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Research paper

Combining mixture design of experiments with phase diagrams in the evaluation of structural ceramics containing foundry by-products

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ABSTRACT

This work compares the effects of the incorporation of foundry by-products (foundry sand dust and Waelz slag) on the technological properties of three different ceramic products fabricated with different clays and fired at different temperatures: wall bricks (850 °C), roof tiles (950 °C) and face bricks (1050 °C). To this aim, the mixture design of experiments (M-DoE) methodology was used to define the minimum number of mixtures necessary to model and predict the technological properties (water absorption, flexural strength, open porosity, linear firing shrinkage, weight loss and apparent density) in terms of raw materials contents: clay, foundry sand dust and Waelz slag. The results obtained were interpreted in terms of the chemical and mineralogical compositions of the final products, which showed the importance of combining the M-DoE methodology with the phase equilibrium predictions to better understand the development of the fired products final properties.

1. Introduction

Basic industrial materials processes consume finite resources and generate large quantities of by-products. These industrial by-products have potential for further uses but are nevertheless discarded in landfills. At present, industries are moving from these open processes towards closed production models that convert waste into valuable materials to be used as new resources. From a technological perspective, the possibility of reusing or recycling a given by-product depends on the desired quality of the final products into which the by-product is introduced. In particular, the ceramic industry allows the suitable incorporation of several types of industrial waste as raw materials into ceramic matrices (Jonker and Potgieter, 2005; Raupp-Pereira et al., 2006; Raut et al., 2011; Rozenstrauha et al., 2006; Vieira and Monteiro, 2009; Zimmer, 2010). Several studies have shown that waste–containing alternative ceramic products maintain the mechanical properties without relevant process modifications and the final product quality can even be improved (Alonso-Santurde et al., 2010; Segadaes, 2006).

Mathematical and statistical techniques, such as mixture design of experiments (M-DoE), are modelling tools that enable the prediction of products behaviour with a minimum number of trials and are capable of evidencing various types of joint effects amongst variables. The basic assumption of this technique is that, when the process conditions are kept constant, the properties of the products (response variables) are simply determined by the proportion of the components in the mixture (input variables). Various studies have demonstrated the viability of using the M-DoE methodology in ceramic products, such as triaxial porcelain (Correia et al., 2006, 2008, 2009), bricks or tiles (Menezes et al., 2009). The mixture design approach has also been applied to maximise (Menezes et al., 2007) or optimise (Correia et al., 2004, 2005; Menezes et al., 2008) the by-product content in ceramic mixtures to obtain products with particular desired properties. However, to the best of our knowledge, M-DoE has only been used to model the properties of structural clay ceramics that incorporate mining or mineral by-products that are similar in composition to the natural raw materials commonly used in the fabrication of bricks and tiles, and the effect of other types of industrial by-products, with more distant compositions, has not been investigated.

Previous studies looking for reuse alternatives for two by-products from the foundry industry, Waelz slag (WS) and foundry sand dust (FSD), have demonstrated, not only on the laboratory scale but also through industrial trials, the viability of using Waelz slag (Quijorna et al., 2011), foundry sand (Alonso-Santurde et al., 2011) and mixtures of both by-products (Quijorna et al., 2012) to produce suitable structural ceramics. In this work, the M-DoE methodology was used to obtain critical information about the synergetic or antagonistic effects on various properties of the combination of the two by-products.

The purpose of a design of experiments (DoE) is to obtain appropriate models to correlate the response variables with the input variables. In the particular case of mixture design of experiments (M-DoE), the input variables are the proportions of the components of the mixture,

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which must sum to unity. In the case of ceramic mixtures, the usual raw materials can be divided into three different types of components, with different roles during ceramic processing: plastic (clays), fluxing (feldspar) and filler (quartz). This explains the frequent use of the epithet “triaxial” to describe industrial ceramic products and suggests graphical representations based on triangles, usually equilateral. In such diagrams, composition axes run from each vertex (pure component, weight fraction = 1) to the opposite side (mixtures without the component from the opposite vertex, weight fraction = 0). Thus, raw materials, natural or otherwise, with similar nature, hence having similar roles and capable of replacing each other, should be represented in the same vertex of such triangles (Correia et al., 2009; Menezes et al., 2007, 2008, 2009). This also suggests the use of a ternary simplex-lattice design of experiments to calculate mathematical equations that might model the behaviour of ceramic products using a minimal number of mixtures, strategically located and widely distributed on the equivalent simplex triangle.

