Influence of Surface Area on the Flowability Behaviour of Self-Flow Refractory Castables

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Abstract. Alumina, with high melting point (2050°C), high hardness and mechanical strength, and excellent abrasion resistance, is one of the most common raw materials used in self-flow refractory castables (SFRC) for monolithic linings and is commercially available in various fine to coarse size classes. However, the performance of the refractory lining depends not only on the properties of its ingredients but also on its easy installation (good flowability). The aim of this work was to evaluate the relationship between the flowability index (FI) of fresh castable and the specific surface area (SSA) of its particles, which is mostly determined by the finer particles content. The results obtained showed that, by controlling the proportion between matrix and aggregate, it is possible to control the SSA of the refractory castable and find a mathematical relationship between the specific surface area and the minimum flowability index required to obtain a self-flow refractory castable. It is, thus, possible to optimize the refractory castable size composition and obtain an estimate for FI as a function of SSA. Using a minimum 45 wt.% matrix content in the castable mixture, a SSA value above 2.215 m\(^2\)/g is obtained, which leads to FI ≥ 80%, the recommended value for self-flow.

Introduction

Ceramic materials as a whole and, particularly, engineering ceramics, have an unusual combination of properties which makes them very attractive for the use in aggressive thermo-mechanical environments. Those properties are mainly a consequence of crystal structure and strong ionic and covalent bonds, which lead to special optical and magnetic behaviour, low electrical and thermal conductivity, low density, high mechanical strength even at high temperatures, and good wear and corrosion resistance [1]. The combination of thermal insulation capacity with high mechanical strength and corrosion resistance at high temperature explains the use of ceramic refractory linings in a variety of high temperature industries, from iron and steel metallurgy to glass melting, traditional ceramics and the chemical industry [2,3].

Among refractory linings, monolithic linings do not require energy for shaping and pre-firing, which is a major advantage. Refractory castables for monolithic linings, like a concrete, can be regarded as a composite material containing a mixture of aggregate particles bound by a matrix of fine particles. They are supplied as dry mixed materials to which water, or other specified liquid, is added for in situ mixing and application [4,5]. Refractory castables have, therefore, been gaining growing importance in applications that require simultaneous easy in situ application and good mechanical performance.

During the past two decades, a series of industrial developments were introduced in castable compositions, both in aggregate and binder systems, and application technology, all essentially aimed at simplifying the application procedures while guaranteeing adequate mechanical performance and reduced environmental impact. One such development is the self-flow refractory castables (SFRC) with low, ultra low or no cement content, whose performance ultimately depends on the combination of raw materials used in the castable composition. The proportion between matrix and aggregate is optimized for maximum flowability index (FI) of the fresh castable, for easy application, and minimum water content, to promote the mechanical strength after firing.
Commercial tabular alumina is commonly used as raw material in SFRC without cement. Being a comparatively low cost material, easily available, alumina has rather good high temperature properties. As a comparison with other materials, tabular alumina (calcined, α-alumina) presents higher hardness and mechanical strength, excellent abrasion behaviour and a melting point of 2050°C, which explain its use in refractory applications. Also used in the manufacture of high performance refractory castables as matrix ingredient, is reactive alumina, of fine particle size, which is capable of forming hydraulic bonds at room temperature upon the addition of small amounts (3 to 6%) of water [6,7]. The performance of all-alumina SFRC without cement relies on the rigid control of the particle size distribution of tabular and reactive aluminas, to guarantee sintering and densification, and the appropriate mixing and homogenization process, to produce a fluid fresh paste for easy casting [8].

