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On the Brittle-to-Ductile Transition of Y-PSZ Single Crystals

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Abstract

The fracture behaviour (fracture mechanisms, fracture toughness, and strength) of yttria-partially stabilized zirconia (Y-PSZ) single crystals has been investigated between room temperature and 1700°C. Cleavage was the fracture mechanism acting over the entire temperature interval studied, with some plastic elements arising at temperatures of 1000°C and above, accounting for the increasing fragmentation with temperature observed for the precipitation-hardened materials. Fragmentation is ensured from interdomain cleavage on {1 0 0} and {1 1 0} planes. No subcritical cracks were observed. The strength and fracture toughness follow behaviour typical for materials with a pronounced brittle-to-ductile transition. After a gradual decrease between room temperature and 1000°C, both properties show a sharp increase under elevated temperatures, followed by a sharp decrease. This increase is associated with dislocation plasticity revealed in delaminations along {1 1 0} and {1 0 0} planes. The lower temperature limit of the brittle-to-ductile transition was estimated as 1000°C. A higher limit was not reached and the transition of Y-PSZ [1 0 0] single crystals into a ductile state probably occurs at about 2000–2100°C. The mechanism of ductile fracture is presumably intergranular sliding of grains, formed in the course of plastic deformation, from domain microstructure. © 1996 Elsevier Science Limited.

1 Introduction

For a long time, zirconia as well as zirconia ceramics were considered as materials only for low-temperature application. Mainly for this reason their high-temperature mechanical and fracture behaviour has not been the subject of intensive systematic study. Lack of interest in high-temperature behaviour was due to the belief that the mechanical properties of some zirconia materials drop too drastically with temperature,[1,2] therefore investigators generally did not extend testing to high temperatures to determine the behaviour above 1000–1200°C.

At the same time, an investigation of the brittle-to-ductile transition of different refractory materials, metallic and ceramic, has shown that after some decrease in properties, properly tailored brittle materials can develop a pronounced enhancement of strength and fracture toughness at high temperatures.[3] For polycrystalline materials, this enhancement depends strongly on the chemical composition of the grain boundaries as well as the grain size. Granular structure facilitates the transition of a body from a brittle state into a ductile one via high-temperature intergranular sliding. Before it can fall into a ductile state, the single crystalline material has to form a new dislocation, similar to a polycrystalline structure, during the predeformation stage. Only after this event can the single-crystal material use the intergranular sliding mechanism of failure.[4]

This phenomenon has received little investigation in the case of zirconia ceramics, and is the reason why this study was undertaken. The specific objective was to obtain systematic data on the fracture mechanisms of yttria- partially stabilized zirconia (Y-PSZ) single crystals during the temperature-induced brittle-to-ductile transition, using tests for strength and fracture toughness of a limited number of samples.

2 Material and Experimental Techniques

As materials for the study, partially stabilized (by 6 mol% of yttria) zirconia single crystals, grown by scull melting in the Francevich Institute for Problems of Materials Science, were used.

Since the scull-melting technique gives comparatively small crystals with a high scatter of directions, only a limited number of specimens, that were nearly similar in composition, was available for analysis. We succeeded in finding about 30 samples to study, to a first approximation, the temperature dependence of the strength of smooth and notched samples, using only one sample for each temperature selected in both tests.

Samples of dimensions 3 × 4 × 45 mm, oriented (using the X-ray technique) along the [1 0 0] direction, were cut by a diamond saw, carefully ground and polished with diamond abrasives of up to 1.0 μm grain size. The processed samples, with a discrepancy of [1 0 0] directions of approximately 10' were annealed in air at 1600°C for 8 h and furnace-cooled. They were then tested in a three-point bend in a vacuum of ~10^{-3} Pa at temperatures between room temperature and 1700°C, with a loading frame rate of ~10^{-3} m min^{-1}. Samples for fracture toughness testing were notched with a diamond saw to a radius of ~50 μm.

The structure of the thin sample sections was examined using transmission electron microscopy (TEM; Jeol JEM 100 CX II). Fracture mechanisms were studied by scanning electron microscopy (SEM; Jeol Superprobe-733).

