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A three-scale homogenisation approach to the prediction of long-time absorption of radiation induced interstitials by nanovoids at interfaces



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ABSTRACT

Various nanoscale materials that contain high density of interfaces highlight an optimistic perspective of discovering radiation tolerant materials. However, given the huge dimensional contrast between the core structural components of nuclear reactors and many nanoscale treatments for improving materials radiation resistance, the corresponding predictive models are required to possess a delicate balance between resolution and efficiency. Motivated by this, a three-scale homogenisation scheme is introduced in this article, and a continuum model for the long-time interstitial-sink behaviour at interfaces is derived with all important nanoscopic parameters and mechanisms properly retained. Compared with the existing works alike, the derived model shows its advantage in at least two aspects. First, it incorporates the collective effect of multiple sinks on interstitial migration which is not fully taken into account in conventional works, and the accuracy of the continuum description to the underlying mechanisms is thus improved substantially. Second, the derived model naturally formulates a sink saturation condition under which sinks no longer absorb point defects. The present work originates from developing a long-time predictive model for a recent proposal for improving the radiation tolerance of materials by Chen et al. (2015), and the derived three-scale homogenisation approach can be naturally generalised to model the collective behaviour of other types of sink-defect interactions.

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1. Introduction

Radiation induced damage in the core structural materials that are made of crystalline materials leads to the creation of microstructural/crystal defects (Rosinski et al., 2001). Conceptually, these defects can be visualised as regions where there is either a deficiency of lattice atoms (voids, vacancies, edge dislocations, vacancy type dislocation loops) or an excess of lattice atoms (self interstitial atoms, interstitial type dislocation loops). All these defects lead to changes in the mechanical properties of the material (Konings et al., 2015; Rosinski et al., 2001). For example, radiation induced swelling (by vacancies and voids) causes dimensional distortion and embrittlement, and is a life-limiting issue for structural materials in nuclear

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power reactors. Austenitic steels were used in fast reactors but could not reliably serve beyond 150 d.p.a. (displacement per atom) (Gelles, 1984). Ferritic and ferritic-martensitic (FM) steels have been found to swell much less than austentitic steels (Pasebani, 2014). Nano-structuring of both austenitic and FM steels appears to be a promising avenue for further improvement of swelling resistance because of high density of interfaces, providing such structures are stable under ion irradiation (Little and Stow, 1979).

Interfaces (interphase boundaries and grain boundaries) have been demonstrated to be ideal sinks that can absorb radiation induced defects and a platform that can accelerate annihilation of radiation induced interstitials and vacancies (Beyerlein et al., 2015). Interfaces between the metal matrix and nanoscale oxides in oxide dispersion strengthened (ODS) steels systems prove to benefit swelling resistance and creep resistance (Ginley and Kahen, 2012). In metallic nanolaminates, interfaces including twist grain boundary in Au (Di et al., 2011), twin boundary in Cu (Chen et al., 2015; Li et al., 2013a, 2013b), and interphase interfaces in Cu/Nb (Bringa et al., 2012; Demkowicz et al., 2008; Han et al., 2012; Misra et al., 2007), Cu/V (Fu et al., 2013), Fe/W (Zhang et al., 2012) etc., have shown strong defect sink strength and the ability to suppress helium bubble formation. Molecular dynamics (MD) simulations reveal that the sink strength is proportional to the amount of free volume created by interfaces (Bai et al., 2010; Chen et al., 2016; Shao et al., 2015, 2013). Recently, a treatment by deliberately introducing nanovoids on the grain boundaries (GBs) of nanotwinned metals is found effective in regard to extending the lifetime of irradiated materials (Chen et al., 2015). The idea behind can be briefed as follows. Under irradiation, pairs of interstitials and vacancies are generated, and interstitial atoms diffuse faster into grain boundaries. In the presence of interface dislocation networks that are implemented on both coherent/semicoherent twin boundaries, the interstitial atoms arriving at the GBs are quickly directed into nanovoids, which delays the formation interstitial loops inside grains. With regard to such a treatment that has proved to work experimentally, a naturally-arising question is, how to choose (within a manufacturable range) the nanoscopic design parameters (e.g. nanovoid radius, nanovoid spacing etc.), so as to maintain the interstitial absorption rate by nanovoids at a high level for a given period. Here the term GB interstitial is used to denote the interstitial atoms migrating in the vicinity of GBs. Trying to answer this question with molecular dynamical or *ab*-initial calculations is almost computationally intractable when the observation period spans for seconds or longer. In fact, even at a coarser-scale where GB interstitial concentration is the quantity of interest, the resulting computational intensity is still too high to afford, mainly due to the following two reasons. First, in order to resolve nanovoids, the evolution equation for GB interstitial concentration has to be discretised with nanoscale mesh grids. Second, the problem is actually of free-boundary type as the nanovoid sizes keep shrinking. Therefore, we still highly desire a model to formulate the aforementioned GB interstitial - nanovoid dynamics, and the model should possess a good trade-off between resolution and efficiency. To be more specific, on one hand, the model can be used for predicting the long-time behaviour of GB interstitial concentration, while on the other hand, all important nanoscopic parameters and mechanisms should be properly retained in the predictive model. One effective way to reconcile this dilemma is to homogenise the underlying nanoscopic mechanisms so as to derive a model where nanovoids are treated as a sink continuum to GB interstitial atoms.

The key to the derivation of such a continuum model is to develop a scheme which effectively "upscales" the considerably large GB interstitial concentration gradient in the vicinity of the densely distributed nanovoids. The most widely-used approach for upscaling nanoscale interactions between point defects (e.g. vacancies and interstitials) and sinks (e.g. dislocations, voids etc.) follows the pioneering works by Mansur, Yoo and other contemporary researchers (e.g. Olander, 1976; Yoo and Mansur, 1976; Mansur and Yoo, 1978; Mansur, 1978). Although the (two-dimensional) problem studied here displays a strong similarity with the mechanism of vacancy diffusion among dislocation "forests", that has been investigated by Mansur and other researchers, there are essential differences lying between the presented work and the conventional ones (such as Mansur and coworkers'), and they are summarised in two aspects as follows. Firstly, the conventional models are established based on a (microscopic) situation where the interstitial diffusion around isolated sinks is considered. In this scenario, the effect of neighbouring sinks on the migration of point defects gets fully neglected. From an asymptotic viewpoint, the conventional treatment provides a leading-order approximation to the underlying mechanism, and the approximation is supposed to be sufficiently accurate provided that the ratio of inter-sink spacing to sink size is large. However, such an approximation is not always accurate. It will be shown in Section 5.1 that the omission of higher-order terms amounts to a deviation of 30%, even when the sink spacing-to-size ratio reaches as large as 100. The second issue that limits the conventional models being used for the present study is that they do not fully take into account the role played by the inert gas in bubbles. It is widely agreed that the migration of point defects deviates from pure diffusion given a hydrostatic pressure gradient in materials. Due to the presence of inert gas, a hydrostatic pressure gradient is generated in the neighbourhood of a nanovoid, which counters the "diffusive force". As a result, sinks become "saturated" and shade themselves from GB interstitial migration. A proper formulation of the conditions under which sinks get saturated is important for designing relevant radiation resistant materials. However, such saturation states are not a natural outcome of the conventional models where the gas pressure is simply included to vary the equilibrium concentration of point defects on sink surfaces, while its effect on the distribution of the hydrostatic pressure field surrounding nanovoids is still ignored. Therefore, a continuum model that properly summarises the underlying mechanism governing the GB interstitial-nanovoid interaction is still highly desired

For this purpose, a three-scale homogenisation approach is proposed in this article, and the method can be envisaged as a revamped version of the traditional multiscale treatment (e.g. see Pavliotis and Stuart, 2008), where two distinguished length scales are concerned. If the traditional two-scale treatment is adopted to homogenise the above-mentioned nanovoid problem, the fast scale would likely be chosen as the length scale associated with nanovoid spacing, while the slow scale

Table 1 Three cases studied in this paper.

	Case I	Case II	Case III
Interstitial type	Self-interstitial	Self-interstitial	Inert gas
Elements in voids	Vacant	Inert gas	Inert gas
Governing equations	(5)–(12)	(5)–(12)	(5), (6), (13)

would be associated with the size of structural materials. As a result, the information about individual nanovoids, such as their size, gets smeared out in the two-scale homogenisation formulation. To overcome this difficulty, an extra length scale associated with nanovoid size is introduced. With use of matched asymptotic techniques, all important (nanoscale) parameters and mechanisms are properly maintained in the homogenised model.

The present work is expected to provide an initial (and also relatively successful) attempt towards the integration of nanoscale models to a timescale that is useful for engineering applications. With the proposed homogenised formulation, the effect of nanoscale design on the long-time behaviour of GB interstitial concentration can be quantitatively studied. It is worth noting that the derived formulation effectively covers the following three different types of interstitial dynamics on grain boundaries. In the first case, nanovoids are vacant and GB interstitial atoms are of self-interstitial type. In the second case, nanovoids become bubbles filled with inert gas (Di et al., 2011). In the third case, GB interstitials are helium solute atoms. In the two latter cases, the inert gas in bubbles generates an extra hydrostatic pressure field to the system, which resists further GB interstitial inflow towards nanovoids. With the derived homogenised model, a condition governing the onset of sink saturation to GB interstitial atoms is derived. We remark that the focus of the present work is to demonstrate the effectiveness of the three-scale homogenisation approach, and to apply it to the investigation of the long-time behaviour of GB interstitial concentration in the presence of nanovoids at interface. Hence the dynamic processes regarding generation, annihilation, and diffusion of radiation induced defects inside grains are simply integrated into a source term and will be systematically formulated in forthcoming works. It is also noted that given the similarities between the mechanism investigated here and the vacancy-dislocation mechanism widely studied elsewhere (e.g. Mordehai et al., 2008; Keralavarma et al., 2012; Ayas et al., 2014; Gu et al., 2015; Huang and Li, 2015; Gu et al., 2017; Rovelli et al., 2017), the proposed threescale homogenisation scheme can be generalised to improve the existing continuum model of dislocation incorporating vacancy diffusion that is deduced using the conventional approach for upscaling defect-sink interactions (Geers et al., 2014).

