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# Swelling modification by one-dimensional diffusion of cascade-produced small interstitial clusters

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## Abstract

The paper deals with the influence of one-dimensional diffusion of small interstitial clusters produced in collision cascades on the swelling of irradiated structural materials. It is demonstrated that the resulting swelling modification is very sensitive to microstructural parameters of irradiated materials. Qualitative trends of swelling modification in well annealed materials correlate with experimental observations; however for cold-worked materials the model predicts noticeable decrease in swelling rate by one-dimensionally diffusing clusters, which is in contrast with the observed material behaviour. © 1998 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Irradiation of structural materials with high energy neutrons typical for fast fission and fusion reactors results in radiation damage, which occurs mainly in the form of collision cascades. As discovered recently in molecular dynamics simulations [1–6], such high-energy cascades produce not only isolated point defects (vacancies and interstitials), but also vacancy and interstitial clusters containing up to 10–20 defects. The creation of such clusters can be straightforwardly incorporated in the classical rate theory of swelling (as formulated in Ref. [7]) in terms of the modification of overall sink strengths (see e.g. [8,9]). For example, when the clusters collapse to faulted (and thus immobile) dislocation loops (which is very often the case for vacancy clusters and is typical for interstitial clusters with more than approximately ten interstitials [10]), the in-cascade cluster creation leads only to the renormalization of the dislocation density.

An interesting feature of the cascade produced damage, disregarded in the above mentioned analytical ap-

proaches, is the mobility of very small interstitial clusters (SICs). As indicated by some computer simulations [1] the clusters with the number of interstitials  $n \leq n^* \sim 4-6$  can perform one-dimensional motion along low-index crystallographic directions, the number  $M$  of equivalent directions being defined by crystal symmetry (e.g.  $M=4$  and  $M=6$  for clusters moving along close packed directions in bcc and fcc lattices, respectively). The mobility of SICs should be appropriately taken into account in the rate theory description of radiation-induced phenomena. In particular, if the mobility of small interstitial clusters is sufficiently high in order to provide the accommodation of their concentration to the evolution of extended sinks, they should be considered as a third kind of mobile defect in addition to vacancies and self-interstitials.

The one-dimensional motion of small interstitial clusters (SIC) is often interpreted in the current literature (see e.g. [11–13]) in terms of the dislocation loop glide. However, the glide, i.e. the directed motion of SICs under the effect of applied stress, can hardly be of importance for microstructural evolution. First of all, glissile loops become energetically favored compared to faulted configurations at much larger loop sizes than those typical for cascade-produced SICs. Moreover, in order to provide the matter transport over noticeable distances

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via the loop glide, the long-scale stress gradients should be acting in the material. Indeed, the glide force  $F$  acting on a small circular loop that is able to glide along, say, the  $x$ -axis of a Cartesian coordinate system can be easily derived from the well known Peach–Koehler formula [14] to be

$$F \simeq -n\Omega \frac{d\sigma_{xx}}{dx}, \quad (1)$$

where  $n$  is the number of interstitials in the loop,  $\Omega$  is the atomic volume and  $\sigma_{xx}$  is the corresponding component of the stress tensor acting in the loop center. Such long-scale gradients can hardly be provided by strongly fluctuating internal stresses.

It seems more reasonable that the movement of SICs occurs via one-dimensional stochastic jumps, being a diffusional process. The aim of the present paper is to investigate how such one-dimensional diffusion of interstitial clusters affects the rate of material swelling under the cascade-forming irradiation. Consequently, among a number of various defects produced in collision cascades we consider only those mobile, namely, isolated point defects and SICs. Larger cascade-produced interstitial clusters (with  $n > n^*$ ) provide a negligible modification of the total dislocation density and are not considered here. Additionally, we neglect vacancy clusters (loops and stacking fault tetrahedra) forming during the cascade cooling down, which is a reasonable approximation at not too short irradiation doses, when the steady-state size distribution of dissolving vacancy clusters has already been established. Correspondingly, all vacancies surviving the in-cascade recombination are mobile and thus the estimate of the SIC effect on swelling, as obtained below, is the maximum one.

In Section 2 we formulate a set of rate theory equations that take into account the presence of one-dimensionally diffusing small interstitial clusters, while in Section 3 the sink strengths are specified. The rate of void growth and the swelling kinetics modification due to SIC generation are discussed in Section 4.

