

The materials having high dielectric constant and low dielectric loss, is an important in material science due to its applications in electrical and electronic devices such as multilayer capacitor (MLCC), microwave devices, aircraft, dynamic random excess memory (DRAMs) and automobiles, gas sensors, capacitors, humidity sensors as well as memory devices. Ceramic capacitor materials have more attracted due to increasing demand of high dielectric constant and also because of good thermal stability. Thurnauer was firstly reported in 1941 for high dielectric constant ($\epsilon_r \sim 10^3$) of BaTiO₃ ceramic material but have limitation towards temperature dependent of ϵ_r in industrial applications. Further, temperature independent of dielectric constant materials were played very important role in ceramic capacitor industry. The perovskite oxides with the general formula ABO₃ have been widely investigated for their excellent ferroelectric, dielectric, optoelectronic and piezoelectric properties. Their properties can be improved by the partial substitutions of various substituents in either A or B metal sites or by simultaneous substitutions like as $A_{1-x}A'_xB_{1-x}B'_xO_3$, $AB_{1-x}B'_{1-x}O_3$, and $A_{1-x}A'_xBO_3$, respectively. The substitution in the perovskite oxide materials may be possible in different ways i.e., heterovalent, isovalent and valency, radius and the co-ordination number of the substituents are the important factor to decide the site where it occupies in these oxide materials. The ideal and distortion of the structure of the perovskite were determine with the help of tolerance factor (t), if the tolerance factor is exactly 1, then the perovskite having ideal cubical structure and if greater than 1 then distortion from cubic to hexagonal is taking place in the perovskite structure. The hexagonal ferrite with the general formula MFe₁₂O₁₉ (Where M= Ca⁺², Sr⁺², Pb⁺², Ba⁺² etc.) is the ferromagnetic materials and having magnetization along c axis. BaFe₁₂O₁₉ having tolerance factor 1.0073, adopted hexagonal crystal structure.

Ferroelectric materials having permanent dipole moment due to arrangement of cations and anions under the applications of strong electric field which causes spontaneous polarization within the materials. Barium titanate (BaTiO_3 or BT) is a normal ferroelectric materials and having high dielectric constant. Its higher value of dielectric constant is due to ferroelectric behaviors. In ferroelectric material, displacement of the atoms take place from their original lattice point in a particular orientation in the presence of electric field due to change in symmetry of the crystal structure. Therefore, all the domains i.e., orientation of polarization present in volume region aligned in electric direction and permanently reorientation of polarization within the materials was observed at strong electric field. These types of materials have been widely used in the applications of high dielectric capacitor, ultrasonic transducers, piezoelectric sonar and actuators, etc.

Magnetic properties of ceramic materials are mainly depends on the morphology, particle size, synthesis procedure as well as their compositions which is very important for practical applications such as video recorder, disk driver, tunnel junction, computer data storage, magneto-optice recording media. These types of materials contain the some special properties such as low loss and magnetic coupling, and high electrical resistivity which can be improved by changing the structure and composition of the material. Magnetic properties in the nanoscale region changed in comparison to the bulk material because surface to volume ratio increases. As a result of this, number of atoms on the surface of nanocrystals will be increases and their structure will be changed when particle size of nano-particles exist less than 100 nm.

In the present work, barium hexaferrite, cobalt and nickel –doped barium hexaferrite and composite of barium hexaferrite with $\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BCTO) ceramic synthesized by chemical route. All the synthesized materials were characterized by various physicochemical techniques to describe microstructure, crystal structure, particle size and shape, elemental analysis, surface roughness and their dielectrics as well as magnetic properties were studied in detail.

The objective of the present work is to concerned on (a) crystal structure (b) microstructure (c) elemental analysis (d) particle size (e) electrical and dielectric (f) magnetic behaviour of the following system synthesized by chemical route.

- $\text{BaFe}_{12}\text{O}_{19}$ (BHF)
- $\text{BaFe}_{12-x}\text{Co}_x\text{O}_{19}$ ($x = 0.0, 0.05, 0.1, 0.2$)
- $\text{BaFe}_{12-x}\text{Ni}_x\text{O}_{19}$ ($x = 0.0, 0.05, 0.1, 0.2$) and its composite with $\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BCTO) was also included in the thesis.

A brief description of the research work presented in the thesis divided into six chapters has been given as follows:

Chapter I A brief description of introduction of Perovskites and distortion of Perovskites to hexagonal were reported in this chapter and effect of various substitutions such as heterovalent, isovalent, and valence compensated substitutions on the dielectric and magnetic properties were also mentioned in this chapter.