3 Results and Discussion

TEM shows that the materials under study consisted of three {1 1 2} type tetragonal variants or ferroelastic domains[5] with sizes of 2 × 0·2 μm, as seen in the dark-field images of Figs 1(a), (b) and (c) and the electron diffraction pattern of Fig. 1(d). The twin boundaries lay in the (1 1 0) planes of a pseudo-cubic symmetry. The dark-field image of {1 1 2} type reflections for the [1 1 1] beam reveals that {1 1 2} tetragonal variants have slightly different volumes due to the release of mechanical stresses.

The temperate dependences of flexural strength and fracture toughness for the partially stabilized tetragonal zirconia single crystals, derived using only one sample for each temperature, are shown in Fig. 2. Nevertheless, it can be seen that the data lie on curves that are typical for materials with a pronounced brittle-to-ductile transition.[3] Strength decreases from ~1000 to 400 MPa within a temperature range between 300 and 1000°C, after which it increases rather sharply up to ~1.2 GPa with the maximum at 1300–1500°C. At 1700°C the single crystals again have a bend strength of ~700 MPa. Similarly to bend strength, the fracture toughness decreases from 7 to 4 MPa m$^{1/2}$ in the temperature interval between room temperature and 1300°C. After 1300°C K_{Ic} increases rather sharply, reaching 18 MPa m$^{1/2}$ at 1700°C. The high-temperature maximum as well

