Structural, Electrical and Dielectrical Properties of Copper Doped Strontium Titanate

Dr. C. Sudheendra  
Department of Physics,  
SBSYM Degree College, Kurnool, India.

Abstract
Strontium titanate (SrTiO$_3$, STO) is an ABO$_3$-type perovskite and well-known incipient ferroelectric, characterized by a very high dielectric constant at low temperature. This perovskite, either in bulk or in thin films, is a favorite substrate (buffer layer) for the growth of high-$T_c$ superconducting oxide layers. Important properties of these superconducting structures depend in an essential way on the substrate dielectric response which can be used as a convenient tuning mechanism. In this connection effect of copper doping into STO was taken for the present study and structural properties were investigated. Cu$_{1-x}$Sr$_x$TiO$_3$ ($x=0.1$ to $0.9$) powders were prepared by conventional bulk preparation method. Structural properties of the prepared samples were investigated by X-ray diffraction SEM and EDAX. From XRD profiles, effect of Cu concentration on the crystal structure, average particle size and lattice parameter was investigated. At low Cu concentration a phase of Cu$_3$Sr$_4$TiO$_{12}$ was identified from the XRD profiles which crystallizes into cubic structure with a=7.426Å. The less solubility of Cu in STO was confirmed from XRD and EDAX profiles. Variation of electrical conductivity ($\sigma$) with temperature ($T$) of the system Sr$_{1-x}$Cu$_x$TiO$_3$ ($x=0.1$ to $0.9$) and for the samples. Al$_2$TiO$_5$CuAl$_2$TiO$_6$ and CuTiO$_3$ was observed. It is obvious that electrical conductivity increases with increase of temperature. A.c conductivity also increases with the increase of temperature ($T$).

INTRODUCTION
Oxide ceramics such as SrTiO$_3$ (STO) are widely used for passive devices in high frequency systems such as filters and antennas, and also in active devices such as tunable rf filters and phase switches. STO-based compounds are also used in grain boundary layer capacitors, in which conductivity differences increase the device performance. The electronic structure of strontium titanate can be changed by the introduction of dopants, e.g. changing it from insulator to metallic conductor. For
applications in electronic devices, the contact between the ceramic and the electrode material plays an important role in device performance. In particular, band alignment at the metal–oxide interface can determine the electrical properties of the device [1].

High dielectric permittivity lead-free compositions are currently of great interest due to the need for environmental harmless applications. The perovskite strontium titanate (SrTiO$_3$, ST) is an incipient ferroelectric material, i.e. it possesses polar soft modes but does not exhibit any ferroelectric phase transition down to 0 K. It is known that the dielectric response and other related properties of SrTiO$_3$ can be modified by controlling the Sr/Ti ratio and oxygen vacancy concentration, by oxygen isotope exchange and by doping and substituting Sr and/or Ti ions. In contrast, isovalent Sr-site dopants, such as Ca$^{2+}$, Ba$^{2+}$ Mn$^{2+}$ as well as heterovalent Bi$^{3+}$ induce a dielectric anomaly [2]. There is considerable renewed interest in the low-temperature properties of SrTiO. This perovskite, either in bulk or in thin films, is a favorite substrate for the growth of high-T$_c$ superconducting oxide layers. Important properties of these superconducting structures depend in an essential way on the substrate dielectric response which can be used as a convenient tuning mechanism. On the other hand, unusual phenomena were uncovered in recent low-T investigations of bulk SrTiO$_3$.

Oxide ceramics such as SrTiO$_3$ (STO) are widely used for passive devices in high frequency systems such as filters and antennas and also in active devices such as tunable rf filters and phase switchers and other devices. STO-based compounds are also used in grain boundary layer capacitors, in which conductivity differences increase the device performance. The electronic structure of strontium titanate can be changed by the Introduction of dopants, e.g. changing it from insulator to metallic conductor. Recently, polycrystalline STO has been under discussion in the Semiconductor industry as candidate material for DRAM memories. STO is also important as a model system for more complex oxides. The Solid solution STOBaTiO$_3$. For example, is used as a capacitor material. For applications in electronic devices, the contact between the ceramic and the electrode material plays an important role in device performance. In particular, band alignment at the metal-oxide interface can determine the electrical properties of the device (3). High dielectric permittivity lead-free compositions are currently of great interest due to the need for environmental harmless applications. The perovskite strontium titanate (SrTiO$_3$, ST) is an incipient ferroelectric material, it possesses polar soft modes but doesn't exhibit any ferroelectric phase transition down to 0 K. It is known that the dielectric response and other related properties of SrTiO$_3$ can be modified by controlling the Sr/Ti ratio and oxygen vacancy concentration by oxygen isotope exchange.

