Materials Science and Engineering A 498 (2008) 208–215

Contents lists available at ScienceDirect

Materials Science and Engineering A

journal homepage: www.elsevier.com/locate/msea

Effect of the addition of mullite–zirconia to the thermal shock behavior of zircon materials

N.M. Rendtorff*, L.B. Garrido, E.F. Aglietti

Centro de Tecnologia de Recursos Minerales y Cerámica (CETMIC: CIC-CONICET-UNLP), Camino Centenario y 506. C.C.49 (B1897ZCA) M.B. Gonnet. Buenos Aires, Argentina

A R T I C L E I N F O

Article history:
Received 18 February 2008
Received in revised form 21 July 2008
Accepted 25 August 2008

Keywords:
Zircon
Mullite–zirconia
Composites
Mechanical properties
Thermal shock resistance

A B S T R A C T

Ceramics and refractories are subjected to local temperature and atmosphere gradients due to the successive application of heating cooling cycles, during service. These situations originate thermal stresses that cause certain degree of damage to the material defining the potential usage of the refractory. The favorable effect of the addition of a second phase for improving the thermal shock resistance (TSR) of dense ceramics have been proved previously. However the addition must be controlled because it can excessively affect the densification and microstructure. The objective of this work is to study the TSR of pure zircon ceramic materials and to quantify the influence of the addition of mullite–zirconia (MZ) grains in a range between 15 and 45 wt.%. Cool water quenching test was carried out on slip cast prismatic bars in order to study the TSR. For the thermal shock tests temperatures differences (ΔT) between 200 and 1200 °C were applied. The elastic dynamic modulus (E) was evaluated by the impulse excitation technique as a function of the temperature difference of the quenching test. The decrease in the E modulus was also correlated with the number of thermal cycles. The materials were characterized by density measurements and mechanical properties. The XRD and Rietveld method were employed for determining the crystalline phase composition. Finally, the microstructure of the materials was examined by SEM. Significant changes in the E modulus were observed for ΔT over 400 °C for all composites studied.

© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Zircon ($ZrSiO_4$) is good refractory material because it does not undergo any structural transformation until its dissociation at about 1700 °C. It exhibits many attractive properties such as excellent chemical stability, a very low thermal expansion coefficient ($4.1 \times 10^{-6}\,^{\circ}C^{-1}$) from room temperature to 1400 °C and low heat conductivity coefficient 5. 1 W/m °C at room temperature and 3.5 W/m °C at 1000 °C.

Sintered zircon with high purity can retain its bending strength up to temperatures of 1200 and l400 °C [1]. These properties make zircon a potential candidate as a useful structural ceramic, especially in such fields where a sudden change of temperature may occur. These materials are widely used in the steel and glass industries [2–4]. The presence of impurities decreases the dissociation and usage temperatures. Moreover, pure zircon dense materials degradation by a thermal shock is sometimes the limiting property for its applications.

On the other hand mullite–zirconia (MZ) electrofused powders, commercially available, have demonstrated good refractory properties in structural applications and as an additive to other phases forming a composite material [5–10].

The thermal shock resistance is improved by the deliberate introduction of thermal stress concentrators in the form of microstructural inhomogeneities in the material. Thus, the dispersion of zirconia grains in a mullite matrix, an improved thermomechanical behavior is obtained as a result of microcracks formation and by the dissipation of elastic energy related to zirconia martensitic transformation [11].

The first approach to determine the thermal stresses of brittle material is the thermo-elastic theory [12,13], which is focused in the initiation of the fracture. A second approach focuses on crack propagation for conditions of thermal shock more severe than those for crack initiation [13]. A unified theory of the thermal shock resistance considering the initiation and crack propagation in brittle ceramics was developed by Hasselman [14] who presented analytical solutions for the extent of the crack propagation as a function of the severity of the thermal shock. Therefore, several theoretical thermal shock parameters were proposed previously [15] to predict the thermal shock resistance (TSR) of a certain material. These are functions of the global properties of the material.

The principal objective of the present work is to study the influence of the added amount of mullite–zirconia on the mechanical properties and the thermal shock resistance of dense zircon

---

* Corresponding author. Tel.: +54 2214840247; fax: +54 2214710075.
E-mail address: rendtorff@hotmail.com (N.M. Rendtorff).

0921-5093/$ – see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.msea.2008.08.036

materials. The elastic modulus (E) was the property chosen to follow the material damage. This can be done because E is related to the microstructure integrity, meaning the number, size and shape of cracks developed. E was measured by a dynamic method, using the impulse excitation technique (sonic velocity), that is a simple and non destructive standard test [16]. In this way the severity of the treatment was studied by determining the relative drop in elastic modulus after exposure at a given thermal cycle. Also the retention of E was evaluated after application of repeated thermal cycles. Finally, the TSR was compared with the behavior predicted from parameters R, R'''' and R_{st} which were obtained from the thermoelastic and energy balance theories respectively.