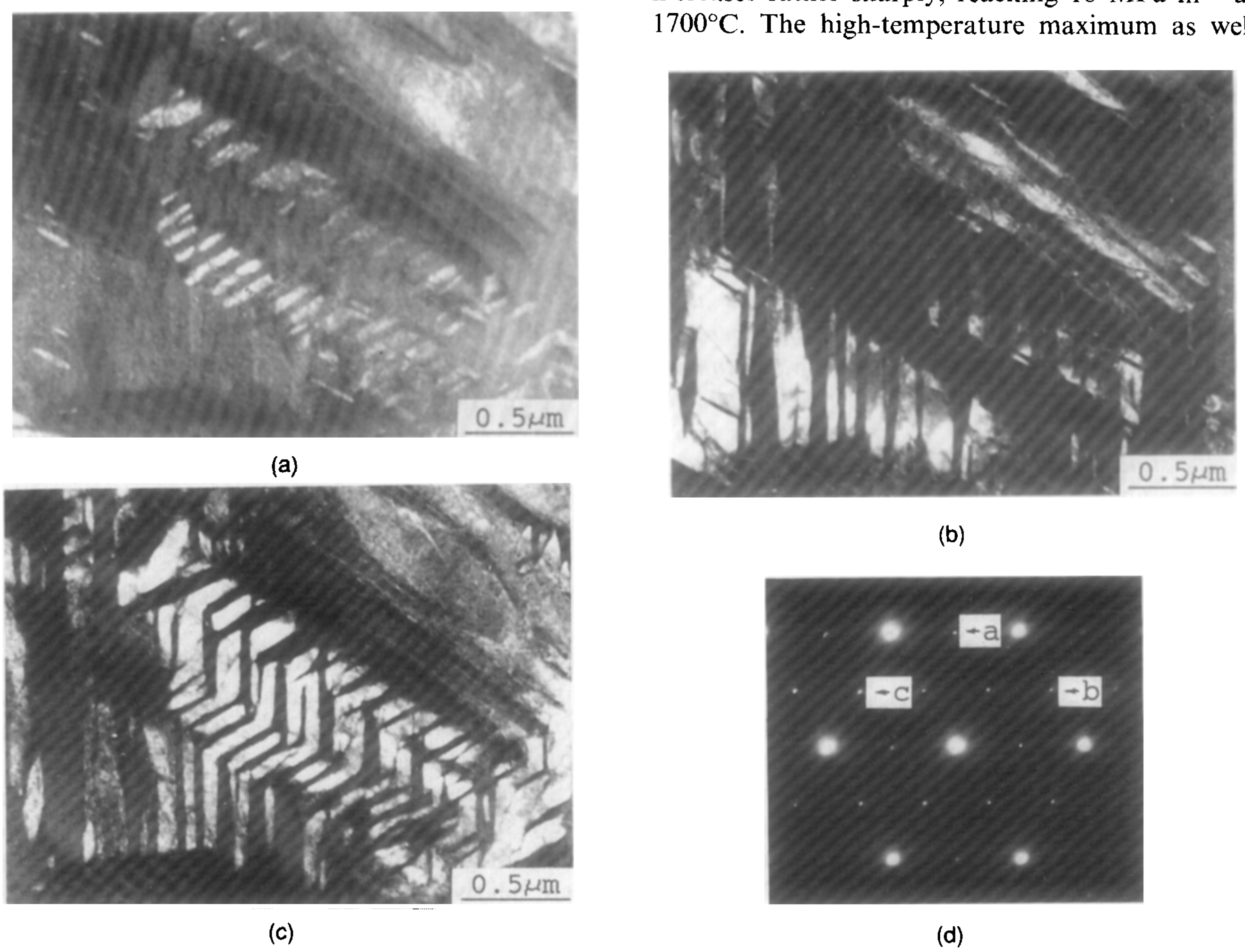


Fig. 1. (a), (b), (c) TEM dark-field images and (d) diffraction pattern of three {1 2 2} tetragonal twin variants of partially stabilized zirconia single crystals in a [1 1 1] zone axis.

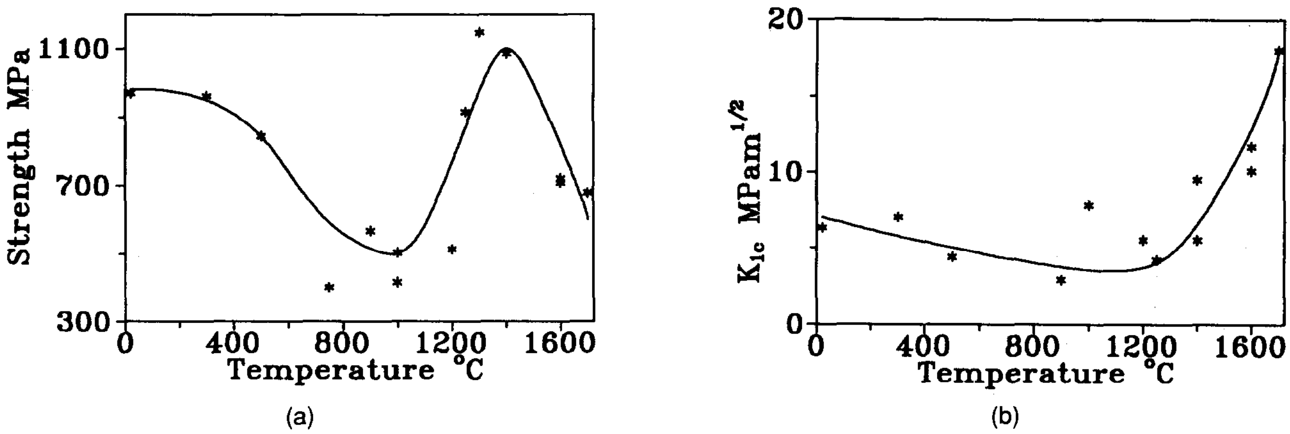


Fig. 2. Temperature dependences of the flexural strength and fracture toughness of partially stabilized zirconia single crystals.

as the following decrease of fracture toughness will probably be achieved at higher temperatures. This means that the maximum of fracture toughness is achieved at a temperature 300–400°C higher than that of bend strength. The low-temperature part of our data is close to that published earlier.[1]

Fractographical analysis shows that the main fracture mechanism is cleavage. This acts over the entire temperature interval investigated, from room temperature up to 1700°C, in both types of samples: notched and unnotched. No subcritical cracks preceding cleavage were observed.

3.1 Unnotched samples

The fracture surface features of the unnotched samples may be detailed as follows.

