



HAL
open science

Effect of sink strength dispersion on cluster size distributions simulated by cluster dynamics

Denise Carpentier, Thomas Jourdan, Pierre Terrier, Manuel Athenes, Yann Le Bouar

► **To cite this version:**

Denise Carpentier, Thomas Jourdan, Pierre Terrier, Manuel Athenes, Yann Le Bouar. Effect of sink strength dispersion on cluster size distributions simulated by cluster dynamics. *Journal of Nuclear Materials*, 2020, <https://doi.org/10.1016/j.jnucmat.2020.152068>. 10.1016/j.jnucmat.2020.152068 . cea-02495660

HAL Id: cea-02495660

<https://cea.hal.science/cea-02495660>

Submitted on 2 Mar 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Effect of sink strength dispersion on cluster size distributions simulated by cluster dynamics

D. Carpentier^a, T. Jourdan^{a,*}, P. Terrier^{a,b}, M. Athènes^a, Y. Le Bouar^c

^a*DEN-Service de Recherches de Métallurgie Physique, CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France*

^b*Université Paris-Est, CERMICS (ENPC), INRIA, F-77455 Marne-la-Vallée, France*

^c*LEM, Université Paris-Saclay, ONERA, CNRS, 29 av. de la division Leclerc, 92322 Châtillon, France*

Abstract

The microstructure evolution in irradiated materials can be conveniently modelled, at large scale, by cluster dynamics (CD). In this approach, the effect of the local environment of defect clusters is neglected. In this article, we first check the validity of this assumption by comparing CD to object kinetic Monte Carlo (OKMC) simulations. We show that for microstructures produced under irradiation, taking into account in CD only the average dependency of clusters' growth rate on volume fraction does not permit to reproduce reference OKMC results. Accordingly, the sink strength dispersion, quantified using OKMC, is introduced in CD, using a new formalism depending on the Voronoi volumes of defect clusters. CD calculations including sink strength dispersion are shown to be in better agreement with reference OKMC simulations and experimental observations than are classical CD calculations.

Keywords: Cluster dynamics ; object kinetic Monte Carlo; diffusional interactions; spatial correlations

1. Introduction

2 Long term evolution of microstructures containing second phase particles or
3 defect clusters can be efficiently simulated by cluster dynamics (CD). Its raw

*Corresponding author

Email address: thomas.jourdan@cea.fr (T. Jourdan)

4 output consists in size distributions at a desired time, and this approach can
5 be applied, for example, to the description of precipitates [1, 2, 3], voids and
6 dislocation loops [4, 5, 6]. Its mean field character makes it particularly efficient
7 compared to other methods which describe the position of clusters and thus take
8 into account spatial correlations naturally, such as atomistic and object kinetic
9 Monte Carlo (A/OKMC) methods. Under thermal aging, CD reproduces well
10 AKMC results provided it is carefully parametrized with the same atomistic
11 data [2, 3] and that potential corrections are added to the classical formalism
12 to handle concentrated alloys [7, 8]. For materials under irradiation, it has
13 been shown that provided input parameters in CD and OKMC are consistent,
14 cluster size distributions are very similar for low volume fractions, even if dis-
15 tributions obtained with OKMC can be slightly broader than those obtained
16 with CD [9]. Tests at higher volume fractions have not been performed, to our
17 knowledge. Comparison of CD and experimental results show that the mean
18 field approach becomes less precise at larger dose, the experimental distributions
19 being broader [4].

20 Such discrepancies in particle size distributions have been extensively stud-
21 ied in the context of Ostwald ripening [10]. Indeed it is known that experimen-
22 tal distributions can be broader than the prediction of Lifshitz-Slyozov-Wagner
23 (LSW) theory, which is itself in overall good agreement with CD [2]. LSW ap-
24 proach and CD in its simplest form both neglect the effect of volume fraction of
25 particles. However, the diffusion field around a particle can be modified by the
26 presence of other particles in the surroundings, thereby modifying the growth
27 rate of the particle [11, 12]. For Ostwald ripening, numerous attempts have been
28 made to determine the *average* growth rate of particles, taking into account the
29 presence of other particles [10]. The dependency of the average growth rate
30 of particles on volume fraction can be rationalized in terms of microstructure-
31 dependent “sink strengths” [13]. Such dependency has been early recognized
32 in the simulation of radiation-induced clusters such as voids and dislocation
33 loops [14]. However, CD calculations with sink strengths depending on the vol-
34 ume fraction of second phase particles remain rather scarce [2, 15]. Beyond the

35 effect on the average growth rate, the variety of local environments of particles
36 can lead to a *dispersion* of sink strengths for a given particle size. This disper-
37 sion is neglected in CD and to our knowledge, the validity of this assumption
38 has never been checked.

