# Technique for optimization of ceramic bodies using mixture design

# *(Técnica para otimização de corpos cerâmicos usando projeto de misturas)*

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## Abstract

In the production of wall and floor ceramic tiles, mixtures of raw materials from several origins with different physical and chemical characteristics are used. Those changes of raw materials alter the quality of the finished product, what implicates in the constant reformulation in the composition of the ceramic mass through try and error, consuming time and materials. This configures the ideal circumstance to apply the techniques of experiments design, often used in many areas to model properties of such ceramic bodies. In the present study, 21 formulations of six raw materials, namely talc, quartz, calcareous, phyllite, dolomite and clay were selected and used as control factors in the experiments design. Those formulations were processed under conditions similar to those used in the ceramics industry: powder preparation (wet grinding, drying, granulation and drying), green body preparation (pressing and drying), firing (at 1180 ºC) and characterization. With the experimental results, regression models were calculated, relating bending strength, linear firing shrinkage and water absorption. After statistical analysis and verification experiments, the significance and validity of the models were confirmed, and one technique for optimization of ceramic bodies was developed: a mathematical expressions denominated loss of quality function. The regression models and the developed technique of the loss quality can then be used for the best combination of those six raw materials to produce a ceramic body with specified properties.
**Keywords:** mixture experiments, statistical analysis, mixtures optimization, experimental design.

## *Resumo*

*Na produção de revestimentos cerâmicos para parede e piso, são usadas misturas com matérias-primas de diversas regiões com características físicas e químicas diferentes. Essas mudanças de matérias-primas alteram as qualidades do produto final, o que implica em constante reformulação na composição de massas cerâmicas através de tentativas e erros, consumindo tempo e material. Isso configura a circunstância ideal para aplicar as técnicas de projeto de experimentos, freqüentemente usadas em outras áreas para modelar as propriedades mecânicas de corpos cerâmicos. No presente estudo, 21 formulações foram desenvolvidas a partir de seis matérias-primas utilizadas como fator de controle no projeto de experimento: talco, quartzo, calcário, filito, dolomita e argila. Essas misturas foram processadas em condições semelhantes aquelas usadas na indústria cerâmica: preparação da massa (moagem a úmido, secagem, granulação) preparação do corpo verde (prensagem e secagem) aquecimento (a 1180 ºC) e caracterização. Com os resultados experimentais foram calculados os modelos de regressão relacionando resistência à flexão, retração linear e absorção de água. Depois da análise estatística e verificação dos experimentos, o nível de significância e validades dos modelos foram confirmados, e uma técnica para otimização dos corpos cerâmicos foi desenvolvida, com expressões matemáticas denominadas função perda de qualidade. Os modelos de regressão e a técnica desenvolvida de perda de qualidade podem então ser usados para a melhor combinação daquelas seis matérias-primas para produzir um corpo cerâmico com propriedades específicas.*
***Palavras-chave:*** *projeto de misturas, análise estatística, otimização de misturas, projeto de experimentos.*

## INTRODUCTION

In the industrial processing of ceramic bodies such as floor and wall tiles, due to the sensitivity to raw materials and/or processing changes and the simplicity of their laboratory determination, the bending modulus of rupture, linear firing shrinkage and water absorption are frequently used as quality and process control parameter in the development and manufacture stages [1, 2]. Under constant processing conditions, these properties are basically determined by the mixture of raw materials and can, therefore, be modeled using the optimization methodology specific to the design of mixture experiments. Such procedure is common practice in the chemical industry [3-5] and is becoming popular in the field of glasses and ceramics [6-8]. It has proven, in all cases reported, to lead to greater efficiency and confidence in the results obtained, and to be less demanding in time, both material and human resources.

The design of mixture experiments configures a special case in response surface methodologies using mathematical and statistical techniques, with important applications not only in new products design and development, but also in the improvement of the design of existing products. The basic assumption is that there is a given mixture property which depends solely on the fractions (xi, summing up to unity) of specific components, or ingredients, of the mixture, and not on the amount of the mixture; thus, the changes in (or the response of) the property is entirely determined by the proportions of those components. To this aim, it is necessary, first to select the appropriate mixtures from which the response surface might be calculated; having the response surface, a prediction of the property value can be obtained for any mixture, from the changes in the proportions of its components [9-11].

In a system with $q$ independent variables (or components), there will be $(q - 1)$ independent composition variables $x_i$, and the geometric description of the factor space containing the $q$ components consists of all points on or inside the boundaries (vertices, edges, faces…) of a regular (q-1) dimensional simplex.

