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Prediction and fuzzy synthetic optimization of process parameters in heavy clay brick production

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Abstract

Many factors influence final clay brick properties, since the raw materials used are highly heterogeneous. Statistical analysis is rarely used, according to literature, but it would improve understanding of the overall system behavior and the quality of products.

In this study, analysis of variance (ANOVA) showed that the most important parameters influencing compressive strength (*CS*) were the quadratic terms of firing temperature, CaO and SiO_2 content in developed second order polynomial (SOP) models. Water absorption (*WA*) was mostly influenced by quadratic terms of CaO and SiO_2. The most influential interchange terms in all the models were $SiO_2 \times CaO$, $SiO_2 \times Na_2O$, $Fe_2O_3 \times Na_2O$, $CaO \times Na_2O$ and $CaO \times K_2O$. Developed SOP models, which connected the influence of major oxides content and firing temperature on *CS* and *WA*, showed the highest r^2 values (0.926–0.967) obtained in the literature so far, for these naturally occurring heavy clay raw materials. Developed models were able to predict *CS* and *WA* in a wide range of chemical composition and temperature treatment data. The implementation of the SOP model is simple using the set of equations in a spreadsheet.

The focus of this study was to determine the optimal composition and firing temperature, depending on final usage of the raw material in heavy clay brick industry. The study was conducted using fuzzy synthetic evaluation, through membership trapezoidal function, with pre-defined optimal interval values for every group of heavy clay products. The optimal samples chemical composition and firing temperature were chosen regarding the kind of the heavy clay product (I—solid bricks, II—hollow blocks and ceiling elements, and III—roof tiles).

Keywords: Prediction; Optimization; Process parameters; Clay brick

1. Introduction

Many factors influence final clay brick properties, since the raw materials used are highly heterogeneous. Complexity of these natural occurring systems is proved by an extensive never-ending research towards better understanding of the materials themselves, as well as their possible use [1–8].

Heavy clay raw materials (139 samples) used in this study contained mostly of quartz, illite, chlorite, smectite and feldspar. Calcite was detected in most of the samples, while dolomite was found in about 50% of them. Calcite and quartz are reported as a predominant phases in the Tertiary clays [9]. Kaolinite was present in minor quantity in about 20% of the samples. During firing of these materials, the most of the matrix vitrified, and only a few crystalline phases could be observed and detected using XRD. Hematite formed from the re-crystallization of iron after clay minerals (chlorite and smectite) breakdown [10], and its content slightly increased with firing temperature, as reported earlier [11]. Chlorite and smectite decomposed before 920 °C [12]. Besides hematite, no new crystalline phases could be detected. All the fired samples were constituted of residual quartz and a small amount of partly dehidroxilated mica minerals. Feldspars content

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decreased, but they were still detectable by XRD. Around quartz clasts, feldspars can also be formed from illite and calcite during firing [6]. Vitreous phase content increased with firing temperature by decomposition of phases presented in the starting material [12,13]. Having in mind that the raw materials contained generally not so high clay content (on average 22.24%), XRD of the fired samples did not reveal important conclusions. Such systems are rarely presented in the literature [9,14], even being appropriate for use in heavy clay brick industry for a range of different products. The previous was the reason that analysis presented in this study was based only on the major chemistry of raw materials, and firing temperature. Chemical composition can be taken as an important parameter, partly separately from minerals present in the material, as reported by Malaiskiene et al. [15], where the conclusion drawn was similar as in our previous work [14]: the highest effect on physical–mechanical properties of ceramics mostly depend on the amount of carbon, silicon and aluminum. The present study complements our previous research and gives more details about the influence of inputs over the outputs.

Mathematical modeling is a powerful tool, which can save time and costs for doing experimental research. Moreover, the prediction models allow better understanding of the overall system behavior, while improving the quality of products. Surprising is that the literature on the subject is very limited when heavy clay is concerned [14,16–20]. Artificial neural networks (ANN) are one of the most powerful modeling techniques [20–22], and r^2 values that were obtained by this method are 0.923–0.958 [14], for the same input and output parameters concerning heavy clay. In this study we observed that second order polynomial (SOP) models give slightly higher r^2 (0.926–0.967). Knowing that ANN's are often called "black box" [21], SOP models give more visible mutual influence of parameters concerned, and therefore amend ANN.

Polynomial models were used to investigate the effect of chemical composition and firing temperature on *CS* and *WA* of laboratory samples. The SOP equations describe effects of the input variables on the outputs, determine input variables interrelationships and represent the combined effect of all test variables in the observed outputs, enabling the experimenter to make efficient exploration of the parameters concerned. The performance of SOPs was compared to experimental results, and it is showed that they are the effective tool for optimizing the process parameters.

Achieving sustainability in manufacturing requires optimization, not just of the product, but also the manufacturing process involved in its fabrication. Improved models and optimization techniques are necessary in order to achieve optimized technological improvements and process planning for reducing energy and resource consumptions. Since the brick industry spends large amounts of energy in drying and firing processes, the proposed analysis could reduce the pollution of the environment. In this work, non-linear optimization problems were constrained by a fuzzy synthetic evaluation (FSE) method. FSE process all the parameters on the basis of predetermined weights and decrease the fuzziness by using the membership function, giving quite high sensitivity compared to other index evaluation techniques [23].

The application of second order polynomial model to natural occurring heavy clay in predicting brick properties, and later optimization of the products using fuzzy synthetic evaluation was never presented before. This combination of methods can be used as a complementary for different material research areas with non-linear parameters relationship.

