Optimization Of Clay-Bonded Graphite Crucible Using D-Optimal Design Under Mixture Methodology

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Abstract: The aim of this study is to optimize clay-bonded graphite crucible from the mixture of clay and graphite and selected additives (MgO and SiC) using D-optimal under the Mixture Methodology of the Design-Expert version 6.08. The samples were dried forced under the normal atmospheric condition for two weeks and were oven dried at 100^oC and later fired in a muffle furnace at 1100^0C. The samples were subjected to the following mechanical test bulk density, linear firing shrinkage, and apparent porosity. The results obtained shows that experimental Run 7 gave the least value of bulk density (1.80) with Clay (80 %), Graphite (10 %) and MgO and SiC (10 %) respectively but Run 9 gave the lowest apparent porosity with Clay (70 %), graphite (10 %) [MgO and SiC (10 %)]. Further investigation conducted indicated that Run 13 with percentage composition of 80 % clay, 10 % graphite, 5 % MgO and 5 % SiC gave the highest value of the bulk density of 2.91 and the least value of linear shrinkage of 2.15 and the apparent porosity value of 28.20. These values fall within the experimental range given by literature. Though by natural intuition and from the literature survey, run 14 would have been selected but based on values obtained, run 13 was confirmed and concluded upon to have a better composition as a result of the computer analysis using design expert considering the values obtained from the selected mechanical test.

Keywords: Bulk density, D-Optimal design, Graphite crucible, Optimization, Shrinkage.

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1.0 INTRODUCTION

Foundry crucibles are a refractory product used for melting iron, glass as well as other metals and alloys. There are various types of foundry crucibles depending on the constituent materials, these include clay crucibles, silicon carbide crucibles and graphite crucibles, among these, graphite-containing refractories, graphite crucibles are the most demanded locally (Nwobi, *et al.,* 2002). Furthermore, the crucible is a container that can withstand very high temperatures and is used for metal, glass, and pigment production as well as a number of modern laboratory processes. While crucibles historically were usually made from clay, they can be made from materials that can withstand temperatures high enough to melt or otherwise alter its contents (Rehen and Papakhristu, 2000). The important raw materials required for their manufacture are natural graphite and clays which occur abundantly in Nigeria (Aigbodion*, et al.,* 2014). However, crucible production requires a proportionate mixture of the various constituents plays a vital role. Response surface methodology (RSM) consists of a set of statistical methods that can be used to develop, improve or optimized products (Cihon, 2003). RSM typically is used in situations where several factors influence one or more performance characteristics or responses. RSM used to optimize one or more responses or to meet a given set of specification (e. g a minimum strength specification or an allowable range of values). There are three general steps to that which comprise: experimental design, modeling, and optimization (Gihan, 2013). In order to obtain a higher and homogeneous mixture of crucible composition, it is important that the optimal additives to be added to clay and graphite are investigated. Design of experiment is useful in the analysis and optimization of the mixture effects of several process variables influencing responses with a minimum number of experimental runs while varying the variables concurrently (Oladosu *et al*, 2016). In the mixture experiments, the measured response is assumed to depend only on the relative ratio of the components in the mixture and not on the amount of the mixture, compared to Taguchi and factorial methods (Anderson and Whitcomb, 2014). The focus of this study is therefore to investigate the influence of optimizing the mixture of clay, graphite and selective additives such as MgO and SiC on the production of crucibles with the view of developing an effective and suitable mixing ratio for clay bonded crucible production.

2.0 MATERIAL AND METHOD

2.1 Materials

Crude graphite ore used in this research was sourced from a mine site in Sama-Burkano village in Ningi Local Government Area of Bauchi state; about 20 km away from Bauchi along Kano road with a coordinate of 11^o 11$^'$ 0$^{''}$N, 9^o 45$^'$ 0$^{''}$E, and has an area coverage of about10,000 m^2 of land. The kaoline used was obtained from a site in Tsaragi village in Edu Local Government Area of Kwara State. Both samples were crushed, ground and pulverized before been beneficiated using froth floatation method.

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2.2 Sample Preparations

The weight of clay + kaolin + Silicon carbide + Magnesium Oxide as calculated were thoroughly mixed together using a turbulent mixer at a creed of the mixer. About 1% of water was added for proper plasticity. The mixture was carefully packed in the die where it was pressed. This was repeated for all the other composition. The pressed samples were air-dried for about two weeks, and then oven dried at 100 °C for 8 days. And finally fired at 1150 °C for proper sintering. The essence of air-drying and Oven drying was to remove all the moisture in the pressed samples. Before the sintering, the dimensions of all the samples were taken (i.e. the height and diameter) using digital Vernier caliper. This was also repeated after sintering.

