# USE OF SIMPLEX METHOD IN DETERMINATION OF OPTIMAL RATIONAL COMPOSITION OF TITLES PRODUCIBLE FROM NIGERIAN CLAYS

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**ABSTRACT**

*Using Simplex method and Scheffe's experimental design, the bending strength, water absorption capacity and other properties of titles from two Nigerian clays were investigated for the whole range of imposition recommended in literature. One of the days from Nsu was found suitable for tile production and the region of application was indicated. The optimal rational composition was found to be: Nsu Clay = 47.8%, quartz = 33.7% and $CaCO_3$ = 18.5%. The other clay from Ukpor was found unsuitable at the firing temperature (l000°C) used. It showed bending strength lower than the standard requirement for all compositions studied. To improve the strength an increase in firing temperature is recommended.*

## INTRODUCTION

Clays have been object of interest to researchers both in Nigeria [1 - 3] and abroad [4 - 6]. Each researcher has always gone for the properties that interest him most. Publications therefore abound in the area [1-12] and books [13, 14] and theses [15, 16] have been written. Yet research on clays continues. More deposits are being discoverer: and investigated. In USA and Europe, work on identification and application of clays abound in the thirties and even up to sixties [4, 2,5,17]. In Nigeria, some work has been done towards identification of her clay deposits and their possible applications [1 - 3]. Yet many of the Nigerian deposits remain unidentified. Even some of those identified have not been systematically studied. The author has therefore taken it upon himself to characterize and systematically investigate the possible application of some known clay deposits in Nigeria. The present work is a result of such effort and the result obtained will be both of interest both to industrialists and researchers.

Two clays from Ukpor in Anambra State and Nsu in lmo State, Nigeria which have been identified in earlier work [2, 3] are subjected to further investigation to ascertain their suitability for the production.

First the rational composition of the clays was calculated based on their chemical composition. Then the prescription for the tile production was established for a composition falling within the range recommended in literature [18]. Then followed a systematic an; detailed investigation of the region using simplex method of experimental design. The plan of experiment was that recommended by H. Scheffe [19]. From the result of the experiment, the coefficient of the regression equation was calculated and isolines plotted by means of computer from the result the region of composition satisfying tile standard requirements was recommended The Optimal rational composition was found together with the corresponding optimal properties.

## THEORY

Tiles: Definition, Types and Means of Production

The term *tile* is used for white fine ceramic ware with high porosity. Their water absorption capacity is between 8 to 22%. The surface of tiles gets dirty easily. For this reason, tiles are glazed

According to their composition, tiles are of different types: the hard (feldspar type); the soft (carbonate type), the mixed and clay tiles.

Their rational composition is given in Table I [18]

TABLE I - RATIONAL COMPOSITION OF VARIOUS TYPES OF TITLES

| COMPOSITION | FELDSPAR | CARBONATE | MIXED | CLAY |
|---|---|---|---|---|
| clay substance | 40-60 | 35-55 | 45-55 | 75-85 |
| feldspar | 5-10 | - | 3-4 | - |
| quartz | 30-50 | 30-45 | 35-50 | 15-25 |
| carbonate ($C_aC0_3$ dolomite etc) | - | 5-20 | 3-10 | - |

Feldspar (hard) tiles, in comparison with other types of tiles, are fired at highest temperature. They have the lowest water absorption capacity, and the highest mechanical indices: compressive strength 100-120 MPa, and Bending strength 12- 25MPa.

In carbonate (soft) tiles, alkaline earths carbonates replace feldspar. Firing is done at lower temperature ($1000^0$C - $1150^0$C). In comparison with other type, this tile has the highest porosity-20-22 %. As a result of this its mechanical properties are lower. It is however a lot lighter, has brighter colour, and the glazing adheres better to the surface because of its high CaO content.
In composition, firing temperature, and technical indices, mixed tiles take average position between the hard and the soft.

Clay-type tiles is the oldest. In essence it is modified potteryware. It is fired at 1000°C and has low indices.

The type of quartz material used in tile production is not of great significance since at the comparatively low firing temperature quartz almost does not react. Of great significance to the property of tile however is accurate control of the fineness of the grinding of the quartz. By regulating the fineness, one can have effective influence on the plasticity, shrinkage, coefficient of thermal expansion and the adherence of the glazing to the surface of the tile.

Firing of tiles can be done as follows: First firing (called biscuiting) to be done at lower temperatures (800-900°C), while glazing to be done at higher temperatures (1200-1300°C). More often, however, the opposite is practiced: first firing is done at high temperature while the second-glazing is done at lower temperatures. This gives the opportunity to utilize low melting glazing materials.