## 2. Formulation of the problem

Let us assume that cascade-produced small interstitial clusters (consisting of  $2 \leq n < n^*$  interstitials) can perform one-dimensional diffusional motion along a set of  $M$  crystallographically equivalent directions with corresponding diffusion coefficients  $D_n$ , the latter being dependent on the SIC size  $n$ , but not on its particular orientation  $m$  ( $1 \leq m \leq M$ ). Then the mean-field concentration  $C_n^m$  of SICs of a chosen orientation  $m$  can be described by one-dimensional rate equation

$$G_n^m + I_n^m - (k_n^m)^2 D_n C_n^m + \sum_{m' \neq m} (R_{mm'} C_n^{m'} - R_{m'm} C_n^m) = 0, \quad (2)$$

where  $G_n^m$  is the generation rate of SICs of size  $n$  and orientation  $m$ ,  $R_{mm'}$  is the rate of on-site “rotation” of  $m'$ -type clusters to the  $m$ -type clusters and  $I_n^m$  is the rate of cluster creation due to the interaction of SICs with point defects,

$$I_n^m = L_{(n-1)i} D_{(n-1)i} C_{n-1}^m C_i + L_{(n+1)v} D_{(n+1)v} C_{n+1}^m C_v - (L_{ni} D_{ni} C_i + L_{nv} D_{nv} C_v) C_n^m, \quad (3)$$

$D_{\alpha\beta} = D_\alpha + D_\beta$  is a sum of diffusion coefficients and  $L_{\alpha\beta}$  is the recombination factor for corresponding defects  $\alpha$  and  $\beta$  ( $\alpha, \beta = i, v, n$ , where  $i$  denotes interstitials,  $v$  the vacancies and  $n$  the SICs of size  $n$ ). In writing down Eq. (2) we neglect the creation of SICs due to the dissolution of larger dislocation loops, which means that  $L_{(n^*+1)v} \equiv 0$ .

In the case of isotropic (or cubic) materials that will be discussed below we can additionally assume that the sink efficiencies are the same for all orientations of SICs and equal to  $k_n^2$  for  $n$ -atom cluster absorption (i.e.  $(k_n^m)^2 = k_n^2$  for all  $m$ ). Then one can describe SICs in terms of their overall concentration

$$C_n = \sum_{m=1}^M C_n^m, \quad (4)$$

which is determined by an equation obtained via summation of Eq. (2) for all SIC orientations:

$$G_n + I_n - k_n^2 D_n C_n = 0, \quad (5)$$

where  $G_n = \sum_m G_n^m$  and  $I_n = \sum_m I_n^m$ . Note that in the latter equation all terms accounting for the on-site SIC rotation have cancelled each other and the one-dimensional nature of the diffusion process is kept only in  $k_n^2$ .

The rate equations for the mean-field concentrations  $C_v$  and  $C_i$  of three-dimensionally diffusing vacancies and interstitials are only slightly modified by one-dimensionally diffusing SICs,

$$G_\alpha - k_\alpha^2 D_\alpha C_\alpha - L_{iv} D_{iv} C_i C_v - \sum_{n=2}^{n^*} L_{n\alpha} D_{n\alpha} C_n C_\alpha = 0 \quad (\alpha = i, v), \quad (6)$$

where  $G_\alpha$  is the generation rate,  $D_\alpha$  the diffusion coefficient,  $k_\alpha^2$  the sink strengths for  $\alpha$ -type point defects ( $\alpha = i, v$ ), respectively. In writing down Eq. (6) it is assumed that the irradiation temperature is sufficiently low in order to neglect the thermal evaporation of point defects from SICs and extended sinks.

The number of vacancies created by irradiation is exactly equal to the number of displaced atoms, implying the relation

$$G_v = G_i + \sum_{n=2}^{n^*} n G_n. \quad (7)$$

### 3. The efficiencies of defect absorption by extended sinks

In order to determine the defect concentrations from the equations of Section 2, one has to specify for all moving defects the sink strengths  $k_x^2$ . For three-dimensionally diffusing defects these are calculated as sums of strengths of different sink types present in the material [7]. Usually the principal sink types governing vacancy and interstitial loss are voids and dislocations (including loops), which have the well-known strengths, namely,

$$k_{V_x}^2 = 4\pi Y_x R N_V, \quad (8)$$

$$k_{D_x}^2 = Z_x \rho_D, \quad (9)$$

where  $N_V$  is the number density of voids,  $R$  the average void radius,  $\rho_D$  the dislocation density,  $Y_x$  and  $Z_x$  ( $x = i, v$ ) the bias factors for voids and dislocations, respectively. Correspondingly,  $k_x^2 = k_{V_x}^2 + k_{D_x}^2$ .