(1) Fracture originates from the side surfaces

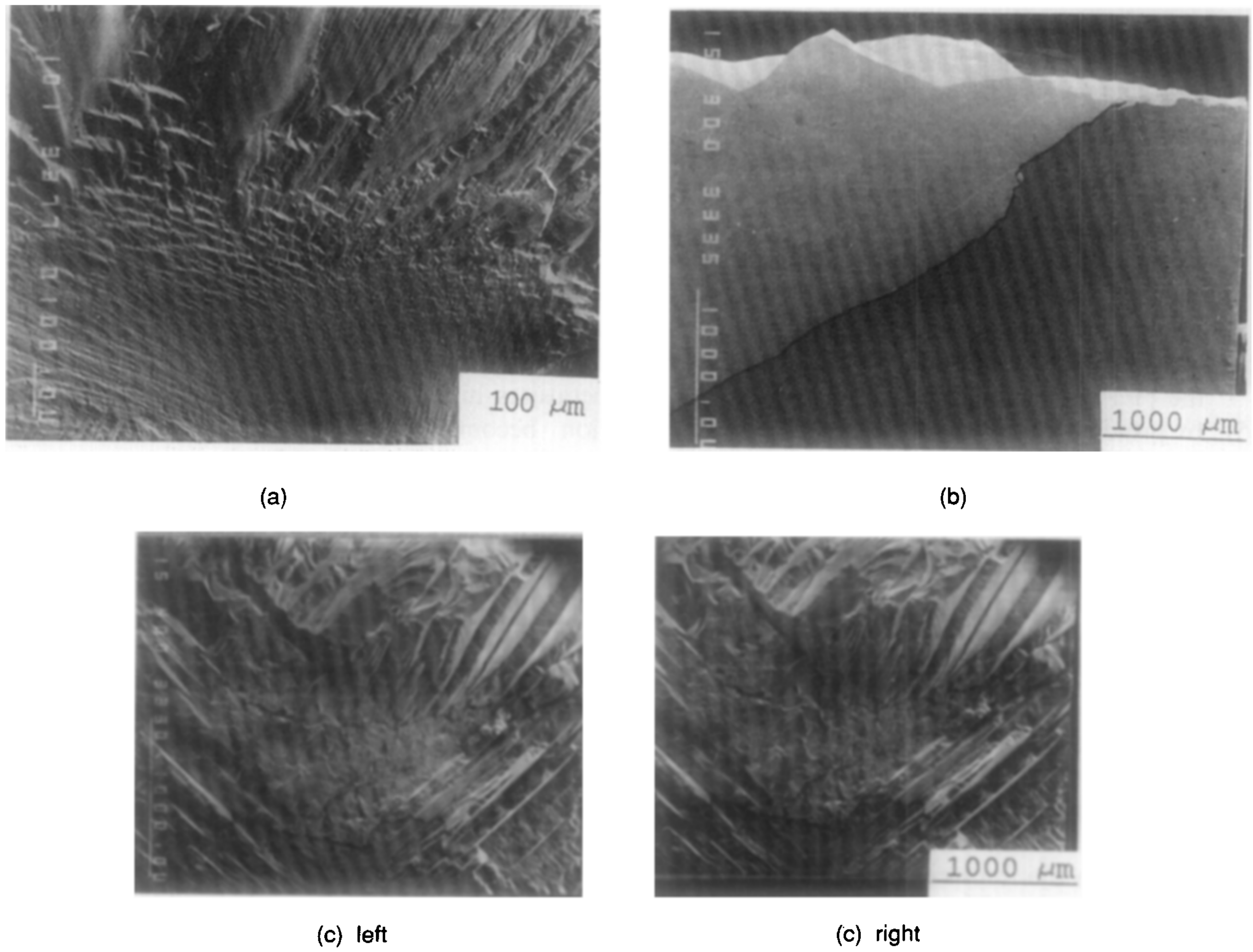
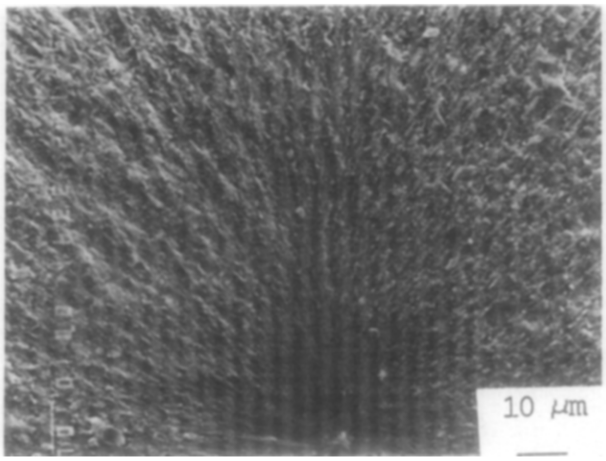
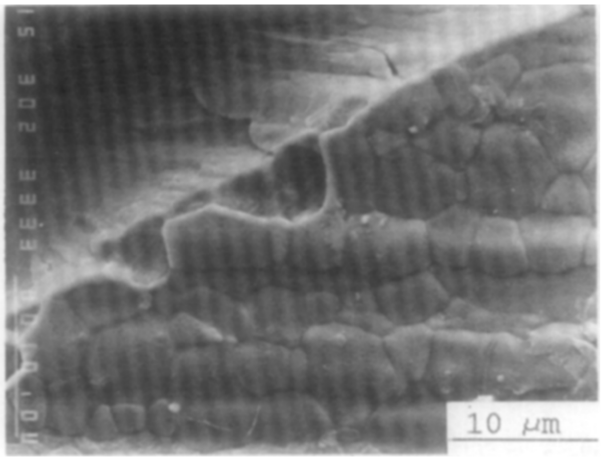


