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DIELECTRIC PROPERTIES OF STRONTIUM TITANATE FILLED MULLITE COMPOSITES AT 10 Hz - 1 MHz

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ABSTRACT

This research was designed to form better dielectric composite material using one steady state dielectric (mullite) with a good dielectric material (strontium titanate). Distinct dielectric composites were successfully produced using locally sourced kaolinite clay. The samples were made using mullite as the base matrix and strontium titanate (ST) added at varying ratios. Strontium titanate was synthesized via solid-state reaction using strontium carbonate and rutile titanium (IV) oxide sintered at 1300°C. Local white kaolinite was used to fuse the strontium titanate material at varying weight ratios. The powders were dry-mixed and made into pellets for calcination at 1000°C. XRD and SEM characterizations were made. The dielectric measurements were carried out using the HP 4192A LF Impedance Analyzer dielectric setup in the frequency range of 10 Hz to 1 MHz. Three samples were prepared, namely ST 60%, ST 70% and ST 80%. The dielectric measurements were carried out in a controlled LT furnace at 30°C - 400°C. Measurements showed distinct varying interfacial encapsulation and dipolar relaxation for all composite samples.

INTRODUCTION

Kaolinite and perovskite are two distinct naturally occurring minerals, which have profound usage in today's industries. The physical and chemical properties of kaolinite have determined its use as an industrial mineral. Kaolinite with its relatively low viscosity at high solid concentrations has made it a substantial raw material in paper and paint processing besides the traditional ceramic refractory uses [1]. It is also an excellent insulator with stable dielectric response over a thermal operating range. Perovskite, especially barium titanate and strontium titanate, however are dielectric and piezoelectric materials that are developed specifically for the electronic and insulation purposes. It has high quality dielectric response and simple production. Recent research papers have focused on the development of various detecting sensors using perovskite material [2]. So far, the bulk of scientific knowledge has not explored the potential of ceramic matrix composite (CMCs) in relation to dielectric studies despite the insulator behavior for some of its configurations. The concept of merging ceramic materials is applied in this research where kaolinite and perovskite (barium titanate and strontium titanate in particular) are fused at varying ratios. These CMCs are subjected to

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frequency and temperature dependent dielectric measurements at low frequencies. The measured signal showed that the blended materials introduced new compounds with varying mix ratios and these have distinct behaviors in the dielectric research.

METHODOLOGY

The main focus of this research is to report on the preliminary dielectric results of the effect of ST filler in kaolinite matrix. The raw materials used are strontium carbonate, titanium (IV) dioxide and Kaolinite. The ST materials are mixed thoroughly by solid state ionic mixing for oxide powders [3]. Stoichometric amounts of $SrCO_3$ and TiO_2 are mixed thoroughly in 1:1 ratio for 24 hours using a porcelain-mixing chamber. The resulting dry-mix are loaded into an alumina boat and pre-sintered at 1200°C in a carbolite open furnace for 20 hours with one intermittent grinding. ST powders were then mixed with the kaolinite matrix at varying weight ratios before pre-sintering at 800°C. The powders were then mixed with polyvinyl alcohol (PVA) and pressed into discs at a pressure of 5 kilopascals before being subjected to final sintering stage at 1000°C [4]. This sintering temperature was chosen to form mullite structure and limit ST-mullite fusing. XRD measurements were done on the samples to identify the composite compounds using PAnalytical instrument. For the dielectric measurements, the prepared samples were placed between two gold-coated metal plates test bench connected to a Hewlett-Packard HP 4192A Impedance Analyzer in the frequency range of 10 Hz to 1 MHz. The sample measurements were made at controlled temperature increments using aprogrammable induction furnace.

RESULTS AND DISCUSSION

In this paper, we place importance on the XRD and the dielectric data. The former for investigating the resultant compounds and the latter is used to characterize the dielectric permittivity response at low frequency.

