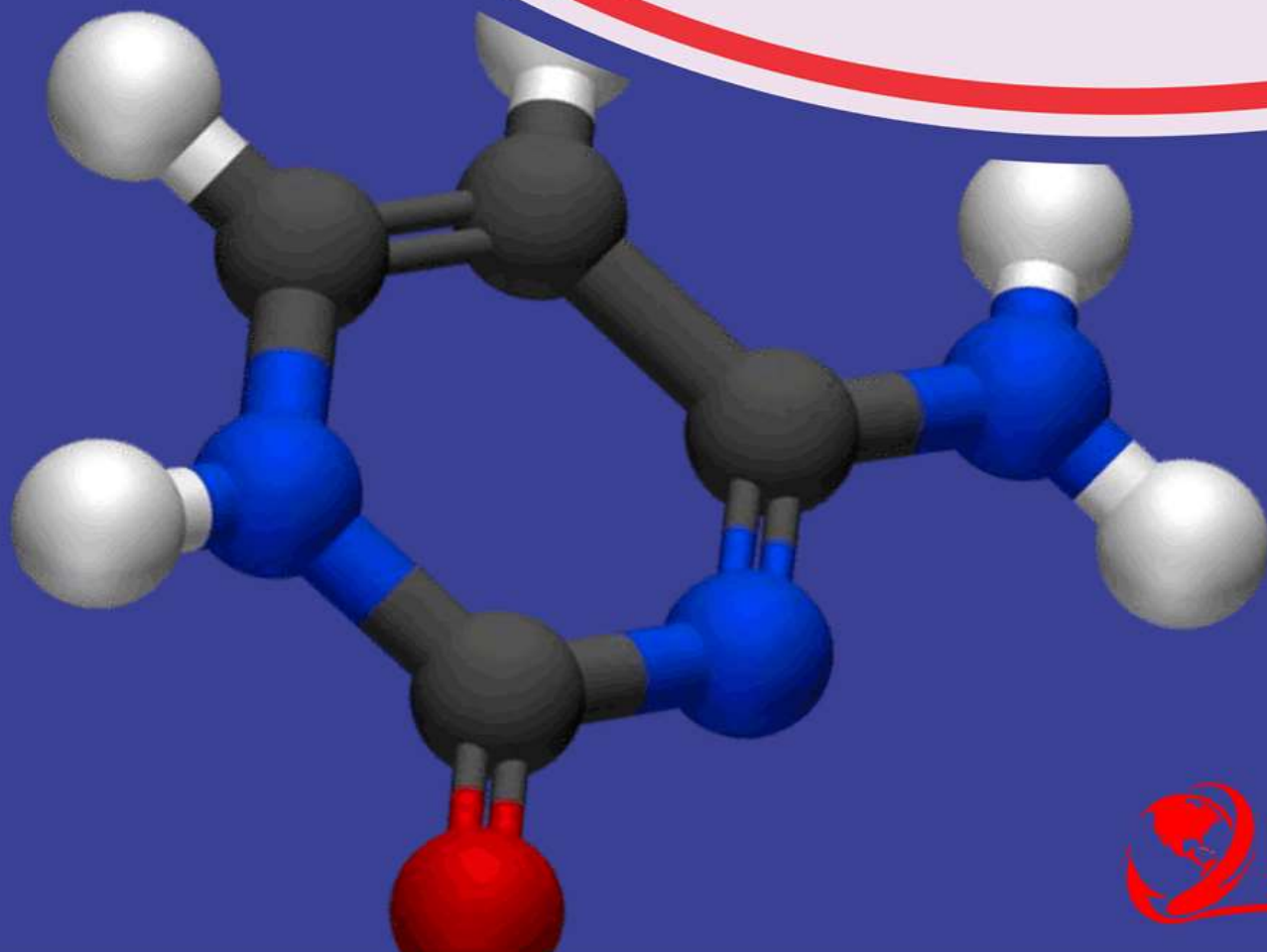


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(A Case study of Sama Borkno Graphite in Warji Local Government Area of
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Abstract

Purpose: This research work investigated the use of graphite as material for production of crucibles by using clay slip casting method.

