

**LOW-FIELD MAGNETORESISTIVE AND MAGNETIC PROPERTIES IN
(La_{1-x}Er_x)_{0.67}Sr_{0.33}MnO₃ MANGANITES PEROVSKITE**

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ABSTRACT

Polycrystalline manganites of (La_{1-x}Er_x)_{0.67}Sr_{0.33}MnO₃ (x=0.00, 0.05 and 0.10) had been prepared by conventional solid-state reaction method. X-ray diffraction analysis confirms that all samples are in single phase with distorted perovskite rhombohedral structure. Scanning electron microscope shows that small amount of Er substitution in La site affect the grain formation and this might affect the grain boundaries layer which resulting the reduction of T_c. All sample shows quite similar Low-field magnetoresistance (MR) effect with a large negative MR at low field (0-0.1T) region followed by a slower varying MR at high field (0.1-1T) region. The highest low-field MR value of -3% (at 0.1T, 300K) and high-field MR value of -8.3% (at 1T, 300K) are observed for sample X=0.10.

INTRODCUTION

Colossal magnetoresistance (CMR) effect in hole-doped manganites has attracted significant interest by many researchers in the past decade [1-3] due to their rich physical properties and potential application in magnetic sensing devices. The CMR phenomenon in this system is usually attributed to the magnetic coupling between Mn³⁺ and Mn⁴⁺ ions as described by the Zener's double exchange mechanism as well as to the strong electron phonon coupling arising from the Jahn-Teller effect. Dependent on the doping level, this compound exhibits an insulator-metal transition (T_{MI}) associated with a paramagnetic-ferromagnetic transition (T_c). Under an applied magnetic field, the T_{MI} is shifted to higher temperature, resulting in a CMR effect. The basic early understanding of the CMR effects, which is now known as intrinsic MR, seems to be the same in single crystal bulk and thin film [4]. However, recent reports confirm that polycrystalline compound not only shown intrinsic MR, it also exhibits extrinsic MR in relatively lower field, which commonly known as Low-field Magnetoresistance (LFMR) [5-6]. This effect is believed to be due to the spin-polarized tunneling or spin-dependent scattering across the disorder grain boundaries layers [7-8] which is mostly affected by the grain distribution and size by fine tuning the preparation conditions. Recent report [7-8] has shown that by introduce a dopant in the La site lowered the T_c and affected the magnetoresistance at low-field. In this paper, we report the influence of Er doping at La site on the magnetic and magnetoresistance properties of La-Sr-Mn-O

system.

METHODOLOGY

Polycrystalline sample of $(La_{1-x}Er_x)_{0.67}Sr_{0.33}MnO_3$ with $x=0.00, 0.05$ and 0.10 had been synthesized via conventional solid-state reaction method. Appropriate amounts of high-purity La_2O_3 , Er_2O_3 , $SrCO_3$ and MnO_2 powders (purity >99.5%) were mixed and pre-sintered at $900^\circ C$ for 12 hour in air. After pre-sintered the powder was ground, pressed into pellet and then sintered at $1200^\circ C$ for 24 hours in air. X-ray diffraction (XRD, Philips) with the $Cu\ K\alpha$ radiation and scanning electron microscope (SEM, LEO1455 VPSEM) was used to characterize the structure and microstructure of the samples. The temperature dependent of magnetization of the sample was measured at 313K to 393K by a Vibrating Sample Magnetometer (VSM, LakeShore) and the magnetoresistance effect were measured using DC four-point probe technique with an applied filed of 1 Tesla from 100K to 300K.

