Observation of Both Debye Type and Maxwell-Wagner Relaxation in $Y_{1.9}Ce_{0.1}CoMnO_6$

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Abstract.The dielectric and resistivity measurement have been observed for polycrystalline sample of double pervoskite $Y_{1.9}Ce_{0.1}CoMnO_6$. Dielectric constant shows Maxwell-Wagner and Debye both type of mechanism. Semiconducting nature of resistivity has been observed for present sample.

INTRODUCTION

In past few years double pervoskite materials have drawn tremendous attention due to their exotic properties and their scope of practical applications. These properties include near room temperature ferromagnetism, spin driven ferroelectricity, magnetodielectric, magnetoresistance etc. Y_2CoMnO_6 in this field have drawn considerable interest due to its interesting properties such as E type magnetic ordering driven ferroelectricity, metamagnetic transition, relaxor like dielectric behaviour etc. For a perfectly ordered double perovskite, the B-site ions are ordered in a rock-salt type pattern. However, during sample preparation anti-site disorder becomes inevitable and a site exchange between two B-site ions i.e. Co^{2+} and Mn^{4+} occurs leading to this anti-site disorder. The anti-site disorder has been seen to affect the physical properties of the double perovskites, particularly the magnetic properties. J. Blascoet. al have shown systematically how anti-site disorder can affect the magnetic properties of Y_2CoMnO_6 [1]. However, an appreciable amount of contribution towards the dielectric behavior of ordered double perovskite materials is believed to be arising from the asymmetric hopping of electrons between B-site ions. The dielectric relaxations may be known with two type of contribution in any material intrinsic and extrinsic. As the name says intrinsic effect is due to materials inherent properties. This effect is associated with permanent dipole moment which leads to debye type of relaxation which is very essential for storage device. The extrinsic effect is due trapped charges, defects dipoles [2-4]. This effect leads to Maxwell-wagner relaxations [5]. For case of Y_2CoMnO_6, the ferroelectricity is driven by E-type of spin ordering which is manifested as a peak at ~100 K in the pyroelectric current. However, the higher temperature dielectric behavior is of ferroelectric relaxor type which is coming from the electronic ferroelectricity due to the charge ordering of Co^{2+} and Mn^{4+} ions. In this work, we are interested in investigating how the Ce^{4+} doping in Y^{3+} site affects the electrical properties of the system. It is expected that Ce^{4+} substitution will create mix-valence states in the B-site ions i.e few Mn^{4+} ions will be converted to the Mn^{3+} states which will eventually alter the electronic structure of the present system. This is expected that introduction of Mn^{3+} ions will effectively affect the charge ordering process which will change the dielectric behavior of the system in turn. This was our primary motivation for investigating the electrical property of this system with Ce doping.

On the other hand the study of electron doping at A site of double pervoskite materials are comparatively less and there can be many scope to explore its effects on their physical properties. In this present, we investigated dielectric, structural and transport properties of $Y_{1.9}Ce_{0.1}CoMnO_6$ system.

EXPERIMENTAL METHOD

The polycrystalline powder sample of $Y_{1.9}Ce_{0.1}CoMnO_6$ has been synthesized by conventional solid state reaction method. The high purity oxides(>99.99%) of CoO, Y_2O_3, Mn_2O_3, CeO have been weighted in proper molecular ratio and mixed thoroughly for an hour in a mortar. The well grinded mixture was subjected to initial heat treatment at 1000°C in air. The resulting powder was re-ground and put again in furnace at 1200 °C for 24hours. This process was repeated several times. The resulting powder thus obtained was pelletized and finally kept at 1300°C for 36 hours followed by a slow cooling to reduce anti-site disorder.

Phase purity and structural has been checked by powder X-ray diffraction by RigakuMiniflex 2 X-ray diffractometer (CuKα) and was refined using FULLPROF suite software by rietveld method. The sample thus obtained is free from any chemical impurity and of single phase. Sintered pellets of thickness 1.6mm and diameter 9.5mm was polished on both sides for dielectric measurements. Keysight E4980A LCR meter was used for dielectric measurements in He based closed cycle refrigerator.

RESULTS AND DISCUSSIONS

X-ray Diffraction Study

The Reitveld refinement of X-ray diffraction pattern confirms the single phase of monoclinic structure with $P2_1/n$ space group.Fig 1 shows X-ray diffraction along with its Reitveld refinement of the system at room temperature.
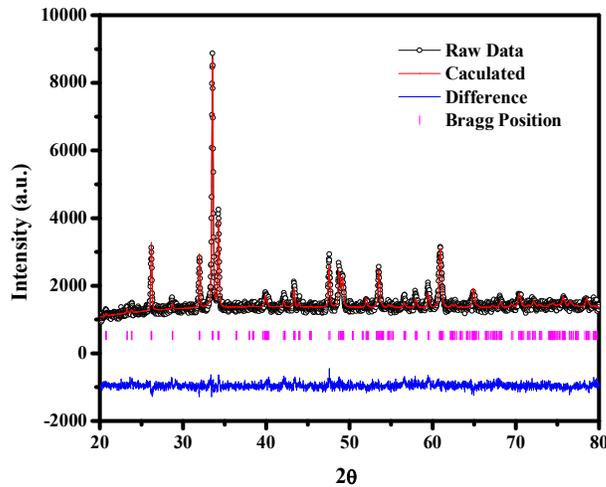