39 In this article we investigate the effect of local neighborhood on the sink
40 strengths of dislocation loops in microstructures generated by OKMC. We show
41 that using the average growth rate for a given cluster size in CD is not sufficient
42 to reproduce reference cluster distributions determined by OKMC. It is therefore
43 necessary to introduce explicitly the sink strength dispersion in CD due to the
44 neighborhood effects. For this purpose, a simple sink strength expression is
45 proposed, which reproduces the dispersion. The CD formalism is modified to
46 introduce the sink strength dispersion.

47 The paper is organized as follows. In section 2, we start by describing the
48 method used to generate microstructures in OKMC simulations, and to calcu-
49 late the sink strengths in these microstructures. By comparing microstructures
50 produced by equivalent OKMC and CD simulations, volume fraction effects and
51 the influence of sink strength dispersion are quantified. Then a sink strength
52 expression is derived in section 3 to reproduce the dispersion obtained in mi-
53 crostructures. In section 4, we present a new method to introduce the sink
54 strength dispersion in CD. Extension of the method to more complicated cases,
55 in particular if elastic interactions between sinks and point defects are consid-
56 ered, is discussed in section 5. This model is then used to simulate electron
57 irradiation experiments performed on aluminum thin foils (section 6).

58 **2. Volume fraction effects and sink strength dispersion in microstruc-** 59 **tures**

60 In order to quantify the sink strength dispersion and show its link with neigh-
61 borhood effects, sink strengths are evaluated in dislocation loop microstructures
62 using OKMC. To consider microstructures as realistic as possible, the forma-
63 tion of dislocation loops by agglomeration of self-interstitial atoms (SIAs) is also

64 simulated by OKMC. Cluster distributions thus obtained can be compared to
 65 distributions given by equivalent CD calculations, relying on some classical sink
 66 strength expressions which describe volume fraction effects at different levels of
 67 accuracy. This enables us to quantify the effect of non-zero volume fraction and
 68 sink strength dispersion on cluster distributions.

69 *2.1. Creation of microstructures*

70 The microstructures are created by OKMC simulations [16, 17], starting
 71 from a simulation box containing no defects. SIAs are introduced with a given
 72 creation rate G . They migrate by atomic jumps until reacting with another SIA,
 73 thus creating a dislocation loop, or with a loop. We consider immobile Frank
 74 loops in $\{111\}$ planes with Burgers vector \mathbf{b} of type $1/3\langle 111 \rangle$. The orientation
 75 is randomly chosen among the four variants when the loops are created.

76 Two models are considered for the absorption of SIAs. In the simplest model,
 77 which is studied in detail in this article, an SIA is absorbed by a loop containing
 78 n SIAs when it enters the sphere which encloses the loop, *i. e.* if the distance
 79 d between the loop center and the SIA verifies

$$d \leq r_{L,n} + r_{PD}, \quad (1)$$

80 where r_{PD} is the point defect radius, set to the atomic radius, and $r_{L,n}$ is the
 81 loop radius. The loop radius is related to n through

$$r_{L,n} = \sqrt{\frac{nV_{\text{at}}}{\pi b}}, \quad (2)$$

82 with V_{at} the atomic volume and b the norm of the Burgers vector. We also
 83 assume that SIAs do not interact elastically with the sink. The choice of the
 84 absorption on an encapsulating sphere instead of a torus, without elastic inter-
 85 actions, enables us to use CD with a larger number of sink strength expressions.

