

# Fabrication of high-k dielectric Calcium Copper Titanate (CCTO) target by solid state route

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**Abstract.**  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  (CCTO) ceramic pellet of 10mm diameter has been synthesized by adopting solid state route. The structural and morphological characterization of the ceramics sample was carried out by X-ray diffraction (XRD) and scanning electron microscope (SEM), respectively. XRD pattern revealed the CCTO phase formation, where as SEM micrograph shows the sample consisting of well defined grain and grain boundaries. The room temperature dielectric constant of the sample was found to be  $\sim 5000$  at 1kHz. After successful preparation of CCTO pellet, a 2 inch diameter CCTO sputtering target is also fabricated in order to deposit CCTO thin films for microelectronic applications.

**Keywords:** Ceramic, X-ray diffraction, Dielectrics, Electron Microscopy

## 1. Introduction

In recent years, development of high-k dielectric thin films for microelectronics has drawn tremendous attention in order to replace  $\text{SiO}_2$ . The colossal dielectric constant (CDC) phenomenon in  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  (CCTO) has drawn a great deal of interest. It possesses a distorted cubic perovskite ( $\text{ABO}_3$ ) structure with lattice parameter of  $7.391\text{\AA}$ . It also shows a negligible temperature dependence on dielectric constant [1, 2]. The origin of the giant dielectric constant in CCTO is not well understood yet. The origin of high dielectric constant in CCTO may be due to stoichiometric variation [3], oxygen vacancies [4] and/or presence of secondary phases which forms layered structures which can be explained by Internal Barrier Layer Capacitance (IBLC) [5]. For fabrication of microelectronic devices, the synthesis of high quality CCTO thin films is highly required. CCTO thin films can be deposited by different techniques notably PLD [6], Sol-gel route [8], CVD, RF sputtering [9] etc. Out of them RF sputtering have some advantages over other deposition techniques such as low cost, low processing temperature, good compositional homogeneity, and large area deposition [10]. For sputtering, a high pure CCTO target is required. In this report, an attempt has been done to fabricate 2 inch diameter target for deposition of high-k thin film.



## 2. Materials and Methods

Stoichiometric amount of  $\text{CaCO}_3$ ,  $\text{CuO}$ ,  $\text{TiO}_2$  were mixed thoroughly in acetone medium by using ball milling in a sealed plastic bottle with Zirconium oxide beads for 12 hour. The powder was calcined at different temperature varied from  $900^\circ\text{C}$  to  $1050^\circ\text{C}$  for 10 hours. The final product was grinded into fine powder with Agate mortar. Formation of single phase CCTO was confirmed by XRD studies. The powder was mixed with 3wt. % polyvinyl alcohol (PVA) solution and was pressed into pellets of 10mm diameter and 1.5mm thickness using uniaxial compression with pressure 20MPa. Finally the pellet was sintered in air ambient at  $1100^\circ\text{C}$  for 8 hours.

The structural and morphological properties were studied by X-Ray diffractometer (Rigaku ultima IV) and scanning electron microscope (JEOL JSM- 7001F). For electrical measurements, the sample surfaces were polished and silver paint was applied on both the surfaces. Electrical properties have been investigated using a LCR meter (Wayne Kerr 6500 B). In order to fabricate 2 inch diameter and 3mm thickness sputtering target, the calcined powder was mixed with 5wt.% PVA solution and was pressed into a pellet of 2 inch dia and 3mm thickness using uniaxial compression with pressure 40MPa. Finally the sample was sintered at  $1100^\circ\text{C}$  for 8 hours.

## 3. Results and discussion

Fig. 1. shows the XRD pattern of CCTO powder calcined for 10 hours for the temperatures ranging from  $900^\circ\text{C}$  to  $1050^\circ\text{C}$ . The single phase CCTO was obtained for the calcination temperature of  $1050^\circ\text{C}$ . XRD pattern matches well with body centered cubic perovskite CCTO (ICDD card no – 751149). The diffraction pattern reveals the formation of polycrystalline sample. Fig. 2 shows the SEM micrograph of CCTO pellet sintered at  $1100^\circ\text{C}$  for 8 hour. Smaller grains were observed at the grain junctions, which might be due to decomposition of partial CCTO during sintering at high temperature.

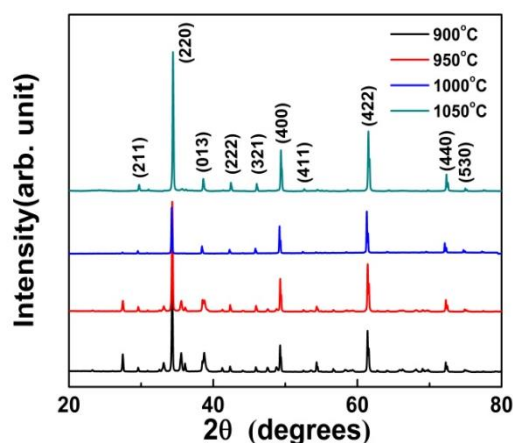


Fig. 1:- X-ray diffraction patterns of CCTO powder calcined at different temperature

