

Structural and Magnetization Behaviors of Ni Substituted Li-Mg Ferrites

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Abstract Magnetization behaviors of Ni-substituted $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{Fe}_{2+x}\text{O}_4$ ferrites, where $x = 0.25, 0.20, 0.15, 0.10$ and 0.00 synthesized by standard ceramic technology sintered at 1300°C in air for 5 hours has been presented in the present study. The bulk density and lattice constants of the samples found to be decreased with the increase in the Ni-content for the x values from 0.25 to 0.00. DC electrical resistivity has found to show highest magnitude at room temperature and decreases with further increase in temperature. Magnetization of the samples has been measured as a function of the field using hysteresis loop tracer at 30°C . It was observed that addition of nickel in polycrystalline Li-Mg ferrites plays an important role in modification of structural and magnetization characteristics.

Keywords: magnetization, ceramic technology, bulk density, hysteresis loop, saturation magnetization

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1. Introduction

Properties like high permeability, low loss feature, high stability of permeability with temperature and time, high wear resistance, controlled coercive force, low switching coefficient etc. have appositely placed Li-Mg ferrites as highly demandable ferrites to both researchers and manufacturers [1]. Li-ferrites became commercially important as computer memory core materials in the early 1960's. The high Curie temperature, leading to unparallel thermal stability and high saturation magnetization all prompted this commercial interest. For many of the same reasons, there was considerable development effort aimed at providing microwave quality Li-ferrites [2,3,4,5]. The principal interest in microwave Li-ferrites is a low cost replacement for the rare-earth iron garnets, offering improved temperature performance. Li-ferrites with magnetizations comparable to the garnets are very refractory due to the high concentration and nature of the substitution elements, which requires relatively high sintering temperatures. This type of heat treatment causes the volatility of Li_2O [6] which results in reduction of iron. For this reason, Li-ferrites were considered difficult to prepare with magnetization characterized by high porosities. As a result, high coercive forces and broad resonance line-widths were experienced [7]. The properties of Li-Ni ferrites studied by Bhatu et al. [8] reported an increase in magnitude of elastic constants, elastic wave velocities and infrared spectral analysis is

found easier, valid and suitable for spinel ferrite. The Magnetic studies of Co-substituted Li-Zn ferrites in Ref. [9] reported that cobalt shows anomalous behavior when substituted in Li-ferrites in the presence of Zn [10]. Structural and magnetic properties of Mg-doped $\text{Li}_{0.41}\text{Fe}_{2.41}\text{Mg}_{0.17}\text{O}_4$ ferrites were investigated by Widatallah et al. [11] while the Fe doped NiO ferrites have been reported in Ref. [12]. More recently magnetic, dielectric and electrical properties of La_2O_3 and simultaneous addition of 1 wt% ($\text{CaO} + \text{SiO}_2$) doped Ba-hexaferrites has been studied [13].

Over the years, it has been observed that pure and mixed Li-ferrites are highly potential candidates for microwave applications, especially as replacements for garnets, due to their low cost. The squareness of the hysteresis loop and superior temperature performance are other application in microwave devices [7,8]. Effect of various substitutions in Li-ferrites exhibit very good relaxation and anisotropy properties than other type of ferrites [9,10]. Studies of Li-Mg-Fe spinels show that it can be useful in microwave applications with improved magnetic properties [11,14]. Besides pure Li, Li-Mg and Li-Ni ferrites [6,7,8], many researchers have extensively studied Li-Zn [15,16], Li-Co [17], Li-Cd [18] and Li-Cr [19] ferrites. However, reports on the magnetization behaviors of nickel substituted Li-Mg ferrites are still very little. In view of the great interest of Li-Mg ferrites, both for their technological applications and theoretical understanding of the mechanisms involved, the present work is aimed at finding the effect of Ni-addition on the structural and magnetization behaviors of Li-Mg ferrites.