86 In section 5, we consider a more realistic model. Absorption of SIAs by
 87 loops occurs on the torus of radius r_L and pipe radius $r_p = 2b$. This means that
 88 a point defect is absorbed by the loop when the distance d between the point

89 defect and the dislocation line verifies

$$d \leq r_p + r_{PD}. \quad (3)$$

90 Some calculations have been performed with elastic interactions in this case.

91 In OKMC simulations, the computation time increases with the number of
92 defects. To decrease the computation time, one way is to reduce the box size
93 as much as possible. However, two difficulties can arise. The first one is related
94 to the potentially large fluctuations of cluster distributions from one simulation
95 to another, due to the moderate number of clusters. To obtain well-converged
96 cluster distributions, we average cluster distributions over a large number of
97 independent simulations. In this study, approximately a thousand of simulations
98 are used. The second problem is that the limited box size can itself lead to a
99 distortion in the cluster distribution. Indeed, in insufficiently large boxes, a
100 single cluster rapidly absorbs all the migrating point defects and remains alone.
101 Moreover, the periodic boundary conditions force the point defects to interact
102 with each other or with the remaining cluster. Therefore clusters can become
103 abnormally large and final cluster size distributions, obtained by averaging over
104 several simulations, vary with the box size for too small boxes. An example is
105 given in Fig. 1, with boxes of side length $l = 50$ nm, $l = 100$ nm and $l = 200$ nm,
106 a creation rate $G = 10^{-1}$ dpa.s $^{-1}$ and a physical time $t = 10^{-3}$ s. It can be
107 seen that the smallest box size gives an inconsistent distribution, while the box
108 sizes $l = 100$ nm and $l = 200$ nm give similar results. The convergence has been
109 checked for all the studied cases and sufficiently large boxes have been chosen.

110 In this work we have considered three different creation rates: 10^{-3} dpa.s $^{-1}$,
111 10^{-2} dpa.s $^{-1}$ and 10^{-1} dpa.s $^{-1}$. We only report the results for $G = 10^{-1}$ dpa.s $^{-1}$,
112 since similar results are obtained for the other dose rates. The case $G =$
113 10^{-3} dpa.s $^{-1}$ can be found in the Supplementary Material and all results are
114 available in Ref. [18]. The high creation rates considered here and in the rest of
115 this work are to simulate high doses with the short simulation times accessible
116 with the OKMC simulations. Other parameters are provided in Table 1. Given
117 the low temperature and high binding energies of clusters [19], thermal emission

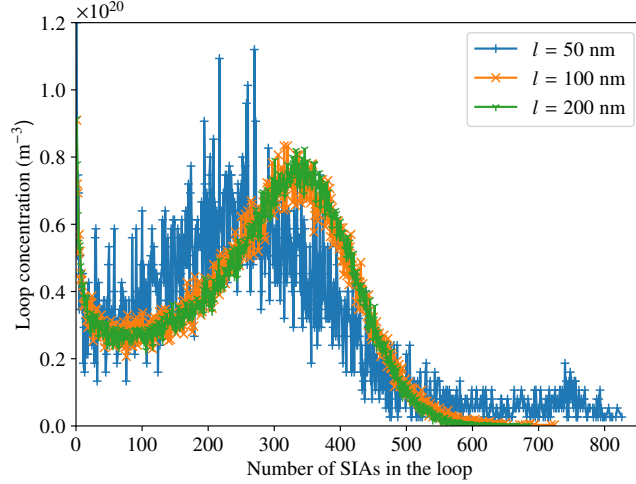


Figure 1: Example of cluster size distributions obtained with OKMC simulations, with $G = 10^{-1}$ dpa.s $^{-1}$, $t = 10^{-3}$ s and with the simulation conditions described in Table 1, for different box side lengths l .

118 of SIAs by loops is negligible.

119 2.2. Cluster dynamics without dispersion of sink strengths

120 The OKMC simulation of SIA agglomeration is reproduced by CD simula-
 121 tions using the CRESCENDO code [6]. The following equations are solved:

$$\frac{dC_n}{dt} = \beta_{n-1}C_{n-1}C_1 - \beta_n C_n C_1 \quad n \geq 2 \quad (4)$$

$$\frac{dC_1}{dt} = \frac{G}{V_{\text{at}}} - \beta_1 C_1 C_1 - \sum_{n \geq 1} \beta_n C_n C_1, \quad (5)$$