Those SFRC were produced from discrete narrow particle size classes, using an aggregate with a specific particle size distribution modulus and 30, 40 or 50 wt% of a constant composition matrix of fine particles, whose particle size distribution was optimized (using statistical techniques, mixtures design and triangular response surfaces) for minimum water content and maximum flowability index. To this aim, a ternary system of three independent size fractions (ingredients) was defined, and a regular array of uniformly spaced composition points (simplex lattice) was set on the composition triangle [8]. The value of the property of interest was experimentally determined for each of the simplex compositions and, based on those results, a mathematical equation (model or response surface) was calculated. After statistical validation, the model can be used to estimate the value that the property will assume for any composition [8-10]. The various response surfaces can then be combined, so that an optimum composition range (i.e. combination of particle sizes) can be determined, to produce an ideal matrix with the desired performance.

This work was aimed at extending previous knowledge to commercial alumina size classes and finding out the effect on the properties of the changes in the proportions between matrix and aggregate, whose size compositions were kept constant.

Materials and Methods

Commercial tabular alumina T60 (Alcoa), available in four different size classes (<0.2mm, 0.2-0.6mm, 0.5-1mm and 1-3mm), was used as raw material. Based on previous work [8], a constant aggregate size composition was selected (21.7% [0.2-0.6mm], 21.7% [0.5-1mm] and 56.7% [1-3mm]). Fig. 1 illustrates the morphology of the three coarser powders used as aggregate in this work. This aggregate composition was added in various proportions to an optimized matrix, also of constant size composition, containing three fine size classes [10], namely: 20% of <230 mesh class; 20% of <500 mesh class and 60% of CT3000SG reactive alumina (Alcoa).

![Fig. 1. General aspect of the three different size classes of commercial tabular alumina powders that compose the aggregate: a) [0.2-0.6 mm]; b) [0.5-1.0 mm]; and c) [1.0-3.0 mm].](image)

The <230 mesh matrix size class is the powder that passes through a series of vibrating standard sieves (100, 140, 200 and 230 mesh), in a dry screening separation of the <0.2mm size class of commercial tabular alumina. The <500 mesh matrix size class is the powder that passes through a series of vibrating standard sieves (200, 230, 325 and 500 mesh), in a wet screening separation of the <230 mesh size class (Fig. 2). After screening, the resulting suspension is left to settle, decanted and dried.
The mixture of the appropriate amounts of powders of matrix and aggregate with water was done in a mortar mixer (Fig. 3a), as described in the Portuguese patent #103432 (2008). Constant contents of 28mg water/m$^2$ SSA and 0.36mg citric acid/m$^2$ SSA (dispersant) were used for all the mixtures [3,8,11]. 87% of the calculated water requirement [8] was added to the powder mixture (Fig. 3b) at a rate of <8g/min in an intermittent way (Fig. 3c). This procedure is fundamental to minimize segregation and improve thorough wetting of powder surfaces, thus reducing the volume of needed water. The final 13% water is just enough to take the mixture through the self-flow “turning point”. Further details of the mixing procedure can be found elsewhere [9,10].
Results and Discussion

Table 1 presents the specific surface area (SSA), flowability index (FI) and flexural strength (MoR) values obtained for the various mixtures investigated (matrix and aggregate proportions are shown for each composition).

Table 1. Composition, specific surface area (SSA), flowability index (FI) and flexural strength (MoR) of the studied castables.