Methodology: The graphite used was sourced from a graphite deposit located at Sama Borkno in Warji Local Government Area of Bauchi state. Clay, kaolin, fireclay silica, lime were used to bind the materials together and to make the slip more plastics and to strengthen the crucible. The investigation covered the assessment of chemical compositions of the graphite sample obtained, processing of the graphite obtained into crucible, determination of physical properties of crucible such as shrinkage, porosity, refractoriness, shock resistance and heat conductivity by ASTM standard methods.

Findings: The result of the chemical analysis for the graphite sample showed that the beneficiated graphite has enough percentage of carbon content suitable for crucible production. The shrinkage tests conducted on the samples showed percentage shrinkage of the sample ranges from 2-17 %. The results of the thermal shock tests showed that all the samples can withstand sudden change in temperature when exposed to different temperatures. The refractoriness test for the sample showed that all the samples can withstand temperatures above 1200⁰C. Conductivity test showed good heat conductance of the samples. After the tests, it was found that sample “B” possess all the requirements required for graphite crucible production. The Crucible was produced by slip casting forming method of ceramic bodies. The crucible produced was tested by melting a brass.

Unique Contribution to Theory, Practice and Policy: The study recommends that government should encourage the full time mining of indigenous graphite by providing a conducive atmosphere and assistance to miners. Some additives such as grog should also be added to the samples mixtures so as to improve the properties.

Keywords: *Graphite, Clay, Crucible, Kaolin, Silica, Lime*

INTRODUCTION

The present economic situation of Nigeria calls for the local sourcing of raw materials used in the production of engineering materials. Refractories are materials which can withstand high temperatures above 1580⁰C, the physical and chemical action of molten metal slag, and gases in the furnace without deformation, failure or change in composition under their own weight (Aiyedun *et al.*, 2012). Refractory being one of the materials largely used in metallurgical plants, foundries and power generating plants are at presently sourced by importation. The metallurgical industry alone accounts for 80% of refractory consumption. If these valuable inputs in the foreign industry are sourced locally, there will be great conservation in the foreign exchange earnings of the country.

Typical refractory materials found in Nigeria are: carbon (graphite and coke), clay, oxides like: alumina (Al₂O₃), magnesia (MgO), Zirconia (ZrO₂), non-oxides like carbides: Silicon carbide (SiC), boron carbides (B₄C): Nitrides (Si₃N₄), etc. The development of our local materials for the production refractories is justified by the need to meet the technological requirements of the country, and to conserve the much needed foreign exchange. Graphite is found naturally in the earth crust though it can be manufactured artificially from petroleum coke using electrical furnace. Graphite is a crystalline and most stable form of carbon that occurs as mineral. It can be used in many ways because of its excellent properties for applications where resistance to thermal shock, basic slag at high temperatures, resistance to corrosive chemicals and oxidants is needed (Nwobi, 2006). Therefore, they are used in production of crucibles. Basically, crucibles are free – standing vessels, used for high temperature operations such as metal smelting, melting and casting (Chirikure and Hall, 2013). Crucible is a cylindrical shaped container placed in the inner cavity of the furnace (Osarenmmwind, 2015). These crucibles are open mouth vessels made of graphite and ceramic materials such as clay. These qualities accounts for its effectiveness and high efficiency in graphite crucibles.

