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Dielectric Properties of Φ (CCTO)-(1-Φ) Silicone Resin Composites with 0-3 Connectivity

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ABSTRACT

CaCu3Ti4O12 –silicone resin (CCTO-silicone resin) composites with various CCTO volume fractions up to 0.9 were prepared. Relatively high dielectric constant (ε=151) and low loss (tan δ=0.35) of the composites with CCTO volume fraction at 0.9 were observed. Various theoretical models were employed to predict the dielectric constant of these composites, the dielectric constant obtained via Maxwell-Garnett model were in close agreement with the experimental data. Dielectric measurements were performed from 10Hz to 106Hz and 30 to 2000C. Dielectric constant of CCTO-silicone resin composites showed weak temperature dependence at the measuring temperature range. The results show that the CCTO-silicone resin composites prepared in this study could find some practical applications in the electronic industry.

Keywords: *Silicone Resin Polymer; CaCu3Ti4O12 Ceramic; Composites; Dielectric Constant.*

1.0 Introduction

Ceramic-polymer composites with high permittivity constant have attracted considerable attention recently due to their good fabrication process, low cost and potential applications ranging from embedded capacitors and high electric energy-storage devices. Requirements for real applications of embedded capacitors include high dielectric constant, low dielectric loss, low leakage current, high breakdown voltage and sufficient stability. To meet the stringent requirements for the composites, considerable attention has been devoted to the development of the candidate materials with high dielectric constant [1].

In previous studies, BaTiO3-polymer composites for embedded capacitors were introduced and fully characterized [2-3]. However, the dielectric constant of such polymer based composites is rather low (about 50) because of the lower dielectric constant of the matrix [4-5].

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. As the volume fraction of ceramics increased, the composite, unfortunately, lost its flexibility. Recently, some perovskite type materials, non-ferroelectric, such as CaCu3Ti4O12 (CCTO) with giant dielectric constant up to 105 at room temperature has been extensively investigated. The unusually high dielectric constant of CCTO as suggested is attributed to an internal barrier layer capacitor (IBLC) model, which consists of semiconducting grains and insulating grain boundaries.

The unique dielectric behaviour of CCTO gives an interesting on making up of CCTO ceramic-polymer flexible particulate composites for high density energy storage and capacitor applications [6]. In this study, an attempt has been made to study dielectric properties of CaCu3Ti4O12 –silicone resin composites as a function of frequency, temperature and filler volume fraction.

2.0 Experimental

CCTO: silicone resin composites of 0-3 connectivity were prepared by mixing pre sintered powder of CCTO ceramic. The first set of samples were prepared in such a way that the material contains 90 percent (90%) by volume of CCTO ceramic and 10 percent (10%) by volume of silicone resin. A paste of the ceramic and resin is formed; one assumes that the CCTO powder has

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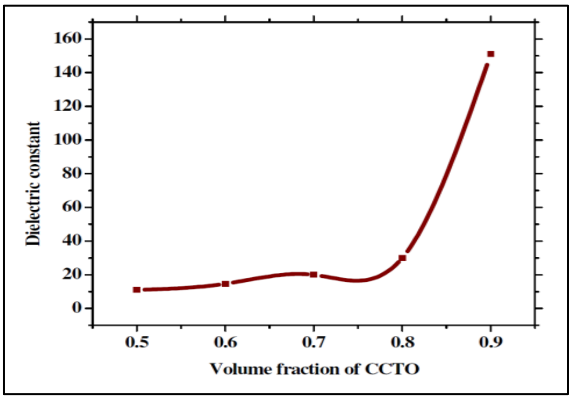
been evenly distributed into a matrix of silicone resin. To 0.5 % by weight of the prepared paste, dibenzoyl peroxide was added and the paste was again mixed so that the peroxide distributes evenly throughout the volume of the mixture. Dibenzoyl peroxide acts as a cross linking agent between the resin molecules. The paste is now injected into a steel die (mould) and the mould loaded with the paste is then heated to 140oC. The temperature was held for 30 min after which the heater was turned off and the mould was allowed to cool to room temperature, opened and the material inside the cavity is removed. We obtained 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. This is now a 0:3 ceramic polymer composite sample. The resulting sample yields a thickness of not more than 1.5 mm. The procedure mentioned above was repeated for samples of compositions 80%, 70%, 60% and 50% by volume of CCTO. The dielectric properties of the sample were determined using the HP 4192A LF Impedance Analyzer.

