Improvement of tensile ductility in 3 mol% yttria-stabilized tetragonal zirconia (3Y-TZP) by prestraining

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There have been numerous investigations of the effect of material testing variables such as strain rate, temperature and grain size on the elongation to failure of superplastic ceramics. This paper presents information on the effect of rapid prestraining on superplastic ductility in a fine-grained 3 mol % yttria-stabilized tetragonal zirconia (3Y-TZP), using two testing programmes: (i) prestraining up to 130% at a prestrain rate of $1 \times 10^{-3} s^{-1}$ followed by elongation to failure at a test strain rate of $1 \times 10^{-4} s^{-1}$ and (ii) prestraining to 60% at prestrain rates of $1 \times 10^{-3} s^{-1}$ and $2.5 \times 10^{-4} s^{-1}$ followed by elongation to failure at a slower test strain rate of $1 \times 10^{-4} s^{-1}$. The results showed that prestraining at the above conditions considerably improved superplastic ductility as well as reducing the time required to achieve a given elongation. The reason for this ductility enhancement is explained in terms of suppression of grain growth. © *1998 Chapman & Hall*

1. Introduction
The demonstration of superplastic flow in fine-grained tetragonal zirconia by Wakai [1] in 1986 led to considerable research on the generality of this phenomenon in other oxide and non-oxide ceramics. Superplasticity has now been demonstrated for many ceramics and ceramic composites, including yttria-stabilized tetragonal zirconia [1, 2], alumina [3, 4], silicon nitride [3], hydroxyapatite [5], zirconia–alumina [6] and silicon nitride–silicon carbide [7]. Large ductilities that have been recorded for some of the above ceramics, thereby promoting the commercial feasibility of superplastic forming of crystalline ceramics.

To encourage the commercial applicability of superplastic forming of ceramics, it is desirable that sufficient superplastic elongation can be attained at high strain rates and at relatively low temperatures. The current state of art is that for $\sim 100\%$ elongation, temperature of $> 0.8\ T_m$ and strain rates of $< 1 \times 10^{-4} s^{-1}$ are required for most ceramics, compared to $\sim 0.6\ T_m$ and $1 \times 10^{-2} s^{-1}$ for metals. A large amount of work has been carried out to assess the effect of test temperature, strain rate, grain size or doping with transition metal oxides on superplastic behaviour in a range of ceramics [1, 8–13]. The effect of prestraining, prior to superplastic testing has not been explored in ceramics although it has been extensively investigated in metals. In superplastic metals, prestraining is effective in improving superplastic ductility through refining the grain size at the early stage of deformation; the present study was aimed to determine whether similar effects could be manifest in a ceramic, using 3Y-TZP as a model material.

2. Experimental: materials and procedures
The ceramic used was a fine-grained 3 mol % yttria-stabilized tetragonal zirconia (3Y-TZP) powder, supplied by Mandoval Ltd. Zirconia Sales (UK) Ltd. The chemical composition of the powder was 5.4 wt% Y_2O_3 (equivalent to 3 mol %), 93.8 wt % ZrO_2 and the following impurities (in wt %): Al_2O_3 0.25, SiO_2 0.11, TiO_2 0.12, Fe_2O_3 0.003, Na_2O 0.02, and CaO 0.06. The material had an initial grain size of $\sim 0.2\ \mu m$.

A slip-casting method was used for the preparation of tensile specimens; this method allowed the economical production of complex, net shapes that required no machining. The slip casting slurry was prepared by dispersing the powder (3Y-TZP) in distilled water with a dispersing agent (Dispex A40); the slurry was then wet ball milled for 4 h to obtain a good dispersion. The milled slurry was injected by a syringe into a plaster mould. Cast specimens were released from the mould after ~ 10 min and then air dried at $\sim 25\ ^{\circ}C$ for a few days. These specimens were presintered at 1123 K to make them more handleable and smooth surfaces were obtained by carefully grinding off any casting protrusions. Specimens were then pressureless sintered in air at 1523 K for 30 min; specimens with $\sim 95\%$ of theoretical density were regularly produced [14, 15]. Density measurement were made by the Archimede's displacement method.

High temperature, uniaxial, tensile tests were carried out in air using an Instron 4505 testing machine. A single zone vertical split furnace with molybdenum disilicide elements was mounted on the crosshead of the test frame; tensile load was applied using high density sintered alumina rods in a pin loading mechanism. Careful specimen alignment was essential to avoid fracture on loading. After achieving the desired (uniform) test temperature, usually at a heating rate of 423 K h^{-1}, the specimen was held at that temperature for ~ 10 min before loading. The small tensile load was then applied on the specimen as a preload and the alignment checked before testing.

