

High dielectric constant material is an interesting topic in materials science due to its applications in capacitor materials, which leads to miniaturization of electrical and electronic devices. Increasing demands of high dielectric constant material, such as ceramic capacitors leading to the development of ceramic capacitor technology. The dielectric constant of barium titanate ($\epsilon_r \sim 10^3$) was first reported by Thurnauer in 1941. The temperature dependent dielectric constant of barium titanate has limitation towards its applications in industry. Temperature independent dielectric constant materials over a wide frequency range are playing very important role in ceramic capacitor industry. The perovskite oxides (ABO_3) have very interesting dielectric, ferroelectric, piezoelectric and optoelectronic properties. These properties are monitored with partial substitution of either A or B site metal ions with other transition metal ions having similar ionic size. The valency, radius and coordination number of substituents are the important parameter for determining their dielectric properties. The perovskite having tolerance factor one exhibits ideal cubic structure, whereas if tolerance factor is more than one indicates the structural deviation from cubic to hexagonal. The $AMnO_3$ (A= Ba and Sr) type of perovskites adopt hexagonal perovskite structure in which AO_3 layer stacked along c axis. Tolerance factor of $BaMnO_3$ was 1.089 shows hexagonal perovskite structure in which BaO_3 layers are hexagonally close packed. The intermediate size of strontium cation acquired 50% hexagonal close packing and 50% cubic close packing.

The materials having permanent dipole moment which can be align in presence of electric field comes under ferroelectric materials. High dielectric constant of barium titanate was due to its ferroelectric behavior. In ferroelectric material atoms are displaced in a particular orientation from their original lattice point resulting spontaneous polarization due to non-centrosymmetric structure. Similar polarization orientation present in volume region of

material is known as ferroelectric domains. The energy of domains is similar at zero field and in a strain free condition, whereas on application of electric field all domains are align in the direction of the field. At very high field permanently reorientation of polarization of domains was observed. Ferroelectric materials are very important, this category of materials shows wide range of applications such as actuators, ultrasonic transducers, paraelectric detectors, piezoelectric sonar, high dielectric capacitors and ferroelectric memories.

Magnetic properties of ceramic materials are also very important. This class of materials possesses variety of application such as data storage, tunnel junction and spin valves. The magnetic ceramic materials have some special properties like magnetic coupling, low loss and high electrical resistivity, which can be tuned by varying the structure and composition of material. With the smaller sizes of nanoparticle less than 100 nm surface to volume ratio increases resulting the number of atoms on the surface of nanocrystals, therefore variation in magnetic properties with change in structure in nanoscale region is observed in comparison to bulk material. The magnetic properties of nanoparticle are affected by particle size, morphology, and chemical composition.

In the present work synthesis of hexagonal perovskites oxides with different composition using semi-wet route and chemical route. All the synthesized ceramics were characterized by various physicochemical techniques to study the crystal structure, particle size and shape whereas dielectric, ferroelectric and magnetic properties of materials were studied in detail.

The aim of the present work is to investigate (a) crystal structure (b) microstructure (c) elemental analysis (d) particle size (e) electrical and dielectric behaviour of the following materials prepared by different routes.

- $\text{Ba}_4\text{YMn}_3\text{O}_{11.5-\delta}$ (BYMO) by chemical route
- $\text{Ba}_6\text{Y}_2\text{Ti}_4\text{O}_{15}$ (BYTO) by semi wet route
- $\text{Ba}_4\text{YMn}_{2.95}\text{Fe}_{0.05}\text{O}_{11.5-\delta}$ (BYMFO-05) by chemical route
- $\text{Ba}_4\text{YMn}_{2.90}\text{Fe}_{0.1}\text{O}_{11.5-\delta}$ (BYMFO-1) by chemical route
- $\text{Ba}_4\text{YMn}_{2.80}\text{Fe}_{0.2}\text{O}_{11.5-\delta}$ (BYMFO-2) by chemical route
- $\text{Ba}_6\text{Y}_2\text{Ti}_{3.95}\text{Fe}_{0.05}\text{O}_{15}$ (BYTFO-05) by semi wet route
- $\text{Ba}_6\text{Y}_2\text{Ti}_{3.90}\text{Fe}_{0.1}\text{O}_{15}$ (BYTFO-1) by semi wet route
- $\text{Ba}_6\text{Y}_2\text{Ti}_{3.80}\text{Fe}_{0.2}\text{O}_{15}$ (BYTFO-2) by semi wet route

Chapter I This chapter contains a brief introduction of the subject describing briefly the technical investigations reported in the field of perovskite oxides and hexagonal perovskite oxides. This includes the effect of isovalent, heterovalent and valence compensated substitutions on the dielectric, ferroelectric and magnetic properties.

