ABSTRACT

Double perovskite systems with fascinating physical properties have a wide range of applications like special interfaces in spintronic devices, solid state refrigerants, digital data storage with spin based memories, etc. Considering this, the present work aims to provide a comprehensive investigation of some new and interesting cobalt-manganese based $Ln_{2-x}(Ca/Sr)_xCoMnO_6$ (Ln=La,Sm,Gd) double perovskite systems, based on detailed studies of their structural disorder, magnetic behaviour, exchange bias effect, magnetic entropy change, Griffiths phase formation and electronic- and magneto-transport properties. The thesis also significantly contributes to the underlying physics behind (1) spin dynamics in phase separated $Ln_{2-x}(Ca/Sr)_xCoMnO_6$ (Ln=lanthanides) systems (2) magnetic frustration and giant spontaneous exchange anisotropy in $Sm_{1.5}Ca_{0.5}CoMnO_6$ system and (3) magnetocaloric and relative cooling power behaviour in $Gd_{2-x}Sr_xCoMnO_6$ systems. To illustrate these ideas, the chemically prepared samples are characterised by X-ray diffraction, Neutron powder diffraction, FESEM, HRTEM, XPS and electrical resistivity measurements followed by rigorous data analysis. The phase separation and magnetic field induced phenomena have been examined using VTI cryostat and SQUID/PPMS magnetometer. In this work, the exchange bias effect has been studied in the $Ln_{2-x}Ca_xCoMnO_6$ (Ln=La and Sm) polycrystalline bulk and nanoparticles to explain the intrinsic unidirectional anisotropy at the magnetic interfaces. Such analysis implies that multi magnetic phases and disorder play a substantial role in the exchange bias phenomena. A phenomenological antiferromagnetic-ferromagnetic interface type structure has been proposed to describe this observed effect. The formation of Griffiths phase arising from short-range ferromagnetic phases/clusters in the paramagnetic matrix, has been investigated in $La_{2-x}Ca_xCoMnO_6$ ($0 \leq x \leq 1$) and $Sm_{1.5}Ca_{0.5}CoMnO_6$ polycrystalline samples, through non-linear AC and DC magnetic susceptibility measurements. The electronic-transport studies for the $La_{2-x}Ca_xCoMnO_6$ (($0 \leq x \leq 1$) samples confirm that the electronic phase of the samples gets enhanced to a high resistive state with Ca doping till x=0.5. The x=0.5 Ca-doped sample exhibits highest magnetoresistance~ 67 % at 40 K. The electronic phase then gets arrested to a low resistive state for x>0.5. Magnetoresistance of all the $0 \leq x \leq 1$ Ca-doped samples strongly depends on temperature and Ca^{2+} concentration, therefore suggesting that disorder plays a crucial role here. A detailed magnetic study on Gd_2CoMnO_6, obtained by replacing La/Sm with rare-earth magnetic element Gd having a comparatively smaller ionic radius, has revealed large magnetocaloric effect at the ordering temperatures because of complex magnetic couplings. This material also shows large entropy change around T_{Gd} (Gd spin ordering temperature). Sr^{2+} doped Gd system ($Gd_{1.5}Sr_{0.5}CoMnO_6$) exhibits enhanced ferromagnetism and inverse magnetic entropy change near antiferromagnetic ordering temperature as a result of negative 3d-4f exchange interactions. Also, a huge entropy change and large relative cooling power are observed around the Curie temperature. Finally, a correlation of structural disorder, magnetic properties and spin dynamics of all the systems has been established.

Keywords: Exchange bias effect, Magnetocaloric effect, Griffiths phase, phase separation, Spin-glass, Magnetoresistance, antisite disorder, Magnetic relaxation and double perovskites.