

Research Article

Microstructural and Dielectric Properties of Zr Doped Microwave Sintered $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ Synthesized by Sol-Gel Route

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Polycrystalline samples with the chemical formula $\text{CaCu}_3\text{Ti}_{4-x}\text{Zr}_x\text{O}_{12}$ ($x = 0, 0.02, 0.1, 0.2, 0.5,$ and 0.1) CCTZO were synthesized from metal nitrate solutions by the sol-gel method, followed by conventional and microwave heat treatments. The X-ray diffraction pattern of powder calcined at 800°C in conventional furnace for 3 h showed formation of a single phase. The crystal structure did not change on doping with zirconium and it remained cubic in the five studied compositions. The surface morphology of samples sintered at 1000°C in microwave furnace for 10 min was observed using a high resolution scanning electron microscope (HR-SEM). The grain sizes were in the range of $250\text{ nm} - 5\ \mu\text{m}$ for these samples. HRSEM results show that doping with Zr enhanced grain growth or densification. Energy dispersive X-ray spectroscopy (EDX) confirmed the presence of Zr. The dielectric characteristics of Zr doped CCTO were studied with an LCR meter in the frequency range of $50\text{ Hz} - 1\text{ MHz}$. A very high dielectric constant 21,500 was observed for the sample doped with Zr (0.02 mol%) at 50 Hz.

1. Introduction

It has been a long-term effort in the electronic industry and scientific community to seek novel dielectric materials. High dielectric constant leads to miniaturization of capacitive components, thus offering the opportunity to decrease the size of electronic devices. For high dielectric constant, materials based on ferroelectric BaTiO_3 are commonly used. BaTiO_3 is a ferroelectric material exhibiting high dielectric constant. In 1967, a new class of materials having the general formula $\text{ACu}_3\text{Ti}_4\text{O}_{12}$ ($A = \text{Ca}, \text{Ba}, \text{Sr}$) was discovered [1]. Among them, $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ has an edge over BaTiO_3 in practical applications in terms of temperature stability [2]. Due to their unique dielectric properties, both the $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ (CCTO) ceramics [3, 4] and the doped CCTO [5–7] have attracted ever-increasing attention for their practical microelectronic applications including capacitors and memory devices [8–10].

Extensive investigation has been made to identify the origin of the colossal dielectric property of CCTO [11–13]. The dielectric property of CCTO has been related to its

microstructure [14]. The studies, concerning the different processing and preparation conditions such as sol-gel derivation [15, 16], sintering method, temperature and time [17, 18], doping [6, 19], electrode material, pellet thickness [5, 20], and the dielectric characteristics of CCTO, demonstrated their strong influence on the dielectric properties of CCTO ceramics.

Developing high dielectric materials synthesized with lower calcination/sintering temperature has become the subject of study for many researchers. While some recently published reports concentrate on different synthesis techniques to reduce the processing temperature, other reports focus on substitutions to change the material properties. However, very limited work on the effects of substituting impurity along with lower processing temperature CCTO has been reported [21, 22]. The purpose of the present work is to dope Zr (0, 0.02, 0.1, 0.2, and 0.5 mol%) into CCTO ceramics and to investigate the effects of doping on the surface morphology and dielectric properties of Zr-substituted CCTO ceramics. Microwave sintering is adopted in this work due to its advantages

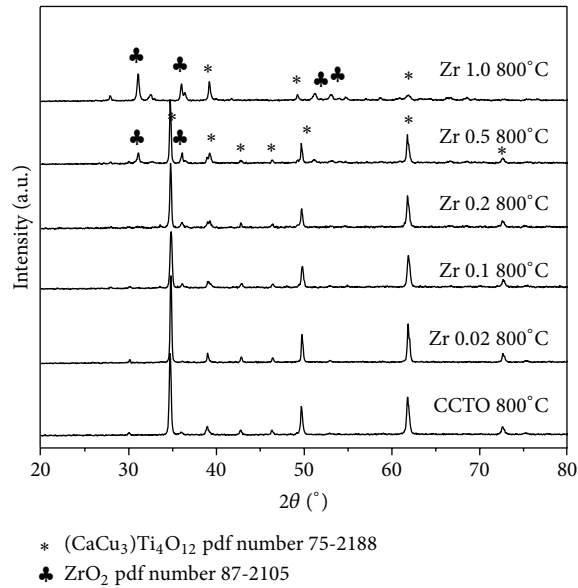


FIGURE 1: XRD patterns of CCTO and $\text{CaCu}_3\text{Ti}_{4-x}\text{Zr}_x\text{O}_{12}$ ($x = 0.02, 0.10, 0.20, 0.50,$ and 1.0 mol%) calcined at 800°C for 3 hrs.