The porous surface of tile is hygroscopic. It gradually absorbs moisture and is partially hydrated. This leads to swelling of the surface. The glazing is subjected to tension and can crack. This phenomena is called wet expansion [8]. The process is slow. It is assumed that wet expansion is dangerous if it is more than 0.07%.

It is established [18], that wet expansion depends more on the composition, than on porosity and firing temperature. Tiles containing more quantities of alkalis (feldspar), tend to show wet expansion. In them during firing, compounds of zeolite type are formed which during wetting increase their volume. With addition of materials containing alkaline earth oxides (CaO, dolomite) the formation of zeolite is hindered and wet expansion is reduced to safe limits.

Carbonate (soft) and mixed tiles, which contain alkaline earth oxides show little wet expansion. This is their advantage over feldspar tiles. Tiles are stable towards deformation because of small amount of glass phases in their surface. Tiles are used for lining internal walls of buildings and are usually of size 150 x 150 mm.
The technological flow diagram [18] for tile production is as shown in FIG I:

**SIMPLEX METHOD [20]**
When studying the properties of a q-

component mixture, which are dependent on the component ratio, the factor space is a regular (q-l) simplex, and for the mixture the relationship holds:

$$\sum_{i=1}^{q} x_i = 1 \quad (1)$$

Where $X_i \geq 0$ is the component concentration, q is the number of components.

For binary system, (q=2) the simplex of dimension 1 is a straight line segment, and at q=3, the regular 2-simplex is an equilateral triangle with its interior. Each point in the triangle corresponds to a certain composition of the ternary system and conversely each composition is represented by one distinct point.

In designing the experiment to attack mixture problems involving composition-property diagrams, the property studied is assumed to be a continuous function of a certain argument. With a sufficient accuracy the function can be approximated by a polynomial. When investigating multicomponent systems the use of experimental design methodology substantially reduces the volume of an experimental effort. Further, this obviates the need for a spatial representation of complex surfaces as the wanted properties can be derived from equations, while the responsibility to graphically interpret the result is retained.

As a rule, the response surfaces in multicomponent systems are very intricate. To describe such surfaces adequately, high degree polynomials are required and hence a great many experimental trials. A polynomial of degree n in q variable has $C^n_{q+n}$ coefficients

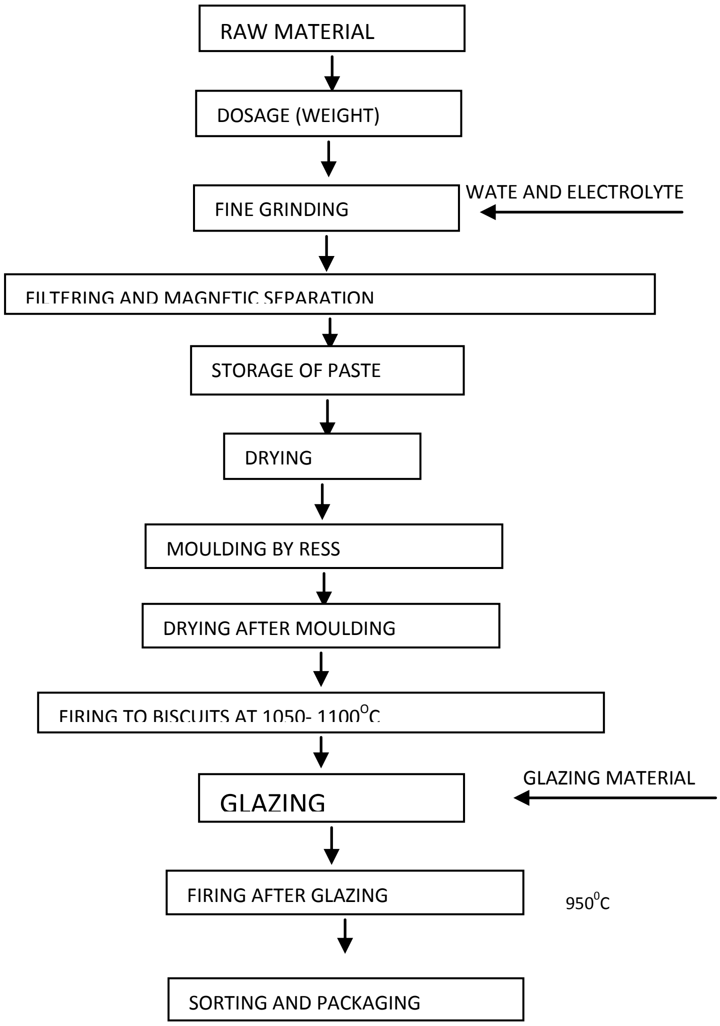


FIG.1 Flow diagram for tile production

$$\hat{Y} = b_0 + \sum b_i x_i + \sum b_{ij} x_i x_j + \sum_{1 \leq i \leq j \leq k \leq q} b_{ij} x_i x_j x_k \sum b_{i,} \dots \dots \dots i_n x_{i1} x_{i2} \dots \dots x_{in} \quad (2),$$

