Dielectric Breakdown Strength and Energy Storage Density of CCTO-ZBS Electroceramic

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Abstract. The effect of ZnO-B₂O₃-SiO₂ (ZBS) glass additives to the microstructure and electrical properties of CaCu₃Ti₄O₁₂ (CCTO) electroceramic has been successfully investigated in this research. CCTO and ZBS glass additives were prepared via solid state reaction and melt quench techniques, respectively. Raw materials of both CCTO and ZBS were wet mixed separately for 24 hours, dried overnight and the CCTO powder was calcined at 900 °C for 12 hours using an electrical carbolite furnace. After that, the ZBS powder was melted at 1400 °C for 2 hours using an elevator hearth furnace. The ZBS glass was grinded to form fine powder. Different weight percentages (0, 1, 3, 5, 7 and 10 wt%) of ZBS glass powder were added into CCTO (CCTO-ZBS powders), then the powders were wet mixed for 24 hours. The CCTO-ZBS mixtures were dried overnight, compacted at 300 MPa using hydraulic pressure of 6 to 9 mm diameter and 1 to 2 mm thickness (for dielectric properties test) and at 200 MPa of 50 mm diameter and 3 mm thickness (for dielectric breakdown strength test), then sintered at 1040 °C for 10 hours using an electrical carbolite furnace. The addition of a small amount of ZBS glass about 1 wt% was able to increase the dielectric constants (33.99%) and reduce the dielectric loss (5.14%) of CCTO measured at 1 MHz. This addition has also increased the relative density to the maximum value (95.90%), helped the formation of single phase of CCTO, increased the grain size (0.35%) and reduced the porosity as compared to pure CCTO. Meanwhile the dielectric breakdown strength (58.0%) and volumetric energy storage density (80.9%) has also improved with 7 wt% of ZBS glass addition.

1. Introduction

The titanate compound of CaCu₃Ti₄O₁₂ (CCTO) is one of the dielectric material, belongs to a family of the type ACu₃Ti₄O₁₂ (where A=Ca, Cd and etc.) and has been reported in the year of 1967 [1]. Calcium copper titanate, CaCu₃Ti₄O₁₂ (CCTO) is one of the ceramic compounds with perovskite structures that are found to be very interesting dielectric properties. CCTO has attracted much interest because of its
high dielectric constant (up to $10^5$) over a broad temperature range (100 to 600 K) and microwave frequency ($10^2$ to $10^6$ Hz) [2] without show any structural phase transition, making it an ideal material for wide range applications. The advantage of high dielectric permittivity is the fact that the physical volume of capacitive components can be reduced and thereby a decrease in the size of devices products [3]. Moreover, their relative dielectric loss is about 0.15 at 1 kHz, which is considerably low as compared with other dielectric and ferroelectric materials such as BaTiO$_3$, making it a highly potential material for practical applications in microelectronic such as capacitors and memory devices [4, 5].

This compounds have been extended by Joubert, (1979) for many years until now. Patterson et al. (2005) report that the additions of ZrO$_2$ into the CCTO electroceramic could maintain a high permittivity and reduces the dielectric loss. The study of relaxor behavior in CCTO electroceramic has been reported by Ke et al. (2006), the dielectric properties of CCTO have been studied from 143 to 573 K and show that a relaxor ferroelectric behavior occurs at high temperature in CCTO electroceramic. The microstructural and dielectric properties of the CCTO with the changes in the composition of Copper (Cu) have been studied by Lee et al. (2007). The change of Cu composition will affect the grain size and grain growth, in which will affect the dielectric constant of CCTO electroceramic. Babu and Govindan, (2014) report that the dielectric properties of CCTO with the modification of solid state reaction route to prepare the CCTO electroceramic, then the results showed that the dielectric properties of CCTO are frequency ($10^2$ to $10^6$ Hz) and temperature (30 to 300 °C) dependent. The processing temperature are very sensitive to the end results of CCTO, such as sintering temperature play an important role to the microstructure evolution and dielectric properties response of CCTO electroceramic. This statement has been proved by Johari et al. (2016). In another study, sintering at temperature of 1040 °C will give the dielectric constant about ~3748, while sintering at temperature of 1020 °C will give the dielectric loss about ~0.025' have been reported by Fariz et al. (2016).