The rest of the article is arranged as follows. After setting up the problem at the nanoscale level in Section 2, the threescale homogenisation approach is introduced so as to derive the coarse-grained formulation in Section 3. In Section 4, the model is further simplified to consist of a system of ordinary differential equations (ODEs), and simulation results based on the derived ODEs are compared with experimental data by Chen et al. (2015). In Section 5, discussion will be conducted in the following three aspects: comparisons of the sink capture efficiency formulated here with that in conventional works, the derivation of a sink saturation condition and searching for an optimal solution for nanovoid implementation at interfaces. The article concludes in Section 6.

2. Dynamics on nanoscales

In the present study, three cases will be studied as summarised in Table 1:

- Case I. GB interstitial atoms are of self-interstitial type, and nanovoids are vacant.
- Case II. GB interstitial atoms are of self-interstitial type, but nanovoids become bubbles filled with inert gas, e.g. helium. Case III. Both GB interstitial atoms and the elements inside nanovoids are inert gas atoms.

These three types of problems can be studied under a unified set-up as shown schematically in Fig. 1. The interface, say, a grain boundary, is set to be the x - y plane (z = 0) and is denoted by Γ . On the grain boundary, nanovoids are distributed with a density of $\rho(x, y)$ (of unit m⁻²). Here we assume ρ takes a smooth profile. This means that the spacing between two neighbouring nanovoids can be approximated by $1/\sqrt{\rho(x, y)}$. In the present study, all nanovoids are assumed to take spherical shapes, and the (three-dimensional) region occupied by the kth nanovoid is denoted by Ω_{nv}^k .

$$\Omega_{\rm nv}^k = \{ (x, y, z) | \sqrt{(x - x_k)^2 + (y - y_k)^2 + z^2} \le r_k \},\tag{1}$$

where $(x_k, y_k, 0)$ is the coordinate of the kth nanovoid center and r_k denotes the kth nanovoid radius. Here we further assume that r_k also varies smoothly in space, i.e. one can find a smooth function $r_{nv}(x, y)$, such that

$$r_k = r_{\rm nv}(x_k, y_k). \tag{2}$$

Here the slow-varying assumption on r_{nv} etc. is reasonable, because the experimentally implemented nanovoids usually locate at the intersections of the (often regularly distributed) interface dislocation networks on coherent/semicoherent twin boundaries (Shao et al., 2013).

In case II and III, nanovoids become bubbles filled with helium gas. If the number of helium atoms within the kth bubble is denoted by N_{He}^k , we introduce another smooth field variable N_{He} , such that

$$N_{\rm He}^k = N_{\rm He}(x_k, y_k). \tag{3}$$



Fig. 1. A family of nanovoids of density ρ (of unit m⁻²) is distributed on a flat grain boundary of size *L* and denoted by Γ . The nanovoids are sinks to GB interstitial atoms deposited from grain interiors. All nanovoids are assumed to take spherical shapes with the *k*th nanovoid centered at (x_k , y_k) and of radius r_k . Three distinguished length scales are concerned, and they are characterised by r_{nv} , $1/\sqrt{\rho}$ and *L*, respectively. The length-scale parameters satisfy the hierarchic relation given by Eq. (14).

Let c(x, y) denote the average GB interstitial concentration in a vicinity of a GB point (x, y). The GB interstitial flux is then expressed by Hirth and Lothe (1982)

$$\mathbf{J} = -D_{\mathrm{i}}\nabla c - \frac{D_{\mathrm{i}}c\Delta V_{\mathrm{i}}}{k_{\mathrm{B}}T}\nabla p,\tag{4}$$

where D_i denotes the diffusivity coefficient of GB interstitial atoms; k_B is the Boltzmann constant; T is temperature; ΔV_i represents the dilatation volume of generating a GB interstitial site; p is the hydrostatic pressure. The first term of Eq. (4) arises from first Fick's law, which accounts for GB interstitial diffusion. The second term of Eq. (4) formulates the interaction between GB interstitial atoms and the elastic stress field existing in materials. It should be noted that given the interface dislocation networks on GBs which act as fast channels for GB interstitial flux towards nanovoids, the actual GB interstitial diffusion is (microscopically) anisotropic. However, for simplicity, we still use an isotropically defined D_i , which properly integrates the fast-channelling effect caused by interface dislocations on GBs.

With Eq. (4), the evolution of fractional GB interstitial concentration is formulated by

$$\frac{\partial c}{\partial t} = -\nabla \cdot \mathbf{J} + s = D_i \nabla \cdot \left(c + \frac{c \Delta V_i}{k_{\rm B} T} \nabla p \right) + s,\tag{5}$$

on Γ outside nanovoids, where s (of unit s⁻¹) is a source term accounting for interstitial deposition from grain interiors. On nanovoid boundaries, we let

$$c|_{\partial\Omega_{\nu}\cap\Gamma} = 0. \tag{6}$$

For both case I and II, Eq. (6) means that a self-interstitial atom gets crystalised on nanovoid surfaces, while for case III it means that the solute helium atoms take their fluidic phase as they enter the bubbles. It is noted that, for case I or II, the crystalline formation energy of atoms on nanovoid surface can also be taken into account, but the boundary condition needs to be re-written in that scenario. To derive a closed system, we also need to determine the expressions for the hydrostatic pressure p and the source term s.

The hydrostatic pressure field within materials is related to the internal elastic stress field σ by its first stress invariant (Balluffi et al., 2005):

$$p = -\frac{\sigma_{11} + \sigma_{22} + \sigma_{33}}{3}.$$
(7)

It is shown in Appendix A that the hydrostatic pressure field should satisfy the three-dimensional Laplacian equation:

$$\frac{\partial^2 p}{\partial x^2} + \frac{\partial^2 p}{\partial y^2} + \frac{\partial^2 p}{\partial z^2} = 0$$
(8)

for linearly elastic and isotropic materials. In order to fully determine p in a specimen containing nanovoids, the boundary conditions on nanovoid surfaces are also needed. The pressure exerted on the material in the vicinity of a nanovoid, which is denoted by p_0 , is given by

$$p_0 = p|_{\partial \Omega_{nv}^k} = p_{\rm g} - \frac{2\gamma}{r_{\rm nv}},\tag{9}$$

where p_g is the pressure due to the inert gas in bubbles and γ is the nanovoid surface energy density. It is remarked that similar as the definition of r_{nv} in Eq. (2), both p_0 and p_g in Eq. (9) are defined to be smoothly-varying field variables in space, and their evaluations at point (x_k , y_k) return the values of the corresponding physical quantities associated with the

*k*th bubble. The gas pressure p_g should be a function of nanovoid radius r_{nv} and the number of helium atoms within N_{He} . For example, p_g can be expressed by using the ideal gas formula:

$$p_{\rm g} = \frac{3k_{\rm B}TN_{\rm He}}{4\pi \,(r_{\rm nv})^3},\tag{10}$$

when N_{He} falls below a certain value.

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In theory, the internal hydrostatic pressure field p can be computed by (numerically) solving Eq. (8) along with boundary condition (9) (and the macroscopic loading conditions). However, it will be seen later that, direct computation for p is too time-consuming. Hence extra treatment is needed so as to make resulting simulations fall in a computationally tractable range.

The source term *s* appearing in Eq. (5) should equal the rate of the total number of interstitial atoms deposited from grain interiors, multiplied by the area occupied by a single interstitial site S_i and divided by the total area of the grain boundaries. In theory, the interstitial deposition rate on grain boundaries should depend on the behaviour of interstitials and vacancies inside grains. Here for better addressing the main point of the article, we assume that the interstitial deposition rate from grain interiors is a constant, and all grains take an identical cuboid shape. Hence when the defects are of self-interstitial type (case I and II), the interstitial deposition rate from grain interiors is given by $dN_0P_1I_0^3$ (Was., 2017), where d is the d.p.a. rate; N_0 is the number of atoms per volume in perfect matrix crystals; P_i is the probability of a displaced atom that finally arrives at the grain boundaries. Since each cuboid facet is shared by two grains, the surface area assigned to each grain is actually $3I_0^2$. Therefore,

$$s = \frac{dN_0P_1S_1l_0}{3}.$$
(11)

The expression for *s* in case III can be obtained through comparing with experimental data or small-scale simulation results, and this will not be discussed in full details here.