2. Materials and methods

2.1. Experimental procedure

The data used in this research were obtained experimentally. A new group containing 139 heavy clays was used to prepare laboratory tiles, blocks and cubes according to the procedure described in our previous work [14]. All the samples were dried and fired in the same way, in order to have comparable results. Final firing temperatures were 800 °C, 820 °C, 850 °C, 870 °C, 900 °C, 930 °C, 950 °C, 1050 °C and 1100 °C. Major oxides chemical content was determined using old, but precise classical silicate analysis [14]. Loss on ignition (LOI) did not enter the analysis because it contained mostly of carbonates loss (already presented with CaO and MgO content) and water evaporation from clay minerals (other major oxides content), but also a small amounts of organic matter (up to 0.5%) [14,24]. Therefore, the trail analysis, which included this parameter, showed lower r^2 values. The chemical contents were recalculated to a null value of LOI, as described in the literature [24]. Major oxides content and firing temperature entered the analysis as inputs. Outputs, such as, water absorption (*WA*) and compressive strength (*CS*) of the samples were determined in the usual way [14].

2.2. Second order polynomial model

An assumption was made, that the parameters were in non-linear relationships in these kinds of materials, and it was attempted to find their inter-relationships. The SOP model behaved the best and estimated the main effect of the process variables on *CS* and *WA*, during the production of heavy clay bricks. The independent variables were: the content of major oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, K_2O, MnO and TiO_2), and firing temperature (in the range of 800–1100 °C), while the dependent variables observed were: compressive strength (for blocks *CSB* and *CSC* for cubes) and water absorption (for tiles *WAT*, for blocks *WAB*, and for cubes *WAC*). All SOP models were fitted to data collected by experimental measurements. Five models of the following form were developed to relate five dependent outputs (Y) to ten process variables (X):

$$Y_k = \beta_{k0} + \sum_{i=1}^{10} \beta_{ki} X_i + \sum_{i=1}^{10} \beta_{kii} X_i^2 + \sum_{i=1}^{10} \sum_{j=i+1}^{10} \beta_{kij} X_i X_j \quad (1)$$

where β_{kn} are constant regression coefficients; Y_k, either *CSB*, *CSC*, *WAT*, *WAB*, or *WAC*; while X_k are either metal oxides content or production temperature. The significant terms in the model were found using ANOVA for each dependent variable.

ANOVA and regression analysis for the SOP model were performed using StatSoft Statistica for Windows (version 10). The model was obtained for each dependent variable, where factors were rejected when their significance level was less then 95%.

2.3. Fuzzy synthetic optimization

The fuzzy composite operator is critical factor that will affect the final optimization results. Two fuzzy composite operators are widely used in a variety of evaluation systems. The supremum – infinum operator M (Λ, V) and the multiplication – summation operator M (●, +) determine the fuzzy algorithm of the comprehensive relative importance sets. In fuzzy synthetic avaluation models "Λ" and "V" denote the supremum and the infinum operator respectively, while "●" and "+" are the notations for the algebraic multiplication and summation separately. According to study [25], the fuzzy model operator M (Λ, V) will lose more information then M (●, +), and therefore the optimization function chosen is $O = M$(Λ, V).

An optimization was carried out using Microsoft Excel 2007. Among many existing optimization theories [26], FSE method was implemented, using the results of models proposed to represent *CSB*, *CSC*, *WAT*, *WAB*, or *WAC*, using Eq. (1). FSE is commonly used technique to solve problems with constraints involving non-linear functions. These methods aim to solve a sequence of simple problems whose solutions converge to the solution of the original problem.

Trapezoidal membership function used could be written as

$$A(x,a,m,n,b) = \begin{cases} a \le x < m, & \dfrac{x-a}{m-a} \\ m \le x < n, & 1 \\ n \le x < b, & 1 - \dfrac{x-n}{b-n} \end{cases} \quad (2)$$

x is whether *CSB*, *CSC*, *WAT*, *WAB*, or *WAC*, and the values of a, b, m and n are function parameters. Interval a–b represent the range in which measured values occurred, while range m–n is the expected optimal values range for output variables, chosen for certain products groups.

An optimization with procedure was performed according to FSE algorithm, using MicroSoft Excel 2007 to determine the workable optimum conditions for the thermal processing of heavy clay bricks.

3. Results and discussions

3.1. ANOVA and SOP models

Table 1 shows the output variables as a function of independent variables for the regression analysis. ANOVA table exhibited the significant independent variables, as well as their interactions. ANOVA was conducted to show the effects of independent on the output variables, and to point out which outputs in the varying treatment combinations were significantly affected. The effect of each variable is quantified by its sum of squares (*SS*). All the variables that entered the analysis showed significant impact on outputs, either as linear, quadratic or interchange term (or more then one of them). These results amended sensitivity analysis conclusions [14].

CS was significantly affected by most of the process variables: content of major oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, K_2O, MnO and TiO_2), and firing temperature, as shown in ANOVA table, and the most influential were quadratic members of the equation and the interchange parameters. The most important parameters influencing *CS*, according to ANOVA analysis, were quadratic terms of firing temperature, CaO and SiO_2 content, significant at $p < 0.05$ (95% confidence level). Increase in temperature affects the strength of products made of heavy clay, as stated in the literature [1–3], but it should be noted that the optimal value is required for a specific product type, not the maximum. The most influential linear term in all the models was Na_2O content, which also confirmed sensitivity analysis results [14].

Significant impact of CaO and MgO on CS is reflected in the fact that their content comes from the presence of calcite and dolomite in the raw material, which burn during the firing process. The one of the possible effects is increasing porosity and *WA* of the fired samples, while increasing *CS*. Actually, CaO can react at the calcite grain boundaries with quartz or Al_2O_3 and SiO_2 from clay mineral dehydroxylation to form calcium-sillicates, which increase the compressive strength [8,9,13,27,28]. This occurs in the case of raw materials rich in clay minerals, and consequently crystaline phase content after firing [9], which is not the present case for most of the tested samples. Raw materials tested in this research contain 22.24% of clay, and the content varies between 0.69% and even 49.31%. For raw materials with lower and average clay minerals content, CaO can partly react to form sillicates, and the rest will stay in the oxide form, especially if the grains of calcite present in the raw materials are >1 mm [29]. MgO shows similar behavior, according to literature, but can improve *CS* to a lower extent then CaO, owing to the presence of unreacted MgO [8], which is confirmed by stronger effect of CaO (quadratic terms) on *CS* and *WA*.

