THERMAL SHOCK RESISTANCE OF YTTRIA-DOPED TETRAGONAL ZIRCONIA POLYCRYSTALS

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ABSTRACT

Thermal shock fracture behavior of various kinds of zirconia ceramics such as magnesia partially stabilized zirconia (Mg-PSZ), yttria and ceria doped tetragonal zirconia polycrystals (Y-TZP and Ce-TZP), Y-TZP/Al_2O_3 composites and yttria doped cubic stabilized zirconia (Y-CSZ) was evaluated by water quenching method. The tetragonal to monoclinic phase transformation behavior of zirconia based ceramics around cracks introduced by thermal stress was investigated by using Raman microprobe spectroscopy. Thermal shock resistance of zirconia based ceramics increased with increasing the fracture strength, but that of Y-TZP and Y-TZP/Al_2O_3 composites was anomalous lower than the predicted value. From the results of thermal shock fracture behavior of Y-TZP with various grain sizes, the critical quenching temperature difference (ΔTc) increased from 250°C to 425°C with increasing grain size of zirconia from 0.4μm to 3.0 μm, while the fracture strength decreased from 900MPa to 680MPa. The improvement of ΔTc of Y-TZP with increasing grain size of zirconia corresponded with the quantity of tetragonal to monoclinic phase transformation around cracks introduced by thermal stress.

INTRODUCTION

Since brittle ceramics are susceptible to catastrophic failure under conditions of thermal stress introduced by temperature difference, thermal shock resistance of ceramic materials is an important property for high temperature structural applications. It is well known that silica glass with low thermal expansion coefficient, cordierite and aluminum titanate ceramics show excellent thermal shock resistance. However, these ceramics are not applied as high strength structural ceramics because of their relatively low fracture strength. One of the most excellent ceramics with high fracture strength and high thermal shock resistance is Si_3N_4 ceramic. Many studies have been carried out to elucidate the basic principles governing the thermal stress fracture of brittle ceramics.[1-3] The quenching test into liquid media such as water has been used extensively for characterizing the thermal shock resistance of ceramics, because this test provides relatively simple quantitative thermal shock resistance, e.g. a critical quenching temperature difference, ΔTc, and the residual strength for $T<\Delta Tc$. It was reported[4] that the observed thermal shock resistance was generally in good agreement with the predicted value by using the thermal shock resistance parameter, $R_0=\sigma_f(1-\nu)/\alpha E$, where σ_f is the fracture strength, ν is Poisson's ratio, E is Young's modulus and α is the coefficient of linear thermal expansion. Since yttria doped tetragonal zirconia polycrystals (Y-TZP) show extensively high fracture strength such as 800-2500MPa by stress-induced phase transformation from tetragonal to monoclinic phases[5,6], they are expected to show high thermal shock resistance. However, the thermal shock resistance of Y-TZP is caused by a thermal stress significantly smaller than the original fracture stress [7-9]. The thermal shock fracture behavior of Y-TZP has not been clarified in details, but Ishitsuka et al[10] reported that the toughening mechanism by the stress-induced phase transformation did not function well against the thermal stress in Y-TZP based ceramics because the degree of the tetragonal to monoclinic phase transformation around cracks introduced by the thermal stress was much smaller than that introduced by the mechanical stress by using Raman microprobe spectroscopy. Since the resistance to the tetragonal to monoclinic phase transformation of zirconia is expected to decrease with increasing grain size, the increase in grain size of Y-TZP may result in increasing the degree of the tetragonal to monoclinic phase transformation around cracks introduced by the thermal stress. From both experimental results of the thermal shock fracture behavior and Raman microprobe spectroscopy in 2mol%Y_2O_3 doped tetragonal polycrystals with various grain size, the ΔTc increased from 250°C to 425°C with increasing grain size of zirconia from

0.4µm to 3.0µm[11]. In the present paper, the thermal shock fracture behavior of Y-TZP is discussed in details.