<table>
<thead>
<tr>
<th>Mixtures</th>
<th>Matrix [wt%]</th>
<th>Aggregate [wt%]</th>
<th>SSA [m²/g]</th>
<th>FI [%]</th>
<th>FI Relative Error [%]</th>
<th>MoR [MPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>M1</td>
<td>38.5</td>
<td>61.5</td>
<td>1.983</td>
<td>9.049</td>
<td>0.990</td>
<td>41.955</td>
</tr>
<tr>
<td>M2</td>
<td>40.0</td>
<td>60.0</td>
<td>2.060</td>
<td>26.190</td>
<td>0.203</td>
<td>38.345</td>
</tr>
<tr>
<td>M3</td>
<td>42.0</td>
<td>58.0</td>
<td>2.163</td>
<td>65.646</td>
<td>0.001</td>
<td>44.442</td>
</tr>
<tr>
<td>M4</td>
<td>42.5</td>
<td>57.5</td>
<td>2.188</td>
<td>75.170</td>
<td>0.032</td>
<td>45.647</td>
</tr>
<tr>
<td>M5</td>
<td>45.0</td>
<td>55.0</td>
<td>2.317</td>
<td>94.218</td>
<td>0.091</td>
<td>45.035</td>
</tr>
<tr>
<td>M6</td>
<td>46.0</td>
<td>54.0</td>
<td>2.368</td>
<td>121.769</td>
<td>0.074</td>
<td>45.083</td>
</tr>
<tr>
<td>M7</td>
<td>47.5</td>
<td>52.5</td>
<td>2.445</td>
<td>128.571</td>
<td>0.026</td>
<td>43.669</td>
</tr>
<tr>
<td>M8</td>
<td>50.0</td>
<td>50.0</td>
<td>2.573</td>
<td>134.969</td>
<td>0.021</td>
<td>49.609</td>
</tr>
</tbody>
</table>

Table 1 shows that the SSA values lie between 1.983 to 2.573 m²/g, which agrees with previous work that showed that, for self-flow behaviour at a minimum water content, this value should be close to 2.0 m²/g. Fig. 7a shows that there is a clear relationship between the FI and the SSA. The effect of SSA on FI can be closely described by Eq. 1 (quadratic polynomial, coefficient of multiple determination, R²=0.9839). The deviation (relative error) of each measured FI value from that calculated using Eq. 1 is also presented in Table 1.
Fig. 7. Effect of specific surface area (SSA) of the powder mixture on the castable properties: a) fresh castable flowability index (FI); and b) sintered castable flexural strength (MoR).

\[ FI \% = -276.74 \times (SSA)^2 + 1486.6 \times (SSA) - 1855.1 \]  

FI increases with SSA (increase in fine particles content) but Fig. 7a also shows that for SSA values above 2.4 m\(^2\)/g, a large increase in SSA is required to produce only a slight improvement in FI. In practice, an increase in fine particles content entails an increase in castable cost but, to guarantee self-flow behaviour, FI should be above 80% [12]. Fig. 7a shows that, in the present case, it is necessary that the refractory castable has SSA > 2.215 m\(^2\)/g.

The effect of SSA on the fired castables flexural strength (MoR) is not so clear (Fig. 7b). Nevertheless, the measured MoR values are above 38 MPa in all cases and tend to increase with the matrix content.

Conclusions

In particulate systems like refractory castables, there are two factors that promote high flowability: the lubricating effect of high water contents and the optimised particle size distribution (low interference contact between particles). However, when the water content is constant the distance between coarse particles controls flowability mechanism. This research showed that it is possible to optimize the refractory castable size composition by adjusting the fine particles volume (matrix content) and obtain an estimate for the flowability index (FI) as a function of the specific surface area (SSA), while keeping the added water content to a minimum. To obtain FI ≥ 80%, required for self-flow, a SSA value above 2.215 m\(^2\)/g is needed. That corresponds to a 45 wt.% minimum matrix content in the mixture (Table 1). This result is very important in the optimization process of the size composition of the alumina self-flow refractory castable.

The importance of SSA has already been demonstrated in other works [7-10,13], and the correct amount of added water and dispersant should be calculated based on it. In this research the relation between the amount of matrix and the SSA value is significantly clear. When the fraction of fine particles increases, the flowability also increases, because the higher matrix fraction contributes to increase the distance between coarse particles, thus reducing their interference.

Acknowledgements

The research team is grateful to FCT – Foundation for the Science and Technology of Portugal (Project: PTDC/CTM/66302/2006), for providing resources for this research.
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Advanced Materials Forum V
doi:10.4028/www.scientific.net/MSF.636-637

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doi:10.4028/www.scientific.net/MSF.514-516.604


