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Research paper

Comparative Study on Structural, Electrical Transport and Magnetic Properties of Cr-Doped in Charge-Ordered Pr_{0.75}Na_{0.25}Mn_{1-X}cr_xo₃ and Nd_{0.75}Na_{0.25}Mn_{1-Y}cr_yo₃ Manganites

Rabiatul Adawiyah Zawawi¹, Nurul Nasuha Khairulzaman², Suhadir Shamsuddin^{3*}, Norazila Ibrahim⁴

^{1,2,3}Ceramics and Amorphous Group, Faculty of Applied Sciences and Technology, Pagoh Higher Education Hub, Universiti Tun Hussein Onn Malaysia, Pagoh Campus, 84600 Pagoh, Johor, Malaysia.
⁴Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia.
*Corresponding author E-mail:suhadir@uthm.edu.my

Abstract

Cr doping in charge-ordered $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ and $Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO_3$ have been synthesized using conventional solid-state method to investigate its effect on structural, electrical transport and magnetic properties. X-ray diffraction (XRD) analysis for both compounds showed that the samples were crystallized in an orthorhombic structure with *Pnma* group. The unit cell volume value decrease as the Cr-doped increased indicating the possibility of Mn^{3+} ion was replaced by Cr^{3+} due to the different of ionic radius. The temperature dependence of electrical resistivity showed an insulating behavior down to the lower temperature the both parent compound (x = 0) and (x = 0) and (x = 0) successive substitution of Cr at Mn-site in $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ manganites induced the metal-insulator (MI) transition temperature around Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are Pr_{MI} and Pr_{MI} and Pr_{MI} are $Pr_$

Keywords: Charge-Ordered; Cr-Doped; Double-Exchange Mechanism; Electrical Transport; Magnetic Properties.

1. Introduction

The discovery of colossal magnetoresistance (CMR) effect in rare-earth perovskite-type manganites with general composition of type $Re_{1-x}A_xMnO_3$ (Re=a trivalent rare-earth ion, A=a divalent alkaline-earth) have attracted much attention in these recent years [1–7]. In addition, studies on these rare-earth manganites have revealed that CMR effect which is commonly attributed to the double- exchange (DE) mechanism is also suggested to be related to the Jahn-Teller (JT) effect and charge ordering (CO) [8] and the lattice distortion [9].

In particular, the $Pr_{0.75}Na_{0.25}MnO_3$ and $Nd_{0.75}Na_{0.25}MnO_3$ compound has attracted due to the existence of CO transition at relatively high temperature, $T_{CO} \sim 220~\rm K$ for $Pr_{0.75}Na_{0.25}MnO_3$ and $T_{CO} \sim 170~\rm K$ for $Nd_{0.75}Na_{0.25}MnO_3$ compared to antiferromagnetic (AFM) interaction [10–12]. The presence of CO state was suggested to be related to the presence of an equal ratio of Mn^{3+} and Mn^{4+} where the ions align themselves in an ordered pattern [13–15].

Mn site substitutions of Cr on perovskite manganites have attracted great interest as the Cr³⁺ion is iso-electronic with the Mn⁴⁺ ion [16–20]. It is reported that among the different studied doping elements, Cr is the most efficient one to induce a metal insulator

transition in the CO undoped insulators and leads to a much higher CMR effect [16]. For example, in Nd_{0.7}Sr_{0.3}Mn_{1-x}Cr_xO₃ and La_{0.7}Ca_{0.3}Mn_{1-x}Cr_xO₃ the electrical transport behaviour shows that the metal-insulator (MI) transition shift to lower temperature and weakened gradually as the Cr content increased due to DE mechanism between Mn³⁺–O–Mn⁴⁺ [17].

Meanwhile all samples for Cr doped up to 0.3 in compound $La_{0.67}Ca_{0.33}Mn_{1-x}Cr_xO_3$ exhibit a ferromagnetic-paramagnetic (FM-PM) transition with T_C decreased rather slowly with increasing Cr content for $x\leq0.2$ [18]. When doping on Mn sites with Cr^{3+} ions, the transport and magnetic properties of the manganites will show some interesting behaviors. Thus, considering all the above, the studies on the effects of Cr doping on Mn site is expected to understand the physical nature of doping effect in manganites. However, to the best of our knowledge, there are only few studies on $Nd_{0.75}Na_{0.25}MnO_3$ and the effect of Cr doped in monovalent manganite of $Pr_{0.75}Na_{0.25}MnO_3$ have not been reported yet on structural, transport and magnetic properties.

