

Perovskite oxide having general formula ABO_3 has been widely investigated because of their excellent electrical, dielectric, optoelectronic and piezoelectric properties. The properties of the perovskite can be modified by a variety of partial substitutions either on A-site or B-site or simultaneous substitutions. Such substitutions may be represented as $A_{1-x}A'_xB_3O_3$, $AB_{1-x}B'_xO_3$ and $A_{1-x}A'_xB_{1-x}B'_xO_3$ respectively. These substitutions may be heterovalent, isovalent or valence compensated perovskite oxide. It is known that the valency, radius, and coordination number of the substituent are an important parameter to determine the site, it occupies in the perovskite oxide with a limit range of composition to form a solid solution. Ceramic materials with high dielectric constant and good thermal stability have attracted a special attention mostly for their applications in electronic devices such as memory devices, gas sensors, humidity sensors and capacitors. Recently, high dielectric constant with weak temperature-dependent has been observed in calcium copper titanate, $CaCu_3Ti_4O_{12}$ (CCTO) having complex cubic perovskite structure with space group $Im\bar{3}$ and lattice parameter 7.391 Å. Subramanian *et. al.* first time reported CCTO compound, which has high dielectric constant ($\sim 10^4$) which is independent of temperature (100 - 600 K) and frequency ($10^2 - 10^6$ Hz) and no phase transition was observed in the CCTO ceramic. A number of family members of CCTO compound having a general formula $ACu_3Ti_4O_{12}$ (A= Ca, $Bi_{2/3}$, $Y_{2/3}$, $Gd_{2/3}$, $La_{2/3}$, $Na_{1/2}$) have been studied since last few years. CCTO may be widely used in the electronic applications such as microwave devices, multilayer capacitor (MLCC), dynamic random access memory (DRAM), aircrafts and automobiles. High dielectric constant and low dielectric loss is always important for practical applications for the capacitor materials. The main drawback of undoped CCTO and its family members is slightly high dielectric loss at room temperature. Scientists around the world have been

working on CCTO and its family to reduce its dielectric loss to make ideal capacitor materials.

$\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BCTO) is members of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ (CCTO) family shows a similar structure and also have high dielectric constant. There was no works have been done on this system. At present, many research groups have been working on this system. The other Bi and Ti based compound is undoped and lanthanum doped $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ (BTO) ceramics have been observed great interest in ferroelectric materials due to their possible application in the production of non-volatile random access memories. This application is based on the existence of two opposite polarization states which can be used to store binary information in a non-volatile electric field. In practice, the materials have to satisfy several requirements, e.g., a low coercive field, a large remnant polarization and a low processing temperature which is compatible with Si-based IC technology.

Magnetic properties of ceramic and composite materials depend on particle size, morphology and composition of materials. Many magnetic materials have been used in variety of applications and have some special properties such as low loss, high electrical resistivity and magnetic coupling. People have been aware composite materials since several hundred years before Christ and applied innovation to improve the quality of life contemporary composites results from research and innovation from past few decades have progressed from glass fiber for automobile bodies to particulate composites for aerospace and other applications. A composite material is a material made up of two or more materials that are combined in a way that allows the materials to stay distinct and identifiable have different properties. Most of composite materials contain fiber of one material tightly bound into another material called a matrix. The matrixes bind the fiber together a bit like on adhesive

and make them more resistant to external damage, where as the fiber make the matrix stiffer, stronger and help it resist to cracks.

Presently, many research groups have been working on polymers based and metal composites but perovskite oxide based composites are not studied extensively. The aim of the present work is to synthesized BCTO and BLTO and some of their composites and described the crystal structure, microstructure, elemental analysis, surface roughness, dielectric and magnetic behavior of the following systems:

- $\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BCTO)
- $\text{Bi}_3\text{LaTi}_3\text{O}_{12}$ (BLTO)
- $0.5 \text{ Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12} + 0.5 \text{ Bi}_3\text{LaTi}_3\text{O}_{12}$ (BCLT-55)
- $0.9 \text{ Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12} + 0.1 \text{ Bi}_3\text{LaTi}_3\text{O}_{12}$ (BCLT-91)
- $0.1 \text{ Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12} + 0.9 \text{ Bi}_3\text{LaTi}_3\text{O}_{12}$ (BCLT-19)

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

Chapter I contains a general introduction of complex perovskite, ceramic and composite materials. Ceramic and composite materials have received great interest in ferroelectrics, ionic conductors, superconductors, multiferroic and magneto resistant materials result from their mutual interaction. The details of complex perovskite and composite materials considered in the current study and the purpose of the thesis is mentioned in this chapter.

Chapter II described the experimental procedure used for preparation and characterization of the perovskite oxides and its composites. The semi-wet route was used for the preparation of materials. X-ray diffraction and transmission electron microscope (TEM)

have been studied for determination of crystalline size and particle size respectively. Scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDX) analysis have been used for micro-structural studies and elemental analysis of materials respectively. Study of surface roughness and maximum peak height of the materials have been determined by atomic force microscopy (AFM). Magnetic properties of the materials was measured by superconducting quantum interference device with the temperature and applied field. Dielectric properties of materials were measured on a LCR meter with the variation of temperature and frequency.