Quadratic term of SiO_2 content is the next significant for *CS*. Actually, the problem can arise from free quartz presence, since it has been reported that the higher amount

Table 1
Analysis of variance for the five output variables.

Source	Term	CSB	CSC	WAT	WAB	WAC
Linear	SiO_2	105.2[a]	371.5[a]	0.08 [c]	4.82 [c]	0.47 [c]
	Al_2O_3	1331.8[a]	922.6[a]	129.04[a]	198.29[a]	161.57[a]
	Fe_2O_3	312.0[a]	951.9[a]	0.00 [c]	5.67[b]	2.22
	CaO	600.5[a]	2409.6[a]	1.52 [c]	16.14[a]	4.28[b]
	MgO	1951.3[a]	5585.1[a]	104.54[a]	151.28[a]	182.26[a]
	Na_2O	2715.6[a]	1271.4[a]	569.21[a]	436.40[a]	548.01[a]
	K_2O	967.3[a]	3417.0[a]	5.31[b]	0.09 [c]	3.67[b]
	MnO	47.7[b]	1248.6[a]	0.61 [c]	3.45 [c]	3.21 [c]
	TiO_2	588.8[a]	1940.9[a]	7.19[b]	4.72 [c]	10.75[a]
	Temp	1046.8[a]	1349.3[a]	104.18[a]	192.67[a]	347.01[a]
Quad.	SiO_2	1569.3[a]	8808.5[a]	237.01[a]	261.94[a]	295.95[a]
	Al_2O_3	4.3 [c]	329.3[a]	44.40[a]	66.72[a]	54.99[a]
	Fe_2O_3	236.3[a]	817.0[a]	7.53[a]	24.97[a]	15.96[a]
	CaO	6681.3[a]	20551.4[a]	681.35[a]	671.21[a]	764.65[a]
	MgO	1433.6[a]	3232.5[a]	129.07[a]	131.51[a]	195.70[a]
	Na_2O	0.7 [c]	17.7 [c]	27.76[a]	38.34[a]	25.27[a]
	K_2O	114.9[a]	1113.2[a]	107.94[a]	145.20[a]	111.54[a]
	MnO	189.9[a]	474.1[a]	103.23[a]	105.84[a]	74.60[a]
	TiO_2	524.0[a]	135.1[b]	46.18[a]	61.31[a]	66.52[a]
	Temp	6560.2[a]	42442.1[a]	28.25[a]	85.83[a]	583.75[a]
Interchange	$SiO_2 \times Al_2O_3$	94.2[a]	215.8[a]	173.36[a]	228.03[a]	221.52[a]
	$SiO_2 \times Fe_2O_3$	187.7[a]	1338.6[a]	5.73[b]	22.15[a]	10.74[a]
	$SiO_2 \times CaO$	3970.4[a]	13456.5[a]	555.86[a]	564.11[a]	645.53[a]
	$SiO_2 \times MgO$	1878.3[a]	1995.0[a]	163.62[a]	144.59[a]	199.61[a]
	$SiO_2 \times Na_2O$	3619.0[a]	1562.5[a]	448.74[a]	360.60[a]	694.03[a]
	$SiO_2 \times K_2O$	1568.1[a]	6820.7[a]	282.17[a]	322.49[a]	340.05[a]
	$SiO_2 \times MnO$	165.3[a]	972.3[a]	47.48[a]	65.07[a]	46.13[a]
	$SiO_2 \times TiO_2$	462.0[a]	28.0 [c]	201.92[a]	260.87[a]	297.96[a]
	$SiO_2 \times$ Temp	1153.2[a]	551.5[a]	76.57[a]	110.28[a]	71.45[a]
	$Al_2O_3 \times Fe_2O_3$	814.2[a]	92.6 [c]	151.54[a]	243.64[a]	177.80[a]
	$Al_2O_3 \times CaO$	59.6[a]	46.3 [c]	159.32[a]	206.24[a]	188.09[a]
	$Al_2O_3 \times MgO$	436.2[a]	1658.3[a]	32.42[a]	18.99[a]	30.87[a]
	$Al_2O_3 \times Na_2O$	81.7[a]	5.0 [c]	71.40[a]	34.98[a]	100.06[a]
	$Al_2O_3 \times K_2O$	601.5[a]	1159.7[a]	258.00[a]	368.86[a]	327.39[a]
	$Al_2O_3 \times MnO$	26.2 [c]	433.8[a]	51.10[a]	60.29[a]	53.05[a]
	$Al_2O_3 \times TiO_2$	3.1 [c]	984.1[a]	52.05[a]	75.77[a]	87.88[a]
	$Al_2O_3 \times$ Temp	1666.0[a]	4676.5[a]	21.17[a]	43.55[a]	43.28[a]
	$Fe_2O_3 \times CaO$	832.5[a]	3709.6[a]	1.43 [c]	15.32[a]	3.37 [c]
	$Fe_2O_3 \times MgO$	2226.0[a]	5428.3[a]	108.45[a]	134.35[a]	170.39[a]
	$Fe_2O_3 \times Na_2O$	3499.3[a]	1916.9[a]	482.84[a]	403.22[a]	498.75[a]
	$Fe_2O_3 \times K_2O$	649.6[a]	4018.9[a]	1.95 [c]	2.85 [c]	0.14 [c]
	$Fe_2O_3 \times MnO$	183.3[a]	4424.0[a]	3.73 [c]	1.63 [c]	2.07 [c]
	$Fe_2O_3 \times TiO_2$	344.5[a]	852.8[a]	1.25 [c]	0.55 [c]	3.01 [c]
	$Fe_2O_3 \times$ Temp	21.3 [c]	52.3 [c]	24.14[a]	72.08[a]	101.84[a]
	$CaO \times MgO$	1757.2[a]	4637.4[a]	95.01[a]	97.39[a]	131.64[a]
	$CaO \times Na_2O$	3123.6[a]	2308.5[a]	606.22[a]	504.75[a]	679.32[a]
	$CaO \times K_2O$	1733.5[a]	5506.2[a]	322.14[a]	368.72[a]	366.38[a]
	$CaO \times MnO$	230.6[a]	10.6 [c]	83.77[a]	107.34[a]	77.04[a]
	$CaO \times TiO_2$	98.6[a]	83.9 [c]	184.63[a]	263.16[a]	308.94[a]
	$CaO \times$ Temp	232.1[a]	78.3 [c]	67.96[a]	86.82[a]	39.90[a]
	$MgO \times Na_2O$	866.3[a]	53.7 [c]	8.59[a]	0.18 [c]	69.60[a]
	$MgO \times K_2O$	105.9[a]	361.2[a]	15.78[a]	23.43[a]	15.45[a]
	$MgO \times MnO$	38.2 [c]	1674.6[a]	9.31[a]	0.74 [c]	0.60 [c]
	$MgO \times TiO_2$	11.4 [c]	31.2 [c]	6.04[b]	11.84[a]	19.26[a]
	$MgO \times$ Temp	591.5[a]	1274.2[a]	24.44[a]	65.04[a]	16.14[a]
	$Na_2O \times K_2O$	144.9[a]	35.9	24.69[a]	10.52[a]	68.21[a]
	$Na_2O \times MnO$	1599.9[a]	2025.5[a]	75.77[a]	38.58[a]	75.15[a]
	$Na_2O \times TiO_2$	138.6[a]	331.1[a]	5.08[b]	14.90[a]	3.91[a]
	$Na_2O \times$ Temp	190.2[a]	3367.0[a]	30.58[a]	103.24[a]	6.16[a]
	$K_2O \times MnO$	170.1[a]	317.2[a]	245.43[a]	290.62[a]	191.92[a]
	$K_2O \times TiO_2$	1041.2[a]	0.9 [c]	262.25[a]	354.74[a]	413.58[a]
	$K_2O \times$ Temp	544.7[a]	3551.0[a]	0.66 [c]	13.98[a]	5.32[a]