On the other hand, the sink strengths for SICs depend very much on the general nature of SIC diffusion, as determined by the ease of SIC on-site rotation. When the distance covered by a SIC between two consecutive on-site rotations,  $l_R$ , is noticeably shorter than the typical inter-sink spacing,  $l_S$ , SIC diffusion can be considered as essentially three-dimensional and the SIC sink strengths will be given by equations similar to Eqs. (8) and (9). In other words, in this case the incorporation of SICs into consideration will result in minor quantitative modification of the swelling rate, but the qualitative trends of the swelling behavior remain unaltered. On the contrary, in the opposite limiting case of  $l_R \gg l_S$ , the functional dependence of the SIC sink strength on the sink parameters is changed and the swelling behavior becomes qualitatively different from that in the absence of SICs. Therefore, in what follows we restrict ourselves to the case when the on-site rotations of SICs are neglected and the clusters move strictly one-dimensionally.

In the limiting case adopted we may use a general expression for the sink strengths for one-dimensionally diffusing defects [15]:

$$k_n^2 = 2\lambda_n^{-2}, \quad (10)$$

where

$$\lambda_n = \left( \sum_S \sigma_{nS} N_S \right)^{-1}, \quad (11)$$

$N_S$  is the volume concentration of sinks of the type  $S$ ,  $\sigma_{nS}$  is the sink cross-section for capture of clusters of size  $n$ , factor 2 in Eq. (10) takes into account that the defects are captured from two sides of a sink and summation in Eq. (11) is over the different sink types. Note that although  $\lambda_n$  in Eq. (10) has the physical meaning of a screening length of the sink ensemble [16,17], it is defined by the same relation as the defect mean free path before the capture by sinks, as predicted from the particle beam scattering analogy [12,18].

The summation in Eq. (11) for a particular case when the considered sinks include spherical voids of capture radius  $R_n$ , dislocations with the capture radius  $r_n$  (the latter may noticeably exceed the dislocation core radius  $r_D$  if the capture of defects is affected by their elastic interaction with dislocations [12]) and planar grain boundaries gives [15]

$$k_n^2 = 2(\pi N_V R_n^2 + 2\rho_D r_n + \gamma_G d_G^{-1})^2, \quad (12)$$

where  $d_G$  is the average grain size, and the factor  $\gamma_G \sim 1$  accounts for the geometry of grains. An estimate for the application relevant values of dislocation density  $\rho_D \approx 10^{12} - 10^{14} \text{ m}^{-2}$  and  $d_G \approx 10 \text{ }\mu\text{m}$  shows that for any reasonable value of  $r_n$  ( $\sim 1 - 10 \text{ nm}$ ) dislocations are much weaker sinks for one-dimensionally diffusing defects than grain boundaries. On the contrary, voids with typical densities  $N_V \approx 10^{19} - 10^{21} \text{ m}^{-3}$  become quite comparable in strength to grain boundaries when the void size reaches  $R_n \approx 10 - 100 \text{ nm}$ .

### 4. Void growth rate and swelling

The growth rate of a void with radius  $R$  due to the absorption of vacancies, interstitials and SICs is given by

$$\frac{dR}{dt} = \frac{1}{R} (Y_v D_v C_v - Y_i D_i C_i) - \frac{1}{4\pi R^2} \sum_{n=2}^{n^*} n J_n, \quad (13)$$

where the first two terms on the r.h.s. of Eq. (13) represent the vacancy and interstitial currents to the void and  $J_n$  is the current of  $n$ -atomic SICs, defined as [15]

$$J_n = 2\pi R^2 \frac{D_n C_n}{\lambda_n}. \quad (14)$$

To simplify the analysis, we assume below  $Y_i = Y_v = 1$ , since the deviations of void bias factors from unity are important only for void sizes smaller than 10–20 nm [19].