Calcium copper titanate (CCTO) has attracted much interest because of its extraordinarily high dielectric constant of 100,000 at room temperature and very small temperature dependence in a broad temperature range, from 100 to 400 K. Furthermore, the dielectric constant of CCTO is almost frequency independent below $10^6$ Hz, which is desirable for many microelectronic applications. However, the remains problem of CCTO is the dielectric loss is relatively high for commercial applications. The average value of dielectric loss of CCTO is about 0.5 at 1 kHz at room temperature [11, 12]. This value is considered as high for commercial application (Wang et al., 2018). Therefore, this high dielectric loss of CCTO need to be reduced in order to improve the performance of CCTO based devices.

It is necessary to know the origin of dielectric loss in CCTO ceramics which is still being discussed among researchers, to find an effective method to reduce it (Boonlakhorn et al., 2018; Cortés et al., 2018; Pal et al., 2018; Thiruramanathan et al., 2018). There are many factors which cause dielectric loss in this complex ceramic ingredient such as the contributions of an extrinsic nature which associated with imperfections in the crystal structure such as microstructural defects, micro-cracks, grain boundary, porosity and impurities (Alford & Penn, 1996; Ferrarelli et al., 2006). The distribution of a small amount of particles or secondary phase tend to give an effect to the electrical properties of the material [19].

There are many methods to decrease the dielectric loss of CCTO including addition, substitution, doping and two-phase composite process. One of the method currently being used among the researcher by adding a glass material in ceramic systems such as addition of single component of B$_2$O$_3$ and multi component BaO-B$_2$O$_3$-SiO$_2$ (BBS) glass in CCTO electroceramic as reported by Prakash and Varma, (2007). Wang et al. (2012) report that the effect of SrO-B$_2$O$_3$-SiO$_2$ (SBS) glass addition in CCTO are able to increase the dielectric constant (up to $10^6$) and lowered the dielectric losses of CCTO over the frequency range of 450 Hz to 40 kHz at the relatively lower sintering temperature for relatively shorter sintering time (1,050 °C, 12 hours). Addition a glassy material in ceramics can build grain–grain bridges through liquid–phase formation at the grain boundary during sintering. The glassy phase could facilitate the grain growth and help in the elimination of inter–grain pores [23]. Generally glassy phases
have high electrical resistance and low dielectric loss, so that the formation of this kind of phases, surrounding grains, could assist in the lowering of dielectric loss and in improving the overall dielectric properties of CCTO (Amaral et al., 2010). There are some reports about the effect of addition of glass materials into the CCTO, but there is no systematic study of the microstructure and dielectric properties as function of addition with ZnO-B₂O₃-SiO₂ (ZBS) glass.

The present study aims at obtaining high-density CCTO ceramics in the presence of proper amount of liquid phase. The reason for choosing ZBS as glass additive, owing to its low temperature sintering additive around 560 °C (Liu et al., 2012; Liao et al., 2016), as it might form grain to grain bridges (Shangguan et al., 2016), ability to reduce the densification temperature (Hsiang et al., 2012) and pores eliminations [29]. Hsiang et al. (2008) report that, addition of ZBS glass content into BaTiO₃ could improve the dielectric constant, lowering dielectric loss without shown secondary phases. According to M. Li et al. (2013), the surface morphology of the ZBS glass-ceramic showed that the amount of the crystal was dense as it might helping in improving the relative density. Therefore, this work was focused on the effect of addition of ZBS glass powder in CCTO electroceramic to the phase formation, microstructure, density, dielectric properties, dielectric breakdown strength and energy storage density.

