Study of the Effect of Rare-earth Oxide Addition on the Magnetic and Dielectric Properties of Sr-hexaferrites

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***Abstract*-- Magnetic and dielectric properties of M-type Sr-hexaferrites of composition $(SrO)_{1-x}$ $(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 prepared by conventional ceramic technology at different sintering temperatures using magnetite of Cox's Bazar beach sand, Bangladesh have been investigated in the present work. Investigations have been carried out by permeability, loss tangent, quality factor and dielectric constant measurements of various samples of Sr-hexaferrites with the addition of La_2O_3. In our entire study, magnetic and dielectric properties of Sr-hexaferrites have been found to be strongly dependent upon the additives, temperature and frequency.**

***Index Term*-- Magnetite, hexaferrites, permeability, loss tangent, quality factor, dielectric constant.**

I. INTRODUCTION

Hexagonal ferrites fall in the category of hard magnetic materials. Now a day, hard ferrites pervade almost every sphere of modern technology. Hard ferrites play heavy role in electronic industry, electronic information industry, car industry, motor cycle industry etc, mean while, they are also widely used in medical treatment, mining and metallurgy, industrial automation, oil industry and civil industry [1]. They are broadly used in the cores of inductors and transformers. Ferrites are also used as memory core devices in computers, cassettes and videotapes. Since their discovery ceramic materials with hexagonal structure have always enjoyed a significant market share thanks to their main applications in the DC motors for the automotive industry and consumer goods. Cores of inductors use ferrites. To control and minimize various loss factors, ferrites become the very best inductor cores having high initial permeability and attractive small physical size. To fulfill the demands, hexagonal ferrites with optimum magnetic properties have always been developed. Work based on partial substitutions with rare-earth elements and transition metals [2-4] have been considered as the most significant one for the Sr-hexaferrites. The improvement of magnetic properties of hexaferrites arises from a drastic change of the magneto-crystalline anisotropy [5].

Various efforts have been made by researchers to improve the structural and magnetic properties of M-type hexaferrites because of their good energy product and the best performance to cost ratio incorporating cationic substitution [6]. Liu et al. [7] prepared *La*-substituted M-type *Sr*-hexaferrites and reported that the suitable amount of La^{3+} substitution remarkably increase saturation magnetization M_s and intrinsic coercivity H_c. After several decades of applications, hexaferrites cover more than 50% of the permanent magnet market. Rare-earth based permanent magnetic materials have been found to be chemically more stable and hence any improvement of the properties is of relevance for the large market of course. The substitution of small amounts of *SrO* by La_2O_3 in the *Sr*-ferrites has been reported to enhance their magnetic properties [8-12].

In the present study, locally available raw materials (magnetite) which are abundant in the beach sand of Cox's bazar, Bangladesh have been used to synthesize *Sr*-hexaferrites. Our aim is to look on the possibility of developing Sr-hexaferrites using magnetite from Cox's Bazar beach sand. It is also expected that the magnetic and dielectric properties of Sr-hexaferrites might also increase significantly with the addition of La_2O_3. Addition of La_2O_3 would play an important role as grain refiner.

II. EXPERIMENTAL DETAILS

Magnetite collected from the beach sand of Cox's bazar, Bangladesh has been used as the raw materials. In 1961 a geological survey shows Cox's bazaar beach sand are enriched with heavy minerals like magnetite, hematite, rotile, zircon etc. A magnetic separator has been used to collect the magnetite from the beach sand. The purity level of the magnetite was 97%-97.3% [13]. Synthesis of hard ferrites with desired properties requires delicate handling and cautions approach. It is always a complicated job. Control of the chemical composition, homogeneity and microstructure are very crucial. Ferrites are not completely defined by its chemical and crystal structure but also require knowledge and control of parameters of its microstructure such as density,

lattice parameter, porosity and their intra- and intergranular distribution.

