Potentials of Graphite as a Material for the Production of Crucibles

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Abstract

This study looked into the use of graphite as a material for making crucibles using the clay slip casting process. The investigation included determining the chemical compositions of graphite samples sourced from Sama Borkono in the Warji local government area of Bauchi State, processing the graphite into crucibles using the slip casting technique, and determining the physical properties of the crucible using ASTM standard methods such as shrinkage, porosity, refractoriness, shock resistance, and heat conductivity. The graphite sample's chemical analysis revealed that the beneficiated graphite has a sufficient amount of carbon content (80 to 85 percent) for crucible manufacturing. The percentage shrinkage of the samples ranged from 2% to 7%, according to the shrinkage tests performed on them. Thermal shock testing revealed that all of the samples could tolerate rapid temperature changes when subjected to various temperatures. The samples' refractoriness test revealed that all of the samples can tolerate temperatures of more than 12000 °C. According to the conductivity test, sample B had a decent heat conductance of 1.49 W/mK. Following the tests, sample "B" was discovered to meet all of the requirements for graphite crucible manufacture (high refractoriness, high conductivity, good thermal shock resistance, low shrinkage, and low porosity). A brass crucible was melted to a temperature of 9270C to test the crucible. According to the findings of the study, non-ferrous metals with melting temperatures below 12000C can be melted in the crucible made from sample B.

Keywords: *Graphite, Crucible, Clay, Kaolin, Fireclay, Lime, Slip casting, Brass, Non – ferrous metals.*

Introduction

Refractory materials can survive high temperatures and the physical and chemical action of molten slag and furnace gases without deformation, failure, or composition change. Refractory, a material widely used in metallurgical, foundry, and power plants, is now imported. The metallurgical industry alone consumes 80% of refractory. If these resources were acquired locally, the country's foreign exchange profits would be greatly reduced.

Typical refractory materials in Nigeria include carbon (graphite and coke), clay, oxides (alumina, magnesia, zirconia) and non-oxides (carbides, boron carbides, nitride, etc.). However, it can be produced artificially from petroleum coke using an electric furnace. Graphite is a solid form of carbon that is resistant to thermal shock, basic slag, and corrosive chemicals and oxidants (Nwobi, 2006). So they are used to make crucibles. They are used for high-temperature activities, including metal smelting, melting, and casting (Chirikure and Hall, 2013).

A crucible is a cylindrical container put into the furnace (Osarenmmwind, 2015). These crucibles are open-mouthed jars made of graphite and clay. This makes it effective in graphite crucibles. Previous research on the use of indigenous (Bauchi) graphite for jewellery crucibles was not successful. Muhammed (2014) examined Sama Barkono graphite beneficiation, but did not use it for jewellery crucible manufacturing, despite the graphite's suitability for crucible production. Nwobi (2006) used Bauchi graphite to manufacture 30 kg steel crucibles using pressing methods. The 30 kg crucible is too large for the local foundries. Defects included failure to heat the crucible to the proper working temperature. So a proper crucible must be made from diverse materials and methods.

Materials and Methods

Materials

The graphite samples were gathered from graphite mining sites in the Warji local government of Bauchi state, specifically in the Sama-Barkono and Dutsenhayar districts. The site was discovered with the help of local miners and data from Muhammed (2014) and Adewale (2014) research (2009). The graphite samples were extracted from an abandoned mine pit that had become a tiny pond due to rainwater collection.

Clay, kaolin, plaster of Paris (P.O.P), all sourced from Alkaleri local government area in Bauchi state, spine oil, keroseene, magnesium oxides (MgO), calcium oxide (CaO), chlorium oxides (CrO), aluminium oxides (Al2O3), sodium hydroxide (NaOH), and distilled water are among the other raw materials used.

Equipment

The following tools were utilised in the research:

Pascallengg Ltd 17747 sieve shaker, Pascallengg Ltd 17769 freq.50hz, Pascallengg Ltd 17949 ball mill crusher, jaw BJAE 19282/1 Type D12 Flotation Machine, Weight Balance PC180Drying oven, Genlanwidnee, England, BJAE19822/1, D12, Carbon Sulphur Analyzer CSA–996Altico, Ltd., BJAE 19282/1 Type D12 Flotation Machine, Weight Balance PC180Drying oven, Genlanwidnee, England, BJAE19822/1, D12, Carbon Sulphur Analyzer CSA–99 (kick wheel, electric furnace).