The mean-field concentrations of point defects and SICs are to be defined from the set of equations presented in Section 2, which is complicated and does not allow simple analytical solution. In order to follow qualitative trends of the SIC diffusion influence on the swelling kinetics, we neglect in the rate equations all terms of the second order in defect concentrations. This assumption is reasonable at temperatures close to that of the peak swelling. Then the concentrations of defects are found easily and the void growth rate is given by

$$\frac{dR}{dt} = \frac{1}{R} \left( \frac{G_v}{k_v^2} - \frac{G_i}{k_i^2} \right) - \frac{1}{4} \sum_{n=2}^{n^*} n G_n \lambda_n. \quad (15)$$

Since the one-dimensionally diffusing clusters are small compared to the linear dimensions of sinks, we can assume that the SIC capture by sinks is insensitive

to cluster sizes and thus  $\lambda_n$  for all  $n$  is equal to a certain value  $\lambda_s$ , which is defined by Eq. (12) with the SIC capture radii of voids and dislocations being set equal to their geometrical sizes. After substitution of Eqs. (8)–(11) into Eq. (15), the latter can be rewritten as

$$\frac{dR}{dG_v t} = \frac{(Z_i - Z_v)\rho_D}{Rk_v^2 k_i^2} + \varepsilon \frac{(4k_{As} - Z_i \rho_D R)\lambda_s}{4Rk_i^2}, \quad (16)$$

where  $\varepsilon = \sum_n nG_n/G_v$  is the relative portion of in-cascade interstitials agglomerated in SICs and  $k_{As} = 2\gamma_D \rho_D r_D + \gamma_G d_G^{-1}$  is the inverse of the SIC mean free path defined by all sinks excluding voids. It is immediately seen that the first term on the r.h.s. of Eq. (16) gives the rate of void growth in the absence of SIC production, while the second term defines the correction due to SIC creation in collision cascades. In order to estimate its contribution, it is convenient to introduce non-dimensional values for void radius,  $r = R/R_c$ , and irradiation dose,  $\phi = G_v t / \Phi_c$ , where  $R_c = Z_i \rho_D / 4\pi N_V$  is the void radius at which the sink strength of voids for point defects is equal to that of dislocations and  $\Phi_c = (Z_i \rho_D)^3 / (4\pi N_V)^2$  is the characteristic dose of void growth. For typical parameter values summarized in Table 1, the characteristic void radius and irradiation dose vary from  $R_c \sim 250$  nm and  $\Phi_c \sim 10^{-5}$  dpa in well annealed materials ( $\rho_D \sim 10^{12}$  m $^{-2}$ ) to  $R_c \sim 2$  nm and  $\Phi_c \sim 10$  dpa in cold-worked materials ( $\rho_D \sim 10^{14}$  m $^{-2}$ ).

Using the non-dimensional variables, we may reduce Eq. (16) to

$$\frac{dr}{d\phi} = \frac{dr}{d\phi} \Big|_{\varepsilon=0} \left( 1 + \frac{\varepsilon}{B} \Delta \right), \quad (17)$$

where  $B = 1 - (Z_v/Z_i)$  is the dislocation bias, the void growth rate in the absence of SICs is given by

$$\frac{dr}{d\phi} \Big|_{\varepsilon=0} = \frac{B}{r(r+1)(r+1-B)} \quad (18)$$

and

$$\Delta = \frac{(r_s - r)(r + 1 - B)}{(r^2 + r_s)}, \quad (19)$$

where

$$r_s = \frac{16\pi N_V k_{As}}{(Z_i \rho_D)^2} \approx \frac{16\pi \gamma_G N_V}{Z_i^2 \rho_D^2 d_G} \approx 5 \frac{N_V}{\rho_D^2 d_G}. \quad (20)$$

Table 1

Parameters used for numerical estimates

Parameter	Value
Void number density $N_V$ , m $^{-3}$	$10^{20}$
Average grain size $d_G$ , $\mu$ m	10
Dislocation bias $B$	0.25
Dislocation bias factor for interstitials, $Z_i$	3
Cascade efficiency, $\xi$	0.1

At the relevant parameter values,  $r_s$  varies in a broad range of  $10^{-2}$ – $10^3$ .

