Abstract

Complex perovskite oxides having chemical formula $A(B'^{1/2}B''^{1/2})_3$ (where $A = \text{Ba, Sr, Ca}; B' = \text{Fe, Al}; B'' = \text{Sb, Nb}$) and $A(B'^{1/3}B''^{2/3})_3$ (where $A = \text{Ba, Sr, Ca}; B' = \text{Mg}; B'' = \text{Nb}$) have been synthesized by solid state reaction technique. The X-ray diffraction study is performed to confirm the formation of single phase of these materials. From the analysis of the XRD results it has been observed that the compounds BFS, CFS, BAN, SAN, CAN, BMN and SMN have monoclinic structure, SFS has cubic structure and CMN has orthorhombic structure. The grain size distribution of the samples is determined by scanning electron microscope. The frequency dependence of the dielectric response of these complex perovskite oxides and one series of disordered soda-lime-silicate glass has been investigated using alternating current impedance spectroscopy at various temperatures. A relaxation is observed in the studied temperature range as a gradual decrease in $\varepsilon'(\omega)$ and/or as broad peak in $\varepsilon''(\omega)$ or $\tan\delta(\omega)$. The dipolar and conductivity-relaxation mechanisms have been employed to investigate the dielectric relaxation in these materials. The most probable relaxation times obtained for these materials are found to obey the Arrhenius law. An analysis of real and imaginary parts of the dielectric constant has been performed by considering the distribution of relaxation times as confirmed by Cole-Cole plots. The scaling behaviour of the loss spectra for these materials suggests that the relaxation describes the same mechanism at various temperatures for these materials. The observed electrical data are also analyzed in the framework of impedance, electric modulus and conductivity formalisms. The frequency dependent conductivity spectra at various temperatures follow the Jonscher's power law. A comparison of the frequency dependent spectra of the imaginary electric modulus with imaginary impedance is used to investigate both long range and localized conduction responsible for dielectric relaxation in these materials. We have also analysed the conduction mechanism in BMN in time domain. In time domain, the time decay function $\phi(t)$ is obtained from the values of $M''$ by using a relaxation function in frequency domain, which is found to be a Cole-Cole distribution function. The Kohlrausch-Williams-Watts (KWW) function is used to analyse the time dependent behaviour of $\phi(t)$. 
The electronic structure calculations of the complex perovskite oxides considering ordered structure have been performed by the first principles full potential linearized augmented plane wave method using density functional theory under the generalized gradient approximation. The electronic structure calculation reveals that the electrical properties of the ordered perovskite oxides are dominated by the interaction between the transition metal and oxygen ions. The x-ray photoemission spectroscopy (XPS) study of these oxides has been performed over a wide energy range to verify the electronic structure calculations experimentally. The calculated total and partial density of states (DOS) data are convoluted to explain the observed valence band XPS spectra in these materials. The $^{27}$Al and $^{93}$Nb nuclear magnetic resonance studies of Ba(Al$_{1/2}$Nb$_{1/2}$)$_3$ are also performed at 78 and 73 MHz respectively in the temperature range from 4 K to 295 K to confirm the chemical ordering of Al$^{3+}$ and Nb$^{5+}$ cations in this material.