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Influence of Soda Lime Glass Addition on the Dielectric Properties of CCTO Ceramics

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Abstract. The dielectric properties of CCTO ceramics with soda-lime glass addition was investigated. The addition of soda lime glass was varied from 0 - 0.05 wt.% by solid-state reaction. Both XRD patterns of CCTO and CCTO-glass sintered samples showed the formation of CCTO phase with minor peaks of CuO phase, respectively. The phase area analysis of CCTO and CCTO-glass proved that the CCTO phase was found to be decreased while the grain boundaries area of CuO + glass was then increased after the addition of soda-lime glass. The dielectric properties of corresponding samples indicated that both ε_r and tan δ were reduced with glass addition. Thus, it shows that the soda lime glass addition has a significant effect on dielectric properties of CCTO ceramics.

1. Introduction

Nowadays, materials with high dielectric constants and low dielectric loss have attracted a big interest for technological electronic applications such as capacitors, resonators and filters. Hence, $CaCu_3Ti_4O_{12}$ (CCTO) perovskite-like compound has received increasing interest since it has reported to obtain high dielectric constant ($\varepsilon_r \sim 10^4$) at room temperature [1, 2]. However, CCTO also suffering high dielectric loss (> 0.05 at 1 kHz) [3, 4]. Thus, this prevents CCTO to be commercialized. The densification of ceramics body play a vital role to improve the dielectric properties of ceramics. Addition of another material into stoichiometric ratio of CCTO has been shown as an effective method to improve the dielectric properties of CCTO.

There are several researchers reported on the fabrication of CCTO ceramic composites. Previous researchers had put some glass materials into CCTO in order to improve densification of ceramics body thus can reduce dielectric loss [5-7]. Wang et al. (2016) [6] reported that, at 1 MHz, the tan δ of 1wt.% and 3 wt.% of SBS glass added into CCTO were ~0.5 and ~0.7, respectively. Nevertheless, the effect of soda-lime glass addition into CCTO on the dielectric properties of CCTO are still not been explored. To

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the best of our knowledge, the crystallite structure, surface morphology, and dielectric properties of CCTO have been studied for the first time.

Hence, in this study, the effects of soda-lime glass addition on the dielectric properties of CCTO ceramics characterized at high frequencies (1 GHz) were investigated.

2. Experimental Method

CCTO ceramics was prepared via solid state reaction. The raw materials of $CaCO_3$ (Sigma Aldrich, >99%), CuO (Sigma Aldrich, >99%) and TiO_2 (MERCK, >99%) powder was mixed in wet condition for 24 hours and the mixture was dried for overnight in the oven. The dried mixtures were then calcined at 930 °C for 12 hours. The soda-lime glass was milled by using the planetary ball milling machine to form fine powder. The glass powder was mixed with the calcined CCTO powder for 24 hours with the ratio of corresponding powders to the following chemical composition: (100-x) % $CaCu_3Ti_4O_{12}$ + x % glass additives (wt.%), where x = 0, 0.05 and 0.5, respectively. The mixed powder was pressed into pellets shape under a pressure of 250 MPa. The sample was labelled as CCTO for pure CCTO sample while SLG0.05 and SLG0.5 for the samples of CCTO with addition of 0.05 and 0.5 wt.% of soda-lime glass, respectively. The pellets were then sintered at 1040 °C for 10 hours. The XRD (Bruker Advance D8) was used to analyze the phase formation of pure CCTO and CCTO-borosilicate glass composites while Scanning Electron Microscopy (SEM) (TM 3000) was used to observe the microstructures of the samples. For electrical measurement analysis, the dielectric properties of the samples were measured by using impedance analyser machine (4291B Hewlett Packard) at 1 GHz.

3. Results and Discussion

3.1 X-ray diffraction analysis

Figure 1. shows the XRD patterns of CCTO-glass based composites (0.01-0.05 wt.%) compared to pure CCTO. Since the maximum amount of each glass added was only 1 wt.%, the major peaks observed are belong to CCTO in all composites. For each sample, it is observed that minor CuO phase also appeared in XRD patterns. CuO peaks were seen at ~35.5° and ~38.6°. The CCTO phase is compared with ICDD Data File Card No. 01-075-2188 whereas CuO phase is matched with ICDD Data File Card No. 00-045-0937. The CuO phase existing for each sample is from the CuO compound that precipitate or segregated at the grain boundary of CCTO. Similar kind of results were also reported by Ab Rahman et al. (2016) [8] where the presence of CuO compound was observed at the grain boundary of CCTO after being sintered at 1040 °C for 10 hours. According to Yuan et al. (2013) [9], the intergranular CuO phase is due to the instability (or activity) of Cu ions in the CCTO lattice. Cu ions were first separated out from CCTO at ~1000 °C, then were ousted to the surface layer of the pellet and mostly segregated at the grain boundaries and were finally oxidized to CuO compound.

