Journal of Magnetism and Magnetic Materials 442 (2017) 72–79

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Research articles

Dielectric properties and impedance analysis of polycrystalline Li-Si ferrite prepared by high energy ball milling technique

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ARTICLE INFO

Article history:
Received 7 May 2017
Received in revised form 11 June 2017
Accepted 16 June 2017
Available online 17 June 2017

Keywords:
Ferrite
Dielectric properties
Ac conductivity
Impedance spectroscopy

ABSTRACT

The effect of Si substitution on the ac electrical conductivity, dielectric and impedance properties of $Li_{0.5+0.5x}Si_xFe_{2.5-1.5x}O_4$ ($0 \le x \le 0.6$) ferrite synthesized by high energy ball milling technique has been reported. The ac measurements were performed over a wide range of frequencies from 100 Hz up to 1 MHz and as a function of temperature from room temperature up to 960 K. Ac electrical conductivity as a function of frequency and temperature exhibited a semiconducting behavior. Small polaron hopping conduction mechanism has been observed in all compositions. For the samples with x = 0.0 and 0.1, the correlated barrier hopping mechanism was noticed at first, followed by the small polaron hopping conduction mechanism. The dielectric constant $\acute{\varepsilon}$ and dielectric loss ε'' decrease with increasing frequency and remain constant at higher frequencies, showing the usual dielectric dispersion. The dispersion of the dielectric properties was discussed through Koop's phenomenological theory. The low dielectric loss tangent at higher frequencies makes the opportunity of these ferrites for high frequency applications. The impedance spectroscopy technique was utilized to investigate the effect of grain and grain boundary on the electrical properties of the present series.

1. Introduction

Ferrites assume special significance in the field of electronics and telecommunication industry due to their high electrical resistivity, chemical stability, mechanical hardness and they are available at low cost [1]. Polycrystalline spinel ferrites have very good electric and dielectric properties that are dependent on various factors such as the methods of preparation, substitution of different ions, amount of substitution and sintering temperature, etc. [2,3]. Lithium and substituted lithium ferrites have attracted great interest for their potential microwave applications such as circulators, isolators, phase shifters, etc., which are due to their high resistivity, low eddy current losses and reasonably low costs involved [4,5]. The electrical as well as dielectric properties of lithium ferrites can be changed by substituting them with different metal ions for device applications. Mazen and Dawoud [6] have investigated the effect of frequency, temperature and composition on the dielectric behavior of Li-Cu ferrite synthesized by standard ceramic method. The electrical properties of Li–Mn ferrites have studied by Refs. [7,8]. Abdullah Dar et al. [9] have investigated the preparation and electrical properties of Li-Al ferrite as a function of frequency and composition at room temperature using impedance spectroscopy. Recently, Mazen et al. [10] have reported the effect of silicon substitution and annealing temperature on the structural and magnetic properties of lithium ferrites. However, no report has been found so far in the literature describing the frequency and temperature variation of dielectric properties of Si substituted lithium ferrite. Moreover, there is a requirement for a careful study of dielectric properties of these ferrites possessing desired applications. Therefore, in the present work, influence of Si^{4+} ions substitution on dielectric properties of Li-Si ferrites prepared by high energy ball milling method has been investigated. The structure of the present composition has been investigated by X-ray analysis, IR absorption spectra and scanning electron microscope beside the dc electrical conductivity in our previous work [11].

2. Experimental technique

Li–Si ferrite samples with the general chemical formula $Li_{0.5+0.5x}Si_xFe_{2.5-1.5x}O_4$ ($0 \le x \le 0.6$) were prepared by high-energy ball milling technique. Li_2CO_3, α-Fe_2O_3 and SiO_2 powders were used as raw materials with high purity (99.9 wt%). Stoichiometric amounts of the required powders were mixed thoroughly and then ground into a very fine powder using ball milling machine (Model Fritsch Pulverisatte-6). The milling was executed at room

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http://dx.doi.org/10.1016/j.jmmm.2017.06.085
0304-8853/

temperature by using a planetary ball mill with rotation speed of 100 rpm for milling time of 20 h. The obtained powders were compressed into a disk shape of 13 mm diameter and 3–5 mm thickness, and then finally sintered at 1000 °C for 2 h in static air. Heating and cooling rates were restricted at 5 °C/min and 2 °C/min, respectively. The high energy ball milling technique is simple, does not require high temperature, short duration heating, and gives fine particles [10].

The disk shaped pellets were coated with silver paste to make the parallel plate capacitor geometry for dielectric and impedance measurements. The ac measurements were carried out using the two-probe method over a wide range of elevated temperatures from room temperature up to 960 K using LCR meter bridge (Fluke PM 6306). The ac electrical conductivity, dielectric constant (ε'), dielectric loss (ε'') were measured in the range of frequencies from 100 Hz to 1 MHz. The impedance parameters, namely Z' and Z'' for all the samples, were measured at elevated temperatures from 50 °C up to 250 °C in the frequency range from 100 Hz to 1 MHz using LCR meter bridge (Fluke PM 6306). More details about the measurements were declared in our previous work [12].

