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Temperature Dependent Dielectric and AC Electrical Response of $0.5La_{0.7}Sr_{0.3}MnO_3$ – 0.5Polyvinylidenefluoride (PVDF) Hybrid Nanocomposites

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ABSTRACT
We have investigated temperature dependent dielectric constant and ac electrical properties of $0.5La_{0.7}Sr_{0.3}MnO_3$ – 0.5Polyvinylidene fluoridemultiferroicnanocomposites. The value of dielectric constant is found to ~ 1400. Z, Z' and Z" also increased with increase in temperature. The Nyquist plots have been fitted using parallel combinations of resistances - constant phase element circuits showing dominant role of grain boundaries of $La_{0.7}Sr_{0.3}MnO_3$ and modified grain boundaries due to thin Poly(vinylidene fluoride) layer situated in inter-granular space of two grains of the samples.

Keywords: LSMO, PVDF, Multiferroic

INTRODUCTION
In last few years, polymer based multiferroic composites have gained lots of interest in research field because they are naturally degradable do not produce chemical [1, 2].The multiferroiccomposite possesses simultaneous existence of Spontaneous Magnetization (Ms) and Ferroelectric Polarization (Ps) and is easily tunable with applied electric and magnetic field respectively [3].Such materialsare widely used in data storage device, replacement ofcapacitor and inductor in resonant circuits with a single component, sensors etc. The multiferroic property in single phase material isquite rare so people are trying to preparing composites materials to achieve multiferroicity in room temperature.Recently few research groups are studied the multiferroic properties of polymer based multiferroic composites. C. Thirmal*et al.* have reported the MD effect in PVDF – $La_{0.7}Sr_{0.3}MnO_3$ polymer based nanocomposites film prepared by dispatching LSMO on PVDF gel[2]. O. D. Jayakumar*et al.* have investigated the MD coupling on Fe_3O_4 – SiO_2- PVDF nanocomposites film[4].Though multiferroic materials are associated with electrical and magnetic properties, theknowledge of the detailed ac electrical characteristic and its temperature dependency is requisite for possible implementation for industrial application.

In this context, we have prepared multiferroics$0.5La_{0.7}Sr_{0.3}MnO_3$(LSMO) – 0.5 Polyvinylidene fluoride (PVDF) nanocomposites and carried out the temperature dependent dielectric (ε) and ac electrical properties.The sample possesses high ε value due to the Maxwell- Wagner interfacial polarization at the interphase region of LSMO grain. Nyquist plots are fitted using parallel combinations of resistance (R) – constant phase element (Q) circuit reveals the dominant role of grain boundary and modified grain boundary interface in the composites.

EXPERIMENTAL DETAILS

To prepare multiferroic nanocomposites 0.5 LSMO – 0.5 PVDF first we have prepare LSMO nanoparticles through chemical pyrophoric reaction process [5- 7],then we dispatched LSMO nanoparticles in PVDF gel prepared by dissolving PVDF beads in N,N-Dimethylformamide(DMF). Structural characterization has been done X-ray diffraction (XRD) (Bruker D8 Advanced) with monochromatic Cu-Kα radiation and high resolution Field Emission Scanning Electron Microscopy (FESEM) (Carl Zeiss, Supra TM[40]) technique. Complex impedance studies have been carriedout through impedance analyzer (Keysight, E4990A)using 2 probe measurement technique. For temperature dependentelectrical measurement, we have used proportional–integral–derivative (PID) controlled portable furnace.

RESULTS AND DISCUSSION

The XRD patterns and FESEM micrograph for 0.5LSMO –0.5PVDF nanocomposites has been shown in Fig. 1 (a) and (b) respectively.The XRD peaks indexed using JCPDS software for both LSMO and PVDF phases. The High resolution FE-SEM micrographs show the absence of any phase segregation and good chemical homogeneity of the samples. They also show that particles size is in nanometric regime.

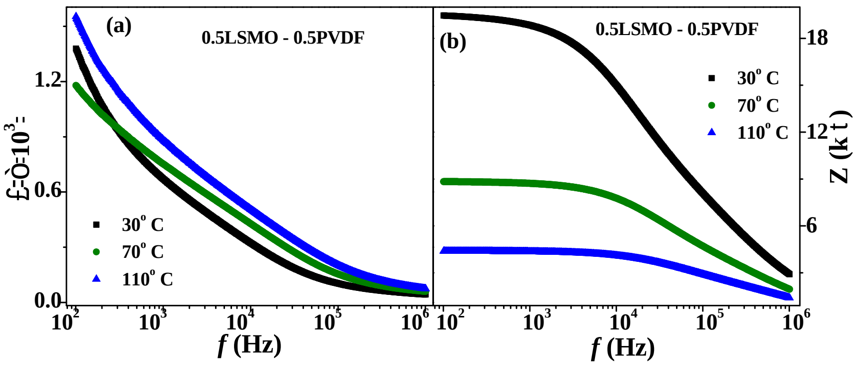

Fig. 1 (a) XRD patterns and (b) FESEM micrograph for 0.5LSMO – 0.5PVDF nanocomposites

ε as a function of frequency (*f*) at different temperature has been shown in Fig 2 (a). ε has been found to ~1400 at room temperature and increases with increase in temperature. The high value of ε at low *f* region is due to the Maxwell – Wagner interfacial polarization or space charge polarization in the grain boundaries of the nanocomposites [8]. Impedance (Z) vs. *f*plots at different temperature for 0.5LSMO – 0.5PVDF sample has been shown in Fig 2 (b). The value of Z has been decreased with increase in temperature may be due to the smooth conduction of thermally activated charge carriers at the grain boundaries of the nanocomposites. With the increase in temperature, the charge carriers associated with conduction process are gets energy to overcome the grain boundaries barrier attributes the decrease in the impedance [9].

