A New Approach for Diffusional Growth of Grain-Boundary Voids

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The diffusion-controlled growth of grain-boundary voids under an applied stress has been examined by considering the change in the overall Gibbs free energy of a material containing a void. The present new approach enables us to derive the void growth rate without knowing a local stress field at a grain boundary. Comparison with previous studies has revealed that when the same basic assumptions are employed, exactly the same expressions of the void growth rate as those given in the previous studies can be derived in a much simpler manner. The origins of the differences in various previous expressions for the void growth rate are also discussed.

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I. Introduction

At elevated temperatures, polycrystalline metals and alloys often exhibit brittle intergranular fracture. The mechanism of this fracture mode has been studied extensively and it is now well known that the fracture occurs as a result of the nucleation, growth and coalescence of voids on grain boundaries. Progress has been made in the theoretical explanation of a series of these processes and, in particular, the diffusion-controlled growth process of voids has been studied by many investigators.

The first detailed analysis for the diffusional void growth under an applied tensile stress was presented by Hull and Rimmer(1). Thereafter, various corrections and refinements of their analysis have been made. For example, Speight and Harris(2), Weertman(3)-(4), Raj and Ashby(5), Raj et al.(6) Speight and Beere(7), Chuang et al.(8) and Needleman and Rice(9) modified the boundary conditions and obtained various expressions for the void growth rate. All of these analyses are based on essentially the same physical concept: The void growth is considered to occur by the stress-induced diffusive flow of atoms from a surface of a void onto a grain boundary and the void growth rates are obtained by solving the differential equations of diffusion. These differential equations are formulated by taking into account the gradient of chemical potential at the grain boundary. In all of these studies, the excess chemical potential of atoms at the grain boundary relative to the stress-free state, \( \Delta \mu \), is determined by the formulation given by Herring(10), \( \Delta \mu = -\sigma_n \Omega \) where \( \sigma_n \) is the normal tension acting across the grain boundary and \( \Omega \) the atomic volume.

The growth of voids occurs since it can decrease the Gibbs free energy of a material containing voids. In other words, the driving force for the stress-induced diffusive flow of matter is identified as the change in the Gibbs free energy. Hence, it is possible to estimate the amount and rate of the diffusive flow if we know the change in the Gibbs free energy of the material during the void growth. In fact, Onaka et al. previously examined the diffusion-controlled grain-boundary sliding by evaluating the change in the Gibbs free energy and obtained the relationship between applied stress and sliding rate in a very simple man-
A New Approach for Diffusional Growth of Grain-Boundary Voids

In this paper, we will use the same approach to study the diffusion-controlled void growth and examine the growth rates of grain-boundary voids with equilibrium shapes. This alternative approach enables us to derive the void growth rate without knowing a detailed stress field at the grain boundary. This is the advantage of the present approach. As will be seen later, the calculations involved are very simple and the basic physical concept is clear.

II. Basic Concept

From a thermodynamic point of view, the growth of voids under a constant temperature and stress takes place since the Gibbs free energy of a material containing the voids monotonically decreases with an increase in the volume of the voids. Such a monotonic decrease in the Gibbs free energy occurs once the volume of a void exceeds a critical value to overcome a thermodynamic barrier (the nucleation process)\(^{(5)(12)-(14)}\), and since the free energy does not show any local minima during the growth process, the volume of the void will continue to increase till fracture.

The change in the Gibbs free energy during the growth of a void, \(\Delta G\) can be written as

\[
\Delta G = -\sigma \Delta V + \gamma_s \Delta S_s - \gamma_B \Delta S_B,
\]

where \(\Delta V, \Delta S_s\) and \(\Delta S_B\) are the changes in the volume of the void, the surface area of the void, and the area of the grain boundary, respectively. We assume that the diffusive flow of matter from the surface of the void to the grain boundary is the only process for the void growth. In this case, work done by the applied stress (\(\sigma \Delta V\)) is partly spent to drive the diffusive movement of atoms and partly to create a new surface area of the void. This means that \(\Delta G\) must be negative for the void growth to occur. Therefore, considering the decrease in the Gibbs free energy as the driving force for diffusion, we can obtain the relationship among the applied stress, the surface energy, the dihedral angle and other constants and parameters involved in the diffusion process, as shown in the following.