**EXPERIMENTAL DETAILS**

The present investigation involves the detailed study of structural properties of Copper doped SrTiO$_3$. The samples with varying Copper concentration were synthesized in air by the high temperature solid-state reaction method. The purity of
the materials was higher than 99%. The calcinations of the samples were carried at 1350°C. Formation of the Cu$_{1-x}$Sr$_x$TiO$_3$ was confirmed from XRD profiles. The XRD studies at room temperature were carried out using SEIFERT X-ray powder diffractometer. The Cu Ka radiation with a wavelength 1.54056 Å was used for recording X-ray diffraction patterns. Ni filter was used as the monochromatic. The X-ray machine was operated with 30 mA beam current and 40 kV of power. For phase identification and for the determination of unit cell parameters, the patterns were compared against standard patterns given in powder diffraction file (The J.C. PDS files). In order to obtain the accurate cell parameters, the data was fed to a computer program EXPO2009 and the cell parameters obtained with an accuracy of >0.1%. Surface morphology of the samples was analyzed from SEM photographs. Composition studies were carried with EDAX.

The present investigation also involves the detailed study of Electrical properties of Copper doped SrTiO$_3$. The samples with varying Copper concentration were synthesized in air by the high temperature solid-state reaction method (4). It is found from experiments that the purity of the materials was higher than 99%. The calcinations of the samples was carried at 1350°C. The resulted powders were made into pellets by using hydraulic press with a pressure of 7 to 10 tons and were finally sintered at 1050°C for 5 hours. Most suitable firing temperatures were determined by studying the shrinkage and apparent density of the products for different firing schedules. The purpose of the sintering is to increase the mechanical strength of the pellet. These pellets were then annealed at ~300°C for about 2 hours under vacuum to remove strain introduced due to mechanical stress. Electrical conductivity of the all the above samples are studied at different temperatures. The TEP is also determined at different temperatures (5). Activation energies are calculated for all the samples and found to be agreeing with literature.

The dielectric properties at fixed frequencies and temperatures are studied (6&7). The dielectric constant, dielectric loss and ac conductivity were determined. The results were interpreted with chemistry of the chosen materials and its structures. In few samples the structural distortion was observed. Transport Properties were explained mainly with scattering mechanism such as lattice-electron and electron-electron interactions are dominated in our samples.

**RESULTS AND DISCUSSION**

Figure 1 shows the XRD profile of Cu$_{1-x}$Sr$_x$TiO$_3$ sample for Cu concentration of 0.1 weight percentage. Particle size was calculated from this profile and found that it decreases with the increase of copper concentration. The average particle size was found to be 600Å. At low Cu concentration a phase of Cu$_3$Sr$_4$TiO$_{12}$ was identified from the XRD profiles which crystallizes into cubic structure with a=7.426Å. The less solubility of Cu in STO was confirmed from XRD and EDAX profiles. At high Cu concentration (>0.2) no specific phase of Cu$_{1-x}$Sr$_x$TiO$_3$ was identified and separate CuO and TiO$_2$ (rutile) phases were identified. This makes Cu$_{1-x}$Sr$_x$TiO$_3$ a candid material as buffer layer in superconducting devices. Figure 2 shows the SEM
photograph of Cu$_{1-x}$Sr$_x$TiO$_3$ samples for Cu concentration of 0.1 weight percentage. Grains are clearly visible in this photograph. Grain size was calculated from this photograph and found that it decreases with the increase of Cu concentration. Figure 3 shows the EDAX Profile of Cu$_{1-x}$Sr$_x$TiO$_3$ sample for Cu concentration of 0.1 weight percentage. Compositional changes are clearly seen in this profile. It is proposed to carry electrical, thermoelectric and dielectric properties of these samples to investigate the conduction mechanism and dielectric response in these materials.

**Figure 1:** XRD profile of Cu$_{1-x}$Sr$_x$TiO$_3$ sample for Cu concentration of 0.1.

**Figure 2:** SEM photograph of Cu$_{1-x}$Sr$_x$TiO$_3$ sample for Cu concentration of 0.1.
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Figure 3: EDAX Profile of Cu$_{1-x}$Sr$_x$TiO$_3$ sample for Cu concentration of 0.1.

Figures 4 depicts experimental observation of variation of dielectric constant with temperature for the system Sr$_{1-x}$Cu$_x$TiO$_3$ (x=0.1 to 0.9) respectively. Figures 2 depicts experimental observation of variation of dielectric constant with frequency for the system Sr$_{1-x}$Cu$_x$TiO$_3$ (x=0.1 to 0.9) respectively. Figures 5 depicts experimental observation of variation of dielectric loss (tand) with temperature for the system Sr$_{1-x}$Cu$_x$TiO$_3$ (x=0.1 to 0.9) respectively. Figures 6 depicts experimental observation of variation of dielectric loss (tand) with frequency for system Sr$_{1-x}$Cu$_x$TiO$_3$ (x=0.1 to 0.9).

Figure 4: Temperature variation of Dielectric Constant of Sr$_{0.1}$Cu$_{0.9}$TiO$_3$
Figure 5: Frequency dependence of Dielectric Constant of Sr0.1Cu0.9TiO₃

Figure 6: Frequency dependence of dielectric loss of Sr0.1Cu0.9 TiO₃
REFERENCES