Fig. 3. SEM micrographs of fracture surfaces of Y-PSZ single crystals tested for strength at different temperatures. (a) Cleavage and Wallner lines at 900°C, cracking starts from the sample side surface; (b) view of crack branching on the sample side surface at 1600°C; (c) double branching and rough delaminating cracks in the fracture surface. Stereopair: (d) fan-like pattern in fracture surface at 1250°C; (e) recrystallized surface layer at 1600°C.



(d)



(e)

Fig 3. Continued.

of the samples in all cases [Figs 3(a), 3(c), 3(d)]. Cracks begin from stress concentrators such as grinding flaws or edges, and at the beginning actually lie in the crystallographic surfaces close to {1 0 0} as shown earlier.[1] Sometimes, cracks lying in {1 1 0} planes may be observed in side surfaces.

(2) Up to 1000°C the fracture surfaces have no distinguishing features except for chop–offs opposite the original side of the sample and Wallner lines [Fig. 3(a)].

(3) Beginning at 1000°C, dynamic crack branching with an angle of about 30–35° may be seen [Figs 3(b), 3(c)]. Some samples show double branching [stereopair in Fig 3(c)].

(4) Also beginning from 1000°C, rough delaminating cracks may be seen [Fig. 3(c)]. Samples tested at 1700°C additionally show fine cracks; these are transverse to the rough and deep cracks. Delaminating cracks lie in the {1 1 0} planes.

(5) In principle, the second phases (as well as some likenesses of them) in zirconia single crystals should give rise to some fragmentation of the cleavage surface.[6] This may actually be seen under higher magnification in all samples, as will be demonstrated later for notched samples [see, for example, Figs 4(a) and (b)].

(6) For all the temperatures studied, the directions of cleavage, crack propagation in the {1 0 0} planes is practically indifferent to the crystallographic orientations of the crystals. This conclusion follows from the fan-like pattern in the fracture surface [Fig. 3(d)].

(7) The surface defects acting as origins of cleavage at temperatures of 1400°C and above are probably intergranular cracks of the recrystallized surface layer, and are clearly seen on the surface of high-temperature samples [Fig. 3(e)]. The thickness of the recrystalized surface layer is ~1–3 μm and its structure inherits the directions of scratches on the sample surface resulting from mechanical processing with diamond powders. Recrystallized grains with a size of ~2–10 μm peel off during crack propagation.

3.2 Notched samples

The predefined location of the process-zone, as well as the suppression of common plastic deformation by the notch, almost completely eliminates some features that are visible on the fracture surfaces of smooth, unnotched samples. First of all, for the notched samples, the fracture surface is rather flat over the temperature interval between room temperature and 1300°C. Only hems and a slight growth of fragmentation sharpness with temperature may be seen ([Figs 4(a) and 4(b)]). Dynamic crack branching is only observed at temperatures higher than 1300°C, whereas delamination becomes slightly visible only at 1700°C [compare Figs 4(c) and (d)].

Thus the fractographical analysis clearly shows that, in spite of the flexural strength passing its high-temperature maximum, the transition of the tetragonal PSZ single crystals of the [1 0 0] orientation into the plastic state is not completed at 1700°C because the fracture mechanism is not changed. Cleavage takes place in the {1 0 0} plane as well as {1 1 0}. The lower-temperature boundary of the brittle-to-ductile transition, in accordance with Ref. 3, may be determined as ~1000°C, at which temperature some marks of plastic deformation can be found in the fracture surface. Such marks are delaminating cracks [Fig. 3(c)]; the number and their delaminating ability grow with temperature. Delaminating cracks are parallel to the {1 1 0} planes and are probably original, lying in those planes where the boundaries of domain microstruc-