Deformation was continuously monitored using a computerized system equipped with a data acquisition facility that allowed tests to be controlled under a constant strain rate. Specimens tested were either pulled to failure without interruption or elongated to a predetermined strain. The work reported here involves test temperature of 1673 K and strain rates in the range of $1 \times 10^{-3} s^{-1}$ to $1 \times 10^{-4} s^{-1}$. Microstructural observations were carried out using a Philips scanning electron microscope (SEM) 525 for grain size measurements and a 300 KV Philips EM430T scanning transmission electron microscope (STEM) for detailed examination of grain boundaries.

3. Experimental results

The experiments were designed to investigate the effect on superplastic ductility in 3Y-TZP of (i) prestraining to different prestrain levels and (ii) prestraining to 60% at different prestrain rates.

3.1. Effect of prestrain on elongation to failure

An initial set of 3Y-TZP specimens were given prestrains of 0, 60 and 130% at a prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-3} s^{-1}$ at 1673 K. Prestraining was followed by deformation to failure at a test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4} s^{-1}$ at the same temperature; true stress–true strain curves are shown in Fig. 1. It is seen that significant enhancements in elongation and slight decreases in flow stress were obtained for all prestrain conditions and that elongation increased with increasing prestrain. Notably, an elongation to failure of 485% after a prestrain of 130% was achieved compared to 283% in specimen not prestrained (at $1 \times 10^{-4} s^{-1}$ and 1673 K). Fig. 2 shows the profiles of specimens deformed under the above conditions; specimen A is untested and specimens B to D were pulled to failure at a test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4} s^{-1}$ after prestrains of 0, 60 and 130%, respectively, at a prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-1} s^{-1}$. The higher ductility is clearly visible in specimen D and there was no evidence of necking within the gauge length of the deformed specimens.

3.2. Effect of prestrain rate on elongation to failure

Superplastic ductility in 3Y-TZP, prestrained 60% at 1673 K, was evaluated in terms of prestrain rate ($\dot{\varepsilon}_1$) in the range 1×10^{-3} to $2.5 \times 10^{-4} s^{-1}$; these specimens were then pulled to failure at a test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4} s^{-1}$. Fig. 3 shows that an improvement in both superplastic ductility and a slight reduction in flow stress was observed for all prestrain rate conditions, compared to unprestrained specimen, e.g. an elongation to failure of 386% was achieved at a prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-3} s^{-1}$ followed by elongation to failure at $1 \times 10^{-4} s^{-1}$. The total elongation to failure was found to be essentially invariant with prestrain rate.

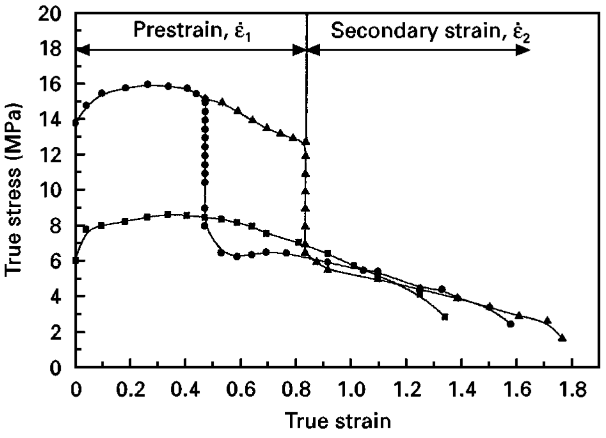

Figure 1 Effect of prestrain on elongation to failure at 1673 K. $\dot{\varepsilon}_1 = 1 \times 10^{-3} s^{-1}$, $\dot{\varepsilon}_2 = 1 \times 10^{-4} s^{-1}$. (■) prestrain 0.0, total elongation 283%; (●) prestrain 0.6, total elongation 386%; (▲) prestrain 1.3, total elongation 485%.

