PLASTIC DEFORMATION IN
OXIDE CERAMICS

MASTER

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Abstract

Research on plastic deformation in oxide ceramics has been undertaken. Specifically, the effects of non-stoichiometry on slip on the "hard" slip systems in Al-rich Mg-Al spinel and oxygen-rich UO_{2+x} single crystals is under investigation. In the case of spinel, dislocation structures in both stoichiometric and alumina-rich specimens deformed in compression at elevated temperatures (1500 to 1900°C) have been studied using transmission electron microscopy. Unusual dislocation structures are observed after large plastic strains, which are attributed to extensive climb and unique node reactions are observed which are the result of climb dissociation. This climb dissociation is attributed to the presence of large concentrations of vacancies produced by non-conservative dislocation motion.

The research UO_{2+x} to date has been primarily concerned with construction of a new high temperature compression testing device useable over the temperature range 600-1600°C. This apparatus permits control and measurement of oxygen partial pressures; precisely metered Co/CO_2 gas flows can be maintained, and the $p\text{O}_2$ measured using a stabilized zirconia oxygen sensor, which is incorporated into the compression apparatus.

I. INTRODUCTION

This document constitutes the first progress report on research on "Plastic Deformation Oxide Ceramics," which has been underway at CWRU under ERDA support since January 1, 1977. The main goals of our work have been to study the effects of non-stoichiometry on plastic deformation and the kinetics of dislocation annihilation in oxygen-rich UO_{2+x} and Al-rich Mg-Al spinels ($\text{MgO} \cdot n\text{Al}_2\text{O}_3, n > 1$).

The rationale for the choice of research topics and the research methodology was specified in detail in our original proposal and will only be stated briefly here. In the case of spinel, we wish to study $\{111\} \langle 110 \rangle$ slip in crystals of differing composition at elevated temperatures; for the Al-rich crystals, this constitutes a hard slip system, as non-stoichiometric crystals preferentially deform by $\{110\} \langle 110 \rangle$ slip. Fortunately, this latter slip system can be completely suppressed if crystals are compressed parallel to $\langle 111 \rangle$, and results of the initial experiments of this type are described in Section II. The preliminary studies on the kinetics of dislocation annihilation in spinel are also described in this Section.

In the case of UO_{2+x} , we again wish to study slip on the hard systems as a function of departure from stoichiometry; in this case, however, it is not known if $\{110\} \langle 110 \rangle$ or $\{111\} \langle 110 \rangle$ is the most important secondary slip system in UO_2 (there is agreement that $\{100\} \langle 110 \rangle$ is the easy slip system). Our initial goal is to determine whether $\{110\}$ or $\{111\}$ is the predominant slip planes when $\{100\}$ slip is suppressed, and whether the preferred secondary slip planes

changes with composition, as occurs for the easy slip system in spinel. Our strategy for resolving this point is discussed in Section III, which also describes the apparatus we have constructed to permit compressive deformation of UO_2 single crystals at elevated temperatures in controlled $p\text{O}_2$ (departures from stoichiometry can be precisely controlled by control of the $p\text{O}_2$), apparatus heretofore unavailable at CWRU.

II. SPINEL

Compression of spinel single crystals along $\langle 111 \rangle$ completely suppresses $\{110\} \langle 110 \rangle$ slip; thus, deforming crystals along this orientation readily permits an assessment of the effect of non-stoichiometry on plastic deformation occurring by $\{111\} \langle 110 \rangle$ slip. A typical resolved shear stress-resolved shear strain curve* for a stoichiometric crystal deformed along this direction is shown in curve c, Fig. 2.1; the noteworthy feature of the high temperature plastic flow is the "work-softening" that occurs after plastic strains of ~5% and persists until a strain of ~15%, whereupon a steady state region of zero work hardening commences. Since such work-softening had been observed in prior work in our laboratory in stoichiometric spinel(1) oriented for both single and duplex slip (curves a and b, Fig. 2.1), and since the resolved flow stresses appear to be independent of whether one, two or six slip systems of the $\{111\} \langle 110 \rangle$ family are activated, it was deemed

*Note that in this orientation, six systems of the $\{111\} \langle 110 \rangle$ family are equally stressed.

Figure 2.1 - The effect of specimen orientation on the resolved shear-stress vs. shear strain curves.

necessary to study the evolution of the dislocation substructure in detail before investigating the composition dependence of $\{111\} \langle 110 \rangle$ slip.

For this purpose, crystals oriented for single slip were deformed at 1800°C to various strains (indicated by the vertical arrows in curve a, Fig. 2.1) and the dislocation structures characterized by TEM.

Figure 2.2a shows that at small strains, long straight edge dislocations belonging to the primary $\{111\} \langle 110 \rangle$ system predominate, indicating that even at these very high temperatures, conventional dislocation glide is an important process, and that the Peierl's force appears to still control the dislocation configuration. As work-hardening proceeds and just prior to the onset of work-softening, a large increase in dislocation density is observed, resulting in a random three-dimensional dislocation network (Fig. 2.2b). The occurrence of work-softening coincides with a large decrease in the dislocation density, i.e. an increase in the network spacing, as shown in Fig. 2.2c.

