



Contents lists available at ScienceDirect

Physics of the Earth and Planetary Interiors

journal homepage: www.elsevier.com/locate/pepi

Short communication

The influence of anisotropic diffusion on the high-temperature creep of a polycrystalline aggregate

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ARTICLE INFO

Article history:

Received 18 June 2010

Received in revised form 6 September 2010

Accepted 7 September 2010

Edited by: K. Hirose.

Keywords:

Diffusion

Creep

D'' layer

Olivine

Post-perovskite

ABSTRACT

The influence of anisotropic diffusion coefficients on diffusion-controlled high-temperature creep is examined. Anisotropic diffusion affects anisotropy of diffusion-controlled deformation of a single crystal. The shape change of a single crystal by diffusional mass flux is controlled directly by the anisotropy in diffusion coefficients. The rate of shape change of a single crystal by diffusion-controlled dislocation glide is controlled by the anisotropy of diffusion coefficients on the plane normal to the dislocation line. Consequently, if a polycrystalline aggregate is deformed uniformly by diffusion creep, then the rate of deformation is controlled by that of diffusion (of the slowest diffusion species) along the slowest direction. In contrast, when a polycrystalline aggregate is deformed by climb-controlled dislocation creep, the rate of deformation is controlled by the diffusion (of the slowest diffusion species) along the direction where the diffusion coefficient has the intermediate value. The results are applied to olivine and post-perovskite. For olivine, the observed large plastic anisotropy and small anisotropy in diffusion suggests that high-temperature power-law creep is controlled not only by diffusion but also by some other factors such as jog density. For post-perovskite, the results of numerical calculations on the mobility of vacancies would suggest that the viscosity of post-perovskite aggregates is higher than or comparable to that of the perovskite aggregates if the defect concentrations were the same among these minerals. However, currently nothing is known about the defect concentrations in post-perovskite and other coexisting phases, and therefore it is impossible to compare the creep strength among coexisting phases from these results.

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1. Introduction

Two recent papers motivated me to examine the role of anisotropic diffusion in controlling the creep strength of a polycrystalline aggregate. Firstly, [Ammann et al. \(2010\)](#) showed, based on numerical calculations, that the mobility of vacancies (at Mg and Si sites) in the post-perovskite phase is highly anisotropic (~ 8 orders of magnitude difference in diffusion coefficient), and the fastest diffusion coefficients of these defects in the post-perovskite phase are much faster than those of Si and Mg in perovskite (and comparable to diffusion of Mg and O in MgO) whereas the slowest diffusion coefficient in the post-perovskite phase is much slower than those of Si and Mg in perovskite. Based on the inferred large diffusion anisotropy, [Ammann et al. \(2010\)](#) discussed that the viscosity corresponding to diffusion creep of a polycrystalline aggregate of post-perovskite is higher than that of perovskite (comparable to that in MgO), but the viscosity corresponding to dislocation creep in post-perovskite is much lower than that of perovskite. Their argu-

ment is based on the premise that diffusion of the slowest species along the slowest direction controls the viscosity of a polycrystalline aggregate deformed by diffusion creep, but the diffusion of the slowest diffusing species along the fastest orientation controls the viscosity of a polycrystalline aggregate deformed by dislocation creep. However, there has been no theoretical analysis on these issues, and consequently the validity of their conclusions is unclear.

Secondly, [Kohlstedt \(2006\)](#) re-examined the connection between diffusion and high-temperature dislocation creep in olivine, and concluded that a classic model of diffusion-controlled dislocation creep (e.g., [Weertman, 1955, 1957, 1968, 1999](#)) applies to olivine. The main argument by [Kohlstedt \(2006\)](#) to support this classic model is (i) the similarity in the activation energy between creep of polycrystalline aggregate and the activation energy of Si (and O) diffusion ([Costa and Chakraborty, 2008; Dohmen et al., 2002](#)), (ii) the near agreement of the amount of water effects on diffusion and creep and (iii) the agreement of predicted strain-rates from a simple model with observed ones. Although the near agreement in activation energy between these processes, based on the recent diffusion measurements cited above, is remarkable, it is not entirely clear if this implies a simple link between diffusion and creep in olivine. There are several issues that need to be