2. Materials and Methods
CaCu₃Ti₄O₁₂ (CCTO) was prepared through solid state reaction method. CaCO₃ (99.99%, Sigma-Aldrich, Japan), CuO (99.99%, Sigma-Aldrich, Japan) and TiO₂ (99.99%, Sigma Aldrich, Germany) are selected as starting raw material. The raw material was wet mixed for 24 hours, dried overnight and then calcined at 900 °C for 12 hours. The ZBS glass powder using ZnO (99.9%, Sigma-Aldrich, USA), B₂O₃ (99.99%, Sigma-Aldrich, USA) and SiO₂ (99.998%, Acros, USA) as starting raw materials and was prepared through melt quench technique with ratio composition of 60wt% ZnO – 30wt%B₂O₃ – 10%SiO₂ (ZBS). The glass materials (50 g) was mixed, sieved and melted at 1400 °C in an alumina crucible about 2 hours [12] in a glass-melting furnace (Lenton EHF 18/3).

The molten glass was manually poured into distilled water forming a glass frits, and then the glass frits were ball milled using planetary mono mill (PULVERISSETTE 6’) to form a fine glass powders. The ZBS glass was mixed with CCTO’s calcined powder for 24 hours following the chemical composition: (100-x) % CaCu₃Ti₄O₁₂ + x % ZBS (CCTO-ZBS), where x = 0, 1, 3, 5, 7 and 10 wt%. The samples were labelled as M1, M2, M3, M4, M5 and M6, represented the CCTO with addition of 0, 1, 3, 5, 7 and 10 wt % of ZBS glass respectively. The mixed powder was pressed into a pellets under a pressure of 300 MPa to form a pellet sample with a diameter of 6mm and 9mm with a thickness of 2 mm by using hydraulic hand press.

The green body pellet was sintered in the temperature of 1040 °C for 10 hours in air with 5 °C/min heating rate. The relative and bulk density of the samples was measured by Archimedes principle. X-Ray Diffraetometer (XRD) analysis was employed to determine the phase formation of CCTO phase. Scanning Electron Microscopy (SEM) was used to observe the microstructure of the samples. The dielectric properties of the samples were measured by using Impedance Analyzer machine (RF Impedance/Material Analyzer 4291B Hewlett Packard) over a frequency range from 1 MHz to 1 GHz.

3. Results and Discussion
3.1. Density of Sintered CCTO-ZBS
The relative density was plotted in the Fig. 1. The densest samples were obtained by the samples added with 1 wt% of ZBS glass addition into the CCTO as represented by samples coded with M2. M2 samples have the highest bulk density (4.846 g/cm³) and relative density (95.9 %). Meanwhile, M6 (10 wt% of ZBS glass addition) sample have the lowest bulk density (4.48 g/cm³) and relative density (88.6 %). The density of M2 sample is increasing about 0.104 g/cm³ (2.19 %) compared to pure CCTO (M1), but it drops when the glass addition is increasing. As a result, a small or proper amount of ZBS glass addition
was helpful in improving the density of CCTO. This trends also reported by other researcher where ZBS act as glass additive to the other dielectric material $\text{Ba}_2\text{Ti}_3\text{Nb}_4\text{O}_{18}$ ceramic by Liu \& Chen (2012) and $\text{Li}_2\text{ZnTi}_3\text{O}_8$ ceramics by Lv et al. (2013). The ZBS glass liquid phase might fill the pores efficiently between the grain contribute to the improved density shown by M2 samples during sintering process. The change in density should closely relate to the microstructure. Addition more than 1 wt% resulting a drastically drops relative density which might attributed from the abnormal grain growth during sintering process, thus increasing the porosity then reduce the density. Lai et al. (2016) mention that, an excess glassy content can lead to the volatilization amounts of boron, resulting in reduction in densification of ceramics compared with a small amount as low as 2 wt% of glass content in their sample.

