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ScienceDirect
Ceramics International 35 (2009) 1077–1081
CERAMICS INTERNATIONAL
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Behavior of silicon carbide/cordierite composite material after cyclic thermal shock

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Received 10 January 2008; received in revised form 7 April 2008; accepted 28 April 2008
Available online 18 July 2008

Abstract

In the present work Mg-exchanged zeolite and silicon carbide were used as starting materials for obtaining cordierite/SiC composite ceramics with weight ratio 30:70. Samples were exposed to the water quench test from 950 °C, applying various number of thermal cycles (shocks). Level of surface deterioration before and during quenching was monitored by image analysis. Ultrasonic measurements were used as non-destructive quantification of thermal shock damage in refractory specimens. When refractory samples are subjected to the rapid temperature changes crack nucleation and propagation occurs resulting in loss of strength and materials degradation. The formation of cracks decreases the density and elastic properties of material. Therefore by measuring these properties one can directly monitor the development of thermal shock damage level. Dynamic Young's modulus of elasticity and strength degradation were calculated using measured values. Level of degradation of the samples was monitored before and during testing using Image Pro Plus program for image analysis. The capability of non-destructive test methods such as: ultrasonic velocity technique and image analysis for simple, and reliable non-destructive characterization are presented.

Keywords: C. Thermal shock resistance; D. Cordierite/SiC composite ceramics; E. Refractories; Ultrasonic velocity; Image analysis

1. Introduction

Furnaces require the use of high temperature ceramics as furnace liners. These ceramic liners undergo rapid temperature changes and high temperature and stress gradients that lead ultimately to the nucleation and growth of cracks by a phenomenon that is generally known as thermal shock. In most cases the subcritical damage prosesses promote a gradual chipping of the refractory ceramic tile until the furnace has to be shut down for the replacement of brick. The result is significant economic losses in lost production time and the brick replacement costs. It also represents the most significant cost in the maintenance of steel production plants.

Thermal shock resistance dictates refractory performance in many applications. In many instances, a twofold approach, i.e. (1) material properties [1–4] and/or (2) heat transfer conditions [5–7] is used to characterize thermal shock behavior of the refractories. As an alternative, information on the thermal shock behavior of refractories can be obtained experimentally. One test for this purpose, which is highly popular because of its simplicity, consists of quenching appropriate specimens from an oven temperature into a medium such as water, liquid metal, oil, or fused salts maintained at a lower temperature. Water quench test is usually applied for thermal stability testing. Thermal quenching of the refractories leads to the crack nucleation and/or crack propagation resulting in loss of strength. Since the formation of the cracks has a profound influence on the ultrasonic velocity and the Young modulus of the material, measuring either of these properties may be applied to monitor the development of the thermal shock

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0272-8842/$34.00
doi:10.1016/j.ceramint.2008.04.029

damage level. The goal of this work is to use non-destructive testing methods and their advantages for prediction of thermal shock behavior. Destruction of the samples was analyzed using Image Pro Plus Program. This is a very convenient method for determining the damage surface level in sample due to the thermal shock. In this paper relationship between change in mechanical characteristics (strength and Young modulus of elasticity) and behavior of the samples during thermal shock will be given.

2. Materials

Cordierite and silicon carbide are ceramic materials suitable for high temperature application with good chemical resistance. Their target application is in furnaces for use at temperatures over 1000 °C. Cordierite has a superior thermal stability, thermal shock resistance and low thermal expansion coefficient. Silicon carbide has superior hardness, good chemical resistance, high values of conductivity, low values of thermal expansion coefficient, and excellent thermal stability and thermal shock resistance. An overview of typical properties of cordierite and silicon carbide are given in Table 1. The ceramic multi-component composite material could exhibit advantages of its constituents when the components have optimized properties and they are mixed in the proper ratio.

A mixture of Mg-exchange zeolite, alumina (Al_2O_3) and quartz (SiO_2) corresponding to a cordierite stoichiometry was attrition milled in ethyl alcohol media for 4 h.

Cordierite/SiC composite ceramics with weight ratio 30:70 (samples KZ 30) were prepared by milling with Al_2O_3 balls in DI water in polyethylene bottle for 24 h and firing at 1160 °C.

