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EFFECT OF VISCOUS GRAIN-BOUNDARY SLIDING ON HIGH-TEMPERATURE DEFORMATION OF NANO-SIZED GRAINS

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Abstract. For steady-state deformation caused by grain-boundary diffusion, the macroscopic creep rate and the effect of viscous grain-boundary sliding in a polycrystal of nano-sized grains are analyzed by the energy-balance method in three dimensions. At decreasing grain sizes, the influence of the viscous grain-boundary sliding becomes increasingly important, which explains the recent experimental observations that the creep rates of nano-sized grains are much lower than those predicted by grain-boundary diffusion.

1. INTRODUCTION

When a polycrystal is stressed at high temperatures, deformation may occur by diffusional flow of atoms. The stress-directed diffusion, which occurs through grain and/or along grain boundary, induces grain elongation, grain-boundary sliding and macroscopic strain. Theoretical analyses of diffusional creep have been conducted for various microstructures including single spherical grain [1-3], single ellipsoidal grain [4], hexagonal grains with identical size and shape [5-9], square grains with different size [10], two-dimensional irregular microstructures [11-14], three-dimensional polycrystal with spherical grains [15] and particle-embedded composites [16].

All of the above studies, except some works [9,15,16], assumed that the shear stress against grain-boundary sliding is fully relaxed at the steady state, and that sliding can occur at any rate. This is not correct. Even an atomically flat grain boundary must be considered to be viscous in the same way that a liquid is viscous. Hence, the rate of diffusional creep should correspondingly decrease with increasing grain-boundary viscosity.

The effect of viscous grain-boundary sliding on the overall creep rate was first formulated by Mori *et al.* [16]. By utilizing the energy-balance method, they analyzed the contributions to energy dissipation by both diffusion and viscous grain-boundary sliding, then predicted the overall creep rate. Using this method, Onaka *et al.* [15] later predicted the creep rate of a three-dimensional polycrystal with equiaxed grains of various sizes. In these models, an analysis was conducted for a single spheroidal particle surrounded by a uniformly deforming matrix, then the creep rate of the polycrystal was predicted by increasing the volume fraction of the particles to unity. A spheroidal particle, however, is not space-filling, so that an aggregate of them does not yield polycrystalline microstructures.

In the diffusional creep of a polycrystal, grain-boundary diffusion and grain-boundary sliding occur along the common boundary facet between two neighboring grains, and the net flux of matter into

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the boundary is determined by grain-boundary diffusion across the surrounding triple junction. Recently, we analyzed such diffusional deformation in two-dimensional hexagonal grains by modifying the energy-balance method [9]. According to the analysis, the creep rate $\dot{\varepsilon}$ of hexagonal grains is represented as

$$\dot{\varepsilon} = 36\frac{\Omega\delta D\sigma}{kTL_0^3}\left[1+\eta^*\right]^{-1}, \tag{1}$$

where Ω is the atomic volume, δ is the thickness of grain boundary, D is the diffusion coefficient of grain boundary, σ is the external stress, k is Boltzmann's constant, T is the absolute temperature, L_o is the grain size, η is the viscosity of grain boundary, and η^* is the normalized viscosity of $36\eta\Omega D/(kTL_o^2)$. This equation indicates that viscous grain-boundary sliding decreases the creep rate and the influence becomes increasingly important at decreasing grain sizes. In the present study, we apply the energy-balance method to a three-dimensional polycrystal consisting of space-filling grains, and discuss further the influence of the viscous grain-boundary sliding for nano-sized grains. We assume that grains in a polycrystal have flat and circular boundary facets of both identical size and uniform distribution in all orientations.