3.0 Results and Discussion

Figure 1 illustrates the dielectric constant (ε) and the loss tangent (tan δ) of the composites measured at 1 kHz and room temperature. Figure 2 and 3 show dielectric constant as a function of frequency and temperature, respectively. As it can be expected, dielectric constant rose as the ceramic volume fraction increases.

Fig 1: Dependence of Dielectric Constant and Loss Tangent of the CCTO-Silicone Resin Composites on the Volume Fraction of CCTO at 1 khz and Room Temperature.



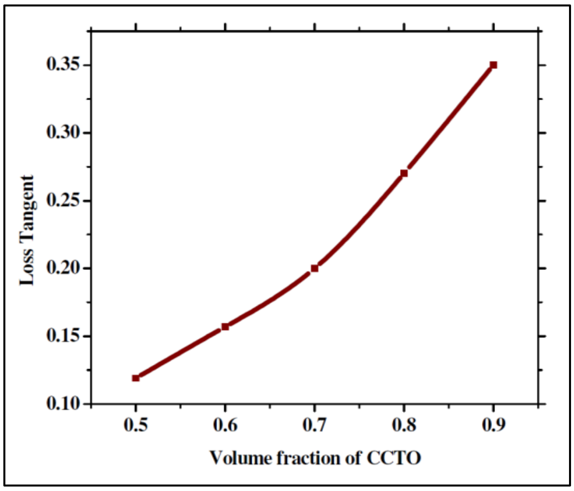


Fig 2: Dielectric Constant Vs. Frequency for Composites with Different CCTO Volume Fraction at 30°C Temperature.

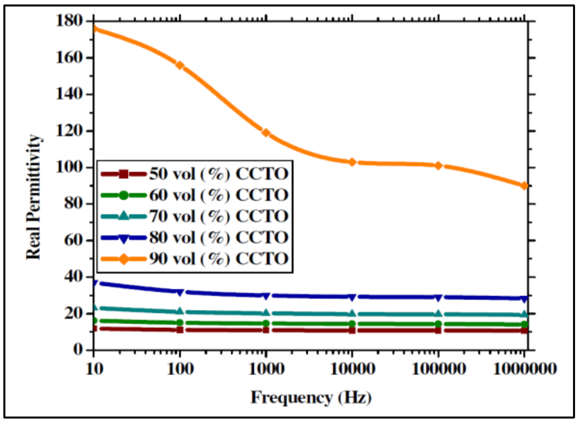
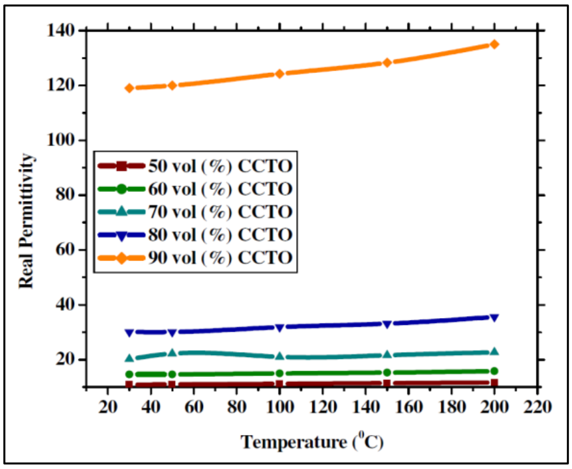


Fig 3: Dielectric Constant Vs. Temperature for Composites with Different CCTO Volume Fraction at Frequency103 Hz.



A number of numerical relations have been put forward by researchers to predict the dielectric constant of the composite. In this study, the following equations have been used to calculate the effective dielectric constant of the CCTO-silicone resin composites.

$\log \varepsilon c = Vm \log \varepsilon m + (0.7)Vf \log \varepsilon f$ -------------------- (1)

$\varepsilon c = \varepsilon_m [1 + 3Vf\{(\varepsilon f-\varepsilon_m)/(\varepsilon_f+2\varepsilon_m)\}]$ ----------------------(2)

$\varepsilon_c = \varepsilon_f [1 + \{3V_f (\varepsilon_m-\varepsilon_f)\}/\{2\varepsilon_f + \varepsilon_m - V_f (\varepsilon_m-\varepsilon_f)\}]$ ---- (3)

Equations (1)-(3) are the expressions of Lichtenecker, Clausius Mossotti and Maxwell-Garnett models respectively, where εc, εf and εm are the dielectric constants of the composites, CCTO ceramic powder and silicone resin respectively and Vf and Vm are the volume fractions of the CCTO ceramic and silicone resin respectively.

