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Impedance Spectroscopy and Dielectric Study of LSM Thin Films as Cathode for SOFC

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Abstract: *Recently, the great research attention on strontium doped lanthanum manganite (LSM), due to their significant report of transport and electrical properties. When LSM compound material synthesized in thin film form for solid oxide fuel cell (SOFC) cathode it helps to minimize its operating temperature. In the present study LSM thin films were synthesized by spray pyrolysis technique and heat treated at* $900^{O}C$ *for 2 hours. This heat treated samples were used for characterization such as impedance measurement with frequency variation and temperature dependent measurement of dielectric constant. The variation of impedance with frequency indicates the presence of relaxation time and space charge polarization in the system.*

Keywords: *Spray Pyrolysis, impedance, dielectric constant Cathode, LSM, SOFC.*

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I. Introduction

Solid oxide fuel cell (SOFC) has been great research attention due to their high efficiency and easy fuel flexibility [1-4]. Lanthanum strontium manganite are the best known perovskite compound, which is used as cathode for SOFC. But SOFC cathode material in bulk form will be hindered due to high operating temperature of SOFC. When SOFC use for commercialization it is essential to reduce operating temperature of SOFC up to $600^{O}C - 700^{O}C$. For significant performance of SOFC, the cathodic polarization resistance is the important factor which affects working temperature of SOFC [5-6]. To reduce the cathodic polarization resistance it is essential to synthesize LSM in thin film form to provide the higher degree of triple phase boundaries (TPB). In the present study the LSM thin films were synthesized by cost effective spray pyrolysis techniques. The advantage of cathode material synthesize in thin film form are low ohmic resistance, fast start- up time and reduction of operating temperature [7]. In this paper impedance and dielectric constant of sintered LSM thin films were investigated in the frequency range 1Hz to 5MHz and dielectric constant measured with varying temperature.

II. Experimental

The impedance measurement of LSM thin films deposited by spray pyrolysis on alumina substrates were carried out to understand the electrochemical properties of LSM thin films. The LSM thin films were deposited on alumina substrate which heat treated at $900^{O}C$ for two hours. An A.C. two point impedance measurement were done using HIOKO LCR- Q meter, 1mv test signal over the frequency range 100 Hz to 5 MHz and measure phase angle simultaneously. Silver paste was used for making electrical contacts, which served the purpose of electrodes for films deposited alumina substrates.

III. Results And Discussion:

3.1 Impedance spectroscopy:

The complex impedance technique was employed to study the electrical response or transport properties, grain boundary and electrode effect of the material as a function of frequency. Complex impedance technique was also be used to separate the contributions of electrical properties due to grain boundaries, grains and electrode/sample interface in a polycrystalline material [8]. This study involved measurement of real and imaginary parts of impedance with respect to wide range of frequency.

The variation in the real and imaginary components of impedance with frequency for LSM samples at room temperature are shown in fig.1 (a) and (b). The value of Z' is higher at lower frequency region and as the frequency increased the value of Z' decreased rapidly and attained a constant value at high frequency region [9]. The magnitude of Z' decreased with increase of frequency, indicating an increase in A.C. conductivity of the film. The variation of Z” with frequency revealed that Z” values first increased reach a maximum (Z”$_{max}$), towards low frequency showing a peak. Such behavior indicates the presence of the relaxation time in the system. After maximum peak further increased the frequency, Z” decreased and attained constant value at high frequency. This behavior indicates the presence

of space charge polarization in the system[10].

Fig.2 & 3 shows the Bode plots for LSM thin films which reveal that phase angle versus logω. We observed one flat region at low frequencies in impedance versus logω plots.

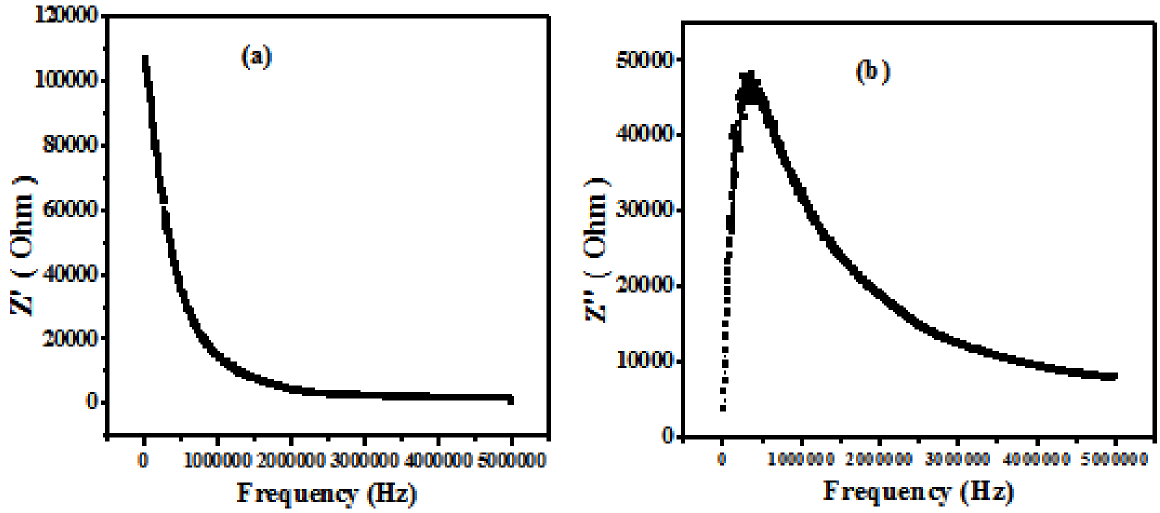

Fig. 1 Variation of (a) real and (b) imaginary components of impedance as a function of frequency for LSM films

