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Dielectric relaxation in combustion synthesized perovskite $SmAlO_3$

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Abstract

$SmAlO_3$ perovskite material was prepared by solution combustion technique. The ac conductivity and dielectric properties of the sintered pellet of the sample have been investigated in the frequency range 1Hz to 1MHz and temperature range 303K-783K. The experimental results indicate that the ac conductivity $\sigma_{ac}(\omega)$, dielectric constant (ε') and loss tangent (ε'') depends on depends on the temperature and frequency. The ac conductivity was found to obey the power law ω^n with and the behavior of exponent n with temperature suggested that the conduction mechanism in the ceramic follows OLPT model. The obtained results are compared to the principal theories that describe the universal dielectric response behavior. The value ε' and tan δ were found to be temperature and frequency dependent.

Key words: Perovskite materials, ac conduction, dielectric relaxation, rare-earth aluminate.

Introduction

The study on the perovskite rare earth aluminates is important as they find several applications in non-linear optics, memory devices, solid state lasers, solid electrolytes, chemical sensors, dielectric resonators etc. apart from the academic point of view due to physical properties they exhibits. Perovskite materials with high dielectric constants have been used for technological applications such as wireless communication system, cellular phones and global positioning systems in the form of capacitors, resonators, and filters[1-5]. Since, most of the devices are operated in the ac electrical mode, the investigation of ac electrical conduction of these materials is interesting. Further, characterization of this family of compound is novel as very few works have been reported.

Various relaxation processes seem to coexist in perovskite crystals or ceramics, which contain a number of different energy barriers point defect appearing during the technological process. There for the departure from the ideal Debye behaviour in solid state samples, resulting from the interaction between the dipoles cannot be disregarded[6]. There are defective perovskite materials that exhibit high dielectric permittivity[7,8]. In general, they also show strong dispersion of dielectric spectra in the radio frequency range as well as marked electrical conductivity[9]. Further, defects or non-stochiomertry in perovskite may lead to a mixed ionic and electronic conductivity[10]. These phenomenons were also discussed within the Maxwell-Wagner model in connection with the non-homogeneous structure of compounds[7,11-13]. Oxygen vacancies, resulting from technological processing, influences the electrical properties of the perovskite materials and the oxygen vacancies play an important role in relaxation and transport phenomena[14].

In this paper, we investigate the structural, electrical and dielectric relaxation properties of the perovskite $\mathbf{SmAlO_3}$ ceramics in the temperature range from 303K to 763K and in the frequency 1 Hz to 1 MHz by means of dielectric spectroscopy.

Experimental Procedure

In analogy to our previous work[15], $\mathbf{SmAlO_3}$ was synthesized by solution combustion technique using citric acid as the complexing agent and nitric acid as the reducing agent. Powders of Sm_2O_3 (99.99% purity, Sigma Aldrich), and $Al(NO)_3.9H_2O$ (99.99% purity, Sigma Aldrich) were taken as primary raw materials. The combustion product was annealed at 1000°C for 2 h for the desired phase formation. X-ray powder diffraction (XRD) patterns were recorded with a X'pert Pro Bruker D-8 diffractometer with CuK_α radiation (λ= 0.15418 nm). Micrographs of the polished and thermally etched samples were taken with the help of high resolution scanning electron microscope (FEI Quanta FEG 200). The dielectric measurements were then performed on these samples as a function of frequency at various temperatures using NOVO-CONTROL (Alpha-A) high-performance frequency analyzer.

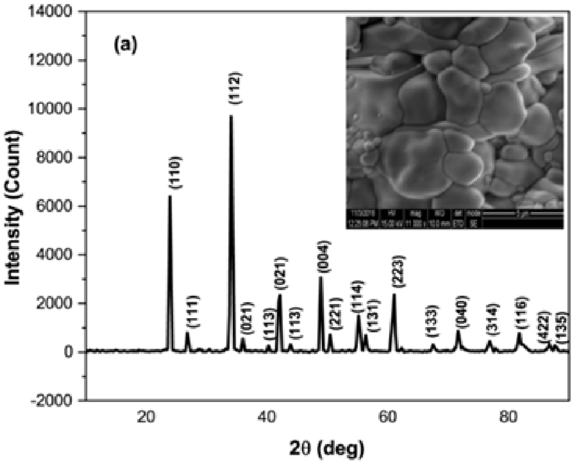

Fig. 1. XRD pattern for $SmAlO_3$ (inset shows HR-SEM of sintered pellet)

