Influence of Flat Cavity Formation on Stress vs. Strain and Strain-Rate Relations of Superplastic Deformation in 3Y-TZP

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During superplastic deformation (SPD) of tetragonal zirconia polycrystals containing 3 mol% yttria (3Y-TZP) at high strain-rates, a number of crack-like flat cavities having very narrow gaps lying along grain boundaries mostly normal to the tensile axis are produced in addition to conventional cavities. In order to see when (from which strain point) the flat cavities get started to form, the tensile deformation of the 3Y-TZP was interrupted at different strain points including a yield point, and then a small angle neutron scattering (SANS) technique was applied to examine the existence of the flat cavities. It was found that no flat cavities were produced at the yield point but they got started to form soon after the yielding, i.e., after a superplastic flow began. The formation of the flat cavities can decrease the cross-sectional area of the specimen, resulting in a decrease in the flow stress apparently. It is therefore recommended that the formation of the flat cavities should be taken into consideration for an accurate evaluation of the flow stress in SPD at high strain-rate deformations.

1. Introduction

As well known, cavities can be formed and grown in polycrystalline materials subjected to superplastic deformations. In tetragonal zirconia polycrystals (Y-TZP) pulled under high strain-rate superplastic condition, the formation of fine and crack-like flat cavities having very narrow gaps lying in grain boundaries mostly normal to the tensile axis has been found by means of a small angle neutron scattering (SANS). The formation of these flat cavities seemed to be responsible for the strain softening that appeared on the true stress-strain curves. It has been found by means of SANS that strain point the flat cavities will be produced during the tensile deformations by means of SANS; the SANS technique is a quite effective method to identify the existence of flat cavities in a bulk specimen.

2. Experimental

2.1 Specimen preparation

Fine-grained 3Y-TZP specimens with an initial average grain size of 0.39 μm, produced by sintering powder of ZrO₂ containing 3 mol% Y₂O₃ in solid solution at 1773 K under atmospheric pressure, are prepared. The chemical composition of the 3Y-TZP is as follows: Y₂O₃ = 5.15, Al₂O₃ ≤ 0.10, SiO₂ ≤ 0.02, Fe₂O₃ ≤ 0.01, Na₂O ≤ 0.04, H₂ ≤ 1.7 and ZrO₂ = balance, in mass%.

Flat tensile specimens were made each with a gage length of 25 mm, 5 mm width, and 3 mm thickness. Tension tests at constant crosshead speeds were carried out at 1723 K at different initial strain-rates, &k, ranging from 1.67 × 10⁻⁴ to 8.0 × 10⁻³ s⁻¹ in air. Each specimen was heated to the test temperature by using a MoSi₂-element furnace. The temperature deviation was ±5 K. Each specimen was pulled to predetermined strains and then was air-cooled to room temperature. Microstructural evolution was observed with a scanning electron microscope (SEM).

2.2 SANS measurement procedure

Small-angle neutron scattering (SANS) experiments were carried out at two different instruments working in different resolution ranges. Low resolution measurements were obtained at the pin-hole diffractometer V4 installed at the research reactor of the Helmholtz Zentrum Berlin. Wave-length of incident neutrons was defined by velocity selector at the mean value of λ = 0.605 nm. Specimens were oriented with the applied strain axis parallel to the vertical component of momentum transfer vector, Qz, as illustrated in Fig. 1. Reduction of raw experimental data was done with the use of the BerSANS software package. It included corrections for background, transmission and normalization to absolute values of scattering cross-section using water as a calibration standard.

High resolution small-angle neutron scattering (HR-SANS) was measured at the double bent crystal (DBC-SANS) instrument DN-2 at the Nuclear Physics Institute in Rež near Prague (Czech Republic). Neutron wavelength at this instrument was set to λ = 0.201 nm by bent Si (111) crystal monochromator.

For both instruments, data analysis was carried out using the same fitting program, SASProFit. Evaluation of absolute values of pore surface and volume fractions requires...
the knowledge of scattering length density of the bulk material. We have used for this purpose the value of $5.54 \times 10^{-10}$ cm$^{-2}$ resulting from the table values of mass density and nuclear scattering lengths of constituting atoms.

