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COLOSSAL MAGNETORESISTANCE OF $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}$ MnO_3 PEROVSKITE

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Abstract In this work, colossal magnetoresistance (CMR) of $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ ceramic, with x= 0.00, 0.20 and 0.40 were prepared using the solid-state reaction technique. The structures, surface profile, magnetic and electrical properties of the samples were investigated. The X-ray diffraction (XRD) spectrum indicated that all samples exhibit single phase rhombohedral distorted perovskite structures, which were caused by the Jahn-Teller distortion. Atomic Force Microscopy (AFM) analysis showed no significant change of grain size when Dy was substituted in La site. Magnetization analysis showed that T_c drop as the substitution concentration increased. The highest CMR value of 24.3% and 16.6% at 100K and 300K was observed in sample with x = 0.4 at 1 Tesla. Low field magnetoresistance effect had been observed for x = 0.00 and x = 0.20 sample. However, at high substitution concentration (x = 0.40) LFMR effect tend to vanish.

Introduction

Perovskite rare earth manganites doped with bivalent or monovalent cation of $La_{1-x}A_xMnO_3$ (where A= Ca, Ba or Sr) were intensively studied over the last decade due to its potential technology application as a sensing element. The magnetic and electrical properties of this compound have been examined within the framework of double exchange and Jahn-Teller effect (Tokura and Tomioka, 1999; Dagotto *et al.*, 2001). It is realized that high field is needed to receive a considerable high CMR value and it is too high for practical application. Thus, efforts have been taken to try to bring it down by engineering of heterogeneous structure (i.e., granular materials, multilayer films or ferromagnetic tunnel junction). Most of the investigated single crystal sample and epitaxial thin film (Ping *et al.*, 2006) show a very high intrinsic magnetoresistance effect (up to 1000% changes) near its Curie transition temperature (T_C) but the MR value is almost negligible at far below or above T_C. However, in recent studies, extrinsic low field magnetoresistance (LFMR) had been observed in polycrystalline materials due to the influence of the formation of a small amount of (Ca,La)O rock-salt or non-ferromagnetic (amorphous) layers dispersed in crystalline manganites grain boundaries (GBs) (Nam *et al.*, 2001; Yu *et al.*, 2003). These findings suggest that polycrystalline grain boundaries play a vital role in low temperature MR or LFMR effect (Xia *et al.*, 2002; Zhao *et al.*, 2006;). Hwang *et al.*, (1996) proposed that, at low temperature, a nearly complete spin polarized state causes tunneling through grain boundaries and contributes largely to magnetoresistance in low fields. In this paper, we report the LFMR effect observed in the polycrystalline La-Dy-Sr-Mn-O system.

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Material and Methods

Highly pure La_2O_3, $SrCO_3$, $MnCO_3$ and Dy_2O_3 powder were mixed in acetone media according to the nominal composition $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ (x = 0, 0.20 and 0.40). The powder was prepared via conventional solid-state reaction. The mix powder was heated at 900°C for 12 hrs in air. Later, the obtained powder was grounded, pressed into pellets and sintered at 1300°C for 24 hour in the air. The structure and surface profile was analyzed using powder X-ray Diffraction (XRD) and atomic force microscope. The magnetic properties were measured using Vibrating Magnetometer (VSM). The magnetoresistive properties were measured as a function of temperature (100K to 300K) and applied magnetic field of 1 Tesla using a four-point probe embedded in a liquid nitrogen cryostat.

Results and Discussion

Figure 1 shows the XRD patterns for all samples. As compared to the standard reference of $La_{0.67}Sr_{0.33}MnO_3$ compound (ICDD No.: 50-0308), similar number of peaks had been identified and were indexed as shown in Fig. 1. The result shows that single-phase compound with no impurity had been prepared and the structure was still in rhombohedral system with low concentration of Dy substitution ($x \leq 0.4$) in La site.

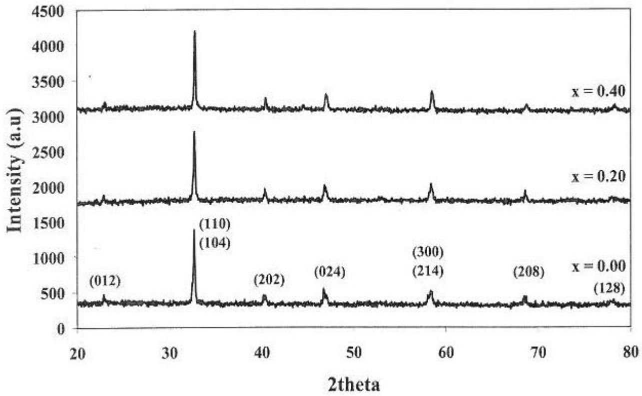

Figure 1. XRD pattern of $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ powder

The surface profile of the bulk sample with the scanning area of 10 μm x 10 μm is shown in Fig. 2. All samples are compact and no obvious pore was observed. No large variation of grain size was observed for of all the samples. The average grain size of 2 to 3 micron was observed for all samples.