Chapter II This chapter describes the details of the experimental procedure used for synthesis, characterization and application of the hexagonal barium hexaferrite. Chemical

route was used for synthesis of materials. Phase formation and the particle size of the materials were done with the help of X-ray diffraction (XRD), Lebeil analysis and transmission electron microscope (TEM). Scanning electron microscope (SEM) provides information about the microstructure and energy dispersive X-ray spectroscopy (EDX) analysis has been used for elemental compositions of materials. Maximum valley peak height and surface roughness of the materials can be measured by using atomic force microscopy (AFM) techniques. Magnetic measurement of the materials was carried out using a superconducting quantum design MPMS-3 with applied magnetic field and temperature. Magnetic properties of the materials was measured by superconducting quantum interference device with the temperature and applied field. Dielectric measurement of silver coated cylindrical pellet of the sample was calculated by LCR meter (PSM 1735, NumetriQN4L U.K.) with the variation of temperature and frequency.

Chapter III The detailed description of synthesis, characterization and application of $\text{BaFe}_{12}\text{O}_{19}$ (BHF) ceramic was included in this chapter. Single phase formation of barium hexaferrite sintered at $1050\text{ }^{\circ}\text{C}$ for 12 h was confirmed by X-ray diffraction (XRD) and Retveld analysis. The presence of Fe-O, Ba-O and Fe-O-Fe stretching band in the BHF ceramic was confirmed by Fourier transform infrared (FTIR) study. The average grain size observed by the SEM analysis was in the range of $0.3\text{--}1.0\text{ }\mu\text{m}$ and EDX spectrum shows the presence of Ba, Fe and O elements. Hexagonal shape of the particle with the particle size 175 nm was confirmed by transmission electron microscopy (TEM). Magnetic hysteresis loop confirmed the ferromagnetic nature of the material. Temperature and frequency dependence of dielectric constant and dielectric loss as well as conductivity measurement were elaborated

in detailed. Impedance studies exhibit the semiconducting nature of materials with the insulating grain boundaries.

Chapter IV This chapter contains electrical, magnetic and dielectric properties of cobalt doped barium hexaferrite ($\text{BaFe}_{12-x}\text{Co}_x\text{O}_{19}$, BHFC) ceramic with compositions $x= 0.0, 0.05, 0.1$ and 0.2) prepared by the chemical route. X-ray diffraction (XRD) pattern emphasized the single phase formation up to $x = 0.0- 0.1$ without any signature of secondary phase, however small amount of secondary phase of $\alpha\text{-Fe}_2\text{O}_3$ was also deducted by XRD with the composition $x = 0.2$. This impurity phase was also confirmed with the help of Le-bail analysis of XRD data. The presence of Fe-O, Fe-O-Fe stretching bands in the BHFC ceramic were observed by FT-IR spectra for few selected compositions. Transmission electron microscope (TEM) analysis shows the hexagonal nature of the particles with the particle size 213, 185 and 52 nm, respectively for cobalt concentrations $x = 0.0, 0.05$ and 0.1 . The value of dielectric constant and dielectric loss were found in decreasing order with increasing cobalt substitution of BHFC ceramic. From magnetic measurement, it was observed that both of saturation magnetization (M_s) and remnant magnetization (M_r) decreases with increasing cobalt substituents, however their coercivity values increases with increasing Co-content in the barium hexaferrite ceramic and shows ferromagnetic behaviors.

Chapter V This chapter describes dielectric and magnetic properties of $\text{BaFe}_{12-x}\text{Ni}_x\text{O}_{19}$ BHNF ($x = 0.0, 0.05, 0.1$ and 0.2) ceramic synthesized via chemical route. Single phase formation with hexagonal phase of the sintered material at $1200\text{ }^\circ\text{C}$ for 6 h was confirmed by X-ray diffraction (XRD) analysis. Hexagonal structure of BHNF ceramic was confirmed by Le-bail analysis and the image observed from TEM, HR-SEM and 2D AFM analysis. EDX spectrum and EDX mapping of the BHNF ceramic shows the presence of Ba,

Fe, Ni and O elements with their atomic percentage as well as weight percentage. The lower value of squareness ratio (M_r/M_s) obtained from the magnetic measurement confirm the presence of single domain non-interacting particles which emphasized hard magnetic materials. Temperature and frequency dependence of dielectric properties and conductivity of BHNf ceramic were disclosed in detail of this chapter.

Chapter VI Synthesis of nanocomposite $0.5\text{BaFe}_{12}\text{O}_{19}-0.5\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BHF–BCT) via chemical route for the improvement of dielectric and magnetic properties is discussed in this chapter. Nanocomposite phase formation of the sintered materials at 1173 K for 8 h was confirmed with the help of X-ray diffraction (XRD) pattern and Le-bail analysis. The existence of Fe-O-Fe stretching bands in $\text{BaFe}_{12}\text{O}_{19}$ (BHF), Cu-O and Bi-O bonds in the $\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BCTO) ceramic observed by FT-IR spectra supported composite formation of the materials. The bright field TEM image revealed the presence of bimodal faceted particles with average particle size 31 ± 5 nm. SEM morphology indicates hexagonal and round shape of the grains in the nanocomposite. A detail discussion of temperature and frequency dependent of dielectric constant and dielectric loss, electrical and impedance studies were widely discussed in this chapter.