Results and Discussion

The room temperature X-ray diffraction pattern of $\mathbf{SmAlO_3}$ ceramics is shown in Fig. 1. All the peaks in the XRD pattern are in good agreement with JCPDS card No. 71-1597, demonstrating the formation of $\mathbf{SmAlO_3}$ phase with orthorhombic perovskite structure. No diffraction peak that could be related to impurity or secondary phase was observed in the XRD pattern. The microstructure of the ceramics was examined by the high resolution scanning electron microscopy (HR-SEM), and shows the polycrystalline nature of the samples with different grain sizes, which are inhomogeneously distributed through the sample surface.

In the frequency domain, the dielectric response of the system subjected to an external oscillating field is fully characterized by complex permittivity,

$$\boldsymbol{\varepsilon^*(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega)},$$

where the real and imaginary components are store and loss, respectively. The relation of frequency (f) with dielectric constant (ε') and loss tangent **(tanδ)** at various temperatures for $\mathbf{SmAlO_3}$ is described in Fig. 2(a) and 2(b) respectively. The value of ε' decreases to a constant value with increase in frequency in the $\mathbf{SmAlO_3}$ compound, which may be

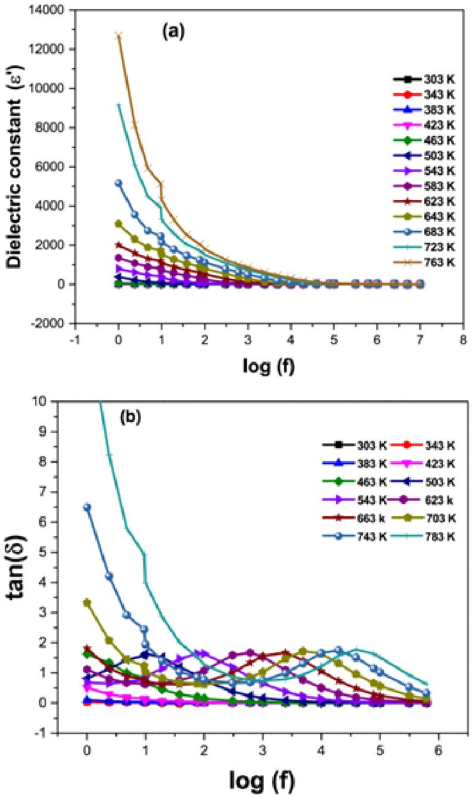

Fig. 2. (a) Variation of dielectric constant (ε') and (b) loss tangent **(tanδ)** with frequency for $\mathbf{SmAlO_3}$.

attributed to the fact that, at lower frequency region the permanent dipoles align themselves along the direction of the field and contribute to the total polarization of the dielectric material. The variation of ε' with frequency explains relaxation phenomena of the material, which are associated with a frequency-dependant orientation polarization. On the other hand, at higher frequency the variation in field is too rapid for the dipoles to align themselves in the direction of field, i.e., dipoles can no longer follow the field, so their contribution to the total polarization and hence to the dielectric permittivity become negligible. Therefore the dielectric constant (ε') decreases with increase in frequency. The high value of dielectric constant (ε') at lower frequencies, increases with decreasing frequency and increasing temperature correspond to bulk effect of the system. At all the frequencies, the dielectric constant increases with rise in temperature. The increase in ε' is more prominent at lower frequencies. The loss tangent spectra (Fig. 2(b)) characterized by peak appearing at a characteristic frequency suggest the presence of relaxing dipoles in the samples. The strength and frequency of relaxation depend on characteristic property of dipolar relaxation.

The tangent loss peaks shift towards the higher frequency side on increase in temperature. The high values of ε' interestingly observed only at very high temperature and very low frequencies may be attributed to the fact that the free charges buildup at interfaces within the bulk of the sample (interfacial Maxwell–Wagner (MW) polarization)[16] and at the interface between the sample and the electrodes space charge polarization .

