Grain-Size Sensitive Viscoelastic Relaxation of High-Purity MgO

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High-temperature mechanical spectroscopy experiments have been performed on newly prepared polycrystalline MgO samples with average grain sizes of 3, 9 and 100 μm. Viscoelastic relaxation is strongly grain size dependent above temperatures of 800°C with a frequency and temperature-dependent 'background' consisting of a broad anelastic relaxation band intensified by the onset of viscous deformation. Data analysis using a master variable approach suggests that grain boundary sliding processes dominate viscoelastic relaxation.

1. Introduction

The high-temperature breakdown of strictly elastic behaviour in fine-grained materials is usually attributed to grain-boundary sliding facilitated by the low effective viscosity of the thin grain-boundary region or to diffusional mechanisms. Both grain boundary sliding and diffusion are grain size dependent deformation mechanisms. This study aims to determine which viscoelastic relaxation mechanism is responsible for the viscoelastic relaxation in high-purity ceramic MgO material. The new data will ultimately contribute to constructing improved micro-mechanical models for grain boundary sliding [1,2].

2. Sample preparation

For fabrication of the samples, high-purity (> 99.99 wt.%) MgO nanopowder with a grain size of 45-60 nm (supplied by Ube Materials Industries Ltd.) has been used. Samples with low porosity (ϕ < 1%) and controlled grain sizes have been obtained by a multi-stage fabrication process: (1) Cold isostatic pressing at 200 MPa for 0.5 hour to yield samples of ~50% porosity; (2) Pressureless sintering at 900-1100°C for 2 hours in an N_2-rich environment to evaporate residual CO_2 and H_2O followed by slow cooling at 2 °C/min to prevent sample cracking (ϕ~40%); (3) Hot-isostatic pressing of the sample in a steel jacket in a gas-medium apparatus at 300 MPa argon confining pressure at elevated temperatures. For the finest grained sample (1-3 μm). Hot-pressing for 24 hours at 1100°C resulted in minor further densification of the sample (ϕ ~1%) and minor grain growth to an average grain size of 1-3 μm. The second sample has been made using two-stage hot-pressing; first at 1100°C for 24 hours for densification followed by 1300 °C for 24 hours to allow grain growth to 5 μm. The third sample had to be hot-isostatically pressed at a confining pressure of 30 MPa and a temperature of 1600°C for 2 hours in facilities at Okayama University in Japan to obtain a sample with a much larger grain size of 100 μm.

Torsional forced-oscillation experiments[3] were conducted at 200 MPa confining pressure over the temperature range 20-1300°C. Sinusoidally varying stresses were applied at 10 different periods between 1s and 1000s at torque amplitudes equivalent to maximum shear stresses of 0.3 MPa, resulting in maximum shear strains of 3.10^{-5} at the highest temperature. Before performing routine torsional forced-oscillation measurements, the assemblies were thoroughly annealed at the highest temperature and the mechanical behaviour was monitored. Small changes in shear modulus G and strain-energy dissipation Q^{-1} with time at the

beginning of the experiment reflect minor microstructural changes (grain size increase from 1 to 3 and 5 to 9 µm, Fig.1). After stabilization of the microstructure, torsional forced-oscillation experiments were performed over the complete range of oscillation periods in temperature steps of 50°C, starting at the highest temperature. Mechanical data in the form of shear modulus and dissipation were determined for each oscillation period at each temperature.

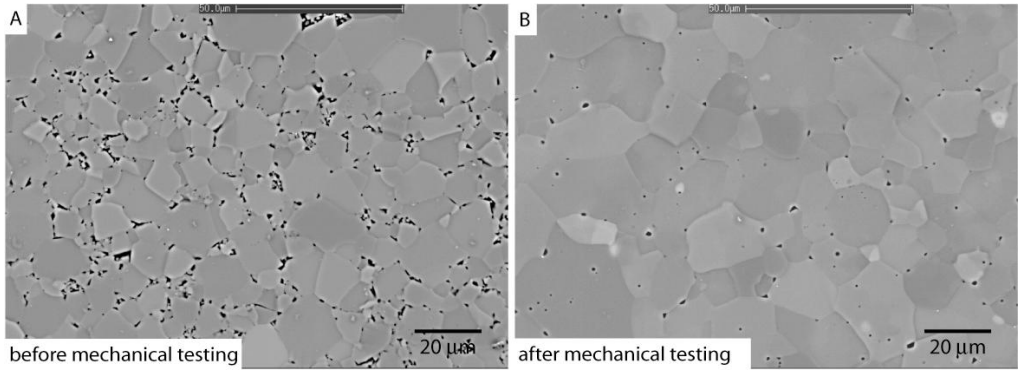

Fig. 1. Backscattered electron image of (a) hot-isostatically pressed sample (total 48 hours at high-temperature) and (b) the same sample after further annealing and mechanical testing involving several days at temperatures > 1000°C resulting in slow continuous grain growth and reduction of porosity.

