Response surface method as a tool for heavy clay firing process optimization: Roofing tiles

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Abstract

Heavy clay samples collected in close vicinity of Toplička Mala Plana, Serbia, were surveyed to examine their possible use in heavy clay industry. The representative raw material, which contained the lowest content of clay minerals and the highest content of carbonates, was enriched with two more plastic clays. Chemical and mineralogical composition, as well as particle size distribution, were determined to distinct the samples. The samples in the form of tiles, hollow blocks and cubes were prepared following the usual practice in ceramic laboratories. The effect of process parameters, such as temperature (850–950 °C) and concentration of the added clays (both in the range of 0–10 wt.%), were investigated in terms of compressive strength, water absorption, firing shrinkage, weight loss during firing and volume mass of cubes. The optimal conditions were determined by the response surface method, coupled with the fuzzy synthetic evaluation algorithm, using membership trapezoidal function, and showed that these materials can be used for roofing tiles production.

Keywords: *optimization, heavy clays, firing*

I. Introduction

Heavy clay products properties depend on raw material characteristics, especially on mineralogical composition [1], chemical composition [2], particle size distribution [3] and plasticity [4]. Firing conditions (temperature, heating rate and kiln atmosphere) also influence final product properties to a great extent [5–7]. Recently mathematical tools have been used intensively to describe ceramic systems behaviour more precisely, and to define the link between input and output parameters [8,9]. The response surface method (RSM) has been proven as useful method for determining the influence of process variables on a group of responses of interest for the process and effects studied [10]. The main advantage of RSM is reduced number of experimental runs that provide sufficient information for statistically valid results. RSM is proven to be an effective tool for optimizing ceramic systems [9,11,12].

Local brick factory, in Toplička Mala Plana, Serbia, uses neighboring raw materials to produce various types of heavy clay hollow blocks and ceiling elements. The aim of this research was to test the possible use of these materials in the production of roofing tiles, based on laboratory tests. The second order polynomial (SOP) models for determination of ten response variables were developed and correlation coefficients between them were found. The response surface method (RSM), coupled with the fuzzy synthetic evaluation (FSE) algorithm, was used to determine the optimal process conditions and define the kind of heavy clay product that best suited the studied raw materials.

II. Experimental

2.1 Sample preparation

Heavy clay samples were collected from Toplička Mala Plana area in Serbia. Representative raw material (MP6) was chosen, to which two more plastic clays (MP1 and MP2) were added in the amount between 0–10 wt.%. The samples were prepared following the usual practice in ceramic laboratories. Laboratory samples were produced in the form of tiles (120×50×14 mm), hollow blocks with vertical voids (55.3×36×36 mm) and cubes (30×30×30 mm). After

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shaping, the samples were dried in air, and later in a laboratory dryer at 105±5 °C to a constant mass. Firing was done in the oxygen atmosphere kiln, with average heating rate of 1.4 °C/min up to 610 °C, and later with the rate of 2.5 °C/min until the final given temperature is reached, at which the samples were treated for 2 h [8,11]. Firing was conducted at 850 °C, 900 °C and 950 °C

2.2 Characterization techniques

The content of major oxides in the clay samples was determined by using classical silicate analysis [8,13], while all the measurements were performed in triplicate. The mineralogical analysis was carried out by X-ray diffraction (XRD) using a powder diffractometer (Philips PW-1050), with λCu-Kα radiation and scanning speed of 0.05 °/s, both on powder (bulk samples) and oriented aggregates [14]. Particle size distribution (PSD) was determined by granulometry analysis. Due to the size of particles, it was necessary to do sedimentation analysis (fractions under 0.063 mm) [11,15].

Compressive strength (*CS*) was determined with the laboratory hydraulic press Alfred Amsler, CHD [8]. Three specimens for each combination of sample shape (blocks and cubes) and firing temperature were tested. The samples were flattened to ensure that the surfaces were parallel. Compressive strength is then tested on single samples (without mortar usage), with bottom area of 0.002 m^2 for blocks and 0.0009 m^2 for cubes, and a loading rate of 0.6 kN/s. The strength results reported were the average of three specimens with a variation of no more than 10%.