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examined. First, Bai et al. (1991) showed that creep behavior of olivine single crystal is complex having a range of activation energy depending on the orientation and thermodynamic conditions. Consequently, Bai and Kohlstedt (1992a), for example, proposed the role of jogs to control the creep rate (see also Karato, 1989). Second, the experimental studies on diffusion also showed very weak anisotropy compared to anisotropy in single crystal deformation of olivine. Third, a diffusion-controlled dislocation creep model is difficult to explain the observed fabric transitions in olivine (Karato et al., 2008).

In order to address these issues, it seems important to examine the link between plastic anisotropy and anisotropic diffusion coefficient. In this paper, I will first briefly review the role of anisotropic diffusion in diffusion creep in a polycrystal, and then develop a model of dislocation climb in a material with anisotropic diffusion coefficient to clarify the link between anisotropic diffusion and the creep strength of a polycrystalline aggregate deformed by dislocation creep. I will show that rate-controlling diffusion in diffusion creep is diffusion along the slowest direction, whereas that in dislocation creep in a polycrystalline aggregate is the diffusion coefficient along the direction in which diffusion coefficient has an intermediate value. Some implications for deformation of post-perovskite and olivine will be discussed.

2. Theory

2.1. Diffusion creep

Although theory of diffusion creep was developed long time ago (e.g., Herring, 1950; Lifshitz, 1963; Nabarro, 1948), there has been no theoretical analysis to examine the influence of anisotropic diffusion. In all of these papers, isotropic diffusion coefficient was assumed. Only paper where the influence of anisotropic diffusion in diffusion creep that I am aware is Yang and Li (1995) where some very specific cases were discussed.

Extension of diffusion creep model for anisotropic diffusion is, however, straightforward. We use a concept that for a polycrystalline material to deform by diffusion creep, the continuity in displacement at grain-boundaries needs to be satisfied (Lifshitz, 1963). A simple case where this condition is met is homogeneous deformation, and I assume that this condition is met. The classic model of diffusion creep shows that for a material that deforms by diffusion creep, strain-rate and stress are related by

$$\sigma = \alpha \frac{RT}{\Omega} \frac{L^2}{D} \dot{\epsilon} = \eta \dot{\epsilon} \quad (1)$$

where σ is stress, $\dot{\epsilon}$ is strain-rate, R is the gas constant, Ω is the molar volume, L is grain-size (I consider volume diffusion for simplicity), D is diffusion coefficient and η is viscosity.

In order to extend this model to anisotropic diffusion, let us consider, first, a single crystal subjected to stress along three directions (x_i , $i=1, 2, 3$). In the following, I will use the coordinate system where the main axes are parallel to the main axes of a crystal where the diffusion coefficient tensor is diagonal. General conclusions will not be affected by this choice of the coordinate system. By extending the treatment such as a model by Lifshitz (1963) to anisotropic diffusion, I obtain,

$$\sigma_{ii} = \alpha \frac{RT}{\Omega} \frac{L^2}{D_{ii}} \dot{\epsilon}_{ii} \quad (i=1, 2, 3) \quad (2)$$

where σ_{ii} is normal stress acting on the grain-boundary normal to the x_i direction, $\dot{\epsilon}_{ii}$ is the shortening rate along the x_i direction, D_{ii} is diffusion coefficient along the x_i direction when the force is acting along that direction.