III. Growth Rate for a Long Cylindrical Void

To clarify the physical concept, we will first examine the growth of a simple cylindrical void. Figure 1(a) shows an element of a

Using McCartney's results\(^{(15)}\), it can be shown that if the shape of a void is approximated as a spheroid whose axial ratio is not much different from unity, contribution [iv] is small compared with contribution [i] as the applied tensile stress perpendicular to the grain boundary, \(\sigma\), is much less than Young's modulus, \(E\) (actually, for fcc metals, \(\sigma/E < 10^{-4}\) under the condition of diffusional creep\(^{(16)}\)). Contribution [iv] is also small compared with contribution [i] when a void is not so large \((r_o/r_c << E/\sigma)\), where \(r_o\) is the void radius in the plane of the grain boundary, and \(r_c\) is the critical radius in the plane of the grain boundary for a nucleus of the void. Experimentally observed voids on the fracture surface satisfy this condition\(^{(17)}\). Therefore, as done in previous studies\(^{(1)-(9)}\), contribution [iv] is reasonably neglected when a void has an equilibrium shape.

Then, the change in the Gibbs free energy during the growth of a void, \(\Delta G\) can be written as

\[\Delta G = -\sigma \Delta V + \gamma_s \Delta S_s - \gamma_B \Delta S_B,\]
material containing a cylindrical void of diameter $2r_o$ on a planar grain boundary. The element has a unit depth (measured perpendicular to the sheet), the width $2l$ being equal to the void spacing, and the height $g$ equal to the grain size. Since matter is conserved in this material element, the void growth and diffusion processes can be fully discussed in this element. A remote tensile stress, $\sigma$, is applied to this element perpendicularly to the grain boundary. The geometrical parameters of the void with an equilibrium shape per unit depth of the element, i.e., the volume, $V$, the surface area, $S_s$, and the replaced grain-boundary area, $S_B$, can be expressed in a similar manner to the axisymmetric void \(^{(5)}\) as

\[
V = F_\psi r_o^2, \\
S_s = F_S r_o, \\
S_B = F_B r_o,
\]

where $F_\psi$, $F_S$ and $F_B$ are the functions of $\theta$ as

\[
F_\psi = 2(\theta - \sin \theta \cos \theta) / \sin^2 \theta,
\]

\[
F_S = 4\theta / \sin \theta, \\
F_B = 2.
\]

The relationship among $F_\psi$, $F_S$ and $F_B$ is given by

\[
2F_\psi \sin \theta = F_S - 2F_B \cos \theta.
\]

Consider that during a time interval, $\delta t$, the diameter of the void changes from $2r_o$ to $2(r_o + \delta r)$ and, thus, the volume of the void increases by $\delta V = 2F_\psi r_o \delta r$ per unit depth of the element (see Fig. 1(b)). As done by previous investigators\(^{(1)-(9)}\), we assume that matter deposits uniformly on the grain boundary between voids by diffusion. Then, an increase in the volume of the void occurs by two processes; one is the flow of atoms from the void to the grain boundary by the grain-boundary diffusion and the other is the jacking action of atom-plating\(^{(7)}\). When atoms uniformly plate onto the grain-boundary area, the abutting two grains are moved apart and thus a part of the volume increase occurs without the flow of matter. In other words, because of this jacking action, $\delta V$ becomes larger than the total volume of atoms transported by diffusion\(^{(7)}\). Then, the relationship between $\delta r$ and the number of atoms flowing out of the void by diffusion per unit depth of the element, $\delta n_o$, can be written as

\[
\delta V = 2F_\psi r_o \delta r = \frac{\Omega \delta n_o}{(1 - r_o / l)}.
\]

Here, the term $1 / (1 - r_o / l)$ expresses the effect of the jacking action.

