

### 7.1 Summary of the Present Work

In the present thesis work, structure, phase transition and physical properties of multiferroic solid solution (1-x)BNT-xPT and its composite with  $\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$  have been investigated. The samples were synthesized by conventional solid state ceramic method. The phase purity and crystal structures were characterized by using x-ray powder diffraction in conjunction with Rietveld crystal structure refinement. Microstructure was characterized by scanning electron microscopy. Nature of phase transitions was investigated by temperature dependent dielectric and x-ray diffraction measurements. P-E, M-H and magnetoelectric measurements were carried out to investigate structure property correlations.

The important findings of the present research work are listed below:

(1) The structure and stability region of various crystallographic phases across morphotropic phase boundary region were determined precisely. We have discovered for the first time, a monoclinic (space group Pm) phase in the MPB region of BNT-PT. The room temperature crystal structure of (1-x)BNT-xPT solid solution is shown to be pseudocubic in the  $\text{Pm}\bar{3}\text{m}$  space group for  $x < 0.41$ , monoclinic with Pm space group for the compositions  $x = 0.41, 0.42$  and tetragonal in the  $\text{P}4\text{mm}$  space group for  $x > 0.49$ . For the composition range  $0.43 \leq x \leq 0.49$ , the structure is found to be coexistence of monoclinic (Pm) and tetragonal ( $\text{P}4\text{mm}$ ) phases.

(2) We have discovered for the first time an electric field induced phase transition in BNT-PT. Centrosymmetric pseudocubic phase exhibits significant value of planer electromechanical coefficient ( $k_p$ ) after poling due to this electric field induced phase transition. After poling, the pseudocubic phase transform into monoclinic phase.

(3) We have investigated systematically the nature of dielectric relaxation at high temperatures as well as below room temperatures for various compositions of BNT-PT across MPB. We have shown for the first time that the freezing of polar nanoclusters above room temperature obeys the Vogel-Fulcher law. We have discovered for the first time frequency dependent low temperature dielectric anomaly in all the compositions of BNT-PT across MPB. The dielectric relaxation is found to obey Vogel-Fulcher freezing at cryogenic temperatures, also.

(4) Particulate composite of BNT-PT with NZFO fabricated by conventional solid state ceramic route, exhibits two types of grain morphology having separate ferroelectric (BNT-PT) and magnetic (NZFO) phases like ideal composite. High magnetoelectric coefficient is obtained in the BNT-PT/NZFO composite, which can be very useful for multiferroic based device applications.

## 7.2 Suggestions for Future Work

1. High resolution neutron and synchrotron powder diffraction measurements should be carried out to study the structural phase transition at high temperature in (1-x)BNT-xPT solid solution. It will help to locate the exact position of various phase boundaries and draw the temperature-composition phase diagram.
2. The dielectric constant and loss tangent of (1-x)BNT-xPT was found to be very high at low frequencies. P-E hysteresis also shows the lossy nature for BNT rich compositions. Substitution of compound like  $\text{MnO}_2$ ,  $\text{Mn}_2\text{O}_3$ , etc. should be tried to reduce the hopping conductivity and increase the resistivity of sample.
3. Atomic pair distribution function (PDF) analysis should be carried out to investigate the local structure of (1-x)BNT-xPT ceramics in the pseudocubic composition region  $0.35 \leq x \leq 0.40$ .
4. First principles density functional calculations should be carried out to understand the nature of phase transition at low temperatures where anomaly in the dielectric permittivity is observed but no clear signature of structural phase transition is seen in the diffraction study.
5. It will be interesting to investigate the ME response below room temperature in BNT-PT/NZFO composite where superparamagnetic character is anticipated in magnetic phase.