Table 1 (*continued*)

$MnO \times TiO_2$	26.1 [c]	84.4 [c]	7.19[b]	24.06[a]	15.28[a]
$MnO \times Temp$	383.1[a]	20.5 [c]	7.63[a]	7.40[a]	5.73[a]
$TiO_2 \times Temp$	975.9[a]	102.0 [c]	17.33[a]	19.59[a]	18.47[a]
Error	20338,1 [c]	62992.0 [c]	2497.94 [c]	2427.44 [c]	1701.93 [c]
Total SS	232663,0	609959.5	42952.40	44573.78	43189.34
r^2	0.913	0.897	0.942	0.946	0.961

[a]Significant at 95% confidence level.
[b]Significant at 90% confidence level.
[c]Not significant.

of SiO_2 in a ceramic body lowers its strength [25], and can even cause loosing of primary shape and product melting [30]. Increase in firing temperature causes occurence of a greater amount of liquid phase and a drop in liquid-phase viscosity. This enables a gradual eliminating of pores, increasing shrinkage, lowering porosity and increasing of the density. As temperature increases, the smallest pores coalsecence and form bigger ones. This phenomenon is also related to destruction of illites and chlorites [8,9,13].

At the same time the gas can occur in the liquid phase, one part of which can stay trapped in the material and cause the open pores formation, decreasing density [31]. It is important to note that large-sized quartz particles, together with the allotropic transformation of quartz around 573 °C, during cooling adversely affect fired mechanical strength, because of great shrinking and separation from the surrounding clay matrix [8]. Moreover, in a case of products with a dense, sintered matrix, destruction mostly begins at the location of the glassy phase, or across the grain boundaries [26].

The notable impact between interchange terms was observed by $SiO_2 \times CaO$, $SiO_2 \times Na_2O$, $Fe_2O_3 \times Na_2O$, $CaO \times Na_2O$ and $CaO \times K_2O$. CaO is more influential when interfering with SiO_2, Na_2O and K_2O content. CaO content influence level was related to the presence of free quartz (SiO_2), since highly dispersed carbonates are intense fluxes due to the formation of lowmelting eutectics [7]. Alkaline oxides also promote liquid phase formation that facilitates densification in reaction with silica and alumina [32]. The interchange terms in the polynomial equation (SiO_2, CaO, Fe_2O_3, Na_2O and K_2O) are fluxing agents. The more of them are present in the system and the higher their content, sintering process happens at the lower temperatures [31].

Also shown in Table 1 is the residual variance, where the lack of fit variation represents other contributions except the terms presented in table. A significant lack of fit would show that the model failed to represent the data in the experimental domain, at which points were not included in the regression. All SOP models had insignificant lack of fit tests, which means that all the models represented the data satisfactorily.

r^2 is also the proportion of the variability in the output variable, which is accounted for by the regression analysis. A high observed r^2 indicates that the variation was accounted and that the data fitted satisfactorily to the proposed model (SOP in this case). The r^2 values for *CSB* (0.927), *CSC* (0.926), *WAT* (0.948), *WAB* (0.951) and *WAC* (0.967) were very satisfactory and show the good fitting of the model to experimental results, even higher than those found using artificial neural networks [14].