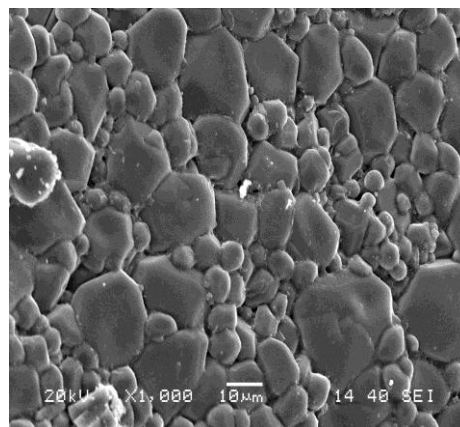


Fig. 2:- SEM micrograph of CCTO pellet

Fig. 3. shows the frequency dependence of dielectric constant in a temperature range of  $30^\circ\text{C}$  -  $310^\circ\text{C}$ . From figure 3, the dielectric constant of CCTO increased rapidly below 1kHz and in the frequency range of 1kHz to 100kHz it is nearly constant. The dielectric constant at  $310^\circ\text{C}$  is very high with a value of 16000 at 100Hz. The  $\epsilon_r$  value decreased with an increase in frequency, but for a particular frequency the dielectric constant increased with increase in temperature. This can be attributed to the contribution of charge accumulation at the interface. The high permittivity value of CCTO ceramic is due to the presence of large grains and grain boundaries, which supports internal barrier layer capacitance (IBLC) mechanism [11]. Fig. 4 shows the frequency dependence of dielectric loss ( $\tan\delta$ ) in the temperature ranging from  $30^\circ\text{C}$  to  $310^\circ\text{C}$ . The value of  $\tan\delta$  for CCTO ceramic at 1kHz at room

temperature is about 0.14. The value of  $\tan \delta$  is nearly frequency independent at high frequencies (10kHz -1MHz). There is a sharp increase in dielectric loss at 100Hz with increase in temperature. This may be due to exponential increase in conductivity with temperature [12].

A 2 inch diameter and 3mm thick sputtering target was fabricated from calcined CCTO powder.

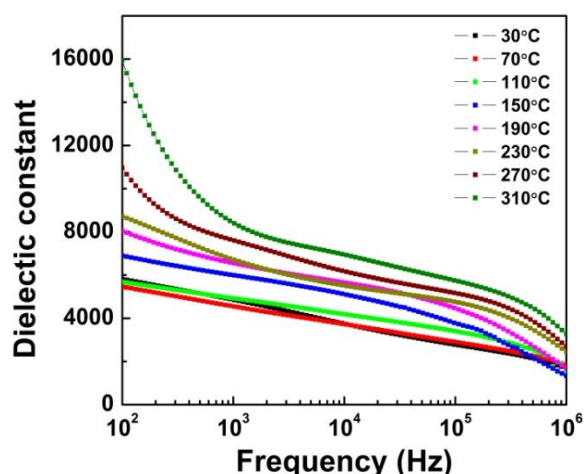


Fig. 3:- Frequency dependence of dielectric constant of CCTO pellet at different temperature

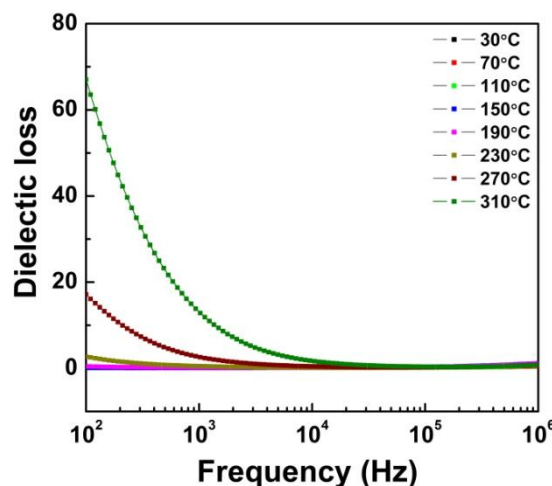


Fig. 4:- Frequency dependence of dielectric loss of CCTO pellet at different temperature

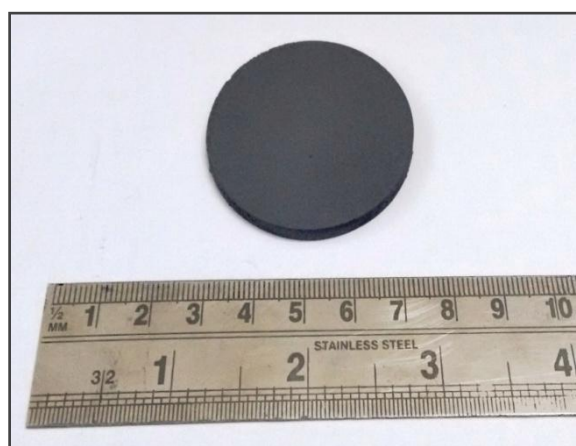


Fig. 5:- Optical image of 2 inch diameter fabricated CCTO sputtering target

#### 4. Conclusions

Perovskite  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  ceramic sample was synthesized by solid-state reaction method. XRD pattern confirms the CCTO phase formation at calcinations temperature of 1050°C. SEM results depict formation of microstructures with well defined grain and grain boundaries. The sample has shown a dielectric constant of around 5000 at 1kHz frequency with loss factor of 0.14. The high permittivity value is related to capacitance of the grain boundaries which is consistent with the IBL model.

#### 5. Acknowledgement

This work was supported by Science and Engineering Research Board (SERB), DST project (SR/FTP/PS-099/2012) under Fast Track Scheme for Young Scientist.

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