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# Effect of Calcination Temperature to the Dielectric Properties of $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ using Enhanced Microwave Processing

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**Abstract.**  $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  or calcium copper titanite (CCTO) electroceramics is viably produced with an enhanced process in order to tackle the dielectric loss problem in microelectronic industry. In this study, the influence of microwave radiation temperature during calcination to the electrical properties was investigated. The CCTO samples undergo synthesis process using a solid-state reaction route. The calcination process was conducted for 1 hour at different calcination temperatures (500-800°C) using microwave furnace set at the frequency of 2.45GHz. An enhanced silicon carbide (SiC)-based susceptor was used as crucible to accelerate the process. The result of X-Ray Diffraction (XRD) pattern shows that the phase formation of cubic perovskite CCTO is partially formed after calcination at more than 500°C for 1 hour, but the single-phase CCTO does not form completely during this time duration. The Scanning Electron Microscopy (SEM) analysis shows that with the increasing of calcination temperature, there are patterns of reduction in porosity and grain growth of the sintered CCTO pellets. Dielectric properties also increase within the frequency range of 1 GHz to 10 GHz with the increasing calcination temperature.

## 1. Introduction

$\text{CaCu}_3\text{Ti}_4\text{O}_{12}$  or Calcium Copper Titanite (CCTO) is an electroceramic with tremendously high dielectric properties. It belongs to the perovskite family and its dielectric constant value reaches 100,000 at room temperature with very low-temperature dependence in the range from 100 to 600K [1-2]. With these unique properties, miniaturization of microelectronic devices are possible to fulfill the market demand, provided that its high dielectric loss properties are overcome. Many aspects have been identified and studied by the researchers in order to reduce the high dielectric loss due to long production time and procedures. It is proven that conventional solid-state processing technique that is widely used to produce CCTO is not efficient and has caused the high dielectric loss [3-5]. The production cost is usually high due to the conventional heating system of the process require high temperature with long duration of heat treatment [6]. Hence, several other methods are introduced in order to synthesis the



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CCTO such as microwave technique, mechanical alloying, polymeric citrate precursor route, sol-gel technique, and others [7-10].

Microwave technology is known as a process in which the materials couple with microwaves, generates heat within the materials first and then heats the entire volume. The microwave processing technique in ceramic processing is discovered since the 1950s and further investigation is conducted to date [11]. It is one of the effective choices to reduce long processing time while increasing its densification, lower sintering temperature, low energy-consuming, high efficiency with high dielectric properties and low dielectric loss [12]. In this study, the enhanced microwave is used for the calcination process, while the sintering process is done conventionally. High energy conversion of the microwave is utilized by using high efficient silicon carbide (SiC)-based susceptor crucible to assist the process.

## 2. Materials and Methods

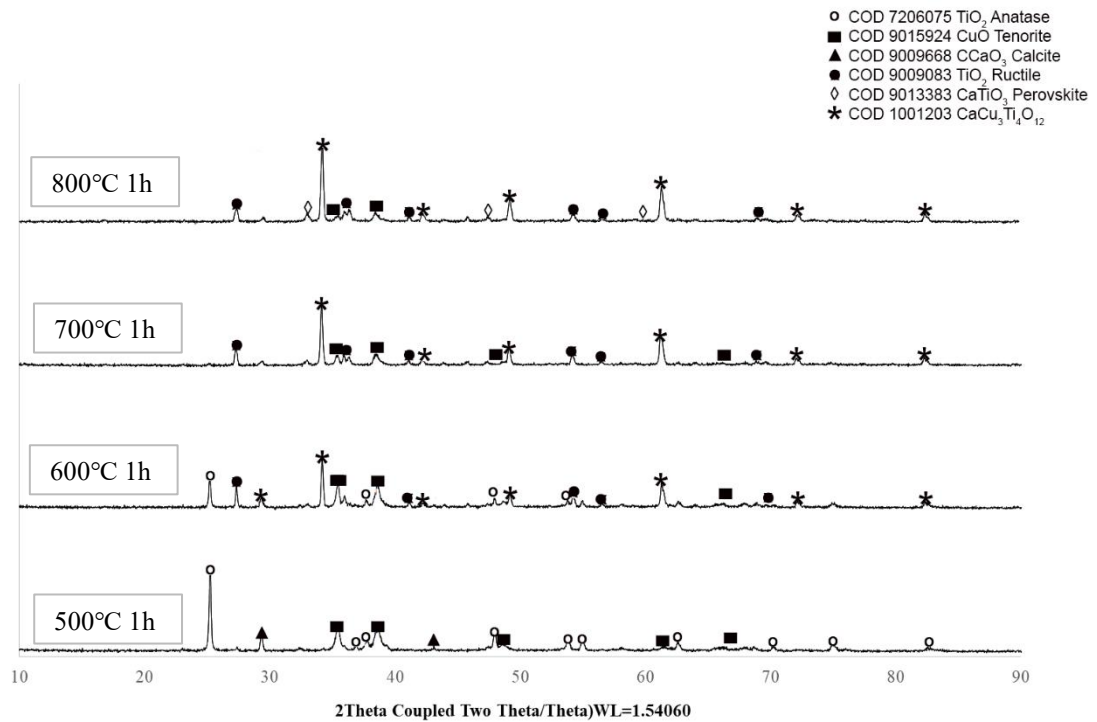
Three types of reagents were weighed according to the stoichiometric ratios to prepare the CCTO sample powder. The powders were mixed with zirconia ball and ethanol using a milling machine at 24 hours [1]. The ratio between raw materials and zirconia ball milling is 1:10, while the ethanol, which acts as medium was poured until the mixture was wet enough for the wet milling process. Then, the sample powder of CCTO formed undergo calcination process assisted by the enhanced SiC-based susceptor in a crucible using microwave auto-controller machine (dawnyx, 2.45 GHz) at 500°C, 600°C, 700°C, 800°C and 900°C for 1 hour. After that, all the calcined sample powders were pressed into cylindrical shapes with 300MPa of pressure using a hydraulic hand press machine. The mold was used to press the sample is 5 mm diameter. The pellets formed were then sintered using a conventional furnace at 1040°C for 10 hours.