Using ball milling technology, we have prepared *Sr*-hexaferrite samples with the following compositions: $(SrO)_{1-x}(La_2O_3)_x 5.7Fe_2O_3$, where x = 0.00, 0.04, 0.08. 100 gm of *Sr*-hexaferrite powders were prepared using $SrCO_3$, La_2O_3 and Fe_3O_4 with appropriate amount of the constituents. Overall synthesis process could be described by the following four major steps [14]: preparing a homogeneous mixture of the materials, prefiring the mixture to form ferrites, conversion of raw ferrite into powders and pressing into the desired shapes, and sintering to produce the final products. The finished products were sintered at a constant temperature of 1235 ^{0}C in air for 4 hours.

III. RESULTS AND DISCUSSION

Fig. 1 shows the real part of the complex initial permeability of Sr-hexaferrite samples of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 as a function of frequency sintered at 1235 °C in air for 2 hours. Complex permeability has been measured using 3255B Precision Magnetic Analyzer that provides a 2-terminal measurement of inductors and transformers over the frequency range 20 Hz to 500 kHz at room temperature using conventional technique based on the determination of complex impedance of circuit located with toroid shaped sample. It is clearly evident from the permeability curves that with addition of La_2O_3 the permeability of the Sr-hexaferrite increases comparing with the undoped one. The general characteristic of the curves is that the real part of the initial permeability remains fairly unaltered on frequency throughout the frequency range considered for the measurement process. It is well known that the permeability of polycrystalline ferrites is determined by the superposition of two magnetization mechanisms, i.e., spin rotation and domain wall motion. As the density of the materials increases at a high sintering temperature the grain size as well as the permeability of the samples might have been increased, which appears to be connected with an increase in the contribution from the domain wall displacement. Permeability of ferrites depends on several other factors e.g., reversible domain wall displacement, domain wall bulging, microstructure, intragranular porosity, chemical composition, average grain size and the presence of second phase etc [15]. Generally, at high sintering temperature and frequency, permeability of ferrites decreases due to the decrease in average grain size which in turn causes to the removal of the domain wall pinning sites at the grain boundary.

Fig. 2 shows the variation of loss tangent of Sr-hexaferrites with and without additives as a function of temperature. According to the data, it is noticed that the loss tangent increases with the increase in temperature which is general characteristic of hard magnetic materials. It is also observed that the loss tangent decreases with the addition of La_2O_3 to pure *Sr*-hexaferrites. This decrease in loss factor might have been related to the fact that additives cause to increase the uniaxial magnetocrystelline anisotropy which in turn causes to decrease the loss tangent. The frequency dependence of loss tangent presented in Fig. 3 shows that loss tangent decreases rapidly with increasing frequency. At high frequency region, the loss tangent is almost remains invariant. Loss tangent in ferrites arise due to various domain defects, which include non-uniform and non-respective domain wall motion, domain wall bowing, localized variation of the flux density and nucleation and annihilation of domain walls [16].

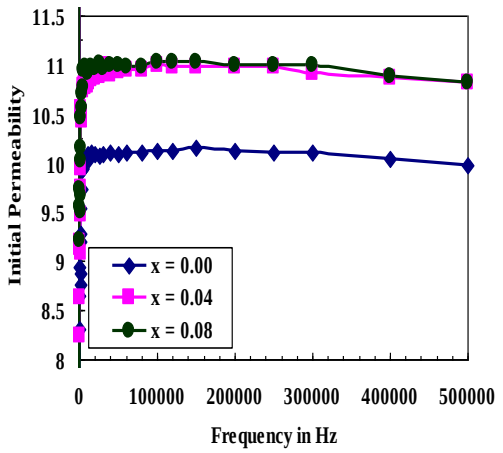

Fig. 1. Real part of permeability Vs frequency of Sr-hexaferrites of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 sintered at 1235°C.

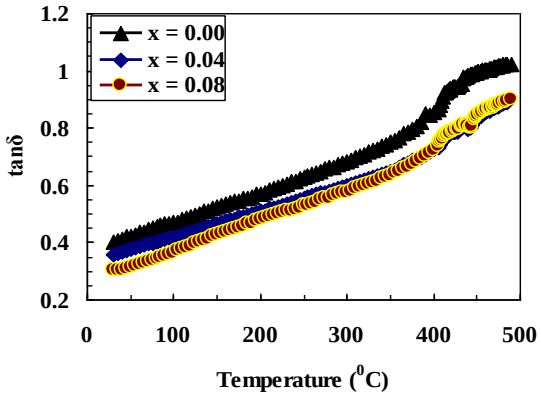

Fig. 2. Temperature dependence of loss tangent of Sr-hexaferrites of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 sintered at 1235°C in air for 2 hours.