In order to elucidate this point, the frequency dependant ac conductivity (σ_{ac}) at various temperatures for $\mathbf{SmAlO_3}$ ceramics is plotted in Fig. 3(a). The conductivity increases with increasing frequency and increasing temperatures. The conductivity curve shows dispersion in the low frequency region. This type of trend in conductivity is very similar to that in ionic conducting ceramics[17].

Notice that at low frequencies, random diffusion of charge carriers via hopping gives rise to a frequency independent conductivity. The increasing trend of σ (ω) in the low frequency range may be due to the disordering cations between the neighboring sites and the presence of space charges that vanishes at higher temperatures and frequencies. The dispersion is narrowed at higher frequencies. The ac conductivity of any dielectric or semiconducting material can be expressed in terms of Jonscher's power law[6]: $\boldsymbol{\sigma(\omega) = \sigma_0 + A\omega^n}$,

where ω is angular frequency, n is a constant $(0 < n < 1)$ and σ_0 is the low-frequency conductivity. We have extracted the value of exponent n by fitting ac conductivity data using Jonscher's power law. The value of 'n' decreases with temperature, reaches a minimum and then increases is depicted in Fig 3(b). This behavior is in accordance with the variation of 'n' in overlapping large polaron tunneling model (OLPT) of ac conduction[19].

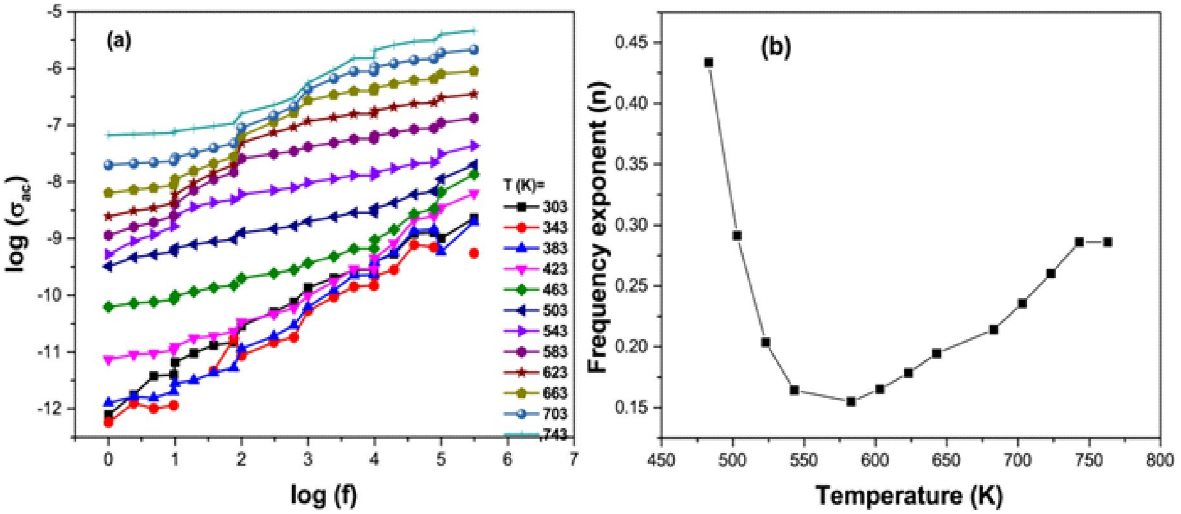

Fig. 3. (a) Variation of ac conductivity (σ_{ac}) with frequency and (b) power exponent n with temperature for $SmAlO_3$.

Conclusion

The frequency dependent dielectric dispersion of perovskite $\mathrm{SmAlO_3}$ ceramics were prepared by solution combustion technique and is investigated in the temperature range at 303 to 743 K and in the frequency range at 1 Hz to 1MHz. The crystal structures of ceramics determined by powder X -ray diffraction shows orthorhombic phase at room temperature. The variation of dielectric constant (ε′) and loss tangent (tanδ) may be attributed to hopping of trapped charge carriers, which resulted in an extra dielectric response in addition to the dipole response. The behavior of exponent n suggested that the conduction mechanism in these ceramic follows OLPT model.

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