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DIELECTRIC VARIATIONS OF BARIUM TITANATE ADDITIONS ON MULLITE- KAOLINITE SAMPLE

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ABSTRACT

This research was designed to form better dielectric composite material using one steady state dielectric with a good dielectric material. Distinct dielectric composites were successfully produced using locally sourced kaolinite clay. The samples were made using kaolinite as the base matrix and Barium Titanate (BT) added at varying ratios. Barium Titanate was synthesized via solid-state reaction using Barium Carbonate and rutile Titanium (IV) Oxide sintered at 1300°C. Local white kaolinite was used as the matrix at varying weight ratios. The powders were dry-mixed and made into pellets for calcination at 1000°C. The dielectric measurements were carried out using HP 4291B Impedance Analyzer dielectric setup. Three samples were prepared, namely 50%BT, 60%BT, and 70%BT. The dielectric measurements were carried out in an LT furnace at temperatures 30°C - 400°C with frequencies ranging from 10 Hz to 1 MHz. Measurements showed varying ionic relaxation for all samples.

INTRODUCTION

Electronic technology needs has greatly recently pushed developmental requirements for better materials for circuit operations. Besides higher quality and better response, there is a need for stable materials. This improved the material durability for the rapid frequency oscillation response. 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, it is also an excellent insulator with stable dielectric response over a thermal operating range [1]. Perovskite, especially Barium Titanate, however are dielectric and piezoelectric material that have been developed specifically for the electronic and insulation purposes. It has high quality dielectric response and simple to produce. 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 insulating behavior for some of its configurations. The concept of merging ceramic materials is used in this research where kaolinite and perovskite (Barium Titanate in particular) are fused in varying ratios. These CMCs are subjected to frequency and temperature dependent dielectric measurements at low frequencies. The measured signal shows that the blended materials introduced new compounds in

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varying mix ratios and these have distinct dielectric behaviors.

METHODOLOGY

The main focus of this research is to report on the preliminary dielectric results of the effect of Barium Titanate (BT) filler in kaolinite matrix. The raw materials used were Barium Carbonate, Titanium (IV) Oxide and Kaolinite. The BT materials were mixed thoroughly by solid state ionic mixing for oxide powders [2]. Stoichometric amounts of $BaCO_3$ and TiO_2 were mixed thoroughly in 1:1 ratio for 24 hours using a porcelain-mixing chamber. The resulting dry-mix were loaded into an alumina boat and pre-sintered at 1200°C in a Carbolite open furnace for 20 hours with one intermittent grinding. Afterwards, BT powders were mixed together in varying weight ratios to the kaolinite matrix before pre-sintered at 800°C. The sample 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. [1] This sintering temperature was chosen to form mullite structure and limit BT-mullite fusing. XRD measurements were done on the samples to identify the composite composition using PAanalytical instrument. For the dielectric measurements, the prepared samples were placed between two gold-coated metal plates test bench connected to an HP 4192B Impedance Analyzer in the frequency range of 10 Hz to 1 MHz.

RESULTS AND DISCUSSIONS

In this paper, we focused on the XRD and the dielectric data. The former for investigating the compounds and latter is used to characterize the dielectric permittivity.

