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Mechanical and thermal shock behavior of refractory materials for glass feeders

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ABSTRACT

Refractory materials of the Al_2O_3–SiO_2–ZrO_2 system are widely used in glass industry in forehearth, distributors, feeders, and as expendable materials as they are known to have good thermal shock properties. They are commonly subject to thermal stress during installation. Once installed, the service life is then determined mainly by the corrosion characteristics. In this work three refractories were studied to observe and correlate mechanical properties with thermal shock behavior. The materials and their principal crystalline phases are: AM (Alumina–Mullite 35), Am (Alumina–Mullite 10), and AZ (Alumina–Zircon).

All the materials have similar open porosity and pore size distribution. The mechanical characterization comprises: fracture toughness (K_{IC}), fracture initiation energy (γ_{NBT}) and work of fracture (γ_{WOF}). The dynamic elastic modulus E of the composites was measured by the excitation technique.

The water quenching method was used for the experimental evaluation of the thermal shock resistance (TSR). Thermal cycles with different quenching temperature gradients ΔT were applied and a cyclic water quenching was used for the thermal fatigue resistance (TFR) assessment. The TSR behavior was evaluated by measuring the decrease in E/E_0 ratio where E_0 and E are the dynamic elastic modulus before and after one quenching, respectively. The strength (modulus of rupture, MOR) of materials before and after the TSR test was also measured. The AM material showed the highest E, σ_f (MOR) and K_{IC} values. The elastic modulus remained relatively high (near 80%) up to a ΔT of 500 °C for the three samples. AM showed a higher reduction of E and MOR than Am and AZ. Considering the retained MOR and E with ΔT, Am and AZ have a similar behavior.

Theoretical TS parameters (R, R''' and R_{ST}) were calculated for the refractories. The parameters considering crack initiation (R = theoretical ΔTc) are very similar but their value differs considerably to those ΔTc observed experimentally. This fact can be explained if we consider that the microstructure of refractory materials initially has defects and microcracks. The R''' parameters are the same for all materials. For our materials the R_{ST} parameter reflected the TSR damage.

The best TSR and TFR of AZ followed by Am are due to the microcracks size and their distribution in the microstructure of the materials. In AM refractory the high content and great grain size of Mullite produce the appearance of greater cracks than in the other materials.

The usage of these materials in glass service indicates that the AM material has a low TSR resistance.

1. Introduction

Aluminosilicate based refractories are commonly used in glass processing systems. Other compositions included Zircon to improve corrosion resistance. [1–4] These materials are widely used in the glass industry in forehearth, distributor, feeders, and as expendable materials such as plungers, spouts, tubes, orifice rings. Coarse Alumina and Mullite powders and grains (chamottes) are used in the elaboration of these materials, and during processing and sintering we observe that very little, if any, overall shrinkage occurs. Therefore in order to achieve optimum fired density, and in consequence mechanical strength and resistance to corrosion by molten glass, a very efficient particle packing is required. But currently the open porosity of these materials is of 17–21% with an Alumina–Mullite or Alumina–Zircon matrix.

A material of this kind is prepared by blending controlled proportions of components having particle sizes ranging from multi-millimeter to sub-micrometer. The refractory material can therefore be considered to be developed by the bonding of the finer fractions of particles to each other, and to the surfaces of

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the large grains. These materials are commonly subject to thermal stresses during installation. Once installed, they are known to have good thermal shock properties, so the service life is then determined mainly by its corrosion characteristics when glass contact exists.

The evaluation or testing of the thermal shock resistance (TSR) of refractory materials has been studied by many authors in the passed century, always involving many interacting variable factors and parameters [5,6]. A recent review on refractories TSR testing provides some background regarding the basic material properties and other factors that control TSR of refractory materials [7].

There is not just one simple and universal test to evaluate the TSR of ceramic materials and also capable of extrapolating the actual conditions in service, sample geometry, and thermal cycles. However, some experimental tests consisting in sudden heating and cooling are easily made but they have only a comparative value between similar materials. Testing of TSR can simply be done by visual or microscopic microstructure damage evaluation but this is not a very precise method and it depends strongly on the operator as well as on the experimental conditions.

A practical test to evaluate the thermal shock resistance consists to observe the variation or change of some characteristic properties of the material. The TSR can be evaluated on heating or cooling, but in most methods a sudden cooling step is used owing to its greater severity. A simple method consists in heating the test probe to a desired temperature; followed by rapid cooling to room temperature (referred to us as the quenching method), by immersion in liquids such as water, oil or alcohol [8,9]. A characteristic mechanical property like fracture strength or elastic modulus (E) is measured before and after quenching. In this way the severity of the treatment can be studied by determining the relative drop in mechanical strength (MOR) or elastic modulus after exposure at a given thermal cycle. In addition, the damage in the material can be correlated after application of repeated thermal cycles. This can be done because these properties are related to the microstructure integrity, namely the number, size and shape of cracks developed.