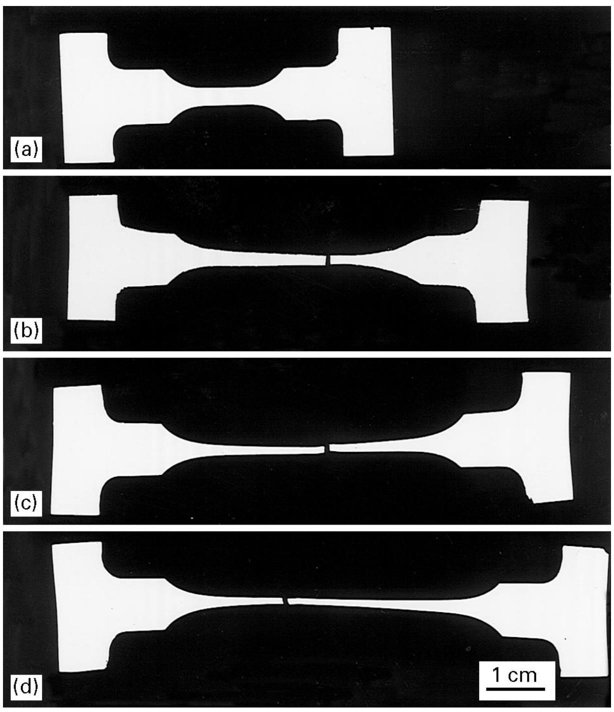

Figure 2 Profiles of 3Y-TZP specimens deformed at 1673 K: (a) undeformed specimen; (b) elongated to failure at $1 \times 10^{-4} s^{-1}$ without prestrain (elongation 283%); (c) elongated to prestrain of 60% at prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-3} s^{-1}$ followed by deformation to failure at test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4} s^{-1}$ (elongation 386%); and (d) elongated to prestrain of 130% at prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-3} s^{-1}$ followed by deformation to failure at test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4} s^{-1}$ (elongation 485%).

4. Discussion

Enhancement of superplastic ductility in 3Y-TZP has been considered to be due to a number of contributing

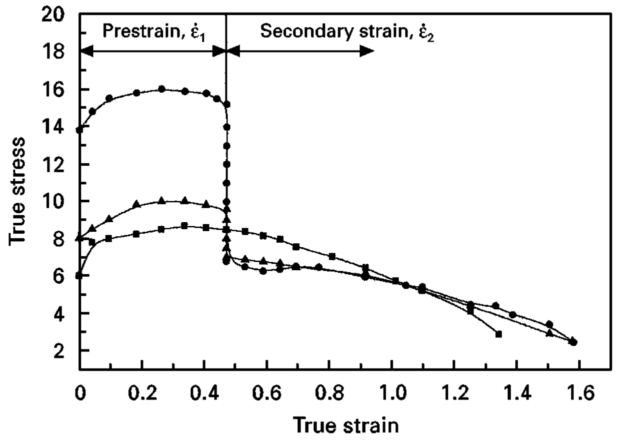

Figure 3 Effect of prestrain rate on elongation to failure at 1673 K. $\dot{\varepsilon}_2 = 1 \times 10^{-4}\,s^{-1}$, prestrain 60%. (■) Total elongation 283%; (●) $\dot{\varepsilon}_1 = 1 \times 10^{-3}\,s^{-1}$, total elongation 386%; (▲) $\dot{\varepsilon}_1 = 2.5 \times 10^{-4}\,s^{-1}$, total elongation 384%.

factors, including the presence of a low viscosity grain boundary (glassy) phase or doping with transition metal oxides. These additives possibly act in a multiple role as sintering aids, grain growth inhibitors and modifiers of grain boundary strength and grain boundary chemistry. Conventionally, reducing the strain rate or increasing the test temperature (but necessarily avoiding grain growth) also promotes larger elongations at lower stress levels. High deformation rates, high ductility and low forming temperatures are primary forming requirements for industrial applications of superplastic ceramics. In the present work, it has been demonstrated that in addition to improving superplastic ductility and reducing flow stresses, prestraining considerably shortened the time required to achieve a given elongation.