The XRD analysis of the CCTO sample were done using Bruker D2 phaser x-ray diffractometer from Bruker (Malaysia) Sdn. Bhd. to investigate the phase formation of CCTO formed. The software used for XRD analysis was via DIFFRAC.EVA software. The phase formation of the sintered CCTO microstructure is observed and compared at different temperature values. The sintered CCTO that calcined in different temperature values were then characterized by using JEOL JSM-IT 100 Scanning Electron Microscope (SEM) to evaluate the microstructure of the CCTO pellets. The analysis of dielectric properties was also conducted using RF Impedance/Material Analyser 4291B Hewlett Packard machine from frequency 1 MHz to 1 GHz and network analyser from frequency 1 GHz to 10 GHz in ambient temperature to observe the dielectric properties behavior.

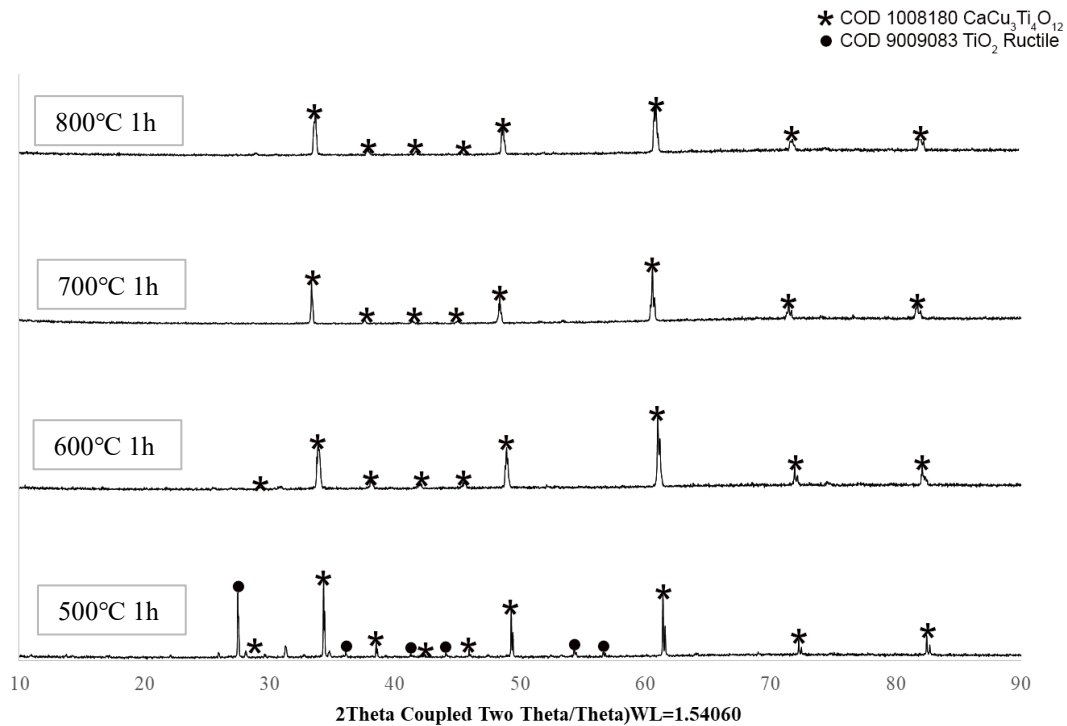
## 3. Result and Discussion

The XRD pattern of calcined powder at different microwave irradiation temperature, which is 500°C, 600°C, 700°C, 800°C, and 900°C in 1 hour is shown respectively in Figure 1. The patterns detected by XRD show that the formation of CCTO has been completed starting at the sample calcined at 600°C. However, there is a presence of multiphase identified at 600°C, 700°C and 800°C. TiO<sub>2</sub> and CuO were detected for sample calcined at 600°C, 700°C and 800°C, while CaTiO<sub>3</sub> was also detected for the sample when calcination is done at 800°C.

After the sintering process at 1040°C for 10 hours, the pellets have undergone XRD analysis. Figure 2 shows the XRD pattern of sintered CCTO pellets at different microwave irradiation temperature. A pure single-phase CCTO has been formed in the sintered pellets, which calcined at 600°C, 700°C and 800°C because all of the peaks detected correspond to CCTO. However, the sintered pellet in 500°C is excluded since there is an existence of the TiO<sub>2</sub> phase that did not fully decompose. It can be concluded that the sintered pellets which calcined at 600°C are enough to produce a single-phase CCTO in which the low calcination temperature is required and the time duration for calcination is shorter than the conventional processing as reported before [4,14].

