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Superplasticity by internal frictional heat under biased cyclic loading

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Abstract A new damping method was developed not only as a testing tool to investigate in situ deformation under stress, but also as a processing method to superplastically deform ceramics. The specific damping capacity (SDC) at low frequencies (<0.2 Hz) decreased with increasing frequencies, which matched previous internal friction results. However, at higher frequencies (0.2–5 Hz) SDC increased with frequencies, which was explained by a new internal frictional heat mechanism. Three different ceramics: a non-superplastic one and two superplastic ones with different activation energies, showed the same behavior at the high frequency damping tests (1–5 Hz). From these results, it was deduced that a cyclic load at high frequencies, superimposed on a static one, has a great potential to enhance superplasticity by specifically heating up grain boundaries from internal frictional heat.

Introduction

Superplasticity is defined as an ability of a material to exhibit large deformation before failure and is a very promising near-net-shape forming process in ceramic industry [1]. Superplasticity of ceramics and ceramic composites has been widely studied in the last two decades [1–7]. It is generally agreed that grain boundary sliding is the main deformation mechanism, accommodated by thermally activated processes, such as diffusion and/or dislocation movements, etc. It is a challenge to activate large grain boundary sliding (the dominant mechanism in superplastic deformation) in ceramics at low temperatures. So far, two kinds of superplastic behavior [2] are established in polycrystalline solids: fine structure superplasticity (FSS) and internal-stress superplasticity (ISS). In FSS, normally a constant strain rate is applied at constant elevated temperatures and thus the sample with fine grains experiences a monotonically changing stress [1–5]. In ISS, a thermal cycling (typically about 0.01 Hz) [2, 6] in specific composites or single phase materials with anisotropic thermal expansion coefficients results in internal stresses, which can cause considerable deformation when a low external stress is applied. A high strain rate at low temperatures has to be realized for industrial application in near-net-shape forming of engineering materials. However, at least 1450 °C is necessary for medium strain rate (10^{-3} s^{-1}) superplastic deformation in ceramics [3–5]. Recently, an example of high strain rate superplasticity (10^{-1} s^{-1}) has been reported for a specific triphase ceramic (zirconia-alumina-spinel) at a rather high temperature 1650 °C [7].

Internal friction [8–13] is an important tool to study grain boundary sliding, yet so far usually it is only applied around zero stress. It aims at studying the grain boundary properties around zero stress, except in a torsion pendulum method, in which, however, the optional applied tensile stress is perpendicular to the cyclic torsional shear stress [13]. In addition, most of the internal friction studies [8–12] were focused in low-frequency (~0.01 Hz) Debye peaks [14], which are closely related with slow mechanisms such as

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diffusion-controlled grain boundary sliding, etc., and thus internal friction is only a testing tool, not practically a process for high strain rate deformation.

Here we developed a damping method, not only able to test in situ material deformation under a certain stress using a common MTS 810 loading frame, but also to be applied as a process to superplastically deform materials at lower temperatures, lower stresses and higher strain rates. A cyclic-compressive-compressive load superimposed on a static one was applied on two superplastic ceramics with different activation energies and an intrinsically non-superplastic ceramic at a low temperature (1300 °C).

Experiments

Materials

Three-mol% yttria stabilized zirconia ceramic was prepared from high-purity powder TZ3Y (Tosoh, Tokyo, Japan) by cold pressing (100 MPa) followed by sintering at 1450 °C in air for 3 h. The sintered compacts with a theoretical density of over 98% were obtained. The samples were then observed by an FEI XL30-SFEG high-resolution scanning electron microscope (SEM) (Philips, Germany) after polishing followed by thermal etching at 1400 °C for 30 min. A typical micrograph of the sintered TZ3Y samples showed an average grain size of about 0.30 μm, which was observed by other investigators.

Superplasticity tests

Dense TZ3Y samples were cut and polished to specimens of a size of $3 \times 3 \times 5$ mm. An MTS 810 controlled by LabView programs was the loading frame and an AST furnace was used to heat specimens to 1300 °C. Once the expected temperature was reached, compressive load was applied and the actual displacements and loads were recorded simultaneously with time. A jump test was performed on the TZ3Y compacts at strain rates from 10^{-4} s^{-1} to 10^{-2} s^{-1} at 1300 °C.