2. Experimental

2.1. Slip casting of dense zircon based materials

A commercial zircon powder was used as principal raw material. With: $ZrO_2 = 64–65.5$ wt.%, $SiO_2 = 33–34$ wt.%, $Fe_2O_3 \leq 0.10$ wt.% and $TiO_2 \leq 0.15$ wt.%, specific gravity of 4.6 g/cm^3, melting point of 2200 °C and mean diameter (D_{50}) of 1.5 μm (Kreutzonit Super, Mahlwerke Kreutz, Germany).

Electrofused mullite–zirconia was used as second raw material. With $Al_2O_3 = 44.0$ wt.%, $SiO_2 = 18.0$ wt.%, $ZrO_2 = 39.1$% $TiO_2 \leq 0.3$ wt.%, $Fe_2O_3 \leq 0.1$ wt.% and $Na_2O \leq 0.2$ wt.%, specific gravity of 3.74 g/cm^3 melting point of 1850 °C and mean diameter (D_{50}) of 5 μm (MUZR, ElfusaLT, Brazil).

Zircon and MZ mixtures were prepared with 0, 15, 25, 35 and 45 wt.% (in weight basis) of MZ and were called Z0, Z1, Z2, Z3, and Z4, respectively. Aqueous 80 wt.% suspensions of the mixtures at pH 9.1–9.2 were prepared by adding the powder to aqueous solutions with suitable content of dispersant (Dolapix CE64, Zschimmers and Schwartz) and NH_4OH. After mixing, the suspensions were ultrasonicated for 20 min. The prismatic bars 7.5 × 7.5 × 50 mm^3 were produced from well dispersed suspensions by slip casting in a plaster molds.

Dried probes were fired at heating rate of 10 °C/min up to 1600 °C for 2 h.

2.2. Characterization techniques

After drying at 110 °C green density of bars was determined by mercury immersion. Density and open porosity of sintered samples were determined by the water absorption method. Theoretical density was calculated taking into account the density of each component: 3.16 g/cm^3 for mullite, 5.89 g/cm^3 for zirconia and 4.56 g/cm^3 for zircon. Crystalline phases formed were analyzed by X-ray difraction (XRD) (Philips 3020 equipment with Cu–Kα radiation in Ni filter at 40 kV – 20 mA). The Rietveld method [17,18], a quantitative analysis, was carried out to characterize the present crystalline phases in the materials. The XRD patterns were analyzed with the program FullProf, which is a multipurpose profile-fitting program, including Rietveld refinement [19]. The starting crystallographic data for each phase was extracted from the literature. The dynamic elastic modulus E of the composites was measured by the excitation technique with a GrindoSonic, MK5 “Industrial” model. For the TSR experiments, the water quenching method was used. Thermal cycles with quenching temperature differentials, ΔT of 400, 600, 800, 1000 and 1200 °C were applied. Sintered sample was heated at a selected temperature in an electrical furnace in air atmosphere for a period of 90 min and then cooled in a water bath at 25 °C. After quenching, samples were dried at 100 °C and then the TS severity effect on E as well as its variation with the number of applied thermal cycles was determined for the different ΔT. Dilatometry of samples sintered at 1600 °C was performed using a Netszch dilatometer up to 1400 °C at a heating rate of 10 °C/min. 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 until to 1 μm diamond paste. Flexural strength (σ_f) was measured on the bars with rectangular section using the three-point bending test. (Universal testing machine INSTRON 4483) 40 mm of span 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 (SENB) [10,20] using a three-point bending universal testing machine. Samples of dimensions (7.5 × 7.5 × 50 mm^3) were notched with diamond saw of 0.3 mm thickness, with depth between 0.3 and 2.5 mm. The three-point test was carried out at room temperature with a rate of 0.1 mm/min. 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 in meters, C is the depth of the notch in meters, D is the thickness of the specimen in meters, W is the width of the specimen in meters, and A_0, A_1, A_2, A_3 y A_4 are functions of the ratio (L/D) described in [10,21]. Eq. (1) can be approximated by the following equation:

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

where 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 propagation (γ_{NBT}) by the subsequent equation [10,20]:

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

where γ_{NBT} can be expressed:

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

Finally, the critical crack length (L_c) was estimated from Eq. (2):

$$L_c = \left(\frac{K_{IC}}{\sigma_f\sqrt{\pi}}\right)^2 \quad (5)$$

The critical length was also proposed by Hasselman [14] in terms of the surface energy less than a constant; the values obtained by the two methods differ in a constant:

$$L_c = R'''' = \frac{EU_{ef}}{{\sigma_t}^2(1-\nu)} \quad (6)$$

This method (SENB) can only be used if the sizes of the defects (pores, cracks etc.) in the microstructure of the materials are smaller than L_c.

3. Results and discussion

3.1. Characterization of the sintered Z0–Z4

In Table 1, the physical characteristics and mechanical properties of the materials sintered at 1600 °C–2 h are shown. All the materials studied had relative densities over the 90% of the theoretical and the open porosity was less than 5%. The values of the elastic modulus and the flexural strength are comparable with literature data [22–24].

Download English Version:

https://daneshyari.com/en/article/1581427

Download Persian Version:

https://daneshyari.com/article/1581427

Daneshyari.com