3. Results

3.1 Results of tensile deformations

3.1.1 Stress vs. strain and stress vs. strain-rate relations

Figure 2 shows true stress, $\sigma_t$, versus true strain, $\varepsilon_t$, curves obtained for specimens pulled at different initial strain-rates, $\dot{\varepsilon}_0$, at 1723 K. The $\sigma_t$ and $\dot{\varepsilon}_t$ values were calculated from the conventional method. It is seen that gentle strain hardening arose on the curves when the specimens were pulled at low strain-rates ($\dot{\varepsilon}_0 \leq 5 \times 10^{-4}$ s$^{-1}$). This is caused by the deformation induced concurrent grain growth.$^{11,12}$ On the other hand, when pulled at high strain-rates, considerable strain softening was observed on the curves (see Fig. 2(a)). It is evident that this softening is not due to the formation of local necking but stems from another phenomena, because no obvious necking was identified on the specimens even after their fractures.$^{12}$

The softening must, therefore, be caused by either or both of the followings: (1) The gradual decrease in the actual cross-sectional area of the specimen, sustaining applied load, due to the formation and concurrent growth of cavities, in particular, flat cavities during the deformation as has been described in our previous papers, and (2) the decrease in a true strain-rate with the progress of the deformation, because the tensile tests are performed at constant crosshead speeds. It has been found$^{12}$ that even though the decrease in the true strain-rate with the deformation due to the constant pulling speed is taken into consideration, the strain softening appears on the stress vs. strain curves of the specimens deformed at high strain-rates.

Figure 3 shows the true stress vs. initial strain-rate relations obtained at different true strain points (see Fig. 2). In the Fig. 3, the yield stress, $\sigma_y$, is the true stress value taken at the maximum stress point when the $\sigma_t$-$\varepsilon_t$ curve showed strain softening, while when the $\sigma_t$-$\dot{\varepsilon}_t$ curve showed strain hardening, the $\sigma_y$ value is taken at a strain point from which steady superplastic flow began (see Fig. 2(b)).

It is evident that the shape of the $\ln \sigma_y$ vs. $\ln \dot{\varepsilon}_0$ relation, i.e., the slopes of the curves vary depending on which true strain point was used, at which the flow stresses were evaluated. It is also evident that the flow stress decreases as the deformation proceeds in a high strain-rate range ($\dot{\varepsilon}_0 \geq 6 \times 10^{-4}$ s$^{-1}$). This phenomenon is caused by the formation of cavities, in particular, that of flat cavities which decreases the actual cross-sectional area of the specimen sustaining the applied load.$^{12}$

It is quite interesting to note that when pulled at $\dot{\varepsilon}_0$ of $6.67 \times 10^{-3}$ s$^{-1}$, the flow stresses particularly the yield stress are seen to be relatively lower than the expected value from the extrapolation of the stress vs. strain-rate curve (see dotted curve in Fig. 3). This implies that the apparent yield stress appeared lower than the actual value because the flat cavities were already produced before the yielding, i.e., there is a possibility that the $\sigma_y$ appeared lower than the actual one. Another probable cause for this phenomenon may be a change in the deformation mechanism in a high strain-rate range. We have therefore conducted tensile tests in which the deformations are interrupted at different strain points including yield point: Detailed conditions for the tests are shown in Table 1. Then the SANS measurements were
performed on the deformed specimens to see whether the flat cavities had already been formed in these specimens.

### 3.1.2 Microstructures

Figure 4(a) and (b) are SEM images of the surface microstructures of specimens pulled to 30% true strain at 1723 K at 
$\varepsilon_0$ of $3.33 \times 10^{-3}$ s$^{-1}$ and $3.33 \times 10^{-3}$ s$^{-1}$, respectively. No flat cavities were seen in the both microstructures though the existence of flat cavities was already identified from the SANS results in the specimen deformed at $3.33 \times 10^{-3}$ s$^{-1}$. This is because the gap each flat cavity has is too small to be identified by SEM.\cite{9}

Table 1  Condition for tensile tests.