The thermoelastic theory is the first attempt to determine the thermal stresses and to introduce the damage resistance parameters of ceramic material, which is focused in the initiation of the fracture (R and R') [5]. A second approach focuses on crack propagation for more severe conditions of thermal shock than those for crack initiation (R''') [10]. A unified theory of the thermal shock resistance considering the initiation and crack propagation was then proposed [11,12] for high strength refractories with short initial cracks (R'''') and for lower strength refractories with larger initial cracks (R_{ST}) [13]. Both models consider γ_{WOF} and E. Aksel [14] studied the TSR behavior of Alumina–Mullite–Zirconia materials using the quenching technique; he concluded that Zircon appears controlling the mechanical properties and the improvement of the TSR.

In this work three kinds of commercial refractories commonly employed in glass feeders were studied to observe and correlate mechanical properties with thermal shock behavior. A complete mechanical and fracture evaluation of the materials was carried out. The TSR behavior was evaluated by two methods: first by mechanical strength decrease (σ_f; modulus of rupture, MOR) of the materials before and after the TSR was measured, and second by measuring the decrease in the E/E_0 ratio where E_0 and E are the dynamic elastic modulus before and after one quenching, respectively. The results of these two methods were then correlated. Microstructure damage was also observed by SEM. Finally the experimental results were related and compared with the theoretical parameters calculated from the experimental mechanical and fracture properties.

2. Experimental procedures

2.1. Characterization techniques

Probes (25 mm × 25 mm × 150 mm) were cut from long refractory pieces. Density and open porosity of the samples were determined by the water absorption method.

Crystalline phases formed were analyzed by XRD (Philips 3020 equipment with Cu-Kα radiation in Ni filter at 40 kV–20 mA). The dynamic elastic modulus E of the composites was measured by the excitation technique with a GrindoSonic, MK5 "Industrial" Model. Dilatometry of samples was performed using a Netzsch dilatometer up to 1400 °C at a heating rate of 10 °C/min [14,15]. The thermal expansion coefficient α of these materials up to 1000 °C was determined. Microstructural examination was conducted with a scanning electron microscope SEM (Jeol JSM 6360 LV) after polishing the probes surface. The MOR (flexural strength, σ_f) was measured on the bars with rectangular section using the three-point bending test (Universal testing machine INSTRON 4483). A span of 120 mm and a displacement rate of 2.5 mm/min were employed. The fracture toughness (K_{IC}) and the fracture initiation energy (γ_{NBT}) were evaluated by the single edge notched beam method [16,17] using notched bars with notches of 0.3 mm wide and depths between 0.5 and 10 mm. In this method K_{IC} is given by:

$$K_{IC} = \frac{3QLC^{1/2}}{2WD^2}\left[A_0 + A_1\left(\frac{C}{D}\right) + A_2\left(\frac{C}{D}\right)^2 + A_3\left(\frac{C}{D}\right)^3 + A_4\left(\frac{C}{D}\right)^4\right] \quad (1)$$

where Q is the load applied to the notched bar in kg, L is the span, C is the depth of the notch, D is the thickness of the specimen, W is the width of the specimen, and A_0, A_1, A_2, A_3 and A_4 are functions of the ratio (L/D) described in [18]. Eq. (1) can be approximated by the following equation:

$$K_{IC} \cong \sigma_f\sqrt{\pi C} \quad (2)$$

Here σ_f is the flexural strength in MPa. The calculated values of K_{IC}, together with E, were used to estimate the surface energy for the area created by the crack initiation (γ_{NBT}) [17,18] which are related by the following equation.

$$K_{IC} = \sqrt{2\gamma_{NBT}E} \quad (3)$$

where γ_{NBT} can be expressed as

$$\gamma_{NBT} = \frac{{K_{IC}}^2}{2E} = \frac{{\sigma_f}^2\pi C}{2E} \quad (4)$$

The work of fracture γ_{WOF} was measured considering the load (σ)–displacement (ε) curve area (energy) divided by twice the fracture surface area (A) (Eq. (5)) [18].

$$\gamma_{WOF} = \frac{\int \sigma\, d\varepsilon}{2A} \quad (5)$$

In this case the bars (25 mm × 25 mm × 150 mm) were notched with diamond saw (0.3 mm wide) between 8 and 10 mm. deep. The displacement rate applied was 0.1 mm/min. Five bars were notched for the γ_{WOF} evaluation. Finally the critical crack length (Lc) was estimated from Eq. (2)

$$Lc = \left(\frac{K_{IC}}{\sigma_f\sqrt{\pi}}\right)^2 \quad (6)$$

The critical length was also proposed by Hasselman [11] in terms of the surface energy. The values obtained by the two methods differ in a constant:

$$Lc = R'''' = \frac{E\gamma_{WOF}}{{\sigma_f}^2(1-\nu)} \quad (7)$$