Chapter III contains synthesis, characterization and application of $\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ (BCTO) ceramic. Fourier transform infrared (FTIR) study of BCTO precursor powder and calcined ceramic showed the presence of alcoholic functional groups and the stretching band of Ti-O and Cu-O respectively. X-ray diffraction (XRD), scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDX) were employed to characterize the structure, surface morphology and purity of the sintered BCTO ceramic respectively. X-ray diffraction study confirmed the single phase formation of BCTO ceramic at 800 °C. The average dimension of grains calculated by SEM and AFM was found to be in the range of $0.73 \pm 0.2 \mu\text{m}$ with clear grain boundaries. Magnetic property was investigated over a wide temperature range 2–300 K at a magnetic field of ± 7 tesla. The Curie temperature was calculated by zero field cooled (ZFC) and field cooled (FC) magnetization at 100 Oe applied field which was found to be 125 K. The sintered BCTO ceramic shows high dielectric constant ($\epsilon' = 2.9 \times 10^4$) at 323 K and 100 Hz.

Chapter IV described the synthesis, characterization and application of $\text{Bi}_3\text{LaTi}_3\text{O}_{12}$ (BLTO) ceramic. The single phase formation of BLTO was confirmed by X-ray diffraction

pattern. The complexation of citric acid with oxygen metal bond was found out by FT-IR studies in the calcined powder at 800 °C for 6 h. Transmission electron microscope (TEM) analysis is used to determine the particle size in the range of 212 ± 20 nm. The average grain size obtained from scanning electron microscope (SEM) and atomic-force microscope (AFM) was almost have the similar values 200 ± 20 nm and 217 ± 20 nm, respectively. The surface roughness and root means square roughness was found to be 46.015 and 58.661 nm respectively, from the AFM analysis. Magnetic properties of ceramic show the magnetic transition of anti-ferromagnetic to ferromagnetic nature in the M-H and M-T curve. The blocking temperature (T_B) and Neel's temperature (T_N) were found to be 155 K and 51 K respectively. The value of dielectric constant (ϵ') for $\text{Bi}_3\text{LaTi}_3\text{O}_{12}$ ceramic was found to be 403 at 100 Hz at 500 K.

Chapter V contains the synthesis, characterization and application of the composite having composition $0.5 \text{ Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12} - 0.5 \text{ Bi}_3\text{LaTi}_3\text{O}_{12}$ (BCLT-55). X-ray diffraction (XRD) analysis showed the presence of BCTO and BLTO phases in the composites sintered at 900 °C for 8 h. Transmission electron microscope (TEM) analysis of the composite shows the presence of nano particles in the range of 55 ± 3 nm. Atomic force microscopy (AFM) study also substantiates the presence of nano particles in the composite. Scanning electron microscope (SEM) images show that the surface morphology consists of plates like and spherical grains. Magnetic behavior of the composite shows the weak ferromagnetic nature in M-T and M-H curve. The observed value of high dielectric constant ($\epsilon' = 13.94 \times 10^3$) of the composite may be due to the presence of space charge polarization.

Chapter VI described the synthesis, characterization and application of the composite having composition $0.9 \text{ Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12} - 0.1 \text{ Bi}_3\text{LaTi}_3\text{O}_{12}$ (BCLT-91). BCLT-91

composite was characterized by X-ray diffraction (XRD) for crystal phase identification and transmission electron microscope (TEM) for particle size distribution. The average particle size was found to be 152 ± 5 nm. Surface roughness and average grain size were found to be 143.08 nm and $0.559 \mu\text{m}$ respectively, by AFM analysis. The elemental composition of the composite was confirmed by energy dispersive X-ray spectroscopy (EDX). M-T curve explains the magnetic transition, paramagnetic to anti-ferromagnetic at 27 K. The high value of dielectric constant was found to be $\epsilon' \approx 23 \times 10^5$ at 463 K, 100 Hz, at low frequency due to the presence of interfacial polarization in the composite.

Chapter VII explain the synthesis, characterization and application of the composite having composition $0.1 \text{ Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12} - 0.9 \text{ Bi}_3\text{LaTi}_3\text{O}_{12}$ (BCLT-19). The phase formation of $\text{Bi}_{2/3}\text{Cu}_3\text{Ti}_4\text{O}_{12}$ and $\text{Bi}_3\text{LaTi}_3\text{O}_{12}$ were confirmed by X-ray diffraction (XRD). Transmission electron microscope (TEM) analysis was determined nano particle size of composite in the range of 14 ± 5 nm. Scanning electron microscope (SEM) image exhibited tubular, spherical and heterogeneous structure of grains. Atomic force microscopy (AFM) study explained root means square roughness, average roughness, and maximum area peak height. Study of magnetic properties was determined the ferromagnetic and anti ferromagnetic in nature. The dielectric constant ($\epsilon' = 3147$ at 100 Hz with 503 K) of nano composite may be due to the existence of space charge polarization.

Chapter VIII described the summary of present research work and its future scope.