Method

The crushing of graphite or sample, ball milling, production of cones, and quartering to generate representative samples are all part of the experimental technique in this work.

Coning and Quartering

Crushed materials are heaped into cones by dumping all of the material onto one spot on a clean surface in such a way that the particles roll down in all directions, distributing the mass's composition into concentric layers of a cone. The top of the cone was then flattened by distributing the material evenly in all directions with the edge of the shovel until a disc was produced.

The materials from the diagonally opposing quadrants were taken as samples from this disc, which was divided into quadrants. The items from the other quadrants were discarded. The mass now holds half of what it did before. The material was then crushed again, and the coning and quartering processes were repeated, decreasing the sample quantity to one-fourth of its original size. The procedure was repeated until the necessary amount was achieved.

Sizing

The sieves were arranged using a sieve scale in which the ratio of neighbouring sieve aperture widths matches the square root of two (2 = 1.414). To collect the finest and tiniest particles of the sample, sieves ranging in size from 355m to 180m were stacked with the coarsest sieve on top and the finest on the bottom. To catch the final undersize, a tight-fitting pan was placed below the lowest sieve, and a lid was placed on top of the coarsest sieve to keep the sample from escaping. The undersized material falls through successive sieves throughout the shaking process until it is kept on a sieve with openings slightly smaller than the diameter of the particle.

Beneficiation of the Sample by Flotation Technique Method

The weighing balance was used to weigh a sample of ground graphite with a sieve size of 90 m. A total of 1000cm3 of distilled water was measured and mixed with the graphite ore before being placed into the flotation cell and agitated for 3 minutes at 2000rpm to generate a froth on the surface. Three drops of pH regulator (sodium hydroxide) were added during the agitation phase, and the mixture was conditioned for seven minutes to produce additional bubbles and froth. After that, 5ml of kerosene and pine oil were added just before the conditioning time ran out in order to make more bubbles.

Air was allowed to travel through the pulp for 2 minutes after conditioning and agitation, and froth arose and was collected until barren foam surfaced. This process was done several times, yielding significant volumes of concentrates. The tailings and concentrate were dried, weighed, and documented. A chemical analysis was performed on a representative graphite sample to establish the percent carbon content before and after beneficiation. Before and after beneficiation, a representative sample was submitted to XRF and a Carbon–Sulfur Analyzer, and the contents in elemental form were revealed and recorded in table 3.

Samples Preparation

A, B, C, D, and E were assigned to the five sections of the beneficiated graphite powder. Portion A was then combined with 150 grammes of refractory clay, 10 grammes of silica, and 45 grammes of kaolin, all of which had been dried and powdered. After that, 150g of refractory clay, 10g of silica, and 60g of fire clay were added to Sample B. Portion C combined 150gof refractory clay, 10g of silica, and 45g of lime. Sample D was made up of 150 grammes of refractory clay, 30 grammes of kaolin, and 30 grammes of lime. Sample E was made up of 150 grammes of clay, 80 grammes of fireclay, and 20 grammes of silica.

After that, each sample part was mixed with water and cast in a 2 cm x 3 cm x 5 cm mould, which was then allowed to cure at room temperature for seven days before being removed. This method yielded twenty-five test samples, or five pieces of each sample. A, B, C, D, and E were then assigned to the rectangular bricks.

Slip Casting

Preliminary studies on the five sample mixtures revealed that sample "B," which is a 50 percent graphite, 30 percent clay, 15 percent fire clay, and 5% silica blend, has the best qualities. A model of a cylindrical crucible with a top and bottom exterior diameter of 10 cm, an internal diameter of 7 cm, a height of 9 cm, and a thickness of 1.5 cm was made from secondary clay and thrown on a kick wheel.

Plate I: A model of crucible being produced for slip casting

The model was then used to make a plaster of Paris mould. As a result, slip was created by dissolving graphite, clay, fire clay, and silica in water. The suspension was then poured into a plaster of Paris mould, where the water from the slip was absorbed, leaving a solid coating on the mould wall. The cast piece was removed from the mould as it dried and shrank.