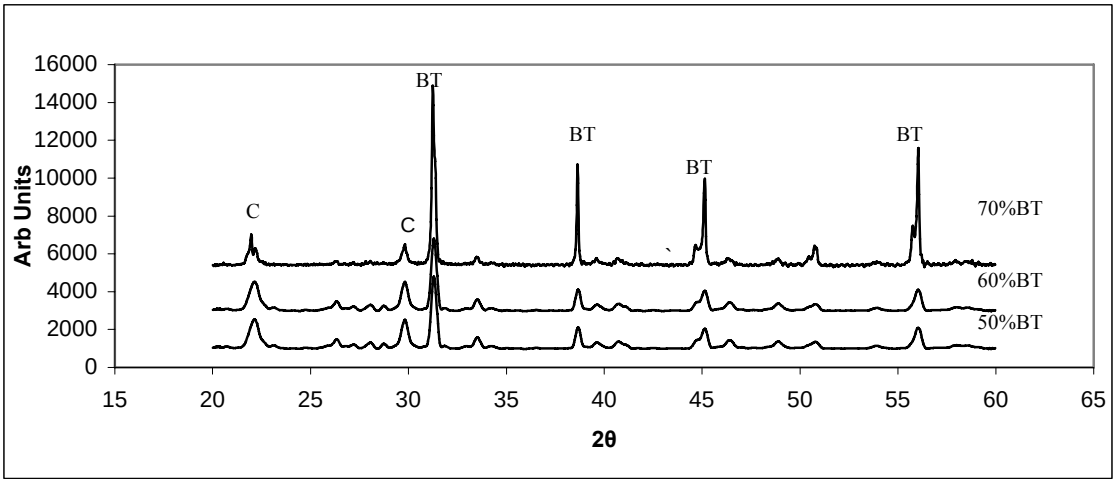

Figure 1: XRD data for the 50%BT, 60%BT and 70%BT samples (C denotes celsian peaks and BT denotes barium titanate peaks described in text).

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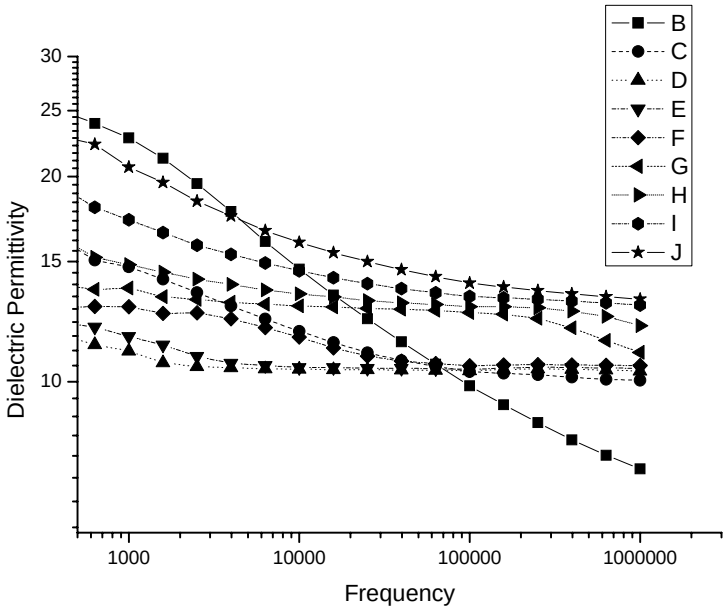
The composites derived from the merging of BT and Kaolinite at 1000°C yielded a series of transitions. The simple kaolinite BT composition was transformed to a poly-crystalline mix of celsian (barium feldspar) and BT. The 50%BT sample has tetragonal BT peaks with low levels of celsian phases in peaks 22.2° and 29.8° [4]. In 60%BT sample, the BT peaks increased in 2θ = 31°, 38°, 45° and 56° [5]. As the BT ratios increases in 70%BT, the celsian peaks decreases in peaks 22.2° and 29.8°, indicating that the low amounts of kaolinite-mullite does not sustain critical mass for the celsian formation in the composite. From what has been reported [6] and the and the results obtained, it is possible that the Ba^{2+} ions are reactive enough to form an immediate chemical bond with the Kaolinite-mullite liquid state during the sintering process. Inorganic chemistry shows that Ba^{2+} ions are electropositive and thus capable of forming a temporary bond with the kaolinite-mullite structure [6]. As the composite cools down slowly with the removal of water layer (cooling rate = 1°C/min), the intermediate liquid state compound rearranges itself to form celsian. However, this reaction is only valid for lower ratios since 50% BT does not increase the celsian formation.