Energy loss in ferrites is related to various kinds of defects such as non-uniform and non-repetitive domain wall motion, localized variation of the flux density, domain wall bowing, nucleation and annihilation of domain walls [17]. It is also noted that losses in ferrites also connected to the several other factors: eddy current loss, hysteresis loss, and residual loss [18]. Eddy current losses are minimized by increasing the resistivity of ferrites in conjunction with the slow cooling rate [18] that reduces the trapping of pores within grains. The hysteresis losses arise due to the irreversible wall

displacements [19] while the residual losses have been found to arise from the domain walls relaxations [19].

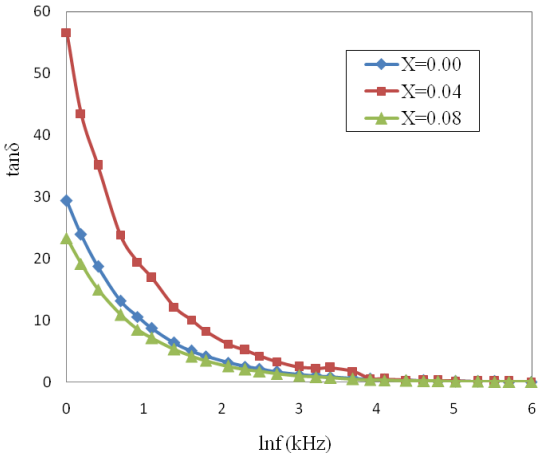

Fig. 3. Frequency dependence of loss tangent (tanδ) of Sr-hexaferrites of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 sintered at 1235°C in air for 2 hours.

The temperature dependence of the effect of *La*-addition on *Q*-factor of *Sr*-hexaferrites sintered at 1235 ^{0}C in air for 2 hours have been presented in Fig. 4. It has been noticed that the *Q*-factor of Sr-hexaferrites decreases with the increase in temperature signifying that the samples work better as permanent magnetic materials at lower temperature region. Fig. 5 shows the frequency dependence of *Q*-factor of the same samples. According to the graphs, it is found that *Q*-factor increases with the increase in frequency. At low frequency region the *Q*-factor increases slightly, but at high frequency it increases sharply.

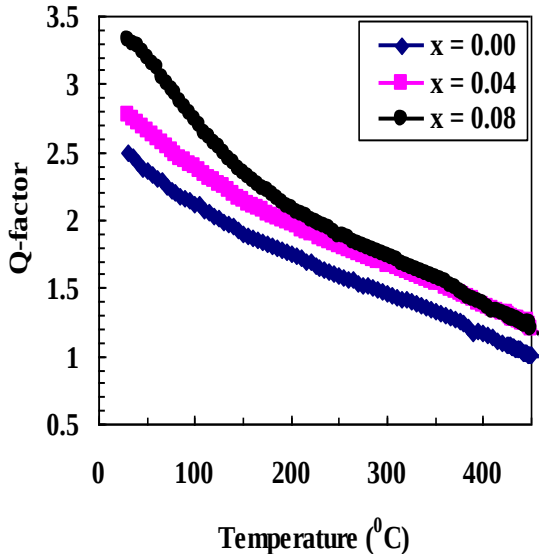

Fig. 4. Temperature dependence Q-factor of Sr-hexaferrites of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 sintered at 1235°C in air for 2 hours.

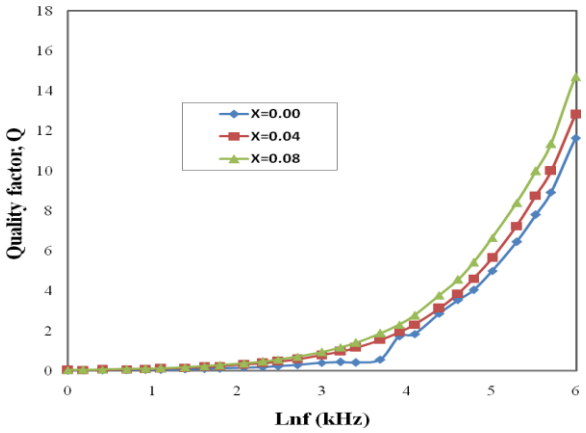

Fig. 5. Frequency dependence Q-factor of Sr-hexaferrites of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 sintered at 1235°C in air for 2 hours.