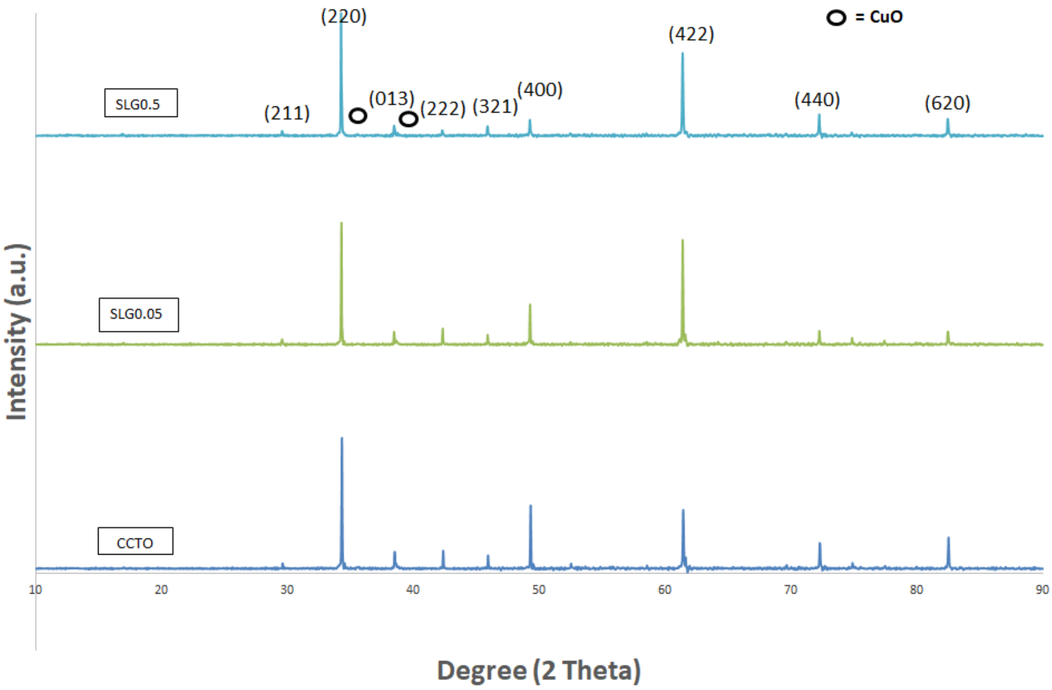

Figure 1. X-ray diffraction patterns of the sintered CCTO and CCTO-soda-lime glass composites

3.2 Phase area analysis

Figure 2. shows the area percentages of the grain and the grain boundary of CCTO and CCTO-soda-lime glass-based composites calculated by using i-Solution DT software where the grain, grain boundaries and the porosity were represented by the orange, green and blue colors, respectively. Based on these figures, it was seen that the grain area was decreased whereas the grain boundaries area was increased by increasing the glass addition into CCTO ceramics. The grain area was decreased from 87.90% (CCTO) to 83.05% (SLG1) while grain boundary area was increased from 9.78% to 15.99% for the same samples. Thus, it shows that the glass addition seems to decrease the grain area and increase the grain boundaries of CCTO. Patel et al. (2015) [10] also have reported where with increasing the glass content, the grain boundary of $BaTiO_3$ ceramics becomes larger and the average grain separation was increased.