For 6 components of the simplex lattice {6, 2}, the experiments numbers are [1, 2]:

$$N = (q + m - 1)\,! \;/\; m!\,.(q - 1)\,! \qquad \text{(A)}$$

where: $q$=6 raw materials and $m$=2 second degree

The response (property) function $f$ can be expressed in its canonical form as a low degree polynomial (typically, first, second or third degree) [1, 2]:

$$\text{Linear: } f = \sum_{i=1}^{q} \beta_i x_i \qquad \text{(B)}$$

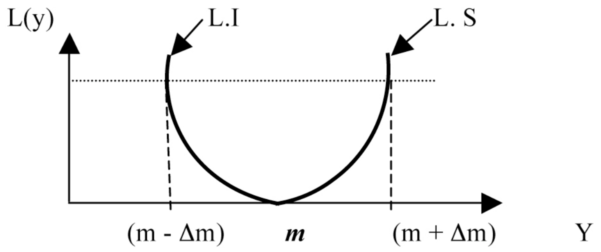
$$\text{Second degree: } f = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q}\sum \beta_{ij} x_i x_j \qquad \text{(C)}$$

Special cubic :

$$f = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j}^{q}\sum \beta_{ij} x_i x_j + \sum_{i<j<k}^{q}\sum\sum \beta_{ijk} x_i x_j x_k \qquad \text{(D)}$$

This polynomial equation has to be evaluated over a number $N$ of points so that it can represent the response surface over the entire region and it is only natural that a regular array of uniformly spaced points (i.e. a lattice) is used. This lattice is referred to as a *{q, m}* simplex lattice, *m being* the spacing parameter in the lattice. Then, a laboratory study consisting of $N$ experiments ($N > q$) has to be carried out and the values of the property on those selected $N$ lattice points evaluated. Regression equations such as B to D are then fitted to those experimental values and the model is considered valid only when the differences between the experimental and the calculated values (error) are uncorrelated and randomly distributed with a zero mean value and a common variance. When some or all the compositions ***xi*** are restricted by either a lower bound and/or an upper bound (i.e. the component fraction is not allowed to vary from 0 to 1.0 only a sub-region of the original simplex is of interest), which is frequently the case, the concept of pseudo-component can be used to define another simplex of new components (pseudo-components) present in the proportions *xi* and to which the *{q, m}* simplex lattice is applied. The fractions ***xi'*** are first calculated from the original ***xi*** { by $xi' = (xi - Li)/(1 - \sum L_i)$ }, where $Li$ is the lower bound for the $i$th component and $\sum L_i$ is the sum of all the lower bounds, < 1 ) and, once the regression equation is obtained, they are reverted back to the original components, so that the mixture can be prepared and the property experimentally determined [1, 2].

From the results of mixtures (planning matrix) and of the regression models, it is possible to select the best mixture using the technique of *function loss* developed by Taguchi [12, 13], reconciling conflicts of responses from types "larger is better" and "smaller is better", seeking always responses for the closest target value $m$ (desired quality). The loss function represents the loss of quality of a product, every time responses (properties) get farther from their value target. To express this function loss, Taguchi developed the quadratic form:



$$L(y) = k\,(y - m)^2 \qquad \text{(E)}$$

L (y) is the loss of quality
m is target value the of a product
Y is the response found in test or analysis
K is a constant that depends on the response.

Figure 1: Graphical representation of the function loss of quality.
*[Figura 1: Representação gráfica da função perda de qualidade.]*

Fig. 1 represents this function. Note that as the quality of the product is away from the target value $m$, the loss of quality increases.

This work describes the use of the design of mixture experiments to calculate initially, a regression model relating bending strength, water absorption and linear firing shrinkage of ceramic bodies, with the proportions (wt.%) of talc, quartz, calcareous, phyllite, dolomite and clay present, under constant processing conditions (wet grinding, moisture content, compaction pressure, firing schedule). The resulting statistical analysis involves fitting of mathematical equations to the experimental results.

An examined technique through the mathematical expressions called *loss function* was developed to allow selecting the best mixture among several of planning matrix. That criterion takes in consideration the types of characteristics of wanted qualities, respecting the properties of the types "larger is better" and "smaller is better", for instance: bending strength (larger is better) and water absorption (smaller is better).