If a polycrystal deforms homogeneously, then strain in each grain must be the same. This means that the stress on individual

grains is different depending on the magnitude of diffusion coefficient, D_{ii} . The total stress of the sample is the arithmetic mean of the local stress, hence,

$$\sigma = \left(\alpha \frac{kTL^2}{\Omega} \left\langle \frac{1}{D_{ii}} \right\rangle \right) \dot{\epsilon} = \eta \dot{\epsilon} \quad (3)$$

where $\langle 1/D_{ii} \rangle$ is the average of $1/D_{ii}$.

$$\eta = \alpha \frac{kTL^2}{\Omega} \left\langle \frac{1}{D_{ii}} \right\rangle \quad (4)$$

Hence I conclude that in diffusion creep of a polycrystalline material made of crystals with anisotropic diffusion coefficient, it is the diffusion coefficient along the slowest direction that controls the viscosity (if one component of diffusion coefficient is zero, a polycrystalline material cannot deform by diffusion creep homogeneously).

2.2. Diffusion-controlled dislocation creep

Let us now consider deformation by dislocation creep that is rate-controlled by dislocation climb. Dislocation climb involves atomic diffusion, and I will consider how anisotropy in diffusion affects the anisotropy in deformation by climb-controlled dislocation creep. The shape change due to dislocation glide is determined by the magnitude of slip for a given slip system, and geometry of shape change is controlled solely by the direction of slip and the orientation of the glide plane. Several slip systems exist for a given crystal and a multiple of slip systems are needed to achieve homogeneous deformation of a polycrystal (von Mises, 1928). At high temperatures, the rate at which dislocation glide occurs is controlled by dislocation recovery that is in turn controlled by diffusion. In this paper, I will consider a case where the rate of deformation by dislocation glide is controlled by diffusion. In such a case, the rate of deformation for each slip system is controlled by the diffusion coefficient relevant to that slip system. Because the homogeneous deformation of a polycrystalline aggregate requires the operation of multiple slip systems (von Mises condition), the rate of deformation of a polycrystalline aggregate is in most cases controlled by that of the most difficult slip system (Kocks, 1970).

In the following, I will examine how the anisotropy in diffusion coefficient affects the choice of the most difficult slip system. I will assume that the creep rate is controlled by the diffusion-controlled climb motion of an edge dislocation (Weertman, 1957, 1999). Under this assumption, the strain-rate for a certain slip system is determined by the rate of dislocation climb for a certain type of an edge dislocation, and the strain-rate is related to the velocity of dislocation climb, v_c , as (e.g., Karato, 2008; Poirier, 1985; Weertman, 1999)

$$\dot{\epsilon} = \rho b v_c \frac{L}{d} \quad (5)$$

where $\dot{\epsilon}$ is strain rate, ρ is the density of mobile dislocations, L is the characteristic length of dislocation glide, d is the characteristic length of dislocation climb, and v_c is the velocity of dislocation climb.

The climb velocity of dislocation is controlled by diffusion and the number density of jogs on the dislocation line, viz.,

$$v_c = \frac{2\pi\sigma\Omega}{RT} \frac{Dc_j}{\log(L/b)} \simeq \frac{\sigma\Omega}{RT} Dc_j \quad (6)$$

where σ is stress, Ω is the molar volume of diffusing species, c_j is jog density (number of jogs per unit length), b is the length of the Burgers vector, L is the mean distance of dislocations, and D is the relevant diffusion coefficient (e.g., Karato, 2008; Poirier, 1985). In case a dislocation line is saturated with jogs, then $c_j = 1/b$ and the anisotropy in climb velocity is solely controlled by the anisotropy

of diffusion. However, when jog density is low (under-saturated case), then the anisotropy in jog density will also contribute to the anisotropy in climb velocity.