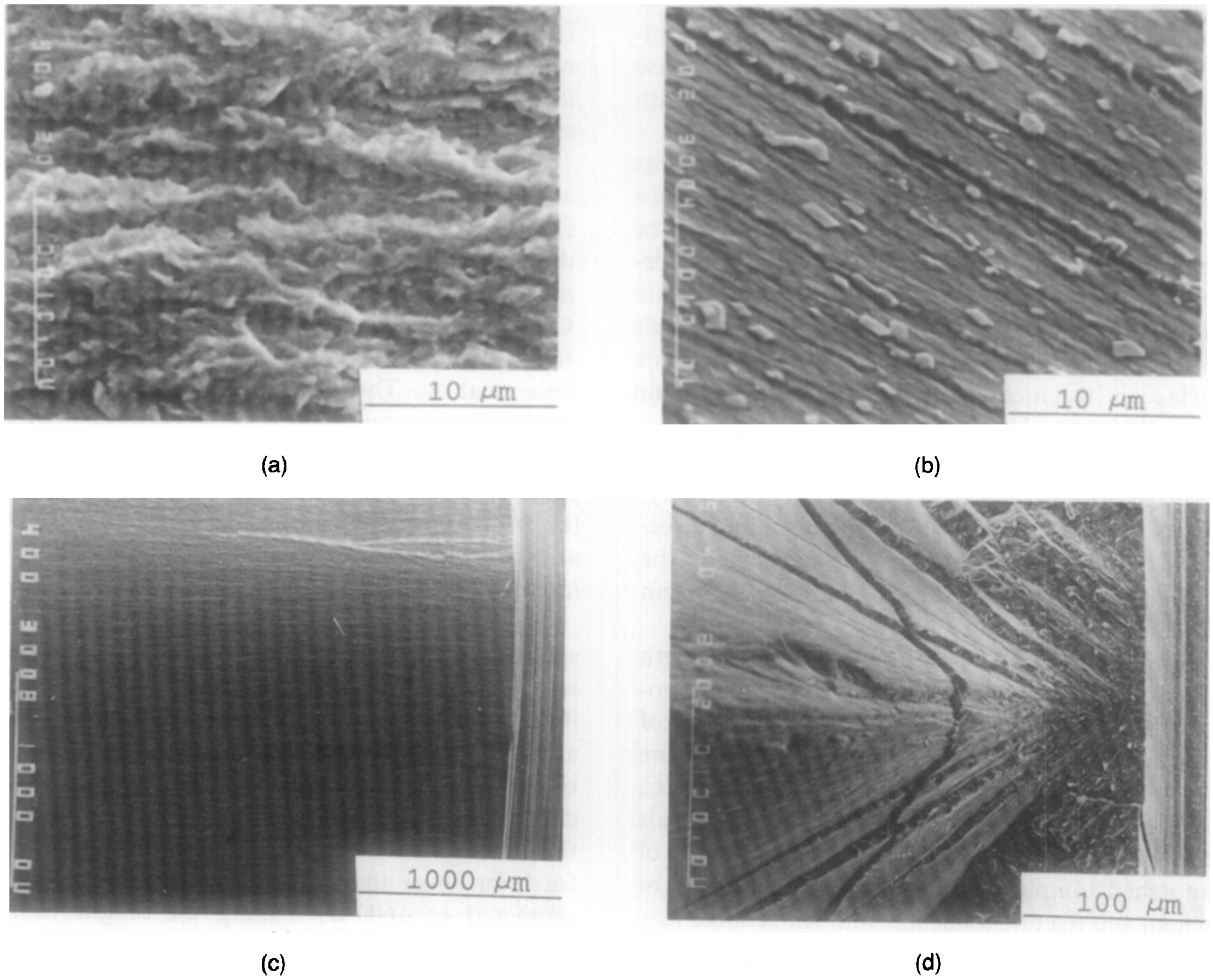


Fig. 4. SEM micrographs of fracture surfaces of Y-PSZ single crystals tested for fracture toughness at different temperatures. (a) Low-temperature fragmentation of cleavage surface at 20°C; (b) high-temperature fragmentation of cleavage surface at 1700°C; (c) crack starts from the notch at 20°C; (d) crack branching and delamination at 1700°C.

ture are located. By analogy with refractory metals, e.g. Mo,[6] microplasticity may result in crack nucleation, namely in boundaries. These cracks divide the crystal into microcrystals separated by delaminating interfacial cracks. Every delaminating microcrystal deforms separately. Some of them may develop rather high plasticity preceding fracture and fail in a ductile manner with the formation of knife-like fracture as seen in the stereopair of Fig. 5. As may be observed, some parts of the knife may fail, equally, as well as the cleavage.