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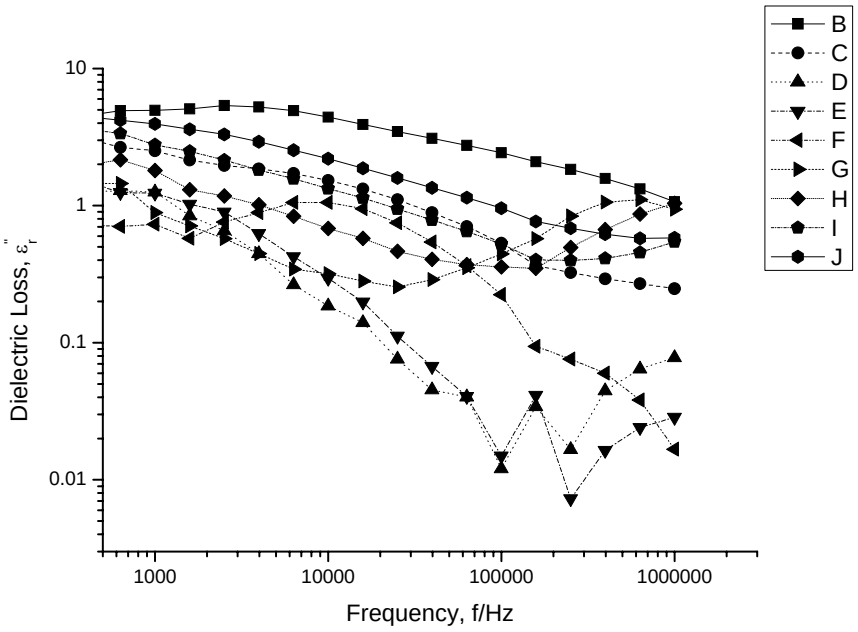

(b)

Figure 2: (a) Dielectric permittivity and (b) Dielectric losses for 50%BT at frequencies 500 Hz –1 MHz and temperatures 30°C to - 400°C.

Dielectric measurements showed that the addition of BT did not increase the dielectric permittivity. Instead, the 50%BT sample exhibit the dielectric response of kaolinite-mullite sample. This polycrystalline sample has immense interfacial boundaries between the compounds, thus impeding polarization. This phenomenon causes the stimulated signal response to favor the kaolinite-mullite path due to the interfacial and vibration effect. This is demonstrated by the convergence of the dielectric permittivity at 10 similar to other BT composites [7, 8]. The losses showed varying low losses with no distinct patterns, which might be due to the heat loss generated by the different components in the samples.

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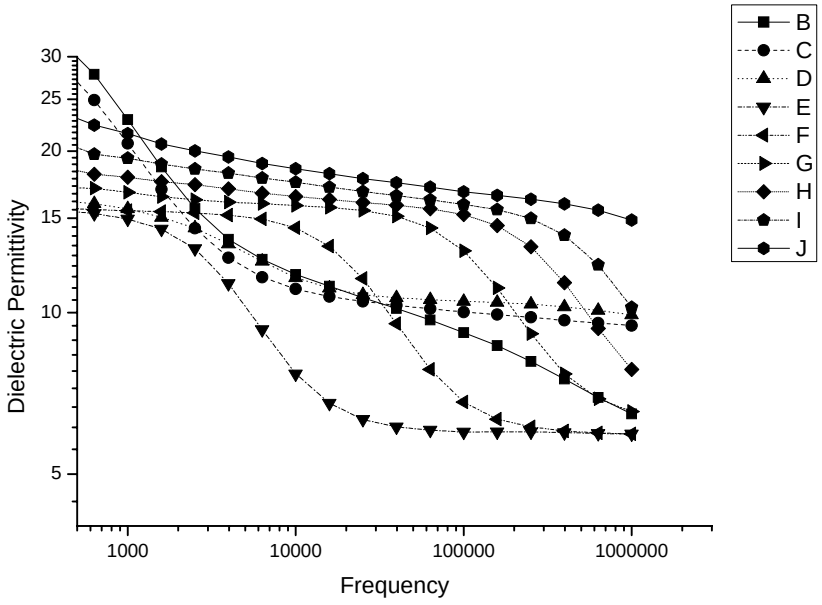