over conventional heating techniques which include reduced processing time, energy saving, fine microstructures, and improved material properties [23–26]. The understanding of the electric and dielectric behavior of these ceramics would facilitate the processing of CCTO to achieve certain electrical properties under desired sintering temperature. To our best knowledge, the dielectric response to the substitution of Zr for Ti in $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ microwave sintered ceramics has not been studied.

2. Experimental Procedure

CCTZO powders were synthesized by the sol-gel method. The starting materials were $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$, $\text{C}_{12}\text{H}_{28}\text{O}_4\text{Ti}$, and zirconium oxychloride. Glacial acetic acid and citric acid were used as chelating reagents. Calcium nitrate and copper nitrate were dissolved in ethanol (solution 1). Titanium tetraisopropoxide stabilized by acetylacetonate was dissolved in isopropyl alcohol. With this solution, zirconium nitrate dissolved in ethanol was mixed (solution 2). After stirring for 40 min, solution 1 was added slowly to solution 2. These two solutions were mixed in a beaker and stirred continuously using a magnetic stirrer at room temperature. Thereafter, acetic acid and citric acid were added to this mixture. The temperature of the magnetic stirrer was increased to 90°C . The ratio of titanium tetraisopropoxide : acetylacetonate : citric acid was kept at 3 : 2 : 0.25, respectively. A transparent solution was obtained after a constant stirring for 3 hrs. When the solution was dried, the viscous liquid began to form a transparent viscous gel. The gel was kept in a hot air oven at 110°C for 2 days. This resulted in a loose and brown ash powder. The dried precursor was calcinated in a conventional furnace at 800°C for complete crystallization. The calcinated powder was uniaxially pressed into pellets of 10 mm in diameter

and 2 mm in thickness with an applied load of 3.5 tons for 4 minutes, using polyvinyl alcohol (PVA) as the binder. Microwave sintering was completed by using a VBCC 2-magnetron microwave heating furnace in which the compacted samples were sintered at $1,000^\circ\text{C}$ at a ramping rate of $50^\circ\text{C}/\text{min}$ and soaked for 10 min followed by fast cooling to room temperature at a rate of $20^\circ\text{C}/\text{min}$. The sintering temperature was measured with a pyrometer and it was standardized with reference to a thermocouple and the emissivity was measured to be between 0.90 and 0.98 for the different samples of CCTZO. The microwave output power level was about 2.2 KW with 2.45 GHz frequency and it was controlled by a Eurotherm PID controller. XRD studies were carried out with an X'PERT-PRO diffractometer (Philips, the Netherlands) using $\text{CuK}\alpha_1$ radiation at 45 kV and 40 ma ($\lambda = 0.154056$ nm) in a wide range of 2θ ($20^\circ < 2\theta < 80^\circ$). The microstructure of sintered pellets was measured by FE-SEM (FEI Quanta 200FEG). Silver paste was coated over the parallel faces of the pellets and the dielectric constant measurements were made using an LCR meter (Hioki 3532-50) in the frequency range of 50 Hz–1 MHz at room temperature.

3. Results and Discussion

3.1. X-Ray Diffraction Pattern of Calcined $\text{CaCu}_3\text{Ti}_{4-x}\text{Zr}_x\text{O}_{12}$ Powder. XRD patterns of CCTZO powder calcinated in a conventional furnace at 800°C for 3 hours are shown in Figure 1. All the XRD diffraction peaks match with the standard powder diffraction pattern (JCPDS card number: 75-2188). The major planes correspond to (211), (220), (013), (222), (400), (422), and (440) of cubic $(\text{CaCu}_3)\text{Ti}_4\text{O}_{12}$. From the XRD patterns it is clear that the concentration of zirconium doped with CCTO is within the limit of its solubility up to $x \leq 0.2$ as an impurity phase is observed in the