2.3 Bulk Density (%)

Bulk density is the weight of a unit volume of loose material (such as powder or soil) to the same volume of water, expressed in kg/cm^3. It is defined as the mass of the particles of material divided by the total volume occupied. The total volume includes particle volume, inter-particle volume, inter-particle void volume, and internal pore volume. Bulk density is an intrinsic property of a material, it can change depending on how the material is handled. The Dried weight which is the weight after firing (DW) was taken. Each of the samples was suspended in a known volume of water in a measuring cylinder where the suspended weight was taken as (SSW). In that cylinder, the volume of water displaced is recorded. The dried weight divided by the volume of water displaced is known as the bulk density of the samples (Thomas *et, al*. 2014).

Bulk Density (g/cm^3) = $\frac{Mass}{Volume\ displaced} = \frac{DW(g)}{Vol.displaced(g/cm3)}$ (1)

2.4 Apparent Porosity (%)

Apparent porosity refers to the amount of void (or pores) within a volume of sediment or porous solid. The value of porosity is measured as a fraction or percentage of the true porosity, this includes the volume of the sealed pores, the ratio of the volume of open pore space in a specimen to the exterior volume. The samples were soaked in boiling water for 30 minutes, after which they were removed from the water, cleaned. Their weight was taken as soaked and suspended soaked weight respectively [SW and SSW (g)], where there is suspended soaked weight (Sugwon *et. al,* 2009). Apparent porosity is calculated from the expression

$$.A = \frac{SW - DW}{SW - SSW} X100 \qquad (2)$$

2.5 Linear Shrinkage (%)

The difference between the initial heights of the samples taken and the final heights after sintering expressed in percentage is the linear shrinkage.

Linear Shrinkage (%)

$$S = \frac{Initial\ height - Final\ height}{Initial\ height} X100 \qquad (3)$$

2.6 Volume Shrinkage (%)

From the initial height of the samples and the final height after sintering, the volumes of the cylindrical samples were calculated, using the initial diameter and final diameter of the cylindrical samples after sintering.

Volume Shrinkage (%)

$$S = \frac{Initial\ volume - Final\ volume}{Initial\ volume} X100 \qquad (4)$$

Volume of a cylinder = $A = \pi r^2$h

2.7 Water Absorption (%)

$$W = \frac{SW - DW}{SW} X100 \qquad (5)$$

2.8 Experimental Design

D-optimal design under the mixture methodology was employed to optimize the mixture and process factor. The design was performed using design expert software (6.0.8). The minimum and maximum of levels of the component (Graphite, Clay, SiC, and MgO) were the range 0-100% while the factor levels were in the range of 1- 4mm (Table 1). The software then gave experimental runs which mixed the additive in various percentage but all aggregated to 100% and at each experimental run for the various particle sizes (Oladosu *et al*, 2016).

Table 1 *Component factor and their levels for D-optimal design*

Component	Name	Level (Low)	Level (High)
A	Clay	70	85
B	Graphite	10	25
C	MgO	0	10
D	SiC	0	10
Factor	Unit	Low	High
Particle size	mm	1.00	4.00

2.9 Statistical Analysis

The data (response) obtained from the mixed ratio of graphite, kaolin, MgO and SiC were analyzed statistically using facilities embedded in the Design-Expert Software version 6.0.8 Stat-Ease Inc. (Vaughn, et al, 2000). The quality of the fit of the model was evaluated using the test of significant and Analysis of variance (ANOVA).

2.10 Optimization Procedure

Based on the process of investigating maximum optimization of the mixing ratio from kaoline, graphite, and selected additive, an algorithm was prepared and translated to design expert software version 6.0.8 version, the flow chart upon which the algorithm was based is shown in figure 1.

1. Collection of Graphite and Kaoline from a local deposit site in Kwara and Bauchi states respectively, Nigeria.
2. The raw materials were beneficiated and characterized for optimum yield, they were also sieved to various particle sizes of about 90µm.
3. An additive such as Magnesium oxide and Silicon Carbide were obtained from the manufacturer and all equipment was well calibrated before use.