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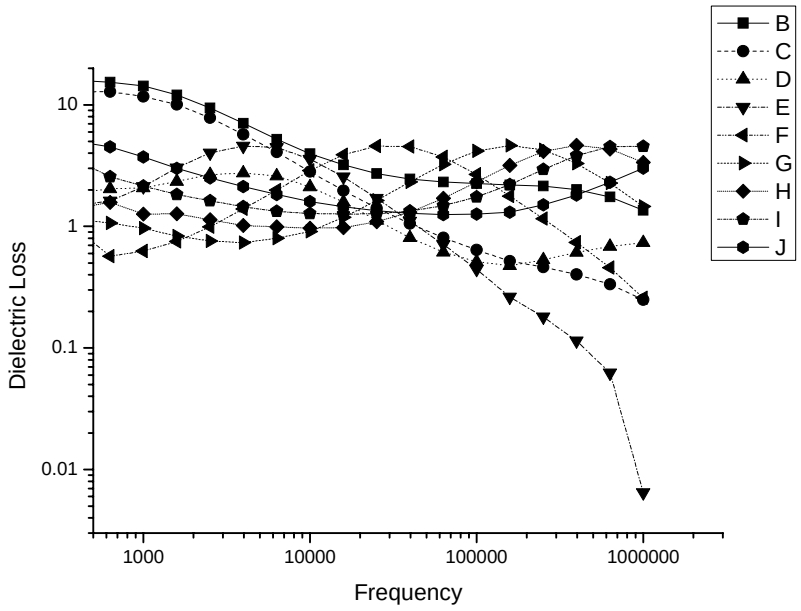

(b)

Figure 3: (a) Dielectric permittivity and (b) Dielectric losses for 60%BT at frequencies 500 Hz –1 MHz and temperatures 30°C to - 400°C.

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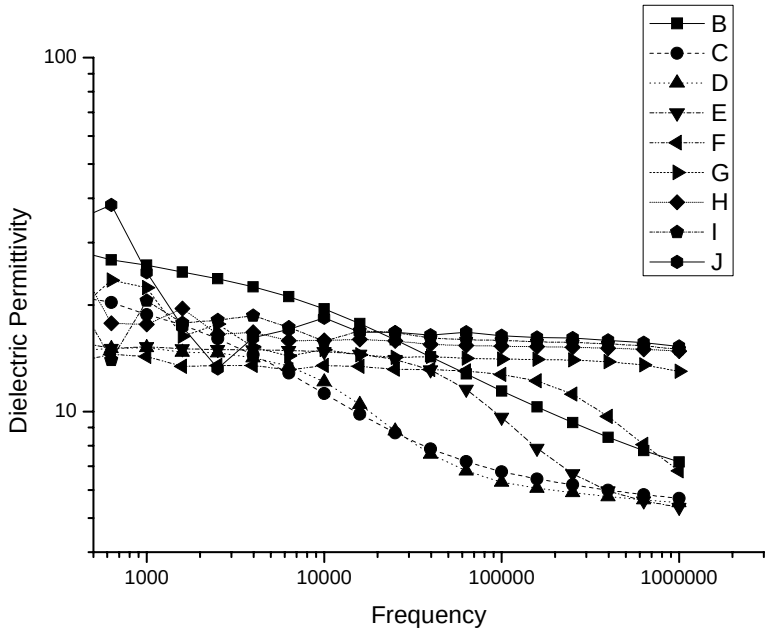