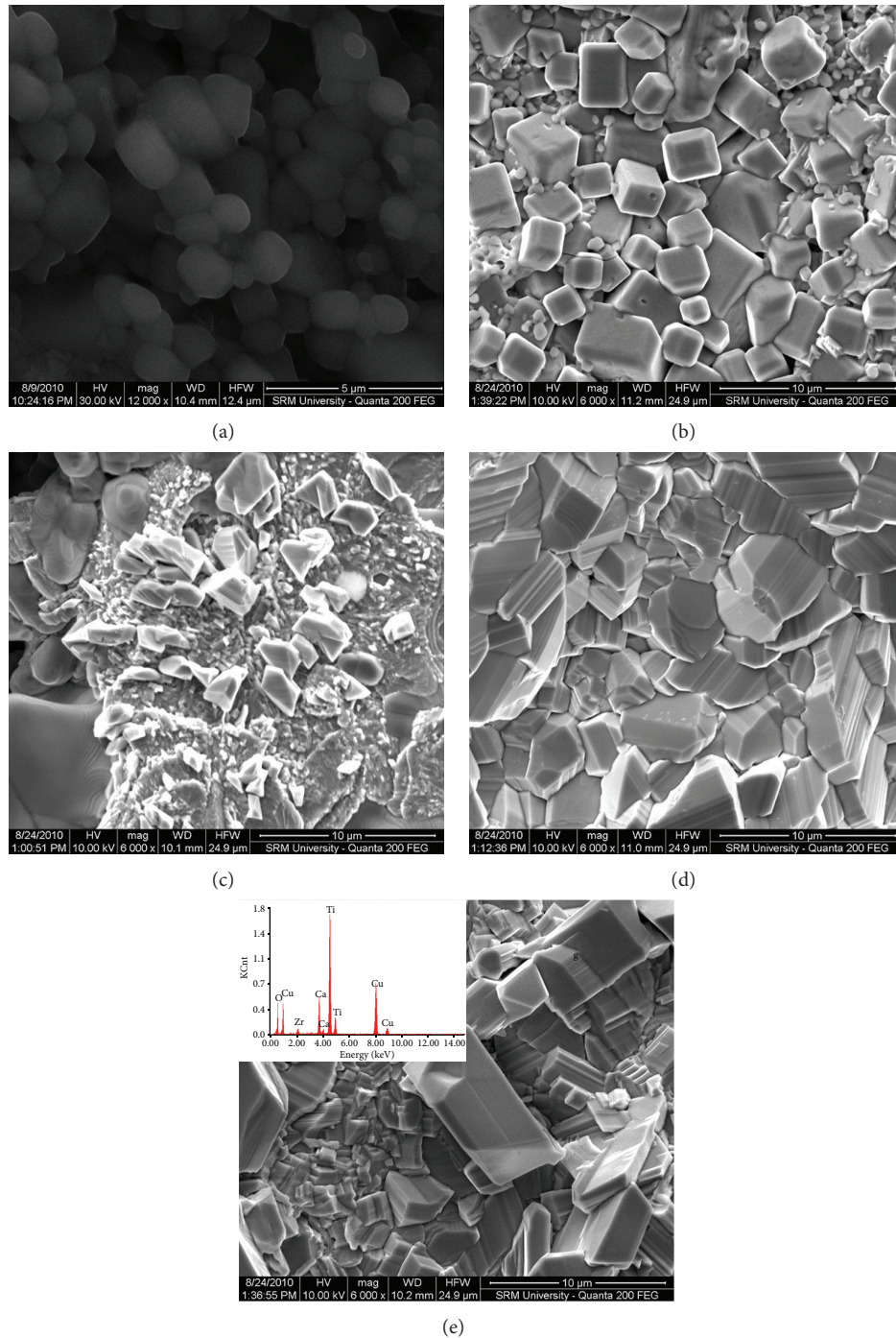


FIGURE 2: FE-SEM micrographs of $\text{CaCu}_3\text{Ti}_{4-x}\text{Zr}_x\text{O}_{12}$ (microwave sintered at 1000°C -10 min) (a) when $x = 0$, (b) $x = 0.02$, (c) $x = 0.10$, (d) $x = 0.20$, and (e) $x = 0.50$.

samples ($x \leq 0.20$). For the sample ($x = 0.5$), extra peaks are present besides the main peaks which indicates that the excess amount of zirconia is not within the limit of its solubility. But the ($x = 0.1$) sample's major peaks belong to the ZrO_2 phase.