Plate II: Slip is poured in the plaster mold

The crucibles were then allowed to dry in the open air for three days before being dried in an oven for twelve hours at 1100C to remove any remaining moisture and avoid cracks during burning. The crucible was then burned for eight hours at 12000C in an electric heating furnace. The crucibles were allowed to cool in the furnace at a rate of 10 degrees Celsius per minute after the fire.

Plate III: Graphite crucibles before and after been fired

Result and Discussion

Table 1: Elemental Composition of Rep Sample, Concentrate and Tailings Using x – ray florescence (XRF)

ELEMENT	REP SAMPLE	CONCENTRATES	TAILINGS
LOI	22.96	69.25	2.23
Al_2O_3	6.35	1.15	17.50
SiO_2	49.14	14.70	75.11
SO_3	< LOD	<LOD	< LOD
K_2O	5.70	4.59	0.05
CaO	3.09	1.80	0.91
TiO_2	0.40	1.30	1.31
V_2O_5	0.38	0.01	< LOD
Cr_2O_3	0.06	1.03	< LOD
MnO	1.86	0.75	0.42
Fe_2O_3	9.03	3.51	0.89
MgO	0.15	0.43	0.50
P_2O_5	0	0	0.18
PbO	0.19	0.10	<LOD
Na_2O	< LOD	< LOD	< LOD
ZnO	0.42	0.63	< LOD
BaO	0.26	0.74	1.11
Total	99.9	99.9	99.9

OI = Loss on Ignition
LOD = Less than level of Detection

Table 1 shows that the graphite sample tested using x-ray fluorescence exhibits high silica (SiO_2) impurities (tailings) that can be used in Portland cement manufacturing, as an additive in the production of glass for doors and windows, and optical fibres for communications. It's also suitable for use in food and pharmaceuticals. Loss on ignition (LOI) is the loss of organic, combustible, or volatile material when heated, and it has an impact on shrinkage and porosity values (Jock, 2013). According to Bojko & Kabala, the loss on ignition obtained after the graphite sample was evaluated is 2.23 percent, which is within the recommended range of 0.93–92.1 percent (2016). The recovery of graphite by floating demonstrates that the floatation method of beneficiation is effective and may be used to produce pure graphite concentrate on a large scale. Mineral gangue tailings, which account for approximately 79 percent of total tailings, will be a significant source of silica production.

Table 2: Result of the Analysis for Carbon and Sulfur content in the Sample before Floatation

S/N	Analysis Time	Carbon%	Sulfur %	Weight(g)	Sample Name	Sample No.
1	2017-05-25 01:42:16	30.01134	0.27210	0.5100	Rep.Sample	1

Using carbon – sulphur analyser in table 2 above, the percentage carbon obtained before beneficiation by froth floatation was 30% approx

Table 3: Result of the Analysis for Carbon and Sulfur content in the Sample after Floatation

S/N	Analysis Time	Carbon %	Sulfur %	Weight (g)	Sample Name	Sample No.
1	2017-05-25 01:47:10	79.02730	0.30231	0.5100	Concentrate	1

As illustrated above, the percentage of carbon obtained after beneficiation was 79 percent, as shown above (approximately 80 percent). Because graphite containing 80 to 85 percent carbon is used to make crucibles, the purity obtained is considered "high purity graphite." This is consistent with Muhammed et al.'s (2014).

The graphite is of good grade as the purity before and after beneficiation falls within the range of high purity graphite, based on the results obtained for pre and post beneficiation. Other tailings, such as AlO_3, TiO_2, and BaO, which make up around 20% of the graphite tailings, are also very useful industrial minerals.

Table 4: Result for the Shrinkage test of the Sample

Sample	Cross Sectional Area Before Oven Dry mm^2	Cross Sectional Area After Oven Dry mm^2	Change in Cross Sectional area mm^2	Shrinkage %
A	1340.77	1260.6	80.17	6.4
B	1362.19	1262.63	99.56	7.8
C	1374.15	1176.55	197.6	16.8
D	1308.20	1278.69	29.51	2.3
E	1385.92	1190.31	195.61	16.4