2. Materials and Method

Ni-substituted $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{Fe}_{2+x}\text{O}_4$ polycrystalline ferrites, where $x=0.25, 0.20, 0.15, 0.10$ and 0.00 were prepared by conventional ceramic technique. The high purity level analytical grade chemical ingredients of NiO (99.9%), MgO (99.9%), Li_2CO_3 (99+%), Fe_2O_3 (99.9%) have been as supplied by the manufacturer. All the raw materials were mixed thoroughly with stainless steel balls in a ball-milling machine with distilled water as fluid. The presintering of the samples was carried out at 900°C for 5 hours in air atmosphere. The presintered samples were milled and pressed at 2 tons pressure into disk shape of 10 mm diameter. A small quantity of polyvinyl alcohol was added as binding material to the powder. The final sintering of the end products was done at 1300°C for 5 hours in air. Elaborate explanations about the specimen preparation technique are available in the literature [13,20-27]. The hysteresis loop measurements of the samples was performed by Automatic Magnetic Hysteresisgraph (Model: HYSTERGRAF IS-300 of Laboratorio Elettrofisico Engineering SRL, Italy) installed at the Materials Science Division, Atomic Energy Centre, Dhaka, Bangladesh. The magnetization versus applied magnetic field measurement has been performed by the Vibrating Sample Magnetometer (VSM), Model: EV9 of Micro Sense, USA at the Materials Science Division, Atomic Energy Centre, Dhaka, Bangladesh. The measurement has been performed by varying the applied magnetic field from -1 to $+1$ kOe. The weight of each sample was approximately 20 mg. The saturation magnetization has been derived from the VSM measurements. Magnetic parameters e.g., coercive field, remnant magnetization, magnetization at the maximum field, etc can be determined from hysteresis loops.

3. Results and Discussion

3.1. Structural Properties of Li-Mg Ferrites

The structural characterization and phase identification of the samples have been done by X-ray diffraction (XRD) technique using Philips X'PERT PRO X-ray diffractometer using Cu-K_α radiation in the $2\theta = 20^\circ$ to 80° geometry in steps of 0.02° . XRD patterns confirmed the single-phase cubic spinel structure of ferrites without having any other intermediate phases. Well-defined diffraction lines along with the crystallization phase also confirm the high purity level of the ferrite powders. XRD data of such measurements have been presented in our earlier publication [26]. The structural characterizations of Li-Mg ferrites have been performed measuring the bulk density, theoretical density, lattice parameters and porosity of the samples. Working principle for the theoretical density, lattice parameters and porosity are given in Ref. [26]. Bulk density and theoretical density of the samples have been presented in Figure 1. According to the data shown in Figure 1, it has been observed that the density of the samples with and without additives observed to vary between 4.47 gm/cc and 4.21 gm/cc. With the increase in Ni-content the the bulk density of Li-Mg ferrite was decreased up to 4.21 gm/cc while the theoretical density has been found to reduce from 5.051 to 4.924 g/cc. The

lattice parameters of the Li-Mg ferrites have been found to be decreased with the progressive addition of the nickel content to the pure samples which has been shown in Figure 2. This variation in lattice parameters is well known to obey the Vegard's law [28]. These new measurements for the samples sintered at 1300°C have been found to be consistent with our earlier study for Li-Mg-Ni ferrites sintered at 1200°C [26]. The decrease in these parameters may be related to the replacement of Li^{2+} ion with larger ionic radius (0.88 Å) by Ni^{2+} ion with smaller ionic radius (0.72 Å). The difference in the lattice parameter may also be explained by the cation migration since Ni^{2+} ions on the A-sites exhibit a smaller ionic radius than on the B-sites because of covalence effects [29]. The remarkable effect is the anomaly shift, due to the redistribution of cations. This behavior is appeared clearly in the physical properties such as density, porosity and magnetic properties. Figure 3 shows that the porosity of Li-Mg ferrites has been found to be increased with the increase in Ni-content to the Li-Mg ferrites. This confirms the fact that the nickel possesses the higher atomic weight than that of lithium and iron. It is also believed that the replacement of Mg^{2+} ion with smaller ionic radius (0.75 Å) by Ni^{2+} ion with larger ionic radius (0.77 Å) may be responsible for the increase in porosity with the increase in Ni-content. Elaborate explanations about the variation in bulk density, theoretical density, lattice parameters and porosity is available in Ref. [26].