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Initial strain rate, $\varepsilon_0$/s</th>
<th>True strain, $\varepsilon_t$/%</th>
</tr>
</thead>
<tbody>
<tr>
<td>FC-01</td>
<td>$3.33 \times 10^{-3}$</td>
<td>30</td>
</tr>
<tr>
<td>FC-02</td>
<td>$3.33 \times 10^{-3}$</td>
<td>10</td>
</tr>
<tr>
<td>FC-03</td>
<td>$3.33 \times 10^{-3}$</td>
<td>5</td>
</tr>
<tr>
<td>FC-04</td>
<td>$3.33 \times 10^{-3}$</td>
<td>0 (yield point)</td>
</tr>
<tr>
<td>FC-05</td>
<td>$6.67 \times 10^{-3}$</td>
<td>10</td>
</tr>
<tr>
<td>FC-06</td>
<td>$6.67 \times 10^{-3}$</td>
<td>0 (yield point)</td>
</tr>
<tr>
<td>FC-07</td>
<td>$3.33 \times 10^{-4}$</td>
<td>0 (yield point)</td>
</tr>
</tbody>
</table>

Test temperature: 1723 K
Heating rate: 25 K/min
Holding time at test temperature: 10 min
Cooling rate: Furnace Cooling

![Fig. 4](image1.png)

Fig. 4  Surface microstructures of specimens pulled to 30% true strain at 1723 K at (a) $\varepsilon_0 = 3.33 \times 10^{-4}$ s$^{-1}$ and (b) $\varepsilon_0 = 3.33 \times 10^{-3}$ s$^{-1}$, respectively.

![Fig. 5](image2.png)

Fig. 5 Fracture surfaces of a specimen deformed to fracture at $\varepsilon_0$ of $3.33 \times 10^{-3}$ s$^{-1}$ at 1723 K. Photos (b) and (c) are enlarged ones corresponding to regions 1 and 2 indicated in the photo (a), respectively.

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Figure 5(a) to (c) show fracture surfaces of a specimen deformed to fracture at $3.33 \times 10^{-3}$ s$^{-1}$ at 1723 K. The fracture surface roughly consists of a fairly flat region and an extremely uneven region (see Fig. 5(a)). Figure 5(b) and (c) correspond to the former and the latter fracture surfaces, respectively. The latter case seems to be caused by a slow crack propagation. The fairly flat fracture surface (Fig. 5(b)) suggests that cracks propagated very rapidly. In the both cases, the fracture was caused by an intergranular one. It appears therefore that initial cracks were produced by the coalescence of the flat cavities: The formation of the flat cavities would therefore be a crucial factor to determine superplastic elongation of 3Y-TZP.\cite{9}
3.2 SANS results

3.2.1 Low-resolution SANS

SANS scattering patterns for the specimens deformed to the yield point ($\varepsilon = 0\%$) are fully isotropic as seen in Fig. 6. These data can therefore be fitted to a simple isotropic model randomly distributed spherical particles. The scattering from the other specimens deformed to above the yield point ($\varepsilon > 0\%$) are clearly anisotropic (see Fig. 7). To analyze these data, we have used the same model as in our previous work focused on 3Y-TZP deformed in high-stress regime.\(^{16}\) This model consisted of two sets of rotation ellipsoids, which are oblate and prolate, respectively, both be oriented with the axis of rotation parallel to the applied strain direction. The prolate ellipsoids should correspond to the cavities elongated parallel to the applied strain, which are commonly present in superplastically deformed 3Y-TZP.\(^{8,16}\) The oblate ellipsoids describe flat cavities often present in strongly deformed specimens and in those deformed in the high-stress regime. Size distributions of all particle types (both the ellipsoids and spheres) were represented by 10 splines fitted as free parameters to the experimental data.

Since the $|Q|$-dependence of the scattering functions obeyed the asymptotic Porod law in the whole range of accessible $Q$ values, only specific surfaces could be evaluated. Analysis of the pin-hole data did not prove the presence of small nanopores (smaller than about 100 nm) in any of the specimens.\(^5\)

3.2.2 High resolution SANS

Figure 8 shows scattering curves measured at the DBC SANS instruments for vertical specimen orientation. Due to the low luminosity of the double crystal diffractometer, only the data obtained on vertically oriented specimens can be evaluated and no information about cavity anisotropy is thus available in the high-resolution range. We can see that the scattering intensities for all $Q$ increase with raising $\varepsilon$ due to the growing volume fraction of cavities. In addition, this measurement (Fig. 8) permitted us to evaluate volume fractions and sizes of cavities measured along the $Q$ vector (i.e. perpendicular to the tensile axis).

In Fig. 9, the total volume fractions and specific surfaces as a function of the true strain are shown. These values are the sum for both the prolate and oblate ellipsoids. An example of size distribution of the cavities is shown in Fig. 8(b) for the FC-01 specimen ($\varepsilon = 30\%$). Maxima is located near $R \sim 0.1 \mu$m, where $R$ should be interpreted as one half of the mean dimension of the cavities measured perpendicularly to the incident beam.

