International Review of Applied Engineering Research.
ISSN 2248-9967 Volume 4, Number 3 (2014), pp. 275-280

http://www.ripublication.com/iraer.htm

Dielectric Properties of $CaCu_3Ti_4O_{12}$ (CCTO) Prepared by Modified Solid State Reaction Method

Sanjesh Babu[1] and Anil Govindan[2]

[1]*Department of Applied Sciences (Physics), IET, Mangalayatan University, Aligarh (U.P.) India.*
[2]*Department of Physics, M.M.H. (P.G.) College, Ghaziabad (U.P.) India.*

Abstract

In this paper, an attempt has been made to study dielectric properties of $CaCu_3Ti_4O_{12}$ (CCTO) ceramics. For this, $CaCu_3Ti_4O_{12}$ was prepared by the modified solid state reaction method. The dielectric constant (ε') and dielectric loss (tan δ = ε''/ε') were measured within the frequency range from 10^2 Hz to 10^6 Hz and the temperature ranging from 30°C to 300°C using HP 4192A LF Impedance Analyzer. The results showed that the dielectric constant and dielectric loss of the sample are frequency and temperature dependent. Dielectric constant and dielectric loss increases with decreasing frequency and increasing temperature due to interfacial polarization.

1. Introduction

In recent years, the perovskite related compound $CaCu_3Ti_4O_{12}$ (CCTO) has attracted considerable interest due to its extremely high dielectric constant (larger than 10^4). The dielectric constant shows small temperature dependence near room temperature, so it has potential applications in the field of energy-storage devices [1]. In $CaCu_3Ti_4O_{12}$, Ca and Cu ions reside at the A-sites, while Ti cations occupy the B-site. $CaCu_3Ti_4O_{12}$ shows great promise in dielectric applications owing to it's essentially temperature and frequency independent dielectric constants ranging from about 10,000 for polycrystalline powders to 100,000 for single crystals [2]. $CaCu_3Ti_4O_{12}$ does not undergo any structural change over the large temperature range 100 to 600K although its dielectric constant abruptly decreases to less than 100 below 100K, showing a Debye-like relaxation [3]. In this paper, an attempt has been made to study the dielectric properties of $CaCu_3Ti_4O_{12}$ by modified solid state reaction method.

2. Experimental Procedure

$CaCu_3Ti_4O_{12}$ (CCTO) polycrystalline ceramics were prepared by solid-state reaction. CaO (99.95%), CuO (99.9%), and TiO_2 (99.7%) 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 sieved powder has been compacted lightly in a crucible made of high grade alumina. Since the particle size of the constituent oxides plays a major part in the physical properties of the ceramic, therefore sieved powder was again sieved before compaction and sintering. 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. The crystal structures of the sample were examined by X-ray diffraction (XRD). The dielectric properties of the sample were determined using the HP 4192A LF Impedance Analyzer in the frequency range from 10^2 Hz to 10^6 Hz at 30^0C and 50°C – 300°C in the intervals of 50°C.

3. Results and Discussion

Table 1 and 2 shows the values of the dielectric constant and dielectric loss for $CaCu_3Ti_4O_{12}$ measured at 30^0C to 300^0C. From the table, it is clear that the values of both dielectric constant and dielectric loss increase as temperature increases, however as frequency increases, the values of dielectric constant and dielectric loss decrease.

Table 1: Dielectric constant (ε') for $CaCu_3Ti_4O_{12}$ measured at different temperatures.

Frequency (Hz)	Dielectric Constant (ε') of $CaCu_3Ti_4O_{12}$						
	Temperature (^{0}C)						
	30^0C	50^0C	100^0C	150^0C	200^0C	250^0C	300^0C
100	177	181	218	443	695	715	1080
1000	139	140	184	234	432	399	459
10000	123	121	133	165	238	309	311
100000	120	119	121	134	154	198	256
1000000	110	109	111	124	144	164	240

