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Factor space differentiation of brick clays according to mineral content: Prediction of final brick product quality

Milica Arsenović [a,*], Lato Pezo [b], Slavka Stanković [c], Zagorka Radojević [a]

[a] Institute for Testing of Materials IMS, Bulevar vojvode Mišića 43, 11000 Belgrade, Serbia
[b] University of Belgrade, Institute of General and Physical Chemistry, Studentski trg 12, 11000 Belgrade, Serbia
[c] University of Belgrade, Faculty of Technology and Metallurgy, Karnegijeva 4, 11000 Belgrade, Serbia

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ABSTRACT

Chemical composition and XRD qualitative analysis were used to calculate mineral contents of 139 brick clay raw materials using LPNORM. The second order polynomial models (SOP) for all the samples, which express the relation between mineral contents and the characteristics of fired laboratory products, did not fit to experimental data satisfactorily, due to low coefficients of determination (r^2). In order to improve the models, the samples are divided into four groups in factor space (four quadrants), according to their mineral content similarity, using principal component analysis (PCA). Predictive models of compressive strength (*CS*), water absorption (*WA*), firing shrinkage (*FS*), weight loss during firing (*WLF*) and volume mass of cubes (*VMC*) are obtained for each of the groups. Second order polynomial (SOP) models are developed, and the influence of certain minerals to brick clay bricks quality within the groups is discussed. Developed models were able to predict the final quality of products in a wide range of mineral content and temperature treatment data, showing coefficient of determination (r^2) in range between 0.704–0.995. In order to estimate the adequacy of these models, the results were applied to the experimental data and compared according to additional statistical tests, so the next values are determined: coefficients of determination, reduced chi-square (χ^2), mean bias error (*MBE*), mean percent error (*MPE*) and root mean square error (*RMSE*).

1. Introduction

Mineralogical composition of brick clay can significantly vary from a deposit to deposit. The quantities of minerals greatly influence the behavior of the products in the stages of forming, drying and firing. The most important components of brick raw materials are clay minerals, which improve strength of products, but also alter plasticity and drying susceptibility, or can cause swelling. The optimal composition of raw materials is important and differs according to the planned final products form (Pansu and Gauthezrou, 2003; Marković et al., 2004; Arsenović, 2013; Arsenović et al., 2014; Bories et al., 2014; Gliozzo et al., 2014; Pezo et al., 2014). It should be inevitably borne in mind that the requirements for the environment preservation are constantly growing (Monteiro and Vieira, 2014), so that optimization of the composition of raw materials in relation to a particular type of product is effective and important in this regard (Arsenović et al., 2013b).

Several authors earlier found that mathematical modeling is useful tool in determining certain behavior laws when bricks and roof tiles are concerned (Dondi et al., 2003; Sidjanin et al., 2007; Meseguer et al., 2009; Gualtieri et al., 2010; Ducman et al., 2011), but these methods are started to be intensively used in 2013. Chemical composition and XRD qualitative analysis of 139 brick clay raw materials from Serbia are impaired to calculate the contents of present minerals using a program developed earlier, and called LPNORM (De Caritat et al., 1994; Pezo et al., 2014). The results are preliminary tested by post-hoc Tukey's HSD test that showed statistically significant differences between the samples and proved that the results are appropriate for further investigation and modeling.

The SOP models, developed in our pre-research (Arsenović, 2013), which presented the relation between mineral contents and the characteristics of fired laboratory products, showed unsatisfactory agreement to experimental results, due to low coefficients of determination (r^2). In this case, it highlighted the need to approach the problem in a different way. In this research, principal component analysis (PCA) is used to decompose the original data matrix into several products of multiplication, loading (different brick clay samples) and score (different mineral content) matrices. Thus, the raw materials are divided into 4 groups according to similarity concerning mineral content (Scull and Schaetzl, 2011). Further, for every quadrant in PCA score plot, a separate SOP model is developed, in order to obtain highly precise prediction of characteristics of fired laboratory products. SOP models were used to investigate the effect of mineral content and firing temperature on compressive strength (*CS*), water absorption (*WA*), firing shrinkage (*FS*),

* Corresponding author.
E-mail address: milica.arsenovic@institutims.rs (M. Arsenović).

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weight loss during firing (*WLF*) and volume mass of cubes (*VMC*). The effects of the input variables on the responses can be realized by analysis of variance (ANOVA) of the SOP equations. ANOVA also enables the determination of input variables interrelationships and the exploration of the combined effect for all input variables in the observed responses. The performance of SOP models was compared to experimental results, using usual statistical tests, such as: coefficients of determination (r^2), the reduced chi-square (χ^2), mean bias error (*MBE*), mean percent error (*MPE*) and root mean square error (*RMSE*). Previously developed artificial neural network model (ANN) for the same observed characteristics also showed good fitting capabilities (Arsenović, 2013), but the models developed in this research are proved to be much simpler and more elegant for practical application.

The specific objective of this study was to investigate the effect of calculated mineral content and firing temperature on compressive strength, water absorption, firing shrinkage, weight loss during firing and volume mass, using SOP models for prediction purposes. The performances of SOP models were compared to experimental results.

2. Materials and methods

Brick clay raw samples were dried at 105 °C and then analyzed for chemical content of major oxides using classical silicate analysis (Rettig et al., 1983), while all the measurements were performed in triplicate. Qualitative mineralogical analysis was carried out by XRD (powder diffractometer Philips PW-1050, λCu-Kα radiation, scanning speed 0.05°/s). The samples were tested in powder, bulk form, and also as oriented aggregates. The quantity of present minerals is calculated using LPNORM, as described before (De Caritat et al., 1994; Pezo et al., 2014).

