Abstract

The material 'manganite' having the formula $AMnO_3$, belongs to the large family of Perovskites and have a large variation in properties like colossal magnetoresistance , ferroelectricity, magnetocaloric effect etc. when the trivalent lanthanide ion A is replaced partially by divalent / monovalent alkaline earth ions. Early studies on manganites were motivated by a need to develop insulating ferromagnets with a large magnetization for high frequency application. The renewed interest in this material started in ~1990's with the observation of large MR effect near Curie temperature. More recent work has been driven by a desire to understand and exploit large magnetoresistance and magnetocaloric effects near Curie temperature (T_C).

In the present program, the polycrystalline bulk samples of $La_{1-x}K_xMnO_3$ have been prepared with Potassium concentration up to 0.15 by chemical 'Pyrophoric method'. The characterization of the samples is done by X-ray diffraction and Rietveld analysis, scanning electron microscopy, oxygen non-stoichiometry, density and ac susceptibility measurements. A detail investigation of electrical, thermoelectric, magnetoresistance and the magnetocaloric effect on this polycrystalline $La_{1-x}K_xMnO_3$ compound has been carried out. A major part of the work is devoted to develop a vibrating sample magnetometer (VSM) in order to carry out the magnetization measurement needed for the study of magnetocaloric property of these materials.

The temperature dependence of the electrical resistivity of the samples, measured from 10 to 300K, exhibits a metal-insulator transition associated with ferromagnetic to paramagnetic transition and reveals increasing conductivity with Potassium doping. In the high temperature insulating zone, the resistivity behavior describes the thermally activated hopping of charge carriers in adiabatic small polaron. However, in the metallic region at low temperature, the conduction mechanism is observed to support the small polaron coherent motion involving a

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relaxation due to soft optical phonon mode, along with contribution of two-magnon scattering and spin wave scattering in this system. A field dependent resistivity minimum below 50K exhibited by these polycrystalline samples can be described by the spin polarized tunneling of charge carriers between antiferromagnetically coupled grains. The temperature dependent two phase behavior of resistivity can be explained by a phenomenological model assuming that two main separated phases coexist in the doped manganites with the energy difference to form the two phases that follows the Boltzmann transport equation for two energy level system.

The thermoelectric power (TEP) data of the same compounds is also analyzed by small polaron hopping model at high temperature whereas at low temperature region, the electron-magnon scattering and spin-wave fluctuation contributes to TEP data of these compounds. The influencing role of magnon in the system is confirmed by a common magnon drag curve as described by all the compositions. This thermal variation of thermopower as well as the resistivity of doped manganites have been tried to explain in terms of an 'effective-medium approach' where the conductivity in the transition region is supposed to originate due to the mixing of the metallic-like region with a matrix of the semiconductive polaronic region and this mixing factor should reproduce the magnetization of the system. This two-component phenomenological model based on an effective medium approximation has been employed to estimate the concentration of the metallic region from the measured resistivity data and is found to provide an excellent description of thermopower of $La_{1-x}K_xMnO_3$ compound over the entire measured temperature range including the transition region.

The temperature dependence of magnetoresistance (MR) of these samples has been carried out at a maximum field of 0.8Tesla and in the temperature range from 50 to 300K. Field dependence of MR both below and above T_C is well accounted by a magnetic cluster model that assumes the spin dependent hopping or tunneling of charge carriers from one weakly localized wave packet to another (magnetic cluster model) with the hopping barrier modulated with the mis-

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orientation between the spins of electrons at an initial state and a final state in an elementary hopping process. However the role of grain boundary in these polycrystalline materials cannot be overlooked and a phenomenological model based on spin polarized tunneling of charge carriers at the grain boundaries have been carried out to model the MR behavior in the ferromagnetic region. The contributions in MR from the intrinsic part arising from DE mechanism, as well as, the part originating from intergrannular spin polarized tunneling are also estimated.

Magnetic entropy change (ΔS_M) in $La_{1-x}K_xMnO_3$ compounds have been studied from isothermal magnetization curves in a temperature span of ~40K around their respective T_C. Analysis of the measured ΔS_M shows excellent agreement with those calculated using Landau theory and confirms the importance of magnetoelastic coupling and electron interaction in MCE of mangenites. Magnitude of the change in magnetic entropy obtained in a magnetic field of 1T in the present system is better than that reported for Gd and for many divalent & monovalent lanthanum manganites. The maximum magnetic entropy change ΔS_M ~ 3.00 J/Kg K is obtained with x=0.15 sample in a magnetic field of 1Tesla. Though the Relative Cooling Power (RCP) of $La_{1-x}K_xMnO_3$ compounds is found to be about 1/3rd to that for Gd, K doped lanthanum compounds are inexpensive, easy to synthesize and chemically stable. Moreover, as their Curie temperatures can be tailored between 260 and 310K by adjusting the dopant (K) concentration, the monovalent (K) doped lanthanum manganites could be a possible candidate for multi-component magnetic refrigerator over a wide temperature range.

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