The radius dependence of the correction term  $\Delta$  is shown in Fig. 1. It can be seen that the sign of the correction term  $\Delta$  is determined by the unique parameter  $r_s$ , which defines the ‘transition’ void size  $R_s = r_s R_c$ , at which the ratios ‘void sink strength/alternative sink strength’ are the same for interstitials and SICs. At the void radii smaller than  $R_s$  the presence of SICs accelerates the void growth, whereas at  $R > R_s$  the growth of voids is decelerated. An estimate with the parameters from Table 1 predicts  $R_s \sim 100$  nm at  $\rho_D \sim 10^{12}$  m $^{-2}$  and  $R_s \sim 1$  nm at  $\rho_D \sim 10^{14}$  m $^{-2}$ , indicating quite different effect of SICs on void kinetics in annealed and cold-worked materials. As low dislocation densities quite noticeable increase in the void growth rate is expected at initial stages of irradiation (while  $R < R_s$ ). On the contrary, in cold worked materials the value of  $R_s$  is comparatively small and the dominant trend is the slowing down of the void growth.

The swelling rate per unit dose  $dS/d\Phi$  (where  $\Phi$  is the NRT-standard value, defined according to  $G_v t = \xi \Phi$  with the cascade efficiency  $\xi \approx 0.1$  [13]) is given by the relation

$$\frac{dS}{d\Phi} = 4\pi N R^2 \frac{dR}{d\Phi} = \xi r^2 \frac{dr}{d\phi}. \quad (21)$$

The dose dependencies of  $S$  predicted by Eq. (21) are shown in Figs. 2–4 for different values of  $\rho_D$  and  $\varepsilon$  (other relevant parameters are from Table 1). Integration of Eq. (21) is performed under simplifying assumption that all voids appear simultaneously at a certain incubation dose  $\Phi_i$ , i.e. the initial condition for the void radius is taken in the form  $R(\Phi_i) = 0$ .

In well-annealed materials the presence of one-dimensionally migrating clusters results in quite noticeable acceleration of swelling, see Fig. 2 (corresponding to  $\rho_D \sim 10^{12}$  m $^{-2}$  and  $r_s \approx 55$ ). This result is immediately

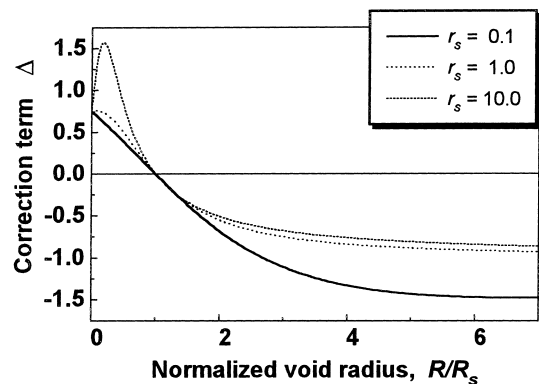


Fig. 1. Correction term  $\Delta$  as a function of the void radius  $R$  (normalized per  $R_s$ ).

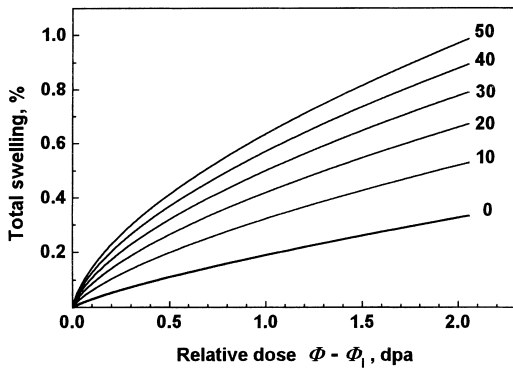


Fig. 2. The swelling kinetics in an annealed material ( $\rho_D = 10^{12} \text{ m}^{-2}$ ) at different relative portion of in-cascade clustered interstitials,  $\varepsilon$ . The values of  $\varepsilon$  (in percents) are indicated at corresponding curves.

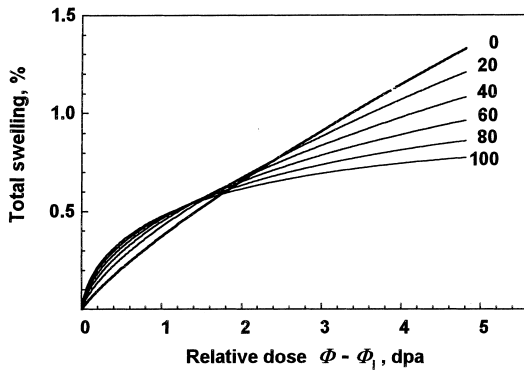


Fig. 3. Same as Fig. 2, but for  $\rho_D = 3 \times 10^{12} \text{ m}^{-2}$ .

