetization of Li-substituted magnesium ferrite has high potential for the application as shielding material at high frequency.

5. ACKNOWLEDGEMENT

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