Fig. 1 and 2 represent the dielectric constant and dielectric loss of $CaCu_3Ti_4O_{12}$ ceramics measured at different temperatures. The sample exhibits giant dielectric constant (1080) at low frequency for 300°C. As frequency increase, dielectric constant drastically decreases and approaching a constant value at 10^6Hz. 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 $CaCu_3Ti_4O_{12}$ 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. This is the famous Maxwell-Wagner effect. Dielectric loss factor of the sample exhibits dc conduction losses. It shows the dielectric loss of $CaCu_3Ti_4O_{12}$ drastically decreases with increasing frequency. The dielectric loss increases with temperature from 30°C to 300°C. The frequency and temperature effect on the dielectric loss illustrates the interfacial polarization of the grain boundaries within the sample. The dielectric constants of $CaCu_3Ti_4O_{12}$ at selected frequencies such as 10^2 Hz, 10^3 Hz, 10^4 Hz, 10^5 Hz and 10^6 Hz with respect to temperature (30°C–300°C) are given in Figure 3. It exhibits the temperature dependence of $CaCu_3Ti_4O_{12}$. The dielectric constant of $CaCu_3Ti_4O_{12}$ increases with increasing temperature. The increment of dielectric constant is low from 30°C–250°C especially during higher frequencies. This means that there exists mechanical limitation where lack of interfacial polarization occurs at higher frequencies. The large variation from 250^0C to 300^0C at low frequencies is due to the increase in interfacial polarization. Figure 4 shows that the dielectric loss drastically increases as the temperature increases.

Table 2: Dielectric loss (tan δ = ε"/ε') for $CaCu_3Ti_4O_{12}$ measured at different temperatures.

Frequency (Hz)	Dielectric Loss (tan δ = ε''/ε') of $CaCu_3Ti_4O_{12}$						
	Temperature (^{0}C)						
	30^0C	50^0C	100^0C	150^0C	200^0C	250^0C	300^0C
100	2.457	2.240	2.200	1.650	6.800	45.450	166.666
1000	0.474	0.500	0.430	0.641	1.440	8.470	38.120
10000	0.081	0.086	0.203	0.272	0.640	1.290	5.850
100000	0.011	0.010	0.0325	0.141	0.240	0.480	0.929
1000000	0.0036	0.0027	0.0063	0.044	0.190	0.230	0.400

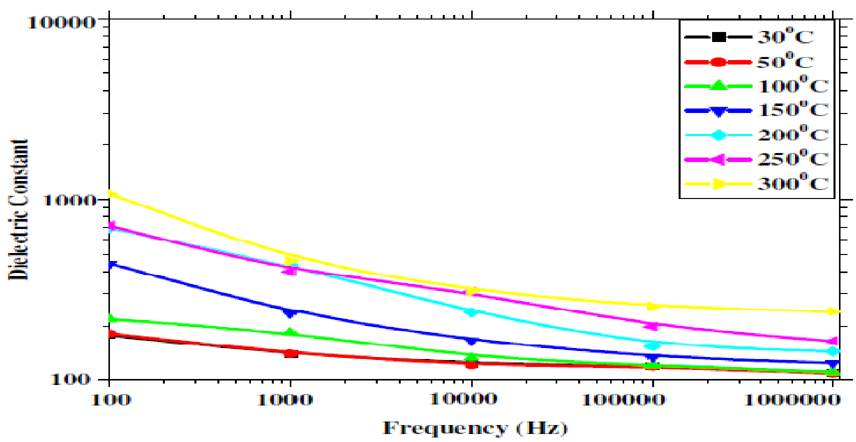

Fig. 1: Dielectric constant of $CaCu_3Ti_4O_{12}$ measured at different temperatures.

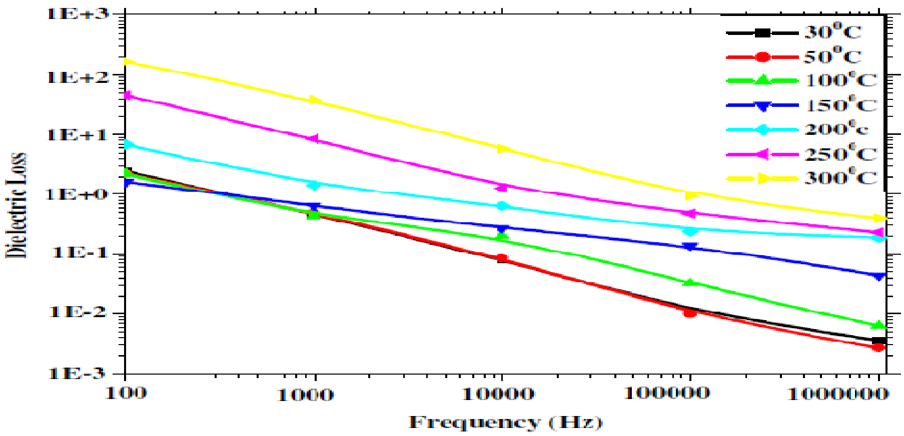

Fig. 2: Dielectric Loss of CaCu3Ti4O12 measured at different temperatures.

