High Dielectric Constant of pure CCTO Ceramic and Silicone Resin Polymer-CCTO Ceramic Composites

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Abstract-In this paper, an attempt has been made to study high dielectric constant of pure CaCu3Ti4O12 (CCTO) ceramics and silicone resin polymer-CCTO ceramic composites. For this, CCTO was prepared by the conventional oxide route. The pure sample of CCTO exhibits giant dielectric constant at low frequency within the studied temperature range. As frequency increases, dielectric constant drastically decreases and approaching a constant value at 1 MHz. The increment of dielectric constant as frequency decreases could possibly be due to interfacial polarization. There is a reduction of dielectric constant by two orders of magnitude just by addition of 10% silicone resin into the ceramic. This implies that in pure CCTO ceramic an interaction between grains is quite significant. This argument is confirmed because further addition of silicone resin does not bring down the dielectric constant as drastically. This indirectly confirms the Inter layer barrier capacitance (ILBC) model for high dielectric constant in CCTO ceramics.

Key Words- CCTO, Silicone Resin Polymer, Composites, Dielectric Constant

I. INTRODUCTION

In recent years, there has been an increasing interest on high dielectric constant flexible composites made up of a ferroelectric ceramic and a polymer for high density energy storage and capacitor applications [1]. However, the dielectric constant of such polymer based composites is rather low (about 50) because of the lower dielectric constant of the matrix [2-3]. For instance, in BaTiO3/epoxy composites, though BaTiO3 has relatively high dielectric constant (>1000), the effective dielectric constant of the composite was as low as 50, even when the highest possible volume fraction of ceramics was incorporated [4]. As the volume fraction of ceramics increased, the composite, unfortunately, lost its flexibility. A new generation of ultrahigh dielectric materials such as CaCu3Ti4O12 (CCTO) can be used in order to obtain composites with better performance [5]. A number of theoretical studies and experimental observations have attempted to elucidate the remarkable dielectric properties of CCTO perovskite-like material [6]. This material has demonstrated to have a dielectric constant as high as 50,000. In this work, an attempt has been made to study high dielectric constant of pure CCTO ceramics and Silicone resin polymer-CCTO ceramic composites.

II. EXPERIMENTAL PROCEDURE

CaCu3Ti4O12 (CCTO) polycrystalline ceramics were prepared by solid-state reaction method. For this, oxides of Ca, Cu and Ti were used as starting materials. Stoichiometric amounts were weighed and mixed by ball milling for 24 hours. The ball milled powder is then sieved using a 5 μm mesh. The mixed powders were poured in a crucible and then calcined in air at 1100°C for 10 hours. The calcined powders were firstly milled and then sieved. They were pressed into pellets in a cylinder shape of appropriate thickness. The pellets were sintered at 1250°C for 11 hours and then cooled to room temperature in the furnace. CCTO: silicone resin composite of 0-3 connectivity was prepared by mixing pre sintered powder of CCTO ceramic. The first set of samples were prepared in such a way that the material contains ninety percent (90%) by volume of CCTO ceramic and ten percent (10%) by volume of silicone resin. A paste of the ceramic and resin is formed, now one assumes that CCTO powder has been evenly got distributed into a matrix of silicone resin. 0.5 % by weight of the prepared paste, dibenzoyl peroxide was added, and the paste was again mixed so that the peroxide distributes evenly throughout the volume of the mixture. Dibenzoyl peroxide acts as the cross linking agent between the resin molecules. The paste is now injected into steel dies (moulds) and the mould loaded with the paste is then heated to 140oC. The temperature was held for 30 minutes after which the heater was put off and the mould was allowed to cool to room temperature and the mould is opened and the material inside the cavity is removed. We get a cured sample which is rubber like, since the silicone resin now acts like an elastic solid, with CCTO ceramic powder distributed within the matrix like filler. The procedure mentioned above was repeated for samples of composition 80% by volume of CCTO and 70% by volume of CCTO. The dielectric constant of the samples was determined using the HP 4192A LF Impedance Analyzer.

III. RESULTS AND DISCUSSION

Figure (1) represents the dielectric constant of pure CCTO ceramic and silicone resin polymer-CCTO ceramic composites measured at different temperatures.