This relationship can be extended to a case of anisotropic diffusion. To do this, recall that Eq. (6) is derived using the relation $v_c = lb^2$ (l is vacancy flux (per unit length of dislocation) and b is the length of the Burgers vector), and the vacancy flux can be calculated from the vacancy concentration that is obtained from the diffusion equation. Let us consider, for simplicity, a case of climb of an edge dislocation oriented parallel to one of the major crystallographic orientations (a similar analysis can be made for a more general case, but the main conclusions remain the same). In order to calculate the rate of dislocation climb, one solves the diffusion problem where a dislocation line is the source/sink of vacancies (atoms). If the dislocation line is along the z -direction, then the diffusion equation for vacancy concentration, c , is

$$\frac{\partial c}{\partial t} = D_{xx}^v \frac{\partial^2 c}{\partial x^2} + D_{yy}^v \frac{\partial^2 c}{\partial y^2} \quad (7)$$

where D_{xx}^v and D_{yy}^v are the diffusion coefficient of vacancies along the x - and y -directions, respectively. This equation can be solved for appropriate boundary conditions to obtain the concentration profile of vacancies, and then the flux of vacancies and the climb velocity of an edge dislocation can be calculated as,

$$v_{c,z} = \frac{2\pi\sigma\Omega}{RT} \frac{c_j}{\log(L/b)} \frac{D_{xx} + D_{yy}}{2} \propto \dot{\epsilon}_z \quad (8)$$

where I used a relation $D = D^v c_0$ (D is the diffusion coefficient of an atom), $v_{c,z}$ is the climb velocity of an edge dislocation parallel to the z -direction, and $\dot{\epsilon}_z$ is the strain rate of deformation controlled by dislocation climb where an edge dislocation is parallel to the z -direction. This means that the diffusion coefficient D in Eq. (5) should be replaced with $(D_{xx} + D_{yy})/2$. Similar relations hold for the climb of edge dislocations along other orientations.

Applying the above analysis together with the von Mises conditions, and assuming that the jog density is isotropic (i.e., jog-saturated conditions), I conclude that it is the smallest of $((D_{xx} + D_{yy})/2)$, $((D_{xx} + D_{zz})/2)$, $((D_{yy} + D_{zz})/2)$ that controls the rate of deformation of a polycrystalline aggregate by climb-controlled dislocation creep ("power-law" dislocation creep). Now, without the loss of generality, one can define the coordinate such that $D_{xx} > D_{yy} > D_{zz}$. Then the slowest effective diffusion coefficient for dislocation climb is $((D_{yy} + D_{zz})/2) \approx (D_{yy}/2)$ if $D_{yy} \gg D_{zz}$. Consequently, it is the diffusion coefficient along the direction where diffusion coefficient has the intermediate value that controls the rate of deformation of a polycrystalline aggregate when deformation occurs by climb-controlled dislocation creep.

Some modifications are needed when the dislocation lines are not straight. This effect is important when the diffusion coefficient along the fastest orientation is much larger than others. However, the corrections for such effects require a precise knowledge of geometry of dislocations and will not be treated in this paper.

3. Applications

3.1. Post-perovskite

A recent first-principles study showed that the mobility of point defects at Mg-site (and Si-site) in the post-perovskite phase of MgSiO_3 is highly anisotropic (Ammann et al., 2010). Since Mg or Si diffusion is likely to control the creep rate in this mineral (oxygen diffusion is much faster), the role of highly anisotropic diffusion (of Mg or Si) in controlling the rheological properties of this mineral needs to be examined.

For diffusion-controlled deformation such as diffusion creep or high-temperature climb controlled dislocation creep, it is the dif-