RESULTS AND DISCUSSION

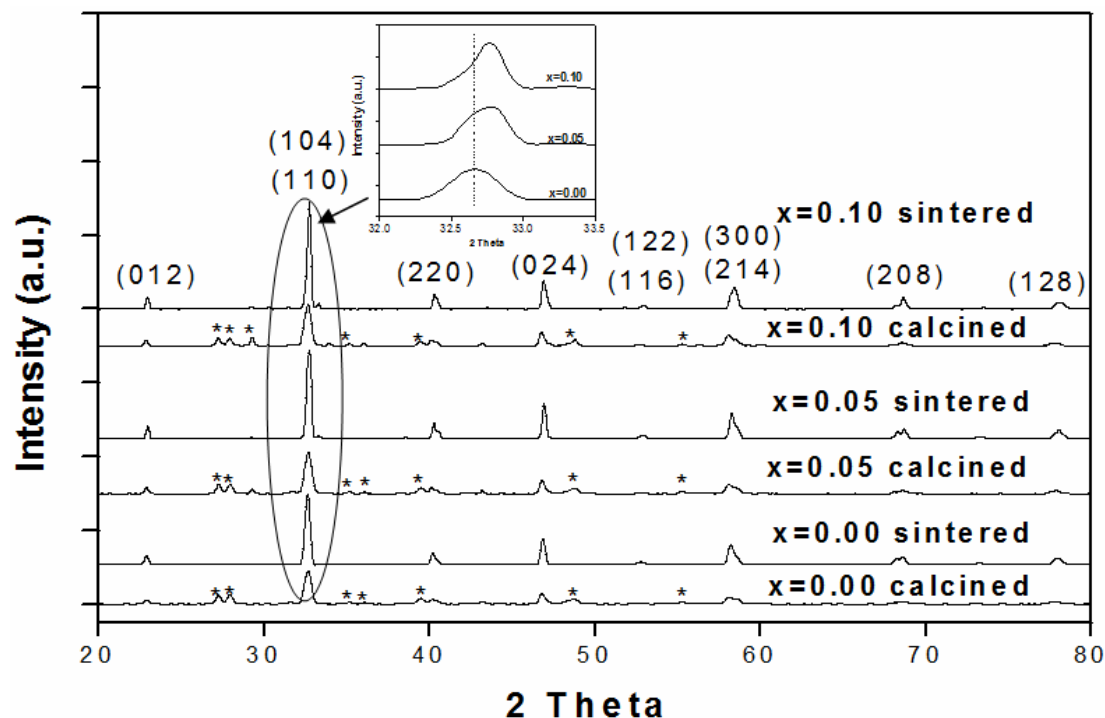


Figure 1: XRD pattern of $(La_{1-x}Er_x)_{0.67}Sr_{0.33}MnO_3$ calcined and sintered at open air. Peak mark with * represent the peak for the starting powder (La_2O_3 , Er_2O_3 , $SrCO_3$ and MnO_2). The insert shows the shift of main peak (32.6°) position due to the substitution of Er.

Figure 1 shows the XRD spectrum for samples before and after sintering process.

Comparing both set of the calcined and sintered powders XRD patterns, the spectrum of the calcined samples show the presence of the peaks of unreacted initial powders. These peaks disappeared when compound sintered at 1200 °C for 24 hours. These remaining peaks (marked with *) were compared with the ICDD standard and found out to be the starting precursor powder (La_2O_3 , Er_2O_3 , SrCO_3 and MnO_2). Meanwhile, single phase of $(\text{La}_{1-x}\text{Er}_x)_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ sintered compound was found matched with the ICDD standard (reference code 00-050-0308) where all samples are in single phase rhombohedral structure with no detectable secondary phase which agrees with the early report [7]. Hence, crystallization had completed after sinter at 1200°C for 24 hours in air. The effect of doping of Er at La sites has shifted the main peak (110) to the right since the ionic radius of Erbium (Er) (0.88Å) is smaller than Lanthanum (La).

Figure 2 show the SEM micrographs at room temperature. Micrograph of pure LSMO ($x=0.00$) showed some degree of liquid phase form when sintered at 1200°C. The particles are well connected and no clear grains or grain boundaries are observed. Big pores are observed through out the sample. However for $x=0.05$, the sample has a comparatively clearer grains or grain boundaries with the grain size of 1µm-2µm and smaller pore size. When more Er ($x=0.10$) doped, the particles are strongly connected to each other and melt-like form microstructure is observed. No clear grains or grain boundaries had been observed. This might be due to the change of the activation energy at higher doping concentration. During sintering, strong diffusion process might have taken place within the particles and the entropy of system increases.