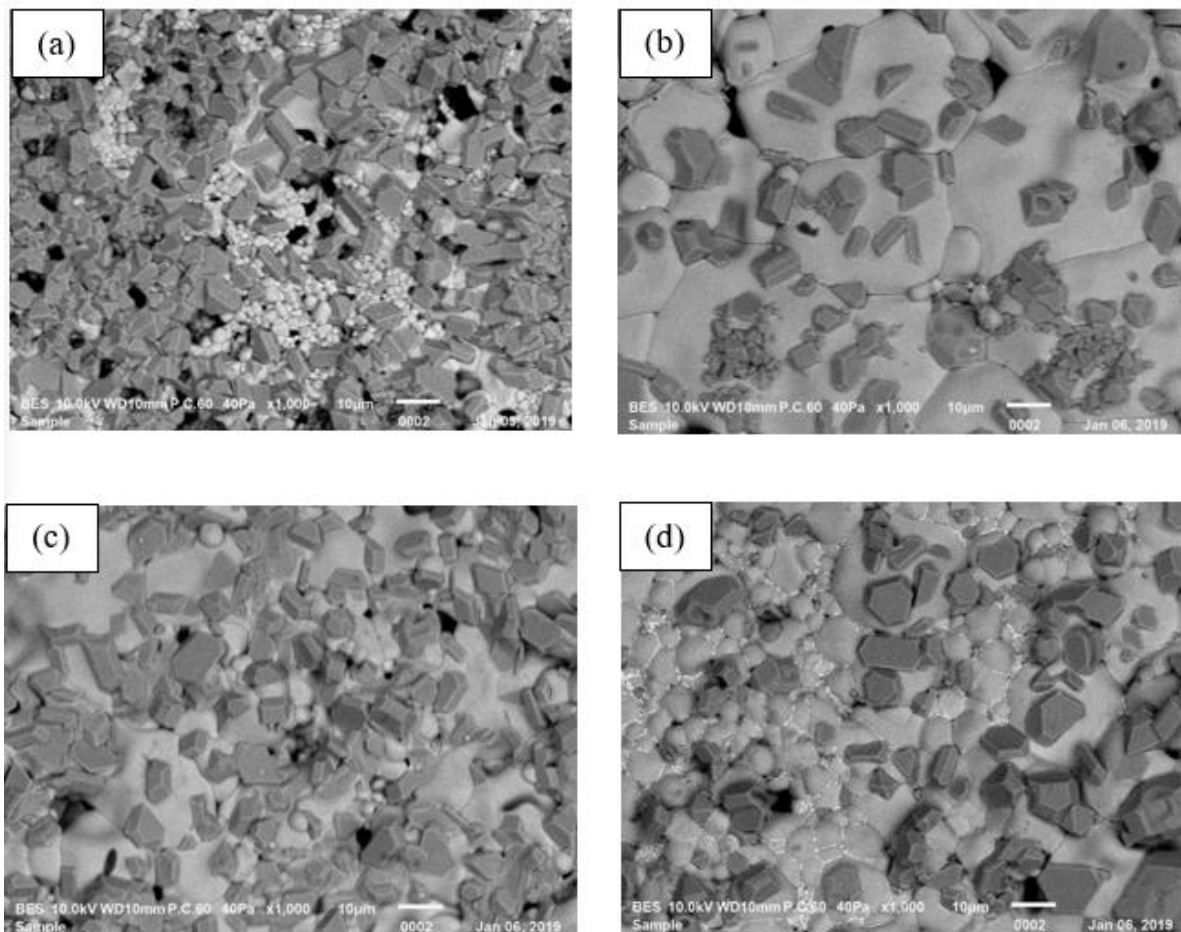


**Figure 1.** XRD pattern of calcined powder in different microwave irradiation temperature (500°C, 600°C, 700°C and 800°C) for 1 hour



**Figure 2.** XRD Pattern of sintered CCTO pellets at 1040°C for 10h of the sample calcined at different microwave irradiation temperature (500°C, 600°C, 700°C and 800°C) for 1 hour

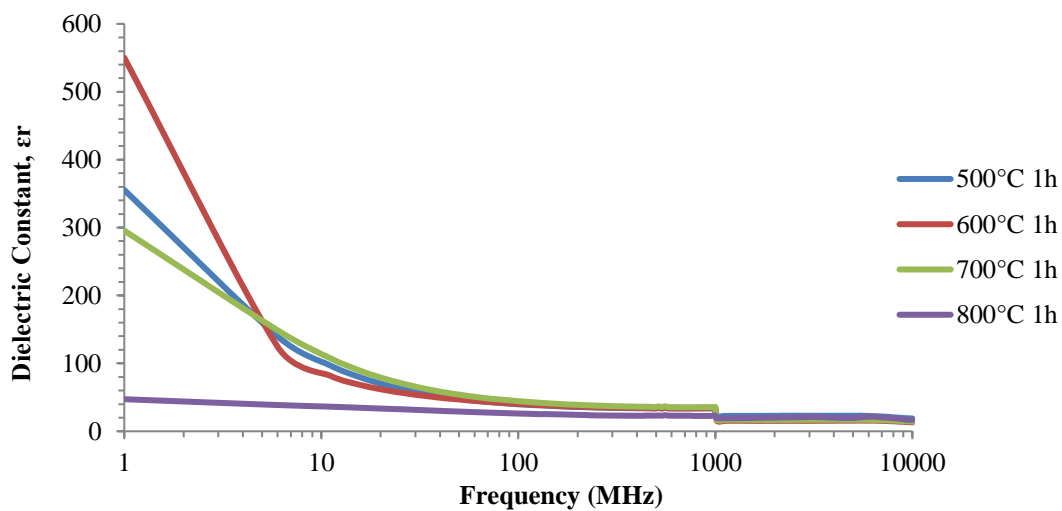
According to Figure 3, the surface microstructure of sintered CCTO pellets at different microwave irradiation temperature at 500°C, 600°C, 700°C and 800 °C for one hour under 1000× magnification which operated in 10 kV. The different morphologies and particles size has been observed clearly by using SEM. During the calcination stage, the increase in temperature promotes the interdiffusion of their ions. Therefore, the amount of CCTO formed is increased. When reached the sintering process, the CCTO particles formed continues the diffusion process as the grain growth happens in order to obtain a homogenous body. There are two types of CCTO can be observed in Figure 3, where the dark region is the CCTO particle that formed to joint together through diffusion with others but not enough time while the light region is the result of grain growth to form a homogenous body. In Figure 3 (d), the small white particles that segregate at the grain boundaries of CCTO ceramics as the causes of massive grain growth have been claimed as CuO since it is the same case has been reported [15].



**Figure 3.** Surface Morphology on sintered CCTO pellets that calcined at different microwave irradiation temperature (a) 500°C, (b) 600°C, (c) 700°C and (d) 800°C for 1 hour