Damping tests

The expected load profile was designed as follows: for a specific load level, first a static compressive load F_0 is applied for 100 s, then 20 cycles of a sine wave compressive load $F_c = F_a \sin(2\pi ft)$ (F_a is amplitude, f is frequency in Hz, t is time in s.) were superimposed on the static load. When the sample was heated to 1300 °C, a package of frequencies from 0.05 Hz to 5 Hz with a step size 0.05 Hz was applied sequentially. After staying at a load level for 20 cycles of loading for each of the 100 frequencies, the specimen was loaded under a higher load level with the same sine wave package superimposed. Typically four levels of loads can be applied before a 0.5 true strain is reached. F_0 was chosen as 178, 297, 356, 445, 534 N (40, 60, 80, 100 lbs) and the amplitude was fixed at 89 N (10 lbs). The corresponding static compressive stresses were 20, 30, 40, and 50 MPa respectively and the amplitude stress was 10 MPa. In order to compare the results by our new method with previous internal friction tests, a sample was tested at 178 N (40 lb) level at two lower frequencies (0.01 and 0.0178 Hz).

Results and discussion

ZrO_2-3mol% Y_2O_3 (TZ3Y) with full density and an average grain size of about 0.3 μm, is the most studied superplastic ceramic [3, 5] with a strain rate sensitivity $m = 0.5$. The results are shown as solid squares in Fig. 1. It is clear that the flow stress-strain rate curve in a log-log scale (the filled squares) is a straight line and the slope is about 0.5, demonstrating superplastic behavior and pointing to grain boundary sliding mechanism. The purpose of the jump test is to verify if the stress-strain relation during cyclic loading would follow the same equation. From Fig. 1, it is clear that the relation between average stresses and average strain rates in damping tests (open circles) is the same as that in a conventional constant strain rate jump test

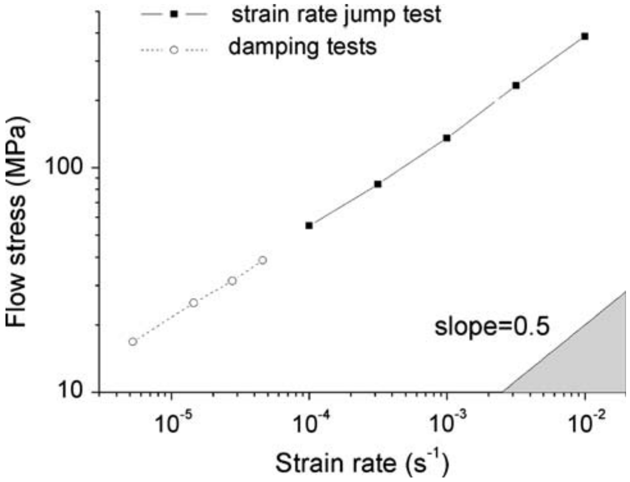

Fig. 1 Flow stress-strain rate relation for the constant strain rate jump test and damping tests at different static stress levels at 1300 °C (TZ3Y)

(solid squares) at 1300 °C. Thus grain boundary sliding is also the primary mechanism during the deformation by damping loads. SEM observations showed no grain growth or elongation at such a low deformation temperature. Also at such a submicron grain size, it is unlikely for intragranular defects such as dislocations and stack defaults to be activated at such a low temperature. More likely the grains remain rigid and grain boundary sliding with some accommodation processes is the main process in TZ3Y at 1300 °C. The static stress and damping stress amplitude in our test were low enough so that the accumulative strain is less than 0.5 after hundreds of cycles of loading.

The specific damping capacity (SDC) [15] is defined as the ratio of the dissipated energy ΔW to the maximum stored elastic energy W_{ealstic} per unit volume in a cycle: $SDC = \Delta W / W_{\mathrm{elastic}}$. It is related to$Q^{-1}$ (internal friction) by a factor of 2π in mechanical loss tests [8]. It is the ability of materials to absorb anelastic and plastic energy. The dependence of SDC on damping frequency at various static load levels is shown in Fig. 2. There is a minimal value of SDC at about 0.2–0.4 Hz for each static stress level. In case of the 20 MPa static stress, at low frequencies (<0.2 Hz), the mechanical loss in terms of SDC increased exponentially with decreasing frequencies, with a very weak Debye peaks (~0.04 Hz) superimposed. The mechanical loss at low frequencies in our tests is widely observed in previous internal friction tests for various ceramics [8–12, 15–18]. Lakki et al. [16] developed a model and interpreted the background as disappearance of the restoring force at the triple junctions. Daraktchiev et al. [17] attributed the exponential background to the sliding of large grains in TZ3Y. However at frequencies greater than 0.2 Hz, SDC increased sharply as the frequencies increased. This result is totally different from that of previous internal friction tests [16, 17], in which the internal friction decreased exponentially with increasing frequency, approaching zero asymptotically.

