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PROGRESS ON THE BREAKDOWN VOLTAGES AND LEAKAGE CURRENTS ANALYSIS OF MULLITE PRODUCED FROM KAOLIN

[*1]Aliyu, A., [1]Imam, R. I., [1]Galadima, A. I., [2]Suleiman, S. A.

[1]Physics Department, Ahmadu Bello University, Zaria, Nigeria
[2]Samaru College of Agriculture, Ahmadu Bello University, Zaria, Nigeria.

*Corresponding authors' email: aliyuelokene@gmail.com

ABSTRACT
Kaolin is a clay mineral that has found its huge use in electrical insulation. In electrical power industries, mullite insulators plays a vital role during electric power transmission. The transformation of kaolin to mullite via sintering method was carried out by varying the temperature during heating at interval of $50℃$ from $1000℃$ to $1200℃$. The mass of the kaolin was observed to have changed during the heating until the complete transformation took place at the temperature of 1150^0C. The dielectric strength (breakdown voltage) was analysed by sending high input voltage through the samples and measuring the output voltage until breakdown occurred. The input voltage of 1kV to4kV was passed through the samples and breakdown occurred at 5kv input voltage for almost all the samples. The leakage current was also determined using Ohm's law. 1150^0C was found to be the optimum temperature for the transformation of kaolin to mullite. The leakage current value (116.47×10^{-3}mV)was found to be negligible making mullite a good dielectric material for insulation.

Keywords: Mullite, breakdown voltage, dielectric strength, leakage current, metakaolin

INTRODUCTION
Mullite materials are generally used as thermal insulating materials due to their low thermal conductivity and a sufficiently cold compressive strength (Rhanim *et al*, 1997). Recently, it become imperative to look at what happens when it is subjected to high voltages while monitoring its leakage current as the sintering progresses (Roy and Maitray, 2014). Mullite is a ceramic material, which is composed of alumina-silicate compound that is extensible used in local refractory and many modern structural, optical, and electrical applications (Arksay *et al*, 1991). Many local ceramic products have mullite as part of their raw material, since most of their product usually contains some quantity of clay and silicon as their initial or starting material (Juliana, 2005). Its occurrence in nature is very rare, and hence providing an alternative way of producing this important material and measure of its electrical values are the aims of this research. Although it is one of the most commonly found phase in industrial ceramics, the importance of mullite as a ceramic phase was only recognized during this century of pioneering work of (Bowen,1924).
It is thermally and chemically stable both in air and under atmospheric pressure. It has very good thermo-mechanical properties, which allow its usage as structural element in systems, such as thermal engines. It has a relatively low thermal expansion co-efficient leading to a good thermal shock resistant (Fauzia *et al*., 2018).
Mullite is becoming increasingly importance in electronics, optical and high temperature structural applications, because of its low dielectric constant, good transparency for mid-infrared light and excellence to creep resistance (Chena *et al*, 2004). Classical uses of Mullite include refractory in the metallurgical industries for electric furnace roofs, hot metal mixers, and low frequency induction furnace (Angel and Prewitt, 1986). In the glass industries, these refractories were employed in the upper structure of the tank in which glass is melt and for constructing the drawing of chambers (Abdezadeh, 2003). Mullite is frequently used as kiln setting slabs and posts for firing ceramic ware as well as for the linings of high temperature reactors (Sardy *et al*, 2012). However, during the last decade, the spectrum of actual potential employment of mullite now include its use as a matrix materials for high temperature composite development, a substrate in multilayer packaging, protective coatings, turbines engines components and infrared-transparent window especially for high temperature application (Juliana, 2005).
This work focuses on the procedures for converting this kaolin to mullite and the subsequent measurement of its breakdown voltage and leakage current. Although several methods are used for synthesizing mullite, this research used sintering method due the nature of the sample used.

MATERIALS AND METHODS
Materials
Sensitive electronic weighing balance, Universal testing machine (TQ), Extrusion die and crucibles, Mortar and pestle, Transparent bottle, Test Cell and Measuring meters, Dropper, Other hand tools, Raw kaolintic clay sample, LCR meter.