The relationship

$$\sum_{i=1}^{q} x_i = 1$$

enables the qth component to be eliminated and the number of coefficients to be reduced to $^{c}n_{q+n1}$

Scheffe [19] suggested to describe mixture properties by *Reduced Polynomials* obtained from equation (2) subject to normalization condition of equation (1) for a sum of independent variables. The reduced second degree polynomial in three variable is given by:

$$\hat{Y} = \beta_1 x_1 + \beta_2 x_2 + \beta_3 X_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \quad (3)$$

While for the same polynomial in q variables we have

$$\hat{Y} = \sum_{1 \le i \le q} \beta_i \times_i + \sum_{1 \le i \le q} \beta_i \times_i \times_j$$

(4)

The reduced incomplete third degree polynomial for ternary mixture has the-form [20]:

$$\hat{Y} = \beta_1 \times_1 + \beta_2 \times_2 + \beta_3 X_3 + \beta_{12} \times_1 \times_2 + \beta_{13} \times_1 \times_3 + \beta_{23} \times_2 \times_3 + \beta_{123} \times_1 \times_2 \times_3 \quad (5)$$

and for q-component mixture

$$\hat{\mathbf{Y}} = \sum_{1 \le i \le j \le k \le q} \boldsymbol{\beta}_i \times_i + \sum_{1 \le i \le j \le k \le q} \boldsymbol{\beta}_{ij} \times_i \times_j + \sum_{1 \le i \le j \le k \le q} \boldsymbol{\beta}_{ijk} \times_i \times_j \boldsymbol{X_k} \quad (\mathbf{6})$$

The non-linear part of the polynomial is called the synergism it gives a higher response as compared with that of the linear part of the equation, and the antagonism if it gives a lesser response.

So the term in the second-degree polynomial is termed the quadratic coefficient of binary synergism of the component i and j.

## SCHEFFE'S SIMPLEX-LATTICE DESIGN

The minimum number of points required to determine the coefficients of a polynomial of degree n in q variables is equal to $C^n_{+q+n+n-1}$.The simplex-lattice design proposed by H. Scheffe [21,22] provides a uniform scatter of points over the (q-l)-simplex. The points form a (q,n)-lattice on the simplex, where q is the number of mixture components, n is the degree of the polynomial. For each component there exists (n+1) similar level $X_i$ = 0, 1/n, *2/n,…*1, and all possible combinations are derived with such values of component concentration.

The design matrix for the simplex (3,2) and (3,3) are shown in Table 2 and Table 3

## EXPERIMENTAL

Materials Used: Nsu and Ukpor clays were obtained from Imo and Anambra States of Nigeria respectively. Quartz sand was washed and $CaCO_3$) was fresh.

Procedure: Two sets of Tiles were produced using Nsu Clay, sand and $CaCO_3$); and Ukpor clay, sand and $CaCO_3$. Grinding was wet and carried out in a ball mill maintaining the ratio: clay: water: Milling balls = 1:1:3:5. after milling for about three hours samples were taken for sieve analysis (The remain sieve of 0.06mm must be below 5 % for the material to be accepted as being milled properly).

The tiles samples (plates were prepared as follows:

Milled sampled was dried both in Gypsum mould and in dried both in Gypsum mould and in drier at a temperature of 100°C. The dry sample was passed through 2mm sieve, moistened 6-8 and kept in closed polytene bag for 24 hours for homogenisation of moisture. Then followed formation (moulding) of the tile plates by press at 25MPa. Tests were carried (out on some of the plates to ascertain their bending strength (in raw state). The rest of the samples were fired at 1000°C. On heating, the samples were held at 350°C for 1/2 hour for dehydration; at 600°C for 1/2 hour for transformation of $\beta \rightarrow \alpha$ quartz; at 800°C for 1 hour for final dehydration and decarbonation. On reaching 1000°C, final holding was done for 1/2 hour.