## EXPERIMENTAL PROCEDURE

The raw materials used as reference in a local industry were: 10 wt.% talc, 4 wt.% quartz, 3 wt.% calcareous, 14 wt.% phyllite, 5 wt.% dolomite and 64 wt.% clay.

A modified {6, 2} simplex-lattice was used to define the matrix planning of 21 elaborated masses for mixtures of these raw materials that should be investigated, according to the equation C.

The selected mixtures were wet processed, following the conventional wall and floor tile industrial procedure: wet grinding for 2 h in a rotating cylindrical ball mill type, with capacity for grinding of 5 kg, using porcelain spheres, drying, moisturizing (6 ± 0.5 wt.%, dry basis), granulation (200 mesh) and uniaxial pressing (50 MPa). For each mixture, six flat specimens (150 x 65 x 8 $mm^3$) were produced, using 120 g of material for each specimen. An automatic hydraulic press was used with capacity of 100 ton and applied a compaction pressure of 50 MPa. After compaction, the test specimens were oven dried type of rolls type of rolls at 110 ± 5 °C until constant weight, and cooled to ambient temperature. These processing conditions were the same ones adopted by the industry.

The mechanical strength of dried specimens was determined in three-point bending tests, using a test machine with 10 kN capacity.

The linear shrinkage was calculated from the change in length (measured with Mitutoyo callipers with a resolution of 0.05 mm), upon firing, of the flat test pieces.

Water absorption was determined via boiling in water for 2 h, in a digital scale, resolution 0.01g.

All these tests were conducted in accordance with the Brazilian standard ABNT 13818 [14], and for each mixture, the test result was taken as the average of the six specimens, affected by the corresponding standard deviation. Those values were then used to iteratively calculate the coefficients of a regression equation such as eqs. B and C, until a statistically relevant model and response surface was obtained, relating the mechanical strength, firing shrinkage and water absorption with the weight fractions of talc, quartz, calcareous, phyllite, dolomite and clay present in the mixtures.

## RESULTS AND DISCUSSION

### *The simplex lattice mixture composition*

The distinctive roles that talc, quartz, calcareous, phyllite, dolomite and clay play during ceramic processing were used to establish the lower bound limits of 8.0 wt.% of talc, 3.0

Table I - Matrix planning of the mixtures created by the augmented {6, 2} simplex.
*[Tabela I - Matriz de planejamento das misturas criadas pelo método simplex {6,2}.]*

| | *A: True formulation* **(wt.%)** | | | | | | | **B: Pseudo-components** | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Raw → material** | **X1** | **X2** | **X3** | **X4** | **X5** | **X6** | **Total** | **X1** | **X2** | **X3** | **X4** | **X5** | **X6** | **Total** |
| **M1** | 18 | 3 | 3 | 12 | 4 | 60 | 100 | 1 | 0 | 0 | 0 | 0 | 0 | 1 |
| **M2** | 8 | 13 | 3 | 12 | 4 | 60 | 100 | 0 | 1 | 0 | 0 | 0 | 0 | 1 |
| **M3** | 8 | 3 | 13 | 12 | 4 | 60 | 100 | 0 | 0 | 1 | 0 | 0 | 0 | 1 |
| **M4** | 8 | 3 | 3 | 22 | 4 | 60 | 100 | 0 | 0 | 0 | 1 | 0 | 0 | 1 |
| **M5** | 8 | 3 | 3 | 12 | 14 | 60 | 100 | 0 | 0 | 0 | 0 | 1 | 0 | 1 |
| **M6** | 8 | 3 | 3 | 12 | 4 | 70 | 100 | 0 | 0 | 0 | 0 | 0 | 1 | 1 |
| **M7** | 13 | 8 | 3 | 12 | 4 | 60 | 100 | .5 | .5 | 0 | 0 | 0 | 0 | 1 |
| **M8** | 13 | 3 | 8 | 12 | 4 | 60 | 100 | .5 | 0 | .5 | 0 | 0 | 0 | 1 |
| **M9** | 13 | 3 | 3 | 17 | 4 | 60 | 100 | .5 | 0 | 0 | .5 | 0 | 0 | 1 |
| **M10** | 13 | 3 | 3 | 12 | 9 | 60 | 100 | .5 | 0 | 0 | 0 | .5 | 0 | 1 |
| **M11** | 13 | 3 | 3 | 12 | 4 | 65 | 100 | .5 | 0 | 0 | 0 | 0 | .5 | 1 |
| **M12** | 8 | 8 | 8 | 12 | 4 | 60 | 100 | 0 | .5 | .5 | 0 | 0 | 0 | 1 |
| **M13** | 8 | 8 | 3 | 17 | 4 | 60 | 100 | 0 | .5 | 0 | .5 | 0 | 0 | 1 |
| **M14** | 8 | 8 | 3 | 12 | 9 | 60 | 100 | 0 | .5 | 0 | 0 | 5 | 0 | 1 |
| **M15** | 8 | 8 | 3 | 12 | 4 | 65 | 100 | 0 | 5 | 0 | 0 | 0 | 5 | 1 |
| **M16** | 8 | 3 | 8 | 17 | 4 | 60 | 100 | 0 | 0 | .5 | .5 | 0 | 0 | 1 |
| **M17** | 8 | 3 | 8 | 12 | 9 | 60 | 100 | 0 | 0 | .5 | 0 | .5 | 0 | 1 |
| **M18** | 8 | 3 | 8 | 12 | 4 | 65 | 100 | 0 | 0 | .5 | 0 | 0 | .5 | 1 |
| **M19** | 8 | 3 | 3 | 17 | 9 | 60 | 100 | 0 | 0 | 0 | .5 | .5 | 0 | 1 |
| **M20** | 8 | 3 | 3 | 17 | 4 | 65 | 100 | 0 | 0 | 0 | .5 | 0 | .5 | 1 |
| **M21** | 8 | 3 | 3 | 12 | 9 | 65 | 100 | 0 | 0 | 0 | 0 | .5 | .5 | 1 |
| **RM** | 10 | 4 | 3 | 14 | 5 | 64 | 100 | .2 | .1 | 0 | .2 | .1 | .4 | 1 |