Now we write down the evolution equations for nanovoid size r_k and the number of helium atoms in the *k*th nanovoid N_{He}^k . For case I and II, as nanovoids keep absorbing GB self-interstitial atoms, their sizes shrink accordingly. The shrinkage in nanovoid volume divided by the volume occupied a single atom (of the structural materials) should equal the number of incoming GB self-interstitial atoms, i.e.,

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{4\pi (r_k)^3}{3} = \frac{1}{N_0 S_\mathrm{i}} \int_{\partial \Gamma_\mathrm{nv}^k} \mathbf{J} \cdot \mathbf{n} \mathrm{d}s,\tag{12}$$

where N_0 is recalled to be the atom number per volume in perfect lattice and S_i is recalled to represent the area of a GB self-interstitial atom occupies on the grain boundary Γ . For case III, nanovoid sizes stay unchanged but the number of helium atoms within bubbles increases, i.e.

$$\frac{dN_{\text{He}}^{k}}{dt} = \frac{1}{S_{\text{i}}} \int_{\partial \Gamma_{\text{nv}}^{k}} \mathbf{J} \cdot \mathbf{n} ds.$$
(13)

The governing equations for the three cases are summarised in Table 1.

In principle, one can evolve the above-listed equations so as to predict the long-time behaviour of the GB interstitial concentration in irradiated materials. Practically, however, direct simulations of the aforementioned equation system are difficult. This is because there are actually three distinguished length scales involved, which is respectively characterised by the nanovoid radius r_{nv} , nanovoid spacing $1/\sqrt{\rho}$ and grain size *L*. In practice, irradiated material GB should not be too "hollow" and this suggests $\sqrt{\rho}r_{nv} \ll 1$. Moreover, the number of nanovoids is often very large and this gives rise to $\sqrt{\rho}L \gg 1$. Hence a hierarchic relation is established as follows

$$r_{\rm nv} \ll \frac{1}{\sqrt{\rho}} \ll L. \tag{14}$$

This means that the numerical discretisation of Eqs. (5)-(13) has to be fine enough in order to resolve nanovoids. If the simulation is conducted on a length scale that is far larger than nanoscales, the resulting computational cost is too huge to afford. Moreover, since nanovoid boundaries may also evolve in time, the problem considered is actually of free-boundary type, which further brings up the computational cost. Therefore, a homogenised description of the presented evolution system is still highly desired. On one hand, it can be used for predicting the long-time behaviour of GB interstitial concentration. On the other hand, important nanoscopic information has to be retained at the coarse-grained level.

3. Homogenisation

3.1. Scale separation

The homogenisation tactics adopted in this work can be summarised in Fig. 2. Nanovoids display various profiles when measured on different length scales. On nanoscales (characterised by r_{nv}), isolated nanovoids are fully resolved. When viewed on a scale of tens of nanometers (characterised by $1/\sqrt{\rho}$), nanovoids are so small that they are considered as locally



Fig. 2. The three regimes are characterised by nanovoid radius r_{nv} , nanovoid spacing $1/\sqrt{\rho}$ and material size *L*, respectively. After non-dimensionalisation (by following Eq. (18)), the regime measured in unit of $\delta\epsilon$ is named as the inner region, where isolated nanovoids are fully resolved. When zoomed out to the intermediate region measured in unit of ϵ , nanovoids are so small that they can be treated as locally periodic point sinks to GB interstitial atoms. When zoomed out even further to the outer region measured in unit of 1, nanovoids look like a sink continuum to interstitials.

periodic point sinks to interstitials. If zoomed out even further to microscales or above (characterised by *L*), the nanovoid family can be treated as a continuous distribution of interstitial sinks. In this work, the behaviour of solutions to the model listed in Section 2 will firstly be analysed on the three length scales individually. Then the information collected on various scales is connected by using the matched asymptotic techniques. For simplicity, we will first look for the governing homogenised equations near the origin. The global representation of the homogenised system is then obtained by changing the inputs of all field variables from the origin to the points of interest. We remark that the idea behind the derived three-scale homogenisation scheme originates from the work by Chapman et al. (2015), where the continuum limit of the shielding effect by Faraday cages is studied.

3.2. Expression for hydrostatic pressure

Before carrying out multiscale analysis, we first look for an expression for hydrostatic pressure p, which should satisfy Eq. (8) with boundary condition (9). Noted that if viewed on the length scale associated with L, p displays a highly-oscillatory profile because of the densely distributed nanovoids. Here we refer to the analytical results by Chapman et al. (2016) to assume that p (near the origin) can be decomposed into a mean-field pressure field p_m and a (fast-varying) locally periodic pressure field p_s :

$$p = p_{\rm m} + p_{\rm s}.\tag{15}$$

Here p_m measures the hydrostatic pressure field (near the origin) due to the external load and faraway nanovoids, and it is smoothly varying. The fast oscillatory feature originally displayed by the hydrostatic pressure field is then captured by the short-range pressure field p_s , which is a periodic function of period $1/\sqrt{\rho}$. Based on the aforementioned decomposition, pnear the origin can be approximated within a small square cell of size $1/\sqrt{\rho}$ by a periodic p_s added by a locally constant p_m (due to its smoothly-varying feature).

For the expression for p_s , one only needs to look for the solution to the three-dimensional Laplacian Eq. (8) in a periodic square with a nanovoid centered at the origin. On nanovoid surface, we let

$$p_{\rm s}|_{\partial\Omega_{\rm nv}} = p_0 - p_{\rm m},\tag{16}$$

where p_0 is given by Eq. (9). The mean-field hydrostatic pressure field p_m can be computed with a global finite element scheme. Since the inclusion of p_m is a trivial task, we let $p_m = 0$ for further analysis. Hence Eq. (16) becomes $p_s|_{\partial\Omega_{nv}} = p_0$. Although the full expression for p_s may not be available, as suggested by a detailed calculation in Appendix B, p_s should look like

$$p_{\rm s} = \frac{p_0 r_{\rm nv}}{\sqrt{x^2 + y^2}} \cdot g(\sqrt{\rho}x, \sqrt{\rho}y),\tag{17}$$

where $g(s_1, s_2)$ is a non-dimensional and bounded function. It is also seen from Appendix B that $p_s \sim \frac{p_0 r_{nv}}{\sqrt{x^2 + y^2}}$ as $\sqrt{x^2 + y^2} \rightarrow 0$. The aforementioned information for p_s is sufficient for further multiscale calculations.

3.3. Multiscale analysis

3.3.1. Nondimensionalisation

To facilitate further multiscale analysis, we non-dimensionalise by

$$\bar{x} = \frac{x}{L}, \quad \bar{y} = \frac{y}{L}, \quad \bar{t} = t \cdot \frac{D_{\rm i}}{L^2}, \quad \bar{s} = s \cdot \frac{L^2}{D_{\rm i}}, \quad \bar{p}_{\rm s} = \frac{p_{\rm s}}{p_0},$$
(18)

where a bar is added to a variable to denote its non-dimensional counterpart. Note that there is no need to non-dimensionalise the fractional concentration *c*.

The non-dimensional version of Eq. (5) is

$$\frac{\partial c}{\partial \bar{t}} - \bar{\nabla} \cdot \left(\bar{\nabla} c + \beta c \bar{\nabla} \bar{p}_{s} \right) = \bar{s}, \tag{19}$$

where $\bar{\nabla}$ denotes taking gradient with respect to (\bar{x}, \bar{y}) and the non-dimensional parameter β satisfies

$$\beta = \frac{p_0 \Delta V_i}{k_B T}.$$
(20)

For better identifying the three regimes shown in Fig. 2, two small parameters are introduced:

$$\delta = \sqrt{\rho} r_{\rm nv} \ll 1, \qquad \epsilon = \frac{1}{\sqrt{\rho}L} \ll 1. \tag{21}$$

In the non-dimensional space, the three regimes characterised by r_{nv} , $1/\sqrt{\rho}$ and L are corresponded to an inner region measured in unit of $\delta\epsilon$, an intermediate region measured in ϵ and an outer region measured in 1, respectively, as shown in Fig. 2.

Noted that the hydrostatic pressure field is expressed by

$$\bar{p}_{s} = \frac{\delta\epsilon}{\sqrt{\bar{x}^{2} + \bar{y}^{2}}} \cdot g\left(\frac{\bar{x}}{\epsilon}, \frac{\bar{y}}{\epsilon}\right)$$
(22)

in the non-dimensional space.

Now we will take turn to analyse Eq. (19) in the aforementioned three regimes shown. Then the homogenised equation is derived by matching the information across different regimes.

3.3.2. Inner region

In the inner region, we rescale by

$$\xi = \frac{\bar{x}}{\delta\epsilon}, \quad \eta = \frac{\bar{y}}{\delta\epsilon}.$$
(23)

The GB interstitial concentration can thus be represented by $c := c_{in}(\xi, \eta)$. It is noted that c_{in} should also depend on the original slowly-varying variables (\bar{x}, \bar{y}) , if higher-order effects are also taken into account. However, it is posteriorly found that investigating the leading-order effect of c in the inner region is sufficient for obtaining the homogenised formulation. In the inner region, Eq. (22) reads

$$\bar{p}_{s} = \frac{1}{\sqrt{\xi^{2} + \eta^{2}}} \cdot g(\delta, \delta).$$
(24)

Since $\lim_{(x,y)\to(0,0)} g(x,y) = 1$ (due to Eq. (B.8) in Appendix B), we obtain

$$\bar{p}_{\rm s} \sim \frac{1}{\sqrt{\xi^2 + \eta^2}} \tag{25}$$

to the leading order.