Table 2 shows the regression coefficients for the SOP models of *CSB*, *CSC*, *WAT*, *WAB* and *WAC* used by Eq. (1) for predicting the values at optimum conditions. The analysis revealed that the linear, quadratic and interchange terms contributed substantially in all cases to generate a significant SOP model. The SOP models for all variables were found to be statistically significant and the fitting to experimental data was good. The SOP model showed in the table indicates significant influences, but not the level of it.

3.2. Validation of the model

To determine the adequacy of the SOP models, independent experiments were performed at optimum conditions for validation (Table 3). As shown in the previous ANOVA and regression coefficients tables, the predicted values were comparable to the actual values in the experiment. Very good coefficients of variation (*CV*) of less than 10% for all process variables were calculated. *CV* values higher than 15% for output variables show great influence to the statistically minor significance of its SOP model. The low *CV* values for output variables *CSB*, *CSC*, *WAT*, *WAB* and *WAC* indicated the adequacy of these models.

3.3. Fuzzy synthetic optimization

It is known that water absorption, along with open porosity and linear shrinkage, are physical parameters that can be used for optimizing the production of materials [33]. After thermal production of bricks it is essential to gain optimal values of *CSB*, *CSC*, *WAT*, *WAB* and *WAC*, depending on the final product application. It is not necessary to, for example, spend a lot of energy and get an extra hard product. It is enough to find the optimal firing temperature which would contribute to satisfying

Table 2
Regression coefficients of the SOP models for the five responses.