Dielectric measurement of Sr-hexaferrites as a function of frequency in the range of 1 kHz - 600 kHz at room temperature has been carried out using 3255 Precision Magnetic Analyzer in conjunction with a laboratory made furnace which maintain the desired temperature with the help of a temperature controller. The results of this measurement have been presented in Fig. 6. As the frequency increases the dielectric constant decreases, which is rapid at lower frequencies and slower at higher frequencies. At much higher frequency the change becomes very small and above 250 kHz it becomes constant. This is a normal dielectric behavior observed in most of the ferromagnetic materials, which may be due to the interfacial polarization reported by Maxwell and Wagner [20]. The dielectric behavior of ferrites may be explained on the basis of dielectric polarization process, which is similar to that of the conduction mechanism that takes place mainly by the hopping of electrons. According to Ref. [21] electrical conductivity in ferrites is due to hopping of electron between ions of the same element present in more than one valence state and distributed randomly over crystallographically equivalent lattice sites. The observed decrease in dielectric constant might be attributed to the fact that above certain frequencies the electronic exchange between Fe^{2+} and Fe^{3+} ions becomes independent of frequency of the applied ac electric field. It has been also noticed that the decrease in ε' becomes slow at the higher frequency region from 100 kHz to 500 kHz. The decrease in dielectric constant with the increase in frequency is a normal characteristic of hexagonal ferrites.

IV. CONCLUSIONS

Influence of rare-earth element (La^{3+}) on the magnetic and dielectric properties of *Sr*-hexaferrites have been discussed in present work. Single phase M-type hexagonal structures, homogeneity and purity level of the ferrite powders have been confirmed by X-ray diffraction study of our samples. Over the frequency range of observation, the permeability has been found to be almost constant which means that no structural

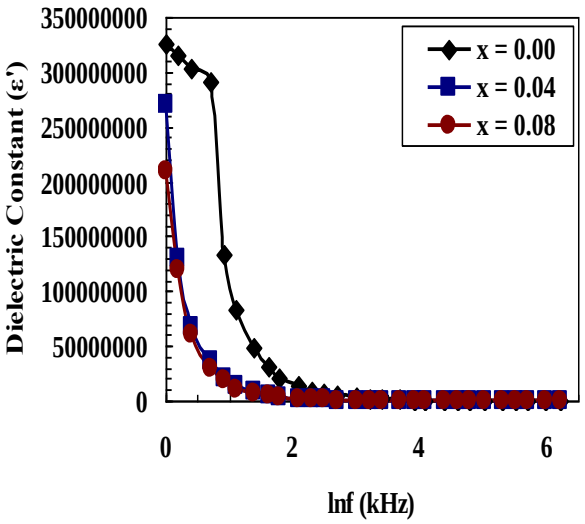

Fig. 6. Frequency dependence dielectric constant of Sr-hexaferrites of compositions $(SrO)_{1-x}(La_2O_3)_x$ 5.7 Fe_2O_3, where x = 0.00, 0.04, 0.08 sintered at 1235°C in air for 2 hours.

relaxation or resonance has been occurred in the domain walls of hexaferrites. Loss tangent has high value at low frequency region and decreases with increasing frequency. At low frequency region, the quality factor increases slightly, however at high frequency side it increases sharply. The observed decrease in dielectric constant with increase in frequency is due to the fact that above certain frequencies the electronic exchange between Fe^{2+} and Fe^{3+} ions cannot follow the frequency of the applied ac field. Dielectric constant decreases rapidly with frequency at law frequency region while in the high frequency region becomes approximately constant. Rare-earth oxides are used to act as grain the refiner. Addition of La_2O_3 might causes to enhance the magnetocrystalline anisotropy of the hexaferrites resulting in new magnetic interactions which in turn causes to improve the magnetic properties.

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