Inserting Eq. (25) into (19) and replacing the spatial variables by (ξ, η) , we have

$$\frac{\partial c_{\rm in}}{\partial \bar{t}} - \frac{1}{\delta^2 \epsilon^2} \cdot \left(\tilde{\nabla}^2 c_{\rm in} + \beta \tilde{\nabla} \cdot \left(c_{\rm in} \tilde{\nabla} \left(\frac{1}{\sqrt{\xi^2 + \eta^2}} \right) \right) \right) = \bar{s}, \tag{26}$$

where the differentiation chain rule is used and " $\tilde{\nabla}$ " represents taking gradient with respect to (ξ , η). We remark that there is no need to rescale time, because the evolution process taking place on a timescale of \bar{t}/ϵ or above does not appear in the final homogenised formulation. Taking the leading-order term of Eq. (26) gives the governing equation for c_{in} :

$$\tilde{\nabla}^2 c_{\rm in} + \beta \tilde{\nabla} \cdot \left(c_{\rm in} \tilde{\nabla} \left(\frac{1}{\sqrt{\xi^2 + \eta^2}} \right) \right) = 0.$$
⁽²⁷⁾

In the inner region, the boundary condition (6) is re-written by

$$c_{\rm in}|_{\rho=1} = 0,$$
 (28)

where $\rho = \sqrt{\xi^2 + \eta^2}$. One way to solve Eq. (27) along with condition (28), is to express the solution as a series expansion in terms of β . Thus one write

$$c_{\rm in} = A \cdot (\log \rho + h(\rho; \beta)), \tag{29}$$

where *A* is a constant to be determined. The expansion form of $h(\varrho; \beta)$ is detailed in Appendix C. It is noted that only the information about c_{in} as $\varrho \to \infty$ is of use when being matched with the solution from the intermediate region. Also as detailed in Appendix C, the behaviour of c_{in} at infinity is approximately given by

$$c_{\rm in} \sim A \cdot \left(\log \sqrt{\xi^2 + \eta^2} + \frac{\beta}{4 - \beta} \right),\tag{30}$$

as $\varrho \to \infty$.

3.3.3. Intermediate region

When zoomed out from the inner region, nanovoids degenerate to a family of locally periodic point sinks to GB interstitials. The local periodicity is due to the fast-varying feature of nanovoids (when viewed on a length scale of O(1) in the non-dimensional space). In this scenario, the effect of nanovoids to the GB interstitial concentration (near the origin) can be incorporated into Eq. (5) by the sum of a family of periodically centered Dirac- δ functions:

$$\frac{\partial c}{\partial \bar{t}} - \bar{\nabla} \cdot \left(c + \beta c \bar{\nabla} \bar{p}_{s} \right) = \bar{s} - \sum_{i, j \in \mathbb{Z}} B_{1} \delta_{d} (\bar{x} - i\epsilon) \delta_{d} (\bar{y} - j\epsilon), \tag{31}$$

where a subscript "d" is added to a Dirac function so as to distinguish from the small parameter δ defined in Eq. (21); B_1 is a constant (associated with the origin) to be determined.

If still measured in terms of (\bar{x}, \bar{y}) , the GB interstitial concentration *c* behaves highly oscillatory. To cope with such a high oscillation, we follow the general multiple-scale analysis procedure to introduce a pair of intermediate-scale variables

$$(X,Y) = \left(\frac{\bar{x}}{\epsilon}, \frac{\bar{y}}{\epsilon}\right).$$
(32)

Then the GB interstitial concentration can be approximated by a function $c_m(\bar{x}, \bar{y}; X, Y)$, which is assumed periodic over X and Y with period 1. By doing this, the fast-varying feature of c is captured in terms of X and Y, while the slow-varying feature (on the length scale associated with O(1)) is still described in terms of \bar{x} and \bar{y} . Thus variations taking place on different length scales get separated with use of the chain rule:

$$\frac{\partial}{\partial \bar{x}} = \frac{\partial}{\partial \bar{x}} + \frac{1}{\epsilon} \frac{\partial}{\partial X}, \quad \frac{\partial}{\partial \bar{y}} = \frac{\partial}{\partial \bar{y}} + \frac{1}{\epsilon} \frac{\partial}{\partial Y}.$$
(33)

Here the book by Pavliotis and Stuart (2008) is referred to if one is interested in getting more knowledge about the traditional multiscale techniques.

The hydrostatic pressure \bar{p}_s given by Eq. (22) now becomes

$$\bar{p}_{\rm s} = \delta \cdot \frac{g(X,Y)}{\sqrt{X^2 + Y^2}}.\tag{34}$$

Eq. (34) suggests that the effect due to \bar{p}_s away from (X, Y) = (0, 0) (~ $\mathcal{O}(\delta)$) is weak compared to GB interstitial diffusion (~ $\mathcal{O}(1)$). Near (X, Y) = (0, 0), the singularities of \bar{p}_s has been taken into account by the undetermined coefficient B_1 of the family of Dirac- δ functions in Eq. (31). Therefore, if only the leading order effect is of interest, the hydrostatic pressure term \bar{p}_s can be neglected on the intermediate scale. Thus under the multiple-scale framework, Eq. (31) is written by

$$\frac{\partial c_{\rm m}}{\partial \bar{t}} - \bar{\nabla}^2 c_{\rm m} - \frac{1}{\epsilon^2} \left(\frac{\partial^2 c_{\rm m}}{\partial X^2} + \frac{\partial^2 c_{\rm m}}{\partial Y^2} \right) = \bar{s} - \frac{B_1}{\epsilon^2} \cdot \delta(X) \delta(Y), \tag{35}$$

with $(X, Y) \in \Gamma_m = [-1/2, 1/2] \times [-1/2, 1/2].$

Keeping the leading-order terms of Eq. (35) gives the governing equation in the intermediate region:

$$\frac{\partial^2 c_{\rm m}}{\partial X^2} + \frac{\partial^2 c_{\rm m}}{\partial Y^2} = B_1 \delta_{\rm d}(X) \delta_{\rm d}(Y) \tag{36}$$

with c_m periodic in Γ_m . The general solution to Eq. (36) can be expressed by

$$c_{\rm m} = \frac{B_1}{2\pi} \cdot \left(\log \sqrt{X^2 + Y^2} + \chi(X, Y) \right) + B_2, \tag{37}$$

where B_2 is another coefficient to be determined and the function $\chi(X, Y)$ is the solution to a cell problem defined by

$$\left(\frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2}\right)\chi(X, Y) = 0,$$
(38)

with

$$\frac{\partial \chi}{\partial X}\Big|_{X=\pm\frac{1}{2}} = \mp \frac{2}{1+4Y^2}, \quad \frac{\partial \chi}{\partial Y}\Big|_{Y=\pm\frac{1}{2}} = \mp \frac{2}{4X^2+1}.$$
(39)

In order to uniquely determine χ , we let

$$\lim_{\sqrt{X^2 + Y^2} \to 0} \chi(X, Y) = 0.$$
(40)

Although the explicit form of χ is not available, it will be shown later that the numerical solution to Eqs. (38)–(40) is sufficient for further analysis.

3.3.4. Outer region

When zoomed out from the intermediate scale, individual nanovoids will no longer be resolved. What is seen is a sink continuum to GB interstitial atoms. Actually, the macroscopic GB interstitial concentration should equal the intermediate solution c_m averaged over its associated periodic cell Γ_m :

$$c(\bar{x},\bar{y}) = \frac{1}{|\Gamma_{\rm m}|} \int_{\Gamma_{\rm m}} c_{\rm m}(\bar{x},\bar{y};X,Y) dXdY.$$
(41)

Therefore, by integrating Eq. (35) over Γ_m and using (41), the homogenised equation for *c* is obtained:

$$\frac{\partial c}{\partial \bar{t}} - \bar{\nabla}^2 c = \bar{s} - \frac{B_1}{\epsilon^2}.$$
(42)

3.3.5. Matching between solutions from various regimes

In order to fully determine the homogenised equation for *c*, one needs to eliminate B_1 in Eq. (42). This can be achieved by inserting Eq. (37) into (41):

$$c(\bar{x},\bar{y}) = \frac{B_1}{2\pi |\Gamma_{\mathrm{m}}|} \cdot \left(\int_{\Gamma_{\mathrm{m}}} \log \sqrt{X^2 + Y^2} \mathrm{d}X \mathrm{d}Y + \int_{\Gamma_{\mathrm{m}}} \chi(X,Y) \mathrm{d}X \mathrm{d}Y \right) + B_2.$$
(43)

Based on Eqs. (38)-(40), it can be numerically calculated that

$$\int_{\Gamma_{\rm m}} \chi(X,Y) dX dY \approx -0.2515. \tag{44}$$

Combining this with the fact that $\int_{\Gamma_m} \log \sqrt{X^2 + Y^2} dX dY \approx -1.0612$ and $|\Gamma_m| = 1$, we obtain

$$c(\bar{x},\bar{y}) = -\frac{k_0 B_1}{2\pi} + B_2,\tag{45}$$

where $k_0 \approx 1.3127$. Now another coefficient B_2 is introduced in Eq. (45). To determine it, asymptotic matching between the inner and intermediate solutions is performed. The underlying idea is that the solution of the inner problem at infinity should agree with the solution of the intermediate problem at the origin.