Water absorption (*WA*) was evaluated by the soaking samples in water for 24 h, according to the standard SRPS EN 771-1, and later volume mass of the cubes (*VMC*) was calculated as weight of fired samples divided by the volume of water displaced by the sample (previously saturated with water) in the measuring cylinder [11]. Weight loss during firing (*WLF*) was determined by measuring the samples on a scale with 0.001 g precision, and was calculated as a ratio between the weight lost during firing and the starting weight of sample, and was expressed in weight percent [wt.%]. Firing shrinkage (*FS*) was obtained by the relative variation in length of the tiles using a caliper (precision of ±0.01 mm).

2.3 Optimization study

Processing variables play a very important role on the characteristics of the final ceramic products [7]. In our study the chosen independent variables were temperature (850, 900 and 950 °C) and concentrations of MP1 and MP2 clays (0, 5 and 10 wt.%). The accepted experimental design was taken from Box and Behnken [16]. The dependent variables were the responses: compressive strength of blocks - *CSB* and cubes - *CSC*; water absorption of tiles - *WAT*, blocks - *WAB* and cubes - *WAC*; firing shrinkage *FS*; weight loss during firing of tiles - *WLFT*, blocks - *WLFB* and cubes - *WLFC*; and volume mass of cubes - *VMC*. The process variables were coded according to Box and Behnken's central composite full factorial design (2 level-3 parameter) with 15 runs (1 block), where "-1" denotes low value of the independent variables (850 °C and 0 wt.% addition), "0" was used for medium values (900 °C and 5 wt.% addition), and "+1" for high values (950 °C and 10 wt.% addition).

The experimental data used for the optimization study were the obtained parameters using central composite full factorial design (3 level-3 parameter) with 27 runs (1 block) [16]. A model was fitted to the response surface generated by the experiment. The model used was a function of the process variables:

$$Y_k = f_k(temperature, MP1, MP2) \tag{1}$$

The second order polynomial (SOP) models were developed to relate ten responses (*CSB*, *CSC*, *WAT*, *WAB*, *WAC*, *FS*, *WLFT*, *WLFB*, *WLFC* and *VMC*) to three process variables, i.e. temperature and concentrations (MP1 and MP2) [11,16], according to equation:

$$Y_k = \beta_{k0} + \sum_{i=1}^{3} \beta_{ki} X_i + \sum_{i=1}^{3} \beta_{kii} X_i^2 + \sum_{i=1}^{2} \sum_{j=i+1}^{3} \beta_{kij} X_i X_j \tag{2}$$

where β_{kn} are constant regression coefficients. The significant terms (linear, quadratic and multiplied terms) in the model were found using ANOVA for each response.

The response surface method (RSM), coupled with the fuzzy synthetic evaluation (FSE) algorithm, was used to determine the optimal process conditions and defines the kind of quality heavy clay product.

The analysis of variance (ANOVA) and response surface method (RSM) were performed using StatSoft Statistica program. The model was obtained for each dependent variable (or response), where factors were rejected when their significance level was less than $p<0.05$, confidence limit 95%. The same program was used for generation of graphs and contour plots. The fuzzy synthetic optimization method was implemented using the results of the proposed models, to represent *CSB*, *CSC*, *WAT*, *WAB*, *WAC*, *FS*, *WLFT*, *WLFB*, *WLFC* and *VMC*, according to Eq. 2. 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 [11].

Trapezoidal membership function used, could be written as:

$$A(x,a,m,n,b) = \begin{cases} a \le x < m, & \dfrac{x-a}{m-a} \\ m \le x < n, & 1 \\ n \le x < b, & 1 - \dfrac{x-n}{b-n} \end{cases} \tag{3}$$

where x is whether *CSB*, *CSC*, *WAT*, *WAB*, *WAC*, *FS*, *WLFT*, *WLFB*, *WLFC* or *VMC*, and the values of a, b, *m* and *n* are function parameters. Interval *a* – *b* represent the range in which measured values occur, while range *m* – *n* is the expected optimal values range for response variables, chosen for certain products groups.