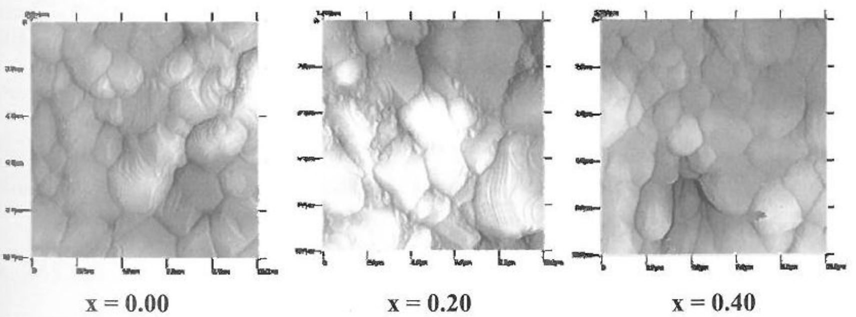

Figure 2. AFM image of $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ compound

Figure 3 shows the magnetization curve measured as a function of temperature ranging from 100K to 300K for all samples. The curve indicates that magnetic phase transition temperature, T_c (Curie temperature) for sample x = 0.20 and 0.40 are 230K and 210K, respectively. From the previous report (Dagotto *et al.*, 2001), T_c for this material dropped at 360K. Therefore, the T_c for sample x =

0.00 cannot be identified due to the limitation of the maximum measurement range (300K). Hence the substitution of Dy at La site will move the T_c to lower temperature.

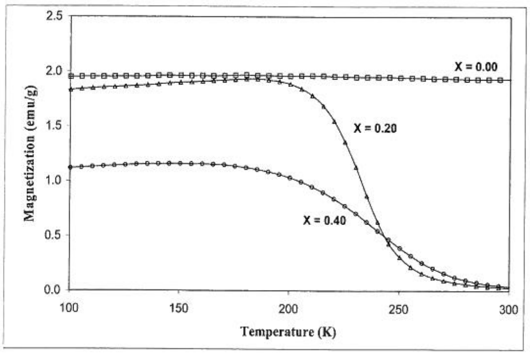

Figure 3. Magnetization as a function of temperature of $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ compound

The CMR curve as a function of magnetic field at various temperatures is shown in Fig. 3. All samples give negative CMR value, which is due to the reduction of the resistance when magnetic field is applied. This phenomenon is caused by the alignment of magnetic moment of localized t_{2g} electron in Mn atom, which lead to the enhancement of the hopping process of e_g electron and hence increases its conductivity. The results also shows that sample x = 0.00 and 0.02 has two regions of linearity at 0 to 0.1T and 0.1T to 1T with different gradient except for sample x = 0.40.

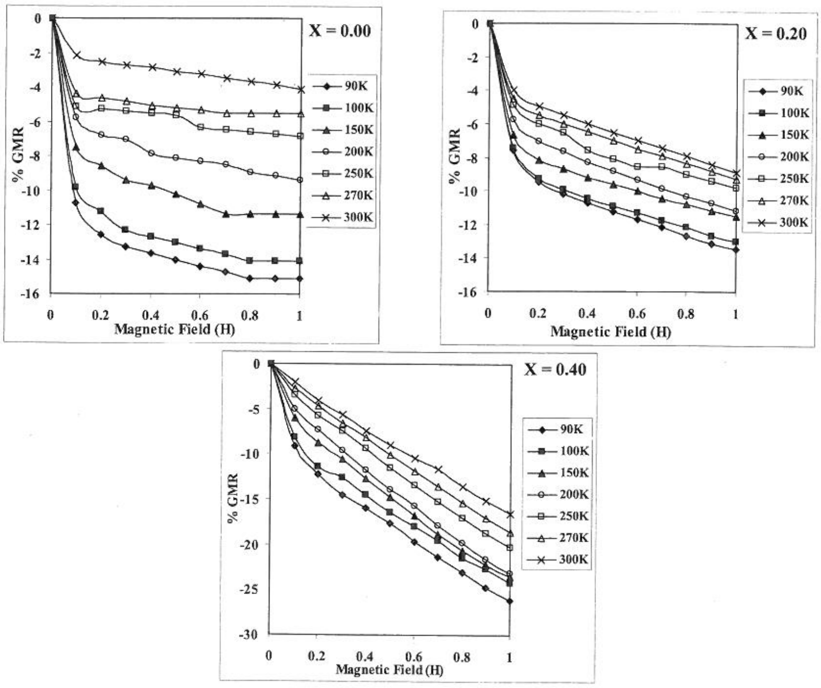

Figure 4. CMR value as a function of magnetic field of $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ compound at different temperature

The sudden drop of the resistance at low applied field is believed to be due to the presence of non-ferromagnetic layer at the grain boundaries. When small field is applied, localize spin moment at the GBs layer tends to align and this encourages the electron hopping. Consequently, there is a significant drop of resistance and hence the magnitude of MR increases dramatically.

Conclusions

In this study, the influence of Dy substitution in La site of $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ compound (x = 0.00, 0.20, 0.40) had been evaluated. The XRD analysis indicated that all samples are in single phase orthorhombic distorted perovskite structure. The surface profile obtained from AFM analysis shows that no significant change of grain size occurred when Dy was substituted. However, the magnetization curve indicated that T_c drop as the amount of Dy substitution increases. Low-field Magnetoresistance (LFMR) effect had been observed for x = 0.00 and 0.20 samples which is believed to be due to the influence of disorder layer at the grain boundaries. At higher substitution concentration (x = 0.40), the LFMR effect tends to vanish. This LFMR effect is one of the properties that is provided by polycrystalline compound. The highest CMR value of -16.6% (at 300K) and 24.3% (at 100K) had been observed for sample x = 0.40 even though this sample did not show any LFMR effect.

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