Experimental Materials

The graphite samples were collected from the graphite mining sites around Sama – Barkono / Dutsenhayar area of Warji local government of Bauchi state. The site was located with the help of local miners and the information obtained from the studies of Muhammed (2014) and Adewale (2009). The graphite samples were dug out from an abandoned mining pit which contains water collected from rainfall, which turned it into a small pond. Other raw materials used include: Clay, Kaolin, Plaster of Paris (P.O.P), spine oil, Kerosene, Magnesium oxides MgO, CaO, Chromium Oxides, Aluminium Oxides Al₂O₃, Sodium hydroxide NaOH, Distilled waters

Equipment

Ball mill Pascalleng Ltd 17949, Sieve shaker pascalleng.ltd 17747, Jaw crusher pascalleng Ltd 17769 freq.50hz, Pulverizer Altico Ltd, Flotation machine BJA E 19282/1 Type D12, Weight balance pc180, Drying oven Genlanwidnee England BJA E 19822/1 D12, Carbon Sulphur analyser CSA – 996, kick wheel, electric furnace.

METHODOLOGY

The graphite samples were crushed and ball milled, then coned and quartered to yield a representative sample.

Coning and Quatering

Crush materials is heaped into a cone by shoveling all the material to one point on the clean table in such a manner that the particles roll down in all directions and the composition of the mass are thus distributed as concentric layers of a cone. The top of the cone is then flattened with the edge of the shovel by spreading the material equally in all directions until a disc is formed. This disc is made into quadrants and the materials from the diagonally opposite quadrants are taken as sample. The materials in the other quadrants are removed and rejected. The mass now contains half the original quantity. The material so sampled is then further crushed and coning and quatering are repeated, thus reducing the quantity of the sample to one fourth of its original quantity. The cycle is repeated until desired quantity is obtained.

Sizing

The arrangement of the sieves was done using a sieve scale in which the ratio of the aperture width of adjacent sieves is the square root of two ($\sqrt{2} = 1.414$). Sieves sizes from $355\mu\text{m}$ to $180\mu\text{m}$ were arranged in a stack with the coarsest sieve on the top and the finest at the bottom. A tight fitting pan was placed below the bottom sieve to receive the final undersize and a lid was placed on top of the coarsest sieve to prevent escape of the sample. During the shaking, the undersize material falls through successive sieves until it is retained on a sieve having apertures which were slightly smaller than the diameter of the particle.

Beneficiation of the Sample by Flotation Technique Method

Sample of ground graphite, sieve size of $90\mu\text{m}$ was weighted up to 500g using the weighing balance. Distilled water of up to 1000cm^3 was measured and mixed with the graphite ore, poured into the flotation cell and agitated for 3 minutes at the speed of 2000rpm.

During the agitation process, three drops of regulator (sodium hydroxide) was added to it as a pH regulator and conditioned for seven minutes. Thereafter, 5ml of kerosene and pine oil added before the expiration of conditioning time.

Immediately after the conditioning / agitation, air was allowed to pass through the pulp at a reasonable rate and froth emerged and collected till barren froth surfaced.

This process was repeated and large quantities of the concentrates obtained. The concentrate and tailings dried, weighed and recorded. The chemical analysis was conducted on the representative graphite sample to determine the % carbon present before and after the beneficiation. The representative sample was subjected to XRF and Carbon – Sulfur Analyser, before and after the beneficiation, and the constituents in elemental form revealed and recorded in a table 3

The beneficiated graphite powder was divided into five portions and labeled as A, B, C, D, and E. Each portion is 200g graphite, with exemption of E which is 150g graphite. Portion A was then mixed with 150g of refractory clay, 10g of silica and 45g kaolin all in their dried and powdered form. Sample B was then mixed with 150g refractory clay, 10g of silica and 60g Of fire clay.

