Structural and Magnetic Properties of La_xSr_{1-x}MnO₃

A. A. Gomaa* and A. A. Mohamed**

* Giza Higher Institute of Engineering and Technology, Basic Science Department, Giza, Egypt. **Physics Department, Faculty of Science, Cairo University, Giza, Egypt. Corresponding author: M.A.Ahmed, Materials Science lab.(1), Phys.Dept.Faculty of Science, Cairo University, Giza, Egypt, moala47@hotmail.com, fax:00202 35693738

Abstract: Samples of $La_xSr_{1-x}MnO_3$ (x = 0.5, 0.55, 0.6, 0.66, and 0.7) were prepared by the citrate-nitrate autocombustion method. The prepared nano-particles were investigated and characterized using X-Ray diffraction (XRD) and Transmission Electron Microscopy (TEM) to confirm the formation of the samples in single phase without any impurities and to calculate the particle size. The magnetic susceptibility χ_M was measured as a function of temperature and magnetic field intensity. From $\chi_M(T)$ and M(H) the saturation magnetization (M_s), remanent magnetization (M_r) and coercivity (H_c) of the samples were estimated. The correlation among physical properties and the ratio of divalent cation substitution was discussed and reported to shed more light on technological applications of the investigated samples.

Keywords: Perovskite; LaSrMnO₃, Nanostructure, Magnetic susceptibility, Hysterysis.

1. INTRODUCTION

The behavior of antiferromagnetic - insulator of the parent compound LaMnO₃ is changed to compounds that can exhibit ferromagnetism and metallic conductivity materials due to the replacement of trivalent La³⁺ ions instead of divalent ions (A²⁺). This will creates a mixed-valence state (Mn³⁺ and Mn⁴⁺)[1–2]. The electrical resistivity of the Perovskite likes lanthanum manganites La_{1-x}A_xMnO₃ (A; Ca, Sr, Ba, etc.) were decreased on application of magnetic field which known as giant magnetoresistive materials (GMR) [3–5]. The manganites are known to be sensitive to chemical composition, such as oxygen deficiency, which affects strongly on the crystal structure and magnetic properties [6]. Different crystal structure spin states and transport properties could be obtained by cation substitution.

The transport and magnetic properties of manganites were explained by the double exchange model which involves the interaction between Mn^{3+} and Mn^{4+} through oxygen anions [7]. Hopping of the d-electrons between the two oxidation States of the Mn^{3+} and Mn^{4+} produces metallic behavior as the materials become ferromagnetic [8–10], giving rise to an insulator–metal transition at temperatures below the ferromagnetic Curie temperature. Understanding of the physical mechanism responsible for the ferromagnetic ordering in manganites remains challenging.

The main objective of the current research is to study a system possesses a fairly large magnetic properties and a high T_C . The study was carriedout for a very important range of compositions $0.5 \le x \le 0.7$.

2. EXPERIMENTAL PROCEDURE

Lanthanum strontium manganites of the formula La_xSr₁₋ $_xMnO_3$ (x=0.5, 0.55, 0.6, 0.66 and 0.7) were prepared by the citrate-nitrate autocombsution method [13, 14]. All chemicals used were obtained in a pure 99.9% from (BDH). Stiochiometric ratio of $La(NO)_3$, $Sr(NO)_3$ and $Mn(NO)_3$ were mixed together with citric acid in ratio of 1:1 from the nitrates and citric acid. The PH value of the mixture was adjusted from 7 - 8. Heating the mixture on a hot plate untill self ingnition takes place and the gray powder was obtained. The obtained nanopowder was sintered at 973 K with a heating rate of 5 C /min for 5hrs. The powder was characterized by X-Ray Diffraction (XRD) to verify the formation of the manganites in single phase without any impurities. The average crystallite size (L) of nanometric lanthanum strontium manganites were calculated using the Debve-Scherer's formula [15]. L= $0.89\lambda/\beta \cos\theta$, where λ is

the wavelength of X-Ray, Θ is the diffraction angle and β is the full width at half maximum (FWHM). The shape and morphology of the fine particles was analyzed using Transmission Electron Microscope (TEM) model (JEOL-1010).

The dc magnetic susceptibility measurements were carried out using Faraday's method, where the measurements were performed from 85K up to 450K in FC (Field Cooling) and ZFC (Zero Field Cooling). Magnetization M Vs. magnetic field (M-H) hysteresis was investigated at room temperature for all samples using VSM model ().

3. RESULTS AND DISCUSSION:

3.1. Structural properties

The data show that the investigated samples were crystallized in single phase pervoskite structure with space group R3c and, All planes in XRD chart were compared and indexed using the International Centre of Diffraction Data (ICDD) card No. (51-0118). The broad XRD peaks confirm of the samples is in nano crystalline form. The tolerance factor for the investigated samples was calculate from the relation

$$t = \frac{R_o + R_A}{\sqrt{2}(R_o + R_B)}$$

Where R_o , R_A , R_B are the ionic radii of oxygen, A and B cations respectively

the radii of La, Sr [16] were take 9-fold coordinate while 12 coordinate of Mn is taken in 4-f.