2. GRAIN-BOUNDARY DIFFUSION AND VISCOUS SLIDING

When a polycrystal is deformed by diffusion under uniaxial stress, the relative displacement between two neighboring grains can be divided into normal and tangential components to the common boundary facet. Using Stevens' analysis [17], we can obtain the relative velocities, v_n and v_t, as

$$v_n = \left(1-1.5\sin^2\theta\right)L\dot{\varepsilon}$$
and
$$v_t = \left(1.5\sin\theta\cos\theta\right)L\dot{\varepsilon}, \tag{2}$$

where θ is the angle between the stress axis and the vector normal to the boundary facet, L is the distance between the centers of the two neighboring grains, and the subscripts n and t mean the normal and tangential components, respectively (Fig. 1). These relative velocities are constant on each boundary facet, because the boundary facets are assumed to remain flat and not rotate during deformation.

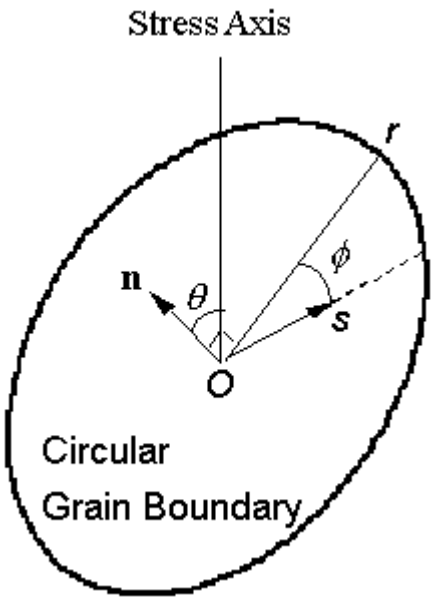

Fig. 1. Circular grain-boundary facet with a radius r and an angle θ.

Consider diffusion on the flat and circular boundary facet with a radius r and an angle θ. If the boundary diffusion is represented in the cylindrical coordinate system $(\sigma-\phi)$ shown in Fig. 1, the condition of the volume conservation of diffused matter yields

$$v_n = -\frac{\Omega}{s}\left[\frac{\partial(J_s\cdot s)}{\partial s}+\frac{\partial J_\phi}{\partial\phi}\right], \tag{3}$$

where s is the distance from the center of the boundary, ϕ is the angle on the boundary, and J_s and J_ϕ are the flux of matter in the radial and tangential direction, respectively. In addition, since the rate of the volume increase due to the total flux of matter into the boundary with a radius $s(-\Omega\int J_s s d\phi)$ is equal to $\pi s^2 v_n$, we can obtain another equation for the volume conservation as

$$v_n = -\frac{\Omega}{\pi s}\int_0^{2\pi} J_s d\phi. \tag{4}$$

We next obtain J_s and J_ϕ on the boundary facet by solving Eqs. (3) and (4) with appropriate boundary conditions. Consider two additional grain boundaries connected to the triple line around the circular boundary facet at an equilibrium angle of 120°. During grain-boundary diffusion, the sum of the radial fluxes on the three grain boundaries through the triple line is zero, and the average contribution of the tangential flux J_ϕ to the relative velocity (v_n) is zero on

respective boundaries. Moreover, for axisymmetric diffusion ($\partial J_\phi/\partial\phi$=0), the radial flux J_s on each boundary is proportional to the relative velocity v_n. Hence, assuming that the radial flux around the triple line is proportional to the difference of the relative velocities between grain boundaries and ignoring the tangential contribution, we can estimate the radial flux J_s at the periphery (s=r) of the circular boundary facet to be $a(v'_n+v''_n-2v_n)$, where a is a constant, and v'_n and v''_n are the relative velocity of the additional grain boundaries connected to the triple line. Another boundary condition is found from the symmetry of the circular boundary; J_s=0 at s=0 and J_ϕ=0 at ϕ=0. Then, the value of a is determined from Eq. (4), and J_s and J_ϕ are obtained from Eqs. (2) and (3) as

$$J_s = \frac{sL\dot{\varepsilon}}{2\Omega}\left(\sin^2\theta\cos^2\phi - \cos^2\theta\right), \tag{5}$$

$$J_\phi = -\frac{sL\dot{\varepsilon}}{4\Omega}\sin^2\theta\sin 2\phi. \tag{6}$$

The velocity of diffusing atoms v_a is obtained by using a relationship of $\Omega J=\delta v_a$.