Portion C was mixed with 150g of refractory clay, 10g of silica, and 45g lime. Sample D was mixed with 150g refractory clay, 30g kaolin, and 30g lime. Sample E was mixed with 150g clay, 80g fireclay, and 20g silica. Each sample portion was then mixed with water and cast in 2cm x 3cm x 5cm mould and then allowed to dry at room temperature for seven days after which they were removed from the mould. Twenty five test samples were produced in this way i.e. five pieces for each sample. The rectangular bricks were then labeled as A, B, C, D and E. Preliminary tests conducted on the five sample mixtures confirmed that sample ‘B’ which is a combination of 50% graphite, 30% clay, 15% fire clay, and 5% silica, give the best properties. A model of a cylindrical crucible of size 10cm top and bottom external diameter, 7cm internal dia., 9cm height, 1.5cm thick, was produced from secondary clay by throwing method on a kick wheel. The model was then used to produce a mold of plaster of Paris. Therefore, Slip was formed by suspension of graphite, clay, fire clay, and silica in water. The suspension was then poured into a mold (made of plaster of Paris), water from the slip was absorbed into the mold, leaving behind a solid layer on the mold wall. As the cast piece dried and shrank, it was pulled from the mold. The crucibles produced in this way were then allowed to dry in open air for three days, followed by drying in oven for twelve hours at 110⁰C to expel any moisture left in the samples and to avoid cracks during firing. The crucible was then fired in electric heating furnace to 1200⁰C for eight hours. After firing, the crucibles were allowed to cool in the furnace at a cooling rate of 1⁰C/minute.



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



Plate XIV: Slip is poured in the plaster mold



Plate XV: Graphite crucibles produced

RESULT AND DISCUSSION

This section is the presentation of all the results and outcomes obtained from the analyses and tests carried out in this research. The analyses and tests conducted on the samples are: Chemical Analyses, Shrinkage Test, Thermal shock resistance Test, Porosity Test, Refractoriness Test, and Thermal Conductivity Test.

Results of Chemical Analyses

The chemical analyses were conducted at the Department of Mineral And Petroleum Engineering, Kaduna Polytechnic, and. XRF model X- MET5100, XMDS26700, and Carbon-Sulphur Analyser model Shadong CSA-996 were used. The results of the beneficiation of the graphite before and after floatation shows the carbon content for each and is presented in a table.

Results of the Physical Properties Test of the Samples

The shrinkage test, porosity test, thermal shock resistance test, conductivity test, and refractoriness test, were conducted at the Department of Civil Engineering of College of Engineering, Kaduna Polytechnic, Kaduna. The results of the tests mentioned above are presented in tables:

LOI = Loss on Ignition

LOD = Less than level Of Detection

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 ₂ O ₃	6.35	1.15	17.50
SiO ₂	49.14	14.70	75.11
SO ₃	<LOD	<LOD	<LOD
K ₂ O	5.70	4.59	0.05
CaO	3.09	1.80	0.91
TiO ₂	0.40	1.30	1.31
V ₂ O ₅	0.38	0.01	<LOD
Cr ₂ O ₃	0.06	1.03	<LOD
MnO	1.86	0.75	0.42
Fe ₂ O ₃	9.03	3.51	0.89
MgO	0.15	0.43	0.50
P ₂ O ₅	0	0	0.18
PbO	0.19	0.10	<LOD
Na ₂ O	< LOD	<LOD	<LOD
ZnO	0.42	0.63	< LOD
BaO	0.26	0.74	1.11
Total	99.9	99.9	99.9

Table 2: Result of the Analysis for Carbon and Sulfur Content in the Sample before Flotation

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

Table 3: Result of the Analysis for Carbon and Sulfur Content in the Sample after Flotation

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

The graphite sample analyzed using x – ray fluorescence in table 3 has the impurities (tailings) of high silica (SiO₂) which is 75.11% which can be used in the Portland cement production, as an additive, glass for doors and windows, and optical fibers for telecommunication. It can also be used for food and pharmaceutical applications.

The loss on ignition (LOI) is the organic or combustible or volatile matter loss on heating and this affect the values of shrinkage and porosity to some extent (Jock, 2013). The loss on ignition obtained after the graphite sample was analyzed is 2.23% which fall within the recommended range of 0.93 – 92.1% as stated by Bojko & Kabala (2001).

