International Journal of Engineering Research-Online
A Peer Reviewed International Journal
Email:editorijoer@gmail.com http://www.ijoer.in

Vol.4., Issue.4., 2016
(July-August)

RESEARCH ARTICLE

ISSN: 2321-7758

EFFECT OF Sm^{3+} AND La^{3+} ADDITION ON DC ELECTRICAL RESISTIVITY AND CURIE TEMPERATURE OF NANOCRYSTALLINE Mg-Cd FERRITES

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ABSTRACT

The ferrites samples with the general formula $Mg_{1-x}Cd_x Fe_2O_4$ + 5wt% R (R= Sm^{3+} and La^{3+}) (x = 0, 0.2, 0.4, 0.6, 0.8 and 1) were prepared by oxalate co-precipitation method. The XRD confirms the formation of cubic spinel structure with orthoferrite secondary phase. The grain size (0.36 to 0.99 μm) increases with increase in Cd^{2+} content and decreases with Sm^{3+} and La^{3+} addition in Mg-Cd ferrites. The DC electrical resistivity increases and the Curie temperature (Tc) decreases with increase in cadmium content (x). Activation energy in paramagnetic region is higher than that in ferromagnetic region. DC resistivity of La^{3+} added samples is higher than Sm^{3+} added samples and it is also higher than for Mg-Cd ferrites.

Keywords - DC resistivity, Rare earth ion, Curie temperature, Activation energy,

INTRODUCTION

Nanosize magnetic particles have important technological application. Magnesium and cadmium ferrite have important electrical and magnetic properties and applications such as humidity and gas sensors [1-4]. The electromagnetic properties of spinel ferrites can be tailored by controlling the type and amount of additives. The rare earth ions are becoming the promising additives for the improvement of ferrite properties. The addition or substitution of rare earth ions into spinel Mg-Cd ferrite can improve their structural, electrical and magnetic properties [2-5]. Many researchers have studied the influence of various rare earth doping ions on the properties of ferrites [3,6-8]. The results of these researches show that different rear-earth ions behave differently in spinel ferrite. Some literature reported the decrease in lattice constant, grain size, density and resistivity of ferrites with rare earth addition. On the other hand, others reported its increase.

In the present work, co-precipitation method were used for synthesis of Sm^{3+} and La^{3+} added Mg-Cd ferrites and the effect of rare earth ions on dc electrical resistivity and Curie temperature was studied.

EXPERIMENTAL DETAILS:

Polycrystalline ferrites having general formula $Mg_{1-x}Cd_x Fe_2O_4$ (0, 0.2, 0.4, 0.6, 0.8 and 1) with 5wt% addition of Sm^{3+} and La^{3+} were prepared by the oxalate co-precipitation method using high purity sulphates of AR grade. The detail experimental procedure is explained elsewhere [2]. The X-ray diffraction (XRD) patterns were obtained at room temperature on Philips PW-3710, in 2θ range 20-80^o using CuK_α radiation (λ= 1.5424 A^o). The micrographs of fractured surface of the pellets were taken on a scanning electron microscope (SEM) (JEOL-JSM 6360 model, Japan). DC electrical resistivity (ρ) measurements of all the samples were carried out by using two probe method in the temperature range of

300-800^{o}K. Curie temperature was measured by modified Loria Sinha method and also from DC resistivity measurements.

RESULTS AND DISCUSSION

Characterization: The typical XRD of Sm^{3+} and La^{3+} added $MgFe_2O_4$(x = 0) is shown in Fig. 1. All the samples confirm a cubic spinel with orthoferrite as secondary phase. The characteristic peaks belong to the (*Fd3m*) cubic spinel space group. XRD analysis shows that the R ions containing Mg-Cd ferrite have besides the spinel phase as major phase, crystalline secondary phases, identified as orthoferrite ($RFeO_3$) in very small amount. Similar orthoferrite phase was reported in case of Gd^{3+} substituted Mg-Cd ferrites [9]. The crystallite size lies in the range 28.69 to 32.66 nm. The SEM microphotograph of Sm^{3+} and La^{3+} added $CdFe_2O_4$ (x = 1) is shown in Fig. 2. The average grain size of the sample is calculated by linear intercept method and lies in the range 0.36 to 0.99 µm. The SEM study shows reduction in grain size due to formation of secondary phase, i. e. the grain size is smaller than unadded Mg-Cd ferrites [2].

DC Resistivity: The conductivity in ferrites occurs mainly due to hopping of electrons between ions of the same element existing in different valence state, distributed over crystallographically equivalent lattice sites. In the investigated ferrites, Fe^{2+} ions were created during sample preparation providing an increase in electron hopping between the Fe ions in B sites in 2+ and 3+ valence states.