wt.% of quartz, 3.0 wt% of calcareous, 12.0 wt.% of phyllite, 4.0 wt.% of dolomite and 60.0 wt.% of clay. These lower limits were determined by evaluating also, the percentage used around of the mixture used in local industry for the production of tiles, here called RM reference mixture (Table I).

By considering the oxides that more influence in the formulation of a ceramic are the alumina, silica, alkali and alkaline earth [13] according to the chemical composition of raw materials above, the ternary diagram of raw materials that make up the tiles may be represented by these oxides. These limits create a restricted composition area of pseudo-components (Fig. 1) on which a {6, 2} simplex lattice was set. It visualizes, therefore in figure, an area bounded by location of raw materials, and any point within this area can be used for the development of the project of experiment.

Table I shows the matrix planning with the 21 elaborated mixtures (Mi), independent components, and the reference mass RM, in terms of true formulations (A) and in pseudo components (B), where: X1: talc, X2: quartz, X3: calcareous, X4: phyllit, X5: dolomite, X6: clay.

Fig. 2 shows the ternary system $SiO_2$ / $Al_2O_3$ / $XO+X_2O$. All mixtures of these raw materials must lie within the triangle they define. Hence, only a part of the composition area will be used.

*The models for responses*

Table II shows the properties (response) obtained for the selected 21 mixtures.

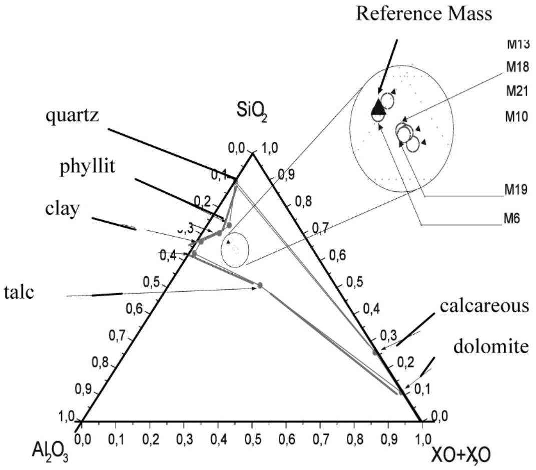


Figure 2: The ternary system $SiO_2$ / $Al_2O_3$ / $XO+X_2O$, showing the raw materials area, the restricted pseudo-components.
*[Figura 2: Sistema ternário $SiO_2$ / $Al_2O_3$ / $XO+X_2O$, mostrando a área de matérias-primas, a restrição em pseudo-componentes.]*

Having a measured value for the response property at specific coordinates, a regression equation can be sought for each property. Regression models were evaluated, eq. C: second degree polynomial, subjected to a significance level of 5%.