4. Clicking D-optimal design under mixture methodology using design expert software version
5. Selection of the number of mixed components and enter (Clay, Graphite, MgO, and SiC) and their corresponding levels (low and high) in the dialog box provided.
6. Selection of the numerical factor and enter particle sizes and its level:
7. Selection of the number of response for both mixture and process factor and then click continue;
8. Entering the response data obtained in the design layout view.
9. Clicking on the analysis and follow the steps displayed at buttons across the top of the view as follow;
10. Clicking on square root under transformation;
11. Clicking on the fit summary to evaluate mixture process crossed model;
12. Clicking model order and desire term from the list;
13. Clicking on analysis of variance ANOVA to analyze the chosen model and view results;
14. clicking on diagnostic to model evaluate fit and transformation choice with the graph;
15. Clicking on a model graph to interpret and evaluate the model (Oladosu *et al*, 2016).

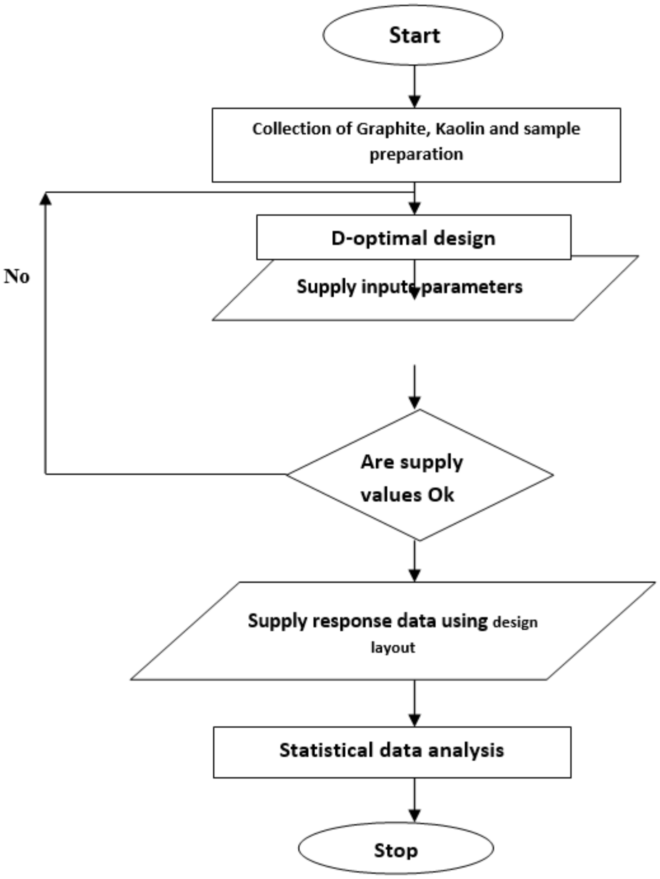

Figure 1. Flowchart showing the operational procedure for the mixture of Graphite, Kaoline, and other additives

3.0 Results and Discussion

Table 2: Result of D-optimal design composition by percentage

Run	Component 1. A: clay (%)	Component 2. B: Graphite (%)	Component 3. C: Magnesium oxide	Component 4. D: Silicon Carbide	Response 1. Bulk Density	Response 2. Firing Shrinkage	Response 3: Apparent Porosity
1.	70.00	20.00	0.00	10.00	1.89	2.29	28.18
2.	70.00	25.00	5.00	0.00	1.88	2.24	34.86
3.	70.00	10.00	10.00	10.00	1.91	2.77	33.76
4.	73.33	13.33	3.33	10.00	1.91	2.50	29.02
5.	75.00	25.00	0.00	0.00	1.90	2.17	30.75
6.	85.00	10.00	5.00	0.00	1.89	2.48	34.82
7.	80.00	10.00	10.00	0.00	1.80	2.21	38.70
8.	70.00	20.00	10.00	0.00	1.87	2.68	33.80
9.	80.00	10.00	0.00	10.00	2.89	2.50	29.00
10	85.00	10.00	0.00	5.00	1.90	2.84	30.00
11	76.36	16.36	3.64	3.64	1.88	2.76	33.33
12	73.33	13.33	10.00	3.33	1.86	2.63	38.24
13	80.00	10.00	5.00	5.00	2.91	2.15	28.20
14	70.00	20.00	5.00	5.00	2.89	2.21	30.07
15	85.00	15.00	0.00	0.00	1.90	2.67	32.81