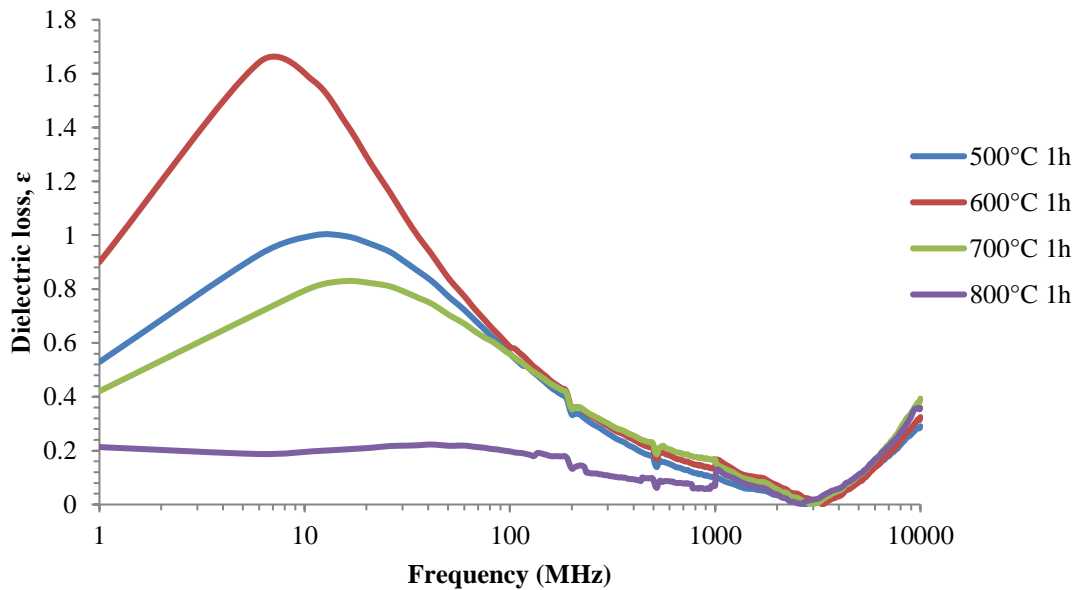
The dielectric properties have been characterized from the frequency range 1 MHz to 10 GHz by using an impedance analyzer and network analyzer. The dielectric constant plot of sintered CCTO pellets, which calcined in different microwave irradiation temperatures is shown in Figure 4. The result shows a significant change of dielectric constant as the function of frequency happens in the

range 1-100 MHz. As the temperature increase, the dielectric constant of the sintered pellets increased. Based on the result in Figure 1, the sintered pellet which calcined in 500°C did not perform single-phase CCTO while the other three parameters had shown single-phase CCTO after the sintering process. Hence, it can be suggested that a sintered pellet that calcined in 500°C can obtain the highest value of dielectric constant value due to the residue of  $\text{TiO}_2$  composition that occupies a higher phase ratio than CCTO composition. By excluding the result of 500°C, as the frequency is in 1 MHz, the highest value of dielectric constant shown in the graph is obtained by sintered pellet calcined at 600°C is about 550.



**Figure 4.** The dielectric constant of sintered CCTO pellets which calcined in different microwave irradiation temperature at 1 hour versus frequency (MHz)

The dielectric loss plot had been shown in Figure 5. It had shown a frequency dependence of dielectric loss of the sintered pellets. In the frequency range 1-50 MHz, there are peaks of all the CCTO sintered pellets. As the temperature increases, the dielectric loss of the sintered pellet decreases. The highest peak value of the dielectric loss is obtained by sintered pellet calcined at 600°C, which is 1.64. According to the results, there is no significant peak has been shown for the sintered pellet, which calcined at 800°C. Based on Figure 3 and 4, it can be concluded that as the calcination temperature increases, the dielectric constant decreases, and the dielectric loss also decreases. As the calcination temperature increases, the polarization to the dielectric constant becomes weak. By comparing calcination time and temperature, low temperature and longer time more effective than higher temperatures but shorter time.



**Figure 5.** The dielectric loss of sintered CCTO pellets which calcined in different microwave irradiation temperature at 1 hour versus frequency (MHz)

#### 4. Conclusions

Enhanced microwave processing is used to investigate the effect of calcination temperature on the properties of CCTO. Based on the XRD analysis, the phase of CCTO was successfully obtained but not in single-phase as it did not wholly form in the calcination process. The phase formation of CCTO is successfully obtained after sintering for all sintered pellets. It is concluded that the sintered pellets which calcined at 600°C are enough to form single-phase CCTO for a one-hour duration. However, the calcination temperature at 900°C is not suitable for microwave processing as the sample melted during the short duration. For the surface morphology of CCTO pellets, as the calcination temperature increased, the amount and particle size of CCTO particles also increased, resulting in the diffusion process had more time duration to generate energy for grain growth. Hence, the grain boundaries can be reduced. For dielectric properties, as the microwave irradiation temperature for calcination increases, the dielectric constant and the dielectric loss also decreases. It is due to the higher dielectric constant sample will suffer a higher effect of the weakening of space charge polarisation.

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