Term	Source	*CSB*	*CSC*	*WAT*	*WAB*	*WAC*
	Mean/Interch.	4615.3 ± 353.6	8039.3 ± 622.3	−1699.0 ± 123.9	−1762.8 ± 122.2	−1994.9 ± 102.3
Linear	SiO_2 (linear)	−89.4 ± 7.6	−178.0 ± 13.4	35.1 ± 2.7	36.7 ± 2.6	39.1 ± 2.2
	Al_2O_3 (linear)	−27.8 ± 12.5	–	35.8 ± 4.4	41.6 ± 4.3	41.2 ± 3.6
	Fe_2O_3 (linear)	−59.5 ± 23.5	−180.9 ± 41.3	−18.4 ± 8.2	−33.9 ± 8.1	−23.4 ± 6.8
	CaO (linear)	−200.3 ± 11.7	−333.8 ± 20.6	76.6 ± 4.1	77.3 ± 4.0	81.7 ± 3.4
	MgO (linear)	177.3 ± 18.2	249.4 ± 32.0	−50.1 ± 6.4	−47.1 ± 6.3	−56.5 ± 5.3
	Na_2O (linear)	−434.9 ± 33.2	−238.7 ± 58.5	164.1 ± 11.7	138.9 ± 11.5	199.8 ± 9.6
	K_2O (linear)	−330.8 ± 31.2	−628.7 ± 54.9	137.1 ± 10.9	149.4 ± 10.8	151.5 ± 9.0
	MnO (linear)	−695.9 ± 217.6	1065.8 ± 383.0	−399.4 ± 76.3	−467.9 ± 75.2	−396.5 ± 63.0
	TiO_2 (linear)	−226.7 ± 53.3	–	176.1 ± 18.7	201.1 ± 18.4	218.0 ± 15.4
	Temp (linear)	−1.1 ± 0.1	−1.8 ± 0.1	0.1 ± 0.0	0.1 ± 0.0	0.3 ± 0.0
Quad.	SiO_2 (quad.)	0.4 ± 0.0	1.0 ± 0.1	−0.2 ± 0.0	−0.2 ± 0.0	−0.2 ± 0.0
	Al_2O_3 (quad.)	–	−0.6 ± 0.2	−0.2 ± 0.0	−0.3 ± 0.0	−0.3 ± 0.0
	Fe_2O_3 (quad.)	1.3 ± 0.3	2.3 ± 0.6	0.2 ± 0.1	0.4 ± 0.1	0.3 ± 0.1
	CaO (quad.)	2.4 ± 0.1	4.2 ± 0.2	−0.8 ± 0.0	−0.8 ± 0.0	−0.8 ± 0.0
	MgO (quad.)	−2.9 ± 0.3	−4.3 ± 0.5	0.9 ± 0.1	0.9 ± 0.1	1.1 ± 0.1
	Na_2O (quad.)	–	–	−0.9 ± 0.2	−1.1 ± 0.2	−0.9 ± 0.2
	K_2O (quad.)	1.7 ± 0.6	5.3 ± 1.1	−1.7 ± 0.2	−1.9 ± 0.2	−1.7 ± 0.2
	MnO (quad.)	177.2 ± 50.5	280.0 ± 88.9	−130.7 ± 17.7	−132.3 ± 17.5	−111.1 ± 14.6
	TiO_2 (quad.)	13.0 ± 2.2	–	−3.9 ± 0.8	−4.4 ± 0.8	−4.6 ± 0.6
	Temp (quad.)	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	−0.0 ± 0.0
Interchange	$SiO_2 \times Al_2O_3$	0.3 ± 0.1	0.4 ± 0.2	−0.4 ± 0.0	−0.4 ± 0.0	−0.4 ± 0.0
	$SiO_2 \times Fe_2O_3$	0.8 ± 0.2	2.2 ± 0.4	–	0.3 ± 0.1	0.2 ± 0.1
	$SiO_2 \times CaO$	2.1 ± 0.1	3.8 ± 0.2	−0.8 ± 0.0	−0.8 ± 0.0	−0.8 ± 0.0
	$SiO_2 \times MgO$	−1.9 ± 0.2	−2.0 ± 0.3	0.6 ± 0.1	0.5 ± 0.1	0.6 ± 0.1
	$SiO_2 \times Na_2O$	4.8 ± 0.3	3.2 ± 0.6	−1.7 ± 0.1	−1.5 ± 0.1	−2.1 ± 0.1
	$SiO_2 \times K_2O$	3.2 ± 0.3	6.7 ± 0.6	−1.4 ± 0.1	−1.5 ± 0.1	−1.5 ± 0.1
	$SiO_2 \times MnO$	6.8 ± 2.1	−16.6 ± 3.7	3.7 ± 0.7	4.3 ± 0.7	3.6 ± 0.6
	$SiO_2 \times TiO_2$	2.7 ± 0.5	–	−1.8 ± 0.2	−2.1 ± 0.2	−2.2 ± 0.1
	SiO_2 × Temp	0.0 ± 0.0	0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0
	$Al_2O_3 \times Fe_2O_3$	−1.4 ± 0.2	–	0.6 ± 0.1	0.8 ± 0.1	0.7 ± 0.1
	$Al_2O_3 \times CaO$	0.4 ± 0.2	–	−0.7 ± 0.1	−0.8 ± 0.1	−0.8 ± 0.1
	$Al_2O_3 \times MgO$	−1.4 ± 0.3	−2.8 ± 0.5	0.4 ± 0.1	0.3 ± 0.1	0.4 ± 0.1
	$Al_2O_3 \times Na_2O$	1.1 ± 0.5	–	−1.0 ± 0.2	−0.7 ± 0.2	−1.2 ± 0.1
	$Al_2O_3 \times K_2O$	2.7 ± 0.4	3.7 ± 0.8	−1.8 ± 0.2	−2.1 ± 0.1	−2.0 ± 0.1
	$Al_2O_3 \times MnO$	–	18.8 ± 6.3	6.5 ± 1.2	7.0 ± 1.2	6.6 ± 1.0
	$Al_2O_3 \times TiO_2$	–	−7.5 ± 1.6	−1.7 ± 0.3	−2.1 ± 0.3	−2.2 ± 0.3
	Al_2O_3 × Temp	0.0 ± 0.0	0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0
	$Fe_2O_3 \times CaO$	3.4 ± 0.5	7.3 ± 0.8	–	0.5 ± 0.2	–
	$Fe_2O_3 \times MgO$	−6.8 ± 0.6	−10.6 ± 1.0	1.5 ± 0.2	1.7 ± 0.2	1.9 ± 0.2
	$Fe_2O_3 \times Na_2O$	12.6 ± 0.8	9.3 ± 1.5	−4.7 ± 0.3	−4.3 ± 0.3	−4.8 ± 0.2
	$Fe_2O_3 \times K_2O$	4.1 ± 0.6	10.3 ± 1.1	–	–	–
	$Fe_2O_3 \times MnO$	−16.8 ± 4.9	−82.8 ± 8.6	–	–	–
	$Fe_2O_3 \times TiO_2$	−5.5 ± 1.2	−8.6 ± 2.0	–	–	–
	Fe_2O_3 × Temp	–	–	−0.0 ± 0.0	-0.0 ± 0.0	−0.0 ± 0.0
	CaO × MgO	−3.1 ± 0.3	−5.0 ± 0.5	0.7 ± 0.1	0.7 ± 0.1	0.8 ± 0.1
	CaO × Na_2O	8.5 ± 0.6	7.3 ± 1.1	−3.8 ± 0.2	−3.4 ± 0.2	−4.0 ± 0.2
	CaO × K_2O	6.0 ± ± 0.6	10.8 ± 1.0	−2.6 ± 0.2	−2.8 ± 0.2	−2.8 ± 0.2
	CaO × MnO	15.0 ± 3.9	–	9.0 ± 1.4	10.2 ± 1.3	8.6 ± 1.1
	CaO × TiO_2	2.6 ± 1.0	–	−3.5 ± 0.4	−4.2 ± 0.4	−4.5 ± 0.3
	CaO × Temp	0.0 ± 0.0	-	−0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0
	MgO × Na_2O	7.2 ± 1.0	-	−0.7 ± 0.3	–	-2.0 ± 0.3
	MgO × K_2O	1.9 ± 0.7	3.4 ± 1.2	−0.7 ± 0.2	−0.9 ± 0.2	−0.7 ± 0.2
	MgO × MnO	–	-60.1 ± 10.2	−4.5 ± 2.0		–
	MgO × TiO_2	–	–	–	1.1 ± 0.4	1.4 ± 0.4
	MgO × Temp	0.0 ± 0.0	0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0	−0.0 ± 0.0
	$Na_2O \times K_2O$	3.7 ± 1.2	–	−1.5 ± 0.4	−1.0 ± 0.4	−2.5 ± 0.3
	Na_2O × MnO	76.8 ± 7.5	86.4 ± 13.3	−16.7 ± 2.6	−11.9 ± 2.6	−16.6 ± 2.2
	$Na_2O \times TiO_2$	−6.3 ± 2.1	−9.7 ± 3.7	–	2.1 ± 0.7	–
	Na_2O × Temp	−0.0 ± 0.0	−0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
	K_2O × MnO	24.2 ± 7.3	33.1 ± 12.8	29.1 ± 2.6	31.7 ± 2.5	25.7 ± 2.1

Table 2 (*continued*)

Term	Source	*CSB*	*CSC*	*WAT*	*WAB*	*WAC*
	$K_2O \times TiO_2$	17.8 ± 2.2	–	-8.9 ± 0.8	-10.4 ± 0.7	-11.2 ± 0.6
	$K_2O \times$ Temp	0.0 ± 0.0	0.0 ± 0.0	–	-0.0 ± 0.0	-0.0 ± 0.0
	$MnO \times TiO_2$	–	–	–	18.3 ± 5.1	14.6 ± 4.3
	$MnO \times$ Temp	0.1 ± 0.0	–	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
	$TiO_2 \times$ Temp	0.0 ± 0.0	–	-0.0 ± 0.0	-0.0 ± 0.0	-0.0 ± 0.0

Table 3
Predicted and observed outputs at optimum conditions.

