DEVELOPMENT OF A METHODOLOGY TO FORMULATE CERAMIC BODIES USING DESIGN OF EXPERIMENTS - TEST CASE: PORCELAIN STONEWARE TILES

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1. INTRODUCTION

The research involved in the development of a proper formulation for a ceramic mass requires a great deal of time, effort and money. Moreover, if the research is not well planned, little information will be obtained about similar or possibly even better or less costly formulations, even if a good one is identified. Porcelain stoneware tiles are fired in short cycles and their formulations usually contain several raw materials, reducing the usefulness of phase equilibrium diagrams to predict the behaviour of slightly different formulations. The work reported here involved the evaluation of a method to aid in the above mentioned tasks, based on the technique of design and analysis of experiments, using porcelain stoneware tiles as a test case.

2. METHODOLOGY

Based on parameters relating to the physical, chemical and mineralogical characteristics of natural raw materials identified as favourable to meet the technical requirements and necessary workability of porcelain stoneware, the methodology consisted of:

1) selecting four raw materials and their lower and upper limits in formulations to be studied, defining a feasible "region of interest";
2) selecting the order of the polynomial to attempt to adjust experimental data and the number of replications, defining the formulations;
3) batching the masses;
4) processing and characterizing the test bodies of the formulations on a laboratory scale under standardized conditions; and
5) analyzing the results according to ceramic and statistical aspects/features.

3. RESULTS AND DISCUSSION

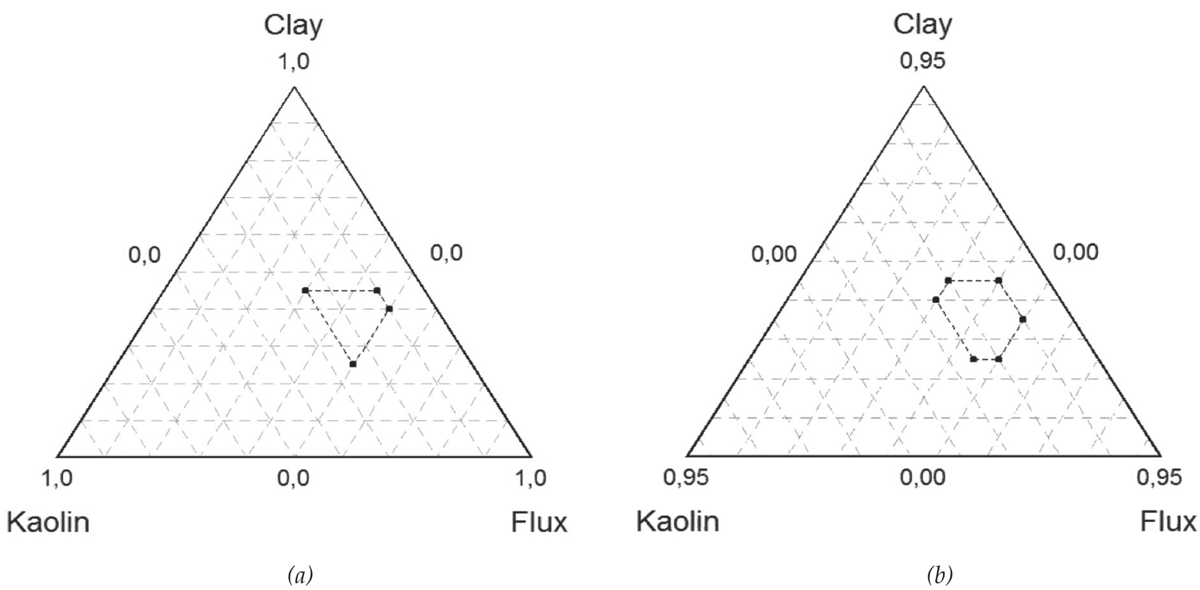

Figure 1. Region evaluated at: a) 0% talc, and b) 5% talc

Thirteen masses corresponding to eleven formulations were examined to evaluate the measured characteristics (responses) in the polyhedral region defined by mixtures of clay, kaolin, flux and talc with a first-degree order polynomial. The lower and upper limits of each constituent were clay: 25% - 45%, kaolin: 10% - 25%, flux: 30% - 50% and talc: 0% - 5%. Figure 1 shows two sections of the 3-D polyhedral region, at 0% and 5% talc, with ten formulations in the vertices. An additional formulation on the centre of the region, replicated twice, was added to allow for the identification of nonlinear variations of responses inside the region.

Pre-milled raw materials were mixed in a mechanical stirrer (70% solid content, dry basis, at 1500rpm, 15 minutes) according to predefined proportions, dried to 8% humidity and granulated to less than 350μm. The chemical compositions of the formulations, normalized to 0% loss on ignition, were SiO_2 57.8-60.7, Al_2O_3 27.9-33.1, $K_2O + Na_2O$ 3.1-3.8, MgO + CaO 1.6-4.0 and Fe_2O_3 2.6-3.3. All the masses had less than 1.0% residues through a 53μm sieve.

The formulated masses were compacted under different pressures (53 - 65MPa) to average apparent compact densities of 1.87 to 1.91g/cm^3. The firing schedules were 65 – 70 min, at maximum temperatures of 1175, 1200 and 1225°C and 5 min soaking time. The fired bodies were characterized for water absorption (WA), linear shrinkage (LS) and modulus of rupture (MOR). Table 1 presents the range of these characteristics for the fired test bodies.