3. Experimental

3.1. Thermal shock

Thermal stability of the refractories was determined experimentally by water quench test (JUS. B. D8. 319.). Samples were cylinders with 1-cm diameter and 1-cm high. Each thermal shock cycle consisted of several consequent steps. Slow heating up at a nominal heating speed of 10 °C/min to the quench temperature set at 950 °C, holding at this temperature for 30 min to reach thermal equilibrium in whole specimen volume and finally quenching into water bath at temperature of 23 °C. Samples KZ 30 were thermally cycled up to 36 cycles. The experimental method is similar to the procedure described in PRE Refractory Materials Recommendations 1978 (PRE/R5 Part 2).

Table 1
Typical values of selected properties of dense constituents used in refractory materials investigated [8,9]

	Coriderite	Silicon carbide
Chemical formula	2 MgO–Al_2O_3–5 SiO_2	SiC
Density (g/cm^3)	2.60	3.1
Modulus of elasticity (GPa)	70	410
Poisson ratio	0.21	0.14
Compressive strength (MPa)	350	460
Fracture toughness (MPa m$^{0.5}$)	–	4.6
Linear thermal expansion coefficient (10^{-6} K^{-1})	1.7	3.1
Hardness, moss	7.5	9.5

3.2. Non-destructive measurements

3.2.1. Monitoring the damaged surface area in refractory specimen during thermal shock

Samples were cylinders 1-cm diameter and 1-cm high. Photographs of the samples were taken, before and after water quench test. Samples surfaces were marked by different colors, in order to obtain a better resolution and difference in damaged and non-damaged surfaces in the material. Non-damaged surface was calculated as ideal surface of the sample using common equation ($Po = d^2\pi/4$). Diameter of the samples was measured using Image Pro Plus program. For this investigation damage of the samples was monitored using Image Pro Plus Program [8]. Some of the photographs are given in Fig. 1.

White area is non-damaged surface and black (dark) area is damaged surface. Obtained results for material destruction as ratio of P/Po, were given as function of number of quench experiments, N (Fig. 2).

3.2.2. Ultrasonic determination of dynamic Young modulus of elasticity

Ultrasonic pulse velocity testing (UPVT) [8] was first used on refractory materials in the late 1950s. Various publications have dealt with the practical application of UPVT to characterize and monitor the properties of industrial refractory materials non-destructively [8–21]. The UPVT method has been considered in detail in ref. [8]. Briefly, pulses of longitudinal elastic stress waves are generated by an electro-acoustical transducer that is held in direct contact with the surface of the refractory under test. After traveling through the material, the pulses are received and converted into electrical energy by a second transducer. Most standards describe three possible arrangements for the transducers:

(1) the transducers are located directly opposite each other (direct transmission),
(2) the transducers are located diagonally to each other; that is, the transducers are across corners (diagonal transmission),
(3) the transducers are attached to the same surface and separated by a known distance (indirect transmission).

The velocity, v, is calculated from the distance between the two transducers and the electronically measured transit time of the pulse as

$$V\,(\mathrm{m/s}) = \frac{L}{T} \quad (1)$$

where L = path length (m) and T = transit time (s).

By determining the bulk density, the Poisson's ratio and ultrasonic velocity of a refractory material it is possible to calculate the dynamic modulus of elasticity using the equation