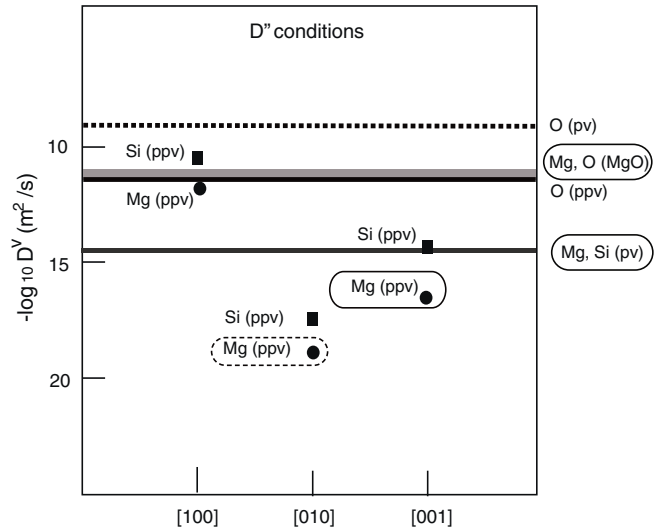


Fig. 1. A comparison of vacancy diffusion coefficients in lower mantle minerals under the D^v layer conditions along various orientations (after Ammann et al. (2010)). Circled values correspond to the rate-controlling diffusion coefficients for creep for each material. pv means perovskite and ppv post-perovskite. For MgO diffusion is isotropic because of the cubic symmetry, and for perovskite diffusion is nearly isotropic (oxygen diffusion in post-perovskite is also nearly isotropic).

fusion of the slowest species that controls the rate of deformation (e.g., Karato, 2008). The present analysis shows that for diffusion creep with anisotropic diffusion coefficients, the rate controlling process is the diffusion of the slowest diffusing species along the slowest orientation, and hence for post-perovskite, it is Mg diffusion along the [0 1 0] orientation (if the defect concentration at Mg-site is the same as that at Si-site). The defect mobility (diffusion coefficient of vacancies) at Si and Mg sites along the [0 1 0] orientation in post-perovskite is slower than those in perovskite and in MgO (Fig. 1). Consequently, if the defect concentration in post-perovskite is the same as those in other minerals as assumed by Ammann et al. (2010), then post-perovskite would have a larger viscosity than other coexisting minerals when it is deformed by diffusion creep. When deformation occurs by dislocation creep controlled by diffusion (dislocation climb), then diffusion along the intermediate direction controls the rate of deformation. The mobility of vacancies at Mg site in post-perovskite along the intermediate direction, [0 0 1], is slower than or comparable to that of perovskite, so one would conclude that viscosity of post-perovskite controlled by dislocation creep would be higher than that of perovskite. However, it should be emphasized that there is no reason to assume the same defect concentrations among coexisting minerals and various sites in a given mineral. The concentration of point defects is controlled by the thermo-chemical parameters and by the properties of a given material (e.g., defect formation energy). Defect formation energy is in general different among different materials. Therefore it is impossible to compare diffusion coefficients of different minerals from the results calculation of defect mobility alone.

Although the absolute values of diffusion coefficients in post-perovskite are unconstrained by the work of Ammann et al. (2010), one could derive a few useful conclusions. First, because anisotropy in defect diffusion is very large, and diffusion coefficient along different directions controls the viscosity corresponding to diffusion and dislocation creep, one can conclude that dislocation creep is relatively easier than diffusion creep in post-perovskite phase. Second, the easiest slip system in post-perovskite is likely the one in which the direction of edge dislocation is normal to [1 0 0]. Considering the length of the Burgers vector as another controlling factor for the slip system, a slip system involving $\mathbf{b} = [1 0 0]$ is likely the

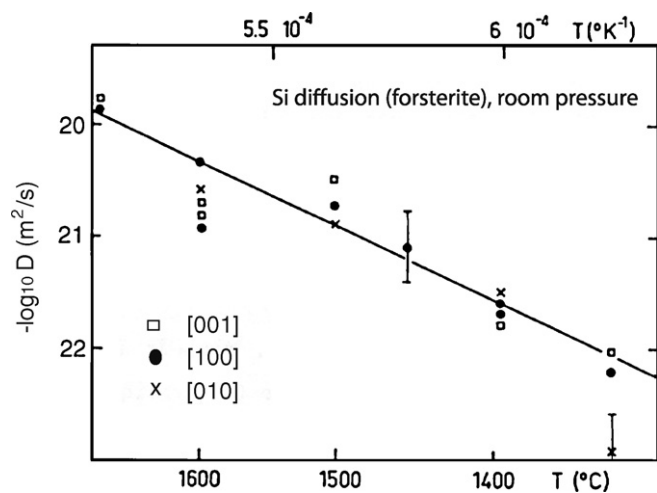


Fig. 2. Anisotropy in diffusion coefficients of Si in olivine (data for forsterite by Jaoul et al. (1981) at room pressure). For San Carlos olivine, less extensive study has been conducted. A comparison of Houlier et al. (1990) with Dohmen et al. (2002), however, suggests a small anisotropy.

dominant (the easiest) slip system that will define the nature of lattice-preferred orientation.