Table III shows the individual modeling of the response for the quadratic model, where the determination coefficients $R^2$ evaluate if the equations obtained by regressions are representative. The effects of the interactions between raw materials are noticed.

Those equations show, for instance, that the quartz associated to the calcareous reduces the linear shrinkage

Table II - Average values of the responses and corresponding standard deviations.
*[Tabela II - Valores médios das respostas e desvios padrão correspondentes).]*

| Mixture | Linear shrinkage (%) | Mechanical strength (Mpa) | Water absorption(%) |
|---|---|---|---|
| M1 | 3.22 ± 0.01 | 15. 14 ± 1.5 | 13.12 ± 0.53 |
| M2 | 2.34 ± 0.09 | 14.49 ± 1.1 | 17.35 ± 0.37 |
| M3 | 0.97 ± 0.01 | 12.53± 0.4 | 20.65 ± 1.64 |
| M4 | 3.20 ± 0.18 | 16.0 ± 1.6 | 15.85 ± 1.33 |
| M5 | 1.39 ± 0.18 | 11.5 ± 0.7 | 22.27± .42 |
| M6 | 2.99 ± 0.14 | 16.1 ± 0.9 | 15.5 ± .55 |
| M7 | 2.87 ± 0.08 | 13.1 ± 0.9 | 13.95 ± 1.05 |
| M8 | 1.72 ± 0.03 | 15.1 ± 2.5 | 17.05 ± 0.39 |
| M9 | 3.20 ± 0.22 | 18.6 ± 2.0 | 14.05 ± 0.83 |
| M10 | 2.10 ± 0.10 | 16.1 ± 1.0 | 16.70 ± 0.87 |
| M11 | 3.15 ± 0.15 | 17.7 ± 1.1 | 13.50 ± 0.16 |
| M12 | 1.26 ± 0.05 | 9.4 ± 1.0 | 20.65 ± 0.61 |
| M13 | 2.37 ± 0.04 | 16.0 ± 1.5 | 15.62 ± 0.29 |
| M14 | 1.61 ± 0.08 | 13.9 ± 1.7 | 16.40 ± 0.84 |
| M15 | 2.84 ± 0.06 | 14.9 ± 1.2 | 14.62 ± 0.46 |
| M16 | 1.67 ± 0.12 | 16.2 ± 0.7 | 18.90 ± 0.29 |
| M17 | 1.65 ± 0.36 | 14.8 ± 0.7 | 20.67 ± 0.17 |
| M18 | 1.64 ± 0.07 | 17.0 ± 0.8 | 18.07 ± 0.51 |
| M19 | 1.78 ± 0.13 | 16.6 ± 0.4 | 16.37 ± 0.93 |
| M20 | 3.48 ± 0.18 | 18.9 ± 0.1 | 14.37 ± 0.41 |
| M21 | 2.37 ± 0.03 | 18.4 ± 0.8 | 18.20 ± 0.44 |
| MR | 2.74 ± 0.08 | 17.2 ± 0.8 | 14.70 ± 0.50 |

Table III - Regression models obtained for the responses Yi.
*[Tabela III - Modelos de regressão obtidos para as respostas Yi.]*

| Response (Yi) | $R^2$ | quadratic model |
|---|---|---|
| Linear shrinkage.<br>(Y1) | 0,97 | $Y1 = 3.26X_1 + 2.42X_2 + 1.1X_3 + 3.23X_4 + 1.48X_5 + 3.07X_6$<br>$- 1.79X_1X_3 - 1.03X_1X_5 - 1.92X_2X_3 - 1.88X_2X_4 - 1.42X_2X_4 -$<br>$2.32X_3X_4 + 2.01X_1X_3X_5 - 1.87X_3X_6 - 2.48X_4X_5 + 1.16X_4X_6.$ |
| Bending Strength.<br>(Y2) | 0,76 | $Y2 = 14.7X_1 + 13.6X_2 + 13.5X_3 + 16.7X_4 + 11.6X_5 + 16.5X_6 +$<br>$14.2X_1X_4 + 10.1X_1X_5 + 11.6X_1X_6 - 16.9X_2X_3 + 11.4X_3X_5 +$<br>$9.6X_4X_5 + 9.6X_4X_6 + 11.9X_5X_6.$ |
| Water Absorption<br>(Y3) | 0,94 | $Y3 = 13.12X_1 + 17.35X_2 + 20.65X_3 + 15.85X_4 + 22.27X_5 + 15.5X_6$<br>$- - 5.15X_1X_2 - 4.0X_1X_5 + 6.6X_2X_3 - 3.9X_2X_4 - 13.65X_2X_5 -$<br>$7.2X_2X_6 - 10.75X_4X_5 - 5.20X_4X_6.$ |

