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Double Peak Behavior of Resistivity-Temperature Curves in $(Nd / Pr)_{0.67}Sr_{0.33}MnO_3$ Manganites

Bharat Singh[1,*], Naresh Kumar[2], Masroor Ahmad Bhat[1], Rajneesh Mohan[3], S. Bhattacharya[4], N.K. Gaur[1]

[1] *Department of Physics, Barkatullah University, Bhopal – 462026 (M.P.) India*
[2] *Department of Physics, Sidharth Govt. College, Nadaun – 177033 (H.P.) India*
[3] *Department of Mechatronics Engineering, Jeju National University, Jeju 690756, South Korea*
[4] *T.P.P.E.D., Bhabha Atomic Research Centre, Mumbai – 400085 India*

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We have reported the synthesis of polycrystalline samples of $R_{0.67}Sr_{0.33}MnO_3$ (R = Pr, Nd) from the precursors of $PrMnO_3$, $NdMnO_3$ and $SrMnO_3$ by using solid state reaction method. These samples were sintered at 1200 °C and 1400 °C. Some of samples were also undergone oxygen annealing at 950 °C for 6 h. All the synthesized samples were characterized by X-Ray diffraction (XRD) technique, scanning electron microscopy (SEM) and low temperature resistivity versus temperature (R-T) measurement. The XRD patterns show the monophasic nature of the sintered and annealed samples. The sintered samples of $Nd_{0.67}Sr_{0.33}MnO_3$ has metal- insulator transition at 250 K, while the sample annealed in oxygen shows two peaks, one sharp peak at 250 K and other broad peak at 200 K. The sintered samples of $Pr_{0.67}Sr_{0.33}MnO_3$ has only one peak at 290 K, while the sample annealed in oxygen shows two peaks, one sharp peak at 290 K and another broad peak at 225 K. The two peak behavior in the annealed samples has been explained by inhomogeneous diffusion of oxygen in the core of the grain and at the grain boundaries region.

Keywords: Manganites, Solid state reaction route, XRD, RT behaviour and SEM.

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1. INTRODUCTION

The discovery of colossal magnetoresistance (CMR) in the perovskite manganites $R_{1-x}A_xMnO_3$ (R is a trivalent rare earth ion and B is a divalent alkaline-earth metal) has attracted considerable interest of physicists because of their exotic electric, magnetic and optical properties [1, 3]. The phenomenon of colossal magnetoresistance (CMR) generally is exhibited in range $0.2 < x < 0.5$ range. For such a range of doping the resistivity (ρ) versus temperature (T) usually exhibits a sharp peak at a certain temperature known as Tc, indicating a transition from metallic behavior below Tc to insulating above Tc. The application of an external magnetic field strongly suppresses resistivity (ρ) and moves the resistivity peak to higher temperatures, thereby producing a CMR near Tc [11, 4]. The unique properties of mixed valence manganites are determined by complex spin, charge, orbital ordered phases and lattice degrees of freedom. Hence manganites are of enormous significance for scientific community. For single crystal and epitaxial films, the electronic and magnetic transitions are observed simultaneously, along with very high value of magnetoresistance [2, 4, 5]. They propose great opportunities for new electronic devices e.g., magnetic sensors, random access memory devices, non-volatile electronic data storage and magneto-electronics. Physical and electronic properties of manganites depends upon the delicate balance of various electronics and magnetic interactions and can be tuned by external disturbance (like magnetic field, electric field and optical radiation etc) due to the breaking of the subtle balance between these interactions in these materials. In our previous study we have reported that on the addition of mercury oxide (HgO) in $La_{0.75}Ca_{0.25}MnO_3$ compound results decrease in the metal to insulator transition temperature (T_{MI}) from 242 K to 232 K has been observed due to cationic disorder created by HgO addition. [12]. We have also shown in our previous report that on varying the electric current values and / or electric field has a strong influence on the electrical properties of manganites [10]. But in case of polycrystalline materials, the magnetoresistance values are over a much wider temperature range [8]. This shows the important role of the grain boundaries in transport properties. Here in this paper, we have reported double peak type resistivity transport behaviour, one sharp peak near T_c and another broad peak below T_c which is attributed to interplay of resistivity transport in the bulk and surface phase of doped manganites.

2. EXPERIMENTAL TECHNIQUE

Initially we have prepared the precursors of $PrMnO_3$, $NdMnO_3$ and $SrMnO_3$ from their corresponding oxides through conventional solid state reaction method. Then stoichiometric $Pr_{0.67}Sr_{0.33}MnO_3$ and $Nd_{0.67}Sr_{0.33}MnO_3$ were prepared from these precursors by solid- state reaction route. The calcination was done at 950 °C for 24 h. The calcinated powders were thoroughly grinded and compacted into circular pellets of 12 mm with the help of hydraulic press. These pellets were heated at 1100 °C, 1200 °C and 1400 °C for 24 h with intermediate grindings. Some samples were also annealed at 950 °C in the continuous flow of oxygen for six hours.