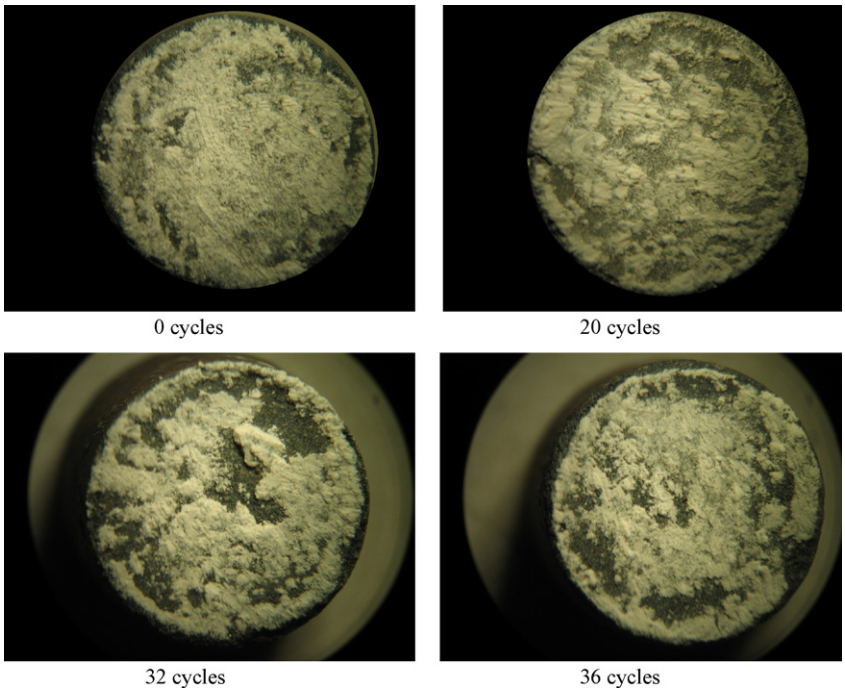

Fig. 1. Samples KZ 30 under thermal shock experiment white area is non-damaged surface, black (dark) area is damaged surface.

below [10–24]:

$$E_{dyn} = V^2\rho\left(\frac{(1+\mu_{dyn})(1-2\mu_{dyn})}{1-\mu_{dyn}}\right) \quad (2)$$

where V is the pulse velocity (m/s), ρ is the bulk density (kg/m^3) and μ_{dyn} is the dynamic Poisson ratio. For the calculation of Poisson ratio the following equation was used.

$$\mu_{dyn} = \frac{(2\alpha^2-1)}{(2\alpha^2-2)} \quad (3)$$

Parameter α is defined as ratio of longitudinal (V_p) and transversal velocities (V_s), described using the following equation:

$$\alpha = \frac{V_p}{V_s} \quad (4)$$

The measurement of ultrasonic velocity was performed using the equipment OYO model 5210 according to the standard testing procedure (JUS. D. B8. 121). The transducers were rigidly placed on two parallel faces of the cylindrical sample having 1-cm diameter and 1-cm height using Vaseline grease as the coupling medium. The ultrasonic velocity was then calculated from the spacing of the transducers and the waveform time delay on the oscilloscope.

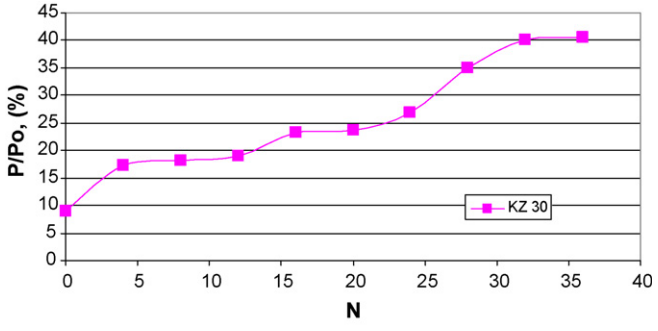

Fig. 2. Damaged surface level (P/Po) versus number of quench experiments (N).

4. Results

4.1. Ultrasonic velocity and strength degradation

Some of the measured values for the samples are given in Table 2.

From the obtained results given in Table 2 and comparison with the selected material properties given in Table 1, it could be observed that material KZ 30 has lower density and Young modulus of elasticity. As goal of our investigation was to synthesize porous material with good thermal stability resistance, our results confirm porosity and very good thermal resistance, which was confirmed by 36 cycles of water quench test.

Results for material KZ 30 will be presented versus quench experiments and level of degradation during thermal cycling.

Table 2
Measured values for using ultrasonic determination of dynamic Young modulus of elasticity

Parameter	Value
Number of cycles, N	0
Density, ρ (kg/m^3)	1.756
Longitudinal velocity, V_p (m/s)	860
Transversal velocity, V_s (m/s)	400
Poisson ratio, μ_{dyn}	0.36
Young modulus of elasticity, E (GPa)	0.872