Figure (2) shows the decreases tolerance factor with increasing x, which means the increase in the distortion of pervoskite structure from the ideal cubic structure, and Mn-O-Mn angle decreases also, therefore the atomic distances were expected to be changed as well. The Transmission electron microscope (TEM) images for the powder samples are shown in Fig. (3a: 3e) where the size of the particles observed are comparable to those from X-ray.

3.2. Magnetic measurements

Figure (4) illustrates the relation between the molar magnetic susceptibility (χ_M) and absolute temperature, ranging from 90 - 420K, at a magnetic field intensity of H (1340 Oe). The results show that, (χ_M) decreases with increasing temperature up to the Curie temperature (T_C) as in Table (1). This reduction in (χ_M) is attributed to the thermal agitation which disturbs the oriented spins. This reduction will continue till it reaches T_C at which the sample behavior is completely changed from ferromagnetic to paramagnetic. This reduction is due to the presence of divalent cations leading to transition of the samples from antiferromagnetic to ferromagnetic state.

Fig. (5) Shows the magnetic moment as a function of the applied field M (H). The data show that, all samples have saturation magnetization and remnant magnetization as well as coercive field. These values are reported in table (2). From the raported data, we can say that, all the samples show ferromagnetic character with small hysteresis area as well as saturation magnetization. For the investigated samples the Curie temperature T_C as in table (1) changed from 331K to 352.5K which was explained in the phase diagram shown Fig.(6).

The values of the Curie temperature of the samples were determined from the first derivative of magnetization curve' [17, 18] as illustrated in the inset Fig. (6). the data revealed a decreased in T_C with increasing La content (x), and returned to increase again.

4. CONCLUSION

LaSrMnO3 samples were prepared by the citrate-nitrate autocombsution method. The single phase of the samples was confirmed by the XRD data. The particle size ranged from 22 to 39 nm.

The values of magnetization parameters, M_s , M_r and H_c , were listed in table (2) and the highest value of Ms was found for the sample 0.6 Conc. of La in LaxSr1-xMnO3. This concentration is considered optimum to be used in technological applications as magnetic sensors, thermistors and transducers.

REFERENCES

- [1] J. Topfer, J.B. Goodenough, J. Solid State Chem. 130(1997) 117.
- [2] R. Mahendiran, S.K. Tiwary, A.K. Raychaudhari, T.V. Ramakrishnan, R. Mahesh, N. Rangavittal, C.N.R. Rao, Phys. Rev. B 53 (1996) 3348.
- [3] W. Cheikhrouh-Koubaa, et al J. of Magn. Magn. Mate. 310(2006) 1481.
- [4] Y. D. Zahoo, et al. J. of Magn. Magn. Mate. 310 (2006) 1481.
- [5] Y. Tokura, Science 312 (2006) 1481.
- [6] Elena Rezlescu, et al. J. Magn. Magn. Mater. 320 (2008) 796- 802.
- [7] C.N.R. Rao, C.R. Serrao, J. Mater. Chem. 17 (2007) 4931.
- [8] J. Rodriguez-Carvajal, FULPROFF 98, Laboratoire Leon Briouillon
- [9] P.G. de Gennes, Phys. Rev. 118 (1960) 141.
- [10] K. Chahara, T. Ohno, M. Kasai, Y. Kozono, Appl. Phys. Lett. 63 (1993) 1990.
- [11] M. Verelst, N. Rangavittal, C.N.R. Rao, A. Rousset, J. Solid State Chem. 104 (1993) 74.
- [12] R. Mahesh, R. Mahendiran, A.K. Raychaudhuri, C.N.R. Rao, J. Solid State Chem. 114 (1995) 297.
- [13] D.D. Sarma, et al., Phys. Rev. Lett. 85 (2000) 2549.
- [14] J. Fontcuberta, et al., J. Magn. Magn. Mater. 290–291 (2005) 974 [14] M.A.Ahmed,S.F.Mansour,S.I.El-Dek,Sol.Stat.Ion181(2010)1149.
- [15] M. A. Ahmed, S. F. Mansour, M. Afifi, J. magnetism and magnetic materials. (2011)..
- [16] S.Ayyappan, J.Philip, B.Raj, Mater. Chem. Phys. 115(2009) 712.
- [17] S. Zemni, A. Gasmi, M. Boudard, M. Oumezzine, Mater. Sci. Eng. B 144 (2007)117–122.

[18] R. Regmi, R. Tackett, G. Lawes, J. Magn. Magn. Mater. 321 (2009) 2296–2299.