122 where C_n is the concentration of clusters containing n SIAs and $\beta_n C_1$ is the
 123 absorption rate of SIAs by these clusters. The sink strength of clusters of size
 124 n is defined by

$$k_n^2 = \frac{\beta_n}{D_1} C_n, \quad (6)$$

125 where D_1 is the diffusivity of SIAs, which means that the loss rate of SIAs, by
 126 unit volume, to clusters of size n is

$$\phi_n = k_n^2 D_1 C_1. \quad (7)$$

Temperature	$T = 300 \text{ K}$
Material: pure aluminum	
Lattice parameter	$a_0 = 0.405 \text{ nm}$
Atomic volume	$V_{\text{at}} = 1.66 \cdot 10^{-2} \text{ nm}^3$ [19]
Poisson's ratio	$\nu = 0.35$
Shear modulus	$\mu = 25.91 \text{ GPa}$ [17]
SIA properties	
Relaxation volume (for elastic interactions)	$\Delta V = 2.35 V_{\text{at}}$ [17]
Diffusion prefactor	$5 \cdot 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$ [19]
Migration energy	0.105 eV [19]
Point defect radius	$r_{\text{PD}} = 0.1503 \text{ nm}$
SIA loop properties	
Norm of the Burgers vector	$b = 0.2338 \text{ nm}$

Table 1: Parameters for the OKMC and CD simulations of SIA agglomeration. The relaxation volume is used in CD, it corresponds to the elastic dipole at stable point used in OKMC, which was computed by density function theory calculations (see Ref. [17]).

127 We also define the absorption efficiency by

$$\kappa_n = k_n^2 / C_n. \quad (8)$$

128 Various expressions for sink strengths, or equivalently absorption efficiencies,
129 exist in the literature, especially when the absorption occurs on a sphere. We
130 consider three of them:

- 131 • “Laplace” expression [20]:

$$\kappa_n = 4\pi r_n, \quad (9)$$

132 where $r_n = r_{\text{L},n} + r_{\text{PD}}$. This formula is derived by assuming that the loop
133 is isolated in an infinite medium, *i. e.* the volume fraction is zero.

- 134 • Wiedersich expression [20]:

$$\kappa_n = 4\pi r_n \frac{1 - \eta^3}{1 - \frac{9}{5}\eta + \eta^3 - \frac{1}{5}\eta^6}, \quad (10)$$

135 where $\eta = r_n/R$ and R is the average half-distance between sinks. This
 136 distance is determined at each time step of the CD calculation by

$$\frac{1}{\frac{4\pi}{3}R^3} = \sum_{n \geq 2} C_n. \quad (11)$$

137 This absorption efficiency is obtained by relating the flux to the loop to
 138 the average concentration, assuming a homogeneous production rate of
 139 defects and zero flux for $r = R$.

140 • Effective medium approach [14]:

$$\kappa_n = 4\pi r_n \frac{1 + k_{\text{tot}}R}{1 + k_{\text{tot}}(R - r_n)} \left[1 + \frac{k_{\text{tot}}^2(R - r_n)}{6(1 + k_{\text{tot}}R)} ((3 + k_{\text{tot}}R)(R + r_n) - 2k_{\text{tot}}r_n^2) \right], \quad (12)$$

141 where k_{tot}^2 is the total sink strength:

$$k_{\text{tot}}^2 = \sum_{n \geq 2} k_n^2 = \sum_{n \geq 2} \kappa_n C_n. \quad (13)$$

142 In practice, κ_n is calculated by iterating until self-consistency is obtained.
 143 Starting with Laplace expression for κ_n ($k_{\text{tot}} = 0$), a converged value is
 144 obtained in a few iterations. This kind of approach has been used in the
 145 context of Ostwald ripening to account for the dependency of growth rate
 146 of particles on the particle's volume fraction [13]. A simpler, approxi-
 147 mate expression [14], derived from Eq. (12), is also used in the context of
 148 irradiation [21, 22], but it will not be considered here.

149 Using the same parametrization as for OKMC (Table 1), the cluster size
 150 distributions obtained in CD are compared to the OKMC distribution, with
 151 $G = 10^{-1} \text{ dpa.s}^{-1}$ and $t = 10^{-3} \text{ s}$, in Fig. 2 (a). In OKMC simulations, point
 152 defects are absorbed when they enter the encapsulating sphere, which is the same
 153 absorption condition as in CD with the three above-mentioned sink strengths.