Historically, the effects of prestraining have been reported for metallic alloys including Pb–Sn alloys [16], Al–Cu–Zr [17] and Al–Li alloys [18]. With the exception of Pb–Sn alloys, where rapid prestraining reduced superplastic ductility, (because of strain inhomogeneities in the form of necking), elongation increased with prestraining. As shown by Geary *et al.* [19], improvement in superplastic ductility in dynamically recrystallized Supral 220 was the result of a decreased (recrystallized) grain size with increasing prestrain rate at the early stage of superplastic testing. Thus, an initial rapid strain rate, which led to a fine recrystallized grain size, was followed by a lower test strain rate that minimized cavitation and improved superplastic ductility. Amichi *et al.* [20] reported that superplasticity under prestrain conditions led to more stable superplastic deformation, as proposed by Hart [21]. Uniform deformation occurred up to a certain strain (defined by an instability parameter I); fast prestrain rates followed by slow test strain rates caused further ductility increase. In contrast to metallic alloys, deformation of 3Y-TZP did not show strain inhomogeneities in the form of necking or recrystallization in the early stages of deformation; the ductility enhancement cannot be due to these. To verify the latter claim, substantial TEM studies were carried out on unprestrained and prestrained specimens (prestrain range 0 to 130%, $\dot{\varepsilon}_1 = 1 \times 10^{-3}\,s^{-1}$ followed by a test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4}\,s^{-1}$). These TEM studies confirmed that prestraining did not cause recrystallization (grain refinement) at the early stage of deformation; grain interiors were also devoid of single or network dislocations, as shown in Fig. 4. Grain growth had occurred in the unprestrained specimen (from 0.2 μm to 0.76 μm). The effect of prestrain and prestrain rate can be seen in Figs 5 and 6, respectively; the differences in elongation are explicable in terms of grain size. Prestraining at the faster prestrain rates

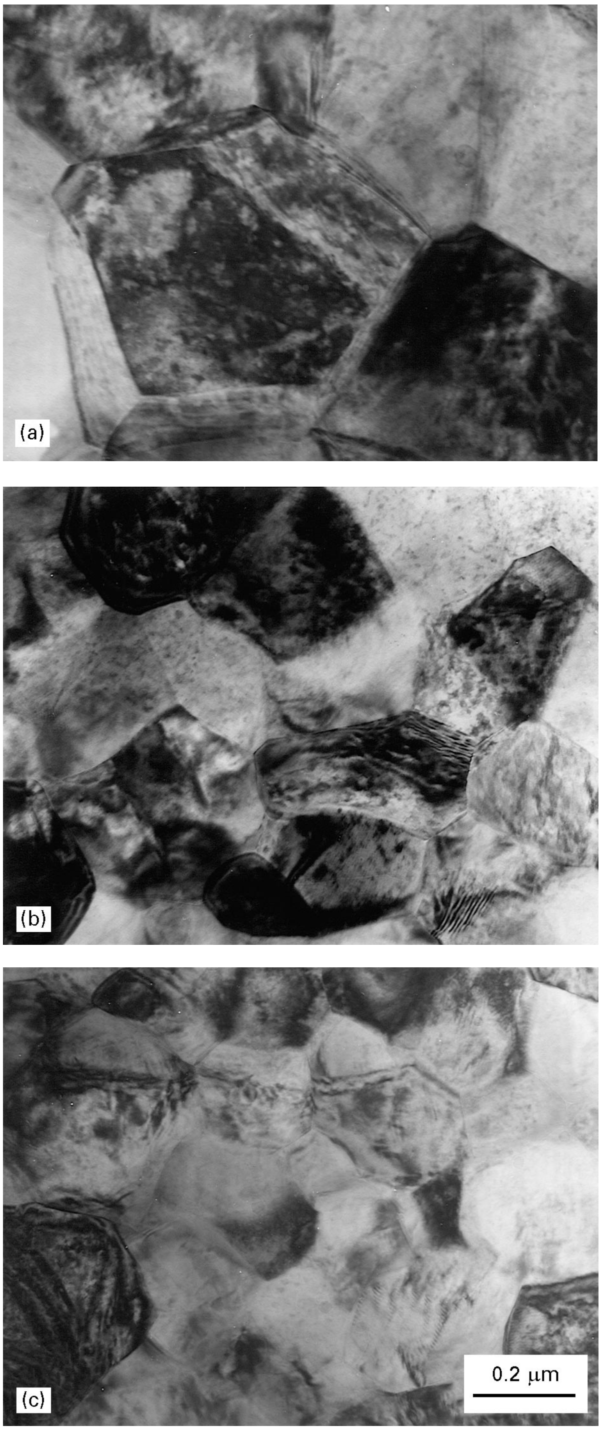

Figure 4 TEM micrographs showing the microstructure obtained after a prestrain of: (a) without prestrain; (b) 60%; and (c) 130% in specimens tested at prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-3}\,s^{-1}$ followed by elongation to failure at a test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4}\,s^{-1}$ at 1673 K. There was no evidence of recrystallization.