and the bending strength simultaneously ($-1.92\ X_2X_3$ and $-16.9X_2X_3$). In the production line, the minimum of linear shrinkage (smaller is better) is important, but with the increase of the bending strength (larger is better). Having conflict is necessary, therefore, to find a conciliatory solution. The simultaneous study that leads the optimization of the response of interest is needed and will be the following subject. Here, $X_1$ is the talc fraction, $X_2$ is the quartz fraction, $X_3$ is the calcareous fraction, $X_4$ is the phillit fraction, $X_5$ is the dolomite fraction and $X_6$ is the clay fraction (i.e. independent component fraction).

### *Variance analysis*

Table IV gives the various statistical properties of the regressions, using the nomenclature commonly found in the relevant texts [1, 2]. Using the P-value approach to hypothesis testing (i.e. P-value 5 significance level), this table shows that the second degree model is statistically significant for three responses, because $P = 0.00000 < 0.05$. The coefficients of multiple determinations $R^2$ for the responses linear shrinkage and water absorption are 0.97 and 0.94 respectively, meaning that the models present a very small variability. The $R^2$ for the response bending strength is 0.83, meaning that the model present a small variability. Low values of $F_{LF}$ mean adjustment of the model.

### *Optimization of the mixture ceramic - The function loss of quality*

The concept of the *loss of quality* developed by Taguchi [12, 13] will be adapted to the present study to select the best mixture of the Table I, taking in consideration the best responses (linear shrinkage, bending strength, water absorption) and at the same time reconciling existing conflicts like: larger is better (example bending strength) and the smaller is better (example: linear shrinkage).

Fig. 3 compares two responses $Y_1$, $Y_2$, - (✳) - inside the range of LS = upper limit (which in this case is the worst response, because the target value is: the smaller is better), and LI = lower limit, response that would be ideal (smaller is better, i.e.; target value). YMR is the response of the ceramic bodies manufactured in a local industry, used as reference mixture in this work. It tries to find an expression that represents the sum of the losses of found response.

Table IV - Statistical properties relevant for variance analysis[a].
*[Tabela IV - Propriedades estatísticas relevantes para análise de variância[b].]*

| Response | Lack of fit / Pure error = $F_{LF} = MS_{LF} / MS_{PE}$ | $F_0 = MS_R / MS_r$ | $R^2$ | $R^2_A$ | P Value |
|---|---|---|---|---|---|
| Linear shrinkage | 0.0 | 122.6 | 0.9750 | 0.9670 | 0.0000 |
| Bending Strength. | 0.0 | 14.6 | 0.8227 | 0.7664 | 0.0000 |
| Water Absorption | 0.0 | 52.5 | 0.9464 | 0.9293 | 0.0000 |

[a] $MS_{LF}$: quadratic average of adjustment lack; $MS_{PE}$: quadratic average of pure error; $MS_R$: quadratic average of the model; $MS_r$: quadratic average of the residues. $R^2$: coefficient of multiple determinations. $R^2$: adjusted $R^2$.

*[[b] $MS_{LF}$: Média quadrática de falta de ajuste; $MS_{PE}$: Média quadrática de erro puro; $MS_R$: Média quadrática do modelo; $MS_r$: Média quadrática dos resíduos. $R^2$: coeficiente de determinação múltipla. $R^2_A$: ajustado $R^2$.]*

It is observed that $Y_1 > Y_2$, whose situation is more favorable for $Y_2$ (smaller is better), and $(Y_1 - IL) > (Y_2 - IL)$, what implicates in a larger loss for $Y_1$ (worst response).

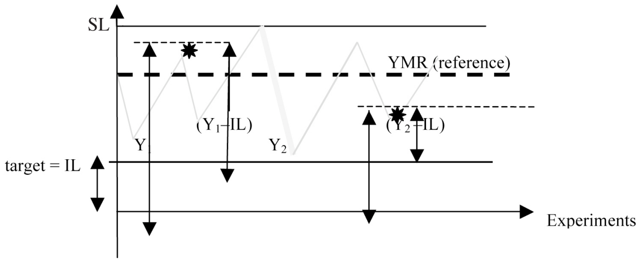


Figure 3: Behavior for response of the type "smaller is better = target value".