The change in the Gibbs free energy per unit depth of the element, $\delta G$, is evaluated from eq. (2) with eqs. (3) to (6) as

\[
\delta G = -\sigma (2F_\psi r_o \delta r) + \gamma_S (F_S \delta r) - \gamma_B (F_B \delta r) \\
= -2(\sigma - \gamma_S \sin \theta / r_o) F_\psi r_o \delta r.
\]

Obviously, both grain-boundary diffusion and surface diffusion along the void surface must occur simultaneously during the growth of the cylindrical void. However, since the void is assumed to maintain its equilibrium shape, the surface diffusion should occur rapidly and easily. Therefore, $\delta G$ is considered to be the energy spent on the grain-boundary diffusion process during the time interval $\delta t$. 

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Fig. 1 (a) The cross section of an element of a material containing a cylindrical void with an equilibrium shape on a grain boundary subjected to an applied stress $\sigma$. (b) After the time interval, $\delta t$, the void diameter increases by $2\delta r$. 

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\[ F_\psi = 4\theta / \sin \theta, \]

\[ F_S = 2. \]
As atoms are assumed to deposit on the grain boundary between voids, the number of diffusing atoms, $\delta n(x)$, is a decreasing function of the distance from the center of the void, $x$, measured along the grain boundary. In the present model, a planar diffusive flow occurs in directions normal to the axis of the cylindrical void. Then, we obtain the following two equations as the boundary conditions of $\delta n(x)$:

$$\delta n(r_o) = \delta n_0, \quad \delta n(l) = 0. \tag{8}$$

The second condition arises since no net flow of atoms is considered to occur just at the midpoint $(x=l)$ between two neighboring voids. The assumption of the uniform deposition of matter on the grain boundary leads to

$$\frac{d(\delta n(x))}{dx} = C_1, \tag{9}$$

where $C_1(<0)$ is a constant. With eqs. (8) and (9), $\delta n(x)$ becomes

$$\delta n(x) = \delta n_0 \frac{l-x}{l-r_o}. \tag{10}$$

From the conservation of flux in the grain boundary, the drift velocity of atoms, $v(x)$, is written as

$$v(x) = \frac{\Omega \delta n(x)}{A \delta t}, \tag{11}$$

where $A$ is the cross-sectional area across which the grain-boundary diffusive flow passes per unit depth of the element. Considering that the diffusion occurs on grain boundaries at both sides of the voids, we have

$$A = 2w, \tag{12}$$

where $w$ is the grain-boundary thickness.

From Einstein’s relation, the thermodynamic force acting on an atom for the grain-boundary diffusion, $f(x)$, is expressed as

$$f(x) = -\frac{kT}{D_b} v(x), \tag{13}$$

where $D_b$ is the grain-boundary diffusion coefficient and $kT$ has its usual meaning. This force is also derived from the gradient of the Gibbs free energy per atom. When $dG(x)$ describes the change in the Gibbs free energy caused by an infinitesimal movement of $\delta n(x)$ atoms from $x$ to $x+dx$, $f(x)$ can be written as

$$f(x) = -\frac{dG(x)}{dx} \frac{1}{\delta n(x)}. \tag{14}$$

Therefore, we have

$$\delta G = \int_{r_o}^{l} \frac{dG(x)}{dx} dx = -\int_{r_o}^{l} f(x) \delta n(x) dx. \tag{15}$$

Combining eqs. (3) to (15), we obtain the rate of the void growth as

$$\frac{\delta V}{\delta t} = 6 \frac{D_b w}{kT} \left( \sigma - \frac{\gamma_S \sin \theta}{r_o} \right) \Omega \frac{1}{l(1-r_o/l)^3}. \tag{16}$$

As will be shown later, Chuang et al. (8) derived a very similar but slightly different expression for the growth rate of the cylindrical void. The origin of the difference between their result and ours will be discussed in detail later.

**IV. Growth Rate for an Axisymmetric Void**

Next, we treat the growth of a void having a three-dimensional axisymmetric equilibrium shape. The rate of the void growth can be obtained by the approach similar to that in the previous section.