The behaviour of c_{in} as $\sqrt{\xi^2 + \eta^2} \to \infty$ has been given by Eq. (30), and it can be re-written in terms of the intermediate variable (*X*, *Y*) by

$$c_{\rm in} \sim A \cdot \left(\log \sqrt{X^2 + Y^2} - \log \delta + \frac{4\beta}{4 - \beta} \right),\tag{46}$$

where $(\xi, \eta) = (\delta X, \delta Y)$ is used. Eq. (46) is then matched with the behaviour of c_m given by Eq. (37) as $\sqrt{X^2 + Y^2} \rightarrow 0$:

$$c_{\rm m} \sim \frac{B_1}{2\pi} \cdot \log \sqrt{X^2 + Y^2} + B_2,$$
(47)

where Eq. (40) has been used. A comparison between Eqs. (46) and (47) suggests

$$B_1 = 2\pi A, \quad B_2 = -A\log\delta. \tag{48}$$

Inserting Eq. (48) into (45) gives

$$B_1 = \frac{2\pi c}{\log \frac{1}{\delta} + \frac{4\beta}{4-\beta} - k_0}$$
(49)

Finally, replacing B_1 in Eq. (42) gives the homogenised equation for the GB interstitial concentration on grain boundaries in the non-dimensional space:

$$\frac{\partial c}{\partial \bar{t}} - \bar{\nabla}^2 c = \bar{s} - \frac{2\pi}{\epsilon^2} \cdot \frac{c}{\log \frac{1}{\delta} + \frac{4\beta}{4-\beta} - k_0}.$$
(50)

3.4. Homogenised system

Now we re-dimensionalise Eq. (50) and obtain

$$\frac{\partial c}{\partial t} - D_i \nabla^2 c = s - \rho D_i \omega_0 \cdot c, \tag{51}$$

where

$$\omega_0 \triangleq \frac{2\pi}{\log\frac{1}{r_{\rm nv}\sqrt{\rho}} + \frac{4p_0\Delta V_i}{4k_{\rm B}T - p_0\Delta V_i} - k_0} \tag{52}$$

and the expression of β by Eq. (20) is used to derive Eq. (52). In theory, Eq. (51) holds near the origin. Actually, it is trivial to generalise the method used in Section 3 to any spatial point (*x*, *y*). Thus Eq. (51) can be used as a homogenised description of the GB interstitial motion throughout the whole region Γ .

Then we derive the evolution equations for nanovoid radius r_{nv} (in case I and II) and the helium number per bubble N_{He} (in case III). It is noted that the last term of Eq. (51) has the physical meaning of GB interstitial absorption rate (by nanovoids) per area. Hence the GB interstitial absorption rate by a single nanovoid (near the point of interest, say, (x, y)) is calculated by the last term of Eq. (51) divided by ρS_i , where S_i is recalled to be the grain boundary area occupied by a single GB interstitial atom.

In case I and II, interstitial absorption results in a shrinkage in nanovoid volume:

$$N_0 \cdot \frac{\partial}{\partial t} \left(\frac{4\pi}{3} r_{\rm nv}^3 \right) = -\frac{D_i \omega_0}{S_i} \cdot c, \tag{53}$$

where N_0 is recalled to be the number of atoms per volume of perfect crystals. Rearranging Eq. (53) gives the governing equation for the nanovoid radius (near the spatial point (*x*, *y*)):

$$\frac{\partial r_{\rm nv}}{\partial t} = -\frac{D_{\rm i}\omega_0}{4\pi N_0 S_{\rm i}} \cdot \frac{c}{r_{\rm nv}^2}.$$
(54)

In case III, GB interstitial absorption results in an increase in the number of helium atoms per bubble:

$$\frac{\partial N_{\text{He}}}{\partial t} = \frac{D_{i}\omega_{0}}{S_{i}} \cdot c.$$
(55)

The above system of partial differential equations (PDEs) is closed if an empirical formula relating p_g to r_{nv} and N_{He} is included. In the simulation results presented in this article, the ideal gas formula (given by Eq. (9)) is employed to express p_g .

Compared to the equation system listed in Section 2, the coarse-grained PDE system obtained here can be numerically solved with mesh grid on microscales or above, while useful nanoscopic information has been incorporated in ω_0 defined by Eq. (52). It is worth noting that during the derivation of the continuum model, the asymptotic behaviour of the underlying mechanism is considered until $\mathcal{O}(1)$. This is in contrast with the conventional works (e.g. Olander, 1976; Yoo and Mansur, 1976) where only the leading-order approximation (up to $\mathcal{O}(\log \delta)$) is employed. A more detailed comparison between the present model and conventional ones will be made in Section 5.1.

4. A predictive model for system long-time behaviour

4.1. Governing equations

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The efficiency of the homogenised formulation (in terms of computational time) can be further improved, if only the system overall effect is concerned. In that scenario, we let all quantities be spatially homogeneous. Here the term "homogeneous" is employed in a macroscopic sense, i.e. all field quantities, such as nanovoid density and GB interstitial concentration, are spatially invariant, while the microscopic heterogeneities induced by the presence of nanovoids have been properly taken into account with use of the three-scale homogenisation scheme. Hence the presented model is essentially different from some of the conventional homogeneous models where GB interstitial concentration gradients are completely ignored. Now *c* measures the overall fractional GB interstitial concentration, and so on for r_{nv} and N_{He} . By letting all spatial derivatives in Eq. (51) vanish, we have

$$\frac{\mathrm{d}c}{\mathrm{d}t} = s - \rho D_{\mathrm{i}}\omega_0 \cdot c. \tag{56}$$

For simulations of either case I or II, Eq. (56) is coupled with

$$\frac{\mathrm{d}r_{\mathrm{nv}}}{\mathrm{d}t} = -\frac{D_{\mathrm{i}}\omega_{\mathrm{0}}}{4\pi N_{\mathrm{0}}S_{\mathrm{i}}} \cdot \frac{c}{r_{\mathrm{nv}}^{2}},\tag{57}$$

which is from Eq. (54); while for case III, Eq. (56) is coupled with

$$\frac{\mathrm{d}N_{\mathrm{He}}}{\mathrm{d}t} = \frac{D_{\mathrm{i}}\omega_{\mathrm{0}}}{S_{\mathrm{i}}} \cdot c,\tag{58}$$

which is from Eq. (55).



Fig. 3. Comparison with the in-situ experimental data by Chen et al. (2015). The experimental measurement is made between the two vertical dashed lines. Both of the key phenomena addressed by Chen et al. (2015) are well captured by derived homogenised model: i) the rate of shrinkage is faster for small voids; ii) an accelerated collapse of nanovoids is widely observed, when void sizes fall below \sim 3 nm (as marked out by grey color). In fact, these phenomena can be well rationalised by the derived model.

In this coarse-grained dynamical system, all important sub-continuum parameters and mechanisms are well retained. Hence the ordinary differential equation system (56)–(58) can be employed to predict the long-time behaviour of GB interstitial concentration under various irradiation conditions and nanoscopic design parameters.

We remark that compared to the PDE system listed in Section 3.4, the derived ODE system is more computationally efficient at the expense of a loss in resolution. How to trade-off between resolution and efficiency should depend on the problem of interest. For example, if the spatial distribution of nanovoids are far from being uniform, the PDE representation should be used. In the present work, the ODE description is adopted for further analysis.

4.2. Comparison with experimental data

Now we refer to the in-situ experimental observation made by Chen et al. (2015), where the evolution of nanovoid sizes starting with different values is recorded as irradiation proceeds. The experimental data are shown by the discrete dots in Fig. 3, and they are compared with the simulation results based on the derived ODEs. Eqs. (56) and (57) are used for computation with $N_{\text{He}} = 0$ (case I), and other parameters are evaluated as follows: $\rho = 10^{-4} \text{ nm}^{-2}$, $l_0 = 50 \text{ nm}$, T = 500 K, $d = 3 \times 10^{-3} \text{ d.p.a./s}$ (all taken or estimated from Chen et al., 2015); $N_0 = 62.5/\text{nm}^3$ (by following conventions); $S_0 = 0.08 \text{ nm}^2$, $\Delta V_i = 10^{-3} \text{ nm}^3$ (both estimated from Freund and Heinloth, 2002); $\gamma = 41.2 \text{ eV/nm}^2$ (Wasserman and Vermaak, 1970); $D_i = 10^3 \text{ nm}^2/\text{s}$; $P_i = 0.03$. It should be noted that there seem no quantities from literature which display a physical equivalence to the GB interstitial deposition probability P_i and the (two-dimensional) diffusivity D_i (which should also integrate the fast-channelling effect in GB interstitial motion caused by interface dislocation networks). Their values have to be determined through a comparison with experimental data. Albeit the inevitable arbitrariness lying in the determination of parameter values, the derived homogenised model still manages to capture the two key phenomena that are addressed by Chen et al. (2015): i) the rate of shrinkage is faster for small voids; ii) an accelerated collapse of nanovoids is widely observed, when void sizes fall below ~ 3 nm (as marked out by grey color in Fig. 3). In fact, these two phenomena can be well rationalised by use of the derived model. According to Eq. (57), the rate of nanovoid shrinkage satisfies

$$\frac{\mathrm{d}r_{\mathrm{nv}}}{\mathrm{d}t} \propto -\frac{1}{r_{\mathrm{nv}}^2 \left(\log\frac{1}{\sqrt{\rho}r_{\mathrm{nv}}} - \frac{4\gamma\Delta V_i}{2r_{\mathrm{nv}}k_BT + \gamma\Delta V_i} - k_0\right)},\tag{59}$$

where the expression for ω_0 in Eq. (52) has been used. It is easily found that the right side of Eq. (59) is a (negatively) decreasing function of r_{nv} . Thus the rate of shrinkage is smaller for larger voids. It is also observed from Eq. (59) that $dr_{nv}/dt \rightarrow -\infty$ as $r_{nv} \rightarrow 0$, and this explains the onset of the collapse of tiny voids.