Microplasticity resulting in delaminations may be the reason for the strength enhancement with temperature, due to relaxation of stresses. Increasing

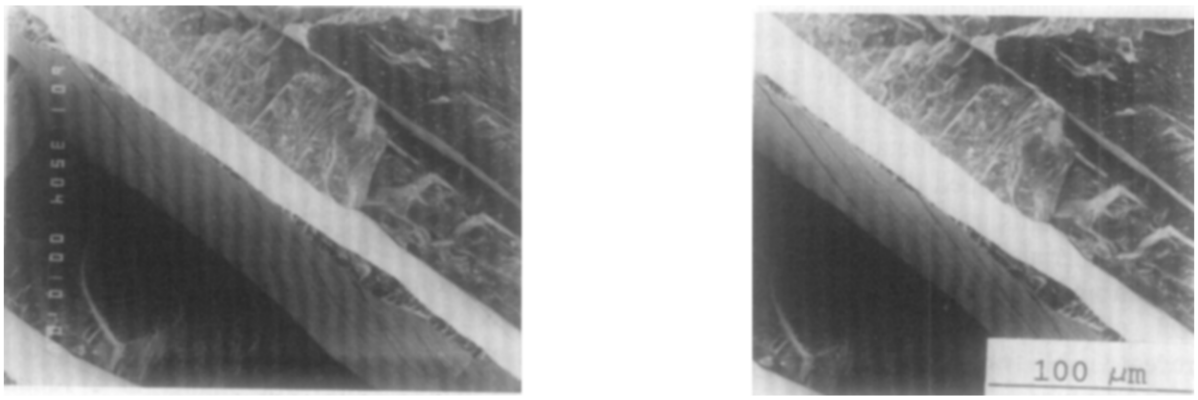


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Fig. 5. The knife-like fracture in Y-PSZ single crystals at 1700°C. Stereopair.

delaminations result in decreasing strength at higher temperatures. During this time, the delaminations promote the fracture toughness increase observed in the fracture toughness behavior with temperature.

Delaminations are practically the only elements of ductile fracture observed on the fracture surface at 1000°C. For this reason, this temperature may be determined as the lower boundary of the brittle-to-ductile transition. Therefore the temperature range from low temperatures to 1000°C corresponds to the area of brittle fracture. The cleavage fracture surface is fragmented by boundaries of domain microstructure. The fragmentation becomes more visible with temperature, revealing the growing role of interdomain boundaries at high temperatures. Cleavage planes are of the {1 1 0} or {1 1 0} family. Cracks propagate in the {1 0 0} plane in any direction without any preference, as evidenced by the fan-like pattern of fracture in the area of origin [Fig. 3(d)].

Comparing fracture toughness and strength behaviours (Fig. 2), it is possible to see their proportional reductions with temperature: the factor of reduction is about 2·3. In the very wide temperature interval from room temperature up to about 1200°C, the fracture toughness of Y-PSZ single crystals behaves in the same manner as the strength of unnotched samples. These identical behaviours of strength and fracture toughness with temperature may indicate the independence of fracture toughness, determined in practice from the concept of $K_{1c} = \sigma\sqrt{\pi c}$, on the parameters of the rather blunt notch. This may be correct in the case where fracture behaviour is controlled by inner, stronger concentrators of stresses than outer ones. The outer concentrators localize the place of failure, whereas actual fracture stresses are produced by inner defects. Such defects may be mainly microcracks; those in our case have an interdomain or interphase nature. Exactly which interphase, interdomain microcracks determine the mechanical behaviour of Y-PSZ single crystals is indicated by the views of cleavage fracture surfaces with intensive fragmentation and delamination [Figs 3(c), 4(a) and 4(b)]. Fragmentation of the cleavage surface by dislocations only cannot give such an intensive and developed fragmentation. Strongly deformed ionic[4] or metallic[6] single crystals do not reveal the developed delaminating relief of fracture surface. This is typical for dispersion-strengthened materials with weakly cohesive particles delaminating from the matrix under loading.[6] Even steels containing well-known iron carbides do not reveal visible fragmentation of the cleavage surface.