3.2. FE-SEM Analysis. The microstructure of $\text{CaCu}_3\text{Ti}_{4-x}\text{Zr}_x\text{O}_{12}$ ($x = 0, 0.02, 0.1, 0.2, \text{ and } 0.5$) ceramics, sintered at 1000°C for 10 min in a microwave furnace, is shown in Figures 2(a)–2(e). The grains are found to be in

the size range of $250\text{ nm} - 5\ \mu\text{m}$. The morphology of the undoped specimen consists of mostly cubic grains. For the dopants with $x = 0.02$, the grains attain cubic shape. When the dopant level increases ($x = 0.1, 0.2, \text{ and } 0.5$), a polyhedral type of grains occurs. Also with the increase in dopant levels, the size of the grains grows bigger and denser. This is in accordance with the earlier report of Jesurani et al. [27]. The porosity is intergranular in nature and is not observed in the samples of higher concentrations of

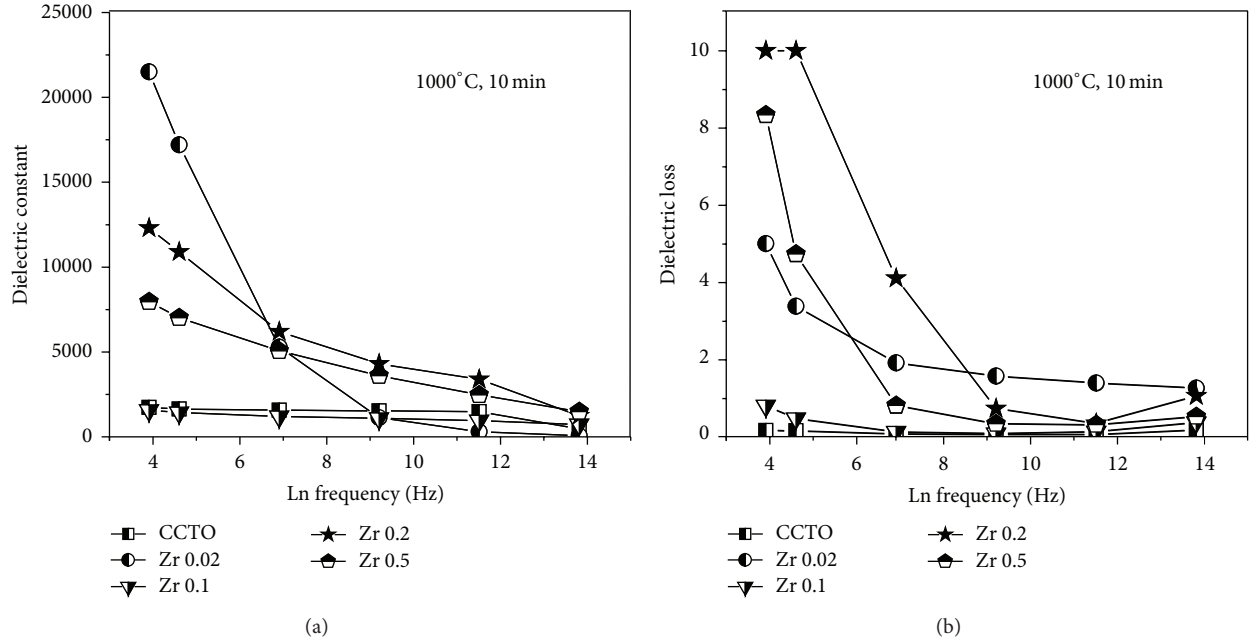


FIGURE 3: Frequency dependence of the dielectric constants and dielectric loss of Zr doped CCTZO ceramics sintered using microwave at 1000°C for 10 min.

TABLE 1: Dielectric constant and loss of CCTO and zirconium dopants using microwave sintering at 1000°C for 10 min.

Frequency Hz	$x = 0$		$x = 0.02$		$x = 0.1$		$x = 0.2$		$x = 0.5$	
	ϵ_r	$\tan \delta$	ϵ_r	$\tan \delta$	ϵ_r	$\tan \delta$	ϵ_r	$\tan \delta$	ϵ_r	$\tan \delta$
50	1730	0.16	21,500	5.00	1570	0.80	12,300	9.99	7960	8.33
100	1633	0.15	17,200	3.38	1430	0.47	10,900	9.99	7020	4.73
1 K	1572	0.06	5290	1.91	1200	0.12	6200	4.11	5060	0.81
10 K	1527	0.06	1140	1.57	1090	0.08	4300	0.73	3610	0.33
100 K	1491	0.06	301	1.39	961	0.13	3390	0.34	2470	0.31
1 M	1462	0.17	74.2	1.25	724	0.37	1240	1.06	1490	0.52

doping due to the high agglomeration of grains. Thomas et al. reported that the dielectric properties of CCTO ceramics have shown a strong dependence on microstructure [28]. The dense microstructure of the sample can possess good dielectric properties. Energy dispersive X-ray (EDX) analysis of $\text{CaCu}_3\text{Ti}_{3.5}\text{Zr}_{0.5}\text{O}_{12}$ ceramic powder shows the evidence for the presence of zirconium in Figure 2(e) (inset).