In this work, we report the effect of magnetic ion, Cr doping at the Mn site on the structural, electrical transport and magnetic properties of Pr_{0.75}Na_{0.25}MnO₃ and Nd_{0.75}Na_{0.25}MnO₃ manganite. The Cr³⁺ ion was suggested to be very effective element to suppress CO as well as induced the phase transition.



Table 1. MI transition temperature (T_{MI}) , Curie temperature (T_{C}) , Neel temperature (T_{N}) , lattice parameters, unit cell volume (V), density (D) and porosity

of Pr Na Mn.	$Cr \cap (0 < r < 0) \cap (1 $	and Nd Na Mn.	$_{v}$ Cr $_{v}$ O ₃ $(0 \le v \le 0.05)$
OI 1 10 751 Van 251VIII	$ _{1}C1_{1}C3 _{U} \geq \lambda \geq 0.04$	1) and ind _{0.751} na _{0.251} viii ₁ .	$_{1}$ $_{1}$ $_{1}$ $_{2}$ $_{3}$ $_{4}$ $_{5}$ $_{1}$ $_{5}$ $_{7}$ $_{1}$ $_{1}$ $_{2}$ $_{3}$ $_{4}$ $_{5}$ $_{5}$ $_{7}$

Samples	$T_{MI}\left(\mathbf{K}\right)$	$T_{C}\left(\mathbf{K}\right)$	$T_N(\mathbf{K})$	Lattice parameter			$V(\text{Å})^3$	D (g/cm ³)	Porosity
	(± 0.1)	(± 0.1)	(± 0.1)	(± 0.001)			(±0.1)	(± 0.01)	(%)
				a (Å)	b (Å)	c (Å)			(±0.1)
x = 0.0	-	-	-	5.446	7.696	5.445	228.2	5.71	11.0
x = 0.02	120.0	132.0	-	5.448	7.669	5.443	227.4	5.68	13.0
x = 0.04	122.0	141.0	-	5.449	7.659	5.444	227.2	5.51	14.0
y = 0.0	-	-	-	5.446	7.696	5.445	228.2	5.97	5.00
y = 0.02	-	-	115.0	5.437	7.697	5.450	228.1	5.96	6.00
y = 0.05	-	-	125.0	5.448	7.669	5.443	227.4	5.88	7.00

2. Experimental Method

2.1. Sample preparation

The polycrystalline samples of $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ (x = 0, 0.02and 0.04) and $Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO_3$ (y = 0, 0.02 and 0.05) from the stoichiometric amount of Pr₂O₃, Na₂CO₃, MnO₂, and Cr₂O₃ powders and Nd₂O₃, Na₂CO₃, MnO₂ and Cr₂O₃ powders with high purity (≥99.99%) respectively were prepared using conventional solid-state reaction method. The starting material were mixed and ground thoroughly in an agate mortar with pestle for approximately 2 hours to get homogeneous powders. The homogeneous fine powders were then calcined in air at 1000 °C for 24 hours at the rate of 15 °C/min on heating with several intermediate grinding followed by cooling at rate of 1 °C/min. The resultant powder was reground for another 2 hours. Such calcined mixtures were then pressed into pellets with 13 mm diameter and 2-3 mm thickness under 6 tonnes and sintered in air at 1200 °C for 24 hours in a Protherm Furnace Model PLF130/15 with the heating rate of the order 15 °C/min and cooling rate 1 °C/min to ensure a better crystallization.

2.2. Sample characterization

The structural characterization of all samples were characterized by X-ray diffraction (XRD) using Bruker D8 Advance model with a CuK_{α} (1.544 Å) radiation at room temperature. The sample was scanned continuously in the range of $20^{\circ} \le 2\theta \le 80^{\circ}$ with a scanning rate of 2°/min and then the structural analyses were carried out by X'Pert HighScore software to confirm the crystalline phase of the samples. Electrical resistivity as a function of temperature was measured by standard four probe method with silver point contact in a Janis model CCS 350T cryostat and controlled by a LakeShore Model 330 under zero magnetic fields in the temperature range 50-300 K. The temperature dependence of AC susceptibility was measured in CryoBIND-T system in conjunction with a 7265 DSP lock-in-amplifier and an oscillator at 240 Hz in the temperature range of 50-300 K to determine the Curie temperatures of the samples. Bulk density of the samples was determined by employing the Archimedes principle using acetone as the liquid buoyant.