According to the Einstein's relationship, the velocity v_a is related to the force F acting to the atom as $v_a=FD/(kT)$. Since the work done by the diffusing atom $\dot{q}$ is given by

$$\dot{q} = \frac{kT\Omega^2}{D\delta^2}\left(J_s^2 + J_\phi^2\right), \tag{7}$$

the total energy-dissipation rate $\dot{W}_{D\theta}$ due to diffusion on the circular grain boundary is

$$\dot{W}_{D\theta} = \frac{\delta}{\Omega}\int_0^{2\pi}\int_0^r \dot{q}\cdot s\,ds\,d\phi. \tag{8}$$

Then, the mean energy-dissipation rate $\dot{W}_D$ due to diffusion on the grain-boundary facets which are uniformly distributed in all orientations, is

$$\dot{W}_D = \int_0^{\pi/2}\dot{W}_{D\theta}\sin\theta\,d\theta. \tag{9}$$

In addition to grain-boundary diffusion, the work done by grain-boundary sliding $\dot{W}_S$ due to viscous boundary sliding can be evaluated;

$$\dot{W}_s = \int_0^r \tau_t v_t\cdot 2\pi s\,ds. \tag{10}$$

Here, τ_t is the shear stress on the grain boundary. If the viscous sliding is described by a Newtonian flow, τ_t is represented as $\eta v_t/\delta$.

In a steady state, the total energy dissipated by both grain-boundary diffusion and viscous grain-boundary sliding is supplied by the external work. Then, an energy balance gives

$$2V\sigma\dot{\varepsilon} = m\dot{W}_D + m\dot{W}_s, \tag{11}$$

where V is the volume of the grain, and m is the number of the boundary facets of the grain. Assuming a tetrakaidecahedral grain (m=14) with an edge length l, the average surface area of the boundary facet S_f is $S_f/l^2=3(1+2\sqrt{3})/7$, and the volume V is $V/L^3=(1+2\sqrt{3})^3/128$. Using Eqs. (9) and (10) to solve $\dot{\varepsilon}$ in Eq. (11), we obtain

$$\dot{\varepsilon} = 14\alpha^3\frac{\Omega\delta D\sigma}{kTL^3}\left[1+\eta_L^*\right]^{-1} = 14\frac{\Omega\delta D\sigma}{kTR^3}\left[1+\eta_R^*\right]^{-1}, \tag{12}$$

where R is the volume-equivalent radius ($V=4\pi R^3/3$), α is the ratio L/R (=1.82), and η_L^* and η_R^* are the normalized viscosities defined as $63\alpha^3\eta\Omega D/(5kTL^2)$ and $63\alpha\eta\Omega D/(5kTR^2)$, respectively. The most important features of the solution are that the viscous grain boundary decreases the strain rate, and that the apparent grain-size exponent changes from three to unity with increasing viscosity and/or decreasing grain size.

3. DISCUSSION

For η_R^*=0, Eq. (12) agrees with the previous solutions for diffusional creep controlled by grain-boundary diffusion, except for the numerical constant. For a three-dimensional polycrystal, Coble [2], Green [3] and Onaka *et al.* [15] predicted the numerical constant to be 18.5/π, 4 and 10, respectively. The major characteristics of the deformation models due to grain-boundary diffusion are summarized in Table 1. Compared with these values, Eq. (12) gives a larger numerical constant 14 which is identical to the number of the boundary facets per grain (m). This is not coincidental. All the previous analyses assumed an axisymmetric spherical grain, and the boundary condition was such that the points of symmetry with zero flux are the north pole, the south pole and the equator. Our model, however, exploits the additional symmetry of a regular polyhedron (tetrakaideca-hedron), in which the center of each facet also satisfies the zero-flux condition. This ap-

Table 1. Grain-boundary diffusion models.