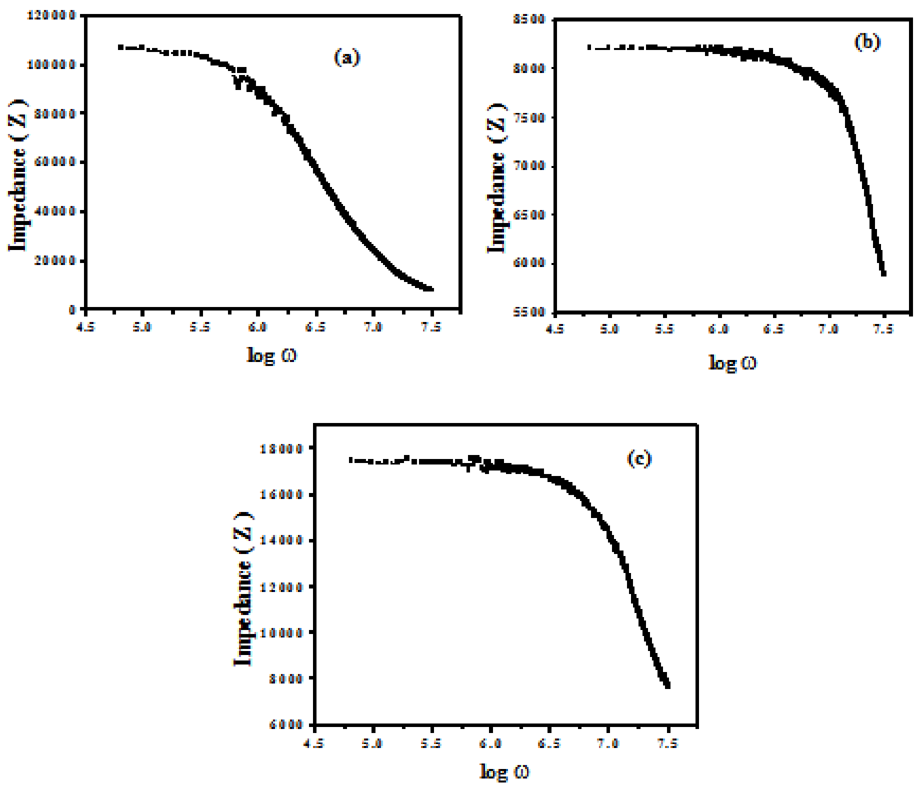

Fig. 2 Bode plots for LSM films (a) X = 0.10, (b) X = 0.20, (c) X = 0.30

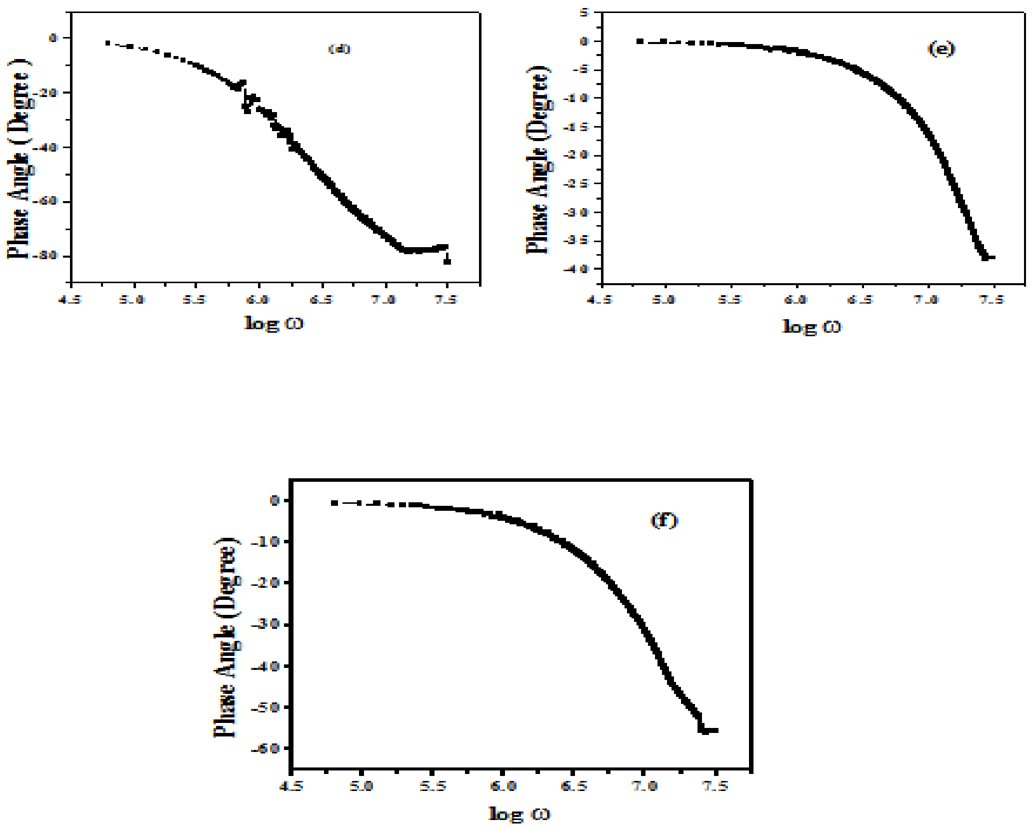

Fig. 3 Phase angle graphs for LSM films deposited on alumina substrate. (d) X = 0.10, (e) X = 0.20, (f) X = 0.30

and two flat regions in phase angle graphs, due to relatively very small ohmic resistance compared to the polarization resistance [11].

3.2 Dielectric constant:

The fig. 4 exhibits the dielectric constant which varies w. r. t. temperature at constant frequency 100 KHz of LSM thin films. The temperature varied from $300^{o}K$ (room temperature) to $813^{o}K$. It is observed from figure that some LSM samples showed sharp transition while some samples broadening of transition, which is characteristic of phase transition from $673^{O}K$ to $723^{O}K$. The dielectric constant value was approximately constant up to temperature $523^{O}K$ but above it there was sharp increase in dielectric constant and reaches maximum at transition temperature (T_C) and then decreases. This was due to accumulation charges at the grain boundaries of LSM particles. For transition temperature $T_C > 0$, the dielectric constant increases with increase in temperature due to growing effect of ionic polarization and dipole polarization which comes in to play relaxation time in the system which was already found in impedance spectroscopy study.