The composites derived from the merging of ST and kaolinite at 1000°C yielded a series of transitions. The simple ST-kaolinite hybrid structure has high intensity counts of strontium titanate peaks. The 80%ST sample has high peaks at 2θ = 32.4°, 39.9, 46.4° and 57.8°. This series correspond to 01-079-0175 of the ICDD database that confirms ST structures [5]. In the 70%ST sample, fore-mentioned peaks have decreased intensity, thus indicating lower ST level. There is some peak broadening of a tertiary compound at point 22.5°. Since the spectra indicate minimal tertiary compounds form through assimilation of the matrix-filler, thus ST does not react strongly with the kaolinite-mullite liquid state during sintering. Comparison of the major ST at 2θ = 32.4°, 39.9, 46.4° and 57.8° between the three spectra lines showed that there is a spectra shift of 0.1°. The shift might be caused by dense amorphous silica oxide layers formed during the mullite composition. The indication of assimilation is only observed in 60%ST where there are some low peaks matched by

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strontium feldspar. The decomposition of kaolinite happened at slow steady temperature rise about 410°C as reported [6]. The resulting compounds are alumina, silica and water. This composition destroys the original kaolinite layer structure irreversibly and with temperature exceeding 1000°C forces the alumina and silica compounds to form mullite [7]. The low peak at 22.5° for the strontium feldspar formation might be due to insufficient energy provided during the sintering process. Since the Sr^{2+} ions are not highly electro-positive, the probability of tertiary chemical reaction was low without sufficient energy mass.

$2Al_2Si_2O_5(OH)_4 \rightarrow 0.66(Al_2O_3 \cdot SiO_2) + 4H_2O + 2.68\ SiO_2$ [8]
Kaolinite Mullite, Water and Silica,