All the samples in the present investigation were subjected to powder X-ray diffraction technique (XRD,

* singh.bharat02@gmail.com

Phillips PW 1710, Cu K_α) for phase evaluation. The low temperature electrical transport measurements in the temperature range of 25 K-300 K were carried out using a close cycled helium cryostat (CTI). The SEM images of annealed samples infer the information of the surface morphology.

3. RESULT AND DISCUSSION

The XRD patterns of the sintered and annealed polycrystalline samples of $Nd_{0.67}Sr_{0.33}MnO_3$ are shown in figure 1 while those for $Pr_{0.67}Sr_{0.33}MnO_3$ sample are shown in figure 2. These patterns are monophasic (orthorhombic unit cell, space group- Pnma) with no detectable impurity phase. This implies that the single phase doped manganites can be prepared from the method described above. From the x-ray diffraction patterns it is quite clear that the oxygen annealing do not have any noticeable effect on the structure and the phase purity of polycrystalline samples. The grain morphology of annealed samples of $Nd_{0.67}Sr_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$ obtained from scanning electron microscopy are shown in figure 3a and figure 3b.

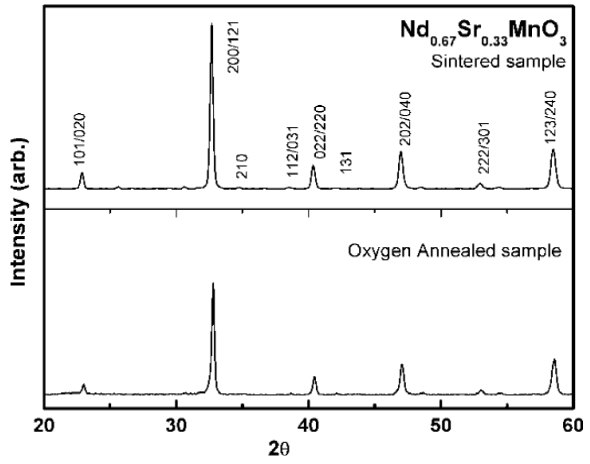

Fig. 1 – Comparison XRD Pattern of annealed and 1400 °C sintered sample of $Nd_{0.67}Sr_{0.33}MnO_3$

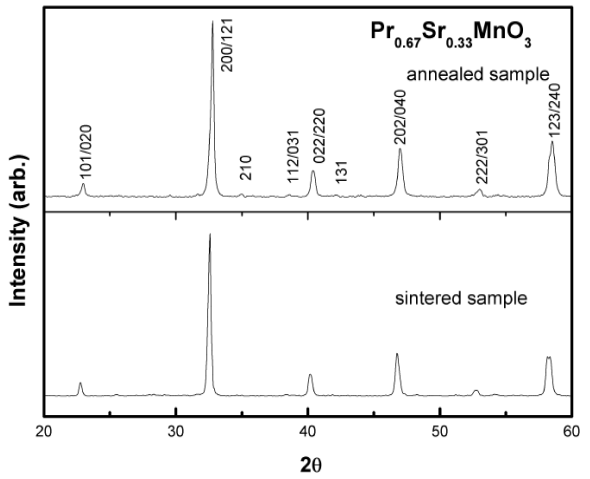

Fig. 2 – Comparison XRD Pattern of annealed and 1400 °C sintered sample of $Pr_{0.67}Sr_{0.33}MnO_3$

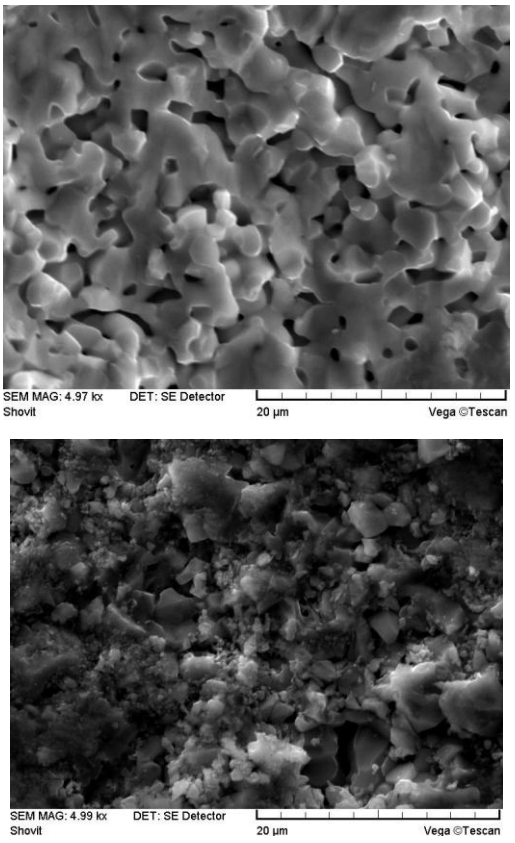

Fig. 3 – SEM image of annealed $Nd_{0.67}Sr_{0.33}MnO_3$ (a) and $Pr_{0.67}Sr_{0.33}MnO_3$ (b)