Here a model based on frictional heat generation and dissipation mechanism is adopted to explain the difference. Friction occurs whenever two solid bodies slide against each other. It is believed that nearly all energy dissipated is transformed into heat and thus increases the temperatures of the contacting areas [19]. The heat generation rate q in sliding friction is related with the friction coefficient μ, the contact pressure P and the relative velocity v as in $q = \mu P v$ [19]. In our tests, the displacement amplitude remained constant under the damping loads at various frequencies, thus the sliding velocity is proportional to frequency. Hence, the heat generation rate in our test should be proportional to frequency. This is confirmed by our experimental results.

From our cyclic loading tests, the energy loss rate P_{loss} is calculated from $P_{\mathrm{loss}} = \Delta W f$, which f is the damping frequency. The dependence of energy loss rate P_{loss} on frequency f is shown in Fig. 3. At higher frequencies, the relative sliding was faster and so was the energy loss rate on friction. The frictional heat is not easy to dissipate through the grains because of the low thermal conductivity of TZ3Y. As a consequence, the temperature of the contacting region-grain boundary would increase. In a mechanical loss test with zero static stress, normally the stress is low and the heating rate is low enough so as not to incur much temperature change due to the frictional heat. However, when the

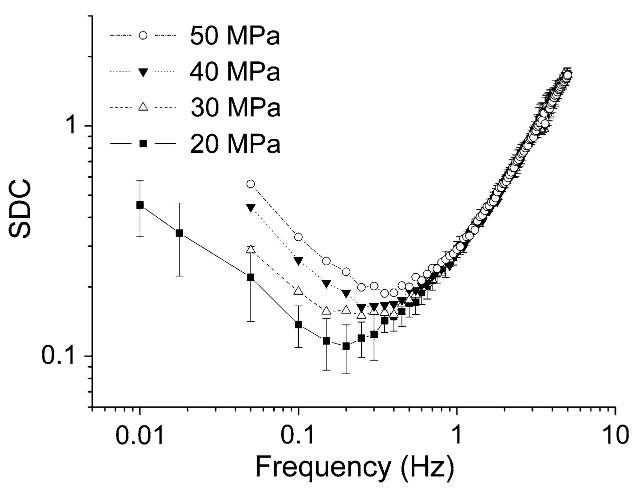

Fig. 2 Dependence of SDC on damping frequency at static stresses of 20, 30, 40 and 50 MPa and at 1300 °C for TZ3Y

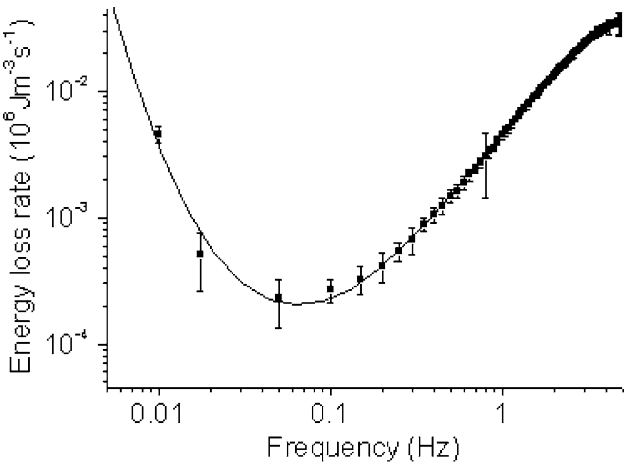

Fig. 3 The energy loss rate dependence on frequency of TZ3Y (at 1300 °C and 20 MPa static stress) (Error bars are determined from standard deviation of the 20 cycles at each frequency)

static stress is not zero, the friction between grains increases, supposing the friction coefficient does not decrease significantly. With an increased friction force and frequency, the energy loss rate increases over the energy dissipation rate and can locally increase the temperature at the grain boundaries and decrease the apparent viscosity of the grain boundaries. Thus the material can absorb more energy and has a high SDC value at higher frequencies under stress.