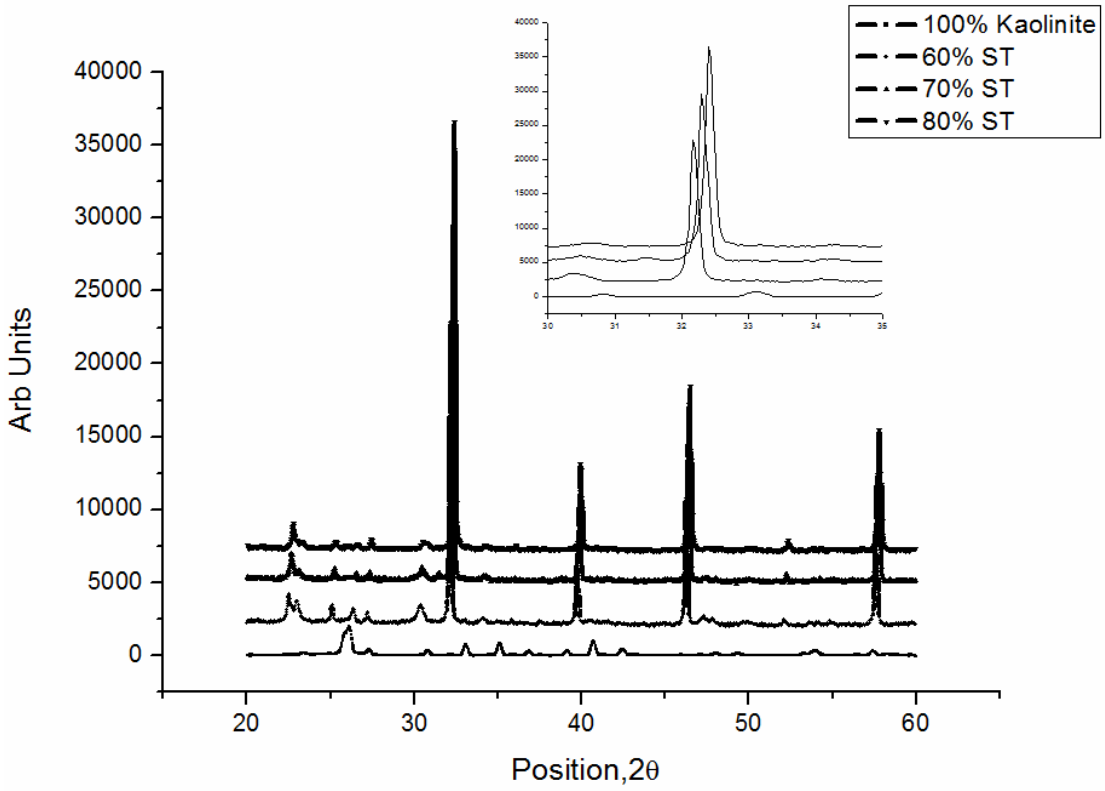

Figure 1: XRD data for 60% ST, 70% ST and 80% ST. The spectra show pattern shifts that depend on the ST amounts. Inset: Magnified image of the 30°-35° spectra showing the gradual XRD shifts of 0.15 *2θ* increments.

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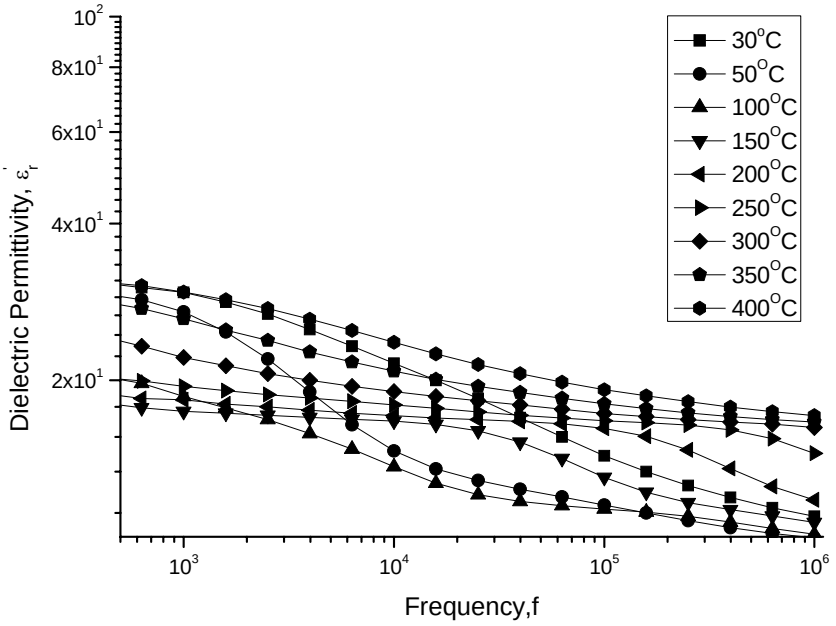