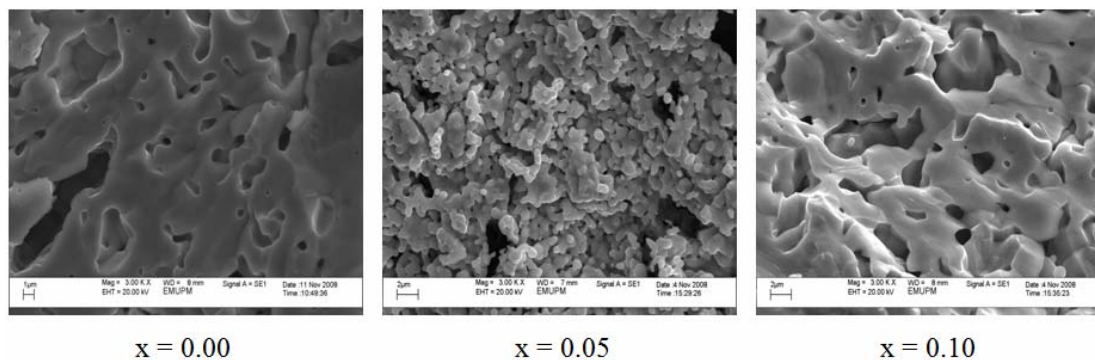


Figure 2: SEM micrograph of $(\text{La}_{1-x}\text{Er}_x)_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ samples

Temperature dependent of the temperature derivative of Magnetization (dM/dT) for all samples are shown in Figure 3. All samples show significant transition of paramagnetic to ferromagnetic phase (Curie temperature, T_c) which assigned as the minimum value in the curve. Curie temperature for pure sample is 370K. This value is in agreement with the report by Cheng et al. [7]. When La is substituted with 5% of Er ($x=0.05$), the Curie temperature dropped to 360K and continued decrease to 345K at $x=0.10$.

The %CMR versus applied magnetic field H curves for all samples at several temperatures all shown in Figure 4(a), 4(b) and 4(c). The %CMR is calculated as $(R_H -$

$R_0/R_0 \cdot 100\%$. All samples showed negative MR with the applied field. This is due to the alignment of the ferromagnetic domains in the sample when magnetic field is applied, the scattering of the polarized electron is reduced and hence resistivity drops when ever there is a applied magnetic field and give rise to negative MR. All curve showed typical behavior for polycrystalline compound where two different gradients are observed, namely a fast drop of negative CMR at low field (0 - 0.1T) region followed by a slower varying of CMR at high field (0.1 - 1T) region. This behavior termed as Low-Field Magnetoresistance (LFMR) is commonly observed in polycrystalline and believed to be due to the influence of the grain boundaries [4-6]. The variation of the %CMR value among all samples might be resulting from the variation of the grain size where different size gives difference surface over volume and hence different grain boundaries. Overall, the CMR is almost linear to the applied magnetic field, H and it decreased continuously without any sign of saturation up to 1 Tesla. This result might be cause by the effect of some smaller grain where higher field is needed to fully align the spins on it. The highest low-field MR value of -10.6% (at 0.1T, 90K) was observed for sample $x=0.05$. However, at room temperature, sample $x=0.10$ gives the highest value of -3%. For high-field MR (at 1 Tesla), the largest %CMR magnitude is given by sample $x=0.05$ with the value of -8.3% (300K).

The temperature dependence of %CMR at 1 Tesla for all samples is shown in Figure 4(d). As the temperature decreases, the %CMR magnitude increases monotonically. Overall, the %CMR magnitude for sample $x=0.05$ is higher followed by $x=0.10$ and $x=0.00$ respectively. This variation might be related to the difference in grain distribution as shown in SEM results where smaller grains enhanced the CMR effect and needed higher field to align the spin contained.