Responses

Response1: firing shrinkage
ANOVA for mixture reduced quadratic model.
Analysis of variance table (partial sum of the square)

Source	*sum of square*	*DF*	*mean square*	*f value*	*pro>f*
Model	0.46	4	0.12	6.26	0.0087 **Significant**
Linear mixture	0.16	3	0.052	2.84	0.0922
AC	0.31	1	0.31	16.53	0.0023
Residual	0.18	10	0.018		
Corresponding total	0.65	14			

The model F-value of 6.26 implies the model is significant, there is only a 0.87% chance that a model f-value this large could occur due to noise. Value of prob>f less than 0.0500 indicate model terms are significant. In this case linear mixture component.AC are significant model terms A value greater than 0.1000 indicates that the model terms (not counting those required to support hierarchy).

Final Equation in Terms of Actual Components:
Firing Shrinkage =+3.38456E-003A - 0.58707 B + 0.042118 C +0.38538 D +8.98451E-003AB - 2.10442E-004AC- 6.76485E-003 AD+ 0.024666 BC- 0.099317 BD +0.12915 C D- 2.36279E-004ABC+1.57651E-003ABD -1.35960E-003 AC D-2.49196E-003 BC D (9)

Where A=Clay; B= Graphite; C=Magnessium Oxide; D= Silicon Carbide and AC= Clay and MgO.

Response 2: Apparent porosity

Analysis of variance table (partial sum of the square)

Source	**sum of the square**	**DF**	**mean square**	**F value**	**prob>F**	
MODEL	189.94	3	63.31	5.87	0.0121	**Significant**
Linear mixture	189.94	3	63.13	5.87	0.0121	
Residual	118.64	11	10.79			
Corresponding total	308.58	14				

The model F-value of 5.87 implies the model is significant. There is only a 1.217% chance that a model f-value this large could occur due to noise. Value of prob>f less than 0.0500 indicate model terms are significant. In this case linear mixture component.AC are significant model terms A value greater than 0.1000 indicates that the model terms (not counting those required to support hierarchy).

Final Equation in Terms of Actual Components: Apparent porosity = +0.33651 A_1 +0.23955 B_2 +0.88281 C_3 -0.094294 D_4

Where A_1 =Clay; B_2 = Graphite; C_3= Magnessium Oxide and D_4 = Silicon Carbide.

The diagnostic plots of Studentized Residuals vs Normal percentage probability for all responses are shown in figure 1-3 to show the adequacy of the model.

Response 3: Bulk density
ANOVA for mixture reduced quadratic model
Analysis of variance table (partial sum of the square)

The model F-value of 7.27 implies the model is significant, there is only a 0.55% chance that a model f-value this large could occur due to noise. Value of prob>f less than 0.0500 indicate model terms are significant. In this case linear mixture component. BC CD are significant model terms A value greater than 0.1000 indicates that the model terms (not counting those required to support hierarchy).

Final Equation in Terms of Actual Components: Bulk density =+0.019329A+0.017759B+3.96704E-003C+0.017440D+5.51234E-004B C+1.13121E-003C D The model shows that the entries for both the firing shrinkage and the apparent porosity are significant but from Table 7,it was discovered that Run 8 with 70% clay,20% graphite, 10% MgO and 0% SiC has the highest shrinkage value of 2.97% while Run 13 with the minimum value of 2.15% contains 80% clay,10% graphite,5% MgO and 5% SiC possesses the lowest shrinkage value, but a suitable foundry crucible should shrink less under the action of heat and pressure (Aigbodion *et.al* 2014). Based on the results obtained from the determination of the apparent porosity, it shows that Run 7 has the highest value of 38.70 (80% clay, 10% graphite, 10% MgO and 0% SiC) while Run 9 has the least value of Apparent porosity with 27.00 (80% clay, 10% graphite, 0% MgO and 10% SiC) and previous discoveries have equally shown that a good refractory device must be less porous (Jong *et al*., 2013). But the values and results from runs in the results could not be used to justify the choice for optimization. Further investigation was therefore carried out to determine the optimum mixture using bulk density.