As shown in Fig. 2, consider a cylindrical element of a material of diameter $2l$ and height $g$, which contains in its center a void of diameter $2r_o$ on a planar grain boundary. A tensile stress, $\sigma$, is applied to this element, and diffusive flow occurs on the grain boundary along radial directions. In this case, geometrical parameters of the void with an equilibrium shape, $V$, $S_s$ and $S_b$, are described as (5)

$$V = F_\psi r_o^3, \tag{17}$$

$$S_s = F_\psi r_o^2, \tag{17}$$

$$S_b = F_b r_o^2,$$

where $F_\psi$, $F_\psi$ and $F_b$ are the functions of $\theta$ as

$$F_\psi = \frac{2\pi(2-3 \cos \theta + \cos^3 \theta)}{3 \sin^2 \theta},$$

$$F_\psi = \frac{4\pi}{1+\cos \theta}, \tag{18}$$

$$F_b = \pi.$$
The relationship among $F^\psi$, $F^\parallel$, and $F^\perp$ is given by

$$3F^\psi \sin \theta = F^\parallel - 2F^\perp \cos \theta. \quad (19)$$

Including the effect of the jacking action, the relationship between the increase in the diameter of the void, $2\delta r$, and the number of atoms flowing out of the void by diffusion, $\delta n_o$, can be written as

$$\delta V = 3F^\psi r_o^2 \delta r = \frac{\Omega \delta n_o}{1-r_o^2/l^2}. \quad (20)$$

The change in the Gibbs free energy due to the growth of the void, $\delta G$, thus becomes

$$\delta G = -\sigma (3F^\psi r_o^2 \delta r) + \gamma_s (2F^\parallel r_o \delta r) - \gamma_h (2F^\perp r_o \delta r)$$
$$= -3(\sigma - 2\gamma_s \sin \theta/r_o) F^\psi r_o^2 \delta r. \quad (21)$$

Similar to the case of the cylindrical void in the previous section, the number of diffusing atoms at the grain boundary, $\delta n(x)$, is a function of the distance from the center of the void, $x$. Thus, we obtain the following two equations as the boundary conditions of $\delta n(x)$:

$$\delta n(r_o) = \delta n_o,$$
$$\delta n(l) = 0. \quad (22)$$

The assumption of the uniform deposition of atoms on the grain boundary leads to

$$\frac{d(\delta n(x))}{dx} = C_2 x, \quad (23)$$

where $C_2 (<0)$ is a constant. From eqs. (22) and (23), $\delta n(x)$ becomes

$$\delta n(x) = \delta n_o \frac{l^2 - x^2}{l^2 - r_o^2}. \quad (24)$$

The cross sectional area across which the grain-boundary diffusive flow passes, $A$, for the present three dimensional model is proportional to $x$ as

$$A = 2\pi wx. \quad (25)$$

As we have assumed that the rate of void growth is determined by the grain-boundary diffusion alone, we obtain

$$\frac{\delta V}{\delta t} = \frac{2\pi D_h w}{kT} \left( \sigma - \frac{2\gamma_s \sin \theta}{r_o} \right) \Omega$$
$$\times \frac{1}{[\ln(l/r_o) - (1-r_o^2/l^2)(3-r_o^2/l^2)/4]} \quad (26)$$

for the growth of a void with an axisymmetric equilibrium shape, using eqs. (11), (13)-(15) and (17)-(25).

V. Discussion

1. Comparison with previous studies

The present analysis treats the diffusional growth of grain-boundary voids by considering the change in the overall Gibbs free energy of a material. As mentioned in Introduction, many researchers have investigated this problem by considering the gradient of the chemical potential of an atom in a grain boundary(1)-(9). This chemical potential has been connected with a normal tension acting on the grain boundary by the equation given by Herring(10).

In order to solve differential equations of diffusion for the derivation of the void growth rate, appropriate boundary conditions to estimate the chemical potential field on a grain boundary, as well as a mechanical equilibrium condition, must be assigned. However, different conditions are adopted in previous studies(1)-(9) and, moreover, the effect of the jacking action
is neglected in some studies. As a result, the derived growth rates are also different. In the present analysis, we have derived the void growth rate without knowing the detailed stress field at the grain boundary. Therefore, except for the assumption that matter deposits uniformly on a grain boundary, we need not establish the appropriate boundary conditions and mechanical equilibrium conditions.