The samples were subjected to the following tests:

1. Bending Strength in raw state, $Y_1$,2.Bending Strength after firing, $Y_2$
2. Water absorption capacity, $Y_3$) and 4. Shrinkage, $Y_4$

## EXPERIMENTAL DESIGN

The experimental design used was that of incomplete polynomial of third degree. The model has the form;

$$\hat{Y} = \beta_1 \times_1 + \beta_2 \times_2 + \beta_3 X_3 + \beta_{12} \times_1 \times_2 + \beta_{23} \times_2 \times_3 + \beta_{23} \times_2 \times_3 + \beta_{13} \times_1 \times_3$$

The composition $X_1$ $X_2$, $X_3$ in their natural units are:

| | | |
|---|---|---|
| $X_1=1$ <br> $(X_2=X_3=0)$ | $\rightarrow$ | Clay = 55% <br> quartz = 40% <br> $CaCO_3$= 5% |
| $X_1=1$ <br> $(X_2=X_3=0)$ | $\rightarrow$ | Clay = 55% <br> quartz = 30% <br> $CaCO_3$= 20% |
| $X_3=1$ <br> $(X_1=X_2=0)$ | $\rightarrow$ | Clay = 35% <br> quartz = 45% <br> $CaCO_3$= 20% |

The area under investigation as represented in the simplex is shown in Fig. 2. The design matrix used for the experiment is given in Table 4.

**RESULTS AND DISCUSSION**

The result obtained for the properties studied: $Y_{1,}Y_2$, $Y_3$, $Y_4$ are shown in Table 5.

The coefficients of regression equation was calculated with the aid of computer and the isolines were plotted for all the properties studied. The results of the changes in the properties with respect to composition are shown in Fig. 3 to Fig. 6. The optimal rational composition is defined as the region of composition possessing the following properties:

Bending strength after firing>11.0 MPa

Water absorption capacity < 22 %

Applying the above specification to the properties studied the optimal compromise region is shown by the shaded area in Fig. 7 for tiles from Nsu clay.

TABLE 4 – DESIGN MATRIX FOR INCOMPLETE THIRD DEGREE POLYNOMIAL

| N | $X_1$ | $X_2$ | $X_3$ | Y | N | $X_I$ | $X_2$ | $X_3$ | Y |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 1 | 0 | 0 | $Y_I$ | 5 | 1/2 | 0 | ½ | $Y_{13}$ |
| 2 | 0 | 1 | 0 | $Y_2$ | 6 | 0 | 1/2 | ½ | $Y_{23}$ |
| 3 | 0 | 0 | 1 | $Y_3$ | 7 | 1/3 | 1/3 | 1/3 | $Y_{113}$ |
| 4 | 1/2 | 1/2 | 0 | $Y_{12}$ | | | | | |

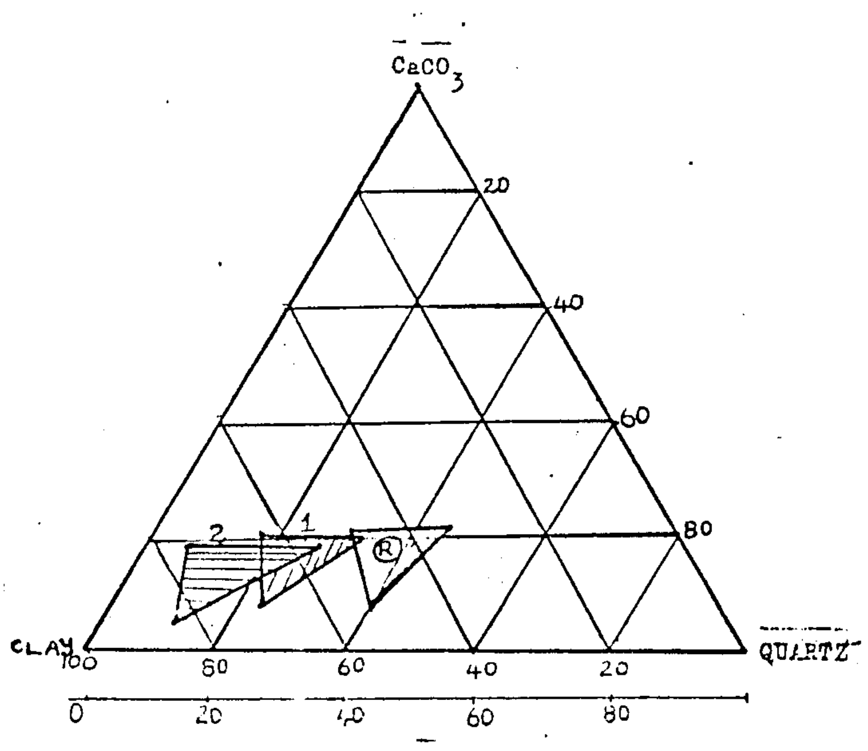


**Fig. 2**

TABLE 5- THE PROPERTIES OF TILES OBTAINED FROM NSU CLAY (NO.1) ANI> UKPOR CLAY (NO.2).