We remark that the experimental measurement starts when the irradiation dose reaches ~0.11 d.p.a. (as marked by the left-side vertical dash line in Fig. 3). This may be one reason for the quantitative difference between experimental and simulation results (for large voids). Due to the pre-existing irradiation, the actual initial GB interstitial concentration may not be zero, while our simulations begin with c = 0. Besides, the experimental measurement stops at ~0.26 d.p.a. (as marked by the right-side vertical dash line in Fig. 3), which indicates the experimental irradiation runs for about 100 s. In contrast, the simulations based on the derived ODEs can literally last for any irradiation period with tiny computational cost.



Fig. 4. Evolution of GB interstitial concentration and nanovoid sizes for case I. (a) All parameter values are taken exactly the same as in Section 4.2. The evolution can be divided into three stages. In the first stage, GB interstitial concentration grows at a rate that is almost the same as interstitial deposition from grain interiors. In the second stage, GB interstitial concentration increases at a much lower speed than the deposition rate, as interstitial sinks become active. In the last stage, nanovoids are fully filled. As a result, *c* rises again at a same rate as *s*. (b)–(d) Simulations are also conducted with different combinations of the two indeterminate parameters D_i and *s* (by means of changing P_i).

4.3. Numerical examples

More numerical examples will be presented so as to provide the readers a clearer overview of the system long-time behaviour predicted by using the derived ODE model. For simulation results presented in the rest of the article, the parameter values are chosen the same as that used in Section 4.2 unless specified.

First we consider case I where nanovoids are vacant ($N_{\text{He}} = 0$). In Fig. 4(a), the evolution of GB interstitial concentration and nanovoid size is shown where the parameter values are chosen exactly the same as that used for generating Fig. 3. It is seen that under irradiation, the GB interstitial evolution can be roughly divided into three stages. In the first stage, GB interstitial concentration increases at a rate almost the same as the rate of interstitial deposition from grain interiors. This is because at a low concentration, few GB interstitial atoms are available for nanovoids to absorb. As more interstitial atoms arrive at GBs because of irradiation, GB interstitial absorption becomes comparable to its deposition. Consequently, GB interstitial concentration rises at an almost constant speed that is far less than its deposition rate *s*, and the sinks become active in this stage. When nanovoids are fully filled by incoming GB interstitial atoms ($r_{nv} = 0$), *c* increases again at the same speed as *s*.

Given the uncertainties in the choice of diffusivity D_i and the interstitial source s (by means of P_0) as discussed in Section 4.2, we also simulate the system behaviour with different combinations of D_i and s, as shown in Fig. 4(b)–(d). As from Fig. 4(b), an increase in D_i brings down GB interstitial concentration before the onset of the third stage. Physically, this means an interstitial atom arriving at GB has a high probability of being transferred to nanovoids by interface dislocation networks that correspond to a higher overall diffusivity. Another interesting observation is that the evolution profiles



Fig. 5. Evolution of GB interstitial concentration and nanovoid sizes for case II. When there are helium gas in bubbles, a similar three-stage evolution is seen. The main difference between case I and II is that interstitial sinks get saturated at a finite radius when they are filled with inert gas. In the simulation for case II, $N_{\text{He}} = 2 \times 10^5$.



Fig. 6. Evolution of the concentration of helium solutes on GBs in case III. (a) When there is no external source of helium atoms depositing to GBs, the helium solutes initially resting on GBs are gradually absorbed by the sinks. (b) When there is helium atoms constantly depositing to the GB (e.g. s = 0.01/s), the GB solute concentration drops initially, but the trend is reversed as more helium atoms arrive at the GB. The simulation for both cases start with an initial GB solute concentration c = 0.2, vacant nanovoids ($N_{He} = 0$) and $r_{nv} = 5$.

depicted in Fig. 4(c) and (d) look identical, while the difference is the time span. This can be rationalised by dividing both sides of Eq. (56) by s:

$$\frac{1}{s}\frac{\mathrm{d}c}{\mathrm{d}t} = 1 - \frac{\rho D_{\mathrm{i}}\omega_{\mathrm{0}}}{s} \cdot c. \tag{60}$$

When the time is rescaled with the interstitial deposition rate *s*, the evolution equation remains the same if the nondimensional parameter $\rho D_i \omega_0 / s$ is fixed.

For case II where nanovoids become helium gas bubbles, the system behaviour is depicted in Fig. 5. Here the number of helium atoms per bubble is set to be $N_{\text{He}} = 2 \times 10^5$. Similar as the previous example, the evolution process can also be divided into three stages. The main difference between the two cases is that sinks become saturated at a finite radius r_{nv}^* in case II. In another word, the inert gas in bubbles "shades" them from GB interstitial migration. This arises from the fact that the "diffusive force" is balanced by the hydrostatic pressure gradient induced by helium gas in bubbles. Detailed discussion on how the presence of helium atoms in bubbles affects the long-time behaviour of GB interstitial concentration is made in Section 5.2.

For Case III where GB interstitials become helium solute atoms, we will consider the cases with and without external source of helium atoms. The simulations for both cases start with an initial GB solute concentration c = 0.2, vacant nanovoids ($N_{\text{He}} = 0$) and $r_{\text{nv}} = 5$. When there is no external source of helium atoms arriving at GBs, the helium solute atoms that initially rest in the vicinity of the GB are gradually absorbed by the sinks, as shown in Fig. 6(a). When there are helium atoms constantly depositing to the GB (e.g. s = 0.01/s), the GB solute concentration drops initially, but the trend is



Fig. 7. The relative deviation due to the omission of higher-order terms during the derivation of the sink capture efficiency ω_0 . In conventional continuum models (e.g. Yoo and Mansur, 1976), ω_0 given by Eq. (61) is derived based on a leading-order approximation (up to $O(\ln(1/r_{nv}\sqrt{\rho})))$ to the underlying microscopic model presented in Section 2, if the hydrostatic pressure *p* is ignored. In contrast, the expression for ω_0 given by (52) obtained based on the three-scale homogenisation scheme, also captures the next-order effect (up to O(1)). The relative deviation due to the omission of higher-order terms is as big as 0.3, even when $r_{nv}\sqrt{\rho}$ falls below 0.01. Here $p_0 = 0$.

reversed as more helium atoms arrive at the GB. It is noted that in practice, helium gas often enters a specimen from its one side, and diffuse throughout it. This means it may be more appropriate to study case III with the PDE system derived in Section 3.4, and the topic will be discussed in greater detail elsewhere.

5. Discussion

The discussion will be made from three aspects: comparison of the sink capture efficiency predicted here with conventional results, identification of the conditions governing the onset of sink saturation and solutions for optimally implementing nanovoids on grain boundaries.

5.1. Sink capture efficiency

The derived homogenised model is firstly compared with the existing models alike. In fact, the (two-dimensional) mechanism considered here has a strong similarity with the works by Mansur and co-workers (e.g. Yoo and Mansur, 1976), where the collective behaviour of point defects (such as bulk interstitial atoms and vacancies) diffusing among dislocation forests is formulated. If we follow the same idea used by Mansur and co-workers to model the present problem, the evolution of GB interstitial concentration is still governed by Eq. (56), while the parameter ω_0 , which is termed as the "sink capture efficiency" (Was., 2017), takes a different form (if compared with Eq. (52)):

$$\omega_0 = \frac{2\pi}{\ln\frac{1}{r_{\rm nv}\sqrt{\rho}}}.$$
(61)

From an asymptotic viewpoint, Eq. (61) is obtained based on a leading-order approximation $(\mathcal{O}(\ln(1/r_{nv}\sqrt{\rho})))$ to the microscopic dynamics formulated in Section 2 with the hydrostatic pressure p ignored, and the approximation is supposed to be accurate enough provided $r_{nv}\sqrt{\rho} \ll 1$. Nevertheless, since the logarithm function tends to infinity relatively slowly, the actual deviation due to the omission of higher-order terms ($\mathcal{O}(1)$ and above) is considerably high. This is seen from a comparison between Eq. (61) and the sink capture efficiency given by Eq. (52) which includes the next-order ($\mathcal{O}(1)$) effect. It is found that the (relative) deviation caused by neglecting $\mathcal{O}(1)$ terms amounts to $|k_0/\ln r_{nv}\sqrt{\rho}|$, where k_0 is recalled to be a fixed parameter ($k_0 = 1.3127$). In Fig. 7, the relative deviation (due to the omission of higher-order terms) is plotted against the non-dimensional parameter $r_{nv}\sqrt{\rho}$. It is observed that for the case when the pressure term is ignored (i.e. $p_0 = 0$ in Eq. (52)), the deviation is still as big as 30%, when $r_{nv}\sqrt{\rho}$ falls below 0.01. Therefore, the accuracy of the continuum description of defect-sink interactions can be greatly improved by use of the three-scale homogenisation approach introduced in this article.