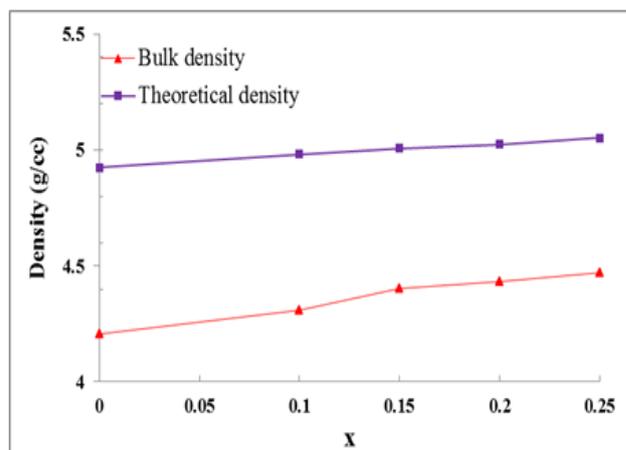


Figure 1. Bulk density and theoretical density of $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{Fe}_{2+x}\text{O}_4$ ferrites sintered at 1300°C in air for 5 hours

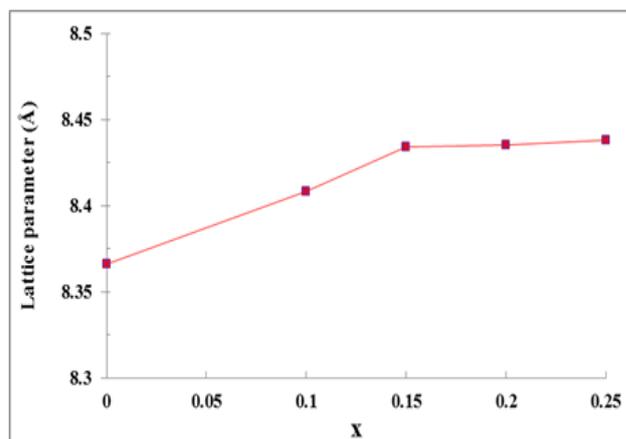


Figure 2. Lattice parameters of $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{Fe}_{2+x}\text{O}_4$ ferrites sintered at 1300°C in air for 5 hours

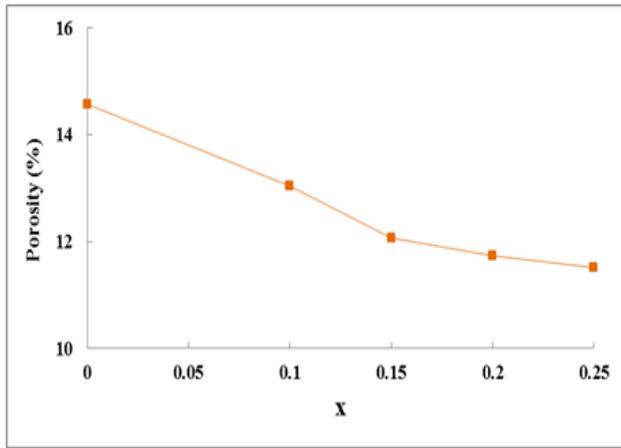


Figure 3. Porosity of $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{Fe}_{2+x}\text{O}_4$ ferrites sintered at 1300°C in air for 5 hours

3.2. DC Resistivity of Li-Mg Ferrites

Figure 4 shows the DC electrical resistivity at room temperature of Li-Mg ferrites sintered at 1300°C in air for 5 hours. From the resistivity data, it is clear that the room temperature DC resistivity of Li-Mg ferrites increases linearly with the increase in Ni-content. DC resistivity data has been found to be consistent with the porosity data (Figure 3) reveals the fact that the rise in resistivity with the increase in Ni-content to the Li-Mg ferrites attributed to the porous nature of this ferrite [30]. It is also believed that a controlled amount of Ni is added to the undoped sample the resistivity because Ni acts as a dead layer over the surface of the Li-Mg ferrites thus reducing the conductivity. It is well established that the electrical conductivity is basically responsible for the exchange of electrons between Fe^{2+} and Fe^{3+} [13,22,23,24,26,27]. The electronic exchange between Fe^{2+} and Fe^{3+} might result in the local displacement of charges and thereby polarization takes place in the ferrites. The magnitude of electronic exchange is dependent on the concentration of the ' $\text{Fe}^{3+}/\text{Fe}^{2+}$ ' ion pairs present on the octahedral B-sites. In ferrites system, the concentration of Fe^{2+} ion depends upon the sintering atmosphere, sintering time and temperature, annealing time and so on. Microstructure of ferrites also play significant role in controlling the electrical resistivity of ferrites as reported in Refs. [31,32,33].