| CLAY1 | CLAY | $Y_1$, MPa | | $Y_2$, MPa | | $Y_3$, MPa | | $Y_4$, MPa | |
|---|---|---|---|---|---|---|---|---|---|
| | | 1 | 2 | 1 | 2 | 1 | 2 | I | 2 |
| 1.1 | 2.1 | 1.82 | 1.31 | 14.9 | 6.8 | 18.29 | 21.0 | -0.58 | -0.78 |
| 1.2 | 2.2 | 1.68 | 1.37 | 8.5 | 5.5 | 22.53 | 19.3 | +0.l3 | -0.5 |
| i.3 | 2.3 | 1.86 | 1.34 | 13.3 | 10.8 | 20.95 | 20.9 | +0.19 | -0.64 |
| 1.4 | 2.4 | 2.15 | 1.36 | 13.0 | 10.2 | 20.36 | 21.4 | -0.3 ) | -0.81 |
| 1.5 | 2.5 | 2.18 | 1.28 | 11.0 | 7.0 | 20.71 | 18.79 | -0.22 | -0.83 |
| 1.6 | 2.5 | 1.81 | 1.34 | 11.1 | 9.8 | 21.31 | 21.44 | +0.31 | +0.19 |
| 1.7 | 2.7 | 1.84 | 1.53 | 11.0 | 8.4 | 21.6 | 19.19 | -0.06 | -0.31 |