5.2. Conditions governing the onset of sink saturation

Another aspect of advantage the derived homogenisation model shows over the existing models alike is that it can be employed to identify the conditions under which sinks become saturated. With reference to Eq. (56), nanovoids should stop absorbing GB interstitial atoms when $\omega_0 = 0$. According to Eq. (52), this indicates $p_0 \Delta V = 4k_BT$. With use of Eq. (9), the



Fig. 8. The value of r_{nv}^* reflects the GB interstitial absorption capability of the corresponding system. A greater r_{nv}^* indicates that it is relatively easy for nanovoids to get saturated. (a) r_{nv}^* against a non-dimensional parameter γ/k_BT ; (b) r_{nv}^* against temperature.

saturation condition is expressed by

$$p_{\rm g}\Delta V_{\rm i} - \frac{2\Delta V_{\rm i}\gamma}{r_{\rm nv}^*} = 4k_{\rm B}T,\tag{62}$$

where r_{nv}^* is saturation radius, that is, the radius at which sinks become saturated. The value of r_{nv}^* reflects the GB interstitial absorption capability of the corresponding system. A greater r_{nv}^* indicates that it is relatively easy for nanovoids to get saturated.

If p_g is further expressed by the ideal gas formula (10), the saturation condition (62) reads

$$\frac{3N_{\text{He}}}{4\pi} \cdot \left(\frac{1}{r_{\text{nv}}^*}\right)^3 - \frac{2\gamma}{k_{\text{B}}T} \cdot \frac{1}{r_{\text{nv}}^*} - \frac{4}{\Delta V_{\text{i}}} = 0.$$
(63)

In Fig. 8(a), r_{nv}^* calculated by using Eq. (63) is plotted against γ/k_BT with the helium number per bubble being 10², 10⁴ and 10⁶. It is seen that an increase in surface energy density γ or a drop in temperature *T* brings down the degree of the GB interstitial absorption capability of the corresponding system. It can also be asserted from Fig. 8(a) that r_{nv}^* grows with N_{He} , because a stronger hydrostatic pressure gradient is induced by more helium atoms per bubble. Given N_{He} , there exits a maximum saturation radius $r_{nv}^* = (\frac{3\Delta V_i N_{\text{He}}}{16\pi})^{\frac{1}{3}}$, which corresponds to $\frac{\gamma}{k_B T} = 0$. In Fig. 8(b), r_{nv}^* is also plotted against temperature *T* under various surface energy density γ .

It is noted that under irradiation, voids in the bulk may also get saturated when they are filled with inert gas, and the method devised here can be generalised to identify the saturation condition for voids in the bulk.

5.3. Optimal nanovoid implementation on grain boundaries

Now we analyse the derived ODE system so as to quantitatively identify how the nanoscopic design parameters (such as the initial r_{nv} , ρ , D_i and N_{He} etc.) affect the long-time behaviour of GB interstitial concentration. As mentioned in Section 1, our goal is to improve the absorption capability of the corresponding nanovoid systems to GB interstitial atoms, for a relatively long period. When nanovoids are implemented on GBs, it can be read from Figs. 4 and 5 that there exists a time interval (stage 1 and 2), when radiation induced interstitial atoms are being absorbed by nanovoids. Here we term this period the "low-concentration" period, and in this subsection, we will look for an optimal solution for nanovoid implementation on GBs so as to extend this low-concentration period.

Here we use a quantity M to denote the GB interstitial absorption rate by nanovoids coming from the last term of Eq. (56):

$$M = \rho D_{i}\omega_{0} = \frac{2\pi \rho D_{i}}{\log \frac{1}{r_{nv}\sqrt{\rho}} + \frac{4p_{0}\Delta V_{i}}{4k_{B}T - p_{0}\Delta V_{i}} - k_{0}},$$
(64)

where the expression for ω_0 in Eq. (52) is used. In order to maintain the interstitial absorption rate at a high level, M should take large values. Then the design problem becomes how to choose ρ , r_{nv} and N_{He} , so as to maximise M. A simple analysis to Eq. (64) shows that the value of M can be brought up by an increase in ρ or r_{nv} , or alternatively by a decrease in N_{He} (by means of p_0). However, the aforementioned design parameters are not in general mutually independent in practice. For example, nanovoids can be introduced by implementing dislocation networks on coherent/semicoherent interfaces



Fig. 9. (a) The quantity *M* defined by Eq. (64) against ρ . If GB nanovoids are introduced by implementing dislocation networks on GBs (Shao et al., 2013), then the total free volume between interfaces per area should be fixed, i.e. ρr_{nv}^3 is a constant. An increase in ρ can effectively increase *M* in this case. Here $p_0 = 0$. (b) Evolution of GB interstitial concentration with various initial nanovoid sizes and ρr_{nv}^3 is fixed to be 0.1nm. All curves converge to a straight line (of slope *s*), as the corresponding low-concentration periods terminate. Here $N_{\text{He}} = 0$.



Fig. 10. Evolution of GB interstitial concentration where nanovoids are filled with helium gas. By following the treatment proposed by Shao et al. (2013) and Chen et al. (2015), the initial ρr_{nv}^3 and p_0 are fixed to be 0.1nm and 14MPa, respectively. The low-concentration period terminates earlier for samples with smaller bubbles.

(Shao et al., 2013). Under this treatment, the total free volume between interfaces per area should be fixed, i.e. ρr_{nv}^3 is a constant. As a result, an increase in r_{nv} inevitably brings down ρ . In order to better trade off between the choice for ρ and r_{nv} , M is plotted against ρ in Fig. 9(a) with ρr_{nv}^3 fixed and $p_0 = 0$. It is observed that a denser nanovoid implementation scheme seems more effective for extending the low-concentration period. In Fig. 9(b), the evolution of GB interstitial concentration is shown under $N_{\text{He}} = 0$ and different initial nanovoid sizes. It is observed that all curves converge to a same straight line (of slope *s*) as the corresponding low-concentration periods terminate. This is because the maximum number of interstitial atoms that can be absorbed by nanovoids per area is already pre-fixed ($\rho r_{nv}^3 = 0.1$ nm).

Then we investigate how the presence of helium atoms in nanovoids quantitatively affects the long-time behaviour of GB self-interstitial concentration. It is noted that when the treatment proposed by Shao et al. (2013) and Chen et al. (2015) is adopted, the initial mean-field hydrostatic pressure p_m generally equals the outside gas pressure. According to Eq. (16), this means p_0 needs to be fixed so as to make meaningful comparisons between specimens with different design parameters. In Fig. 10, the evolution of GB interstitial concentration with various initial bubble sizes is shown, where we require not only $\rho r_{nv}^3 = 0.1$ nm, but also $p_0 = 14$ MPa. It is observed that the low-concentration period terminates earlier for samples with smaller bubbles. This can be rationalised with reference to Eq. (10): provided that the surface effect is stronger for smaller bubbles, more interior initial gas atoms are needed so as to generate a same p_0 . As the bubble shrinks in size, hydrostatic pressure gradient for smaller bubbles grows much faster ($\sim r_{nv}^{-3}$). As a result, systems with initially smaller bubbles saturate earlier.

To summarise, implementing dense dislocation networks on coherent/semicoherent interfaces offers an effective way to raise GB interstitial absorption rate by nanovoids. However, when nanovoids are filled with inert gas, the bubble size should not be too short, as the low-concentration period terminates earlier for systems with smaller bubbles.

6. Conclusion

In this work, a three-scale homogenisation approach is proposed to simulate the long-time behaviour of interstitial concentration on the grain boundaries of irradiated materials embedded with nanovoids. The derived model effectively covers three different scenarios as listed in the beginning of Section 2. With all important nanoscopic design parameters well retained, the derived ODE/PDE system exhibits an excellent balance between efficiency and resolution. Compared with the existing results alike, the derived model incorporates the effect of multiple sinks on interstitial migration, and the accuracy of the resulting continuum description to the underlying mechanisms is substantially improved. Besides, the presented homogenisation result naturally formulates a sink saturation condition under which bubbles shade themselves from GB interstitial migration. With the obtained model, both of the key experimental phenomena reported by Chen et al. (2015) are well rationalised. The long-time behaviour of GB interstitial concentration with various nanoscopic design parameter values is investigated by using the derived model, and optimal solutions for nanovoid implementation on grain boundaries are also discussed.

The successive works of the present study will be conducted along two directions. First, the derived model should be combined with a homogenised description of interstitial-vacancy evolution in grain interiors, so as to derive a more comprehensive model that can be used for prediction of irradiated materials's lifetimes. Second, the three-scale homogenisation approach can be generalised to formulate the collective behaviour of mechanisms that exhibit similarities with the present studies. For example, it can be used to upscale the discrete dislocation dynamics incorporating vacancy diffusion to a continuum level.

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Appendix A

Now we will show that when materials are linearly elastic and isotropic, the hydrostatic pressure p related to stress field σ by Eq. (7) satisfies the three-dimensional Laplacian equation.

Hooke's law reads

$$\sigma_{ij} = \lambda \delta_{ij} u_{k,k} + \mu(u_{i,j} + u_{j,i}), \qquad i, j = 1, 2, 3, \tag{65}$$

where the Einstein summation is employed; $u_{i,j}$ denotes the derivative of u_i with respect to its *j*th entry; λ and μ are two elastic constants; δ_{ij} is the Kronecker delta. Hence the first stress invariant is given by

$$\operatorname{tr}(\boldsymbol{\sigma}) = \sigma_{ii} = (3\lambda + 2\mu)u_{i,i}.$$
(66)

Combining Eq. (65) and the force equilibrium equation ($\sigma_{ii,i} = 0$) gives the Navier–Cauchy equation:

$$\mu u_{i,jj} + (\lambda + \mu) u_{j,ij} = 0, \qquad i = 1, 2, 3.$$
(67)

Taking divergence on both sides of Eq. (67) gives

 $(\lambda + 2\mu)u_{i,ijj} = 0.$

A comparison between Eqs. (66) and (68) indicates

 $\nabla^2 \mathrm{tr}(\boldsymbol{\sigma}) = \mathbf{0},$

which immediately leads to the equation for hydrostatic pressure, which is Eq. (8).