Figure 1. Relative density of CCTO-ZBS samples.

3.2. Phase Structure of Sintered CCTO-ZBS

The XRD pattern for CCTO-ZBS pellets with different amount of ZBS glass addition that sintered at 1040 °C for 10 hours are shown in Fig. 2.0. All XRD pattern shows single phases (purity: 100.0 %) formation of CCTO without trace of un-reacted raw material or ZBS glassy phase which matched with similar ICDD Ref. code number: 98-001-3852 for CCTO and indexed as cubic crystal symmetry with space group of $\text{Im}3$ and space number of 204. This result indicates that the addition of ZBS glass did not modified the cubic perovskite structure of CCTO. Therefore, the addition of ZBS glass did not lead the formation of a secondary phase and expected to obtain better properties in this work. The same trends also reported by the others researcher when the ZBS glass was added into other system of electroceramics (Liu et al., (2012); Lv et al., (2013a)).
In the meantime, the lattice parameter has been distorted after the glass was added. This happens because some of the glassy phase substitute to the host atom in stoichiometry of CCTO crystal structure. The peak shifted on XRD results can be stated as distortion of the lattice parameter of crystal structure after glass addition. Fig. 3.0 shows a close up observation on the highest intensity peak of CCTO-ZBS which is coded to the 220 plane. It was observed, an addition of 1 wt% (M2) and 7 wt% (M5) of ZBS glass tend to shift the peaks to a high angle. The additions of 3 wt% (M3) increase the intensity at the same angle as pure CCTO (M1). Meanwhile, for 5 wt% (M4) and 10 wt% (M6) tend to shifted at low angle. This all shifted angle proves the ZBS glassy phase can substitute into the CCTO lattice structure. However, no secondary phase was detected. Besides that, all ZBS added CCTO sample shows increased intensity higher than pure CCTO. Table 1.0 shows the concentration and net intensity of ZnO, B₂O₃ and SiO₂ in ZBS glass powder characterized through XRF.
Figure 3. Close up XRD pattern of CCTO-ZBS sintered samples (a) M1, (b) M2, (c) M3, (d) M4, (e) M5 and (f) M6.

Table 1. XRF results of ZnO, B₂O₃ and SiO₂.

| Formula | Concentration | Net Intensity |
|---------|---------------|---------------|
| ZnO     | 77.8000       | 56903         |
| B₂O₃    | 7.5940        | 121.3         |
| SiO₂    | 14.606        | 690.8         |

3.3. Microstructure of Sintered CCTO-ZBS

Fig. 4.0 shows the micrograph of the samples for the top surface at 500 and 1000 (inset at upper right corner) magnification for each sample respectively. Fig. 4.0 (a)-(f) shows a clear microstructure feature at top surface that interact with the atmosphere of the furnace during the sintering process. The average grain size of the sample was measured using Image J tools software. All the grain size of the CCTO-ZBS sample is almost in the same size except for M1 and M2 sample. The pure CCTO sample (M1) shows a dense microstructure with low porosity appearance with average grain size of ~ 2.81 μm and it was increased up to ~ 2.82 μm after 1 wt% of ZBS glass addition (M2 sample) with uniform morphology. This can be concluded that a small quantity of additive can increase the average grain size. The M2 sample also displays an improved densification and less porosity. The addition of ZBS resulted in a denser microstructure and a decreased amount and size of pores (Fig. 4.0 (b)). The previous studied by Li et al. (2010) have mentioned that the addition of ZBS glass can increase the densification and grain size of their sample. However, further addition of the ZBS glass in this work reduce the average grain size, increase the presence of porosity and reduce the relative density same as reported by Liu et al. (2012). Liu et al. (2012) found that, ZBS glass addition can reduce the average grain size from 15 μm for pure Ba₂Ti₃Nb₄O₁₈ (BTNO) ceramic to 1 μm due to fast grain growth at maximum amount of ZBS glass concentration. Fig. 5.0 displays the average grain size of all CCTO-ZBS sample.
Figure 4. Microstructures for the top surface (a) M1, (b) M2, (c) M3, (d) M4, (e) M5 and (f) M6.