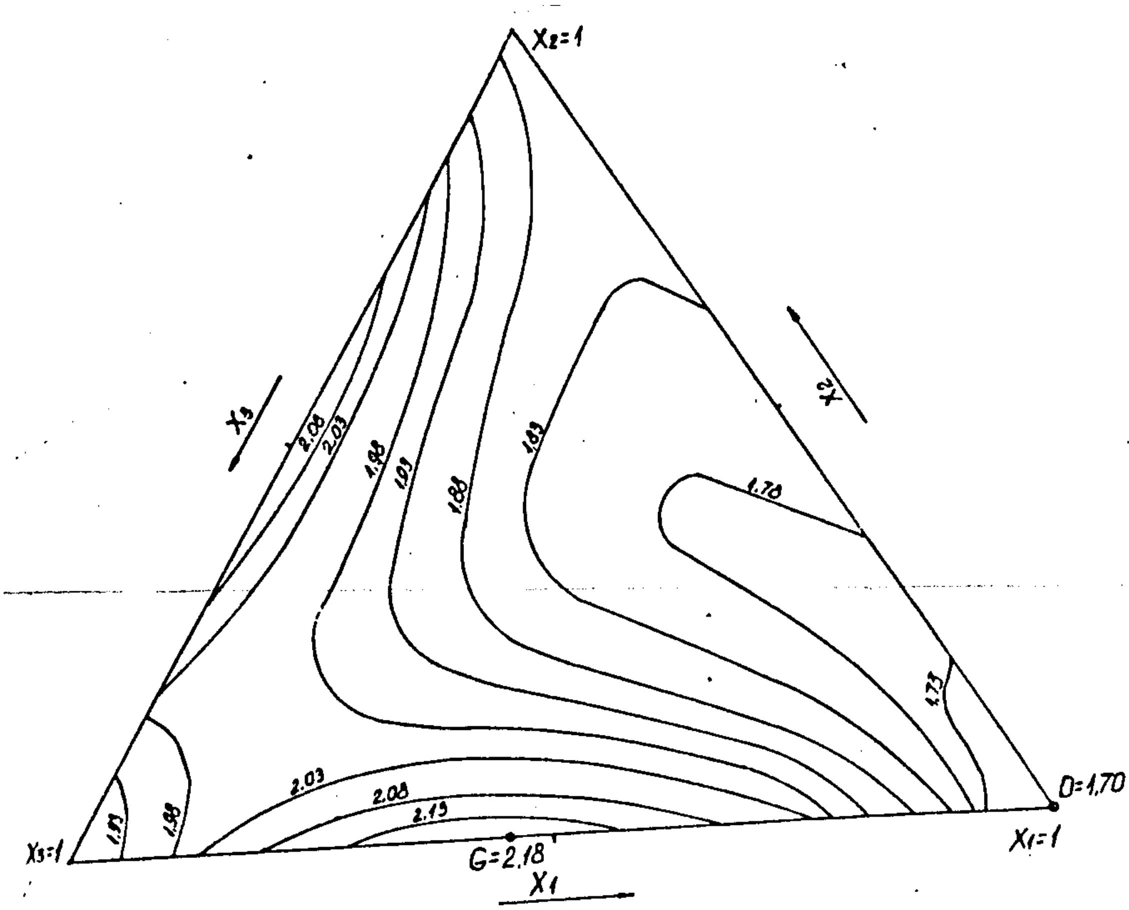


Fig. 3

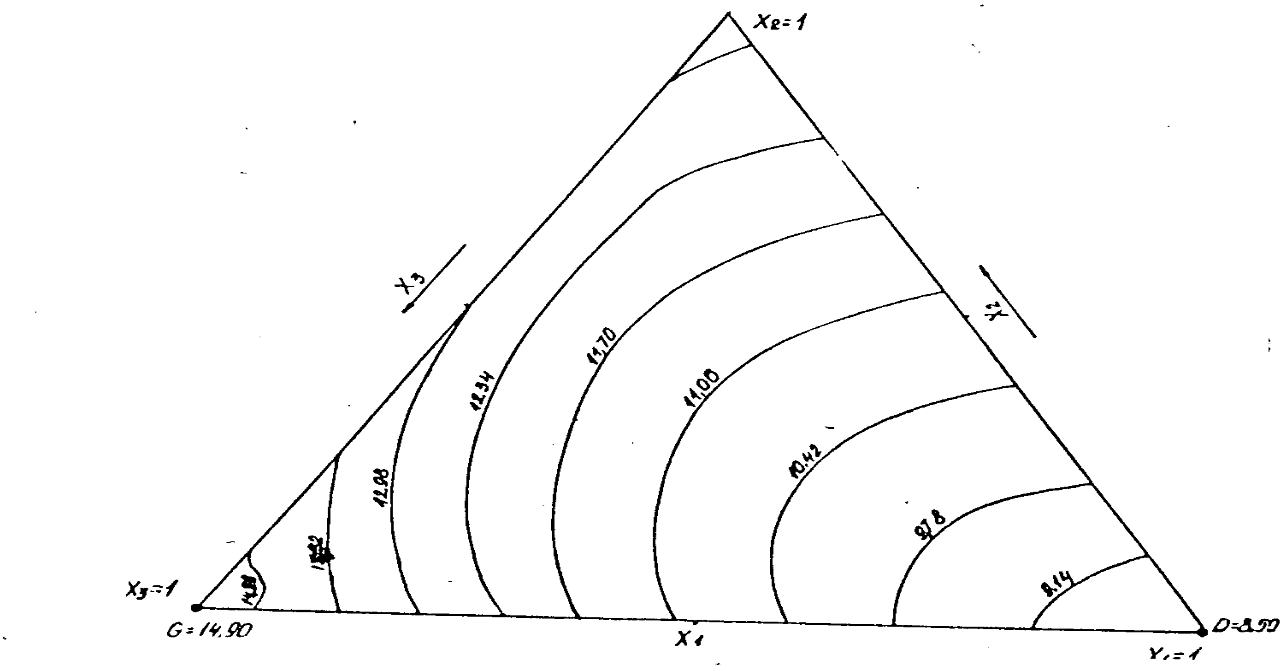
X₂=1
X₃
X₂
12.34
13.70
11.00
12.98
10.42
9.78
8.14
X₃=1
G=14.90
X₁
X₁=1