Among the previous studies (1)–(9), the void growth rates derived by Hull and Rimmer (1) and by Speight and Harris (2) are inexact, as pointed out in refs. (3) to (5) and (9), since they are based on inappropriate boundary conditions for the chemical potential field at a grain boundary. Although the analyses by Raj and Ashby (5) with corrections (6), by Weertman (3)(4) and by Chuang et al. (8) used the proper boundary conditions, they neglected the effect of the jacking action. The jacking action is a natural consequence of the diffusional growth of grain-boundary voids under the present conditions that the rate of volume diffusion is negligibly small compared with the rate of grain-boundary diffusion and that the voids maintain their equilibrium shapes by rapid matter transport around their surface.

For the above reasons, we believe that the two expressions of the void growth rates, one by Speight and Beere (7) and the other by Needleman and Rice (9), are worthy of discussion. For an axisymmetric void, their results are expressed in our notation as

\[
\frac{\delta V}{\delta t} = 2\pi \frac{D_o w}{kT} \left( \sigma - \frac{2\gamma_S \sin \theta}{r_o} \right) \Omega \\
\times \left[ \ln \left( \frac{l}{r_o} \right) - \left( 1 - \frac{r_o^2}{l^2} \right) \frac{3 - r_o^2/l^2}{4} \right] \right)
\]

(Speight and Beere) (27)

\[
\frac{\delta V}{\delta t} = 2\pi \frac{D_o w}{kT} \left[ \sigma - \left( 1 - \frac{r_o^2}{l^2} \right) \frac{2\gamma_S \sin \theta}{r_o} \right] \Omega \\
\times \left[ \ln \left( \frac{l}{r_o} \right) - \left( 1 - \frac{r_o^2}{l^2} \right) \frac{3 - r_o^2/l^2}{4} \right] \right)
\]

(Needleman and Rice) (28)

By comparing the two expressions, it is found that their results are different only in the stress term by as much as the factor \((1 - r_o^2/l^2)^2\). This difference comes from the adoption of different mechanical equilibrium conditions. In our notation, these are given by

\[
\int_{r_o}^l 2\pi x_0(x) \, dx = \sigma l^2 - 2\pi r_o^2 \gamma_S \sin \theta,
\]

(Speight and Beere) (29)

\[
\int_{r_o}^l 2\pi x_0(x) \, dx = \sigma l^2,
\]

(Needleman and Rice) (30)

where \(\sigma(x)\) is the local normal tension acting on the grain boundary. Here, we have included the factor \(\sin \theta\) for the dihedral angle which was not considered in the original equation by Speight and Beere for a spherical void (eq. (3) in ref. (7)).

Let us discuss these two expressions by energy consideration. The movement of matter during void growth is schematically shown in Fig. 3. Because of the effect of the jacking action, a part (designated as the portion P in Fig. 3) of an increase in the volume of the void occurs without the flow of matter. Here, \(\delta u\) is the
thickness of the uniformly deposited matter at the grain boundary and also is the displacement of the material during the void growth along the loading direction. The work done by the applied stress during the growth of a void, $\delta W$, is given by

$$\delta W = \sigma \delta V. \quad (31)$$

However, this work should also be expressed as,

$$\delta W = \left[ \int_{r_o}^\infty 2\pi x \sigma_n(x) \, dx \right] \delta u + \left[ \frac{4\pi r_o \gamma_s}{\sin \theta} \int_0^\theta \cos \alpha \sin \alpha \, d\alpha \right] \delta u. \quad (32)$$

The term in the second square brackets is the normal component of the surface tension to maintain the equilibrium shape of the void. Thus, the second term expresses the work necessary to open up the part $P$ in Fig. 3.

Using eqs. (31) and (32) together with $\delta V = \pi l^2 \delta u$, we have

$$\int_{r_o}^\infty 2\pi x \sigma_n(x) \, dx = \sigma \pi l^2 - 2\pi r_o \gamma_s \sin \theta. \quad (33)$$

This equation is the same as the mechanical equilibrium condition by Speight and Beere(7), eq. (29). Thus, it is clear that when the effect of the jacking action is included, the mechanical equilibrium condition given by Speight and Beere is correct. Comparing our expression of the void growth rate, eq. (26), with that by Speight and Beere, eq. (27), we find that the two expressions are identical.