3.2. Olivine

For olivine, both diffusion and plastic deformation have been studied extensively. Fig. 2 shows the results on diffusion coefficients (Houlier et al., 1990; Jaoul et al., 1981) and Fig. 3 shows results on plastic deformation (Bai et al., 1991; Darot and Gueguen, 1981). It is noted that plastic anisotropy is much larger than the anisotropy in diffusion (of Si). It should also be noted that the activation energy of creep for olivine single crystals varies a lot with orientation of the crystal and/or on the thermo-chemical conditions (230–1000 kJ/mol for San Carlos olivine, 460–600 kJ/mol for forsterite). Therefore I conclude that a large portion of plastic anisotropy in dislocation creep in olivine is caused by the anisotropy of some factors other than diffusion coefficient.

In order to examine possible causes for large anisotropy, let us go back to Eq. (5), $\dot{\epsilon} = \rho b v_c (L/d)$, appropriate for climb-control mechanism. In this equation, dislocation density changes little with slip systems (as far as the Schmid factor (a geometrical factor expressing the relation between applied stress and the effective stress for slip, see e.g., Poirier, 1985) is same), so is the length of the Burgers vector. So the possible sources for large anisotropy is either the anisotropy in dislocation climb velocity, v_c , or the ratio of length of dislocation glide and climb, L/d . However, the length ratio, L/d , is unlikely to be temperature sensitive and therefore is not a likely source for large variation in activation energy observed by Bai et al. (1991) for San Carlos olivine (Darot and Gueguen (1981) also reported different activation energies for different orientations for forsterite). I conclude that because diffusion coefficients are nearly isotropic, anisotropy in creep rate in olivine is likely due to the anisotropy in jog concentration. An alternative mechanism for large anisotropy is the contribution from glide-controlled mechanism where anisotropic Peierls stress likely results in highly anisotropic creep strength.

4. Discussions

In a recent paper, Ammann et al. (2010) discussed that the post-perovskite phase would be the weakest phase in the D'' layer when it is deformed by dislocation creep but it is the strongest mineral when it is deformed by diffusion creep. However, the validity of

