Study of the transport characteristics in LCMO (Perovskite manganite)-rGO nanocomposite System at various temperature for Resistive Switching (RS) Application

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Abstract

Intensive studies of resistive switching mechanisms in oxide systems have been studied extensively due to its great potential prospect in non-volatile memory applications. Several materials are well-explored for the Resistive Switching (RS) phenomenon as binary oxides, polymers, perovskites, chalcogenides and even 2-D materials. The investigation for the best material offering RS behaviour is still going on some magnetite like ($Pa_{0.7}Ca_{0.3}MnO_3La_{0.7}Sr_{0.3}MnO_3$) also depicts the RS behaviour and their performance are limited by the random. Oxygen vacancies perform a vital role in initiating the resistive switching (RS) phenomenon in oxide-based systems. The random oxygen vacancies can be limited by two methods (1) by doping and (2) by forming nanocomposite. Interest has grown in understanding to perovskite material. Among the several oxide materials, oxygen vacancies are introduced in oxide-based systems using the synthesis method itself. So, lanthanum calcium manganites ($La_{0.3}Ca_{0.7}MnO_3$) are taken as active materials due to their fascinating physical and electrical properties and study its RS effects, the small amount of reduced graphene oxide(rGO) in it.

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Introduction

Resistive switching (RS) mechanism in oxide system has been studied extensively due to its prospect in non-volatile memory application [1-5]. Several materials have been well explored for the RS phenomenon like, binary oxides, chalcogenides, polymers, perovskites and even 2-dimensional materials [6-9]. The quest for the best materials offering RS behavior is still going on. The manganites like ($Pa_{0.7}Ca_{0.3}MnO_3$, $La_{0.7}Sr_{0.3}MnO_3$) also depicts the RS behavior, but their performances are limited by the random oxygen vacancies [10-11]. In oxides-based system oxygen vacancies play an important role to initiate the RS phenomenon [12-15]. The introduction of oxygen vacancies into an oxides material may increase the probability of better RS behavior. The oxygen vacancies can be controlled by two approaches by doping or by making nanocomposite. Since nanocomposite approach is better and easier process than the doping and also has an advantage in terms of cost, processability, integrability and flexibility in the standard process [16-19]. Among several oxides materials, perovskite system has the advantages, that oxygen vacancies can be easily induced into the system by synthesis process itself. So, we have chosen a Lanthanum calcium manganites ($La_{0.7}Ca_{0.3}MnO_3$) as an active material because its shows fascinating physical and electrical properties and introducing small amount of reduced graphene oxide (rGO) as a nano-fillers in it to study the RS effects of it [20-22]. In this paper, we have studied the transport properties in LCMO (perovskite manganite)-rGO nanocomposite system at different temperature for RS application.

Experimental setup

$La_{0.7}Ca_{0.3}MnO_3$ (LCMO) polycrystalline sample was prepared using standard solid-state route. The LCMO powder is mixed with reduced graphene oxide (rGO) to make (1-x)LCMO.(x)rGO nanocomposite samples with varying the value of x from 0.001 to 0.005. X-ray diffraction (XRD), Field effect scanning electron microscope (FESEM), Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), Transmission electron microscopy (TEM) measurements are performed to see the structural properties of the samples. Keithley source meter was used to measure I(Current)-V(Voltage) characteristic of all the composite samples using two probe methods. The low temperature measurements were done in the liquid Nitrogen atmosphere using Keithley source meter interfaced with cryogenic probe station.

Results and Discussion

The structural properties of (1-x) LCMO. (x)rGO nanocomposite samples were analyzed by X-ray diffractometer and Raman spectroscopy. The Fig 1(a) and 1(b) shows the XRD pattern and Raman spectra of the nanocomposite samples respectively. The XRD pattern of LCMO shows the presence of secondary phase. As XRD did not detect the small concentration of rGO in the nanocomposite, Raman spectra show the presence of rGO in the nanocomposite with the Raman shift corresponding to D- and G-band at 1352 cm^{-1} and 1583 cm^{-1} respectively.