In the analysis given by Raj and Ashby with corrections, essentially the same mechanical equilibrium condition, eq. (29), as that employed by Speight and Beere was adopted. However, they have neglected the effect of the jacking action in their later discussion. Therefore, the analysis by Raj and Ashby with the corrections is considered to be theoretically inconsistent.

On the basis of the above discussion, the difference in the growth rate for the cylindrical void between our result, eq. (16), and that by Chuang et al. can also be understood. The rate of the growth for the cylindrical void given by Chuang et al. is written, in our notation, as

$$\frac{\delta V}{\delta t} = 6 \frac{D_bw}{kT} \left[ \sigma \left( 1 - \frac{r_o}{l} \right) \left( \frac{\gamma_s \sin \theta}{r_o} \right) \right] \Omega \times \frac{1}{l(1-r_o/l)^2}. \quad (34)$$

Comparing eqs. (16) and (34), we find that one of the differences lies in the stress term as much as the factor $(1-r_o/l)$ in front of $\gamma_s \sin \theta/r_o$. This factor arises in eq. (34) since Chuang et al. used an incorrect mechanical equilibrium condition similar to that by Needleman and Rice. The other difference is found in the effect of the relative void spacing, the factor $(1-r_o/l)$ in the denominator. The second-power effect of this factor in eq. (34) arises since they neglected the additional increase of the volume of the void by the jacking action.

We should like to conclude this part of the discussion by pointing out that our present simple method can also clearly explain the origins of the differences in the expressions of the void growth rate obtained by various investigators.

2. General remarks

Using the present analysis, we can obtain the approximate growth rates of the grain-boundary voids more easily. For an axisymmetric void (Fig. 2), we can assign, as rough approximations, the average diffusion distance of the grain-boundary diffusion, $\lambda_3$, and the average area, $A_3$, across which the diffusive flow passes as

$$\lambda_3 = (l-r_o)/2 \quad \text{and} \quad A_3 = 2\pi \{(l+r_o)/2\} w. \quad (35)$$

Then, the average thermodynamic force acting on an atom, $f_3$, the average drift velocity of the diffusing atoms, $v_3$, and the continuity of the atom flow are given by

$$f_3 = -\left( \frac{\delta G}{\delta n_3} \right)/\lambda_3, \quad (36)$$

$$v_3 = f_3 D_b/kT, \quad (37)$$

$$\delta n_3 = v_3 A_3 \delta t/\Omega. \quad (38)$$

From eqs. (20), (21) and (35) to (38), we can immediately obtain the approximate growth rate of the grain-boundary voids as

$$\frac{\delta V}{\delta t} = 2\pi \frac{D_bw}{kT} \left( \sigma - \frac{2\gamma_s \sin \theta}{r_o} \right) \Omega \Omega.$$
Although this equation gives a larger growth rate compared with the rigorous one (eq. (26)), particularly for large voids or small void spacings, the difference is not very significant.

It is true that the rate equations examined in the present study too much simplify the actual void growth process. Therefore, it does not seem to be very meaningful to directly compare the present rate equations with experimentally observed void growth rate. Even if we assume that the grain-boundary diffusion is the only mechanism of the void growth, we can still point out the following possibilities. First, if matter does not deposit on a grain boundary in an ideally uniform manner, the elastic energy contribution (8) as well as the elastic interaction among voids must be incorporated in the analysis. Secondly, when the surface diffusion is not rapid enough compared with the grain-boundary diffusion, voids cannot maintain the equilibrium shape and, as they grow, the shape becomes more crack-like (8)(10).

Furthermore, not only diffusion but also other processes such as creep or plastic deformation of a matrix is known to contribute to the void growth (8)(19)-(24). However, we believe that the clarification of the fundamental process of the diffusion-controlled void growth is indispensable to the understanding of high-temperature fracture: Unless this process is clarified, the attempts to develop theories for other void growth mechanisms would be like building castles in the air.

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