FIRING TEMPERATURE	WA (%)	LS (%)	MOR (MPa)
1175°C	3.06 – 7.55	6.7 - 8.2	not evaluated
1200°C	0.66 – 5.14	7.6 - 9.1	not evaluated
1225°C	0.12 – 3.01	8.5 - 9.8	68.7 - 97.4

Table 1. Variation range of average water absorption, linear shrinkage and modulus of rupture of test bodies

The statistical models (polynomials) that best fitted the experimental values were first-order (except for WA at 1225°C, which included Kaolin*Talc). The adjusted R^2 values and p-values of regressions are listed in Table 2. The low order of fitted models allows one to easily view the effects of variations in raw material content on the measured responses through graphic techniques such as Contour Plots. Moreover, such planned experiments offer other advantages, such as larger amounts of useful information about the system under study and the possibility of identifying better, less expensive or more robust formulations than the ones evaluated. Figure 2 a) to d) shows the contour plots for water absorption and linear shrinkage of test bodies fired at 1225°C, revealing quite different regions of compositions that attained WA requirements for BIa products and showed LS ranging from roughly 8.8% to 9.6%.

PARAMETER	WA (%)			LS (%)		
	1175°C	1200°C	1225°C	1175°C	1200°C	1225°C
$R^2_{adjusted}$ (%)	86.2	90.2	83.2	95.0	95.6	85.6
p-value	0.000	0.000	0.000	0.000	0.000	0.001

Table 2. Statistical parameters of best fitted models

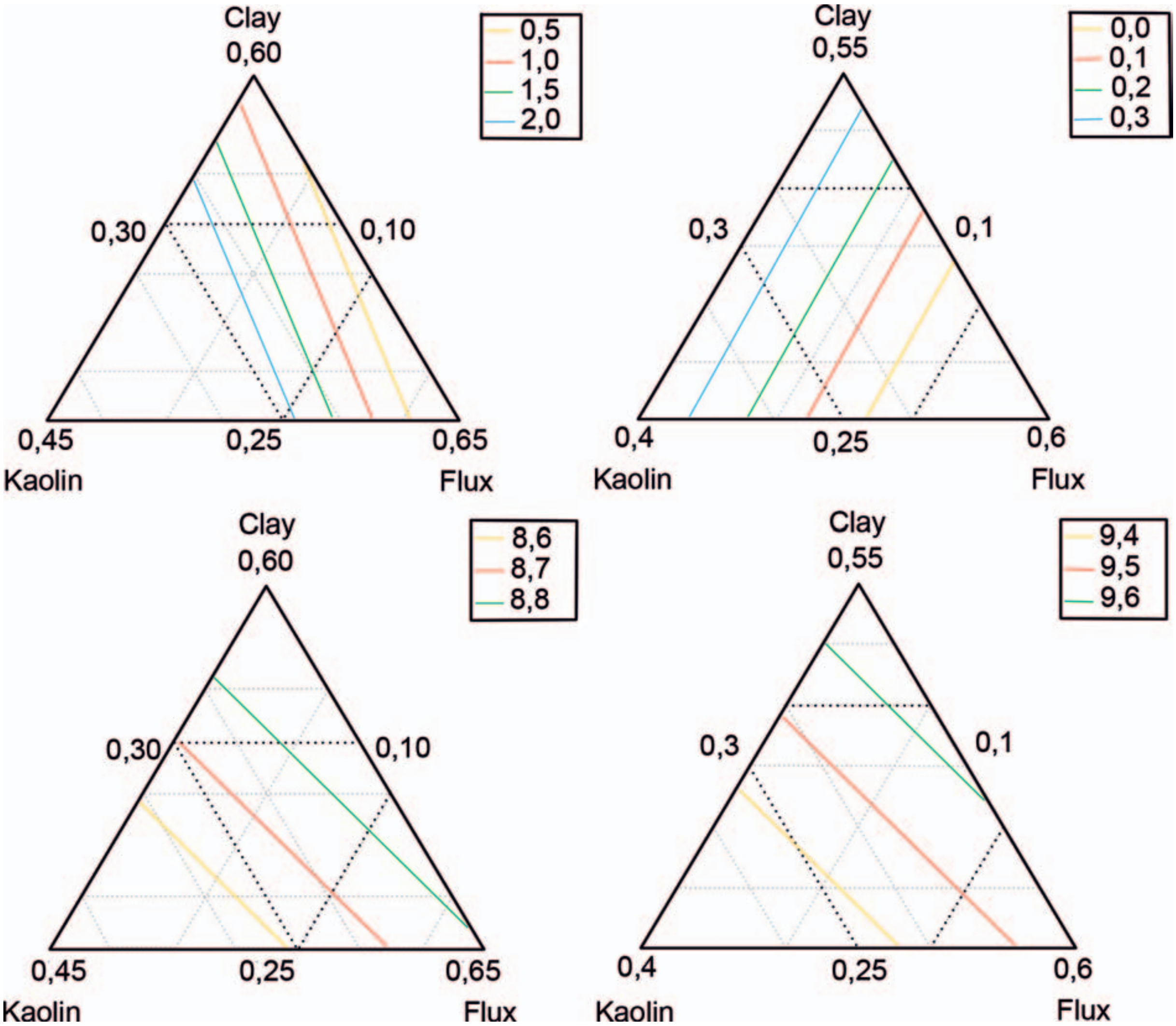

Figure 2. Contour plots of evaluated characteristics of test bodies fired at 1225°C; WA (%) for a) 0% Talc compositions and b) 5% Talc compositions; LS (%) for c) 0% Talc compositions and d) 5% Talc compositions

4. CONCLUSIONS

The methodology developed here enables one, from the first trials, to identify promising formulations of ceramic bodies for a chosen product. Well-planned and controlled experiments are extremely useful for solving problems relating to cost, availability, workability and uniformity.