Methods
For transformation of kaolin into mullite, the samples (kaolin) have to be subjected to extremely high temperature above one thousands one hundred degree Celsius (1100^0C). Heating of kaolin, at a temperature between (650^0C) and (800^0C), result to the transformations of kaolin structure into metakaolin structure. At temperature of about one thousands two hundred degree Celsius (1200^0C), the metakaolin structure is expected to completely transform to mullite.

Figure 1: Electrical furnace

The sample (kaolin) was divided into five different portions using a sensitive electronic weighting balance (*fig: 2)*, each weighing $120g$ approximately. The samples were labelled A, B, C, D and E respectively.
Samples 'A' were put into a high thermal resistant crucible and heated at a temperature of a thousand degree Celsius (1000^0C) using an electrical furnace (*figure 1)*. The heating of the sample continuous at the constant temperature of (1000 ^{0}C) for up to three hours (3hours). subsequently the remaining samples which are B, C, D and E were heated at the temperatures of 1050^0C, 1100^0C, 1150^0C and 1200^0C respectively.

Compaction of the Sample

The powdered sample (kaolin) which was transformed to mullite by heating needs to be compacted to solid form for appropriate measurement to take place, through a process known as compaction (that is to convert the powdered form into solid form). The material (kaolin) is less adhesive and hence the need for an adhesive agent or binder to enable it to be compacted. The adhesive material used in this research was a liquid binder.

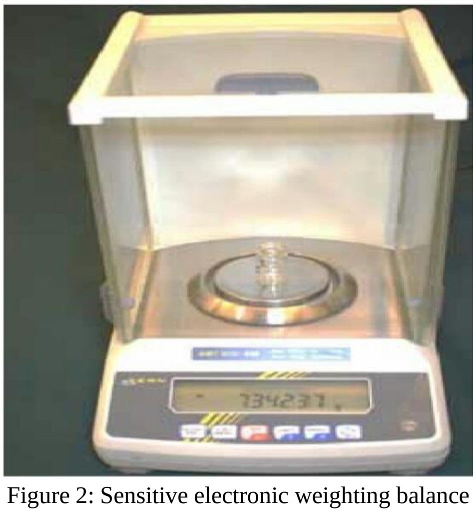

Figure 2: Sensitive electronic weighting balance

Approximately $6g$ of each samples (A, B, C, D and E) was weighed and put into mortar, $3ml$ of the binder was dropped into the mortar containing the sample using a dropper. A small pestle was used to vigorously mix the mullite and the binder until homogeneity was achieved. The homogenous mixture was divided into three parts with slight variation in mass using a sensitive electronic weighing balance (Fig 2)
The mixed mullite was then transferred into the extrusion die and with the aid of Universal testing machine (TQ)(*fig: 3)* This was finally compacted into the circular round shape with the diameter of fifteen millimeter (15mm) and the thickness of approximately two point five millimeter (2.5mm). The amount of load used for compaction was twenty thousand Newton (20,000N) or twenty kilo Newton (20KN). Subsequently, the remaining samples were compacted using the same procedure as above and at the same compaction load as above.

Figure 3: universal testing machine (TQ)

Measurement of Breakdown Voltages and Leakage Currents

The sample (mullite) was place between the two electrodes of the test cell. The test cell was adjusted in order to clamp the sample firmly on the electrodes. The test cell (*fig: 3.4b)* was connected to a high voltage source through a series current limiting resistor of 50 kΩ. The voltage was slowly and carefully raised from the DC Breakdown source (35 kV) stepwise at 1kv/s and the potential difference across the measuring resistor were measured using a Bench Top Digital Multimeter (*fig: 4a)* until the breakdown occurs. Three breakdowns were conducted for every sample and the average values were taken.

Figure 4: (a) Measuring Meters

Figure 4: (b) Test cell

The leakage current was determined from the measured voltage across the measuring resistance of the resistor using Ohm's Law (V=IR) and I=V/R

