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Optimization of drying through analytical modeling: clays as bonding agents in refractory materials

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Abstract

The objective of this study was to investigate the clay drying as a unit operation in the refractory materials processing. Two clays that varied in chemical and mineralogical compositions were experimentally tested in a laboratory dryer. The results obtained on the green samples prior to drying indicated that clays have adequate plasticity and refractoriness for application in shaped refractories. The operating parameters of the dryer were regulated: temperature ranged from 40 to 60 °C, humidity increased in the interval 30–70%; and the airstream rate was 1.3 m/s. The correlation analysis between operating parameters and calculated and/or measured drying outputs was conducted for better comprehension of the clay's role as a refractory binder. Subsequently, a mathematical optimization of the drying regime was conveyed. The effect of the variables (operation parameters) on the drying parameters (critical moisture, equilibrium moisture, dryness degree, etc.) was compared and evaluated. The response surface method, standard score analysis, cluster method, and principal component analysis were used as a means of the drying regime optimization. Assessment of the drying regime impact on the dried samples quality highlighted optimal result for both clay types: SS = 0.95, temperature 50 °C, and humidity 40%. Multiple comparison analyses pointed out that optimized combination of the drying operation parameters decreases the quantity of conducted tests. Furthermore, optimal combination of drying parameters reduces negative effects of clay binder inherent properties on the resulting product which in return improves energetic and economic sustainability of refractories production.

Keywords: A. Drying; B. Composites; C. Thermal properties; D. Clays; E. Refractories; ANOVA

1. Introduction

The clays represent complex mineral mixtures that vary in composition depending upon its geological location [1]. A clay deposit comprises an assortment of main clay minerals (kaolinite, illite, montmorillonite) and accompanying minerals (quartz, feldspar, chlorite, smectite, mica) [2,3]. The fine-grained clay minerals exhibit plastic behavior at appropriate water contents and solidification during thermal exposure (drying and/or firing) [1,4]. The clays can be fabricated into myriad of industrially finished products. Namely, the clay is the principal component of traditional ceramics (bricks, sewer pipes, structural tiles, refractories, insulators, sanitary-ware, lightweight aggregates) [5–7]. The minor binding components used in the advanced ceramics design are mandatory clay based [8,9], while in the production of glass and engineered ceramics the clay represents solely a chemical additive [6,10,11].

The application of refractory ceramics is indispensable in metallurgical industry and facilities with prerequisite fire/thermal protection (e.g. furnace linings, kilns, heat-processing equipment, hot-face materials insulation, laboratory crucible sand setters). Therefore, the refractories are positioned among the highest clay consumers [12]. The shaped refractory materials are mixtures of plastic clays with non-plastic raw materials (e.g. chamotte) [5,13–16]. The performances and technical restraints of the refractories are basically controlled by chemical and mineralogical compositions of the used raw materials.

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Nomenclature

Ts	Drying temperature (°C)
φ	Relative humidity of air (%)
v_a	Rate of the airstream in the dryer (m/s)
w_s	Moisture of clay-water suspension (%)
m_{cs}	Mass of samples/clay slabs (g)
w_i	Initial moisture of material (%)
w_c	Critical moisture content of material (%)
t_c	Time of reaching the critical moisture content (min)
w_r	Equilibrium moisture content of material (%)
t_r	Time of reaching the equilibrium moisture content (min)
C_S	Coefficient of sensitivity
E_B	Bond energy (kJ/mol)
D_D	Degree of dryness

Correspondingly, the physico-chemical nature of the applied clay has to adequately respond to required properties of refractory products [5,17,18]. The basic mineral constituents of refractories are kaolinite ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$), chromite ($FeO \cdot Cr_2O_3$) and magnesite ($MgCO_3$), and the overall composition of the applied raw materials generally comprises 25–45% Al_2O_3 and up to 85% SiO_2 [19,20]. The refractory clays with refractoriness above 1580 °C are used in their crude unfired form as binders [5,15,21]. Chamotte (fired refractory clay) transforms the mixture into the stable high temperature material, and limits the shrinkage and cracking during drying and firing [18,21,22]. Thusly obtained refractories are characterized by relatively common fabrication, good resistance to high temperature, creep, chemical corrosion, low thermal expansion coefficient, and ability to withstand severe service conditions [5,21,23].

The selection of the type and quality of clay suitable for refractory application requires detailed investigation of the clay's chemical and mineralogical composition, properties and thermal behavior [2,21]. The performances of a clay binder are predetermined during the drying stage. Drying is an important step in the refractories fabrication, because the moisture removal combined with simultaneous heat and mass transfer between the material and the surrounding atmosphere can act destructively (e.g. spalling, cracking, explosive failure) [24]. The clay's drying procedure proceeds through stages characterized by: constant rate (initial weight loss and notable shrinkage) and decreasing rate (successive weight loss and minor shrinkage) [25–28]. The shrinkage might arouse the drying cracks in the samples. Drying crack is related to the term "drying sensitivity", and along with moisture content these parameters are regarded as the drying quality indicators [25,26]. The drying related problems can be studied through different techniques, such as numeric modeling, macroscopic weight measurements, and large scale tests [26–34]. Mathematical tools for assessing the impact of raw material properties and processing parameters on the quality of the final product are intensively used in this field [35–37]. Most of the criteria for optimal design of experiments are associated with the mathematical model of the process [38]. The Response Surface Methodology (RSM) is a useful method for determining the influence of process variables on a group of dependent parameters that are significant for the process [39]. RSM is an effective tool for optimizing a variety of processes, especially in the mixture experiments design [40]. The RSM equations describe the effects of the test variables on the observed responses, determine test variables interrelationships and represent the combine effect of all test variables in the observed responses, enabling the efficient process investigation [37–40].

This paper presents the results of an experiment which explored the effect of temperature (Ts) and relative air humidity (φ), as variable parameters applied during drying of clays (Gv and Gk). Principal component analysis (PCA), used as a pattern recognition technique, was applied within assay descriptors to characterize and differentiate various analyzed samples. Simple regression models, using RSM were proposed for calculation of w_i, w_c, t_c, w_r, t_r, C_S, E_B, and D_D by using two operation parameters: Ts and φ. Analysis of variance (ANOVA) was applied to show relations between applied assays. In order to enable more comprehensive comparison between examined samples, particularly the contribution of operation parameters, standard score (SS), assigning equal weight to all assays applied was introduced. The aim of this work was to characterize and monitor properties changes during drying of two different natural clays from Serbia, via a combination of several mathematical techniques, in order to determine the suitability of these clays for refractory applications.

2. Materials and methods

The first step of the investigation implied experimental drying test that was conducted on the two clay types, here labeled as Gv and Gk, in a recirculating dryer. The dryer was provided with the possibility of working/operation parameters regulation. Subsequently, a mathematical optimization of the drying regime of both clays was conveyed and compared. The experimental clays originated from a local source, i.e. two clay deposits in Serbia (*Vrbica* and *Valjevska kamenica*). The chemical composition of the clays was analyzed by a PinAAcle 900 atomic absorption spectrometer (Perkin-Elmer, USA). The results are provided in Table 1.

The basic difference in the chemical composition of the investigated clays is in SiO_2 and Al_2O_3 content, as well as in percentage of alkali metal oxides (Na_2O and K_2O). Namely, the analysis showed a higher Al_2O_3 percentage in Gv sample, while Gk had a higher content of alkalies and a higher SiO_2 percentage.