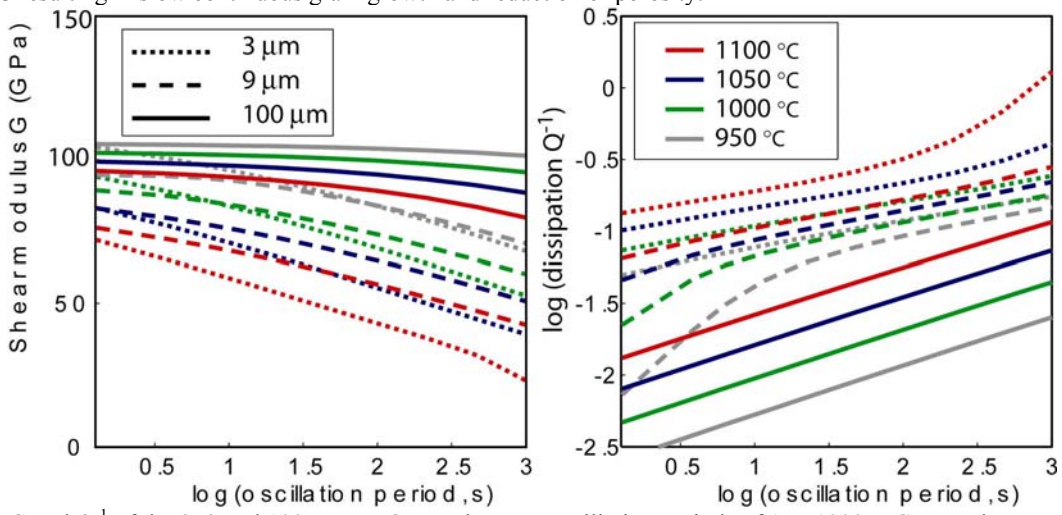

Fig. 2: G and Q^{-1} of the 3, 9 and 100 µm MgO samples over oscillation periods of 1 to 1000 s. Curves shown are least-square fits to an extended Burgers model[4,5]. Line colour and pattern indicate temperature and grain size, respectively, in both diagrams.

3. Results

All three MgO samples with different grain sizes deform essentially elastically at temperatures ≤800°C. At higher temperatures, viscoelastic relaxation occurs with the strongest degree of viscoelastic deformation at the highest temperature, longest oscillation period and smallest grain size (Fig. 2). Maximum reduction in G between 1 and 1000 s at 1300°C is 70%. The systematic monotonic variations in G and Q^{-1} exclude the presence of any localised dissipation peak of significant height on top of the frequency-dependent dissipation background. G and Q^{-1} data for each individual sample were modelled using an extended Burgers model[4,5] describing the full viscoelastic relaxation of MgO resulting in generally small misfit of the experimental data.

Analyses of the Q^{-1} data only using a master variable approach[6] $X \sim \omega d^m \exp(E/RT)$, where E and R are the activation energy and the gas constant. This approach shows that the collapse of all three datasets is best when a linear grain size dependence (m=1) is included in

the approach. When using on optimal activation energy of 330 kJ/mol, both individual datasets and combined datasets collapse on a linear trend. A grain size sensitivity of 3 (m = 3) does not result in a good collapse on a linear trend irrespective of the value for *E*. Only individual datasets collapse on a linear trend. This indicates that the dissipation data of MgO is best described by a grain size dependence of 1, meaning that relaxation mechanisms with linear grain size dependence dominate over relaxation mechanisms with a grain size dependence of 3. We therefore conclude that grain boundary sliding (m=1) dominates over diffusion processes (m=3) in high-purity MgO[1,7].

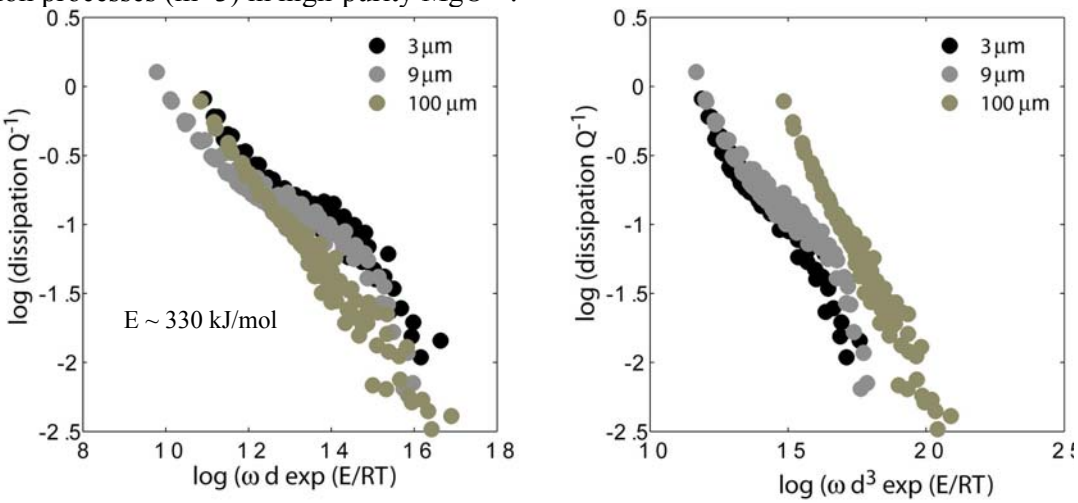

Fig. 3. Master variable approach of the dissipation data[6] $X \sim \omega d^m \exp(E/RT)$, where m=1,3. Collapse of the data with m=1 is narrower than for m=3, indicating grain boundary sliding mechanisms (m=1) dominate over diffusion mechanisms (m=3).

4. Conclusions

- Systematic grain size dependence of viscoelastic relaxation in MgO.
- Grain boundary sliding processes dominate over diffusion processes in high-purity MgO.

Acknowledgments

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