EXPERIMENTAL PROCEDURE

Y-TZP powder containing 2mol%Y_2O_3 was used as starting powder. After uniaxially pressing at 10MPa to form plates $5x30x50mm^3$, these plates were isostatically pressed at 200MPa. The green compacts were presintered at 1400°C to 1650°C for 3hr in air. The presintered samples were hot isostatically pressed at 1400°C to 1700°C and 150MPa for 1hr under Ar gas atmosphere. The samples were cut into bars with size of 5mmx2mmx15mm and polished to parallel mirrorlike planes. The thermal shock resistance of each specimen was determined by a quenching test using water at 0°C as the quenching medium ; the severity of thermal shock was adjusted by varying the temperature of the specimen hold for 30 min, in a tube furnace. The bending strength of the specimen after thermal shock was measured by the 3-point bending test with a crosshead speed of 0.5mm/min and a span length of 10mm. The bulk density of sample was measured by the Archimedes method. The grain size of sample was determined by the line intercept method[12] from SEM micrograph. Raman microprobe spectra in the vicinity of cracks produced on quenching were recorded using a double monochrometer and a direct current amplification method over the Ramanshift range of 100 to $350cm^{-1}$. The 514.5nm line of an argon ion laser was used as excitation. The spectra were recorded for the following optional conditions : probe diameter about 1µm, 100x objective lens, spectrum scan rate of $120cm^{-1}/min$, and resolution power of 2 to $3cm^{-1}$.

RESULTS AND DISCUSSION

A general expression for the thermal stress, S_t, in a ceramic materials is written by equation (1)[2].

$$S_t=\sigma^{*}\alpha E\Delta T/(1-\nu) \qquad (1)$$

where ΔT is the temperature difference between the sample and the environment, α is the coefficient of linear thermal expansion, E is Young's modulus, ν is Poisson's ratio and σ^* is non-dimensional maximum thermal stress. When the thermal shock fracture of ceramics is caused by the thermal stress resulting from convective heat transfer, σ^* can be expressed by equation (2)[2].

$$1/\sigma^*=1.451(1+3.42k/r_0h) \quad (2)$$

where r_0 is the half-thickness of the plate sample, h is the heat transfer coefficient and k is the thermal conductivity. Since the thermal stress fracture may be proceeded by the condition where the thermal stress equals the tensile strength, σ_t, of the materials, the critical quenching temperature difference, ΔTc, above which the fracture strength

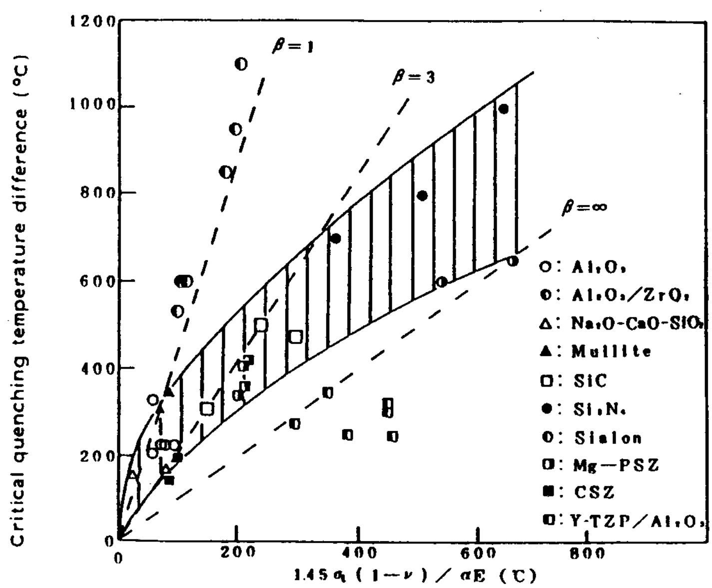

Fig. 1 Relation between Tc and R_0 parameter, $1.451\sigma_t(1-\nu)/\alpha E$[13]

degrades, is expressed by equation (3).

$$\Delta Tc=\sigma_t(1-\nu)/\alpha E\sigma^* \quad (3)$$

Since $1/\sigma^*$ is more than 1.451, Tc is greater than $R_0=1.451\sigma_t(1-\nu)\alpha E$, where R_0 is the thermal shock resistance parameter. The relation between the critical quenching temperature difference, ΔTc, and R_0 parameter in various kinds of structural ceramics is shown in Fig. 1[13]. In this figure, β is Biot's modulus and the dashed lines were calculated by equations (1) and (2) with corresponding values of β. It is seen that ΔTc increased with increasing R_0. It was also noticeable that although Y-TZP and Y-TZP/Al_2O_3 ceramics showed larger value of R_0 than that of SiC, the results of ΔTc were anomalously small ; ie as expected the value of ΔTc of most ceramics were larger than R_0 parameter, but those of Y-TZP and Y-TZP/Al_2O_3 composites were significantly smaller than R_0. These results indicated that the cracks in Y-TZP based ceramics were propagated by the thermal stress being smaller than the original tensile stress. Therefore, it is thought that the toughening mechanism of zirconia by the stress -induced phase transformation did not function sufficiently against thermal stress fracture of Y-TZP based ceramics. From the results of Raman microprobe spectroscopic studies around cracks introduced by thermal shock fracture, it was found that no phase transformation from tetragonal to monoclinic phase was occurred by thermal shock fracture in Y-TZP based ceramics[10].