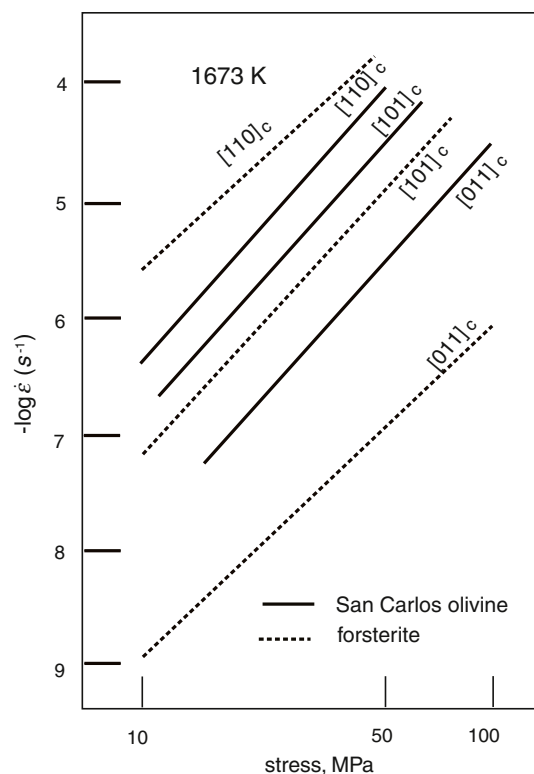


Fig. 3. Anisotropy in creep in olivine single crystals (at 1673 K, room pressure). $[110]_c$, $[101]_c$, $[011]_c$ indicate the orientation of crystals. (a) San Carlos olivine (data by Bai et al. (1991)). (b) Forsterite (data by Darot and Gueguen (1981)). For San Carlos olivine, the results are for opx buffer and at $f_{O_2} = 10^{-1}$ Pa. Deformation experiments for forsterite were conducted in air without any oxide buffer. The activation energy of creep in San Carlos olivine also varies with orientation as well as with some thermodynamic parameters (from 230 to 1000 kJ/mol (the activation energy of creep for forsterite varies from 460 to 600 kJ/mol)). Similarly dependence of creep rate in San Carlos on chemical factors such as oxygen fugacity varies from one orientation to another.

such a conclusion is questionable for two reasons. First, because the calculations by Ammann et al. (2010) are only for the mobility of point defects (vacancies), nothing can be said about the diffusion coefficients that are also proportional to the defect concentration. Second, even under the assumption of common point defect concentrations among coexisting minerals, their conclusion on the viscosity corresponding to diffusion-controlled dislocation creep is not supported by the simplest model of dislocation climb-controlled model presented here.

Let me discuss the role of diffusion in high-temperature dislocation creep in olivine. The importance of diffusion in high-temperature dislocation creep has been recognized for a long time (e.g., Weertman, 1955, 1957, 1968, 1999). There is no doubt that in some cases, there is a close link between diffusion and high-temperature dislocation creep in materials such olivine. However, the extensive studies by Bai and Kohlstedt (1992a,b) and Bai et al. (1991) showed that the link between diffusion and high-temperature dislocation creep is not simple for olivine. The most important observation to show this is the large anisotropy in creep behavior of olivine single crystals. In particular, a large variation in activation energy of creep with orientation (230–1000 kJ/mol for San Carlos olivine), and a variety of dependence on chemical environment such as oxygen fugacity indicates that a simple diffusion-control model does not work for many slip systems. It is still possible that a simple model works for the most difficult slip system under dry low stress conditions, $[001] (010)$, that controls the creep strength of a polycrystalline aggregate for which the activation energy of creep agrees well with the new result on the

activation energy of diffusion. However, recent studies suggest that this slip system will become the easiest slip system under water-rich (and/or high-pressure) conditions (Karato et al., 2008). Under these conditions, the slip system that controls the strength of a polycrystalline aggregate will be other than the [001] (010) slip system, and hence the applicability of a simple model for deformation of an olivine polycrystal becomes less secure.

In addition, observed nearly isotropic diffusion (Costa and Chakraborty, 2008) and highly anisotropic effects of water to enhance deformation (Karato et al., 2008) provide another support for the notion that some factors other than diffusion play important roles in plastic deformation of olivine. Finally, the near agreement in strain-rates calculated from a simple diffusion-controlled model and the observed values cannot be taken as strong support for the validity of the model because the model has an adjustable parameter, L/d .

5. Summary and conclusions

A simple analysis is presented to examine the relationship between diffusion and high-temperature creep of a polycrystalline aggregate when diffusion is anisotropic. I conclude that in diffusion creep, it is the diffusion along the slowest direction that controls the rate of deformation. In contrast, in diffusion-controlled dislocation creep, it is the diffusion along the direction where the diffusion coefficient is intermediate that controls the rate of deformation of a polycrystalline aggregate (if a simple geometry of dislocation lines is assumed). These results are applied to olivine and post-perovskite to interpret some experimental and computational results. I conclude that the currently available results on defect mobility in post-perovskite are insufficient to constrain the magnitude of diffusion coefficients and the creep behavior of this mineral. Additional information on defect concentration is needed. For olivine, the observed large contrast in anisotropy between dislocation creep and diffusion suggests that dislocation creep in olivine involves processes other than diffusion as previously suggested (e.g., Bai et al., 1991; Karato, 1989).

Acknowledgment

This study is supported by National Science Foundation. I thank Jim Van Orman for the helpful review.

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