The dc electrical resistivity (ρ) of the samples under investigation has been measured in the range of 300-800^{0}K. The variation of log ρ against $10^3/$ T for all the samples is shown in Fig. 4 (Sm^{3+}) and Fig. 5 (La^{3+}). It is observed that the electrical resistivity of all the samples decreases with increasing temperature, suggesting the semiconducting nature of the samples. The decrease in resistivity with increase in temperature is found to follow the Arrhenius relation [9].

$$\rho = \rho_o e^{(\Delta E/KT} \qquad (1)$$

where, ρ_o is the pre-exponential factor with the dimensions of Ω cm, K-is the Boltzman constant (8.617×10^{-5} eV/K), ΔE-is the activation energy and T-is the absolute temperature.

From the Fig. 4 and 5, it can be seen that the resistivity decreases with increasing temperature. The decrease in electrical resistivity with temperature may be related to the decrease in drift mobility of thermally activated charge carrier (electron and hole) according to the hopping conduction mechanism. At low temperature, where the resistivity has the highest value and the grain boundary effect is dominant, more energy is required for exchange electrons between Fe^{2+} and Fe^{3+} ions located on the grain boundaries. From figure it is also known that the resistivity increases with increase in Cd^{2+} content for sample under investigation. The pure $MgFe_2O_4$ pellet has the lowest resistivity, which can be increased with the substitution of Cd^{2+}. It is known that Mg^{2+} ions occupy tetrahedral A and octahedral B - sites, then Fe^{3+} ions move from B to A sites. As a consequence, the probability of electron hopping between Fe^{3+} and Fe^{2+} at the B sites decreases, resulting in an increase of resistivity.

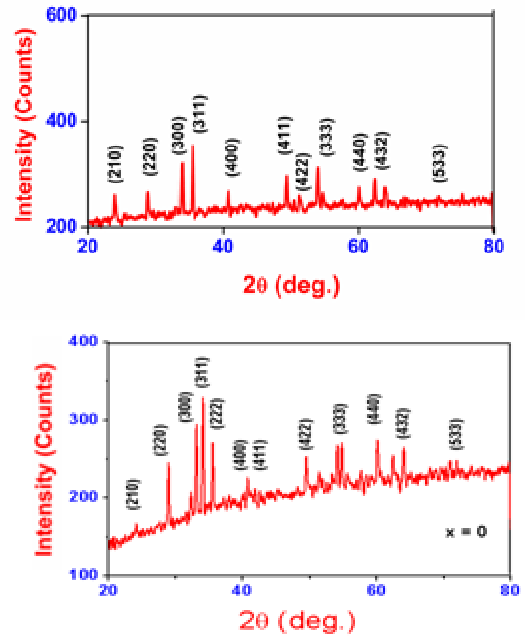

Fig. 1 Typical XRD patterns of 5wt% Sm^{3+} and La^{3+} added $MgFe_2O_4$

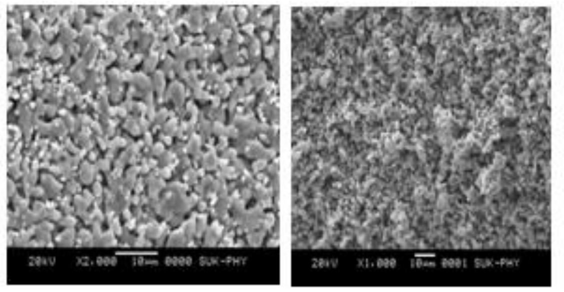

Fig. 2 Typical SEM photographs of 5wt% Sm^{3+} and La^{3+} added $CdFe_2O_4$

The addition of R ions (Sm^{3+} and La^{3+}) also increases resistivity, because R ions have a high preference for octahedral B sites substituting the Fe^{3+}. Therefore the number of Fe^{3+} ions at B sites decreases, the probability of hopping is reduced and thus resistivity increases. Due to low processing temperature, these samples possess smaller grain size (nanocrystallite) and consequently a greater number of grain boundaries. The grain boundaries are the regions of mismatch between energy states of adjacent grains and hence act as a barrier to the flow of electrons [9,10]. This phenomenon leads to the high value of resistivity for the samples under investigation in this work and much higher than those reported for Mg-Cd ferrites prepared by the conventional ceramic method [9,11]. Similar results are reported by Satter *et al* [12] for rare earth ions substituted Cu-Zn ferrite, Stojanovic *et al* [13] for Y^{3+} substituted Ni-Zn ferrites.

The phase transition from ferri to para region of all the sample shows break in resistivity near Curie temperature. It is temperature at which disordering of magnetic spin is completed and the material undergoes, phase transition from ferromagnetic to paramagnetic state. The activation energy (ΔE) in ferri and para regions has been calculated from the plot of log ρ against $10^3/T$. In all the samples the activation energy in paramagnetic region is greater than that in ferrimagnetic region [10]. The activation energy decreases with increasing cadmium content in the Mg-Cd ferrites. This change in activation energy for different compositions is attributed to the hopping of electron. The activation energy of Sm^{3+} and La^{3+} added ferrites in both the region are higher than those corresponding to pure Mg-Cd ferrites.