3.3. Dielectric Studies. Figures 3(a) and 3(b) and Table 1 show the dielectric constant and dielectric loss as a function of frequency of samples sintered at 1,000°C for 10 minutes. The dielectric constants of these samples are 21,500, 1570, 12,300, and 7960 and the corresponding Zr concentrations are 0.02, 0.10, 0.20, and 0.5 for 50 Hz. The dielectric constant does not exhibit a constant variation with frequency for various doping concentrations. However, the dielectric constant (ϵ_r) of pure CCTO ceramic is almost a constant through the whole frequency range with a lower ϵ_r value (1462–1730). This behavior may be attributed to the differences in grain size. According to the internal boundary layer capacitance (IBLC) model, the effective dielectric constant is inversely proportional to the ratio of thickness of the insulating layer

(i.e., grain boundary) to the grain size. A greater number of crystal defects are able to exist in larger grains than in smaller ones. This allows for more internal barrier layers [29]. The dielectric permittivity in CCTO strongly depends on the grain size, and an increase of the grain size leads to an increase of the dielectric permittivity. For IBLC ceramics [30], $\epsilon_{r,\text{eff}} = \epsilon_g b (d_g/e_i)$, where d_g is the grain size and e_i is the thickness of the grain boundary.

Apart from the grain boundary regional defects, the presence of interfacial polarization in these materials also plays an important role for enhancement of the dielectric constant in the low-frequency region. The interfacial space charge polarization arises due to the accumulation of charge carriers at the interface of semiconducting grains and insulating grain boundaries. In the present investigation, CCTZO with zirconium ($x = 0.02$) gives a high dielectric constant 21,500 which is five times larger than that of CCTO [31] and its dielectric loss is 5.

The frequency dependent dielectric loss ($\tan d$) is presented in Figure 3(b) as a function of different Zr concentrations ($x = 0, 0.02, 0.1, 0.2, 0.5$, and 1.0). It can be observed that there is no constant variation of $\tan d$ value with frequency.

The dielectric losses are higher at low frequencies up to 100 Hz. Dielectric loss for all the compositions shows a sharp decreasing trend up to 100 kHz and small rising at 1 MHz. Higher concentration may produce more oxygen vacancies and space charges; thus, more obvious dielectric relaxation can be observed which gives rise to more dielectric losses. These results agree with the previous reports of dielectric behavior of the CCTO ceramics [32, 33].

Choudhury et al. [34] have reported that the dielectric constant of Zr doped BaTiO₃ samples sintered at 1250°C showed a maximum value of about 1000 for 100 kHz frequency. Gómez-Yáñez et al. [35] reported that the dielectric constant and dielectric loss are 576 and 0.33, respectively. When Zr doped BaTiO₃ is synthesized by mechanical alloying [35], its dielectric constant and dielectric loss are 539 and 0.035, respectively. When the sample is sintered in the range of 1200–1400°C, as in the present investigation, a higher dielectric constant (6020) is obtained accompanied by low dielectric loss (0.52). The sintering temperature is also low (1000°C) compared to that in the earlier reports.

4. Conclusion

CaCu₃Ti_{4-x}Zr_xO₁₂ ($x = 0, 0.02, 0.1, 0.2,$ and 0.5) ceramics were successfully synthesized by the sol-gel method. Structural and dielectric characterization of CCTZO ceramics doped with small amounts of Zr was carried out. The use of microwave sintering resulted in the formation of a dense sintered CCTZO in just 10 minutes. When the Zr concentration increased, the grain sizes also increased. The observed dielectric behavior of all the five samples agrees well with the internal barrier layer capacitor (IBLC) model. A high dielectric constant of 21500 was achieved for a moderate Zr doping ($x = 0.02$), that is, for CaCu₃Ti_{3.98}Zr_{0.02}O₁₂ ceramics, and the sintering temperature is also brought down to 1000°C.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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