The low temperature electrical resistivity-temperature behaviors of $Nd_{0.67}Sr_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$ samples with various heat treatments are shown in the figure 4 and 5 respectively. In figure 4, the samples sintered at 1200 °C and 1400 °C of $Nd_{0.67}Sr_{0.33}MnO_3$ show metal to insulator (MI) transition peak at 250 K, while the sample annealed in oxygen shows another broad MI transition peak at 200 K.

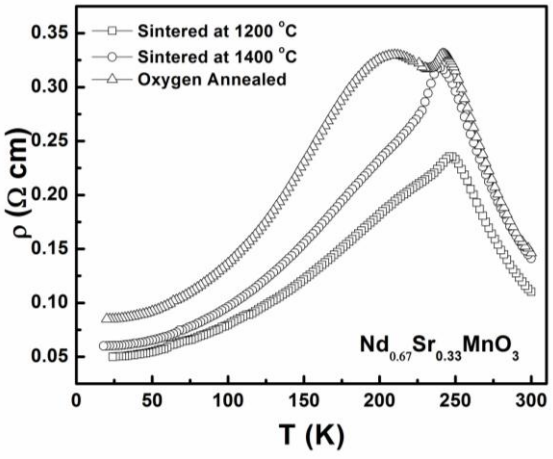

Fig. 4 – The resistivity (ρ) vs temperature (T) behaviour of sintered (1200 °C and 1400 °C) and oxygen annealed $Nd_{0.67}Sr_{0.33}MnO_3$ samples

Similarly, in figure 5, the $Pr_{0.67}Sr_{0.33}MnO_3$ samples sintered at 1200 °C and 1400 °C show metal to insulator (MI) transition peak at 290 K, while the sample an-

nealed in oxygen shows another broad MI transition peak at 225 K. This type of double peak behavior has also been observed in the sintered samples of lanthanum based manganites [7, 9]. This double peak behavior has not been reported in Nd and Pr based manganites in the literature yet. As shown in figures 4 and 5, the sintered samples do not show double peak behaviors. This type of behavior is observed only in the annealed samples.

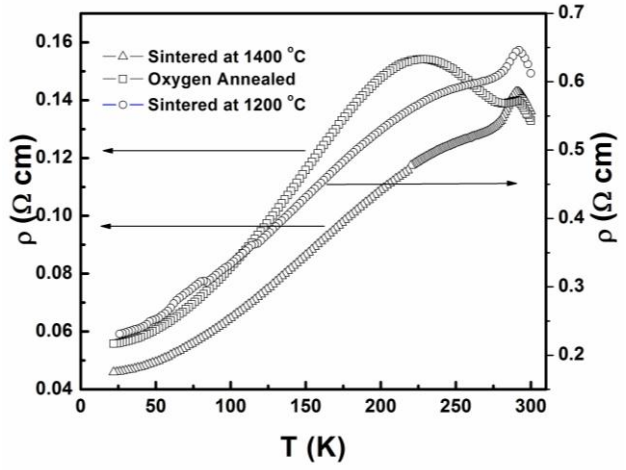

Fig. 5 – The resistivity (ρ) vs temperature (T) behavior of sintered (1200 °C and 1400 °C) and oxygen annealed $Pr_{0.67}Sr_{0.33}MnO_3$ samples

As observed, from the XRD results, the annealed samples are monophasic, without any trace of impurity / secondary phases. Thus, we can say that this second peak do not arises due to any impurity / secondary phase. From the SEM images, it is also clear that there is no phase segregation in the annealed samples.

As it has been shown by Philip & Kutty that the diffusion of oxygen at grain boundaries is faster as compared to that in the bulk of the grains [6]. So we can assume that in the annealed sample, there is inhomogeneous oxygen diffusion in bulk of grains and at the grain boundaries. This inhomegeous oxygen diffusion give rise to two peaks in the resistivity curves one due to intragrain resistivity and other due to intergrain resistivity.

4. CONCLUSION

We have synthesized fruitfully single phase samples of $Nd_{0.67}Sr_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$ through a modified technique from the precursors of PrMnO3, $NdMnO_3$ and $SrMnO_3$. The oxygen annealing at 950°C in sintered samples of $Nd_{0.67}Sr_{0.33}MnO_3$ and $Pr_{0.67}Sr_{0.33}MnO_3$ give rise to double peaks in the resistivity-temperature curves. This behavior can be attributed to different oxygen diffusion coefficients at bulk of grain and at grain boundaries.

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