Fig. 4

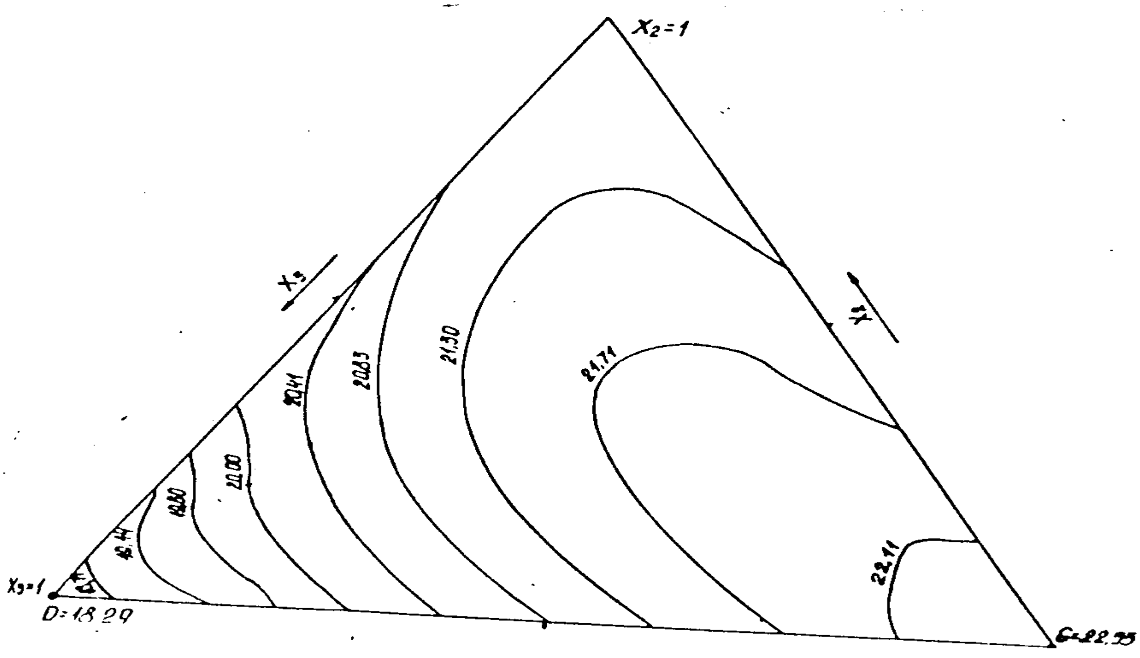
X₂=1
X₃
X₁
21.30
20.83
20.41
21.71
21.00
22.11
X₃=1
D=18.29

Fig. 5

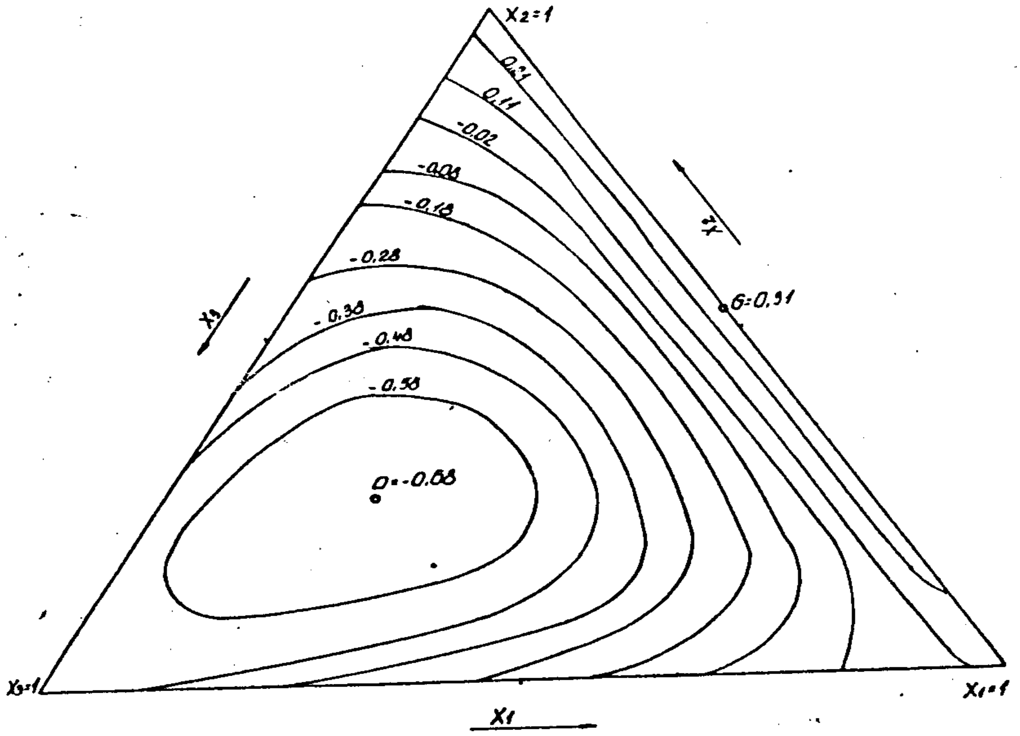
X2=1
0.21
0.11
-0.02
-0.08
-0.18
-0.28
-0.38
-0.48
-0.58
D=-0.68
G=0.31
X3=1
X1=1
X1
X2
X3

