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Using mixture design experiments to optimize the Cambodian clays in ceramic tile manufacturing

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Abstract: *A Cambodian clay deposit was used for the development of tile formulations using the statistical design of mixture experiments. Besides the clays, commercial feldspar and silica sand (quartz) were also used to prepare ten different tile formulations. The specimens were fabricated by pressing (30 MPa) and then fired at 1170 oC. The sintered specimens were characterized for their linear firing shrinkage, water absorption and modulus of rupture. Regression models were calculated, using mixture design experiments method, by relating the sintered properties with the compositions. The experimental results showed that Cambodian clays are suitable to be used as one a raw material in the production of ceramic tiles.*

Keywords: Traditional ceramic; Cambodian clay; Ceramic tile, Mixture design experiments

1 INTRODUCTION

In Cambodia, most widespread clays are found in alluvium plan deposits and those clays are being used for the production of small-scale ceramic products such as potteries, dinner and kitchen wares and souvenirs (Cambodia, 1993).

Ceramic tiles used for walls and floors are a type of structural ceramics and are a multicomponent system consisting of clay, quartz and carbonates, making them one of the most complex ceramic materials.Generally, three components are used and play fundamental roles in optimizing the performance of the final properties of ceramic tiles. Clay is the first component, whereby its plasticity facilitates the shape of the body, while second is feldspar which is used for fluxing and the third is silica which is used as a filler and stabilizer material (Bender et al., 1982 and Kamseu et al., 2007). In the industrial processing of ceramic bodies such as floor and wall tiles, linear firing shrinkage (LFS), water absorption (WA), and modulus of rupture (MOR) have been reported to be important factors in judging the quality and process control in the development and manufacturing states (Correia et al., 2004).

The present work is aimed to provide a comprehensive picture for optimization of the ceramic tile formulations containing Cambodian clays using the mixture design experiments. Regression models were designed and calculated using parameters of LFS, WA and MOR under constant processing conditions and they were used to select the best mixture composition to produce a ceramic tile body with the specified properties.

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2 EXPERIMENTAL PROCEDURE

2.1 Raw materials

A Cambodian clay taken from Prek An Chanh, Kandal province, Cambodia (denoted as C4) and commercial products such as sodium feldspar N325 (Maxum N325) and silica 325 (Cerasil 325, >99 wt.% quartz), all supplied by Sibelco Malaysia SdnBhd, were used to prepare the ceramic tile body.

2.2 Procedure

A modified {3,2} centroid simplex lattice, augmented with interior points, was used to define the mixtures of raw materials. The selected mixtures were prepared according to the industrial ceramic floor tile procedures: wet grinding (residue remained on a #325 mesh sieve below 4 wt.%), drying, moisturizing (6.5 wt.%, dry basis), granulation (using #30 mesh sieve), and uniaxial pressing. The specimens (50 mm × 10 mm × 7 mm) for LFS, WA and MOR measurements were fabricated using 7.0 g of granulated powder per test piece with a compaction pressure of 30 MPa. After pressing, the green specimens were oven-dried at 110 ^{o}C for 24 h and then fired at 1170 ^{o}C for 30 min at a heating rate of 10 ^{o}C/min. After soaking, they were furnace cooled to room temperature.

In all, five replications of the mixtures were carried out for the statistical analysis to iteratively calculate the coefficient of the regression equation until a statistically relevant model and response surface was obtained relating to the LFS, WA, and MOR with the weight fractions of clay,were carried out with Minitab 15 (Minitab Inc., 2007).

3 RESULTS AND DISCUSSION

3.1 The {3,2} simplex-lattice mixture composition and models for LFS, WA and MOR

A modified Three independent components i.e. clay, feldspar and quartz, which define the composition equilateral triangle, were used to establish the lower bound limits of 15 wt.% for each component. Resulting from these restrictions, 10 mixture compositions (Mi, i = 1, 2,…, 10) were generated a restricted composition triangle on which a {3,2} simplex lattice (six points) was set (Montgomery, 2001), as shown in Fig.1 and Table 1.