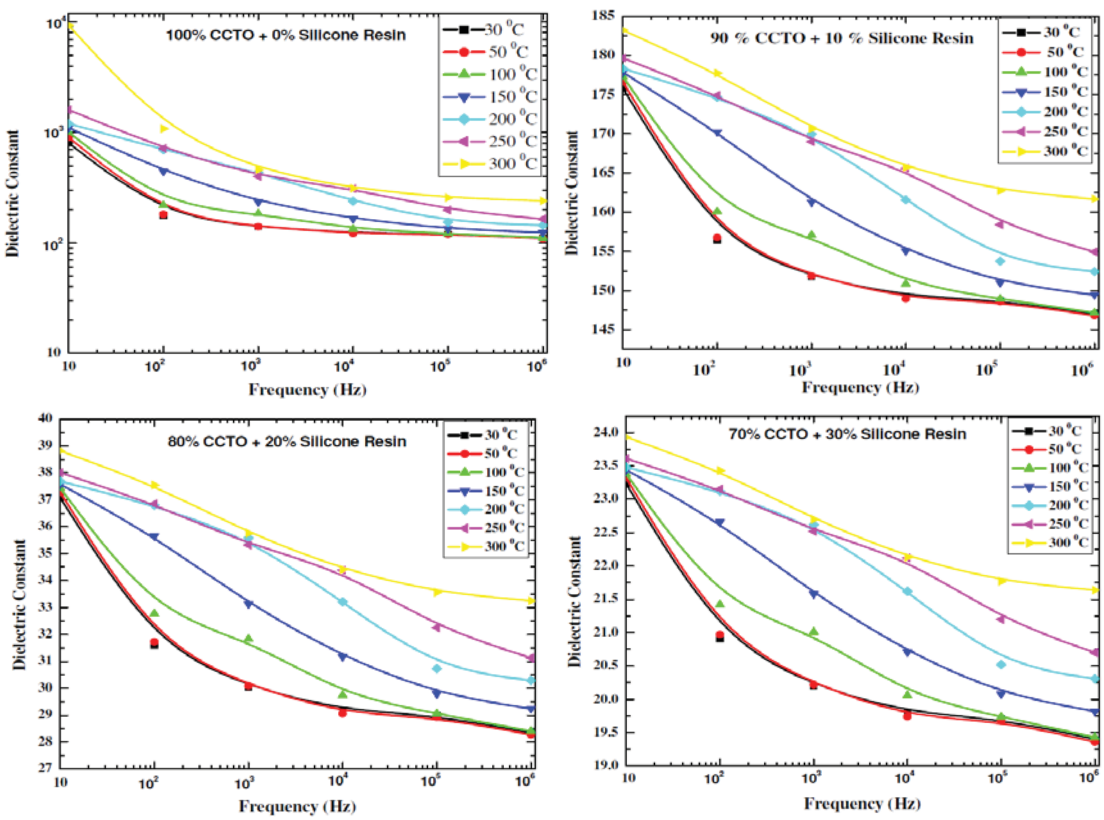

Fig. 1: Dielectric constant of pure CCTO ceramics and silicone resin polymer-CCTO ceramic composites

The pure sample of CCTO exhibits giant dielectric constant at low frequency within the studied temperature range. As frequency increases, dielectric constant drastically decreases and approaching a constant value at 1 MHz. The increment of dielectric constant as frequency decreases could possibly be due to interfacial polarization. The charge carriers may be blocked at the electrode interface under the influence of an electric field. It has been reported that CCTO ceramics consist of insulating grain boundaries and semiconducting grains. The charge carriers accumulated at the interface between semiconducting grains and insulating grain boundaries resulted in an increase in the dielectric constant. One can see that there is a reduction of dielectric constant by two orders of magnitude just by addition of 10% silicone resin into the ceramic. This implies that in pure CCTO ceramic an interaction between grains or the role of grain boundaries is quite significant.

IV. CONCLUSIONS

According to experimental results, CCTO with high dielectric constant was successfully prepared via conventional solid state reaction method. The dielectric constant decreases with an increase in frequency. There is a reduction of dielectric constant by two orders of magnitude just by addition of 10% silicone resin into the ceramic. Therefore in pure CCTO ceramic an interaction between grains or the role of grain boundaries is quite significant. This argument is confirmed because further addition of silicone resin does not bring down the dielectric constant as drastically. This in a way indirectly confirms the Inter layer barrier capacitance (ILBC) model for high dielectric constant in CCTO ceramics.

REFERENCES

[1] L. A. Ramajo, M.A. Ramírez, P.R. Bueno, M. M. Reboredo, M. S. Castro, "Dielectric Behaviour of $CaCu_3Ti_4O_{12}$-Epoxy Composites", Materials Research, Vol. 11, No. 1, pp.85-88, **2008.**

[2] Dias CJ, Das-Gupta DK, "Inorganic ceramic/polymer ferroelectric composite electrets", IEEE Trans. Dielectr. Electr. Insul., 5(3) pp. 706-734, **1996**.

[3] Bai Y, Cheng Z-Y, Bharti V, Xu HS, Zhang QM, "High-dielectric constant ceramic-powder polymer composites", Applied Physics Letter., 25 (76), pp. 3804-3806, **2000**.

[4] Kuo D-H, Chang C-C, Su T-Y, Wang W-K, Lin B-Y, "Dielectric behaviour of multi-doped $BaTiO_3$ epoxy composites", Journal European of Ceramic Society., 21(9), pp.1171-1177, **2001**.

[5] Bueno PR, Ramírez MA, Varela JA, Longo E, "Dielectric spectroscopy analysis of $CaCu_3Ti_4O_{12}$ polycrystalline systems",Applied Physics Letter, 89(19), pp.191117-3, **2006.**

[6] Sanjesh Babu, Anil Govindan, "Dielectric properties of $CaCu_3Ti_4O_{12}$(CCTO) prepared by modified solid state reaction method", International Review of Applied Engineering Research, Vol. 4, No. 3, pp. 275-280, **2014.**