The percentage shrinkage of sample "A" (50 percent graphite, 30 percent clay, 5 percent silica, and 15 percent kaolin) is 6.4 percent, according to the percentage shrinkage values in table 4. Sample "B" (50 percent graphite, 30 percent clay, 5% silica, and 15% fire clay) had a shrinkage of 7.8 percent, which could be attributed to the addition of fire clay rather than kaolin. In sample "C," which contains 50 percent graphite, 30 percent clay, 5 percent silica, and 15 percent lime, the percentage shrinkage is 16.8 percent. Sample "D" (50 percent graphite, 30 percent clay, 10% kaolin, and 10% lime) has a very low concentration of 2 percent, which could be attributable to the inclusion of lime and kaolin. The rise in percentage shrinkage in sample "E" (40 percent graphite, 30 percent clay, 20 percent fireclay, and 10% silica) could be attributable to the decrease in graphite sample and the increase in clay content in the mixture. The comparatively high percentage of shrinkage in the samples shows issues with not adding grog to the refractory clay–graphite mixture. The term "grog" refers to burned clay particles. It is mostly included as an anti-shrinkage component in the form of angular particles of various sizes to improve grain interlocking (Aigbodion et al., 2014). According to Abubakar et al., the shrinkage of samples "A," "B," and "D" is within the permitted range of 4–10 percent (2014)

Table 5: Result for the Thermal Shock Resistance of the Samples

S/N	SAMPLE	Temperature ^0C	Initial colour	Final Colour	Crack Formation
1	A	1000	Dark ash	Light Brown	Slight Crack
2	B	1000	Dark ash	Light Brown	Slight Crack
3	C	1000	Dark ash	Light Brown	Slight Crack
4	D	1000	Dark ash	Light Brown	Slight Crack
5	E	1000	Dark ash	Light Brown	Slight Crack

The thermal stability, also known as thermal shock resistance, of refractory materials declines with rising firing temperature. It is well recognised that products that maintain thermal stability even after high firing temperatures or operating temperatures are the most beneficial in practise (Obikwelu, 2002). Thermal shock is a refractory attribute that is measured when the refractory is subjected to alternate heating and chilling. The samples had a thermal shock resistance of 28 cycles. Table 5 showed excellent thermal shock resistance. Thermal shock resistance is within the acceptable 20–30 cycle range (Abubakar et al., 2014). The results of the samples' thermal shock resistance reveal that all of them can endure a quick shift in temperature after being heated. After 28 cycles, all of the samples developed a very tiny break in their bodies. The inclusion of silica into the combinations provides excellent thermal shock resistance.

Table 6: Result for the Apparent Porosity of the Samples

S/N	SAMPLE	W1 (SOAKED) g	W0 (OVEN DRIED) g	$\frac{W1 - W0}{W0} \times 100$
1	A	38	31	22.58
2	B	38	32.5	16.92
3	C	36	29.6	21.62
4	D	40	35	14.29
5	E	33	26.5	24.53

Porosity is a measure of the open pore space in a refractory into which molten metal, slag, fluxes, vapour, and other substances can penetrate and contribute to the structure's eventual degradation. A refractory material with a higher apparent porosity is less resistant to the eroding action of slag and solid particles that can pass through the pores inside the material (Obikwelu, 2002). Refractory products with low porosity have better mechanical strength and other qualities. As a result, a good refractory should generally have a lower porosity (Kispanai, 2018). The lesser the refractory's perceived porosity, the better. According to the apparent porosity of the samples in table 6, samples "B" and "D" have lower porosity than the other samples. An attribute of a crucible with low porosity and permeability is that gases and liquids cannot easily move through it (Nwobi, 2006). Sample "B" had 16.92 percent apparent porosity, while Sample "D" had 14.29 percent. This could be because their combinations contain kaolin. These results were slightly higher than the suggested range of 11–14% for clay–graphite bonded crucibles. This is consistent with Aigbodion et al.'s (2014). Thermal conductivity will be better in samples with a smaller apparent porosity. Porous refractories have high permeability, poor heat conductivity, low strength, and are less sensitive to temperature variations than solid refractories (Muhammed, 2014).