Figure 2(a): Dielectric Permittivity for 60%ST

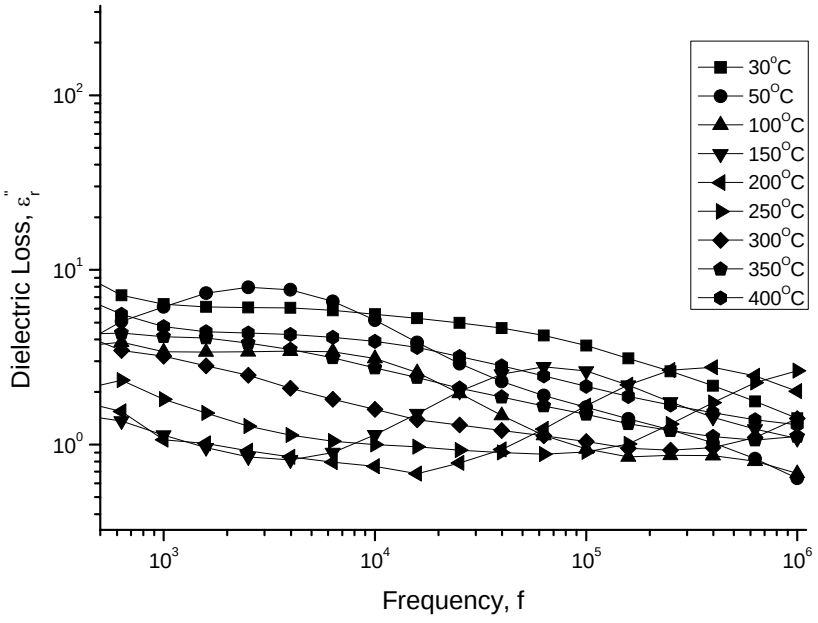

Figure 2(b): Dielectric Loss for 60%ST

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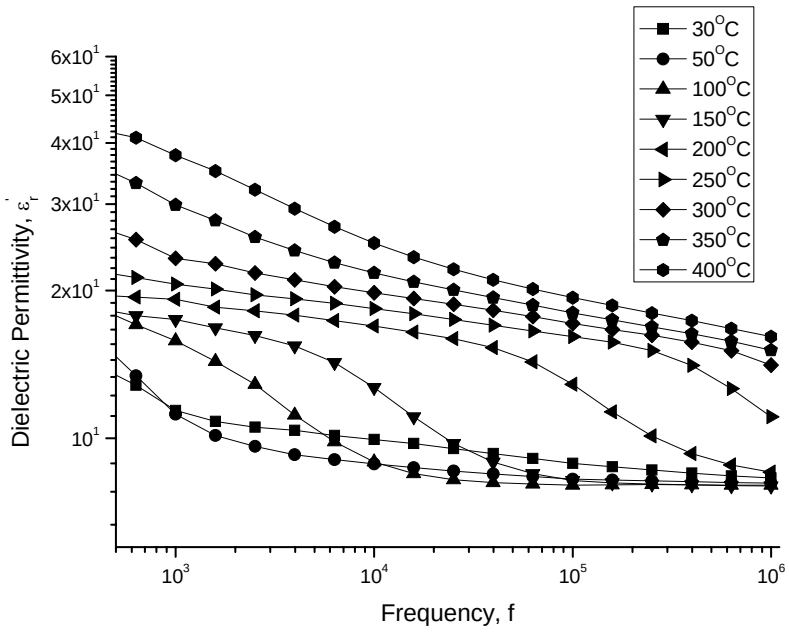