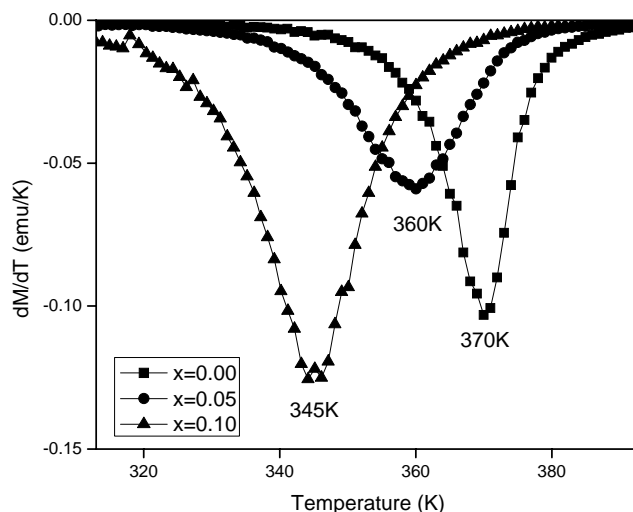


Figure 3: dM/dT vs temperature for $(La_{1-x}Er_x)_{0.67}Sr_{0.33}MnO_3$ sample

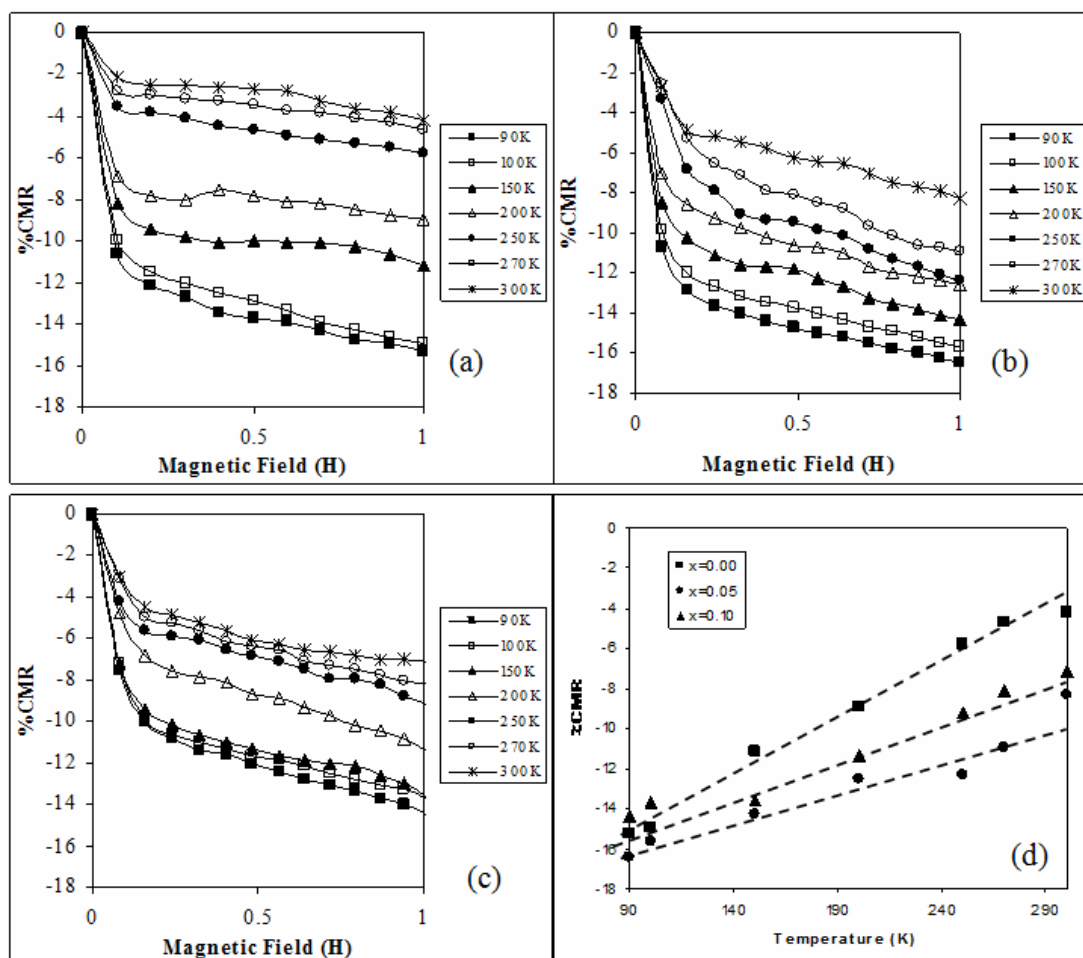


Figure 4: Field dependence of %CMR curve at different temperature for (a) $x=0.00$ (b) $x=0.05$ (c) $x=0.10$ (d) temperature dependence of the MR ratio in 1 Tesla field

CONCLUSION

We have investigated the structure, magnetic and CMR effect in bulk polycrystalline $(La_{1-x}Er_x)_{0.67}Sr_{0.33}MnO_3$ ($X=0.00, 0.05$ and 0.10) samples. The replacement of La by small amount of Er lowered the magnetic phase transition, T_c from 370K ($x=0.00$) to 345K ($x=0.10$). However, the extrinsic MR behavior known as Low-Field Magnetoresistance enhanced. At room temperature, the highest low-field MR value of -3% at 0.1T has been observed for sample $x=0.10$ while at higher applied magnetic field (1Tesla) the highest value of -8.3% is observed for sample $x=0.05$.

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