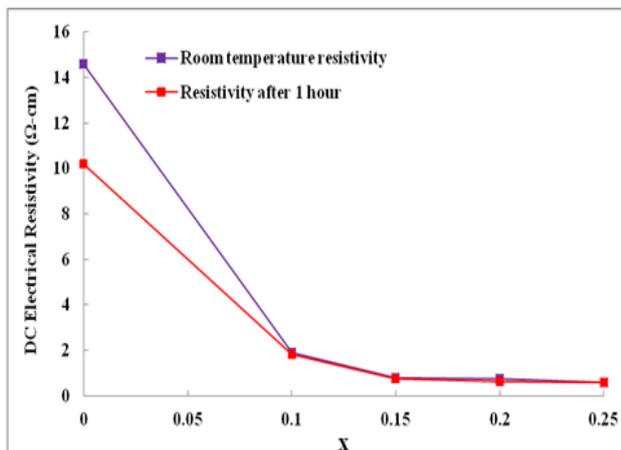


Figure 4. Variation of room temperature resistivity of Li-Mg ferrites sintered at 1300°C in air for 6 hours with content x

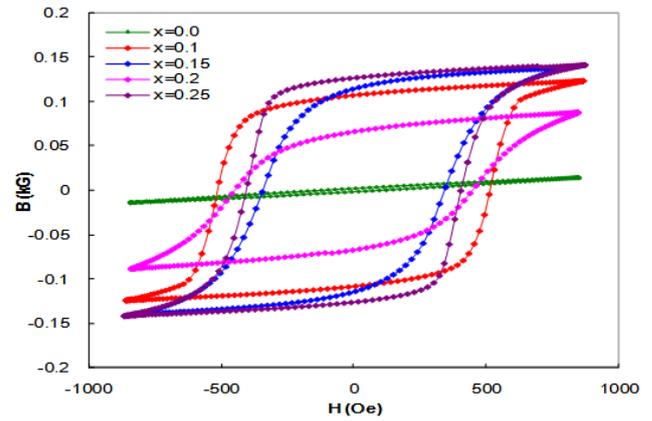


Figure 5. Hysteresis loop for sample $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{O}_4$ for $x = 0.00, 0.10, 0.15, 0.20$ and 0.25

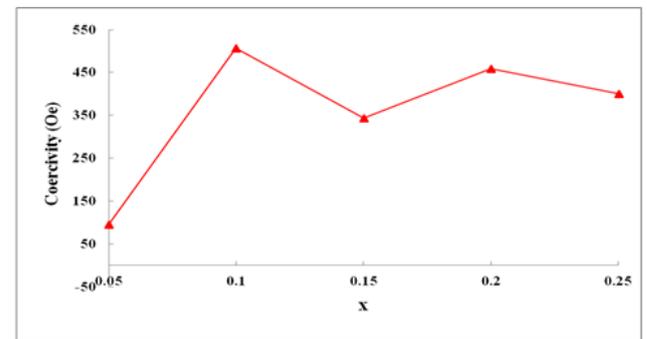


Figure 6. Variation of coercive force of $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{O}_4$ for $x = 0.00, 0.10, 0.15, 0.20$ and 0.25

3.3. Magnetization Behaviors of Li-Mg Ferrites

The lag or delay of a magnetic materials is commonly termed as magnetic hysteresis, relates to the magnetization properties of a material by which it firstly becomes magnetized and then demagnetized. Hysteresis measurements of Li-Mg ferrites with compositions $\text{Li}_x\text{Mg}_{0.5}\text{Ni}_{0.5-2x}\text{O}_4$ where $x = 0.0, 0.10, 0.15, 0.20, 0.25$ were performed by an Automatic Magnetic Hysteresisgraph for a magnetic field ranging from -1000 Oe to $+1000$ Oe are shown in Figure 5. The magnetization of the sample increases linearly with increase in applied magnetic field and then saturation occurs. Using the hysteresis loop presented in Figure 5, the hysteresis parameters viz., saturation magnetization (M_s), coercivity (H_c), remanent induction (B_r) are estimated and presented in Figure 6, Figure 7 and Figure 8. Firstly, with the small introduction of Ni-content the coercivity of the Li-Mg ferrites has been found to be increased and then decreased and increased again with further increase in nickel content as is observed in Figure 6. It is apparent that the softness of Li-Mg ferrites is decreased by the addition of Ni as the remanent induction is increased shown in Figure 7. For $x = 0.00$ the loop tells us that Li-Mg ferrites is a very soft magnet to devise as the remanence and coercive forces both are least. The saturation magnetization of Li-Mg ferrites increases first with the introduction of Ni ions then decreases with the increase in Ni-content for $x = 0.15$ and then increases linearly with the further increase in nickel content which has been shown in Figure 8. The decrease in saturation magnetization is due to the increase in magnetic dilution