The recovery of graphite by floatation shows that the floatation method of beneficiation is efficient and also suitable for large scale production of pure graphite concentrate. Tailings recovered from the mineral gangue, will be a source of producing large amount of silica since it constituted about 79% Of the entire tailings.

From the results obtained for pre and post beneficiation, it can be concluded that the graphite is of good grade, since its purity before and after beneficiation falls within the range of high purity graphite.

Using carbon – sulphur analyser in tables 2 and 3 above, the percentage carbon obtained before beneficiation by froth floatation was 30% approx. and the percentage carbon obtained after beneficiation was 79% (approximately 80%). Therefore, the purity obtained is considered to be within the range of high purity graphite, because graphite containing 80 to 85% carbon is used for crucible manufacture. This is in line with the study of Muhammed *et al.* (2014).

Other tailings like AlO₃, TiO₂, BaO, which constitute about 20% of the tailings are also very useful industrial materials.

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 Sectional area mm ²	Cross 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 results of the percentage shrinkage of the sample in table 4 shows that the percentage shrinkage in sample “A” {50% graphite, 30% clay, 5% silica, and 15% kaolin} is 6.4%. And that of sample “B” {50% graphite, 30% clay, 5% silica, and 15% fire clay} is 7.8% its increase in shrinkage may be due to the addition of fire clay instead of kaolin. The percentage shrinkage in sample “C” {50% graphite, 30% clay, 5% silica, and 15% lime} is 16.8%. Sample “D” {50% graphite, 30% clay, 10% kaolin, and 10% lime} is very low which is 2% this may be due to addition of lime and kaolin. The increase in percentage shrinkage in sample “E” {40% graphite, 30% clay, 20% fireclay, and 10% silica} may be due to the reduction of graphite sample and increase in clay contents in the mixture. The relatively high percentage shrinkage in the samples indicates the problems of not adding grog in the clay – graphite mixture of refractory. Grog is a term used for fired clay particles. It is added mainly as an anti – shrinkage element in the form of angular particles of various sizes to achieve better interlocking of grains (Aigbodion, 2014).

The shrinkage of samples “A”, “B”, and “D” fall within the acceptable value of 4 – 10% as stated by Abubakar *et al.* (2014).

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

S/N	SAMPLE	Temperature °C	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

Thermal stability also called thermal shock resistance of refractory materials often decreases with increasing firing level, it is known that in practice product which retain their thermal stability even after high firing temperature or operating temperature are most useful in practice (Obikwelu, 2002). Thermal shock is the measure of refractory property when the refractory is exposed to alternate heating and cooling.

The thermal shock resistance of the samples was 28 cycles. Thermal shock resistance in table 7 was very good. The thermal shock resistances fall within the accepted range of 20 - 30 cycles (Abubakar *et al.*, 2014).

The results of the thermal shock resistance of the samples show that all the samples can withstand sudden change in temperature when subjected to sudden cooling after heating. All the samples showed a very slight crack formation in their bodies after undergoing 28 cycles. The good thermal shock resistance is due to the addition of the silica in the mixtures.

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 the refractory into which the molten metal, slag, fluxes vapour etc. can penetrate and thereby contribute to the eventual degradation of the structure. At higher apparent porosity, a refractory material is less resistant to eroding action of slag and solid particle which can penetrate through pores inside the materials (Obikwelu, 2006). Low porosity in refractory products improves mechanical strength and other properties of refractories. The lower the apparent porosity of refractory, the better. The result of the apparent porosity of the samples in table 8 indicates that samples “B” and “D” have lower porosity than the remaining samples. Low porosity and permeability is the property of a crucible of not allowing gases and liquids to pass through it easily (Nwobi, 2006).

The apparent porosity of sample “A” was 16.92% and that of sample “D” was 14.29%. These values were little above the recommended standard of 11 – 14% for clay – graphite bonded crucible. This is in line with the study of Aigbodion *et al.* (2014). Samples with lower apparent porosity will have good thermal conductivity. Porous refractories have high permeability, poor heat conductivity, low strength, and less sensitivity to temperature fluctuation (Muhammed, 2013).