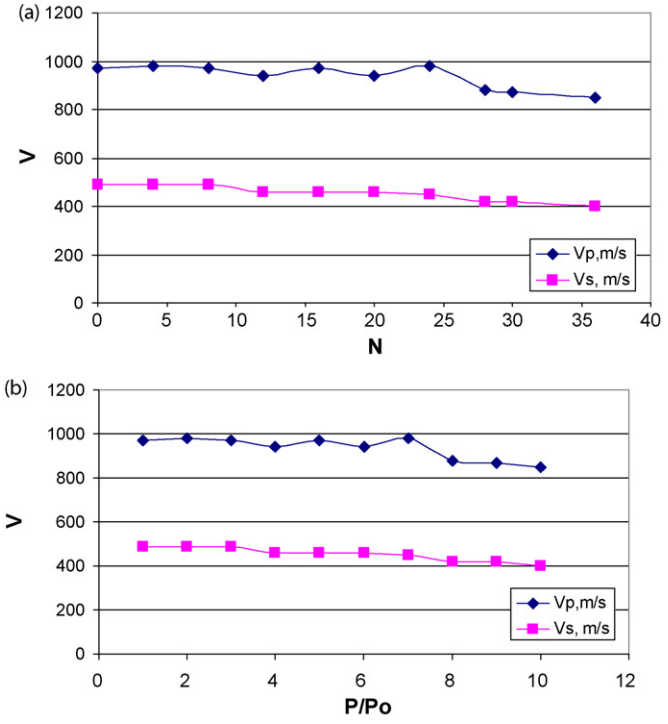

Fig. 3. (a) Values of ultrasonic velocity (V) during testing (longitudinal V_p and transversal V_s) versus number of quench experiments of material KZ 30. (b) Values of ultrasonic velocity (V) during testing (longitudinal V_p and transversal V_s) versus degradation (P/Po) for material KZ 30.

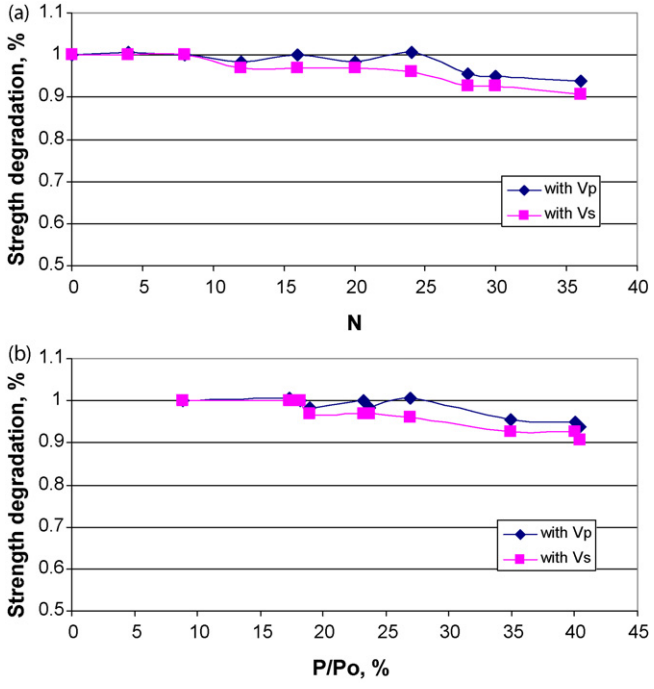

Fig. 4. (a) Strength degradation of material KZ 30 versus number of quench experiment. (b) Strength degradation of material KZ 30 versus degradation (P/Po).

Degradation of ultrasonic velocity, strength degradation and Young modulus of elasticity changes are given in Figs. 2–4.

Obtained results and values of the measured ultrasonic velocity (V_p) abut 1000 m/s indicates porosity of the sample. Results for the velocity changes in both materials suggests that materials were very stable during testing, as degradation of the velocity was not too below from the velocity of the sample before water quench test. These results indicate that number of nucleated cracks and crack propagation did not resolute in rapid degradation of strength and Young modulus of elasticity, and samples exhibited an excellent thermal shock behavior.

The expression for the strength degradation, based on decrease in ultrasonic velocity was used [3,6,11]:

$$\sigma = \sigma_0 \left(\frac{V_L}{V_{L0}} \right)^n \quad (5)$$

where σ_0 is compressive strength before exposure of the material to the thermal shock testing, V_L is longitudinal or ultrasonic velocity after testing, V_{L0} is longitudinal or ultrasonic velocity before testing and n is material constant ($n = 0.488$, Ref. [3]). This equation was used for calculation with longitudinal and transversal ultrasonic velocity. Obtained results for the strength degradation base on results of ultrasonic measurements, and calculated using Eq. (5) were presented in Fig. 3a and b.