Figure 5. Average grain size of CCTO-ZBS samples
3.4. Dielectric Breakdown Strength of Sintered CCTO-ZBS

The dielectric breakdown strength (EBD) of CCTO-ZBS sample as a function of ZBS glass concentration is shown in Fig. 6.0. It was understood that, the EBD is primarily dependent on the density, grain size, porosity distribution and chemistry modification [35]. Usually, highly densified dielectrics material will have high value of EBD. Then, M2 sample was expected to possess the highest EBD because it shows the best results for other measurement, for instance highest density, highest dielectric constant, lowest dielectric loss, CCTO single phase formation, low area of porosity and biggest grains size. As shown in Fig. 6.0, almost all samples seem display conductivity properties. This can be observed when the value of breakdown voltage (VB) of measured sample approaching to zero. As can be seen in Table 1.0 the VB of the all sintered samples improve only a small value from zero. A good dielectric material should have a low conductivity value to be functioned as a good passive component (Li, J. et al., 2013). Moulson, (2003) also mention that a good dielectric material is also a good insulator. These conductivity properties might result from semiconducting grain effect of CCTO which conduct the electricity. One of the reason by selecting the ZBS glass powder as reinforcement in this composites system is to help filling the area of insulated grain boundary to reduce the current flow or blocking the electron mobility since the ZBS glassy phase is a good insulator and have been reported a good result by Li et al. (2010) in their sample of Ba$_{0.3}$Sr$_{0.7}$TiO$_3$ ceramics.

Technically, highly insulated material will show high value of VB under strong electric field. According to the trends of Fig. 6.0, the addition of ZBS as sintering aids shows an increasing trend of EBD of CCTO electroceramic. M5 sample shows the maximum EBD of CCTO which have a high amount of glassy content (7wt %). Hence, the addition of ZBS glass able to improve the EBD of CCTO comparing to the reported value of 1.0–2.0 kV/cm [37]. Even though M5 sample obtain a highest EBD, it is still considered as low EBD which limit further application of CCTO which also reflecting a moderate conductivity. These phenomena have been noted by Barber et al. (2009), the ceramic capacitors have a low inherent breakdown field strength, which results in low energy density. Moreover, it is difficult to manufacture ceramic capacitors with the desired high capacity for energy storage comparing to polymer composites.

Referring to Fig. 4.0 (e), the ZBS glass in M5 sample clearly forming a network of glassy phase coating between the grain might be the reason of the highest EBD. Unfortunately, further addition downgrades the value of EBD showed by M6 sample. This can be suggested that a proper amount of ZBS glass can improve the EBD of CCTO. Table 2.0 shows the value of VB and EBD of the sample.

| Table 2. Dielectric breakdown strength of CCTO-ZBS samples. |
|------------------|------------------|------------------|
| Sample | Breakdown Voltage (V$_B$, kV) | Dielectric Breakdown Strength (E$_{BD}$, kV/cm) |
|--------|--------------------------------|----------------------------------|
| M1     | 0.63                           | 2.1                              |
| M2     | 0.70                           | 2.3                              |
| M3     | 0.90                           | 3.0                              |
| M4     | 0.90                           | 3.0                              |
| M5     | 1.50                           | 5.0                              |
| M6     | 1.23                           | 4.1                              |
3.5. Energy Storage Density ($\tilde{W}$) of Sintered CCTO-ZBS

The $\tilde{W}$ of sintered CCTO-ZBS sample was tabulated and plotted in Table 3.0 and Fig. 7.0 as a function of ZBS glass concentration. From the sub-section of 3.4, the M5 sample was predicted as the highest $\tilde{W}$ since it’s possess the highest $E_{BD}$. Therefore, M5 sample shows the highest $\tilde{W}$ value about 0.02565 J/cm$^3$ which improving relatively up to 80.97 % compared to M1 sample that obtain the lowest value (0.00488). This was proved a proper amount of ZBS glass addition can enhance the $\tilde{W}$ of CCTO electroceramic.

