UNIVERSITI TEKNOLOGI MARA

THE EFFECT OF GLASS ADDITION AND THE GRAIN DIAMETERS ON DIELECTRIC PROPERTIES OF $CaCu_3Ti_4O_{12}$ (CCTO)

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CONFIRMATION BY PANEL OF EXAMINERS

I certify that a Panel of Examiners has met on 5th October 2015 to conduct the final examination of Masturah Binti Mohamed on her Master of Science thesis entitled "The Effect of Glass Addition and the Grain Diameters on Dielectric Properties of $CaCu_3Ti_4O_{12}$ (CCTO)" in accordance with Universiti Teknologi MARA act 1976 (Akta 173). The Panel of Examiners recommends that the students be awarded the relevant degree. The Panel of Examiners was a follows:

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ABSTRACT

Calcium copper titanate, $CaCu_3Ti_4O_{12}$ (CCTO) is well known to have colossal dielectric constant in the range of 10^5. This phenomenon was reported to have a strong dependence on doping condition and processing method. However, while exhibiting high dielectric constant, CCTO also shows a high dielectric loss. This has motivated researchers to conduct several studies in improving the final dielectric characteristic of CCTO. Two separate studies on the effect of doping concentration and the effect of processing method on dielectric properties of CCTO were conducted. For the first part, two types of glasses; electrical conducting V_2O_5-TeO glass (VT) and ionic conducting PbO-B_2O_3 glass (PB), were added into CCTO. Both composites were prepared by conventional solid-state synthesis method. X-ray diffraction (XRD) analysis showed single phase CCTO was obtained with no traces of crystalline phase related to both VT and PB glass. For the first addition; (1-*x*) CCTO - (*x*) VT [*x*, wt.%=0.03, 0.05, 0.10], the scanning electron microscopy (SEM) study showed that low melting point VT glass addition facilitated the grain growth of CCTO. As VT glass amount increased, dielectric constant decreased. The drop, though within reasonable values (~10^5), may be related to the presence of grain boundary glassy phase which itself has low dielectric constant. The decrease can also be interpreted by the decrease in grain conductivity of the samples. However, the dielectric loss of the composite has reduced after the addition of VT glass. The low melting VT glass aided in liquid phase sintering and improved the grain boundary resistance which resulted in decreasing the leakage currents. This has been confirmed by the increase in the resistivity of the grain boundary. Scanning electron microscopy (SEM) study for the second addition; (1-*y*) CCTO- (*y*) PB [*y*, wt.%=0, 0.01, 0.03, 0.05] also showed enhancement in grain diameter as PB glass content increased. However, the grain enhancement did not result in increase of dielectric constant. The drop in grain conductivity might contribute to the decrease of the dielectric constant. The grain boundary resistivity, ρ_{gb} of the samples also drops as PB glass was added. The existence of Pb^{2+} movable ions from PB glass led to the drop of ρ_{gb} which eventually led to the increase in dielectric loss. In the second part, pure CCTO samples prepared by mechanochemical process were characterized. The samples were synthesized by milling corresponding chemical precursors in a planetary ball mill. The samples were sintered at four different temperatures (T=950°C, 1000°C, 1050°C and 1100°C). From XRD analysis, it showed a considerable amount of CCTO phase but with small content of second phase elements. Electron micrographs showed that increase in sintering temperature significantly promoted grain growth. However, the dielectric constant did not show a normal behavior; instead it was higher for both samples with fine grain (sintered at 950°C and 1000°C). It was reported that finer grains and more reactive CCTO powder led to the increase in density which will eventually improve the dielectric constant. The result for dielectric loss showed that this parameter followed the exact pattern of the dielectric constant. The dielectric loss in CCTO sample sintered at 1000°C was the highest. This is a good correlation with grain boundary resistivity, ρ_{gb} where ρ_{gb} for this sample is the lowest.

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