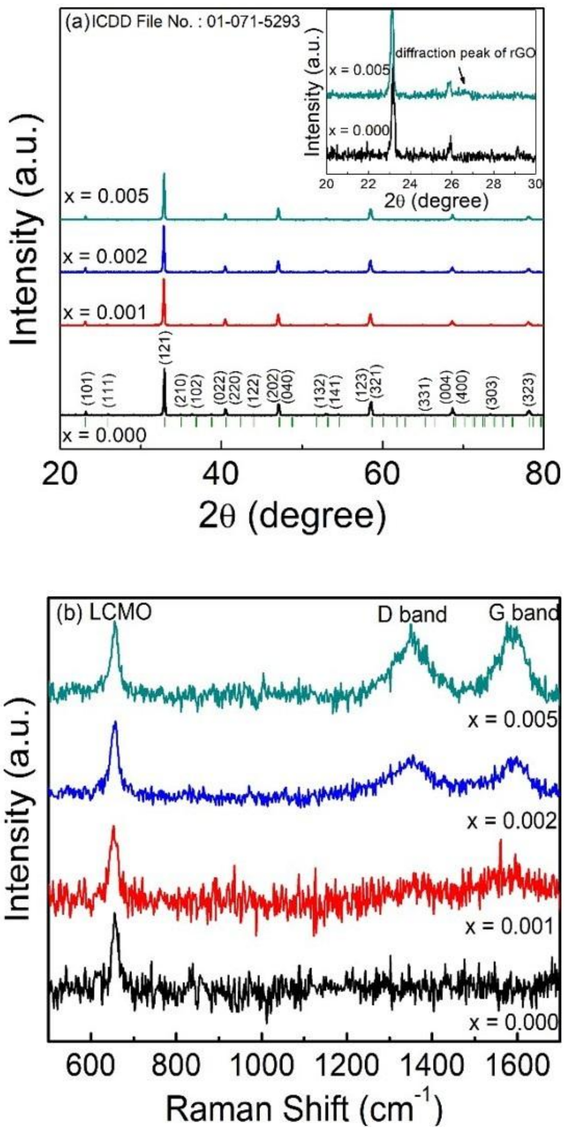

Figure 1: XRD pattern of (1-x)$La_{0.7}Ca_{0.3}MnO_3$.(x)rGO nanocomposite samples [23].

The RS behavior of the samples was study from the I vs V measurement done at room temperature. The I vs V measurement of the LCMO sample is shown in the Fig.2. The I vs V curve is observed after applying a voltage in the forward direction i.e, -3V to 3V. On reversing the direction of voltage, the current follows the same path and hence depicts the Ohmic nature. This exhibits that the LCMO sample does not show any hysteresis.

The I vs V measurement of the sample is shown in Fig.2(a-c). The compliance current is set to 200 mA before starting the experiment to avoid any breakdown. In the given sample, the forming is observed at 10.4 V. Once the sample is activated by electroforming, the I vs V measurement is performed. A pinched hysteresis I vs V loop is observed and is the fingerprint of a memristor.

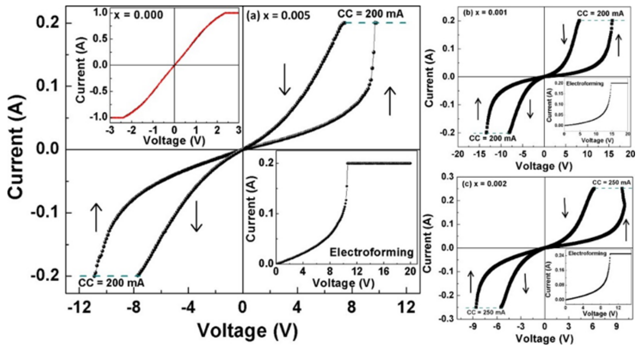

Figure 2: (a) I vs V measurement for (1-x) $La_{0.7}Ca_{0.3}MnO_3$.(x)rGO nanocomposite sample with x=0.005. (b) I vs V measurement of first cycle of x=0.001 and **(c)** x=0.002 [23].

As the voltage is increased the non-linear nature of current is observed and further increase in the voltage leads to a sudden change in the current at 9.8 V (V_{set}). The system changes its states from high resistance state to low resistance state. This is known as the SET process. Further, on application of negative voltage, the samples change its state of resistance from low resistance state to high resistance state at -9.4 V (V_{reset}) through the RESET process. The linear I vsV curve for x = 0.001 and 0.002 are shown in Fig. 2(b) and (c) respectively. It shows the forming process of both the samples. The process is repeated for performing the I vs V measurements of the other samples also. From the I vs V curves of all the samples under study, the steep increase in current region as SET state is observed and becomes more steeper for x = 0.005 as compared with the other samples. This suggests that the numbers of oxygen vacancies are lesser in the small concentration of x sample and hence it is not enough to make the filament type-conducting path within the samples. The steep increase in the current with increasing voltage for x = 0.005 may be due to the increase in the oxygen vacancies in the sample. Increase in oxygen vacancies allows formation of conducting path and it is the reason for the change of resistance state of the samples.