3. Result and Discussion.

3.1. Structural analysis

Room temperature powder XRD patterns of $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ (x=0, 0.02 and 0.04) samples and $Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO_3$ (y=0, 0.02 and 0.05) as shown in Figure 1 and Figure 2 respectively. The analysis revealed that the structure of the both compound were crystallized in orthorhombic with space group *Pnma* and all samples were found in a single phase without the presence of any impurity peaks and it is in line with the structure reported from the previous study [19,20]. MI transition temperature (T_{MI}), Curie temperature (T_C), Neel temperature (T_N), lattice parameters, unit cell volume (V), density

(*D*) and porosity for all samples are tabulated in Table 1. As the Cr content increased, a regular shift toward higher 2θ was observed indicating the decreasing in unit cell volume as reported in previous study by [21]. This is possibly due to the relatively larger of Mn³⁺ ion with higher ionic radius (0.645 Å) partially substitute of Cr³⁺ ions that has a smaller ionic radius (0.615 Å) as proposed by [22] in La_{0.67}Ba_{0.33}Mn_{1-x}Cr_xO₃ compound [22]. Apart from that, the substitution of Cr³⁺ for all samples were also suggested to induce the lattice distortion and rearrangement of atom as a result of smaller cation in the B-site of the ABO₃ perovskite structure is responsible for the reduction in unit cell volume [23–25] and it is also supported by *D* values when Cr doped in these manganite compound.

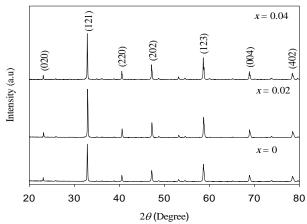


Fig. 1: X-ray powder diffraction (XRD) pattern of $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ ($0 \le x \le 0.04$) samples

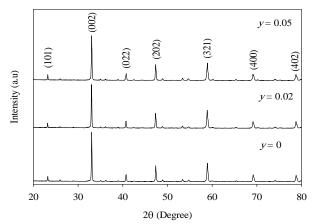


Fig. 2: X-ray powder diffraction (XRD) pattern of $Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO_3$ ($0\le y\le 0.05$) samples

3.2. Electrical transport properties

Figure 3 shows the temperature dependence of electrical resistivity in the temperature range 50–300 K for $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ (x=0, 0.02 and 0.04) under zero magnetic field. The sample with x=0 exhibit an insulating behavior without showing any

transition of MI consistent with the previous study [26]. A MI transition was observed at 120 K and 122 K for x=0.02 and x=0.04 samples respectively. This behavior is attributed to the fact that the doping of Cr^{3+} ion disturbs the lattice and also dilutes the double-exchange (DE) by reducing Mn^{3+}/Mn^{4+} ratio, which is assumed to produce strong DE compared to that DE between Mn^{3+}/Mn^{4+} . The shifting of MI transition temperature, T_{MI} from 120 K (x=0.02) to 122 K (x=0.04) suggest the enhancement of DE-like mechanism of Cr^{3+} –O- Mn^{3+} as a result of weakening the JT effect causing the CO state to be weakened [13,27]

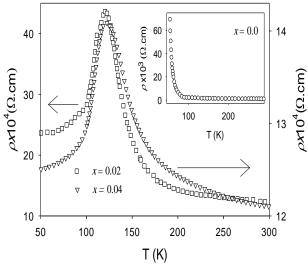


Fig. 3: Temperature dependence of the electrical resistivity of $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ ($0 \le x \le 0.04$). Inset is ρ versus T for x = 0 sample.

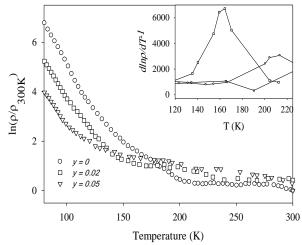


Fig. 4: Temperature dependence of electrical resistivity for $Nd_{0.75}Na_{0.25}Mn_{1.y}Cr_yO_3$ ($0 \le y \le 0.05$) manganites. Inset is $d\ln \rho/dT^1$ vs T for $Nd_{0.75}Na_{0.25}Mn_{1.y}Cr_yO_3$

Meanwhile, Figure 4 shows the effect of Cr-doped on temperature dependence of electrical resistivity in Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO₃ manganites for y = 0-0.05 samples. All the samples with y = 0, 0.02 and 0.05 exhibits an insulating behaviour and did not show any transition as temperature decreases. However, analysis of $d\ln \rho/dT^{-1}$ vs. T (inset Figure 4) curve displays a peak which indicates CO state for sample y = 0 and 0.02 with a CO transition temperature Tco around 210 K and 160 K respectively while no peak was observed for y = 0.05 sample. The decreasing of CO transition temperature indicated the Cr-doped also affected the resistivity data in the region of CO state under zero magnetic field which may be related to the suppression of CO state. Apart from that, the increasing of Cr content affected the intensity values of the resistivity which can be suggested due to the increase of carrier concentration as result of weakening the CO state [12]. The presence of CO and localizing the charge carrier can be suggested

due to JT effect that contributes to the electron-phonon interaction [28]. Insulating behaviour can be related with JT effect as Mn^{3+} is known as JT ion which is the e_g electrons are localized and easier to be trapped in MnO_6 octahedral, hence causing the phonon easier to be scattered by distortion [17].