FIGURE 1. Reitveld refined X-ray diffraction patter of prepared powder

Dielectric Study

From the study of frequency variation of real and imaginary part of dielectric constant, we observed two different types of relaxations present in our system. One is originated from the asymmetric hopping of charge carriers. This is known as Debye relaxation. The other mechanism is due to the presence of accumulated charge carriers. This is known as Maxwell-Wagner relaxation. The study of imaginary part of the dielectric constant as a function of frequency is a powerful technique to distinguish between the Debye relaxation and the Maxwell-Wagner relaxation. For the Debye type of relaxation this curve shows a broad peak showing as it falls to zero at lower frequency and higher frequency. However, in for no-Debye type of relaxations i.e. for Maxwell-Wagner type of relaxations the imaginary part of dielectric constant ($\varepsilon^/$) varies with frequency (ω) with a power law as $\varepsilon^/ \alpha 1/\omega^n$, Where n varies between: 0.5<n<1.

The experimental graphs for our system are shown in Fig. at different temperatures. It can be seen that the ε/ Vs ω curves is not tending to zero as the frequency decreases to zero. This suggests that Maxwell-Wagner type of relaxation is involved in the current mechanism. We tried to fit the data with a combined model of Maxwell-Wagner along with the Debye relaxation model.

The successful fitting of the graphs by this model clearly suggests that both the mechanism is involved in the present case. The Debye type of relaxation is coming from the charge ordering between the Co and Mn ions. The Maxwell-Wagner type of relaxation is coming possibly from the trapped charges or due to the inhomogeinity of conductivity present in the grain/grain boundary regions.
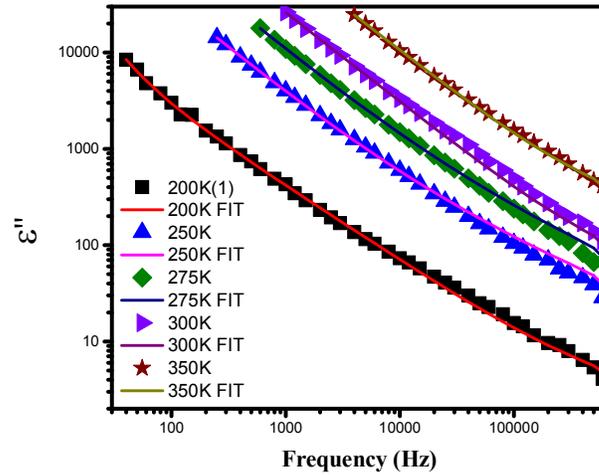

FIGURE 2. Frequency dependent dielectric constant at various temperatures

Resistivity Study

Temperature dependent resistivity of bulk sample has been shown in figure 3. The nature of resistivity suggests semiconducting type of nature. The resistivity in the whole temperature range 100-300 K fits well by the variable range hopping and according to it the resistivity can be written as

$$\rho = \rho_0 \exp\left(\frac{T_0}{T}\right)^{1/4}$$

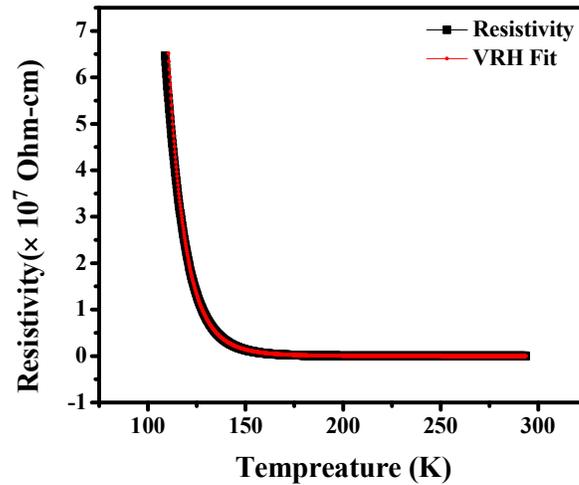Where T_o is the characteristic temperature and $\rho_o$is the pre-factor.

FIGURE 3. Variation of resistivity with respect to temperature

CONCLUSIONS

Pure phase of $Y_{1.9}Ce_{0.1}CoMnO_6$ polycrystalline sample has been prepared successfully. Structural investigations by XRD study confirmed the sample is of single phase and it crystallized in monoclinic structure with P21/n symmetry. The dielectric study shows involvement of both the Debye type and Maxwell-Wagner type of relaxation in the present system. The resistivity measurements of the sample show semiconducting nature. The conduction mechanism involved in the transport property of the system seems to be Variable range hopping mechanism (VRH).

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