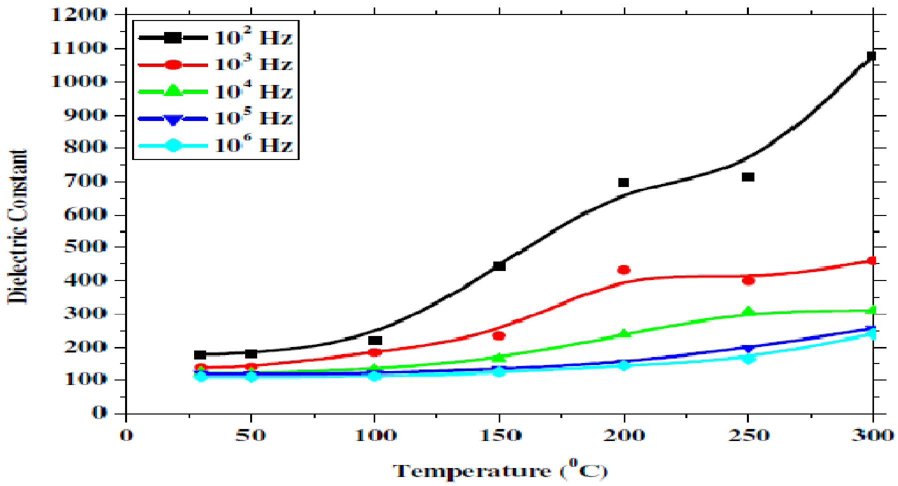

Fig. 3: Dielectric constant of CaCu3Ti4O12 with respect to temperature at selected frequencies.

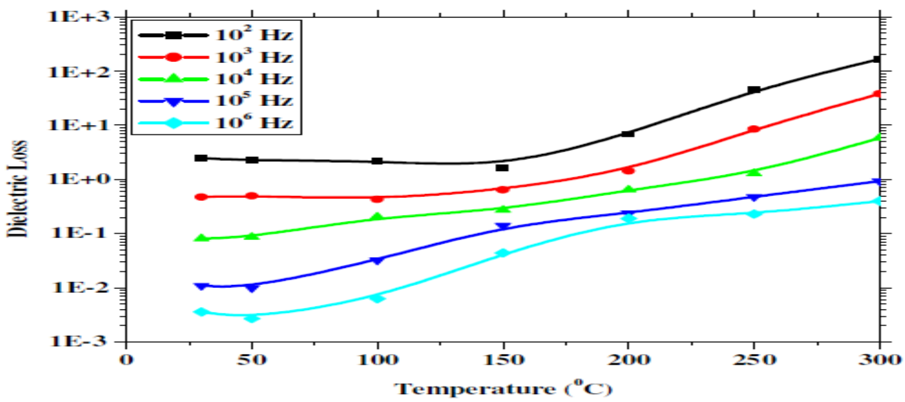

Fig. 4: Dielectric Loss of CaCu3Ti4O12 with respect to temperature at selected frequencies.

4. Conclusions

According to experimental results, $CaCu_3Ti_4O_{12}$ (CCTO) with high dielectric constant was successfully prepared via conventional solid state reaction method. The dielectric properties of $CaCu_3Ti_4O_{12}$ ceramics are very sensitive to processing. The dielectric constant and dielectric loss decrease with an increase in frequency. The dielectric constant and dielectric loss increase with an increase in temperature.

5. Acknowledgments

The authors are very much thankful to the authorities of National Physical Laboratory (NPL), New Delhi for providing necessary facilities.

References

[1] Xuefeng Ruan, Yue Zhang, Zhi Yang, Rui Xiong, Jing Shi (2013), “Electrical conduction and dielectric properties of the $CaCu_3Ti_4O_{12}$ ceramics synthesized by SPS”, Advances in Microelectronic Engineering AIME, 1, pp 24-29.

[2] P. Barber, S. Balasubramanian, Y. Anguchamy, S. Gong, A. Wibowo, H. Gao, H.J. Ploehn and H. Loye (2009), “Polymer composites and nanocomposites dielectric materials for pulse power energy storage”, Materials, 2, pp 1697-1733.

[3] C. Masingboon, P. Thongbai, S. Maensiri (2006), “Giant dielectric response in perovskite-derivative $CaCu_3Ti_4O_{12}$ prepared by polymerized complex method”, Advances in Science and Technology, Vol. 45, pp 2345-2350.