Fig. 6

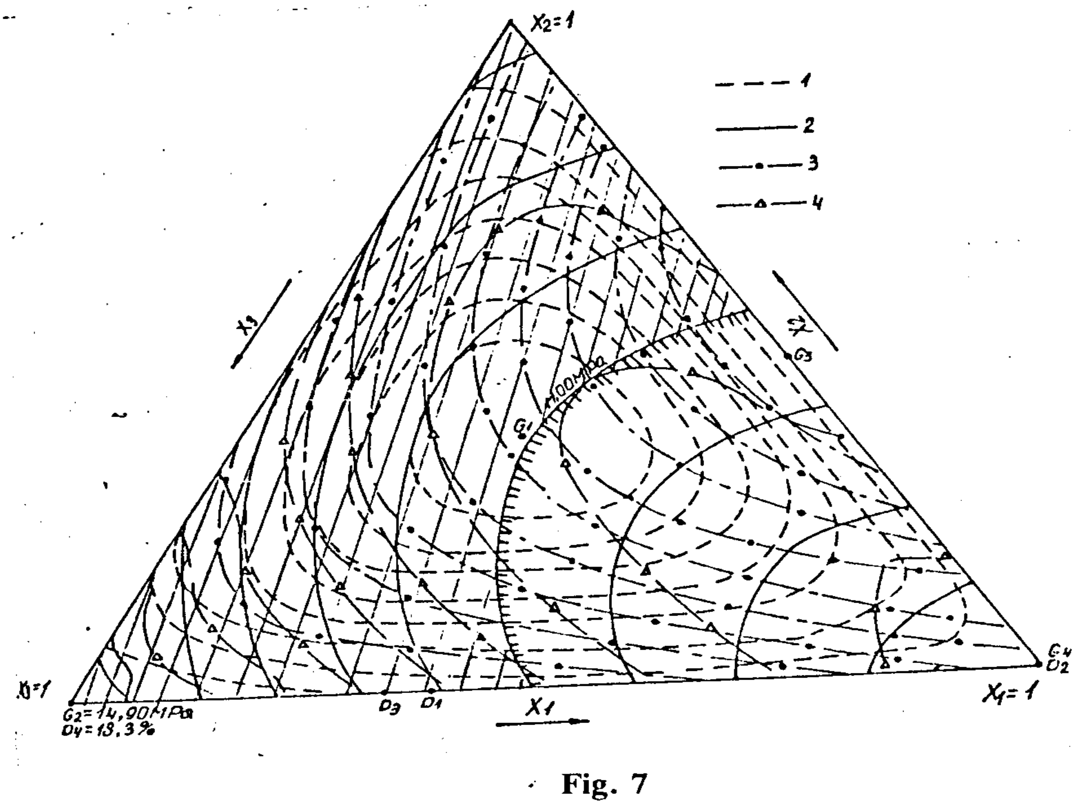
X2=1
1
2
3
4
X3
X2
G3
G1
X1
X1=1
G4
D2
D3
D1
G2=14,90 MPa
D4=13,3%

Fig. 7

For Ukpor clay such region could not be found because for all compositions investigated, the bending strength of the tiles was below 11.0 MPa. Based on the experimental result, it could be seen that Nsu clay is suitable for manufacturing of tiles at firing temperature of 1000°e. The composition enclosed in the shaded region of Fig. 7 could be used. The composition when used will show the optimal properties is as follows:

Nsu clay = 47.8%
Quartz = 33.7%
$CaCO_3$ 18.5%

When this composition is used one expects the bending strength after firing at 1000°C to be 14.9 MPa, and water absorption capacity to be 18.3%.

The maximum bending strength for tile form Ukpor clay is 10.8MPa. This value is lower than the standard requirement. Therefore Ukpor clay is unsuitable for tile making at firing temperature of 1000°C. Knowing however that the bending strength can be increased by increasing the firing temperature [23], a higher firing temperature is recommended for tiles from Ukpor clay, in order to improve the bending strength and thereby making it suitable for tile production.

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**LIST OF FIGURES**

FIG. I Flow diagram for tile production

FIG. 2 The region of the simplex under investigation

R. Recommended (in literature) region for pure clay.
1. Corresponding region (got by calculation) for Nsu Clay.
2. Corresponding region (got by calculation) for Ukpor Clay.

FIG. 3 Change in Bending Strength (in raw state) of tiles from Nsu Clay with respect to composition.

(In Fig. 4 to Fig. 7: G = maximum value of the property studied; D = minimum value of the property studied).

FIG. 4 Change in Bending Strength (after firing) of tiles from Nsu Clay with respect to composition.
FIG. 5 Change in Water Absorption Capacity of tiles from Nsu clay with respect to composition.
FIG. 6 Change in Shrinkage of tiles from Nsu Clay with respect to composition.
FIG. 7 Combined effect of composition on the properties studied for tiles from Nsu clay:
l. Raw Bending Strength, MPa.
2. Bending Strength after firing, MPa.
3.Shrinkage
4.Water Absorption Capacity, %
Gi(i = 1,2,3,4) = max. value of property.
Di(i= 1,2,3,4) = min. value of property