It seems that different mechanisms were dominant during deformation at the two different frequency minimum. In Fig. 4, a mechanism map describes the dominant mechanisms at low and high frequencies. At lower frequencies, slow mechanisms controlled by diffusion play an important role. The long period allows time dependent process of diffusion to take place. The heating rate is low and the energy dissipation is consumed by the diffusive movement of atoms or dissipated through the grains. With increasing frequency and thus with shorter period, diffusional effects diminish rapidly. In the high frequency range, local friction heating and melting becomes dominant because increase in the friction-heating rate with frequency is faster than that in the heat dissipation rate. Hence a minimum appears at certain frequency when there is a transition in the dominant mechanism.

The temperature increase over 100 °C on specimen surface by internal friction heat has been observed for ceramic composites in room-temperature fatigue tests [20–21]. The internal temperature rise should be higher in ceramics because of low thermal conductivities of most ceramics. For TZ3Y, at 20 MPa static stress level with a 10 MPa stress amplitude, the temperature at grain boundaries could increase about 50 and 71 °C at 1 and 2 Hz, respectively (see Appendix I). If heat dissipation through grains were considered, the temperature increase would be reduced. However, at a higher frequency (over 10 Hz), internal frictional heat would make a much-pronounced temperature increase at grain boundaries considering a faster heat generation rate and the low thermal conductivity. Frictional heating serves as continuous heat sources locally at grain boundaries and thus increases the thermally activated processes: diffusion and/or viscous flow. The overall high temperature of the matrix is not a necessity for grain boundary sliding; a localized temperature increase at grain boundary by internal friction is a promising way to lower the external temperature for manifestation of superplasticity.

Similar damping behavior was also found for two other ceramic composites: zirconia-magnesia alumina spinel (ZS) [22] (processed from nanopowder mixtures of TZ3Y, magnesia and alumina) and zirconia-alumina- magnesia alumina spinel (ZAM) [23] (processed from plasma sprayed powder). ZS is superplastic from 1300 °C to 1500 °C with an activation energy 522 kJ/mol (concurrent grain growth is neglected here) and ZAM in this study is not superplastic because of the low-angle grain boundaries by nucleation and growth of α alumina and spinel from metastable tetragonal zirconia phase formed during plasma melting and quenching. Wakai et al. [3] determined the activation energy for TZ3Y to be about 590 kJ/mol (concurrent grain growth is also neglected). From Fig. 5 it is clear that the deformation behavior at constant strain rates can be predicted by extrapolating SDC vs. frequency curve to near 0 Hz (say 0.001 Hz). Highly superplastic materials (low activation energy) also showed high SDC values and a low SDC at low frequency is related to non-superplastic behavior near 0 Hz. More importantly, Fig. 5 also reveals the similarity of all three different ceramics at high frequencies in terms of SDC.

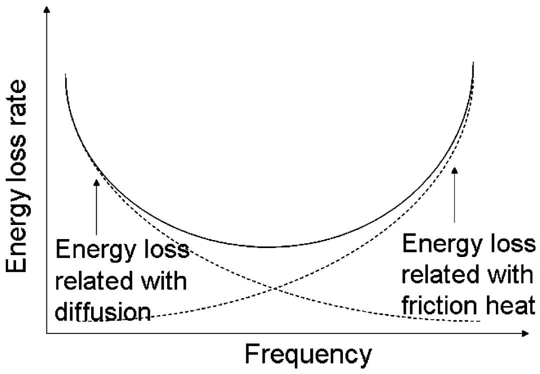

Fig. 4 Mechanism map for biased cyclic loading at low frequencies and at high frequencies

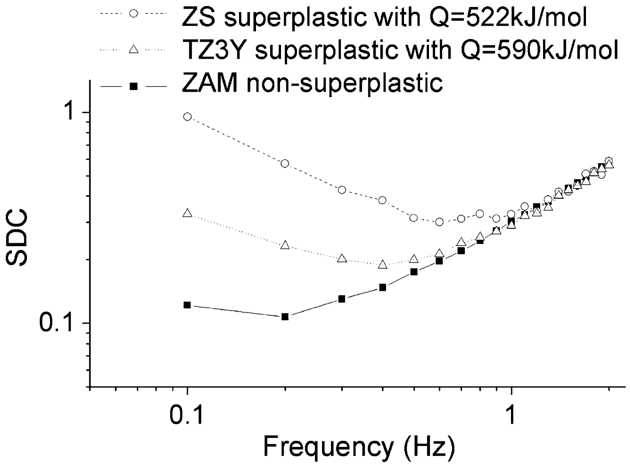

Fig. 5 SDC dependence on frequency for zirconia-spinel [22], TZ3Y and zirconia-alumina-spinel [23] at 30 MPa static stress and 1300 °C

