Magnetic and Dielectric Properties of $R_2\text{CuTiO}_6$ Compounds ($R = \text{Y, La, Pr and Nd}$)

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Abstract Magnetic and dielectric properties of the double perovskite compounds of the type $R_2\text{CuTiO}_6$ (RCTO, where $R = \text{Y, La, Pr and Nd}$) has been studied. $\text{Y}_2\text{CuTiO}_6$ (YCTO) crystallizes in a hexagonal unit cell, whereas the other three compounds form into orthorhombic structure. All four compounds show paramagnetic behavior down to 5 K. The dielectric studies show moderate dielectric constant ($\varepsilon'$) and very small dielectric loss ($\tan \delta$) for YCTO. The orthorhombic members of RCTO compounds exhibit moderate values of $\varepsilon'$ and $\tan \delta$. The dielectric properties are presented and discussed here in the light of the influence of structure and rare-earth ions on the physical properties of RCTO compounds.

Keywords $R_2\text{CuTiO}_6$ perovskites · Dielectric properties

1 Introduction

Layered perovskite oxide materials offer various stimulating and challenging phenomena like superconductivity, colossal magnetoresistance, multiferroicity, colossal dielectric constants, etc. [1–3]. Of these, the last two phenomena have attracted a great deal of attention in recent times [1, 4]. The simultaneous presence of ferroelectricity and magnetism has opened up avenues for the fabrication of new generation random access memory devices. For many devices, such as antennas and transmitters, the size and performance of operating devices are limited by the material properties, and therefore there is great interest in finding new materials with desirable dielectric properties. For example, to miniaturize the size of the device, high dielectric constant is required, whereas to minimize the bandwidth, low dielectric loss at the operating frequency is desirable [5, 6]. For making components for communication devices such as resonators and filters, it is desirable to have a material with high dielectric constant ($\varepsilon'$) so that the size of the ceramic could be reduced since the size of the resonator at any resonating frequency is inversely proportional to the square root of $\varepsilon'$. Another important requirement for ideal filters is that the dielectric material used should not depend strongly on the temperature, i.e., should have a wide temperature window of constant $\varepsilon'$ as a function of temperature.

A large number of materials have been explored in search of above mentioned dielectric properties. From applications point of view, the material should have high $\varepsilon'$, low dielectric loss ($\tan \delta$), narrow temperature and frequency dependence. For example, $\text{SiO}_2$ and $\text{Si–N}$ have a value of $\tan \delta \sim 0.001$ and 0.003 but their $\varepsilon'$ is too low (3.9 and 7, respectively) for any application purposes. The compounds $\text{Ta}_2\text{O}_5$ [7, 8] and $\text{HfO}_2$ [7–9] have high $\varepsilon'(35, 25)$ and low $\tan \delta$ values (0.006, 0.1), respectively, but their temperature coefficient...
of dielectrics is very high. Dielectric constant was found to increase by substituting TiO₂ in Ta₂O₅ [7, 8]. However, the study of perovskite oxide materials has met little success recently as far as the desired dielectric properties are concerned. The perovskite layered CaCu₃Ti₄O₁₂ (CCTO) compound exhibited colossal dielectric constant (CDC) [10]. The values of $\varepsilon'$ and $\tan \delta$ for CCTO are 10286 and 0.067, respectively. Such a large difference in the values of $\varepsilon'$ and $\tan \delta$ as compared to SiO₂ and HfO₂ has made the search for new high $\varepsilon'$ materials more interesting and desirable. Another advantage of the non-ferroelectric CCTO exhibiting large CDC over conventional ferroelectric-based dielectrics is the temperature independent CDC around room temperature. This report has led the research for a new oxide material with high $\varepsilon'$, particularly CDC.

Among the copper–titanium compounds, the dielectric properties of Ho₂CuTiO₆ (HCTO) were reported first in the literature [11, 12]. HCTO exhibits $\varepsilon'$ of 1300 at room temperature. In a recent study, it was found that HCTO has an unusually stable dielectric property, which is attributed to the B site (Cu/Ti) disorder [12]. It is assumed that the B-site disorder in hexagonal compounds such as HCTO can yield such robust dielectric properties [12]. Recently, dielectric relaxation has been studied in the orthorhombic La₂CuTiO₆ as well as the hexagonal, late rare-earth members of $R_2$CuTiO₆ compounds [13–15]. The dielectric relaxation has been attributed to oxygen-vacancy induced mixed valence of Cu$^{+2+}$/Cu$^{+2+}$ and Ti$^{3+}$/Ti$^{4+}$ [13, 14]. The studies on hexagonal RCTO compounds reveal that the temperature stability of dielectric properties is dominated by intermediate-frequency vibration modes [15].

According to the recent reports, depending upon the size of the ionic radii of $R$ ion, RCTO compounds can crystallize in two structures, orthorhombic (space group $Pnma$ or $Pbnm$) for $R =$ La–Gd, and hexagonal (space group $P6_3mc$ for $R =$ Y, Dy–Lu). Detailed structural studies on both structure types have been carried out and reported in the literature [16, 17]. However, the dielectric properties of the orthorhombic structured compounds are very scarcely known [15]. In our studies on the double perovskite copper–titanate compounds, we have synthesized $R_2$CuTiO₆ (RCTO, where $R =$ Y, La, Pr and Nd) series of compounds and carried out detailed structural, magnetic and dielectric property studies. The present work is interesting since we find evidence of dielectric relaxation in the orthorhombic members of the RCTO series of compounds.

2 Experimental Details

The polycrystalline samples of the $R_2$CuTiO₆ compounds were prepared by the standard solid-state reaction method using high purity (better than 99.9%) starting compounds such as La₂O₃, Nd₂O₃, Pr₆O₁₁, Y₂O₃, CuO and TiO₂.