Results for the strength degradation presented in Fig. 3a and b showed that strength degradation at the end of the test was between 0.90 and 0.93% for the material KZ 30. This result indicates minimal strength degradation and explains excellent results for water quench test, as result of 36 rapid temperature changes.

4.2. Results for Young modulus of elasticity

Results for the monitoring changes of the Young modulus of elasticity during quenching are shown in Fig. 4. Results for the dynamic Young modulus of elasticity indicates that values before testing indicates that material is porous, but degradation during testing was very stable, which explained 36 cycles of water quench test.

5. Discussion

Thermal shock behavior of the two materials was investigated. Three different techniques were applied:

- water quench test, as most popular experimental method,
- detection of damaged surface area in refractory specimen during thermal shock and
- non-destructive determination of dynamic Young modulus of elasticity.

Obtained results showed that both materials are excellent candidates for the application where thermal shock resistance is required. Water quench results showed that samples were stable till 36 cycles. Behavior of the samples was monitored during water quench test in order to determine damage of the original surface of the samples. Results given in Fig. 1 showed that during quenching damage of the original surface was not exceed 50%. Original surface showed damage about 8.8% for the KZ 30. This damage before the test explains higher values for the damage at

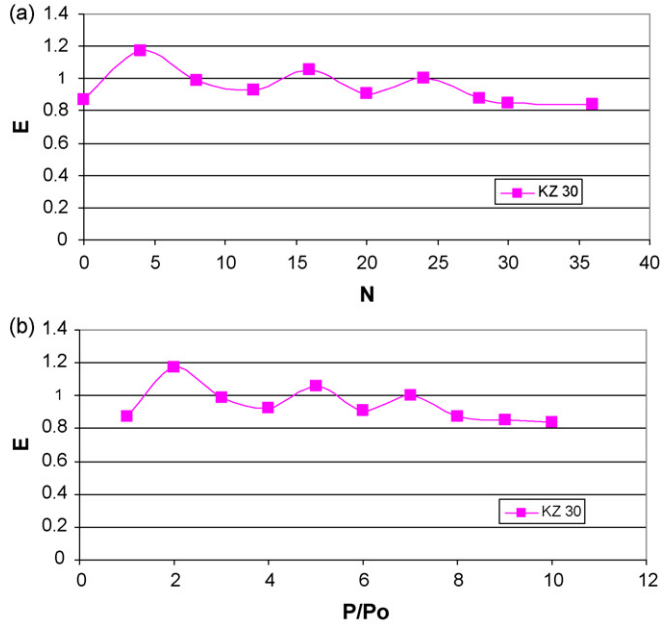

Fig. 5. (a) Dynamic Young modulus of elasticity versus number of quench experiments (*N*). (b) Dynamic Young modulus of elasticity versus degradation (P/Po).

the end of the procedure, which had not overcome level 45% at the end of the test, which is excellent result.

Behavior of the bulk of the sample was monitored using ultrasonic measurements of the Young modulus of elasticity. Results presented in Fig. 5 showed very small changes and degradation of the Young modulus. These results are pointing out that the level of destruction in the bulk of the material and fracture nucleation and growth did not exceed level for material destruction. Results for velocity and strength degradation pointed out these conclusions.

6. Conclusion

Samples under investigation were exposed to certain number of cycles (from 0 to 36). Thermal shock behavior of cordierite/SiC composite ceramics was investigated. Ultrasonic pulse velocity testing was used to determine ultrasonic velocity, Young's modulus of elasticity in cordierite/SiC composite material. Presence of defect in samples during thermal cycling was monitored using Image Pro Plus Program.

As the experimental procedure added to the water quench test for thermal stability behavior determination was described and discussed in detail, it appears that implementation of these methods and their advantages will improve materials characterization and help in preventing and improvement of material properties and synthesis conditions for achieving the best results in thermal stability resistance characteristics of material.

Acknowledgement

This research was financed by the Ministry of Science and Environment of Serbia as part of projects MXT.6717, OI 142016 and TR 16004.

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