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

III. Results and discussion

3.1 Sample characteristics

The chemical composition and fraction content of clay, silt and quartz, calculated according to the measured particle size distribution of the used materials are given in Table 1 and Table 2, respectively. Post ANOVA's Tukey HSD test (honestly significant difference), at the $p<0.05$ significant level (95% confidence limit), was performed in order to access the statistically significant differences within each chemical composition assay. Descriptive statistical analyses, for calculating the means and the standard error of the mean, were performed using Microsoft Excel 2007 software. The obtained results were expressed as the mean ± standard deviation (Table 1).

Tukey's test showed that the similar SiO_2 content was found in all the samples. According to SiO_2, Al_2O_3, Fe_2O_3 and TiO_2 content, which build clay minerals, it is obvious that the the sample MP1 contained the highest clay content and the sample MP6 the lowest. The molar fractions of SiO_2 and Al_2O_3 showed the existence of free SiO_2 (present as quartz) [11,17].

Chemical, mineralogical and particle size distribution analysis results showed that the highest quartz content is found in the sample MP2 and the lowest in the sample MP6. Since silt fraction can contain quartz, and also silt can be of a clay sized fraction, particle size distribution results did not give a clear picture of clay minerals content. All the samples contain similar carbonates composition, while the sample MP6 showed the highest calcite and dolomite content. According to the particle size distribution analysis, all the samples belong to silt loam (Unified Soil Classification System).

Mineralogical analysis revealed similarities between the tested samples (Fig. 1): they consisted mostly of quartz; then illite (mica), chlorite and smectite. Low quantities of calcite, dolomite and feldspar (plagioclase) are detected on the XRD patterns of all samples. Kaolinite is also found in traces in the case of the MP1 and MP2 samples, which were used to enrich the sample MP6.

When adding solely MP1 or MP2 to the raw material MP6, the values of the response variables vary

Table 1. Chemical composition of the used raw materials

	Chemical composition [wt.%]		
	Sample MP6	Sample MP1	Sample MP2
SiO_2	$53.70±0.28^a$	$55.27±0.20^b$	$57.69±0.43^c$
Al_2O_3	$14.13±0.04^a$	$15.02±0.14^c$	$14.72±0.05^b$
Fe_2O_3	$6.94±0.07^a$	$7.85±0.06^c$	$7.68±0.00^b$
CaO	$4.63±0.06^c$	$3.45±0.03^b$	$3.31±0.05^a$
MgO	$4.15±0.02^c$	$3.42±0.06^b$	$2.97±0.02^a$
Na_2O	$1.14±0.01^a$	$1.50±0.01^b$	$2.41±0.03^c$
K_2O	$2.41±0.01^a$	$3.19±0.04^b$	$2.41±0.01^a$
MnO	$0.01±0.00^a$	$0.01±0.00^a$	$0.01±0.00^a$
TiO_2	$0.87±0.01^a$	$1.15±0.01^b$	$1.13±0.01^b$
SO_3	$0.01±0.00^a$	$0.01±0.00^a$	$0.01±0.00^a$
L.O.I.*	$12.00±0.12^c$	$9.12±0.04^b$	$7.67±0.06^a$

[a,b,c] Values with the same letter, written in superscript, are not statistically different at the $p<0.05$ level, 95% confidence limit, according to post-hoc Tukey's HSD test

*L.O.I. represents loss on ignition values while heating from 105 °C to 1000 °C.

Table 2. Fraction content of clay, silt and quartz, calculated according to the measured particle size distribution of the used raw materials

	Clay fraction content [%]	Silt fraction content [%]	Quartz fraction content [%]
Sample MP6	26.26	59.72	14.02
Sample MP1	20.46	61.16	18.38
Sample MP2	18.95	54.60	26.46

Table 3. Experimental design (central composite fractional factorial design, 2 level-3 parameter, with 15 runs, 1 block)