Fragmentation of the cleavage fracture surface in Y-PSZ single crystals results from the special defect structure of these crystals. Defects are localized in the {1 0 0} and {1 1 0} planes, forming a network of boundaries (Fig. 1). The crystal loses its homogeneity and assumes the so-called domain structure with the stressed boundaries lying in the {1 0 0} and {1 1 0}, crystallographic planes, which at that time are revealed as cleavage planes. All this facilitates strong microcracking along these interdomain boundaries, thereby ensuring the important mechanism of fracture toughness enhancement. Crystallographic tetragonal–monoclinic transformation also facilitates microcracking and subsequent delamination. The fact that microcracking is the reason for delamination may be confirmed by the appearance of delaminating cracks containing clearly visible cleavage steps in Fig. 5. Growing with temperature, the dislocation microplasticity intensifies the process of damage accumulation by boundaries and thereby also facilitates the nucleation of interdomain cracks.

The transition to ductile fracture under the specified loading condition begins at ~1000°C with the appearance of rough delaminating cracks [Fig. 3(c)]; the dynamic cleavage crack branching also becomes apparent at this point. The angle of branching, as may be observed in Fig. 3(b), is close to 35°. Taking into account the assumption that twice the elastic deformation energy is needed for branching,[7] the surface energy of fracture was estimated at 1600°C. Taking the length of the cleavage crack before branching to be 0·3 mm [Fig. 3(c)] the surface energy for cleavage was found to be equal to 500 J m^{-1}; this corresponds to a fracture toughness of 11·7 MPa $m^{1/2}$, measured with notched samples.

Unfortunately, we did not succeed in observing the ductile fracture of Y-PSZ single crystals. The upper temperature limit probably lies far above 1700°C and the temperature required could not be attained with testing equipment available. In full analogy with the fracture behaviour of model ceramics, namely NaCl single crystals,[4] and being based on plastic delamination as a forerunner of the plastic nucleation of intergranular (interdomain) cracks, we conclude that intergranular sliding of grains formed in the process of plastic deformation preceding fracture, might be the mechanism by which ductile fracture (like dimple fracture) occurs at elevated temperatures. The transition of Y-PSZ {1 0 0} single crystals into ductile fracture might occur between 2000 and 2100°C.

It was found also that recrystallization of the damaged, mechanical processing surface layer takes place at temperatures of 1600°C (i.e. 0·64 times the melting temperature) and higher in a vacuum of 10^{-3} Pa, leading to peeling of the polycrystalline scale (~1–3 μm) under loading [Fig. 3(e)]. In principle, this indicates that there is a possibility to control the structure as well as the properties of zirconia ceramics by high-temperature plastic deformation and heat treatment.

4 Conclusions

Y-PSZ [1 1 0] single crystals exhibit a pronounced brittle-to-ductile transition that occurs at temperatures between 1000 and 2000–2100°C. At around 1400°C the samples possess a strength that is 20–30% greater than at low-temperature and more than twice the fracture toughness. Cleavage was the fracture mechanism over the whole range of temperatures studied, its action probably extending up to 2100°C. Cleavage planes are the {1 0 0} and {1 1 0} habit ones. The cleavage surface is fragmented by interdomain boundaries as is attributed to precipitation–hardened materials. Fragmentation and delamination at elevated temperatures result from microcracking along interdomain boundaries lying in the {1 0 0} and {1 1 0} planes. Plastic deformation enhances fragmentation. Cleavage crack propagation is independent of crystallographic orientation. Dynamic crack branching takes place in the brittle-to-ductile transition and was observed in both notched and unnotched samples after some plastic deformation. No subcritical cracks preceding cleavage were observed. Recrystallization of a surface layer damaged by mechanical processing occurs at 1400°C and above. Single crystals are covered by a polycrystalline peeling film. Intergranular cracks nucleating in this film originate cleavage in the bulk of specimens. The probable mechanism of ductile fracture at temperatures near 2000°C is intergranular sliding of grains formed during plastic deformation preceding fracture.

In this case of brittle fracture by cleavage, the temperature dependence of fracture toughness is proportional to that of strength. Mechanical behaviour is determined only by the inner defects; these are interdomain microcracks also resulting in fragmentation of the cleavage surface. The notches in samples for fracture toughness tests only localize the place of fracture and prevent general yielding of the samples under elevated temperatures, thereby shifting the upper limit of the brittle-to-ductile transition to higher temperatures (up to 2000–2100°C).

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