The coarsening of the network is clearly associated with increasing amounts of dislocation climb; it appears that deformation by climb becomes increasingly important at large strains because of the difficulty of the (presumably) heavily-jogged dislocations gliding through the "forest" structures typified by Fig. 2.2b. The network dislocations in Fig. 2.2c are also widely dissociated; as will be shown below, we believe that this dissociation occurs by climb rather than by glide

a

b

c

d

Figure 2.2 - Dislocation structure on the primary slip plane in stoichiometric spinel deformed at 1800°C. a,b,c shows the structures at 2%, 3% and 10% shear strain respectively; d, shows the sample of Fig 2.2c after 15 minutes annealing at 1800°C.

and in the stoichiometric crystals shown in Fig. 2.2c, results from a supersaturation of vacancies produced by non-conservative deformation. This latter point is strongly supported by the first experiment performed on the kinetics of dislocation annihilation in deformed spinel. Fig. 2.2d shows the dislocation structure in the same specimen as Fig. 2.2c after a 15 minute anneal at 1800°C. The most striking effect of this heat treatment is the reduction in the width of the dislocation ribbons, indicating that the wide separation of the partials shown in Fig. 2.2c cannot be an equilibrium structure.

Deformation by pure climb has been modeled by Nabarro⁽²⁾; calculated strain rates using Nabarro's model and the diffusion data of Ando and Oishi⁽³⁾ gave order-of-magnitude agreement with the experimental data shown in Fig. 2.1 and preliminary values for the stress exponent n for the steady state flow stress is also in agreement with Nabarro's prediction of $n=3$. The occurrence of extensive climb during high temperature deformation may also explain the lack of an orientation dependence of the stress-strain curves shown in Fig. 2.1.

The first experiments performed on a non-stoichiometric crystal ($\text{MgO} \cdot 3.5\text{Al}_2\text{O}_3$) have confirmed that climb is important in these crystals as well. Fig. 2.3a shows the dislocation structure in a crystal deformed to ~15% plastic strain by compression along $\langle 111 \rangle$ at 1500°C; the three-dimensional network is strikingly similar to that shown in Fig. 2.2c for the stoichiometric crystals, except that the dislocation ribbons are even wider. The large separation of the partials allowed us to perform a high angle tilting experiment, and thus to determine the planes of the stacking faults. By observing the same dislocation

a

b

c

d

Figure 2.3 - a) Dislocation structure in non-stoichiometric (MgO:3.5Al₂O₃) spinel deformed 16% at 1500°C along $\langle 111 \rangle$. The tilting experiment (b-d) of a dissociated dislocation using the sample of Fig. 2.3a permitted identification of the plane of the stacking faults, as discussed in the text.

node through large angles of tilt in the electron microscope (Fig. 2.3b, c and d) the fault planes for the three dislocations comprising the node were found to be different; two of the stacking faults lay on irrational planes near {001} type planes, while the third fault was on a {110} type plane. Since the Burger's vectors of the dislocations do not lie in the plane of the stacking faults, the faults themselves must have been created by climb.

III. UO₂

For unambiguous research on the effects of non-stoichiometry on plastic deformation in UO₂ to be performed, it is mandatory to control the pO₂ precisely and no equipment was available in our laboratory which could be operated under these conditions (or conveniently adapted to allow deformation to proceed under a given gaseous ambient). It was therefore necessary to design and build a suitable compression apparatus, which is shown schematically in Fig. 3.1. The specimen can be deformed between two alumina rods, attached to the testing machine, while a precisely metered mixture of CO/CO₂ is passed through the furnace. An interesting feature of this design is the silicon oil/air seal at the lower end of the alumina tube. A sufficient head of oil is maintained to keep the CO/CO₂ mixture from leaking, and the important feature is that this arrangement will not falsify load measurements as do conventional sealing techniques, e.g. "O" rings or bellows.

Oxygen partial pressures during testing will be measured using yttria-stabilized zirconia tubes. The partial pressure will be

Fig. 3.1 - Schematic of compressi
appartus for use in
controlled oxygen
partial pressure.

measured both directly near the sample and in the effluent gases, which will provide for good control of stoichiometry during each experiment.

Because of the need for a short load train, it was also necessary to build a furnace capable of temperatures between 600 and 1600°C with the smallest possible vertical dimensions. A small "clamshell" type furnace with zirconia and alumina fiberboard insulation and "Super-Kanthal" elements has been built and adapted to ride on the crosshead of the testing machine.

While it is possible to suppress {100} slip completely by compressing samples along [100], this orientation allows a plethora of {110} and {111} samples to be highly stressed. We have chosen an alternative scheme in which samples are oriented such that two specific slip systems of each "hard" slip system e.g. {111} $\langle 110 \rangle$ and {110} $\langle 110 \rangle$ have the same maximum Schmidt factors (0.45), while the Schmidt factor for {100} slip is at most 0.23. This orientation of the compression axis lies about 15° off [001] along a line from [001] to $[\bar{1}11]$.

At present, two single crystal boules, approximately 1.5" long x 0.4" diameter, are being used to fabricate test specimens, and a larger boule, approximately 3" long x 1.5" diameter, is on order from ONC Co., Atlanta, Georgia.

IV. Ancillary Information

The Principal Investigator, Professor A.H. Heuer, has devoted approximately 15% of his time to date on this research and will continue to devote this level of effort on the project for the remainder of the contract period. In addition, he presented a paper, based on research sponsored under this contract, at the 5th International Conference on High Voltage Electron Microscopy, Kyoto, Japan, which was held from August 29 to September 1, 1977. A copy of the conference report, which will be published in the near future, is included as Appendix A of this Progress Report.

Removed

V. References

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