Mineralogical composition of tested clays varies within the deposits, and also in different locations. A large part of the samples belong to the loess raw materials that are characteristic in that they generally contain carbonates, quartz and feldspar, a small quantity of clay minerals, and are usually suitable for the formation of solid bricks. In the case of more plastic raw material (having higher content of clay minerals), it is possible to manufacture hollow blocks, and roof tiles. The results of chemical analysis showed that the examined samples from Serbia generally contain more quartz than clay minerals, enough fluxes and iron, and also often much CaO. Raw samples are found to be representative brick clays, containing free quartz, usually as the dominant ingredient, illite (mica), chlorite, calcite, dolomite, feldspar, as well as small amounts of smectite, kaolinite and rutile, with a satisfactory content of clay minerals (Arsenović, 2013).

The complete procedure of preparing (milling and moist homogenizing), shaping, drying and firing is done using standard laboratory procedure, which is described previously (Arsenović, 2013). 139 samples used in the research were collected in Serbia in order to test new opened deposits near brick factories. After preliminary tests and preparation, shaping and drying, laboratory products in the form of tiles, hollow blocks and cubes, were fired in the broad range of temperatures: 800 °C–1100 °C.

The following parameters were tested on fired samples, in the same way as described before (Arsenović et al., 2013b): compressive strength (*CS*), water absorption (*WA*), firing shrinkage (*FS*), weight loss during firing (*WLF*) and volume mass of cubes – apparent density (*VMC*). These variables were determined in the case of tiles, (hollow) blocks and cubes, so the letters *T*, *B* and *C* were respectively added to the mentioned abbreviations. PCA is a very useful mathematical procedure, which decreases the number of parameters by transforming them to a new orthogonal factor space (Abdi and Williams, 2010; Arsenović, 2013; Pezo et al., 2014). This way, the database is simplified, and, as such, the behavior of the systems involved is easier to understand. Another advantage of this method is that the charts (called score plots) facilitate the conclusions. This approach enabled a differentiation of the samples, according to their mineral contents, into four different groups, to be used in further analysis separately. The biplot diagram represents the projection of the vectors and points on the factor plane 1–2, while the correlations represent the intensities of the vectors, interpretation of the results is much more facilitated in combination with correlation analysis (Bastianoni et al., 2008; Arsenović, 2013).

The second order polynomial model (SOP) was selected to estimate the main effect of the process variables (minerals content and firing temperature) on *CS*, *WA*, *FS*, *WLF* and *VMC*, in every of the four groups. The accepted experimental design was taken from Arsenović et al. (2013b). The independent variables were: the content of minerals (*Q* — quartz, *I* — illite, *Clino* — clinochlore, *Chamo* — chamosite, *Sm* — smectite, *Kaol* — kaolinite, *C* — calcite, *M* — magnesite, *Alb* — albite, *An* — anorthite, *Orth* — orthoclase, *Rut* — rutile), and firing temperature (in the range of 800–1100 °C), while the dependent variables observed were the responses: *CSB*, *CSC*, *WAT*, *WAB*, *WAC*, *FS*, *WLFT*, *WLFB*, *WLFC* and *VMC*. The model was obtained for each dependent variable in every four groups, where factors were rejected when their significance level was less than $p > 0.05$, 95% confidence limit. The significant terms in the model were found using ANOVA for each dependent variable. PCA, ANOVA and regression analysis of the SOP models were performed using StatSoft Statistica for Windows (version 10).

3. Results and discussion

3.1. Correlations and principal component analysis (PCA) of mineralogical contents

Tested brick clay materials consisted of free quartz, mostly as a dominant ingredient, then illite (mica), chlorite, calcite, dolomite, feldspars, and also amounts of smectite, kaolinite and rutile. The empirical formulas of certain minerals that entered the program were taken from the base of natural minerals, and explained in previous research (Pezo et al., 2014). On the bases of the calculated contents of minerals, correlation analysis is done and presented in Table 1. Generally low values of observed correlation coefficients indicated the complicity of interrelations. In this study, most of the correlation coefficients showed statistically significant due to the large number of performed experimental measurements. Given that the correlation assumes linear relationships of variables (Fuks and Kiv, 2013), the obtained coefficients are of generally low values, indicating a non-linearity of the mutual dependence of the observed variables.

The analysis of dissimilarities in the mineral content of brick clay samples was also investigated by means of PCA (Fig. 1). The number of factors retained in the model for proper classification of mineral content data, in original matrix into loading (different samples) and score (mineral content) matrices were determined by application of Kaiser and Rice's rule (Moropoulou and Polikreti, 2009; Ravisankar et al., 2014). This criterion retains only principal components with Eigenvalues > 1. PCA showed that there are 6 statistically significant factors (Table 2), of which only the first two were selected for further interpretation. Factor 1 is mostly affected by calcite, quartz and chlinochlor content, while Factor 2 was affected by orthoclase, rutile, smectite and illite (Fig. 1).

The most intensive negative correlation was found between I and Orth, as expected, since they consist of the same elements (aluminum, silicon, potassium, and oxygen). The correlation between clinochlore–calcite content (positive) and chamosite–calcite content (negative) was also very intensive, as can be seen from Fig. 1. Clinochlore is positively correlated to orthoclase content, while it is not in correlation to magnesite and albite content. It is known that lime in the natural state can be enriched with magnesia and clay, and therefore the positive correlations between calcite–clinochlore and calcite–magnesite content could be explained. Correlations between calcite and chamosite, smectite and kaolinite minerals are found negative, which is in agreement with the results of the PCA (Fig. 1). Calcite and magnesite occur together in the raw material, which is confirmed by the results of a macro oxides