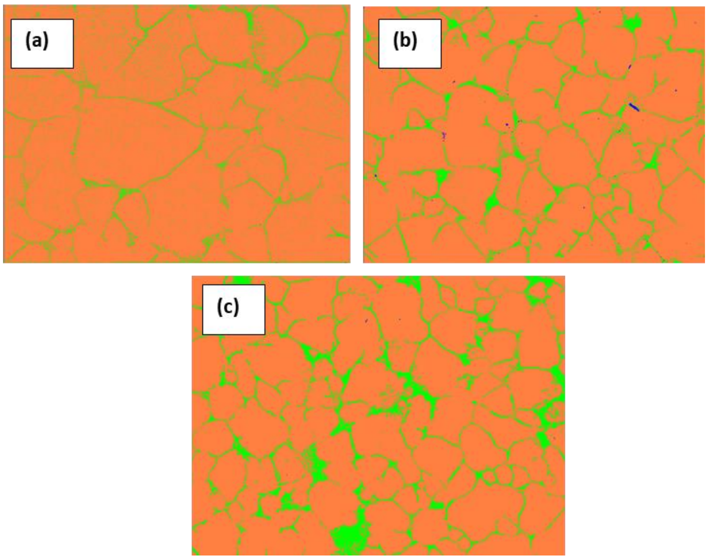

Figure 2. Phase area micrograph of CCTO different soda-lime glass composition (a) 0 wt. %, (b) 0.05 wt. % and (c) 0.5 wt. %.

3.3 Dielectric properties analysis

Figure 3 showed the ε_r and tan δ of CCTO and CCTO-SLG glass composites sample. At frequency 1 GHz, all CCTO-SLG glass composite samples exhibit lower ε_r than CCTO sample where SLG0.5 had lowest ε_r of 59. Meanwhile, the tan δ of CCTO was reduced from 0.76 (CCTO) to 0.54 (SLG0.5). Glass is known to have low tan δ [6]. The addition of low tan δ glass materials into the high tan δ ceramic materials can help to reduce tan δ of ceramic materials.

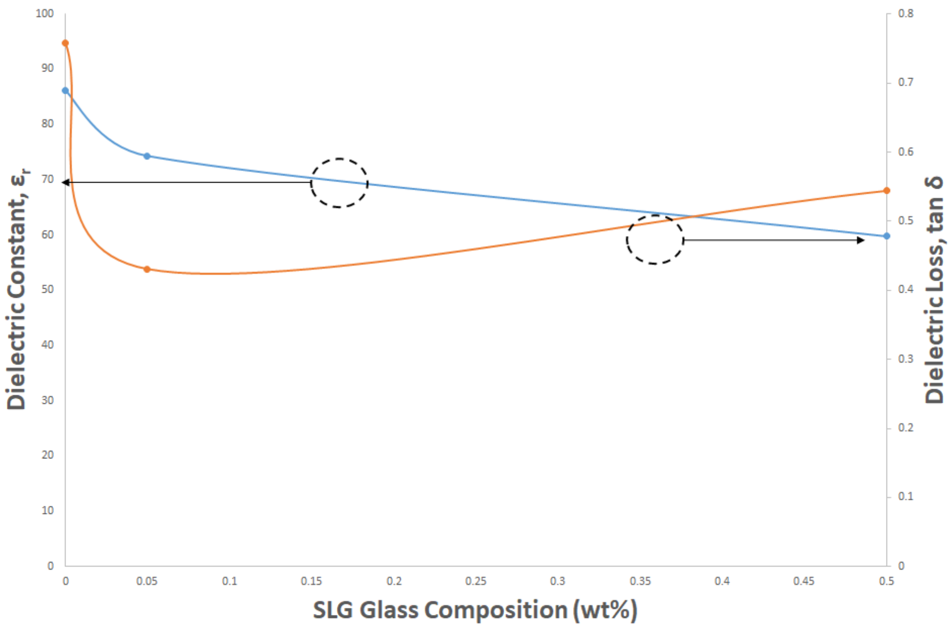

Figure 3. ε_r and tan δ of CCTO and CCTO-soda-lime glass composites at 1 GHz

4. Conclusion

CCTO and CCTO-soda-lime glass composites were successfully prepared via solid state reaction. From the XRD analysis, the formation of CCTO phase alongside with minor CuO phase were found to be seen for each sample which were sintered at 1040 °C for 10 hours. The analysis of area percentage showed that the grain size of CCTO was decreased while the grain boundaries were increased with increasing glass concentration. The ε_r of CCTO was increased while the tan δ of CCTO was decreased with small addition of soda-lime glass. Thus, it shows that the soda-lime glass addition into CCTO can give the greatest effect on the dielectric properties of CCTO ceramics.

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