CONCLUSION

In conclusion, as a result of the further investigation carried out using bulk density, the followings were observed:

i) Run 13(with 80% clay,10% graphite,5% MgO and 5% SiC) gave the highest value of bulk density and least shrinkage and was considered as the optimum mixture or blend followed by Run 14,
ii) Run 7 gave the least value of bulk density.
iii) Finally, computer analysis using designed expert, taking into cognizance the value obtained for linear shrinkage, the bulk density and apparent porosity, which are within the standard range affirmed run 13 to be the best blend for graphite crucible production.

It is hereby concluded that Run 13 with the highest bulk density and the least shrinkage value was therefore considered as the optimum mixture since it is the only run whose values of the investigated property fulfilled the characteristics of a good foundry crucible. Meanwhile, by natural intuition and literature survey Run 14 was suggested (Richards, 1997), but based on the above computer analysis using design expert option 13 was considered and affirmed.

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APPENDICES

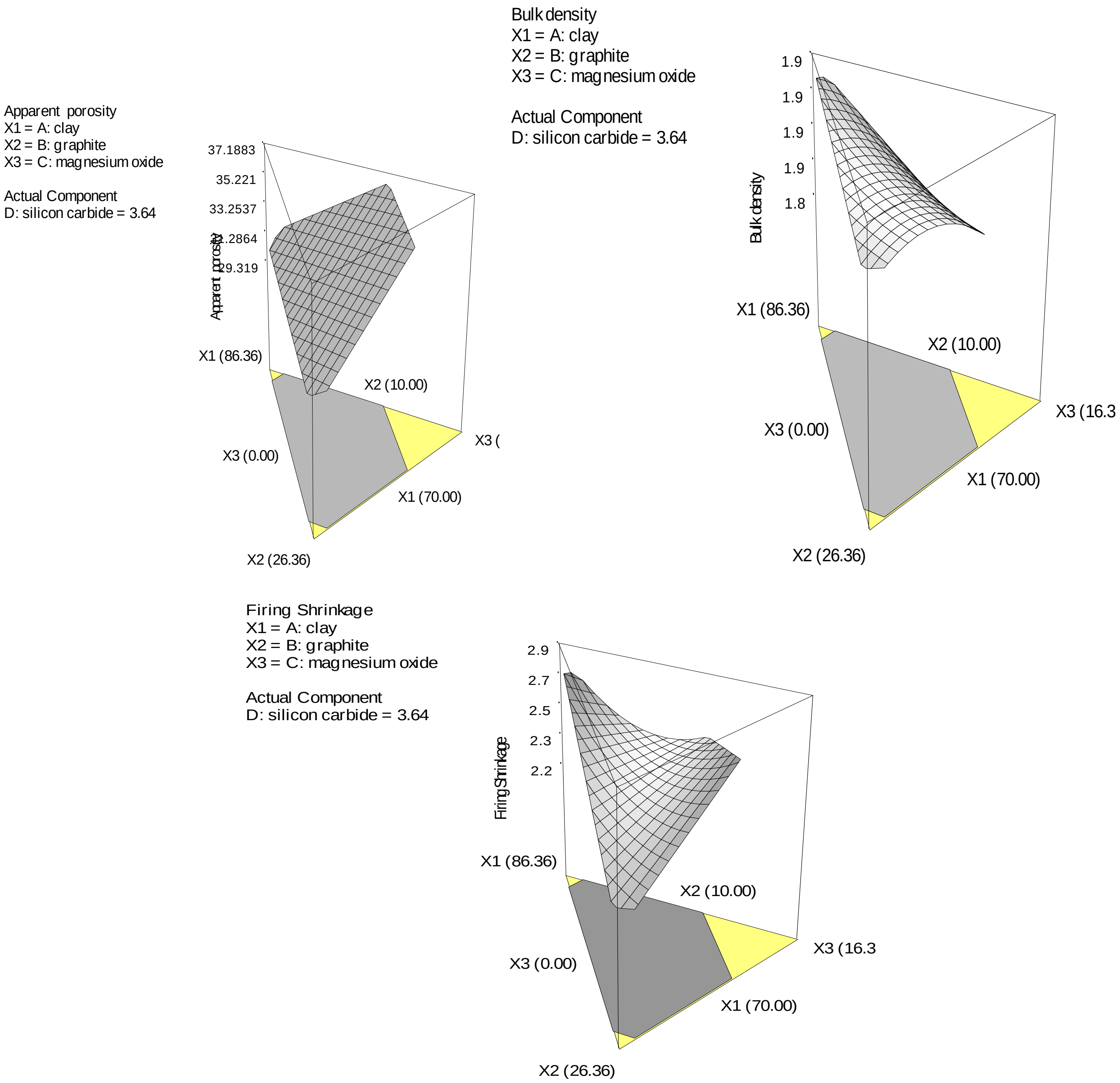

***Figure 3** Response Surface Plots Apparent Porosity, Firing shrinkage and Bulk density.*

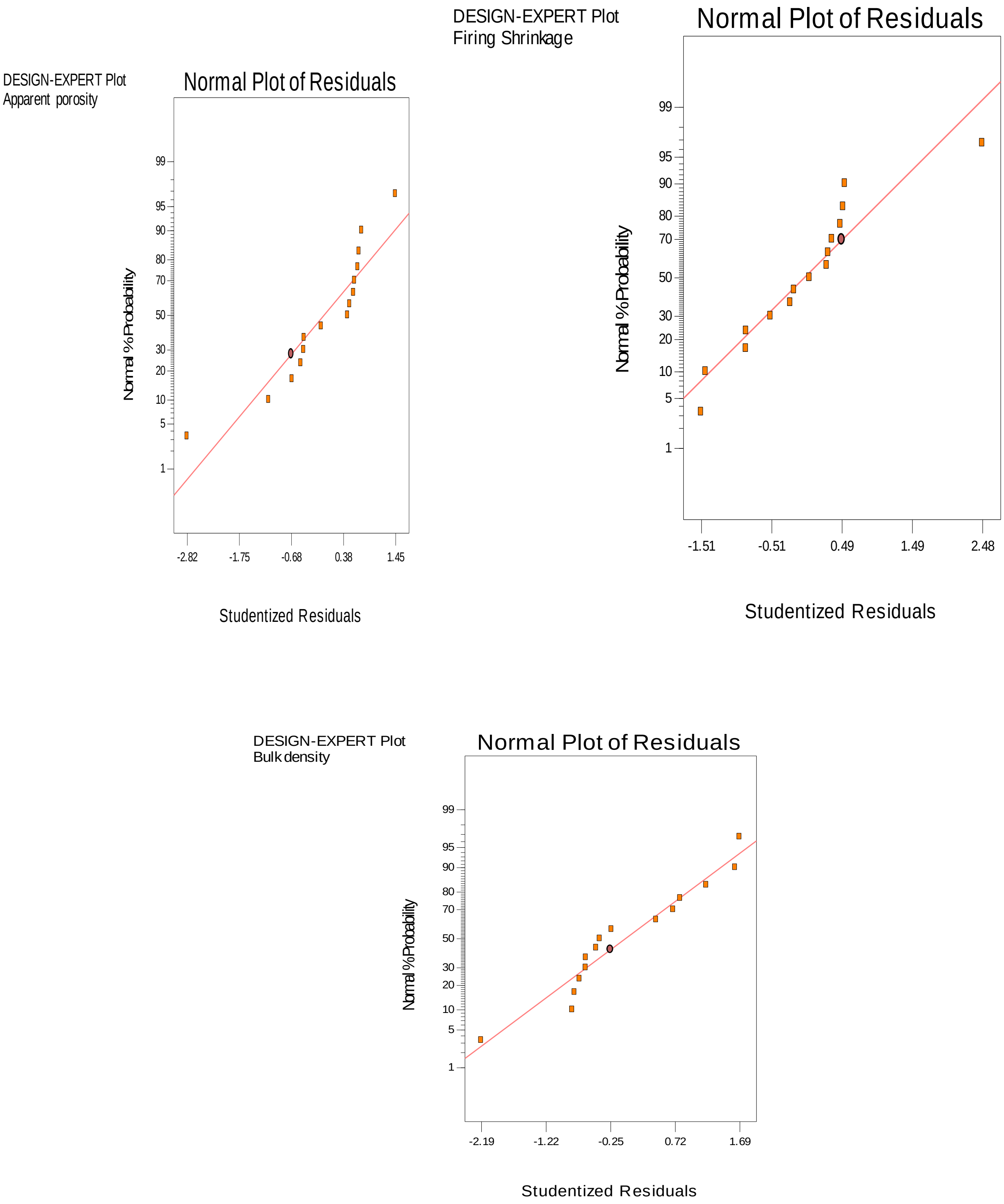

Figure 4: *Response Surface Plots Apparent Porosity, Firing shrinkage and Bulk density*