Parameters	Predicted	Observed	Standard deviation	Coeff. of variation
CSB	13.16–16.91	16.63	2.112	5.67
CSC	28.66–33.33	34.14	1.706	2.73
WAT	19.37–21.44	21.25	0.765	3.37
WAB	19.61–21.63	21.33	1.361	5.95
WAC	19.26–20.96	21.01	2.220	12.08
CSB	27.72	28.85	1.948	6.75
CSC	53.02	52.52	1.926	3.67
WAT	15.13	15.12	0.900	5.95
WAB	15.46	15.13	0.891	5.89
WAC	14.95	14.7	1.832	12.46
CSB	50.6	50.1	2.006	4.00
CSC	79.21	79.43	1.462	1.84
WAT	10.4	10.20	0.627	6.15
WAB	11.15	10.78	1.511	14.02
WAC	10.69	10.5	1.520	11.62

properties of a certain product. The choice of the best process conditions (firing temperature) and raw material chemical content for production of bricks depends on the application that would be given to the product.

Multiple fuzzy synthetic optimization of the five output variables was accomplished in order to find the content of major oxides (SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, K_2O, MnO and TiO_2), and firing temperature that give optimal values of *CSB*, *CSC*, *WAT*, *WAB*, and *WAC*. Trapezoidal membership function was used as optimization method, according to Eq. (2), in which *a*–*b* covered the complete interval where obtained values were found, and *m*–*n* represented the optimal values for observed product group (Table 4). The optimum ranges were given based on our experience with heavy clay, knowing that each ceramic product requires clays having particular and appropriate characteristics [3].

Group I is suitable for the production of solid bricks, due to the low clay content and loess raw material nature, which causes low plasticity. In order to improve the product quality, more plastic clay should be added in the mixture for hollow blocks shaping. *Group II* can be used for producing hollow bricks and blocks, as well as ceiling elements *Group III* can be, after grinding below 0.5 mm to avoid the appearance of lime corns, used in roof tiles and facade elements production. The samples belonging to *Groups II and III* can be also used in a light-weighted bricks, as a primary raw material.

Table 4
Trapezoidal membership function parameters.

Group	Parameter	*CSB*	*CSC*	*WAT*	*WAB*	*WAC*
–	*a*	4.95	8.45	4.48	4.44	4.00
	b	137.18	144.74	30.95	31.38	27.20
I	*m*	13.00	25.00	19.00	19.00	19.00
	n	17.00	35.00	22.00	22.00	22.00
II	*m*	24.00	45.00	14.00	14.00	14.00
	n	28.00	55.00	16.00	16.00	16.00
III	*m*	50.00	65.00	8.00	8.00	8.00
	n	70.00	75.00	13.00	13.00	13.00

The objective function (*F*) is the mathematical function whose maximum would be determined, by summing the FSE results for of the five models, according Eq. (1). Both groups of output parameters (*CS* and *WA*) have the same influence on the function *F* (0.5 each):

$$F(\mathrm{SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2}O, K_2\mathrm{O, MnO, TiO_2, Temp.}) = 0.25(\overline{CSB}+\overline{CSC})+0.16(\overline{WAT}+\overline{WAB}+\overline{WAC}) \quad (3)$$

The maximum of function *F* represents the optimal parameters for major oxides content and processing temperature, and also the optimum *CSB*, *CSC*, *WAT*, *WAB*, or *WAC*. The graphs of the dependent variables with significant parameters were determined using objective function to determine optimum production conditions, plotted on optimization graphic. If the value of membership trapezoidal function is close to 1, it shows the tendency of tested processing parameters of being optimal. After the optimum conditions were established, separate experiments were performed for validations of the models (Table 3). The objective functions, regarding SiO_2/ Al_2O_3, and Fe_2O_3/ CaO content were shown in the surface plots, for *Groups I*, *II and III* (Figs. 1–3). It can be seen that not very high number of tested samples shows optimal behavior.

The combination of working conditions, that simultaneously optimize *CSB*, *CSC*, *WAT*, *WAB* and *WAC* functions, was given in Table 5. Objective function gained values 1.00 for *Groups I and II*, and 0.98 for the *Group III*, which presents very satisfying values.

Group I showed mostly the highest free quartz content (SiO_2/Al_2O_3), and carbonates content, and subsequently the lowest Fe_2O_3 content. This indicates that these were the

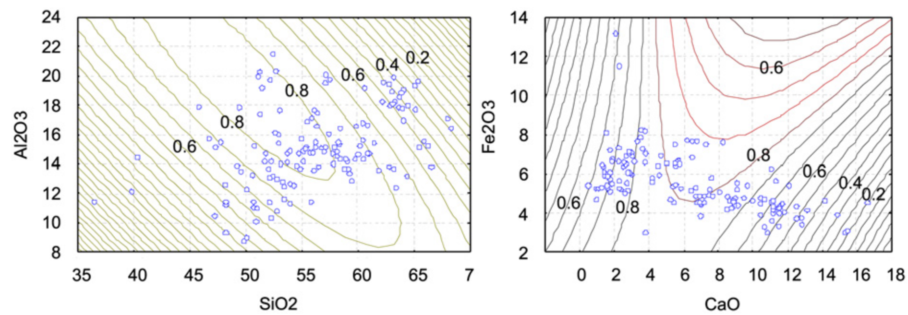

Fig. 1. Objective function for *Group I* products.