Table 7: Result for the Refractoriness test of the Samples

Property	UNIT	A	B	C	D	E
Refractorines	⁰ C	16500	1700	1650	1500	1450

Refractoriness is the measure of fusibility of a refractory material and it shows the temperature at which the material softens or fused. Refractoriness is defined as the temperature corresponding to the moment when a material begin to lose its shape, melting point (Sadik *et al.*, 2013). Refractories are non- metal materials having those chemical and physical properties that make them applicable structures or component of system that are exposed to environment above 1000⁰C (Bolanle, 2011).

Table 8: Result for Thermal Conductivity of the Samples

SAMPLE	UNITS	A	B	C	D	E
Temperature	⁰ C	51	50	48	48	49
Temperature2	⁰ C	42	41	41	42	42
Thermal Conductivity	W/mk	1.49	1.49	1.16	0.99	1.16

Thermal conductivity is the measure refractory regarding its ability to conduct heat from the hot to the cold face when it is exposed to high temperatures. Thermal conductivity in refractories increases with temperature but generally decreases with increasing porosity. The thermal conductivity of refractory material depends on temperature, chemical composition of the raw material for refractory product, the mineralogical structure of the mix, true porosity and pore size, firing temperature.

The results of the thermal conductivity of the samples in table 10 indicate that samples “A” and “B” have the higher thermal conductivity of 1.49W/mk and 1.49W/mk respectively. Sample “C” and “E” also have same thermal conductivity of 1.16W/mk each. The sample with a lowest thermal conductivity is “D” with 0.99W/mk this may be due to the fact that silica was not present in the mixture. In the study of Kleiner *et al.* (1996) the thermal conductivity of the thin film of silicon dioxide (silica) was observed to increase with rising temperature. It is noted that samples “B” and “D” apparent porosity was little above the value of recommended standard of 11 – 14% for clay – graphite bonded crucible as stated by Aigbodion (2014), this means that with this value of porosity, they will have good thermal conductivity to transfer heat better than the other samples. Hence the relationship between apparent porosity and thermal conductivity can be clearly seen.

Conclusion and Recommendation

Conclusively, the utilization of indigenus graphite disclosed great prospect and potential which call for full – time sourcing and making of foundry products worthy of good price. The research concludes that Sama Barkono graphite is a good source of a refractory material used in the manufacture of crucible. The research showed that when graphite powder was mixed with some

readily available additives, a very good refractory mixture can be obtained. In the absence of isotactic press machine, slip casting method can be applied in the manufacture of clay bonded graphite crucibles. Froth floatation method is the best of graphite beneficiation, because the result of the analysis after beneficiation showed that the tailings and gangue materials removed cannot be achieved by other methods like chemical leaching and magnetic separation. Slight cracks on the surface of the crucibles occur when the sample are not properly and completely dried before firing . Based on the findings of this research, the following recommendations are made:

1. Government should encourage the full – time mining of indigenous graphite by providing a conducive atmosphere and assistance to miners.
2. Some additives such as grog should also be added to the samples mixtures so as to improve the properties.
3. Due to unstable electricity supply in the country, the use of kerosene or gas powered kiln for firing is hereby suggested.
4. Properly fitting tongs which apply little force should be used while taking the crucible out of the furnace.
5. Crucible should be properly annealed before being put into production.
6. Charge materials should not be packed tightly in crucible to allow space for expansion.
7. Direct contact of the crucible with a burner flame should be avoided.
8. Minimum quantity of flux should be used to satisfy metallurgical requirements.
9. Rapid heating of the crucible in the furnace should be avoided in order to check cracks.
10. Crucible should be dried properly and devoid of any moisture before being put to use.
11. The use of 2% silica or lime content in the mixture is hereby suggested.
12. Further study on the production of crucible using Bauchi graphite should be encouraged.

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