*[Figura 3: Comportamento das respostas do tipo "menor é melhor = valor alvo".]*

IL = Inferior Limits = target: it is the value that would be ideal, (smaller is better)

SL = Superior Limits, it is the worst found response, (when smaller is better)

Equation (B) represents the evolution in the calculation of quality losses for ceramic mixtures starting from the discussion done based on Fig. 3.

$$\check{Z}(I) = \sum_{j=1}^{j} [1 / (SL - IL)^2 \, . \, [(Y_J - IL_J)^2]] \qquad (B)$$

For the cases of the type "larger is better" (example: bending strength), Fig. 4 shows the behavior of two responses (✸) showing that the smallest losses happen when the largest response is obtained.

It is observed that $Y_1 > Y_2$, whose situation is more favorable for Y1 (larger is better), and $[SL - Y_1] < [SL - Y_2]$ implicates in a larger loss for $Y_2$.

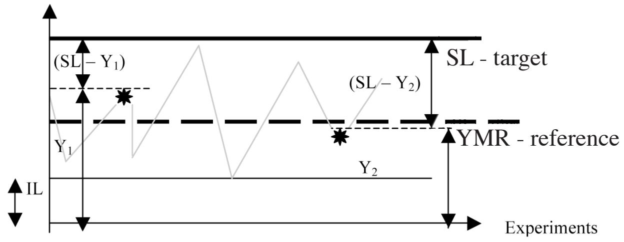


Figure 4: Behavior of the graph for response of the type "larger is better".

*[Figura 4: Comportamento do gráfico para respostas do tipo "maior é melhor".]*

In a similar way to the development of equation C, the equation of the loss function in that situation (Fig. 4) is:

$$\check{Z}(I) = \sum_{j=1}^{j} [1 / (SL - IL)^2 \, . \, [(SL - Y_J)^2]] \qquad (C)$$

(Target =SL, the target is the superior limit)

Table V shows the results of the loss of quality of the masses, using equations ( B ) and (C ).

*L.S.: Linear* Shrinkage W.A.: Water Absorption B.S.: Bending Strength.

Larger losses, in growing order:

Table V - Summary of the losses of quality of the mixtures.

*[Tabela V - Resumo das perdas de qualidades das misturas.]*

| **Mixtures** | *Loss Z L.S.* | Loss Z W.A | Loss Z B.S. | **Loss Z Total** |
|---|---|---|---|---|
| M1 | *2.24* | 0.00 | 0.68 | **2.92** |
| M2 | *0.82* | 0.89 | 1.10 | **2.81** |
| M3 | *0.00* | 2.83 | 2.29 | **5.12** |
| M4 | *2.19* | 0.37 | 0.47 | **3.03** |
| M5 | *0.08* | 4.19 | 3.08 | **7.35** |
| M6 | *1.80* | 0.28 | 0.45 | **2.53** |
| M7 | *1.59* | 0.03 | 1.90 | **3.53** |
| M8 | *0.25* | 0.77 | 0.83 | **1.85** |
| M9 | *2.19* | 0.04 | 0.01 | **2.24** |
| M10 | *0.56* | 0.64 | 0.43 | **1.63** |
| M11 | *2.09* | 0.01 | 0.09 | **2.19** |
| M12 | *0.04* | 2.83 | 5.05 | **7.92** |
| M13 | *0.86* | 0.31 | 0.47 | **1.02** |
| M14 | *0.18* | 0.54 | 1.40 | **2.12** |
| M15 | *1.53* | 0.11 | 0.88 | **2.53** |
| M16 | *0.22* | 1.67 | 0.41 | **2.30** |
| M17 | *0.20* | 2.85 | 0.94 | **3.99** |
| M18 | *0.20* | 1.23 | 0.20 | **1.65** |
| M19 | *0.29* | 0.00 | 0.29 | **0.58** |
| M20 | *2.77* | 0.08 | 0.00 | **2.85** |
| M21 | *0.86* | 1.29 | 0.01 | **2.26** |

M19 < M13 < M10 < M18..........,
(0,58) < (1,02) < (1,63) < (1,65) ……,
M19 is the best mass (smaller loss = 0, 58).

Fig. 5 shows the behavior of the masses in function to the quality losses.