Sample No.	Process parameters *T*	*[MP1]*	*[MP2]*	Responses *CSB* [MPa]	*CSC* [MPa]	*WAT* [%]	*WAB* [%]	*WAC* [%]	*FS* [%]	*WLFT* [wt.%]	*WLFB* [wt.%]	*WLFC* [wt.%]	*VMC* [%]
1	-1	-1	0	41.55	76.25	13.26	12.73	9.46	-0.04	7.18	8.09	8.82	1.93
2	-1	1	0	39.81	73.30	14.01	13.51	9.95	-0.24	7.05	7.91	8.45	1.94
3	1	-1	0	40.58	76.10	13.24	12.39	8.50	0.16	7.29	8.34	9.28	1.95
4	1	1	0	41.78	75.93	12.89	12.10	8.60	0.14	7.29	8.16	9.03	1.93
5	0	-1	-1	38.89	73.22	13.85	12.96	9.08	-0.04	7.28	8.15	8.88	1.93
6	0	-1	1	39.40	71.29	13.92	12.97	9.61	-0.09	7.36	8.24	8.93	1.95
7	0	1	-1	39.17	71.55	14.03	13.18	9.17	-0.06	7.23	8.23	9.09	1.95
8	0	1	1	37.66	68.32	14.31	13.50	10.07	-0.29	7.09	8.02	8.71	1.95
9	-1	0	-1	41.71	76.28	13.29	12.59	9.30	-0.04	7.19	7.96	8.73	1.94
10	-1	0	1	39.18	72.71	14.02	13.46	10.21	-0.24	7.08	7.94	8.55	1.93
11	1	0	-1	41.22	76.62	13.17	12.21	8.34	0.19	7.26	8.17	9.28	1.95
12	1	0	1	39.80	73.01	13.71	12.83	9.37	-0.07	7.22	8.12	8.90	1.94
13	0	0	0	39.49	71.78	13.82	12.89	9.38	-0.08	7.21	8.23	8.94	1.92
14	0	0	0	39.60	72.74	13.63	12.99	9.34	-0.08	7.31	8.10	8.84	1.92
15	0	0	0	39.36	72.78	13.78	12.96	9.45	-0.08	7.17	8.09	8.95	1.92

with the temperature to a small extent (Table 3). Higher content of clay minerals causes better particle packing as well as better sinterability, which can improve compressive strength (*CS*). Carbonates react with clay minerals, giving calcium and magnesium silicates. The rest of the unreacted carbonates burn out and produce pores, thus increasing water absorption (*WA*) and decreasing compressive strength (*CS*) [8,11]. Thus, the observed fluctuations of the responses (increase and decrease with temperature) are determined by the actual number of carbonates grains in contact with clay minerals as well. The highest compressive strength occurred at 950 °C for the block-sample (*CSB*) with 10 wt.% MP1 and 5 wt.% MP2, and the cube-sample (*CSC*) with 5 wt.% MP1 and 0 wt.% MP2 (Table 3).

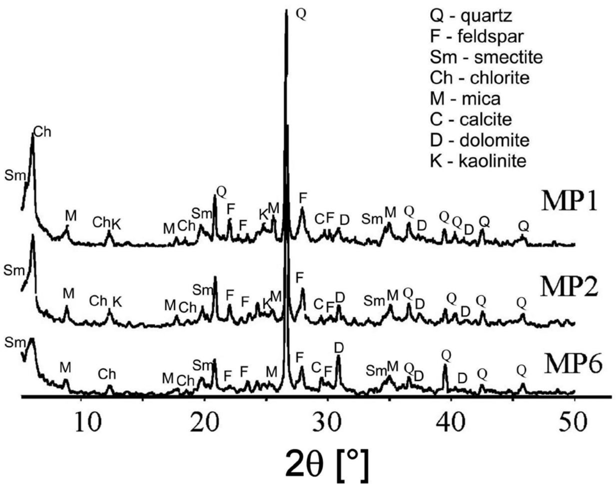


Figure 1. XRD spectra of tested samples

The greatest water absorption is observed at 900 °C in the case of tiles with 10 wt.% of MP1 and 10 wt.% of MP2 (Table 3). Almost all the samples expand except samples 3 and 4, fired at 950 °C. Weight loss during firing at 950 °C was highest in the case of sample 3 blocks and cubes (Table 4). Volume mass was similar in all the fired samples.

