RESUME
There has been a renewed interest in the magnetoelectric (ME) phenomenon in case of ferroic systems due to the occurrence of giant magnetoelectric coefficient exhibited by the bilayer and multilayers of different ferroic compositions. The bilayer and multilayers have exhibited both the longitudinal and transverse ME properties, quite distinct from each other in terms of their dependence on the applied dc magnetic field ‘H’. Current research is being carried out to understand the basic physical processes responsible for the observed giant magneto electric coupling and also to realize the possible configurations of the ME devices. Another interesting feature is the work on nanocomposites, which has shown a possibility of occurrence of enhanced magneto-electric coupling in these compounds.

Owing to these observations, the thesis is an attempt of furtherance the work on ferroic materials, in the form of nanocomposites. The materials exhibiting the magnetoelectric phenomenon could be classified in to four different classes mainly
1) Compounds with perovskite-type structure
2) Compounds with hexagonal structure
3) Boracites
4) BaMF₄ compounds with M to be a divalent cations

All the classes discussed above are the single-phase compounds. It has been observed that the magnetoelectric susceptibility exhibited by these compounds is insufficient to realize ME useful magnetoelectric device. Nevertheless Wood and Austin have proposed various devices based on ME phenomenon, operating at optical, radio frequency and low frequency regions. Since 1993 the new class of magnetoelectric compounds investigated extensively is multiferroics.
The multiferroics could be in the form of particulate composites, nanocomposites or multilayers of ferrimagnetic and ferroelectric species. The magnetoelectric effect in these occurs as a product property. Here the stress induced in the ferrimagnetic/ferroelectric materials, because of application of an external magnetic field gets coupled to the ferroelectric system. In turn the ferroelectric system produces dielectric displacement because of the induced stress. Thus application of an external magnetic field produces useful dielectric displacement in the multiferroic compound, which causes the longitudinal or transverse voltage across the sample depending on the direction of applied magnetic field and output measured from the sample.

There are number of single phase materials as listed in the thesis which exhibits magnetoelectric phenomenon, but amplitude of ME voltage not sufficient. Post 1993 research is mainly on the giant ME coupling observed for MLL and ME composites. Generally ferrite-ferroelectric composites possess the magnetostrictive property and piezoelectric property.

There are various review articles published in literature which cover the properties and applications of different ME compounds. The thesis briefly presents an abstract of these activities. It has been realized that the multilayer magnetoelectric systems exhibits a giant magnetoelectric coupling, but the mechanical strength of these compounds is insufficient to machine the product. The magnetoelectric composites on the other hand exhibit a lower resistivity so that the polling of the piezoelectric phase becomes difficult. These two difficulties could be overcome by employing magnetoelectric nanocomposite. The present thesis is one of the attempts in this direction.
Owing to the discussions above the thesis deals with investigations on LSMO-PZT nanocomposites and MLLs. The nanoparticles of LSMO and PZT are synthesized using hydroxide co-precipitation followed by ceramic processing at relatively low temperature. The composites investigated possess the formula $x\text{LSMO} + (1-x)\text{PZT}$ for $x=0.15, 0.2, 0.25, 0.3$ and $0.4$. The reason for selecting the LSMO as the magnetic phase is the similarity of crystal structure between LSMO and PZT. As the composites are formed using nanoparticles there occurs a possibility of diffusion of Mn and Ti ions between the LSMO and PZT matrices. The inter diffusion of Mn and Ti may exhibits a characteristics change in the physical properties of the composites. Analysis of this feature is interesting in terms of the basic physical properties of the ME composites. Another attempt reported in the present thesis is studies on multilayer laminates of PZT and composite of LSMO+PZT. Details about synthesis of LSMO and PZT by chemical route, their individual electrical and magnetic properties, fabrication of composites and multilayer laminates form an important part of the present thesis.

The X-ray analysis of the composites has been performed to determine the formation of the composite phase. The composites are sintered at three different sintering temperatures $T_s= 1100^\circ C, 1175^\circ C,$ and $1250^\circ C$. The observations of x-ray diffraction suggest that required composite phase is formed, though it is observed that a fractional diffusion of Mn and Ti ions occurs at the interfacial region. The increase in sintering temperature is observed to lead to the formation of micron size grain compacts of LSMO and PZT and a graded interface.

LSMO is a ferromagnetic material, which possesses a moderate to high value of electrical conductivity, while the PZT is a ferroelectric phase having very low conductivity (i.e. an insulator). Therefore the conductivity
as well as complex impedance of the LSMO-PZT composite should depend on the variation of the conductivities of the individual phases and also on the property of the interface region.

The variation of log \((\sigma_{ac} - \sigma_{dc})\) with log (\(\omega^2\)) for the LSMO-PZT is a linear, which confirms that the conduction in composite systems is due to small polarons. The log\(\rho\) verses 1000/T behaviour shows a NTCR and a mild structure in the temperature region 350K to 450K for Ts= 1100°C. This feature is attributed to the occurrence of barrier layer at the LSMO-PZT interfaces. The \(\rho\) decreases with x, while it increases with Ts. These features are attributed to the formation of local regions of 3-0 connectivity and formation of grain compacts of LSMO and PZT.

From the observations of complex impedance spectrum it is confirmed that there occurs a partial diffusion of Mn and Ti ions between PZT and LSMO matrices. The effect of diffusion has been visualized in the drop centered complex impedance spectra.

The variation of electrical permitivity \(\varepsilon_r\) as a function of temperature and x for the sintering temperature Ts=1100°C, 1175°C and 1250°C are analyzed in details. The observations are

1) The \(\varepsilon_r\) at 1KHz is sufficiently high and is characteristic of relaxor material or interfacial polarization.

2) \(Q\) on the other hand is sufficiently low which is also a characteristic feature of relaxor material.

3) The \(\varepsilon_r\) is observed increase as a function of x up to x= 0.3, while the quality factor \(Q\) decreases with increasing x.

4) The \(\varepsilon_r\) at 1KHz passes through a diffused phase transition (DPT) in the temperature range 350K to 450K.

5) Though a DPT is observed, the overall trend of \(\varepsilon_r\) with temperature is a slowly varying \(\varepsilon_r\).
6) It could be seen that the structure as in the εr is observed only at low frequency.

7) The frequencies up to which a structure is seen in the temperature interval 350K to 45K in the behaviour of εr decreases as Ts increases.

8) The εr is observed to decrease with increasing Ts and as expected the Q increases with increasing Ts. In spite of this the magnitude of εr and Q are characteristic of relaxor material.

From the observations above and XRD and complex impedance spectra it is predicted that the LSMO-PZT interfaces become graded as the sintering temperature Ts increases. Also the interfacial polarization is an important feature of the ME composites.

Present thesis mainly discusses the static as well as dynamic magnetoelectric voltage measurement methods. It reports an indigenous and laboratory scale setup to measure the magnetoelectric voltage v as a function of h and H both. The earlier report indicates that the instrumentation used for measurement of α and β is costly and imported test and measurement (TM) equipment are required. On the other hand, the experimental design in our laboratory is an indigenous one and uses cost effective TM components.

Using the ME setup designed in the laboratory magnetoelectric coefficients α and β are determined. From the observations it could be said that the magnitude of α as observed for the composite sintered at Ts=1250°C possess the magnitudes of α comparable with the observation on LSMO-PZT MLL compounds reported earlier. The α is observed to show a systematic variation with x and Ts. An empirical model is devised to explain the variation of α with x.