However, $\tilde{W}$ value of M5 samples still considered as low coefficient. The grain size and other intrinsic effect might the reason of this low value due to small average grain size, high porosity, and low density obtained by M5 sample. Further investigation could help to overcome this obstacle. Naturally, the $\tilde{W}$ value was strongly dependent to both $\epsilon$ and $E_{BD}$. Improving only single dependent variable alone did not help in improving the $\tilde{W}$. According to Equation 1.0, the $\tilde{W}$ is directly proportional to both $\epsilon$ and $E_{BD}$.

It is well known, to obtain a high value for both parameters simultaneously and a predictable value of $\tilde{W}$ is very difficult as shown in Fig. 7.0. The investigation in searching a high value of $\tilde{W}$ in electroceramic is a matter of debate among the researchers today (Young et al., (2007); Li et al., (2013b); Fariz et al., (2017)).

$$W = \frac{1}{2}CV_B^2$$ (1)
Table 3. Energy storage density of CCTO-ZBS sample.

| Sample | Capacitance (F) | Breakdown Voltage (kV) | Energy storage density (J) | Energy storage density (J/cm³) |
|--------|----------------|------------------------|---------------------------|-----------------------------|
| M1     | 0.4439         | 0.63                   | 0.06207                   | 0.00488                     |
| M2     | 0.5913         | 0.70                   | 0.14490                   | 0.01140                     |
| M3     | 0.2750         | 0.90                   | 0.11140                   | 0.00876                     |
| M4     | 0.1957         | 0.90                   | 0.07930                   | 0.00623                     |
| M5     | 0.2900         | 1.50                   | 0.32630                   | 0.02565                     |
| M6     | 0.2650         | 1.23                   | 0.20050                   | 0.01576                     |

Figure 7. Energy storage density of CCTO-ZBS samples

4. Conclusion
The addition of ZBS glass about 1 wt% into CCTO had improved the relative density up to 95.8% and bulk density about 4.846 g/cm³. The single phase of CCTO was obtained for all sintered sample ranging from 0 to 10 wt%, although the lattice parameter has been distorted after addition. ZBS glass addition improves the average grain size up to 2.82 μm with homogenous grain size distribution and low porosity.

For dielectric properties, the addition of 1 wt% of ZBS glass can upgrade the dielectric constant (ε_r) up to 3368.23 (33.99%) and reduce the dielectric loss (tan δ) as low as 1.27 (5.14%) at 1 MHz. Meanwhile at 1 GHz, the addition of ZBS glass can upgrade the ε_r about 59.72 (18.85%) and reduce the tan δ about 0.204538 (21.21%).

The dielectric properties as a function of highest amount of glass addition (10 wt%) reveal that it can reduce the ε_r about 1483.47 (69.40%) and increase the tan δ about 1.44575 (6.83%) at 1 MHz. For high frequency region (1 GHz), this glass addition was improving the ε_r up to 60.4 (19.80%) and also improving the tan δ down to 0.083720 (66.23%). ZBS glass addition also able to enhance the dielectric breakdown strength (E_BD) of CCTO about 5.0 kV/cm (58%) for 7 wt%. Hence, for volumetric energy
storage density (\(\bar{\rho}\)) of CCTO, the addition of ZBS glass powder was upgraded until 0.02565 J/cm³ (80.97%) also for 7 wt%.

Acknowledgement
This work was financially supported by Fundamental Research Grant Scheme (FRGS) under the Grant no. 000284.

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