3.2 Optimization results

In this study, ANOVA was conducted to show the significant effects of the independent variables on the responses (dependent variables) and determine which of the responses were significantly affected by the varying treatment combinations. Table 4 shows the ANOVA calculation regarding the response models developed when the experimental data were fitted to a response surface. The response surface used the second order polynomial models in order to predict the function responses for all the dependent variables. The linear term of temperature was statistically significant for all the responses. MP1 and MP2 concentrations showed great influence on all of the output parameters, whether the linear, quadratic or multiplied term (Table 4). Additives showed most important influence on *VMC*.

The analysis revealed that the linear terms contributed substantially in the majority of cases to the generation of significant SOP models. The SOP models for all variables were found to be statistically significant and the response surfaces were fitted to these models. The quadratic terms for temperature were found insignificant for all SOP models, while most of concentration terms were found statistically significant at $p<0.05$ or $p<0.10$ level. The residual variance also shown in

Table 4. Analysis of variance for the ten responses, 3 factors, 1 block, 27 runs

Term	Source	*dF*	*CSB*	*CSC*	*WAT*	*WAB*	*WAC*	*FS*	*WLFT*	*WLFB*	*WLFC*	*VMC*
Linear	*T*	1	0.68**	6.41*	0.31**	0.76*	1.13*	0.08*	0.06*	0.06*	0.16*	4.17E-06
	[*MP1*]	1	0.97**	0.04	0.04	0.05	0.00	0.00	0.00	0.01	0.01	3.16E-05
	[*MP2*]	1	3.96*	18.34*	0.47*	0.38*	0.39**	0.04*	0.00	0.00	0.01	3.34E-04*
Quad.	*T*	1	0.09	0.42	0.06	0.07	0.03	0.00	0.00	0.00	0.00	1.25E-05
	[*MP1*]	1	3.07*	9.24*	0.20**	0.26**	0.79*	0.03*	0.00	0.00	0.03**	1.10E-05
	[*MP2*]	1	1.16**	4.99*	0.04	0.01	0.01	0.00	0.01**	0.02**	0.02**	4.90E-04*
Product	*T* × [*MP1*]	1	2.59*	3.77*	0.30**	0.36*	0.16	0.03*	0.01**	0.01	0.09*	1.00E-05
	T × [*MP2*]	1	0.23	1.44	0.12	0.13	0.05	0.01	0.00	0.00	0.02**	2.22E-06
	[*MP1*] × [*MP2*]	1	9.13*	39.26*	0.70*	0.38*	0.18	0.03*	0.01**	0.02	0.00	1.44E-04*
Error	Error	17	1.20	1.59	0.07	0.13	0.08	0.01	0.00	0.00	0.00	7.06E-06
r^2			93.87	97.97	88.17	84.08	83.71	91.79	84.59	87.69	94.23	93.47

*Significant at 95% confidence level,
**Significant at 90% confidence level,
Unmarked values are statistically not significant