3.3. Magnetic properties

Temperature dependence of the AC susceptibility (χ') measurement for all samples of $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ manganites was shown in Figure 5. The parent compound (x=0) showed paramagnetic (PM) to antiferromagnetic (AFM) transition with Neel temperature, $T_N \sim 129$ K in line with a previous suggestion for by [26]. Increasing of Cr content induced paramagnetic (PM) into ferromagnetic (FM)-like with Curie temperature, T_C increases from 132 K (x=0.02) to 141 K (x=0.04) indicated the suppression of CO. The T_C was determined from the temperature corresponding to the minimum value from $d\chi'/dT$ versus T plots as shown in Figure 5. Considering those electrical properties, this can be suggested that the samples exhibit a FM-like phase due to double-exchange interaction involving Mn^{3+} –O– Mn^{4+} and Cr^{3+} –O– Mn^{3+} [29].

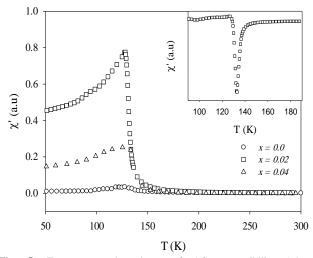


Fig. 5: Temperature dependence of AC susceptibility (χ') of $Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO_3$ ($0 \le x \le 0.04$). Inset is $d\chi'/dT$ vs T for x = 0.02

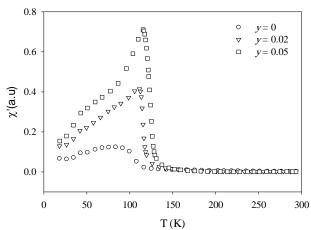


Fig. 6: Temperature dependence of AC susceptibility (χ') for $Nd_{0.75}Na_{0.25}Mn_{1.y}Cr_yO_3$ $(0 \le y \le 0.05)$ manganites.

The AC susceptibility (χ ') versus temperature (T) of Nd_{0.75}Na_{0.25}Mn_{1- γ}Cr_{γ}O₃ compound is plotted in Figure 6. The temperature dependence of AC susceptibility reveals that all the samples shows paramagnetic (PM) to antiferromagnetic (AFM) transition with Neel transition temperature, T_N increased from 115 K for y = 0.02 to 125 K for y = 0.05 indicate the suppression of CO state

which is in line with the analysis of analysis of $d\ln\rho/dT^{-1}$ vs. T as shown in Figure 4.

4. Conclusion

In conclusion, the effects of Cr³⁺ doping on the structural, electrical transport properties and magnetic properties of Pr_{0.75}Na_{0.25}Mn₁₋ $_{x}Cr_{x}O_{3}$ (0 $\leq x \leq 0.04$) and Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO₃ (0 $\leq y \leq 0.05$) samples have been studied. By the XRD analysis for all samples, it is observed that Cr³⁺ could continuously replace Mn³⁺ based on the values of calculated unit cell volume, V decrease continuously for both compound with Cr content suggestively due to the Cr3+ ion has a smaller ionic radius compare to Mn3+ ion. For Pr_{0.75}Na_{0.25}Mn_{1-x}Cr_xO₃, the MI transition temperature, T_{MI} increased with Cr doping from 120 K to 122 K for x = 0.02 and x =0.04 respectively suggested due to the enhancement of DE mechanism as a result of weakening JT effect as well as suppress CO state. Cr³⁺ also suggestively involved in PM-FM transition resulting the increased of T_C from 132 K (x = 0.02) to 141 K (x = 0.04) indicated the suppression of CO. Meanwhile analysis of resistivity data of $d \ln \rho / dT^{-1}$ vs. T in $Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO_3$ manganite showed a peak around 210 K and 160 K for y = 0 and 0.02 samples respectively while no peak was observed for y = 0.05 indicating the Cr-doped affected the resistivity data in the region of CO state. Analysis of susceptibility from Nd_{0.75}Na_{0.25}Mn_{1-y}Cr_yO₃ exhibit PM-AFM as Neel temperature T_N increases from 115 K for y = 0.02 to 125 K for y = 0.05 indicate the existing of CO state.

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