The model of optimization of the loss of quality can be found in function of the independent variables Xi, because, Y=F(X) and Z=F(Y) à Z=F(X). The following expression represents the loss model Z(i) in function of the raw materials. In absolute terms, one can see that the dolomite is the raw material that most contributes with the loss of quality of the masses (10.16 X5), and the clay tends to minimize the quality losses (2,82 X6). In relative terms, phyllite and dolomite minimize the quality loss (–22.26X4X5):

Z(I) = 2.92X1 + 3.66X2 + 7.53X3 +3.41X4 + 10.16X5 + 2.82X6 + 1.08X1X2 – 3.9X1X3 – 3.54X1X4 – 17.44X1X5 – 2.72 X1X6 + 18.02X2X3 – 6.82X2X4 – 15.56X2X5 – 2.48X2X6 – 6.16X3X4 – 8.58X3X5 – 9.54X3X6 – 22.26X4X5 – 0.66X4X6.

Table VI shows comparative data among the best

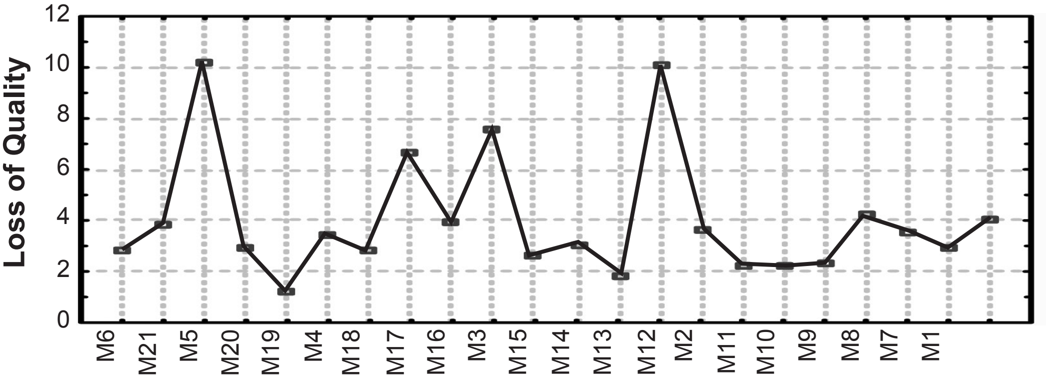


Figure 5: Behavior of the losses of quality of the mixtures.
*[Figura 5: Comportamento das perdas de qualidade das misturas.]*

Table VI - Comparison of the best mixture M19, in relation to the reference mixture RM.
*[Tabela VI - Comparação da melhor mistura M19 em relação à mistura de referência RM.]*

| | A : Formulation (%wt) | | | | | | B: codified Levels | | | | | | L. S | B.S | W.A. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | | | | | % | kgf cm$^2$ | % |
| Mixture<br>e<br>↓ | X1 | X2 | X3 | X4 | X5 | X6 | X1 | X2 | X3 | X4 | X5 | X6 | | | |
| RM | 10 | 4 | 3 | 14 | 5 | 64 | 0.2 | 0.1 | 0.0 | 0.2 | 0.1 | 0.4 | 2.7 | 172 | 14.7 |
| M19 | 8 | 3 | 3 | 17 | 9 | 60 | 0 | 0 | 0 | 0.5 | 0.5 | 0 | 1.8 | 166 | 16.4 |

*X1: Talc, X2:Quartz, X3: calcareous, X4:Phyllit, X5:Dolomite, X6:Clay, L.S. : Linear Shrinkage, B.R: Bending Strength, W.A.: Water Absorption*

mixture selected between those elaborated at the planning matrix, through the function loss (M19); and the mixture manufactured in the industry (RM).

The mixture *M19* had presented a linear shrinkage around 50% smaller than the reference mixture *RM*, without major changes in the bending strength and water absorption. In terms of composition of the raw materials, that mass is characterized by a major percentile of phyllite and dolomite, compared with the reference mixture RM.

## CONCLUSION

The design of mixture experiments enabled the calculation of regression models relating bending strength, linear firing shrinkage and water absorption with composition, which can then be used to select the best combination of six given raw materials (talc, quartz, calcareous, phyllit, dolomite and clay) to produce, under constant processing conditions, a ceramic body with specified properties. Furthermore, the equations loss of quality, developed in this work, allow for the selection of the best mixture among the several ones that are part of the planning matrix, with optimized responses, reconciling conflicts of responses from types "larger is better" and "smaller is better".

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