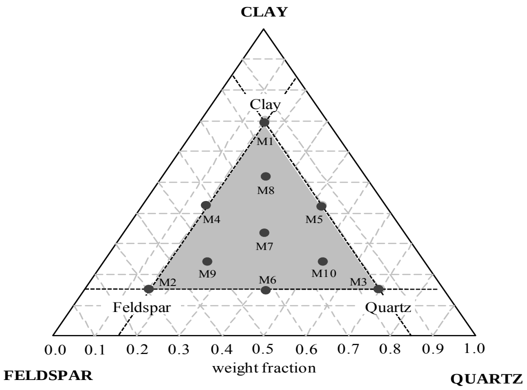

Fig.1.The ternary system clay-feldspar-quartz (independent components), showing the raw materials triangle, the restricted fraction components triangle, and the 10 simplex points which fulfilled those restrictions

Table 1.Composition of the design mixtures created by the augmented {3,2} simplex

Mixtures	Raw materials (wt.%)			Total (wt.%)
	Clay	Feldspar	Quartz	
M1	70.0	15.0	15.0	100.0
M2	15.0	70.0	15.0	100.0
M3	15.0	15.0	70.0	100.0
M4	42.5	42.5	15.0	100.0
M5	15.0	42.5	42.5	100.0
M6	42.5	15.0	42.5	100.0
M7	51.6	24.2	24.2	100.0
M8	33.3	33.3	33.3	99.9
M9	24.2	51.6	24.2	100.0
M10	24.2	24.2	51.6	100.0

Regression equations can be designed for each property (LFS, WA, and MOR) at specific coordinates (Table 1) at a 5% level of significance. The regression models for LFS, WA and MOR of the three mixtures which were found to be statistically the most adequate at the significant level are expressed as:

- $LFS = 20.6C + 29.3F - 20.3Q - 63.1CF + 12.9CQ - 13.6FQ + 182.0CFQ + 51.5CF(C - F) - 127.4CQ(C - Q)$ (Eq.1)
- $WA = 8.5C + 4.4F + 55.9Q + 0.8CF - 116.1CQ - 93.4FQ$ (Eq.2)
- $MOR = 139C - 29F + 161Q - 299CF - 626CQ - 655FQ + 4510CFQ - 1082CF(C - F) + 803CQ(C - Q)$ (Eq.3)

3.2Testing adequacy of the models of LFS, WA, and MOR

Table 2 lists the major statistical properties of the regressions of C4–MC(C4 mixture with feldspar and silica sand). The statistical results obtained from the analysis of variances can clearly prove that the regression models (Eqs. (1)–(3)) are statistically significant (p value ≤ significant level and high coefficients of multiple determination (R^2> 95%).

Table 2.Major statistical properties of variance analysis for significant of regression models

Property	Regression model	df	RMSE	*F* test	*p* value	R^2(%)
C4–MC						
LFS	Full cubic	2	0.190	119.05	< 0.0001	99.55
WA	Quadratic	3	1.244	109.99	< 0.0001	95.98
MOR	Full cubic	2	3.604	25.47	< 0.0001	97.82

Another important tool in validating the adequacies of models' prediction capacity is root mean square error (RMSE). If the value of RMSE is close to zero, then the model has perfectly predicted the properties (Ott and Longnecker, 2001). The values of RMSE in Table 2 prove that the Eqs. (1)–(3) are significant.