RESULTS AND DISCUSSIONS

The result of the heating of the kaolin samples presented in table 1 shows a significant decrease in mass of the samples as the heating temperature is increased. The mass of sample A decreased by about 16.67% after heating at a temperature of 1000℃. The mass of sample B decreased by about 25% after heating1050℃. Sample C had decreased by about 28.33%% in mass after heating it at 1100℃, sample lost about 33.33% of its initial mass after heating it at 1150℃. Sample E was heated at 1200℃ and it lost almost the same amount of mass is that of sample D. these showed that kaolin sample heated at temperatures below 1150℃ will exhibit metakaolin properties (Samples A, B, C) while kaolin fired above the said temperature will most likely transform into mullite. Samples calcined above 1150℃ up to 1200℃ showed no significant reduction in mass implying higher thermal and creep resistance. Generally, mulite's intensity and structure highly depends on firing conditions (firing temperature and dwelling time) as well as quality and quantity of raw materials applied for production (Meng et al., 2016)

The breakdown voltage analysis results were presented in tables 2-6. The breakdown voltage of an insulator is the minimum voltage that can cause a portion of the insulator to become electrically conductive. It is also known as the dielectric strength. As the V_{in} in kV increases v from 1 to 4, the average V_{out} also increases for all the samples. The breakdown occurred at $5kV$ V_{in} for almost all the samples. It was also observed that as the heating temperature increases 1000℃ to 1150℃, the average V_{out} also increases but with slight at the heating temperature of 1200℃. This is a

necessary a understanding because the high insulating character of mullite, electric current measurements have not been performed at room temperature yet (Maximina *et al*,2021).
Sample 'A' breaks at an input voltage of (5KV) with a maximum output voltage of (13.85 $\times10^{-3}$ mV).Sample B also breaks at an input of 5KV with a maximum output voltage of 15$\times10^{-3}$mV. Similarly sample C breaks at 5KV also with maximum output voltage of 20.46$\times10^{-3}$mV. Sample D that breaks at 5KV also have a reasonable amount of output voltage of 5823.50mV, which is the optimum, compared with the other samples. The last sample, which is sample E, also breaks at an input voltage of 5KV produces an output voltage of 4419.50mV.
The result of the leakage current ($I_{leakage}$) were contained in tables 7-11. Ohm's law was used to compute the $I_{leakage}$ as the V_{in} is increased from from 1-4kV for all the samples. The result showed a very small leakage current that can be ignored for all the samples, indicating a very good material for insulating purposes bearing in mind the breakdown voltage. This is in consonant with the work of (Andualem *et al*, 2022) which holds that cullet fired at 1150 ℃ and 1200℃ satisfy the required technical and electrical properties for insulation application
The summary of the breakdown voltage and leakage current were presented in tables 12-13. As the temperature increases from 1000℃ to 1150℃, the maximum output voltage $V_{out\ max}$ also increases reaching a maximum of $5823.50mV$ at 1150℃. But there is a slight decrease at a temperature of 1200℃ (table 12). Also the temperature increases from 1000℃ to 1150℃, the average voltage output increases and the maximum leakage current increased too, all reaching maximum value at 1150℃. Again, there was slight decrease in values of the average voltage output and the maximum leakage current at 1200℃ (table 13)

Table 1: Heating of Kaolin

S/N	Samples	Temperature (^{0}C)	Initial Weight (kg) 10^{-3}	Final Weight (kg) 10^{-3}
1	A	1000	120	100
2	B	1050	120	90
3	C	1100	120	86
4	D	1150	120	80
5	E	1200	120	81

Table 2: 1000^{0}C Breakdown Voltage Analysis for Sample A

V_{in}(kV)	V_{out}(mV) 1st$\times10^{-3}$	V_{out}(mV)2nd$\times10^{-3}$	V_{out}(mV)3rd$\times10^{-3}$	Average (mV)$\times10^{-3}$
1	1.39	2.32	1.23	1.65
2	4.71	7.78	3.25	5.23
3	12.83	12.78	9.04	11.55
4	15.44	14.40	11.72	13.85
5	Bdv	Bdv	Bdv	Bdv

Table 3: 1050^{0}C Breakdown Voltage Analysis for Sample B

V_{in}(kV)	V_{out}(mV) 1st$\times10^{-3}$	V_{out}(mV)2nd$\times10^{-3}$	V_{out}(mV)3rd$\times10^{-3}$	Average (mV)$\times10^{-3}$
1	5.27	2.94	5.46	4.56
2	14.49	8.24	14.23	12.32
3	17.45	12.63	20.15	16.74
4	Bdv	15.67	Bdv	15.67
5	Bdv	Bdv	Bdv	Bdv