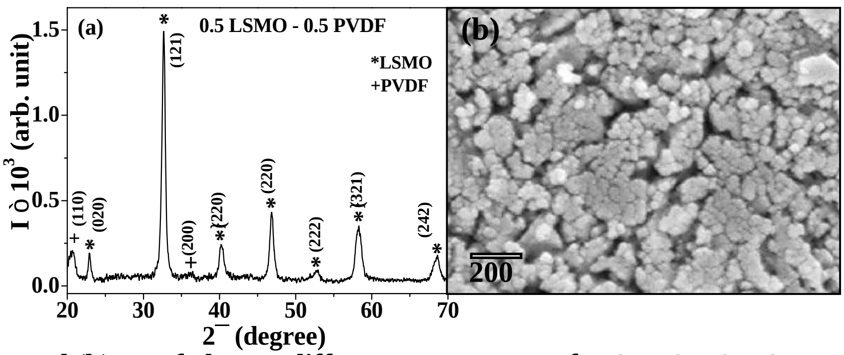

Fig. 2 (a) εand (b) Z vs*f* plots at different temperature for 0.5LSMO – 0.5PVDF nanocomposites

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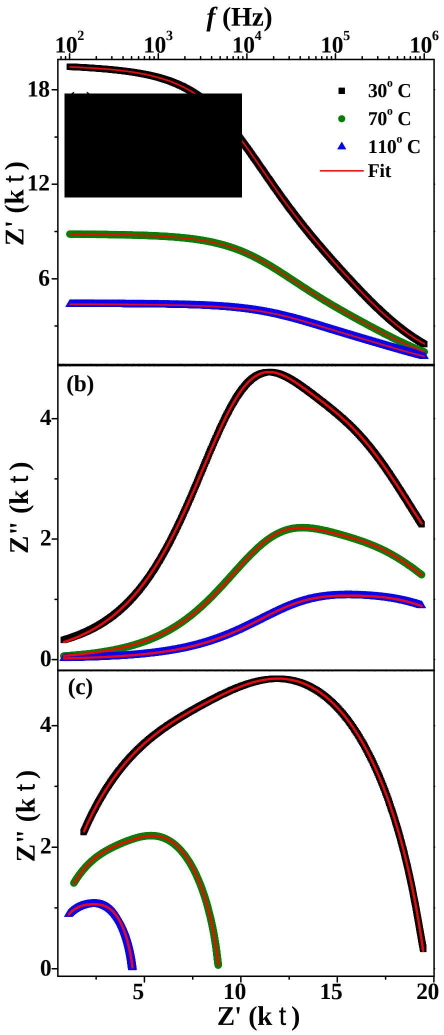

Fig. 3 (a) Z',(b) Z"vs*f* plots and (c) Nyquist plots at different temperature for 0.5LSMO – 0.5PVDF nanocomposites

The value of real part (Z') and imaginary part (Z") of impedance also decreased with increase in temperature (shown in Fig. 3 (a) and (b)).Z' and Z" *vs.f*have been fitted using parallel combinations of R - Q two circuits as shown in inset of Figs. 3(a); first circuit (R_1 - Q_1) corresponds to LSMO grain boundaries and; second circuit (R_2 - Q_2) corresponds to the grain boundaries interfaces in the composites [5, 10, 11].The temperature dependent Nyquist plots for 0.5LSMO – 0.5PVDF nanocomposites are shown in Fig. 3 (c). Nyquist plot shows single semicircle along with a small portion of another semicircle for 0.5LSMO – 0.5PVDF nanocomposites. With the increase in temperature, the semi-circular arc has been decreased due to the temperature dependent conduction process in the grain boundaries in the nanocomposites [12].

CONCLUSIONS

In conclusions, we have prepared the 0.5LSMO – 0.5PVDFmultiferroic nanocomposites and successfully studied the temperature dependent dielectric and ac electrical response. The value of ε_r increased,whereas Z, Z' and Z" deceased with increase in temperature. The high value of ε_r at low *f* region is due to the Maxwell - Wagner interfacial polarization at the LSMO grain boundaries. The decrease in impedance with increase in temperature is due to the enhancement of conduction process through grain boundaries of the LSMO grains.

Equivalent circuit fitting of Nyquist plots reveals the dominant role of grain boundaries and grain boundaries interfaces of the nanocomposites.

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