The plot of resistivity vs. temperature plot show that with the increase in rGO concentration, there is a shift in the metal to insulator transition temperature towards the low temperature range. This is shown in the Fig 3. The decrease in metal to insulator transition may be due to the creation of strain in the sample and hence a change in Mn-O-Mn angle.

To observe the effect of temperature in the RS behavior of the LCMO-rGO samples, the I vs V curve has been studied at different temperature, varying from 100 K to 250 K (Fig 4 a-b). The loss in the hysteretic nature was observed at lower temperature region (< 200 K). Further, as the temperature increases a decrease in the operating voltages are observed and are shown in Fig. 4(c).

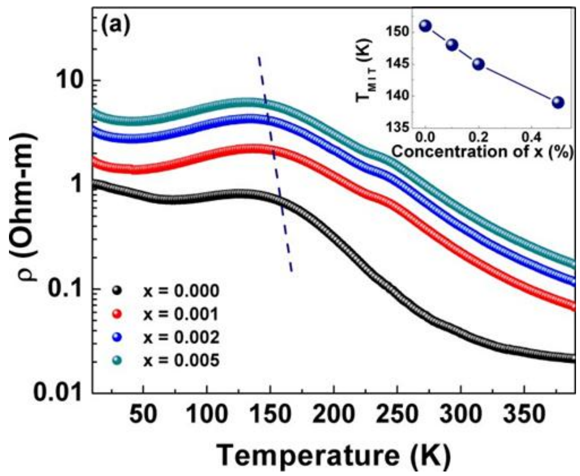

Figure 3: Resistivity vs. Temperature plot of (1-x) $La_{0.7}Ca_{0.3}MnO_3$.(x)rGO samples. The dashed line represents the metal-insulator transition temperature [23].

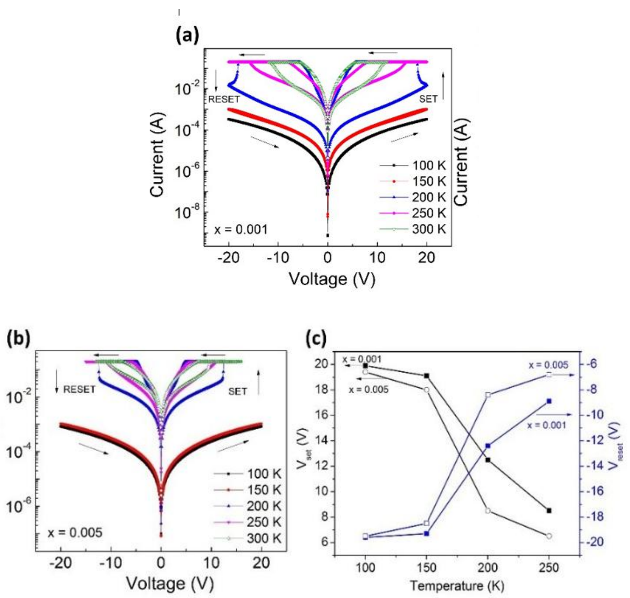

Figure 4: Temperature dependent I-V curve of (1-x) $La_{0.7}Ca_{0.3}MnO_3$.(x)rGO in semilogarithmic scale for **(a)** x=0.001 and **(b)** x = 0.005 varying from 100 K to 300 K. **(c)** Variation of operating voltages V_{set} and V_{reset} as a function of temperature [23].

Conclusion

The presence of both the phases in the nanocomposite is confirmed by structural characterization using XRD and Raman spectra of (1-x)$La_{0.7}Ca_{0.3}MnO_3$.(x)rGO. The I vs V curve of LCMO shows linear nature and retrace its path when the voltage is swept in the opposite direction, however, the nanocomposite sample with x = 0.005 shows the hysteresis in the I vsV curve and can be attributed to the resistive switching behavior. The material under study shows pinched off I vs V nature, which may be due to the memristor-type behavior. The RS behavior in the nanocomposite has been explained using conducting filament model and may be attributed to the creation of oxygen vacancy in the nanocomposite. The resistivity vs. temperature plot suggests that the metal insulator transition temperature decreases with the increase in the concentration of rGO in the sample. This may be attributed to the decrease in the concentration of oxygen and due to change of the Mn-O-Mn angles. The transport mechanism behind the I vs V curve gives better fit for space charge limited conduction in the linear variation of the current for the low bias region (I ∝ V), exhibits quadratic dependence (I ∝ V 2) for the intermediate voltage region (Child's law region) and steep increase in current region for the high bias region. The temperature dependent I-V nature suggests that the sample losses its hysteresis nature with the decrease in temperature. In addition, there is decrease in the V_{set} and V_{reset} is observed with increasing temperature.

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