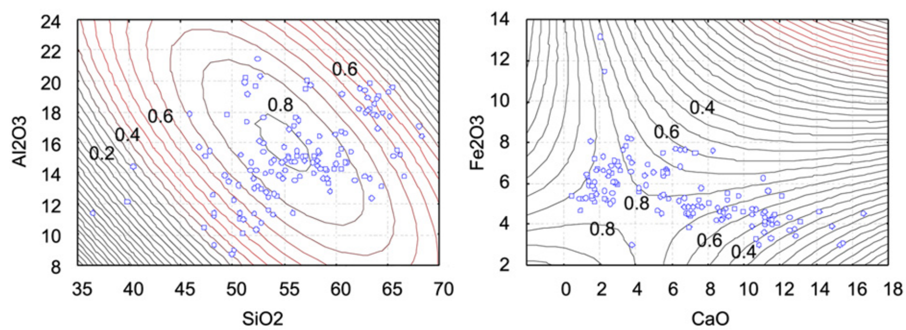

Fig. 2. Objective function for *Group II* products.

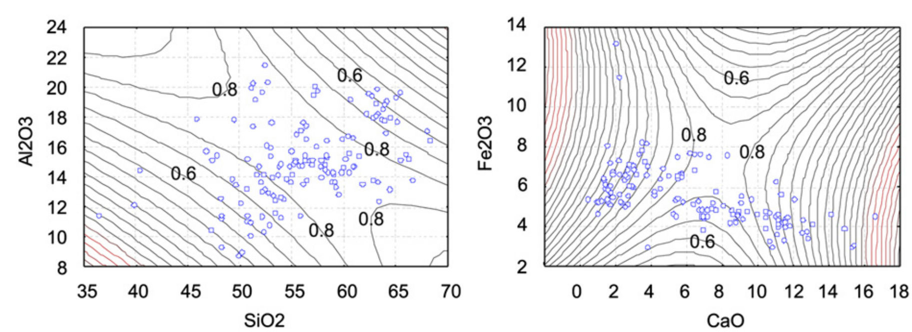

Fig. 3. Objective function for *Group III* products.

Table 5
Optimum process parameters.

Group	Inputs										Outputs				
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	MnO	TiO_2	Temp	*CSB*	*CSC*	*WAT*	*WAB*	*WAC*
I	55.33	15.20	4.26	8.76	2.18	0.88	1.39	0.10	0.53	950	13.92	29.83	20.68	20.44	20.12
	50.93	10.15	4.42	11.58	4.10	1.35	1.44	0.10	0.90	950	15.31	32.22	21.08	21.04	20.2
	50.67	11.47	4.73	10.63	3.58	1.26	2.24	0.08	1.08	950	16.91	33.33	20.14	20.18	19.47
	54.49	13.91	5.09	8.05	3.7	1.14	1.7	0.08	0.46	950	13.32	32.45	19.66	19.61	19.39
	52.08	13.04	4.16	11.13	1.65	1.16	1.64	0.20	0.39	950	16.58	32.39	21.44	21.63	20.96
	52.09	11.41	4.81	8.93	3.57	2.59	3.11	0.15	0.68	1000	13.88	29.69	19.37	19.92	19.37
	54.49	13.91	5.09	8.05	3.7	1.14	1.7	0.08	0.46	1000	15	29.56	19.79	19.85	19.26
	58.34	15.35	3.89	6.93	2.21	1.29	1.83	0.09	0.38	1050	13.16	28.66	20.13	20.85	20.39
II	57.07	19.54	7.13	2.78	1.87	1.65	2.59	0.12	0.38	900	27.72	53.02	15.13	15.46	14.95
III	67.99	17.07	5.41	0.43	1.53	0.70	1.57	0.01	0.78	900	50.60	79.21	10.40	11.15	10.69

samples of the lowest "quality", but that they can still be used in brick industry. *Group II* had the lowest free quartz and the highest Fe_2O_3 content. According to literature, the content above 5% Fe_2O_3 presents high sinter able heavy clays [7]. *Group III* contained the most TiO_2 and Al_2O_3, which is related to clay minerals content and regarded this group as the most plastic.

The ranges of CS and WA values were set on the basis of our experience, and show the highest *CS* and the lowest *WA* for *Group III*. The presence of moderate amounts of carbonates in *Groups II and III* (2–5%) permits manufacture of light clay building products with good mechanical properties.

The optimal firing temperature for *Group I* is mostly 950 °C. Actually, most of these samples are of loess nature, where the organogenic nature of calcite contributes to the formation of a local reducing medium in firing, affecting the transition of Fe^{3+} to Fe^{2+} and facilitates sintering at relatively lower temperatures [7], unlike the last three optimal samples from the same group. Optimal temperature for *Groups II and III* (900 °C) is the lowest because of higher clay, and lower carbonates content, compared to *Group I*.

These conclusions correspond very well to the effects of parameters revealed in the SOP model.

4. Conclusions

An intention of presented research was to promote the use of statistical methods in natural occurring raw materials used in brick industry. Second order polynomial models, which connected major oxides content and firing temperature to compressive strength and water absorption of different products, showed the highest r^2 values. The most influential parameter in the linear part of the models was Na_2O; important quadratic terms were temperature, CaO and SiO_2; and among interchange parameters $SiO_2 \times CaO$, $SiO_2 \times Na_2O$, $Fe_2O_3 \times Na_2O$, $CaO \times Na_2O$ and $CaO \times K_2O$ stand out. It was seen that, regardless of all the other raw material and processing parameters (particle size distribution, shaping moist, plasticity etc.) the proposed models performed the prediction accurately.

ANOVA analysis revealed that all the observed inputs were statistically significant affecting the outputs, while highlighting the importance of certain members of the polynomial, which is complemented by optimizing the chemical content and firing temperature for a variety of heavy clay products. The optimal samples chemical composition and firing temperature, within three heavy clay products groups (*I—solid* bricks, *II*—hollow blocks and ceiling elements, and *III*—roof tiles), were chosen by fuzzy synthetic evaluation.

Second order polynomial models and fuzzy synthetic optimization synergize and can be used together for different material research areas with non-linear parameters relationship.

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