3. Results and discussion

3.1. Ac electrical conductivity

The ac conductivity as a function of frequency ($f = 10^2 \rightarrow 10^6$ Hz) at 323 K for the above investigated composition is shown in Fig. 1. It is observed that the ac conductivity increases slowly at lower frequencies, but after a certain frequency ($\approx 10^3$ Hz) it increases instantly and reaches to the highest value at 10^6 Hz. The dispersion in ac conductivity with frequency has been explained by Koop's theorem [13], which supposed that the ferrites compact acts as a multilayer capacitor. It this model, the dielectric structure is assumed to consist of well conducting grains separated by poorly conducting grain boundaries. At lower frequencies, grain boundaries are effective with high resistance giving a small increment of conductivity. At high frequencies, the increase in conductivity is due to the grain effect. The total conductivity in the form of power law is represented by [14]:

$$\sigma_{tot} = \sigma_o\ (T) + A\omega^s, \tag{1}$$

where, σ_o is the dc conductivity, it is temperature dependent and frequency independent function. $A\omega^s$ is the pure ac conductivity due to hopping process at B-sites. 'A' is little dependent on temperature, ω (=$2\pi f$) is the frequency at which the conductivity was measured and the power s, which is a weak function of frequency is to be determined for all compositions [14]. Fig. 2 shows the relation between $\log\sigma$ and $\log\omega$ for the investigated compositions of Li-Si ferrites at 323 K. This relation gives a straight line with slope equal to the power s. It is observed that the power s is composition dependent, where s increases with increasing Si content up to x = 0.5 and then decreases at x = 0.6 as shown in the set of Fig. 2. It can be seen that s varies between 0.23 and 0.35. It was reported by Pike [15] and Elliot [16] that s values vary between 0 and 1. For s = 0, the electrical conduction is frequency-independent or dc conduction meanwhile for s > 0 the conduction is frequency-dependent [17].

The variation of ac conductivity with the reciprocal of absolute temperature for the above mentioned compositions of $Li_{0.5+0.5x}Si_xFe_{2.5-1.5x}O_4$ was measured at selected frequencies 1 k, 10 k, 100 k, 300 k, 500 k and 1 MHz over a temperature range from 305 to 950 K as shown in Fig. 3. It can be seen that the conductivity shows a metallic-like behavior at lower temperatures ($T < T_m$, T_m was found to vary between 343 and 363 K), where the conductivity decreases with increasing the temperature. This is clearly seen in the samples with x = 0.0 and 0.1 and decreases gradually with the successive substitution by silicon ions. This unusual behavior can be related to the impurity conduction, which plays a main role in this region (relatively low temperature) [14]. This phenomenon has been appeared previously in Li-Ti ferrites [18]. At certain temperature ($T > T_m$), the conductivity begins to increase with increasing temperature indicating the semiconductor behavior as commonly seen in most ferrite samples. The increase in conductivity with temperature may be attributed to the increase in drift mobility and hopping frequency of charge carriers. The conductivity at higher temperatures seems to be frequency independent, while a remarkable dispersion was observed at lower frequencies.

Furthermore, in the semiconductor behavior region the change of $\ln\sigma$ with $10^4/T$ seems likely to exhibit two linear regions (I and II) with different slopes and hence change in the activation energy

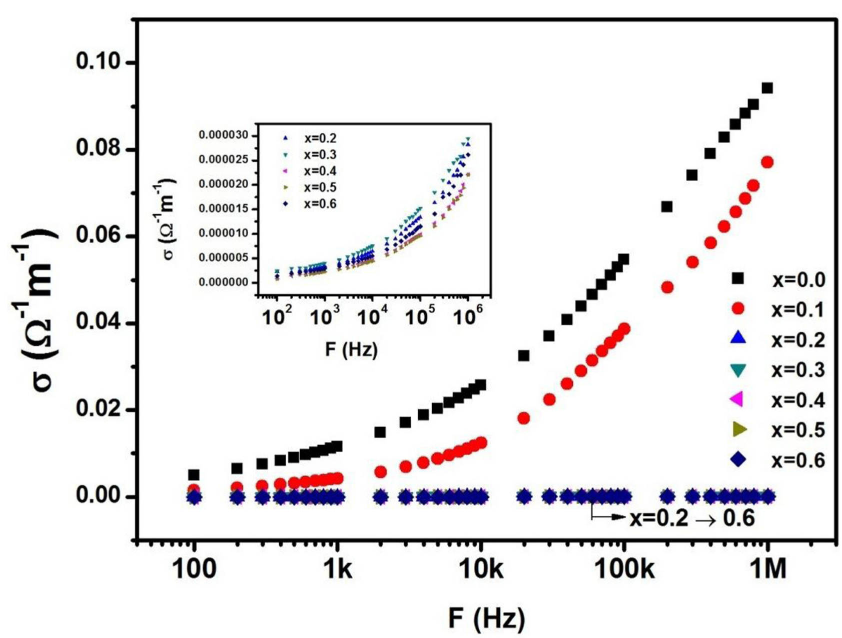

Fig. 1. The variation of the ac conductivity with frequency for $Li_{0.5+0.5x}Si_xFe_{2.5-1.5x}O_4$ ($0 \leq x \leq 0.6$).