Mixture design and response surface analysis of pozzolanic products

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Abstract. In products consisting of several components, physical and engineering properties are a function of the components proportions. A pozzolanic product can be understood as a ternary mixture where two components are ceramic solid particulates (e.g. fly ash and lime) and the third component is liquid (water). In this work experimental data of pozzolanic compositions and respective measured properties were adapted to the methodology of response surfaces using a mixture design of a constrained triangular surface. The results obtained were satisfactory for determination of equations that represent the desired properties as a function of the components variations of the ternary mixture. The use of this mathematical procedure can be an important tool to predict and understand the behavior of such type of ceramic mixtures.

Introduction

Solid wastes, liquid effluents and gaseous emissions are undesired by-products of many industries all over the world, bringing a negative environmental impact. Among the industrial residues, billion of tons of solid particulates have been thrown in the environment, contaminating gradually soil, water and air.

In Southern Brazil, substantial amounts of fly ashes from combustion of powdered coal in heat-generation plants have been thrown away as an industrial reject. This pollutant is characterized as a ceramic material in form of a solid particulate constituted basically of silicon, aluminum and iron.

Many researches works have been showing the possibility of using fly ash in several technological areas, e.g. in cement concrete [1], ceramics [2], glass [3] and in soil stabilization [4] as bases of roads and airports.

The combination of coal fly ashes with lime and water lead to chemical reactions which originate a cement that is processed in room temperature. Generally the compositional studies undertaken with such pozzolanic mixtures employ traditional experimental designs. In these cases, one component or the relationship of two components is held constant while a third component amount is varied. Such techniques usually demand some previous experience with materials and processes and a great number of required runs to yield the desired product characteristics. The mechanical strength of the final product among other properties depends basically on the relative amounts of the three components ash, lime and water.
In many engineering fields designed mixture experiments have been used to predict quantitatively the properties of materials, in a more reliable, fast and easy way. The use of response surfaces – which can be mathematically expressed and statistically tested – allows obtaining a series of fundamental information of the system in study from a minimum amount of samples or runs. These techniques are based on the fact that the properties of a mixture depend exclusively on the proportions among their components, and not of their total amount. [5,6]

The methods of mixture design and response surfaces have been employed in various problems in science and engineering, particularly in industrial applications [7-11]. In this paper, a series of pozzolanic mixtures is regarded as a ceramic system that can be described by response surfaces, using a designed mixture approach in a constricted triangular surface. The property to be mathematically modeled is the mechanical strength from simple compression tests.

**Experimental procedure**

A fly ash obtained from the combustion of powdered coal from a heat-generation plant (Jorge Lacerda, Capivari de Baixo, SC, Brazil) and a hydrated lime (commercial type) were used, Table 1. Potable water was added to ash-lime mixtures and fine grained sand was used as aggregate.

<table>
<thead>
<tr>
<th>Chemical composition of raw materials.</th>
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<tbody>
<tr>
<td>Fly ash</td>
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<tr>
<td>SiO₂</td>
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<tr>
<td>Al₂O₃</td>
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<tr>
<td>Fe₂O₃</td>
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<tr>
<td>CaO</td>
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<td>MgO</td>
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<td>Na₂O</td>
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<tr>
<td>K₂O</td>
</tr>
<tr>
<td>SO₃</td>
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<td>Loss on ignition</td>
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</tbody>
</table>

Sample bodies were compacted with 132 kgf/m² according to DNER-DPT M 48-64 norm, in cylindrical molds of 10 cm diameter and 20 cm height. Sample bodies were submitted to simple compression tests after 28 days of cure.

**Results and discussion**

Mixtures of fly ash, lime and water – the so-called pozzolanic pastes – have as main function to attach solid aggregates and to fill the porosity among them, in such a way that, after hardening, the system has the desired strength to support the required mechanical stresses.

When the pozzolanic paste is added to small aggregates a pozzolanic mortar is obtained. Similarly, when the pozzolanic paste is added to large aggregates a pozzolanic conglomerate mortar is obtained. It is the paste that influences decisively the aggregate cementation. In this way, the
relative amounts of ash, lime and water are determinant for the properties of the pozzolanic product to be analyzed.

The reactions that happen in the ternary system ash-lime-water are complex and not yet completely understood. This complexity is enhanced due to the heterogeneity of physical and chemical compositions of raw materials, primarily of fly ash.

The most important reaction is the one that results in a cement gel, which bind the inert particles of the aggregate. A typical reaction occurs between calcium hydroxide and the silicon and aluminum minerals, producing calcium silicates and aluminates. In the case of fly ash, Fig. 1a, the reactions products, Fig. 1b, resemble CaO·SiO₂·H₂O from the hydration of Portland cement.

![SEM micrographies of (a) fly ash and (b) pozzolanic paste obtained from fly ash.](image)

**Fig. 1** SEM micrographies of (a) fly ash and (b) pozzolanic paste obtained from fly ash.

In this work, laboratorial results of an empiric project were used to build up a triangular surface representing a ternary mixture. The whole extension this area is not used, due the restrictions in the limits of the proportions of the components, so that pseudocomponents are introduced as a function of upper and lower limits of each variable. [5,6] For the model calculations the software Statistica was used (StatSoft, 1999 edition).

Small aggregates were considered to be an inert material, not participating in the pozzolanic reactions and not having cement properties. The proportions of ash, lime and water – the chemically active components – were then normalized to yield 100%. Therefore the analyzed area was from 7.9% to 25.8% for lime, 44.6% to 67.6% for fly ash, and 18.3% to 40.6% for water. The restrictive conditions of these intervals, to be used in the software, were obtained by matrix calculation giving the following equations:

\[ -0.11278L + 0.05205A = 0 \]  \hspace{1cm} (1)
\[ 0.16042L - 0.02468A = 0 \]  \hspace{1cm} (2)
\[ -0.00159L + 0.05590A - 0.07073W = 0 \]  \hspace{1cm} (3)
\[ -0.00058L - 0.03782A + 0.11619W = 0 \]  \hspace{1cm} (4)
where $L$ is lime, $A$ is ash, and $W$ is water.

To obtain the response surfaces for triaxial mixture, a quadratic regression was applied. Next, a minimum number of mixtures was determined, so that the correlation factor $R^2$ was greater than 0.9. The results of simple compression tests are presented in Fig. 2, as function of proportions of the ternary mixture in pseudocomponents. The mathematical model from 15 mixtures presented the desired adjustment factor. The same results are present also in a three-dimensional form, Fig. 3.

![Contour plot of compressive strength [kg/m²] as a function of an ash-lime-water mixture.](image)

**Fig. 2** Contour plot of compressive strength [kg/m²] as a function of an ash-lime-water mixture.
Fig. 3 Response surface of compressive strength [kg/m²] as a function of an ash-lime-water mixture.

The following equation below corresponds the simple compressive strength $CS$ expressed as a function of the proportions of the pozzolanic mixture, calculated by the quadratic model for the original components:

$$CS = 90.173L - 118.454A - 506.969W - 197.564LA + 474.778LW + 1165.461AW$$  \hspace{1em} (4)

Conclusions

After the experiments and calculations it was possible:
- to define a triangular diagram that correspond to a range of potential pozzolanic compositions;
- to obtain a mathematical model that allows, among many other purposes, to describe the resulting product according to specifications;
- to decrease the number of sample bodies and time of experimentation in laboratory;
- to predict variation the mixture behaviour as a function of the component variation range.

References