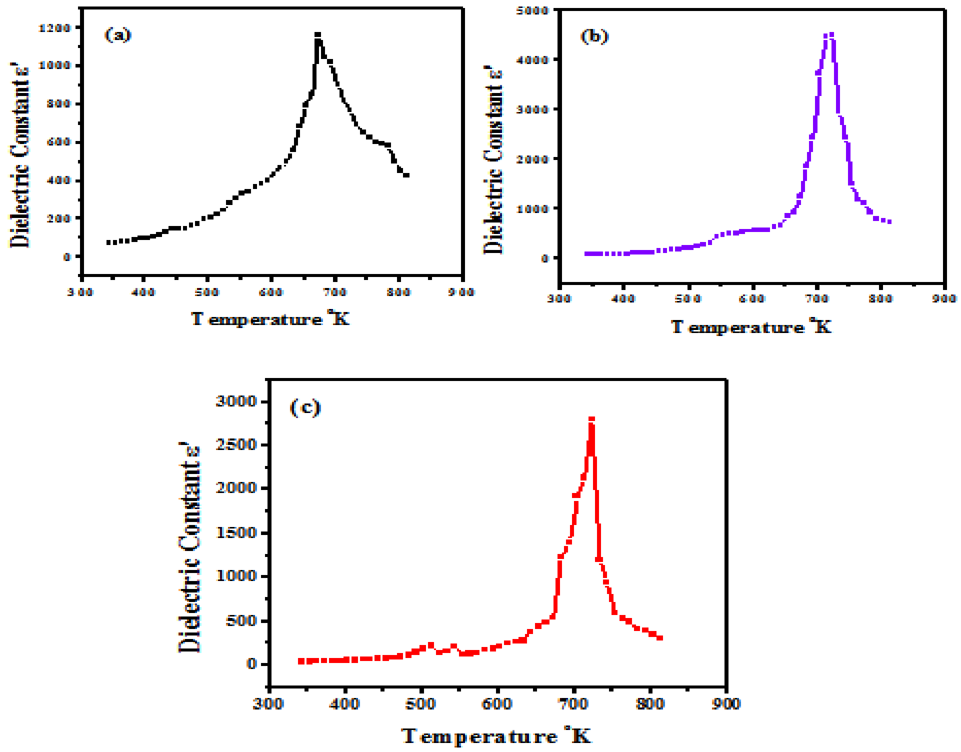

Fig.4. Variation of dielectric constant with temperature (a) X = 0.10, (b) X = 0.20 and (c) X = 0.30

The relaxation time decreases with increases in temperature starting from very low temperature [12-13]. A further increase in temperature however adds to the randam vibrational motion of the molecules, which became less susceptible to the orientation in the field direction and hence dielectric constant decreases. It was also found that as strontium content increased dielectric constant became constant for higher temperature and further increase in temperature suddenly increased and then decreased.

IV. Conclusion

The electrical transport properties of LSM materials used as cathode for SOFC were studied using the technique of impedance spectroscopy and the results were analyzed by Bode plot. As frequency increases A.C. conductivity increases and resistivity decreases and which is applicable for SOFC. The presences of relaxation time were observed both the impedance and dielectric constant graph. The increase in strontium content in LSM was responsible to increase in transition temperature (T_C).

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