3.3Response surface and contour plots of LFS, WA, and MOR

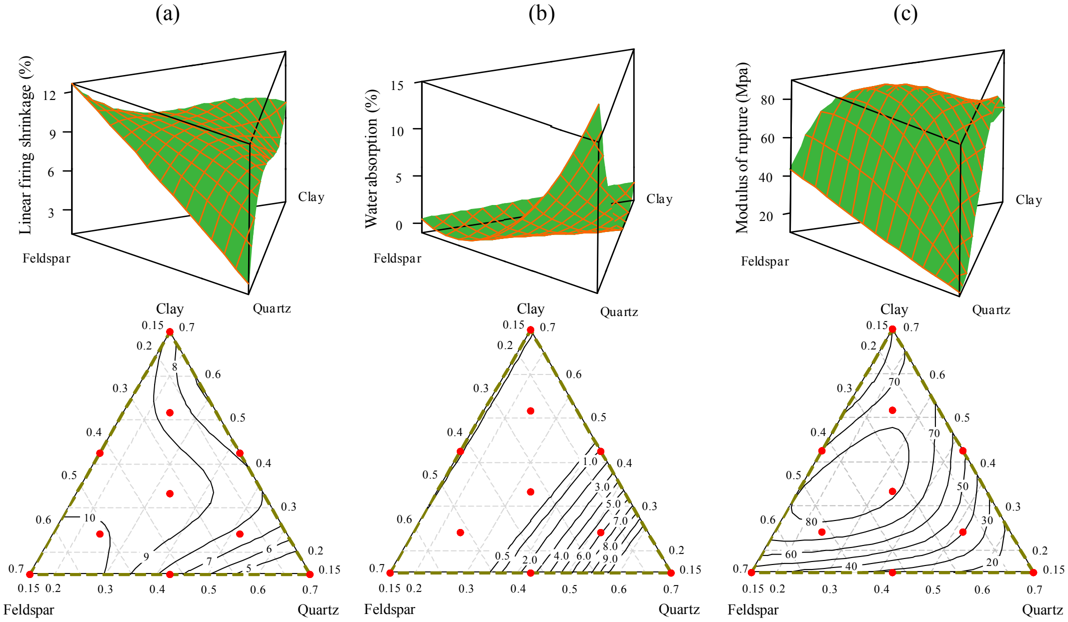

Fig.2. Response surface plots and their mixture contour plots of C4–MC: (a) linear firing shrinkage, (b) water absorption, and (c) modulus of rupture

Fig. 2 shows the calculated response surface plots for LFS, WA, and MOR and their projections onto the composition triangle (contour plots) of C4–MC. The 3D surface plots shown in Fig. 2 was the graphical representation of Eqs. (1)–(3) which allowed the predictive estimates to be conducted easier and faster over the entire composition range under investigation.

Fig. 2(a) shows that higher quartz content in the formulation produced lower LFS. Feldspar and clay were the sensitive components that affected the LFS. However, higher LFS (≥ 10 %) was mainly influenced by the feldspar. It can be seen in Fig. 2(b) that the higher WA is strongly controlled by the quartz content. The mixture compositions that are able to produce lower WA (< 0.5 %) are M2, M7, M8, and M9. Higher quartz content leads to lower MOR (Figure 4.41(c)) and higher MOR is controlled by the ratio between feldspar and clay. All the mixture compositions of C4–MC are able to produce higher MOR (> 30 MPa), except for M3 and M6.

3.4Testing Restrictions of the responses subjected to fulfil the product specifications

The sample preparation conditions of the pressure and firing temperature in this work follow the industrial standard conditions of ceramic tiles in Malaysia which restricts certain adequate properties, especially LFS (5% ≤ LFS ≤ 9%), to fulfill the product specifications. Hence, based onthe BSI standard (BS EN 14411: BSI, 2006) (BSI, 2006), which specifies limits for WA and MOR, several formulations using Cambodian clays can be obtained by following the intersection procedure of the WA and MOR response surfaces to the technical and standard requirements.By doing so, the ceramic body formulations can be classified into the categories or groups based on the BSI standard.Results showed that for the C4-MC, the compositions which can be classified as Group 1 are M1, M5, M6, M8, and M10. Only M10 can be classified as BIIa, M1 and M5 as BIb, and M8 as BIa. Group 1 refers to the limits of WA ≤ 10% and MOR ≥ 18 MPa. Group 2 or BIIa (BS EN 14411): 3% < WA ≤ 6% and MOR ≥ 22 MPa. Group 3 or BIb: 0.5% < WA ≤ 3% and MOR ≥ 30 MPa. Group 4 or BIa: WA ≤ 0.5% and MOR ≥ 35 MPa.

4 CONCLUSIONS

Mixture design experiments using Cambodian claywasperformedto provide a comprehensive picture for optimization of the ceramic tile formulations. The regression models of linear firing shrinkage, water absorption and modulus of rupture, respectively, were calculated from the design of mixture experiments and the use of response surface methodologies. The models were found to be significant at the given level and presented very small variability.

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