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The structural and electrical properties of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ manganites

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Abstract. In this paper, the authors report the electrical properties of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ manganites synthesized using sol-gel method. The X-ray diffraction (XRD) patterns of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ samples reveal an orthorhombic perovskite structure with *Pnma* space group. Analysis using energy dispersive X-ray (EDX) confirms that the sample contains all expected chemical elements without any additional impurity. The measurement of resistivity versus temperature using cryogenic magnetometer was performed to investigate the electrical properties. The results show that the electrical resistivity of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ exhibits metalic behavior below 244 K. The temperature dependence of electrical resistivity dominantly emanates from electronelectron scattering and the grain/domain boundary play a important role in conduction mechanism in polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃.

Introduction

Perovskite manganites with general formula $La_{1-x}A_xMnO_3$ (A = divalent ions) have been a popular research subject due to its high magnetoresistance (MR) and magnetocaloric effect [1-5]. A theoretical explanation of relation among the level of A doping, electrical conductivity and magnetic properties was initially explained by Zener double exchange interaction [6]. It is a transfer of e_g electron from Mn^{3+} ions to Mn^{4+} ions through O^{2-} ions [6,7]. According to this model, the electrical conductivity is proportional to the level of A doping [6].

A perovskite material La_{0.8}Ca_{0.2}MnO₃ has a high MR and magnetocaloric effect but its Curie temperature (T_c) and metal-insulator transition temperatur (T_{MJ}) below room temperature [8–10]. Monovalen ions oxidizes Mn^{3+} ions to Mn^{4+} ions twice as much as divalent ions [11]. For low doping monovalent Ag, Pi et al reported that doping Ag in La site of lanthanum manganite induced an increase of MR, Curie temperature and metal-insulator transition temperature [12]. In this present work, the authors partially subtitute divalent ion Ca in $La_{0.8}Ca_{0.2}MnO_3$ with monovalent ion Ag to form a perovskite manganite $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$. We study the structural, and electrical properties of that perovskite mananites.

Experimental

The perovskite manganite material La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ was synthesized by sol-gel method. Stoichiometric amounts of La₂O₃, Ca(NO₃)₂.4H₂O, AgNO₃, Mn(NO₃)₂.4H₂O were used as precursor materials. La₂O₃ was dissolved in nitric acid solution. Then, lanthanum nitrate solution was mixed



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with other metal nitrate solution. Citric acid was added to the solution with molar ratio of 1.2 between citric acid and metal nitrate. the pH value of solution was adjusted to 7 by adding amonia solution while stirring the solution at temperature of 80 $^{\circ}$ C until the gel was formed. The gel was heated at 120 $^{\circ}$ C for dehidration. It was burned at 600 $^{\circ}$ C for 5 h to remove produced organic materials. The materials obtained from this process were ground by using an agate mortar. The powders were pressed into pellets and then heated at 900 $^{\circ}$ C for 24 h.

Identification of the phase and structural analysis of the sample was determined by X-ray diffractometer (XRD) using monochromatic CuK α radiation. The composition of elements was analyzed using energy dispersive X-ray spectroscopy (EDX). The electrical properties of samples was determined by cryogenic magnetometer at temperature 12-255 K.

Results and discussion



The XRD pattern of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ is shown figure 1. The analysis results using Rietveld refinement method confirm that the sample is a single phase since no peaks belongin to the other phase detected. The diffraction peaks can be indexed in an orthorombic perovskite structure with space group *Pnma*. The refinement results are shown in table 1. The quality of the refinement of the XRD data is evaluated by the goodness of the fit indicator χ^2 . The Energy Dispersive X-ray (EDX) analysis shown in figure 2 reveals the presence of all expected elements. It shows that there is no quantifiable loss of each elements.