Figure 2(e): Dielectric Permittivity for 80%ST

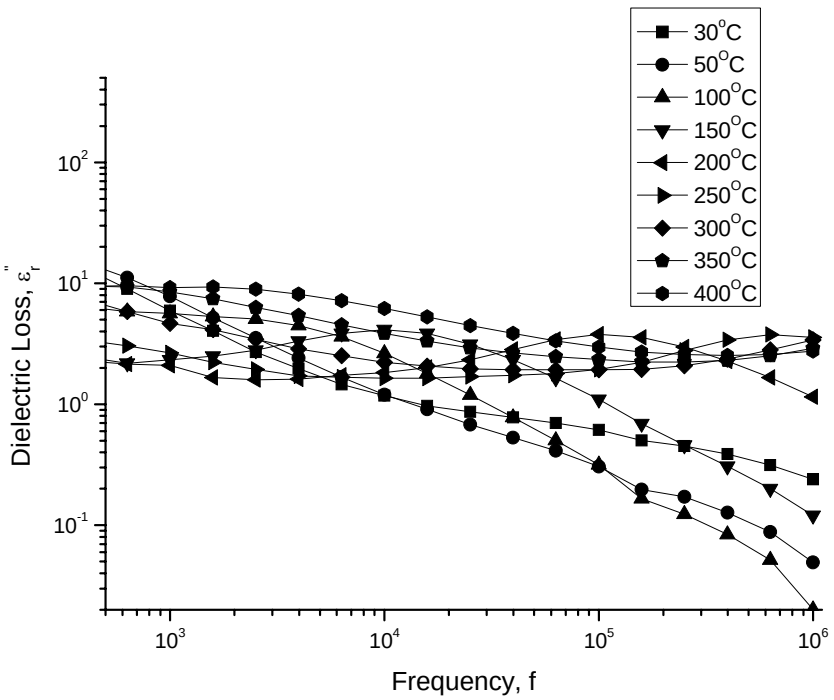

Figure 2(f): Dielectric Loss for 80%ST

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Figures 2(a)-(f) show the dielectric permittivity and losses for 60%ST, 70%ST and 80%ST samples. Close examination of the permittivity figures particularly the 80%ST exhibits two distinct dielectric permittivity regions where 30°C to 200°C represents the α region and 250°C to 400°C represents the β region. In Figure 2(a), the 60%ST has a low α region value of 10 units, and 20 units for the β region at 1 MHz response. Despite the relatively high initial dielectric permittivity at low frequency, the dielectric permittivity exhibits frequency independent response (γ response) at 150°C to 250°C. This actually contradicts with the other two samples. The 70%ST exhibits almost similar properties except that the α region tend to focus onto a singularity at point 11 units. The higher ST concentration disperses the dielectric permittivity response spectra and thus eliminating the γ responses shown in Figure 2(a). In the 80%ST, the high ST concentration exhibit higher initial dielectric permittivity and clearer dielectric response spread. In the 70%ST and 80%ST samples, the α and β are distinct and thus inferring the existence of two dominant responses. The dielectric losses however indicate that all three samples have considerable high losses ($>10^0$) and the higher ST insertion exhibits some low loss response at certain temperatures. The ST has higher dielectric permittivity around 270 [9] whereas the mullite structure exhibits permittivity ranging from 6- 11 [10]. Both the dielectric permittivity and loss responses indicate possible interfacial polarization between the amorphous silica and the ST-mullite particles. Interfacial polarization that occurs could increase the dielectric permittivity between the grains or particles, but the same polarization could induce resonating heat during higher frequency oscillations. The heat energy generated via resonance and vibrational effects will indirectly impede the permittivity. Thus the experimental data proves that interfacial polarization represents dominant impeding factor in building composite dielectric material.

CONCLUSION

CMCs were successfully produced using two ceramic raw materials of distinct dielectric permittivity. Dielectric properties of the composite material do not conform to linear composite mix equation. Instead, the dielectric permittivity is bounded by the interfacial polarization between the matrix-filler boundaries. In the 60%ST sample, there is an occurrence of flat permittivity across the temperature region, thus suggesting both frequency and temperature independent response. Therefore the 60%ST represents the better dielectric composite despite the lower dielectric permittivity.

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REFERENCES

[1]. Cornelius Klein and Barbara Dutnow, 2008, The 23rd Ed.of the Manual of Mineral Science, 23rd Ed., John Wiley and Sons, New Jersey, 521- 523.

[2]. X.Y. Fang, O.K. Tan, Q. Wei, M.W. Yao, and S.C. Tjin, 2006, *Sensors and Actuators B*, No. **119**, 78–83

[3]. Janusz Nowotny, 1992, Electronic Ceramic Materials, Trans Tech Publications, Switzerland, 150-151.

[4]. Rex W. Grimshaw, 1971,The Chemistry and Physics of Clay and Allied Ceramic Material, 4th Ed., Wiley Interscience, London, 723-727.

[5]. S.A. Howard, J.K. Yau and H.U. Anderson, 1989, *J. Appl. Phys.*, **65**, 1492.(Under PAnalytical Database)

[6]. W. D. Kingery, H. K. Bowen and D. R. Uhlmann, 1960, Introduction to Ceramics, 2nd Ed. John Wiley and Sons, New York, 419- 420.

[7]. Rex W. Grimshaw, 1971, The Chemistry and Physics of Clay and Allied Ceramic Material, 4th Ed., Wiley Interscience, London, 446-448.

[8]. H. Schneider and S. Komarneni (editors), 2005, Mullite, Wiley-VCH, Weinheim, 251-253.

[9]. V.S.Nisa, S.Rajesh, K.P.Murali, V.Priyadarsini, S.N.Potty and R.Ratheesh, 2008, Preparation, Characterization and Dielectric Properties of Temperature Stable $SrTiO_3$/PEEK composites for microwave substrate applications, Composite Science and Technology, **68**, 106-112.

[10]. S. Somiya and Y.Hirata, 1991, Mullite Powder Technology and Application in Japan, Amer. Ceram. Soc. Bull., **70**, 1624-1632.

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