

## Synopsis

Lead based materials have shown to be important materials showing high piezoelectricity, colossal dielectric constant, ferroelectricity along with their good mechanical behaviour and resistance to corrosion. As a result, they have wide range of applications such as sensors, actuators, detectors, batteries, lead-painting pigments etc. However, due to high toxicity of lead and its nature to accumulate in the environment with gradually increasing concentrations, increasingly more health hazards are encountered. In this regard, replacing these lead based materials by lead free material with similar/higher level of dielectric properties is the driving force for research on lead free dielectric materials. In this work, we have studied few double perovskites structured dielectric materials and their solid solutions.

The present work attempts to understand synthesis, structure and dielectric property of lead free double perovskite structure  $\text{BaFe}_{0.5}\text{Nb}_{0.5}\text{O}_3$ ,  $\text{BaFe}_{0.5}\text{Ta}_{0.5}\text{O}_3$  and solid solution of (x)  $\text{BaFe}_{0.5}\text{Nb}_{0.5}\text{O}_3-(1-x)\text{KNbO}_3$  (x= 0, 0.2, 0.4, 0.6, 0.8 and 1). This thesis work organized in seven chapters as given below. First two chapters describe the motivation behind the work and experimental technique used for synthesis and characterizes materials, chapter 3, 4, 5 and 6 described investigated material and chapter 7 summarizes the thesis. Details of chapters as follows,

In **Chapter 1**, a brief introduction of field of ferroelectric is given with their everyday life application; emphasis has been on double perovskite structure materials. Use of lead based materials with impact on human and environment is discussed. With detail literature survey motivation behind work is discussed in this chapter.

**Chapter 2** described the various experimental techniques that are employed to synthesis and characterized materials.

**Chapter 3** explains the low temperature dielectric properties of the  $\text{BaFe}_{0.5}\text{Nb}_{0.5}\text{O}_3$  (BFN) double perovskite ceramic. The BFN ceramic sample with pseudo-

cubic  $Pm-3m$  structure was successfully synthesized by solid state reaction route, and the dielectric property was studied between 20 K and 300 K. The activation energies associated with the charge carriers for polarization at the G and GB regions are found to be  $\sim 17.2$  meV and 2.73 meV, respectively. The impedances of grain (G) and grain boundary (GB) regions are observed to be distinctly different from each other. The contribution from the G and GB regions, to the dielectric relaxation time broadens the distribution at 120 K, however, no structural phase transition is observed between 20 K and 300 K. The Negative Temperature Coefficient of Resistance (NTCR) observed in our sample makes it a potential candidate for temperature sensor application. The short range movement of charge carriers are confirmed from the non-overlapping nature of the normalized imaginary impedance ( $Z''$ ) and modulus ( $M''$ ) plots at different temperatures.

**Chapter 4** explains the charge transport mechanism in  $\text{BaFe}_{0.5}\text{Nb}_{0.5}\text{O}_3$  double perovskite ceramic. It observed that the electronic dc conductivity in BFN ceramic originates due to intrinsic point defects in the grains (interstitials and vacancies) and grain boundaries (vacancies and disordered). The ac conductivity originates due to the bound charges (electrons and ions) along with the oscillation of the accumulated charges at the grain and grain boundary interfaces. The contribution of the bound cations ( $\text{Fe}^{3+}$  and  $\text{Nb}^{5+}$ ) to the ac conductivity was found to show different response than the contribution of the charges at the GB region. Conductivity in grain region follows Mott Variable range hopping model. Below 120 K the conductivity of the charge carriers was found to be supported by acoustic phonons and above 120 K is due to both acoustic and optical phonons.

**Chapter 5** describes understanding of ferroelectric nature of  $\text{BaFe}_{0.5}\text{Ta}_{0.5}\text{O}_3$  (BFT), whether it is displacive, order-disorder or relaxor ferroelectric. The structural and microstructural analysis of BFT shows phase pure sample with distinct grain and grain boundary. The M-W interfacial polarization and dipole or orientational type polarization

contributes to the high value of dielectric constant observed in BFT sample. Temperature dependent dielectric constant shows peak maxima for 100 Hz frequency indicating long range ferroelectric ordering at 400 K. The observed decreased value of dielectric constant maxima with increased frequency along with monodispersive nature and obeying Debye type relaxation confirm the order – disorder ferroelectric nature of our BFT sample.

**Chapter 6** describes solution combustion synthesis, structure and dielectric property of  $(x) \text{BaFe}_{0.5}\text{Nb}_{0.5}\text{O}_3 - (1-x) \text{KNbO}_3$  ( $x = 0, 0.2, 0.4, 0.6, 0.8$  and  $1$ ) (BFN-KN) solid solutions. The synthesized BFN-KN solid solutions by solid state route (SSR) and solution combustion route (SCR) are compared. Through the solution combustion route, the solid solution was found to form at lower temperature than SSR. In BFN-KN solid solution, with increase Fe concentration in solid solution a colour change from light blue (for KN) to dark brown for BFN is observed, which corresponds to a decrease in band gap from 3.4 eV to 1.8 eV. The high value dielectric constant observed for KN is attributed to the large pores and low density of sample, origination prominently due to the M-W interfacial polarization. It is observed that with increasing BFN content, the solid solution shows less number of pores, which leads to enhanced dipolar polarization in BFN-KN solid solution.

This thesis ends with summary and conclusion **Chapter 7**, though each chapter is provided with conclusion and complete list of references.