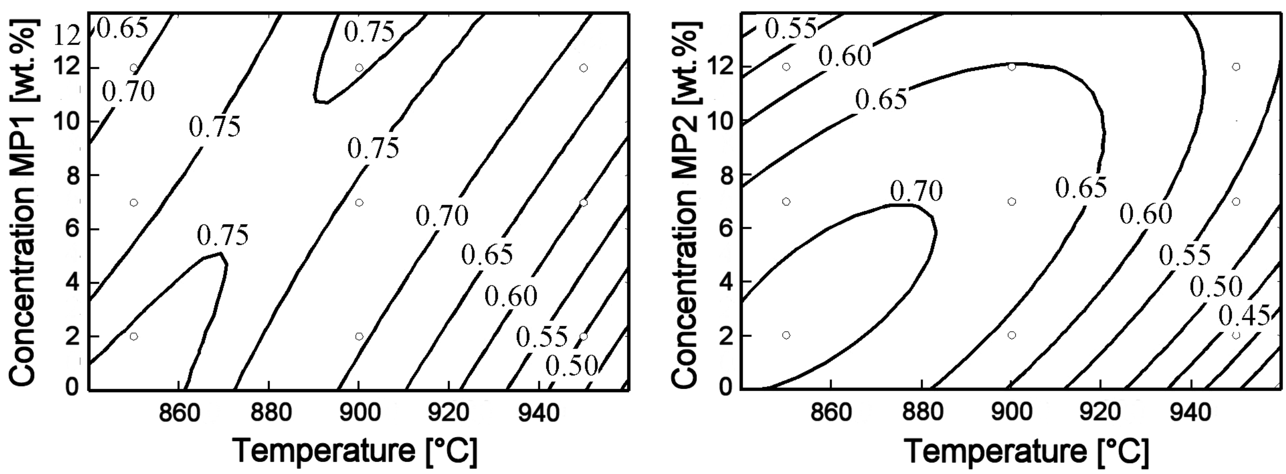
Table 4, where the error term represents the lack of fit variation, i.e. it represents other contributions except the linear, quadratic and cross product terms. All SOP models had insignificant lack of fit tests, which means that all the models represented the data satisfactorily. A high r^2 is indicative that the variation was accounted and that the data fitted satisfactorily to the proposed SOP models. The r^2 values for *CSB* (93.87), *CSC* (97.97), *WAT* (88.17), *WAB* (84.08), *WAC* (83.71), *FS* (91.79), *WLFT* (84.59), *WLFB* (87.69), *WLFC* (94.23) and *VMC* (93.47) were very satisfactory and show a good fit of the model to experimental results.

Water absorption, along with open porosity and linear shrinkage, are physical parameters that can be used for optimizing the production of materials [11]. It is essential to gain optimal values of *CSB*, *CSC*, *WAT*, *WAB*, *WAC*, *FS*, *WLFT*, *WLFB*, *WLFC* and *VMC* after thermal treatment of bricks, 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 properties of a certain sort of a product. The choice of the optimal process conditions (firing temperature and concentration of added heavy clays) for production of bricks depends on the application of the final product.

The objective function (*F*) is the mathematical function whose maximum would be determined, by summing the FSE results of the five models, according Eq. 4. All groups of response variables (*CS*, *WA*, *WLF, FS* and *VMC*) have the same influence on the function *F*:

$$F(T,[\mathrm{MP1}],[\mathrm{MP2}])= \overline{CSB}+\overline{CSC}+\overline{WAT}+\overline{WAB}+\overline{WAC}+\overline{WLFT}+ +\overline{WLFB}+\overline{WLFC}+\overline{FS}+\overline{VMC} \tag{4}$$

The maximum of function *F* represents the optimal processing parameters, and also the optimal *CSB*, *CSC*, *WAT*, *WAB*, *WAC*, *FS*, *WLFT*, *WLFB*, *WLFC*



a) b)

Figure 2. Objective function for roofing tiles: a) concentration of the MP1 addition influence, and b) concentration of the MP2 addition influence

or *VMC*. The graphs of the dependent variables with significant parameters were obtained using objective function to determine optimal production conditions, plotted on optimization graphic. Objective function can gain values between 0 and 1, depending on results obtained using trapezoidal function defined in Eq. 3. If the value of membership trapezoidal function is close to 1, it shows that the tested processing parameters are close to being optimal. Optimal values of *CSB*, *CSC*, *WAT*, *WAB* and *WAC* were published in our previous research [11]. The overall optimal process parameters are gained by summing the membership functions of all responses, and dividing it by 10, according to Eq. 4. The obtained optimal process parameters were: firing temperature of 870 °C, MP1 concentration 8–10 wt.%, and MP2 concentration 2 wt.%, with *F* function value of 0.75. The objective functions, regarding processing parameters, temperature and concentrations of added heavy clays were shown on the surface plots (Fig. 2).

IV. Conclusions

In this research, we used different combinations of three heavy clays from nearby locations in order to find the optimal behaviour to produce roofing tiles. Experimental design and response surface analysis revealed that the sample containing more clay and less quartz caused improvement of the final product. After fuzzy synthetic optimization, it was concluded that the optimal combination of independent variables were firing temperature of 870 °C, MP1 concentration 8–10 wt.%, and MP2 concentration 2 wt.%, with *F* function value of 0.75.

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