Table 7: Result for the Refractoriness test of the Samples

SAMPLE	TEMPRATURE (^{0}c)
A	1650
B	1700
C	1650
D	1500
E	1450

Refractoriness is a measurement of a refractory material's fusibility, or the temperature at which it softens or fuses. The temperature at which a material begins to lose its shape is known as refractoriness. A material's melting point is the temperature at which a material begins to lose its shape (Sadik et al., 2013). Refractories are non-metallic materials with chemical and physical qualities that make them suitable for structures or system components exposed to temperatures beyond 10000 °C (bolanle, 2011).

Table 9 shows that all of the samples are good refractories, suited for the manufacturing of crucibles, and can sustain heat or temperatures exceeding 14000C, making them ideal for melting metals such as gold 10630C, sterling silver 8900C, copper 10830C, brass 9400C, and aluminium 6600C. (Wicks, 2003).

Because of the use of fireclay in the mixture, Sample "B" had a maximum refractoriness of 17000C. A useful refractory substance is fire clay. The lowest refractoriness is seen in samples "E" and "D," which may be related to a decrease in graphite content and an increase in silica and lime in the mixture. The results reveal that as the graphite component in the mixture grows, so does the refractoriness. It reduces as the silica content rises, and graphite decreases.

The refractoriness of samples "A," "C," and

"D" is 16500C, 16500C, and 15000C, respectively. The presence of lime in sample "D" reduced the sample's refractoriness. According to Obikwelu, the acquired refractoriness values are lower than the refractoriness of clay-graphite products at 19000C. (2002). Working temperatures of 16000C, 16500C, 14500C, and 14000C can still be applied to the samples.

Table 8: Result for Thermal Conductivity of the Samples

SAMPLE	TEMPRATURE 1(^{0}c)	TEMPRATURE 2(^{0}c)	THERMAL CONDUCTIVITY(W/mk)
A	51	42	1.49
B	50	41	1.49
C	48	41	1.16
D	48	42	0.99
E	49	42	1.16

When subjected to high temperatures, thermal conductivity is a measure of a refractory's capacity to transport heat from the hot to the cold face. Thermal conductivity increases with temperature in refractories but decreases with increasing porosity. Temperature, chemical composition of the raw material for refractory products, mineralogical structure of the mix, actual porosity and pore size, and firing temperature all affect the thermal conductivity of refractory materials (Harbison, 2005).

Table 8 shows that samples "A" and "B" had the highest thermal conductivity of 1.49W/mK and 1.49W/mK, respectively, according to the results of the thermal conductivity test. The thermal conductivity of samples "C" and "E" is also 1.16W/mK. The sample with the lowest thermal conductivity, "D," is 0.99W/mK, which could be related to the lack of silica in the combination. Kleiner et al. (1996) discovered that the thermal conductivity of a thin coating of silicon dioxide (silica) increases with increasing temperature in a study.

Test Firing the Crucible Produced from Sample" B"

The physical properties tests performed on the sample mixtures A, B, C, D, and E revealed that sample B has better qualities for producing graphite crucibles. Its high refractoriness of 17000 °C means it can be used in environments where temperatures of 15,000 °C are predicted. As a result, it can melt the majority of non-ferrous metals used in jewellery manufacture and casting. In the crucible created, brass fragments were melted. However, tiny cracks were discovered after firing, which could have been produced by firing with moisture in the crucible's body.

Conclusion and Recommendations

According to the findings, Sama Barkono graphite is a good source of a refractory material utilised in crucible manufacturing. The study found that mixing graphite powder with several widely accessible additions yielded a very good refractory mixture. In the absence of an isotactic press machine, clay bound graphite crucibles can be made using the slip casting process.

The optimum method for graphite beneficiation is froth flotation, because the analysis after beneficiation revealed that other methods, such as chemical leaching and magnetic separation, could not remove the tailings and gangue elements. When the sample is not adequately and thoroughly dried before firing, little cracks appear on the surface of the crucibles. The samples' physical characteristics tests revealed that they might be used in our local foundries. The study also revealed that when Sama Borkono graphite is properly analysed and beneficiated, it provides the exact amount of carbon needed for the manufacturing of graphite-clay bonded crucibles.

The following suggestions are given based on the findings of this study:

1. More research on the pressing processes used to make graphite crucibles.
2. Research on the application and use of graphite mineral in fields other than foundry.
3. Additional research into materials that improve the mechanical strength of graphite crucibles made by slip casting.
4. Research on the possibilities of Bauchi graphite for grinding and polishing wheels

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