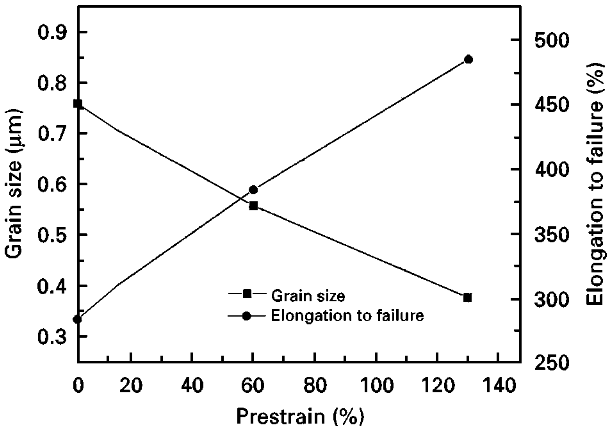

Figure 5 Grain size (■) after testing and elongation (●) of specimens tested to failure at 1673 K as a function of prestrain rate. $\dot{\varepsilon}_1 = 1 \times 10^{-3}\,s^{-1}$; test strain rate $\dot{\varepsilon}_2 = 1 \times 10^{-4}\,s^{-1}$.

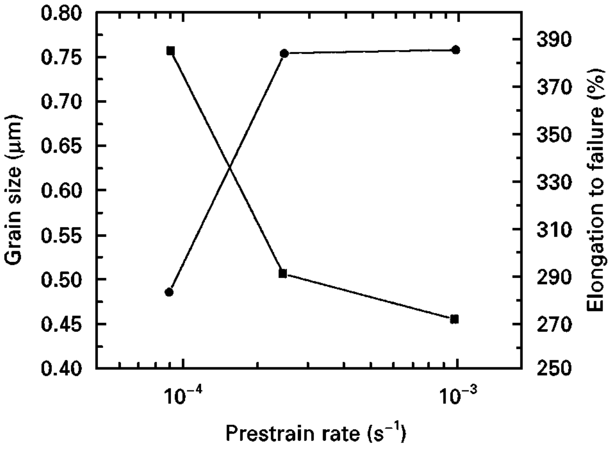

Figure 6 Grain size (■) after testing and elongation (●) of specimens tested to failure as a function of prestrain rate. $\dot{\varepsilon}_2 = 1 \times 10^{-4}\,s^{-1}$.

suppressed grain growth at the early stage of deformation (it is reported that in 3Y-TZP dynamic grain growth is severe at 1673 K and slow strain rates [22]). Increasing the amount of prestrain and increasing prestrain rates stabilized a small grain size that persisted during subsequent deformation at a lower test strain rate. This led to a significant increase in superplastic ductility i.e. 485% was obtained compared to 283% for unprestrained specimen as well as shortening the testing time for a given elongation.

The effect of prestraining on the superplastic deformation may also be expressed in terms of cavitation and the interaction between cavities and grain growth. In fine-grained polycrystalline materials with a narrow but measurable distribution of grain sizes, cavities are most likely to nucleate at triple point junctions of the occasional coarser grains due to incomplete accommodation processes; i.e. when the accommodation process is not rapid enough to meet the displacive requirements imposed by grain boundary sliding; local stresses, which develop at grain boundaries, are not relaxed sufficiently rapidly and grain boundary cavities are seen to nucleate preferentially near large grains. In the present study, grain growth (from ~0.2 to ~0.76 μm) occurred in unprestrained specimens and large cavities were associated with this grain growth; conversely, prestraining suppressed grain growth and a fine and stable grain size was established due to the formation of smaller equiaxed cavities uniformly distributed at triple points throughout the gauge length (the shape, size and distribution of these cavities is discussed elsewhere [23]).

5. Summary and conclusions

The influence of prestrain and prestrain rate on the superplastic deformation and testing time of 3Y-TZP was investigated. The results indicate that (i) prestraining 3Y-TZP at 1673 K at a prestrain rate of ($\dot{\varepsilon}_1$) $1 \times 10^{-3}\,s^{-1}$ followed by elongation to failure at the test strain rate of ($\dot{\varepsilon}_2$) $1 \times 10^{-4}\,s^{-1}$ significantly enhanced superplastic ductility and shortened the time for a given elongation. Elongations to failure of ~485% after a prestrain of 130% were achieved compared to ~283% without prestrain. (ii) Enhancements of ductility (around 390% compared to ~283% without prestrain) was also obtained in specimens prestrained 60% at various prestrain rates (where prestrain rates were faster than the subsequent test strain rate). The increases in ductility were attributed to suppression of grain growth at the early stage of deformation. It is proposed that the prestraining resulted in the formation of small, uniformly distributed cavities at grain boundary triple points; these small triple point cavities were a means of suppressing grain growth.

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