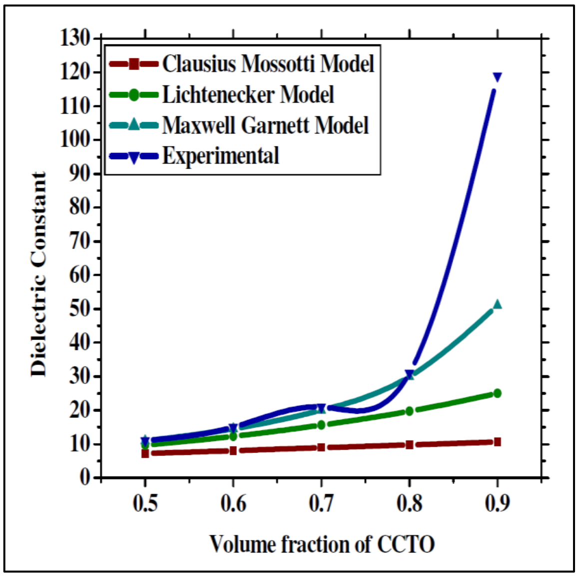
The dielectric constants of silicone resin and CCTO ceramic measured at 1 kHz are 3.0 and 139, respectively.

Figure 4 depicts the comparisons of dielectric constants of the CCTO-silicone resin composites with the values predicted by the above equations at 1 kHz. From this figure, it is clear that the Maxwell-Garnett model is the best fit of the experimental values for the CCTO-silicone resin composites.

Maxwell-Garnett model predicts the dielectric constant of the composites is nearly equal to the experimental values up to 0.8 CCTO volume fractions. However, as volume fraction of CCTO increases beyond 0.8, a deviation from the predicted value of dielectric constant is observed in CCTO-silicone resin composites. It was found that both ε and tan δ increased gradually with the increase in CCTO content. For the composite with CCTO volume fraction of 0.9, the ε and tan δ values at 1 kHz were 151 and 0.35, respectively.

In comparing with pure silicone resin (the experimental values of pure silicone resin, ε = 3.0 and tan δ = 0.03), the ε value of the composite was improved by 50 times, while the tan δ value increased by nearly 12 times. High ε and low tan δ of the CCTO-silicone resin composite made it attractive for practical applications.

Fig 5: Comparison of Experimental and Theoretical Dielectric Constants of CCTO-Silicone Resin Composites at 1 kHz



4.0 Conclusions

A ceramic-polymer 0-3 composite fabricated by using CCTO ceramic particles as filler and silicone resin as polymer matrix is introduced in the present study. Dielectric measurements were performed from 10Hz to 106Hz and 30 to 2000C. Dielectric constant of CCTO-silicone resin composites showed weak temperature dependence at the measuring temperature range. The dielectric constant (ε) and the loss tangent (tan δ) of the composites measured at 1 kHz and room temperature were increased gradually with the increase in CCTO content.

For the composite with CCTO volume fraction of 0.9, the ε and tan δ at 1 kHz was 151 and 0.35, respectively. In comparing with pure silicone resin (the experimental values of pure silicone resin, ε = 3.0 and tan δ = 0.03), the ε of the composite was improved by 50 times, while the tan δ increased nearly 12 times.

The dielectric constant of the composite was simulated based on three different models. The values obtained by the Maxwell-Garnett model are in close agreement with the experimental values up to 0.8 CCTO volume fractions

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References

[1.] Yi Hu, Chia-Hao Yu, Chun-Chi Lin, High Permittivity of CaCu3Ti4O12 /polymer Nano-Composites for High Energy Storage Application, Proceedings of the World Congress on New Technologies (New Tech 2015) Barcelona, Spain – July 15 - 17, Paper No. 337, **2015.**

[2.] Y Kobayashi, T Tanase, T Tabata, T Miwa, M Konno, Fabrication and dielectric properties of the BaTio3-polymer nano-composite thin films, Journal of the European Ceramic Society, 28(1), 2008, 117-122**.**

[3.] ZM Dang, YQ Lin, HP Xu, CY Shi, ST Li, J Bai, Fabrication and Dielectric Characterization of Advanced BaTiO3/Polyimide Nanocomposite Films with High Thermal Stability, Advanced Functional Materials, 18(10), 1477-1596**.**

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

[5.] Y. Bai, Z.Y. Cheng, V. Bharti, H.S. Xu, Q.M. Zhang, "High-dielectric constant ceramic-powder polymer composites", Applied Physics Letter., 25 (76), 2000, 3804-3806.

[6.] Amaral F, Rubinger C, Henry F, Costa LC, Valente MA. Dielectric Properties of Polystyrene-CCTO Composite, J. Non-Cryst. Sol., 35, 2008, 5321-5322.