Chapter II This chapter describes the details of experimental procedure used for the synthesis, characterization and application of these hexagonal perovskite oxides. TG/DTA has been used to characterize the materials that exhibit a weight change due to decomposition or dehydration of the precursor powder. Thermo-grams of the precursor powder materials carried out in static air from room temperature to $1000\text{ }^{\circ}\text{C}$ at a heating rate of $10\text{ }^{\circ}\text{C min}^{-1}$. Powder X-ray diffraction have been used to study crystal structure of the material. Scanning Electron Microscopy gives an idea of formation of microstructure of these materials. Energy Dispersive X-ray

spectroscopy (EDX) (elemental mapping techniques) has been used to determine the composition of elements in the materials. Transmission Electron Microscopy (TEM) has been used for determination of their size and shape of the particle. Dielectric and electric properties which are characteristic of all the ceramics were measured as a function of temperature (300-500 K) in the frequency range 100Hz-1MHz with the help of PSM 1735 (Newton's 4th U.K Limited) LCR Meter. Ferroelectric properties of the material were recorded with the help of PE loop tester (Marine India). Magnetic measurements were performed on a superconducting quantum interference device (SQUID) (Quantum Design, MPMS 3).

Chapter III The detailed synthesis, characterization and application of the $\text{Ba}_4\text{YMn}_3\text{O}_{11.5-6}$ (BYMO) ceramic were described in this chapter. Single phase formation of material was confirmed by XRD sintered at $1100\text{ }^\circ\text{C}$ for 12.hrs. The presence of Ba, Y, Mn and O elements with their atomic percentages were confirmed by EDX spectra. The formation of intermediate are describes by TG/DTA analysis. Transmission electron microscopy shows the material acquired hexagonal shape and having average particle size of $45\pm 10\text{ nm}$. PE hysteresis suggests decrease in remanent polarization with temperature. Temperature and frequency dependent dielectric constant, dielectric loss and conductivity were also extensively studied. Impedance analysis shows that the presence of semiconducting grains and insulating grain boundaries.

Chapter IV Effect of partial substitution of Fe at the site of Mn was reported in this chapter. Samples of $\text{Ba}_4\text{YMn}_{3-x}\text{Fe}_x\text{O}_{11.5}$ with $x= 0.05, 0.1$ and 0.2 were synthesized by chemical route. X ray diffraction pattern confirms single phase formation of ceramics sintered at $1100\text{ }^\circ\text{C}$ for 12 hrs. EDX and elemental mapping shows the presence of corresponding elements with their atomic percentages. Rietveld refinement specifies the space group of material as $R\bar{3}m$.

Dielectric constant decrease very drastically on increasing iron amount. Remanent polarization is also decrease with increasing Fe dopant concentration. A detailed study of temperature dependent magnetization was carried out.

Chapter V In this chapter $\text{Ba}_6\text{Y}_2\text{Ti}_4\text{O}_{17}$ (BYTO) ceramic was synthesized by semi wet route and single phase formation of the material was observed by XRD at $1400\text{ }^\circ\text{C}$. The intermediate formation (Ba_2TiO_4 , BaTiO_3 and Y_2O_3) was confirmed by TG/DTA as well as XRD patterns. Hexagonal perovskite structure of BYTO was described by Le-Bail analysis and images observed from TEM and HR SEM. Increase in remanent polarization and remanent magnetization was observed with the decrease in temperature. A temperature and frequency dependent dielectric property of BYTO ceramic was also discussed in detail.

Chapter VI Synthesis of partial substitution of iron at the site of titanium are discussed in this chapter. $\text{Ba}_6\text{Y}_2\text{Ti}_{4-x}\text{Fe}_x\text{O}_{17}$ ($x= 0.05, 0.1$ and 0.2) abbreviated as BYTFO-05, BYTFO-1 and BYTFO-2 were synthesized by semi-wet route at low sintering temperature and duration than earlier synthesized $\text{Ba}_6\text{Y}_2\text{Ti}_4\text{O}_{17}$ (BYTO) ceramic. Powder XRD of all samples have same diffraction patterns indicates single phase formation materials sintered at $1100\text{ }^\circ\text{C}$. An image observed by TEM confirms the presence of hexagonal shape and the edge boundary of particles shows 120° bond angles. SEM EDX and elemental mapping indicates the presence of Ba, Y, Ti, Fe and O with their atomic percentages. A detailed discussion of temperature dependent dielectric constant, dielectric loss and impedance analysis is explained. Effect of iron doping as well as temperature on remnant magnetization were widely discussed.