Nevertheless, the actual behaviour of ceramic mixtures is ultimately determined by chemical composition and the approach to equilibrium. In terms of chemical composition, the great majority of natural raw materials for the ceramic industry (whether plastic, fluxing or filler) contain, as major components, silica (SiO_2) and alumina (Al_2O_3). All contain minor amounts of compounds whose main effect will be on the colour of the fired product (Fe_2O_3, MnO, TiO_2, Cr_2O_3), but these oxides should not play such an important role during ceramic processing, in air, at low temperatures and short firing cycles. And in all of them the other minor components (K_2O, Na_2O, CaO and MgO) will act as fluxes and, depending on the firing temperature and time, might have a strong effect during sintering. For long firing cycles, particularly if the temperature is high enough (~1200 °C) and the firing atmosphere is slightly reducing, the iron oxide will also have a strong fluxing effect and its presence should not be disregarded (Segadaes, 2006). Thus, in the ceramic industry, the firing behaviour and property development can be anticipated and interpreted based on phase equilibrium relationships expressed in the silica–alumina–fluxing oxide phase diagram, which is another graphical representation in the form of a triangle. It can be argued that in today's normal industrial operating conditions, with very short firing cycles, thermodynamic equilibrium is usually not reached, but the equilibrium phase diagram can still be used to foresee the reactions tendency to completion.

Although the M-DoE methodology is blind as far as ceramic science and phase equilibrium are concerned, its capability of discerning between synergetic and antagonistic effects of variables might be very useful when it comes to exploring reuse alternatives for unusual by-products in the ceramics industry.

The aim of this study is to compare the effects of the incorporation of foundry by-products (foundry sand dust and Waelz slag) on the technological properties of three different ceramic products fabricated with different clays and fired at different temperatures: wall bricks (clay C_1, 850 °C), roof tiles (clay C_2, 950 °C) and face bricks (clay C_3, 1050 °C). The M-DoE methodology was used with the proportions of clay, foundry sand dust and Waelz slag as input variables, the response variables being: water absorption, flexural strength, open porosity, linear firing shrinkage, weight loss and density. The relationships between mineralogy, chemical composition and technological properties of the fired products were investigated and the M-DoE results are also discussed in terms of the phase diagram.

2. Materials and methods

The methodology used in this work (Fig. 1) is divided into six main steps: (i) characterisation of raw materials, (ii) mixture design of experiments (M-DoE), (iii) ceramic processing, (iv) assessment of the technical properties of the fired bodies, (v) modelling the technical properties of the fired products and (vi) establishment of the relationships between the technical properties and the chemical and mineral composition of the fired bodies.

Three illitic clays (C_1, C_2, and C_3), currently used as traditional raw materials in three Spanish industrial ceramic processes (respectively: wall bricks, P_1; roof tiles, P_2; and face bricks, P_3), and two foundry by-products, Waelz slag (WS) and foundry sand dust (FSD), were used in this study. Clay samples were collected from the referred industrial sites. The WS, is a waste obtained from the Waelz slag process which mixes by-products of Electric Arc Furnace Dust (from steelmaking process) with coal and heated the mixture in a rotary kiln to the point where zinc vapour is formed. The zinc fume (ZnO) is carried with the furnace gases and collected in bag filters. The Waelz slag commercialised as Ferrosita®, was supplied by Befesa Zinc Aser, Abengoa Group, from its Electric Arc Furnace Dust recycling plant located in Erandio (Basque Country, Spain). The FSD was supplied by Nissan Motor Ibérica S.A., from the ventilation bag-house systems of its automotive foundry facility located in Corrales de Buelna (Cantabria, Spain).

Elemental analysis and major oxide composition were determined by inductively coupled plasma atomic emission spectroscopy (ICP-

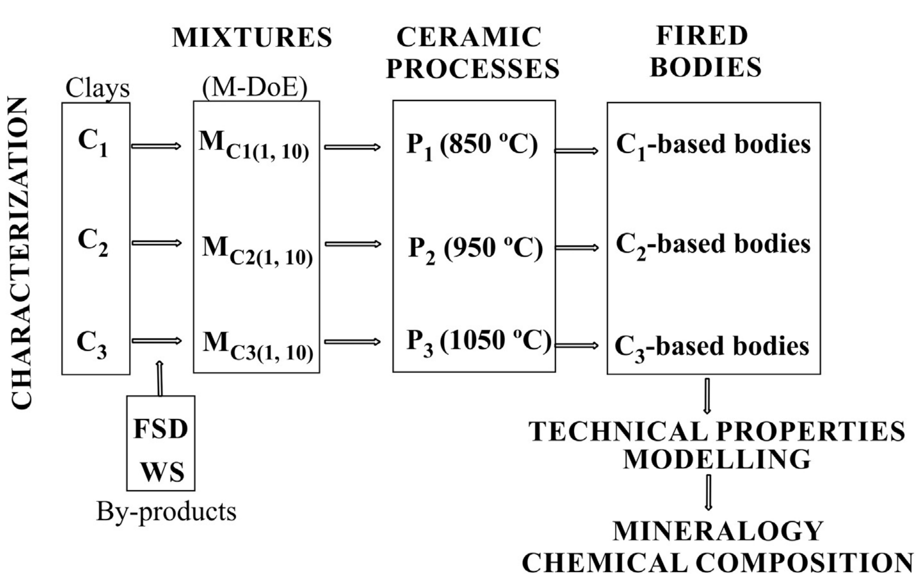

Fig. 1. Methodology based on statistical experiments design approach.

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