If we extrapolate the SDC curve to high frequencies (tens or even hundreds in Hz), all three ceramics may exhibit the same high SDC values, which means we could superplastically deform even ‘‘conventionally non-superplastic’’ materials. A start-up heating may be necessary for the grain boundary to begin sliding; afterwards external heating can be reduced while internal frictional heat can continue promoting desirable grain boundary sliding under the biased cyclic load.

Similar interesting phenomena were found in Figs. 2 and 5: with increasing frequencies, SDC values seemed to be independent on external pressures and materials. In our discussion above, the friction coefficient μ was considered as a constant independent of material and temperature. However, Senda et al. [24] found that in the alumina system, the friction coefficient actually decreased with increasing temperatures. Computer simulation results by Popov [25] also showed that the friction coefficient was a constant at low temperatures but was reduced at elevated temperatures. As the frequency increased, according to $q = \mu P v$ [19], the heat generation rate increased. At low frequencies, the heat generation rate is low and the temperature increase is insignificant and the friction coefficient can be treated as a constant, thus SDC is dependent on pressures and materials. If the temperature increase is significant as in the high frequencies range, the drop of the friction coefficient might compromise the increase of the heat rate caused by increased external pressures, thus SDC curves with different pressures converged. In a similar way, the friction coefficients of the three ceramic materials may show little difference at high temperatures, but depend more on sliding velocities or cycling frequencies. Further studies are needed to clarify the phenomenon. Most importantly, the evidence has shown the possibility to superplastically deform various materials by this special cyclic loading.

Conclusions

The present results show that in situ deformation behavior can be studied by a biased compressive damping load using common mechanical testing equipment. More importantly, it is possible to take advantage of internal frictional heat to locally increase temperatures of grain boundaries by a biased cyclic load at a high frequency. If our conjecture turns out to be true, superplasticity can be enhanced in a wide range of ceramics (including traditionally non-superplastic ones) at comparatively lower deformation temperatures. Although damping tests at higher frequencies are yet necessary to further prove the hypothesis, this paper provides a new way to superplastically shape form ceramic materials at low temperatures.

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Appendix I Estimation of temperature increase at grain boundaries under biased cyclic loading

Temperature increase by internal friction heat can be estimated from tribology theories. Suppose the tribology theory remains valid in the microscopic scale (sub micrometer), and the temperature increase ΔT [19] is:

$$\Delta T = \frac{2\mu P v l}{k\sqrt{\pi\left(1.011 + \frac{v l \rho C_p}{2k}\right)}}$$

in which, the heating rate μ is the friction coefficient, P is the normal stress, v is the relative velocity, l is width of the sliding band, k is thermal conductivity, ρ is the density, and C_p is the specific heat capacity. In our tests,

$$\mu = 0.2, P = 20\ \text{MPa}, \rho = 6050\ \text{kgm}^{-3},$$
$$C_p = 400\ \text{Jkg}^{-1}\ \text{K}^{-1}, k = 2\ \text{Wm}^{-1}\ \text{K}^{-1}$$

For TZ3Y specimens in our test, the average grain size is about 300 nm, the sample size is $3 \times 3 \times 5$ mm, $l = 300\ \text{nm} \times \frac{3\,\text{mm} \times 3\,\text{mm}}{300\,\text{nm} \times 300\,\text{nm}} = 30\,\text{m}$ Supposing the displacement is caused by uniform grain boundary sliding, for a 10 MPa as stress amplitude and 20 MPa as static stress, the relative velocity was determined to be $v = 1.0 \times 10^{-5}\ \text{ms}^{-1}$ from the actual displacement amplitude at $f = 1$ Hz and $v = 2.0 \times 10^{-5}\ \text{ms}^{-1}$ at $f = 2$ Hz, we have $\Delta T = 50$ °C for $f = 1$ Hz and similarly, we have $\Delta T = 71$ °C for $f = 2$ Hz.

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