Appendix B

We now look for an expression for the hydrostatic pressure due to a family of nanovoids of identical radius r_{nv} periodically distributed on $\{(i/\sqrt{\rho}, j/\sqrt{\rho})|i, j \in \mathbb{Z}\}$, i.e. p_s in Eq. (15). First, we look for the pressure field due to an individual nanovoid located at the origin in \mathbb{R}^3 which is denoted by p_{ind} . Since p_{ind} satisfies the three-dimensional Laplacian equation, along with the boundary condition $p|_{\partial\Omega_{nv}} = p_0$, we obtain

$$p_{\rm ind} = \frac{p_0 r_k}{\sqrt{x^2 + y^2 + z^2}}.$$
(B.1)

As we are only interested in the pressure field at interfaces, we let z = 0.

(68)

(69)

Y. Zhu et al./Journal of the Mechanics and Physics of Solids 105 (2017) 1-20

 Table B1

 Truncation error of the finite sum given by Eq. (B.4).

Truncation error	$ A_5-A_1 $	$ A_{10} - A_5 $	$ A_{50} - A_{10} $
(0.1, 0) (0.1, 0.1) (0.25, 0.25) (0.3, 0.1) (0.5, 0.5)	$\begin{array}{c} 3.9\times 10^{-18} \\ 6.2\times 10^{-3} \\ 0.039 \\ 0.0188 \\ 0.15 \end{array}$	$\begin{array}{c} 3.3\times10^{-19}\\ 1.1\times10^{-4}\\ 6.8\times10^{-4}\\ 3.3\times10^{-4}\\ 2.7\times10^{-3} \end{array}$	$\begin{array}{c} 1.3 \times 10^{-19} \\ 1.8 \times 10^{-5} \\ 1.1 \times 10^{-5} \\ 5.4 \times 10^{-5} \\ 4.5 \times 10^{-4} \end{array}$
(, ,			

In order to obtain a locally periodic expression for p_s (of period $1/\sqrt{\rho}$), one can simply sum all repeated images of p_{ind} :

$$p_{\rm s} = p_0 \sqrt{\rho} r_{\rm nv} \cdot g_0(\sqrt{\rho} x, \sqrt{\rho} y), \tag{B.2}$$

where the (non-dimensional) function $g(s_1, s_2)$ is defined by means of the finite part of a divergent series:

$$g_0(s_1, s_2) = \text{f.p.}\left(\sum_{i, j \in \mathbb{Z}} \frac{1}{\sqrt{(s_1 - i)^2 + (s_2 - j)^2}}\right).$$
(B.3)

In order to express g_0 , we consider a family of finite sum A_n first:

$$A_n \triangleq \sum_{k=0}^n \left(\sum_{\max(|i|,|j|)=k} \frac{3(s_1-i)(s_2-j)}{((s_1-i)^2+(s_2-j)^2)^{5/2}} \right).$$
(B.4)

It can be checked that every term from A_n is simply the second derivative of the corresponding term in the series in Eq. (B.3) with respect to s_1 and s_2 . Moreover, compared to Eq. (B.3), the summation in A_n is arranged in a specific order. Hence it is reasonable to have

$$\frac{\partial^2 g_0}{\partial s_1 \partial s_2} = \lim_{n \to \infty} A_n. \tag{B.5}$$

The proof of the convergence of $\lim_{n \to \infty} A_n$ is not trivial. Here for simplicity, we check the truncation error of A_n at various selected points for $0 < s_2 < s_1 < 1/2$, and the results are shown in Table B.1. It can be asserted that $\lim_{n \to \infty} A_n$ is a convergent series.

Now combining Eqs. (B.4) and (B.5), the expression for g_0 is available:

$$g_0(s_1, s_2) = \int_0^{s_1} \int_0^{s_2} \left[\lim_{n \to \infty} \sum_{k=0}^n \left(\sum_{\max(|i|, |j|)=k} \frac{3(\tilde{s}_1 - i)(\tilde{s}_2 - j)}{((\tilde{s}_1 - i)^2 + (\tilde{s}_2 - j)^2)^{5/2}} \right) \right] d\tilde{s}_1 d\tilde{s}_2 + C,$$
(B.6)

where *C* is a constant. In theory, *C* is determined by imposing the boundary condition $g_0 = 1$ on $\sqrt{x^2 + y^2} = r_{nv}$. However, since only the gradient term of p_s is of use, we can simply let C = 0. A comparison between Eqs. (17) and (B.2) suggests that the non-dimensional function *g* in Eq. (17) satisfies

$$g(s_1, s_2) = g_0(s_1, s_2) \cdot \sqrt{s_1^2 + s_2^2}.$$
(B.7)

It can be observed that $g(s_1, s_2) \sim \mathcal{O}(1)$ for $s_1, s_2 \in [-1/2, 1/2]$. Moreover, since $g_0 \sim 1/\sqrt{s_1^2 + s_2^2}$ as $\sqrt{s_1^2 + s_2^2} \rightarrow 0$. Therefore,

$$\lim_{\int s_1^2 + s_2^2 \to 0} g(s_1, s_2) = 1.$$
(B.8)

Appendix C

Now we look for the solution to Eq. (27) with boundary condition $c_{in} = 1$ on $\rho = \sqrt{\xi^2 + \eta^2} = 1$. Since the boundary condition is spherically symmetric, c_{in} should be a function of only ρ . Then Eq. (27) becomes

$$\frac{\mathrm{d}^2 c_{\mathrm{in}}}{\mathrm{d}\varrho^2} + \frac{1}{\varrho} \frac{\mathrm{d}c_{\mathrm{in}}}{\mathrm{d}\varrho} - \beta \left(\frac{c_{\mathrm{in}}}{\varrho^3} - \frac{1}{\varrho^2} \frac{\mathrm{d}c_{\mathrm{in}}}{\mathrm{d}\varrho}\right) = 0. \tag{C.1}$$

As the explicit solution to Eq. (C.1) may not be available, we first consider its solution behaviour for small β . If expanding $c_{in} \sim c^{(0)} + \beta c^{(1)} + \cdots$, the leading order $c^{(0)}$ reads

$$\frac{\mathrm{d}}{\mathrm{d}\varrho} \left(\varrho \frac{\mathrm{d}c^{(0)}}{\mathrm{d}\varrho} \right) = 0 \tag{C.2}$$

with $c^{(0)}|_{\rho=1} = 0$, which indicates $c^{(0)} = A \log \rho$ with A is a constant to be determined.

Thus at the first-order,

$$\frac{\mathrm{d}}{\mathrm{d}\varrho} \left(\varrho \frac{\mathrm{d}c^{(1)}}{\mathrm{d}\varrho} \right) = \frac{A}{\varrho^2} \cdot (\log \varrho - 1), \tag{C.3}$$

which gives rise to

$$c^{(1)} = A \cdot \left(A_1 - A_2 \log \varrho - \frac{1}{\varrho} - \frac{\log \varrho}{\varrho} \right), \tag{C.4}$$

where $A_1 = 1$ because of the boundary condition; $A_2 = 0$ because $c^{(1)}$ decays at infinity. Thus

$$c^{(1)} = A \cdot \left(1 - \frac{1}{\varrho} - \frac{\log \varrho}{\varrho} \right).$$
(C.5)

Sequentially, we can obtain

$$c^{(2)} = A \cdot \left(\frac{1}{4} - \frac{1}{\varrho} + \frac{3}{4\varrho^2} + \frac{\log \varrho}{2\varrho^2}\right);$$
(C.6)

$$c^{(3)} = A \cdot \left(\frac{1}{18} - \frac{1}{4\varrho} + \frac{1}{2\varrho^2} - \frac{11}{36\varrho^3} - \frac{\log \varrho}{6\varrho^3}\right); \tag{C.7}$$

$$c^{(4)} = A \cdot \left(\frac{1}{96} - \frac{1}{18\varrho} + \frac{1}{8\varrho^2} - \frac{1}{6\varrho^3} + \frac{25}{288\varrho^4} + \frac{\log \varrho}{24\varrho^4} \right);$$
(C.8)

.

In principle, one can write $c_{in} = \sum_{n=1}^{\infty} \beta^n c^{(n)}$. Here as the behaviour of $g(\varrho; \beta)$ as $\varrho \to \infty$ is of most use for future analysis, we write

$$\lim_{\rho \to \infty} c_{\rm in} = \beta + \frac{\beta^2}{4} + \frac{\beta^3}{18} + \frac{\beta^4}{96} + \cdots,$$
(C.9)

and $\lim_{\varrho \to \infty} c_{in}$ can be approximated by truncating the series in Eq. (C.9). However, the above series converges only for small β . There exists some $\beta = \beta^*$, where the series in Eq. (C.9) becomes divergent. In fact, this convergent radius β^* is highly related to the saturation state that will be discussed in Section 5. Hence in order to highlight β^* , an approximation to the series in Eq. (C.9) can be made by

$$\lim_{\varrho \to \infty} g(\varrho; \beta) \approx \beta + \frac{\beta^2}{4} + \frac{\beta^3}{4^2} + \frac{\beta^4}{4^3} + \dots = \frac{4\beta}{4-\beta},$$
(C.10)

which leads to $\beta^* = 4$.

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