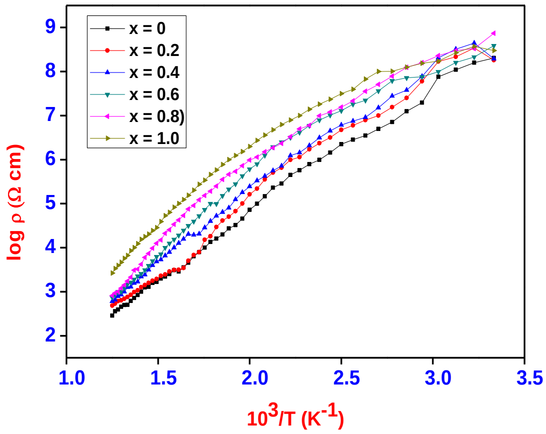

Fig. 3 Variation of log ρ versus 103/T for 5% Sm^{3+} added $Mg_{1-x}Cd_x Fe_2O_4$

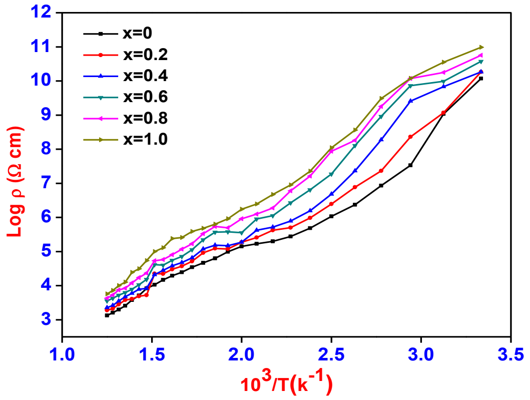

Fig. 4 Variation of log ρ versus $10^3/T$ for 5% La^{3+} added $Mg_{1-x}Cd_x Fe_2O_4$.

Curie Temperature: The Curie temperatures (T_c) are measured by modified Loria Sinha [14] method. Fig. 5 and 6 shows variation of Curie temperature from dc resistivity and Loria Sinha method with cadmium content. It can be seen that the Curie temperature shows decreasing trend with increasing cadmium concentration. This is attributed to occupancy of cadmium on tetrahedral A-site, causing the decrease in A-B interaction (J_{AB}). The addition of rare earth ion lowers the Curie temperature of ferrite due to modification of A-B exchange interaction. For samples with R ions addition, we assumed that R ions reside at grain boundaries. Hence Fe-Fe interactions are still the dominant ones but the number of these interactions is decreased due to the decrease of Fe concentration. Therefore Tc values of the sample with R ions addition are smaller than that of unadded samples. Such reduction in Curie temperature due to rare earth substituted ferrites is reported by [15].

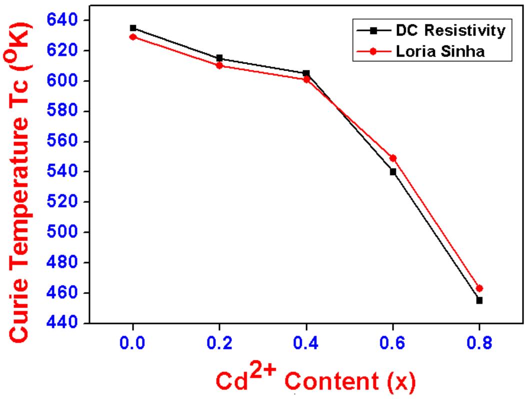

Fig. 5 Variation of Curie temperature (T_c) with Cd^{2+} content (x) and Sm^{3+}

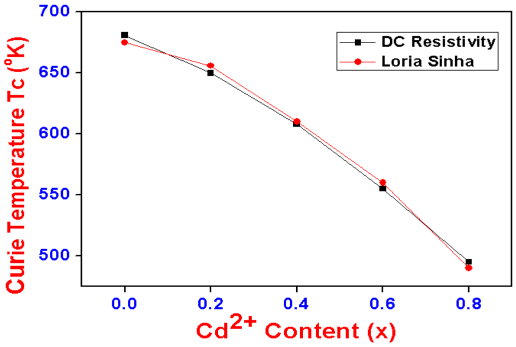

Fig. 6 Variation of Curie temperature (T_c) with Cd^{2+} content (x) and La^{3+}

CONCLUSIONS

The XRD confirms cubic spinel structure with orthoferrite secondary phase. The grain size of Sm^{3+} and La^{3+} added Mg-Cd ferrites is smaller than Mg-Cd ferrites and also prepared by ceramic method. DC resistivity decreases with temperature and increases with Cd^{2+} content. The DC resistivity and Curie temperature of Sm^{3+} and La^{3+} added Mg-Cd ferrites is higher than those reported for ceramic method. The dc resistivity and Curie temperature of La^{3+} added sample is higher than Sm^{3+} added samples because of ionic radius effect. Activation energy in paramagnetic region is higher than that of ferromagnetic region. The Curie temperature decreases with Cd^{2+} content and rare earth ion addition in Mg-Cd ferrite.

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