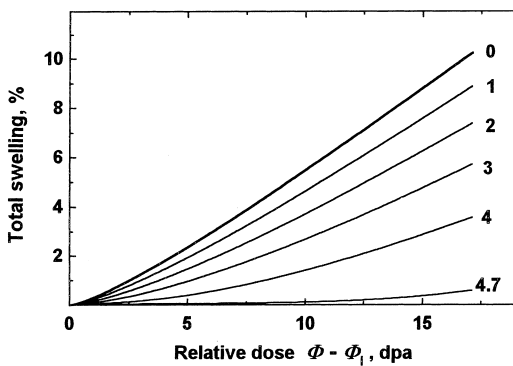


Fig. 4. Same as Fig. 2, but for a cold-worked material ( $\rho_D = 10^{14} \text{ m}^{-2}$ ).

related to the fact that  $R_s$  in such materials is sufficiently high and the voids of any reasonable size fall into the region of accelerated void growth, as indicated in Fig. 1.

At higher dislocation densities the transition void size decreases to the values that can be achieved by growing

voids during reasonable irradiation time. In such situations, exemplified in Fig. 3, the swelling kinetics are more complicated. Like in the case of well-annealed materials, the swelling is initially accelerated, but this acceleration is only transient. At sufficiently high irradiation doses the presence of SICs suppresses swelling. Moreover, when the portion of in-cascade clustered interstitials is sufficiently high, a clear tendency to swelling saturation at high doses is observed. The behaviour results from the fact that at  $r > r_s$  the correction term  $\Delta$  in Eq. (17) becomes negative and at  $\varepsilon > B$  the presence of SICs stops the void growth at the radius  $r_{\text{max}}$  defined from the equation

$$-\Delta(r = r_{\text{max}}) = \frac{B}{\varepsilon}. \quad (22)$$

With even higher dislocation densities the region of accelerated swelling becomes progressively shorter and in cold-worked materials the suppression of swelling dominates, this suppression being manifested in considerable decrease of the steady-state swelling rate. As can be seen in Fig. 4, at sufficiently high dislocation densities only several percent of interstitials clustered in SICs can totally suppress swelling.

We thus see, that the effect of one-dimensionally migrating SICs on swelling is not straightforward and is utterly sensitive to material parameters. In annealed materials the swelling behaviour in the presence of SICs is in quite nice qualitative agreement with experimental observations [20]. However, the predicted swelling behaviour in cold-worked materials seems to be unrealistic. Even though preirradiation cold-working can in principal suppress swelling, such suppression is primarily manifested in the delay of the swelling onset, rather than in the decrease of the steady-state swelling rate [21].

It is interesting to note that only several percent of interstitials clustered in SICs is sufficient to practically suppress swelling in a cold-worked material. According to the already cited results of MD simulations of SIC production in cascades, even under very conservative assumptions one can expect that the portion of interstitials clustered in cascades exceeds those several percent of interstitials that are necessary to practically suppress swelling in a cold-worked material. It seems, therefore, that it is the one-dimensional nature of SIC movement that requires serious justification in order to be applied to the explanation of physical effects in irradiated materials (see e.g. [11,13]).

## 5. Conclusions

1. The rate theory of swelling is extended in order to account for the one-dimensional diffusion of small interstitial clusters created in collision cascades.

2. It is demonstrated that the correction to the swelling rate is directly proportional to the fraction of SICs clustered in cascades. The sign of the correction (and correspondingly the acceleration or slowing down of the void growth rate) is governed by a unique parameter – the transition radius  $R_s$  defined by Eq. (20).
3. Depending on the microstructural parameters, one-dimensionally diffusing SICs can either accelerate or slow down the swelling. In particular, in well-annealed materials an increase of the swelling is expected in accordance with experimental observations, while in cold-worked materials the presence of SICs is predicted to suppress swelling. The mode of swelling suppression (the decrease of the swelling rate) is, however, different from that observed experimentally (the delay of swelling initiation).
4. The disagreement between the predicted effect of one-dimensionally diffusing SICs on swelling and the experimentally observed swelling behaviour in cold-worked materials indicates the necessity of thorough justification of the one-dimensional nature of SIC mobility.

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