Model	Grain-boundary sliding	The numerical constant in Eq. (12)	Reference
Single spherical grain	Non-resistant	$18.5/\pi$	[2]
Single spherical grain	Non-resistant	4	[3]
Single spherical grain + Uniform matrix	Viscous	10	[15]
Space-filling polycrystal	Viscous	14	Present study

parently shortens the diffusion distance, resulting in a higher strain rate. Therefore, we may conclude that the effective diffusion distance is determined not only by the grain size but also by the degree of symmetry of a typical grain.

The strain rate of Eq. (12) for the three-dimensional polycrystal may be compared with experiments. Morita and Hiraga [18] conducted constant-stress creep tests for ZrO_2-3 mol.% Y_2O_3 with different grain sizes, and reported that at 3 MPa and 1400 °C, $\dot{\varepsilon}_1=2.56\cdot10^{-7}$ /s for $L_1=0.58$ μm, and $\dot{\varepsilon}_2=9.27\cdot10^{-8}$ /s for $L_2=1.15$ μm. At such low flow stress and high temperature, it is believed that diffusional deformation occurred in the material. Taking $\Omega=4.67\cdot10^{-29}$ m^3, $\delta D=3.34\cdot10^{-22}$ m^3/s, $\delta=10^{-9}$ m and $\eta=10^8$ Pa·s at $T=1400$ °C, we obtain $\dot{\varepsilon}_1=5.39\cdot10^{-5}$ /s ($\eta_{L1}^*=15.2$) and $\dot{\varepsilon}_2=2.30\cdot10^{-5}$ /s ($\eta_{L2}^*=3.9$) from Eq. (12).

The strain rates predicted by Eq. (12) for ZrO_2 polycrystals are two orders of magnitude higher than the experimentally observed creep rates. The discrepancy may be explained by the following considerations. First, the experimental data of dD may be in error. Many reports on the diffusion coefficient indicated large scatter of data depending on measuring methods and authors. Also, the lack of data on δD, η and $\dot{\varepsilon}$ in the same system may have compounded the problem. Second, the nonuniform microstructure of an actual polycrystal may cause asymmetric fluxes of matter on grain boundaries, reducing the symmetry thus increasing the effective diffusion distance. Lastly, if the grain-boundary facets are non-planar or grain rotation occurs, there would be additional energy dissipation that would also lower the strain rate.

Although the creep rate predicted from Eq. (12) may not be useful at this point, we find the predicted grain-size effect to be more meaningful. Due to the influence of the grain-boundary viscosity, the ratio of strain rates for two different grain sizes is not simply L_2^3/L_1^3, as predicted by the classical theory (Coble [2]), but rather

$$\frac{\dot{\varepsilon}_1}{\dot{\varepsilon}_2}=\frac{L_2^3+cL_2}{L_1^3+cL_1}, \qquad (13)$$

where $c=63\alpha^3\eta\Omega D/(5kT)$ from Eq. (12). Using the above data on ZrO_2, we obtain $c=5.12$ μm^2 and the ratio $\dot{\varepsilon}_1/\dot{\varepsilon}_2=2.34$. This ratio is comparable to the experimental value of 2.76, which is quite different from the ratio of 7.79 given by L_2^3/L_1^3. This means that for small grain sizes, viscous grain-boundary sliding plays an important role in the creep deformation and the actual strain rate is considerably lower than that predicted by assuming frictionless diffusional creep.

4. SUMMARY AND CONCLUSIONS

By utilizing the energy-balance method, we analyzed the steady-state creep rate caused by both grain-boundary diffusion and viscous grain-boundary sliding in three-dimensional polycrystals consisting of space-filling grains. We considered the diffusional interactions between neighboring grains, including those at triple junctions. These considerations have the effect of increasing the symmetry of the diffusion field, thus reducing the effective diffusion distance. The viscous grain boundary decreases the creep rate, and the apparent grain-size exponent changes from three to unity with increasing viscosity and/or decreasing grain size. Our con-

clusion of viscous grain-boundary sliding offers an explanation of the experimental observation that the strain rates of nanocrystalline materials are much lower than those predicted by grain-boundary diffusion.

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