Table 1. Lattice parameters, unit cell volume and googness of fit (χ^2)

Parameters	La _{0.8} Ca _{0.1} Ag _{0.1} MnO ₃		
a (Å)	5.472		
b (Å)	7.739		
c (Å)	5.499		
Volume (Å ³)	232.89		
Goodness of fit	1.055		

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Figure 3. The temperature dependence of electrical resistivity of polycrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$

The temperature dependence of electrical resistivity of polycrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ at zero applied magnetic field and 1 T is shown in figure 3. The electrical resistivity decreases when magnetic field 1 T is applied. The polycrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ shows a metal-insulator transition temperature (T_{M-I}) at 244 K. Below 244 K, The electrical resistivity of policrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ exhibits metalic behavior which electrical resistivity increases with increment of temperature. To analyze the conduction mechanism at metalic region, the experimental data of policrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ at zero applied magnetic field and 1 T are fitted using equation [3,13,14]:

$$\rho(T) = \rho_0 + \rho_2 T^2 + \rho_{4.5} T^{4.5} \tag{1}$$

Where ρ_0 , ρ_2 and $\rho_{4.5}$ are resistivities due to grain/domain boundary scattering, electron-electron scattering, and combinations of electron-electron, electron-magnon and electron-phonon scattering processes, respectively [3,13,14]. The fitting results are shown in figure 4 and the fitting parameters are presented in table 2. The quality of fitting is evaluated by the square of linear correlation coefficients R^2 [3]. The values of ρ_0 , ρ_2 and $\rho_{4.5}$ decrease when a magnet field 1 T is applied. The value of ρ_2 is higher than that of $\rho_{4.5}$ which indicates that the temperature dependence of electrical resistivity dominantly emanates from electron-electron scattering. The value of ρ_0 is the highest indicates that grain/domain boundary play a important role in conduction mechanism in polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃.

	ρ _o	ρ_2	ρ _{4.5}	Adj. R-Square
	(Ohm.m)	$(10^{-3} \text{ Ohm.m/K}^2)$	$(10^{-6} \text{ Ohm.m/K}^{9/2})$	
H=0	0.047	0.223	2.116	99,95%
H=1	0,019	0,054	1,252	99,99 %

Table 2. Fitting paramater of electrical resistivity data of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃

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Figure 4. Theoretical fitting of resistivity data in metallic region at zero magnetic field (left) and applied magnetic field 1 T (right)

Magnetoresistance (MR) as a function of temperature measured at a temperature of 12-255 K is shown in figure 5. The magnetoresistance (MR) is calculated using equation [15]:

$$MR(\%) = \frac{\rho_0 - \rho_H}{\rho_0} \times 100\%$$
(2)

Where ρ_0 and ρ_H are the resistivities in the zero magnetic field and the presence of magnetic field, repectively. The polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ has the highest value of MR at lowest temperature and decrease with temperature increment. At temperature near T_{I-M}, the value of MR increase with increment of temperature. Magnetoresistance (MR) effect in polycrystaline lanthanum manganite emanate to two contribution. First, the intrinsic MR originates from suppression of spin fluctuation when the magnetic field is applied [16]. The intrinsic MR reaches maximum around the paramagnetic-ferromagnetic transition temperature [17]. The second contribution emanates from spin polarized tunneling process at grain boundary which is dominant at low temperature [16,17]. The spin at the grain boundary is highly disorder [18]. That spin aligns to the applied magnetic field direction which reduces electron scattering by the grain boundary and increase electron tunneling [9,16]. The magnetoresistance of polycrystalline La_{0.8}Ca_{0.17}Ag_{0.03}MnO₃ reveals an extrinsic MR at lower temperature and an intrinsic MR around T_{I-M}.



Figure 5. Magnetoresistance as a function of temperature

Summary

The structural and electrical properties of polyicrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ samples (x = 0, 0.03) prepared by sol-gel method are investigated. The X-ray diffraction (XRD) patterns of polyicrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ samples reveale an orthorhombic perovskite structure with *Pnma* space group. Analysis using energy dispersive X-ray (EDX) confirm that the sample contains all expected chemical elements without any additional impurity. Below 244 K, The electrical resistivity of polyicrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ exhibits metalic behavior. The temperature dependence of electrical resistivity dominantly emanates from electron-electron scattering and the grain/domain boundary plays a important role in conduction mechanism in polyicrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$. The magnetoresistance of polyicrystalline $La_{0.8}Ca_{0.17}Ag_{0.03}MnO_3$ reveals an extrinsic MR at lower temperature and an intrinsic MR around T_{I-M} .

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