Chapter 8

Conclusions

In this thesis, we have studied the electrical properties of three series of ordered complex perovskite oxides and one series of disordered soda-lime-silicate glass using alternating current impedance spectroscopy. The first two series of complex perovskite oxides belong to 1:1 ratio having chemical formula \( A(B'_1B''_{1/2})O_3 \) (where \( A = \text{Ba, Sr, Ca}; \ B'^1 = \text{Fe, Al}; \ B'' = \text{Sb, Nb} \)) and the third series belongs to 1:2 ratio having chemical formula \( A(B'_{1/3}B''_{2/3})O_3 \) (where \( A = \text{Ba, Sr, Ca}; \ B' = \text{Mg}; \ B'' = \text{Nb} \)).

The frequency dependence of the dielectric response for these materials are investigated at various temperatures. A relaxation is observed in the entire 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 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 experimental data are modeled with Cole-Cole expression which confirms the deviation from monodispersive Debye process. The scaling behaviour of the loss spectra 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. Data for these materials have been presented to illustrate the benefits of plotting frequency dependent measurements using not only \( \varepsilon'' \) vs \( \varepsilon' \) plots but also \( M'' \) vs \( M' \), \( Z'' \) vs \( Z' \) and \( Y'' \) vs \( Y' \) as well as the frequency explicit plots, namely \( \tan\delta \) vs \( \omega \), \( M' \) and \( M'' \) vs \( \omega \), \( Z' \) and \( Z'' \) vs \( \omega \) and \( Y' \) and \( Y'' \) vs \( \omega \). 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 series of materials.
Due to the change of the values of ionic-radii of the A-site cations and hence the tolerance factor, the crystalline structures as well as the electrical properties of these complex perovskites are found to be different. From the application point of view which is versatile, usually one requires the dielectric materials with high dielectric constant and low loss which varies with the change of the values of frequency and temperature.

The dynamics of the relaxation process (a conduction mechanism) is also studied in the time domain for Ba(Mg\textsubscript{1/3}Nb\textsubscript{2/3})\textsubscript{3}O\textsubscript{3} where a relaxation function which is related to relaxation characteristic of the material is obtained by the time domain method and is found to be a Kohlrausch-Williams-Watts (KWW) function type.

To have a clear information about the transport properties, we have performed the electronic structure calculation of complex perovskite oxides 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 frequency range to verify the electronic structure calculations experimentally. The calculated total and partial density of states (DOS) data are convoluted to explain the observed XPS spectra in these materials. The \textsuperscript{27}Al and \textsuperscript{93}Nb nuclear magnetic resonance studies of Ba(Al\textsubscript{1/2}Nb\textsubscript{1/2})O\textsubscript{3} are performed at 78 and 73 MHz respectively to confirm the chemical ordering of Al\textsuperscript{3+} and Nb\textsuperscript{5+} cations in this material. More electronic structure calculations are required to describe the conductivity mechanism in these materials.