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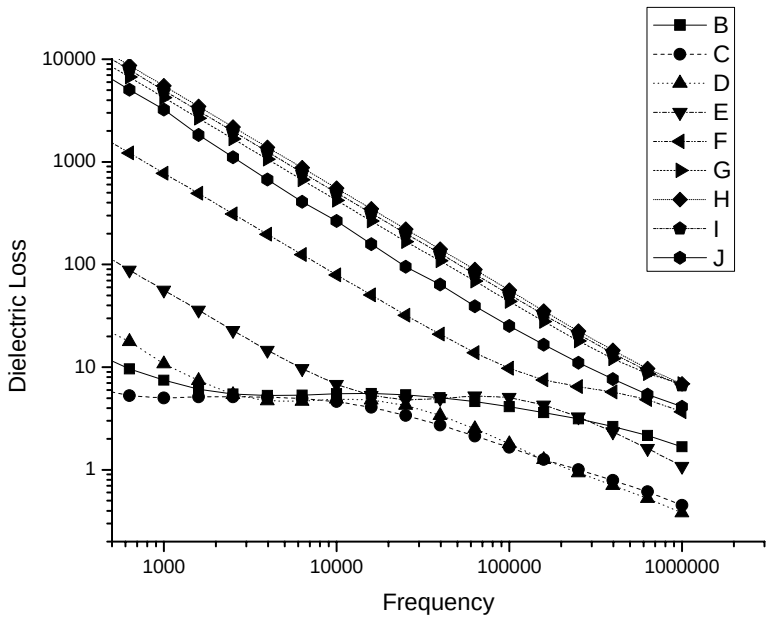

(b)

Figure 4: Dielectric permittivity and losses for 70%BT at frequencies 500 Hz –1 MHz and temperatures 30°C to - 400°C.

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Table 1: Dielectric permittivity, ε_r' at 1 KHz and 1 MHz at varying temperatures 30°C to 400°C.

ε_r' at 1KHz	30°C	50°C	100°C	150°C	200°C	250°C	300°C	350°C	400°C
50%BT	22.8	14.7	11.1	11.6	12.8	13.7	14.8	17.2	20.6
60%BT	22.9	20.7	15.6	14.9	15.5	16.7	17.9	19.3	21.5
70%BT	25.9	18.8	15.1	15.2	14.3	22.3	17.6	20.5	24.7
ε_r' at 1MHz									
50%BT	7.4	10.0	10.3	10.4	10.5	11.0	12.0	12.9	13.2
60%BT	6.4	9.4	9.9	5.9	5.9	6.5	7.8	10.2	14.9
70%BT	7.2	5.6	5.5	5.3	6.8	12.9	14.8	14.9	15.2

In Figure 3, the dielectric response indicated convergence similar to 50%BT sample but this sample exhibits pattern shifts that showed distinct behaviors at two different temperature ranges. The first behavior was observed at 30°C-100°C, which were below the Curie temperature of BT (120°C). The others are indicative of the second behavior. These results, coupled with the relatively high losses, indicate a BT dielectric response. However, the low dielectric permittivity indicates that the interfacial resistance is blocking effective polarization of BT in the composite.

Figure 4 also indicates that the two behaviors occurred in the 70%BT sample. The low dielectric response of the first three curves showed kaolinite-mullite response. The other behavior exhibits high dielectric permittivity at the low frequency end before being reduced to kaolinite-mullite response. The loss factor indicates gradual and increasing shifts in the sample. These results prove the dominant behavior of BT in the composite.

To illustrate further, Table 1 tabulated the dielectric permittivity data for the three samples at varying temperatures. Following ε_r' at 1 KHz, all three samples has a U-shape decrease and increase pattern. These data also proves increased amounts of BT increases the samples dielectric polarization. The interesting part of the response are the gradual incremental relation between ε_r' and high temperatures, especially above 250°C. This behavior does not conform to normal ceramic dielectric permittivity because ε_r' decreases with intense lattice vibrations and thermal interactions. This will be investigated further in future research.

CONCLUSIONS

This research was designed to form and characterize the CMCs produced form merging BT and Kaolinite powder. The resultant material is a composite of BT, mullite and barium feldspar, which was form in two particular mix ratios. The dielectric response for the samples yielded low and erratical responses in varying frequency and temperature conditions. The loss factor for the samples produced were low and indicative of the BT insertion, causing the interfacial and boundaries heat effect losses.

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The 50%BT sample is probably the best sample because of its stable dielectric ε_r' over the test frequency region and low dielectric loss.

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