4. Discussions

The present investigation has shown that (1) no cavities and fine cracks (flat cavities) except residual pores were detected at the yield point from which a superplastic flow began, and (2) it appears certain from the results that the flat cavities were formed in a very early stage of the superplastic
deformation (even at 5% elongation) when initial strain rate
was high ($\dot{\varepsilon}_0 > 3.33 \times 10^{-3} \text{s}^{-1}$), because (a) the scattering
pattern was changed to an anisotropic one that was oriented
vertically, i.e., parallel to the tensile axis and (b) the specific
surface and volume fraction of cavities were increased as the
strain was increased.

These results have clearly shown that the yield stress is
independent of the existence of cavities, but the flow stress
observed during the SPD under a relatively high strain-rate
condition would be affected by the formation of cavities,
especially, by flat cavities which would have decreased the
actual cross-sectional area of the specimen. The present
results indicate that we should take the above things into
consideration when we evaluate true stress vs. true strain and
true stress vs. true strain-rate relations obtained at relatively
high strain-rate deformations; for example, if we take a stress
at 10% true strain point as a flow stress, we should consider
that actual flow stress might be higher than the observed one.
That is, the effect of flat cavities should be taken into
consideration for the discussion of the mechanism of super-
plastic deformation.

Concerning the mechanism of the formation of flat
cavities, it has been proposed that a most probable cause
is a cleavage of grain boundaries existing among conven-

Fig. 7 2D SANS patterns with model (dashed lines) taken at SD = 16 m with $t = 0.605 \text{nm}$ for SPD specimens deformed at initial strain
rates of $3.33 \times 10^{-3} \text{s}^{-1}$ ((a), (b) and (c)) and $6.67 \times 10^{-3} \text{s}^{-1}$ (d) up to non-zero true strains.

Fig. 8 DBC-SANS data with fitted curves.
tional cavities generated at grain boundary triple-points caused by grain boundary sliding (GBS). This idea suggests that a critical strain point from which the flat cavities are produced would be a strain point after the onset of the superplastic flow which would mainly be due to GBS. The present experimental results support this deduction of the formation of the flat cavities, since no flat cavities were identified at the yield point where GBS got started but no conventional cavities would not be produced yet.

There have been different views on whether dislocation motion appears in SPD in 3Y-TZP or not. Munoz et al. have shown that a yield stress for a crystallographic slip in a tetragonal zirconia single crystal is quite high, about 300 to 250 MPa at 1673 to 1823 K. This result suggests that intragranular dislocation motion will not arise under the deformation stresses in the present study: It is seen in Fig. 3 that a σy value at εy of 6.67 × 10^{-3} s^{-1} was around 80 MPa, suggesting that dislocation motion should not occur.

On the other hand, Morita et al. have published a paper in which they have concluded that the dislocation motion appears in the SPD in 3Y-TZP from a fact that they have observed dislocation pile-ups to grain boundaries in a superplastically deformed 3Y-TZP. Our present results have shown dislocation pile-ups to grain boundaries in a tetragonal zirconia single crystal is quite high, about 300 to 250 MPa at 1673 to 1823 K. This result suggests that intragranular dislocation motion will not arise under the deformation stresses in the present study: It is seen in Fig. 3 that a σy value at εy of 6.67 × 10^{-3} s^{-1} was around 80 MPa, suggesting that dislocation motion should not occur.

Conclusions

Results obtained in this study are summarized as follows:

1) The SANS results have shown that no flat cavities exist at the yield point, but soon after the yielding from which superplastic flow begins, the formation of the flat cavities gets started in the specimens deformed at high strain-rates. It seems that the flat cavities are formed by a cleavage of grain boundaries existing among conventional cavities generated at grain boundary triple points induced by GBS after the SPD begins.

2) The yield stress, from its strain point of which a superplastic flow begins, should be taken as a flow stress for discussions of deformation mechanisms and so forth, since in the SPD at high strain rates, flat cavities get started to form soon after the yielding, e.g., even at 5% strain, which can decrease the actual cross-sectional area of the specimen and consequently the observed flow stress may be lower than the actual value.

3) It appears that the region III in which dislocation motion plays an important role exists in the stress vs. strain-rate relation of SPD in 3Y-TZP.

REFERENCES