Table 4: 1100^{0}C Breakdown Voltage Analysis for Sample C

V_{in}(kV)	V_{out}(mV) 1st$\times10^{-3}$	V_{out}(mV)2nd$\times10^{-3}$	V_{out}(mV)3rd$\times10^{-3}$	Average (mV)$\times10^{-3}$
1	13.75	8.49	1.39	7.89
2	29.71	18.19	15.32	21.07
3	25.23	21.63	13.20	20.02
4	Bdv	23.32	17.60	20.46
5	Bdv	Bdv	Bdv	Bdv

Table 5: 1150^{0}C Breakdown Voltage Analysis for Sample D

V_{in}(kV)	V_{out}(mV) 1st	V_{out}(mV)2nd	V_{out}(mV)3rd	Average (mV)
1	483	1259	469	737.00
2	1665	3639	1290	2197.00
3	3250	3696	3532	3492.67
4	5866	Bdv	5781	5823.50
5	Bdv	Bdv	Bdv	Bdv

Table 6: 1200^{0}C Breakdown Voltage Analysis for Sample E

V_{in}(kV)	V_{out}(mV) 1st^3	V_{out}(mV)2nd	V_{out}(mV)3rd	Average (mV)
1	483	203	732	469.67
2	1776	771	2338	1628.33
3	3511	2143	5167	3607.00
4	4420	4419	Bdv	4419.50
5	Bdv	Bdv	Bdv	Bdv

Table 7: 1000^{0}C Leakage current for Sample A

V_{in}(kV)	Average V_{out} x $10^{-3}mV$	Leakage Current I x $10^{-8}A$
1	1.65	3.3
2	5.23	10.46
3	11.55	23.10
4	13.85	27.70

Table 8: 1050^{0}C Leakage current for Sample B

V_{in}(kV)	Average V_{out} x $10^{-3}mV$	Leakage Current I x $10^{-8}A$
1	4.56	9.12
2	12.32	24.64
3	16.74	33.48
4	15	30

Table 9: 1100^{0}C Leakage current for Sample C

V_{in}(kV)	Average V_{out} x $10^{-3}mV$	Leakage Current I x $10^{-8}A$
1	7.89	15.78
2	21.07	42.14
3	20.02	40.04
4	20.46	40.92

Table 10: 1150^{0}C Leakage current for Sample D

V_{in}(kV)	Average V_{out} mV	Leakage Current I x $10^{-3}A$
1	737.00	14.74
2	2197.00	43.94
3	3492.67	69.85
4	5823.50	116.47

Table 11: 1200^{0}C Leakage current for Sample E

V_{in}(kV)	Average V_{out} mV	Leakage Current I x $10^{-3}A$
1	469.67	9.34
2	1628.33	32.56
3	3607.00	72.14
4	4419.50	88.39

Table 12: Summary of Breakdown Voltage result

Sample	Temperature(^{0}C)	Breakdown voltage(KV)	Max. V_{out}(mV)
A	1000	5	13.85×10^{-3}
B	1050	5	16.74×10^{-3}
C	1100	5	20.46×10^{-3}
D	1150	5	5823.50
E	1200	5	4419.50

Table 13: Summary of Leakage Current

Sample	Temperature (^{0}C)	V_{in}(kV)	Avr. V_{out}(mV)	Max. Leakage current I (A)
A	1000	4	13.85×10^{-3}	27.70×10^{-8}
B	1050	3	16.74×10^{-3}	33.48×10^{-8}
C	1100	4	20.46×10^{-3}	40.92×10^{-8}
D	1150	4	5823.50	116.47×10^{-3}
E	1200	4	4419.50	88.39×10^{-3}

CONCLUSION

In thus research, kaolin was transformed to mullite via sintering at a temperature of 1150℃. This is because at a temperature of 1150℃ and 1200℃, the kaolin samples showed no significant change in mass compared to the other samples that were fired at a temperature below 1150℃. the fact that there was no significant change in mass after heating is a clear sign of one of the properties of mullites formation (that is resistance to higher thermal energy). At the 'metakaolin state' the mass decrease as the temperature increases, so also is the voltage output of the breakdown voltage test. The breakdown voltage input for mullite is $5kV$ from this research work. 1150℃ is the optimum temperature for the transformation of kaolin to mullite.

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