of A–B interaction. Since, the Mg^{2+} ions are the same in all the samples, we believe they will not affect the magnetization behavior of our samples. The Li^{1+} ion is nonmagnetic which also does not responsible to the magnetization of the sub-lattice and hence might not impart to the net magnetic moment. Thus, the magnetization behaviour of the ferrite samples arises mainly due to the contributions of the magnetic moments from Fe^{3+} and Ni^{2+} at B-sites [34]. At B-sites, Ni^{2+} ions replace the Fe^{3+} ions and nonmagnetic Li^{1+} ions. Neel's phenomenological model can be successfully applied to explain the increase of saturation magnetization in Li-Mg ferrites with nickel substitution. Neel's model states that the A–B super-exchange interaction is stronger than that of A–A and B–B exchange interaction. The saturation magnetization is given by the vector sum of the magnetic moments of the individual A and B sub-lattices. The decrease of magnetization may be due to the fact that the magnetization of A-sublattice becomes much diluted that the A-B exchange interaction becomes weaker or comparable with the B-B exchange interactions [21]. This gives rise to the canted spins. The existence of canted spin gives rise to Yafet-Kittel (Y-K) angle which compares the strength of A-B and B-B exchange interaction. The decrease of magnetization can be also treated theoretically by triangular arrangement of spins as proposed by Yafet and Kittel according to Ref. [35]. The moderate saturation magnetization values of Li-Mg ferrites with nickel substitution find their potential applications in the high frequency devices [36]. The changes in saturation magnetization, coercivity, and remanence are not linear because of the refractory property of nickel hinders to grain growth as well as the other magnetic properties.

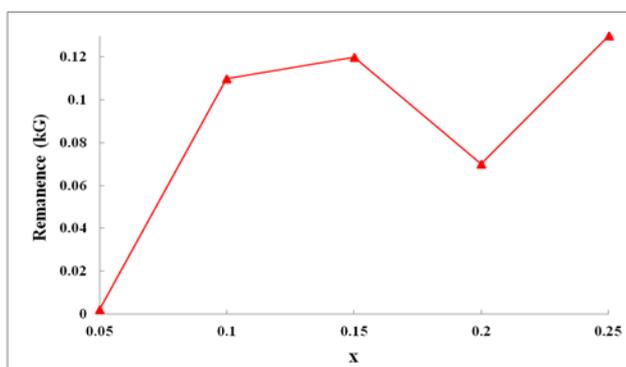


Figure 7. Variation of remanence of $Li_xMg_{0.5}Ni_{0.5-2x}O_4$ for $x = 0.00, 0.10, 0.15, 0.20$ and 0.25

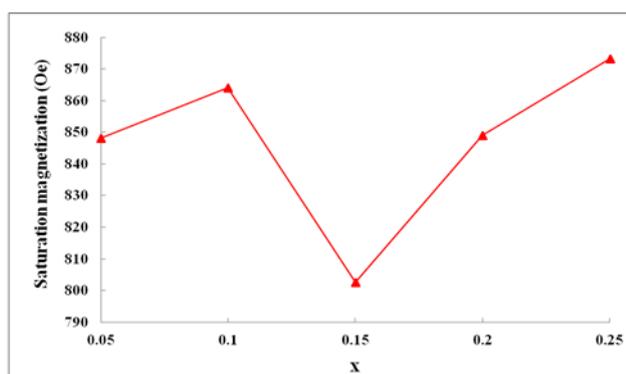


Figure 8. Variation of saturation magnetization of $Li_xMg_{0.5}Ni_{0.5-2x}O_4$ for $x = 0.00, 0.10, 0.15, 0.20$ and 0.25

4. Conclusions

Structural and magnetization characteristics of polycrystalline Li-Mg ferrites have been reported with the addition of Ni-content by replacing Li-content. Throughout the study, it has been noticed that the structural properties of Li-Mg ferrites are improved remarkably with the addition of Ni-content. According to the DC resistivity measurements, it has been observed that at room temperature resistivity is maximum and decreases with increasing temperature. The decrease of magnetization with the introduction of nickel to the pure Li-Mg ferrites might be associated with the refractory behavior of nickel ions. The weakening of the super exchange interaction due to magnetic dilution and disruptions of the collinear spin structure have also found to be responsible for the modification of the magnetization behaviors of the Li-Mg ferrites with the progressive addition of nickel content.

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