Relative density, grain size, and three-point bending strength of 2Y-TZP samples prepared are listed in Table 1.

TABLE 1 Properties and Sintering Conditions of 2Y-TZP

Presintering Temperature (°C)	HIPing temperature (°C)	Grain size (μm)	Bending strength (MPa)	Relative density (%)
1400	1400	0.4	900	100
1450	1500	0.9	630	100
1550	1600	1.6	630	99.8
1650	1700	3.0	680	100

All samples were densified to theoretical density. The grain size increased from 0.4μm to 3.0μm with increasing sintering temperature. The bending strength of sample with grain size of 0.4μm was 900MPa, but the bending strength of samples with grain size of 0.9 to 3.0μm was almost constant at 630 to 680MPa.

The Raman microprobe spectra around cracks formed right after quenching and the critical quenching temperature difference, ΔTc, in 2Y-TZP with various grain size are shown in Fig. 2.

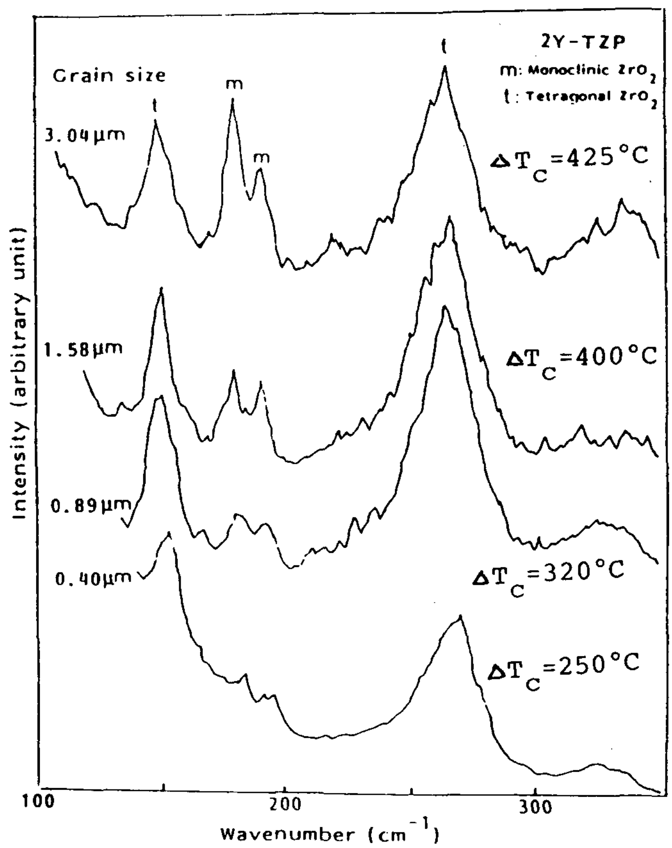

Fig. 2 Raman spectra around cracks introduced by the thermal stress and ΔTc in 2Y-TZP with various grain sizes

Though only peaks at 148 and 264cm^{-1} corresponding to the tetragonal bands[14] were observed in the Raman spectra of the surface without cracks in all samples, a monoclinic doublet[14] at 181 and 192cm^{-1} was observed with the tetragonal bands in the Raman spectra around cracks introduced by the thermal stress. As shown in Fig. 2, the peak intensity of the monoclinic doublet increased with increasing grain size and ΔTc increased from 250°C to 425°C with increasing grain size from 0.4 to 3.0μm. These results indicated that the increment of the grain size of Y-TZP ceramics increased the contribution of the transformation toughening against not only the mechanical stress fracture but also thermal stress fracture, which consequently improved both fracture toughness and thermal shock resistance. The value of 425°C of ΔTc in 2Y-TZP with grain size of 3.0μm was in good agreement with the calculated value of ΔTc in 2Y-TZP with fracture strength of 680MPa.

Y-TZP has excellent fracture strength and fracture toughness by transformation toughening mechanism. As mentioned before, in Y-TZP, high fracture strength requires smaller grain size of zirconia, but larger grain size for high thermal shock resistance. On the other hand, it is well known that the fracture strength of Y-TZP degraded due to the phase transformation from tetragonal to monoclinic phase by annealing at 50-300°C under the presence of water molecule and the thermal stability of Y-TZP strongly depended on the grain size of zirconia[15]. It was found that the thermal stability of Y-TZP significantly decreased with increasing the grain size of zirconia[15]. Therefore, the fabrication of Y-TZP with high fracture strength, high thermal stability and high thermal shock resistance is very difficult. The present results indicate that the research and development in Y-TZP materials corresponds to their application for structural ceramics.

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