154 These results illustrate the fact that the OKMC distributions are broader
 155 than the CD ones, whatever the sink strength model. With the Laplace model,
 156 the density and mean radius are close to the ones obtained in OKMC, but
 157 the distribution is less spread. With the Wiedersich model, which takes into

158 account the effect of the loop density, a difference with the OKMC distribution
 159 is still observed. This model overestimates the mean radius. The effective
 160 medium approach also includes the effect of the surrounding environment on
 161 the sink strength, but fails to reproduce the OKMC distribution. Even though
 162 the average loop radius is less overestimated than with the Wiedersich model,
 163 the distribution is not as broad as the OKMC distribution. Similar but less
 164 pronounced discrepancies were obtained for lower doses (lower values of G) [18].
 165 These results indicate that taking into account only the average effect of particle
 166 volume fraction on sink strength is probably not sufficient to reproduce OKMC
 167 results. Sink strength dispersion also needs to be included in the CD formalism.

168 *2.3. Calculations of sink strengths values*

169 In order to calculate the sink strengths of loops in the OKMC microstruc-
 170 tures, the previously obtained microstructures are frozen: loops do not grow
 171 when SIAs are absorbed and SIAs do not form new loops. Once created, an SIA
 172 diffuses in the matrix until it is absorbed by a loop. It is then removed from the
 173 simulation and the number of SIAs absorbed by this loop is incremented. The
 174 aim of this procedure is to evaluate the loss rate of SIAs to the different sinks
 175 of the microstructure, which is directly related to the sink strength.

176 In practice, a given number M of simulation boxes containing microstruc-
 177 tures of loops are selected, typically 10 boxes for each value of G , and the new
 178 simulation step begins. The SIA creation rate, noted Γ in this context, is set to
 179 ensure a sufficiently large number of point defects in each box at steady state
 180 (typically 50 point defects). The average number \bar{N}_1 of SIAs is calculated as
 181 an average over time and over the boxes. The number N_i^{abs} of SIAs absorbed
 182 by each loop i , during the simulation time t , is recorded. The simulated time is
 183 chosen to ensure good statistics. The point defect loss rate φ_i to the loop i is
 184 then given by

$$\varphi_i = \frac{N_i^{\text{abs}}}{t}. \quad (14)$$

185 Following Eq. (7), the sink strength of each loop is then calculated according to

186

$$k_i^2 = \frac{\varphi_i}{D_1 \bar{N}_1}. \quad (15)$$

187 The sink strength is therefore defined with respect to a global SIA concen-
 188 tration, in agreement with its definition in the CD formalism. From the sink
 189 strength values k_i^2 , we compute the absorption efficiency κ_i according to (see
 190 Eq. (8))

$$\kappa_i = \frac{k_i^2}{1/l^3}, \quad (16)$$

191 where l is the edge length of the cubic simulation box. We note that since φ_i
 192 is the point defect loss rate to a single loop i , the concentration of loops which
 193 has to be used in Eq. (8), to define κ_i (a quantity attached to a single sink), is
 194 $1/l^3$. It is not the concentration of all loops which have the same size as i , *i.e.*
 195 of the class corresponding to i .

196 The values computed in the microstructures obtained with $G = 10^{-1} \text{ dpa.s}^{-1}$
 197 and $t = 10^{-3} \text{ s}$ are presented in Fig. 2 (b). They are compared to the sink
 198 strength models previously mentioned, which were used to obtain the CD cluster
 199 distributions (Fig. 2 (a)).

200 The sink strength values obtained in the microstructures are dispersed: loops
 201 having the same radius can have very different sink strengths. The values are
 202 globally lower than the value given by the Wiedersich model, but can also be
 203 below the value given by the Laplace model. The dispersion of sink strength
 204 values for loops of same radius but with different environments clearly indicates
 205 an environmental effect. We will show in the next sections that this dispersion
 206 is responsible for the difference observed on the cluster size distributions.

207 Similar results were obtained for lower values of G [18]. In that case, the
 208 loop density is lower, and the dispersion is also less important. Thus, it seems
 209 that the dispersion is reduced when the sink density diminishes. To illustrate
 210 this fact on comparable microstructures, the simulation boxes studied above are
 211 dilated to reduce the loop density. The box dimensions and distances between
 212 the loops are increased while the loops radii are preserved. The absorption
 213 efficiencies obtained for different densities are shown in Fig. 3. It is clear from

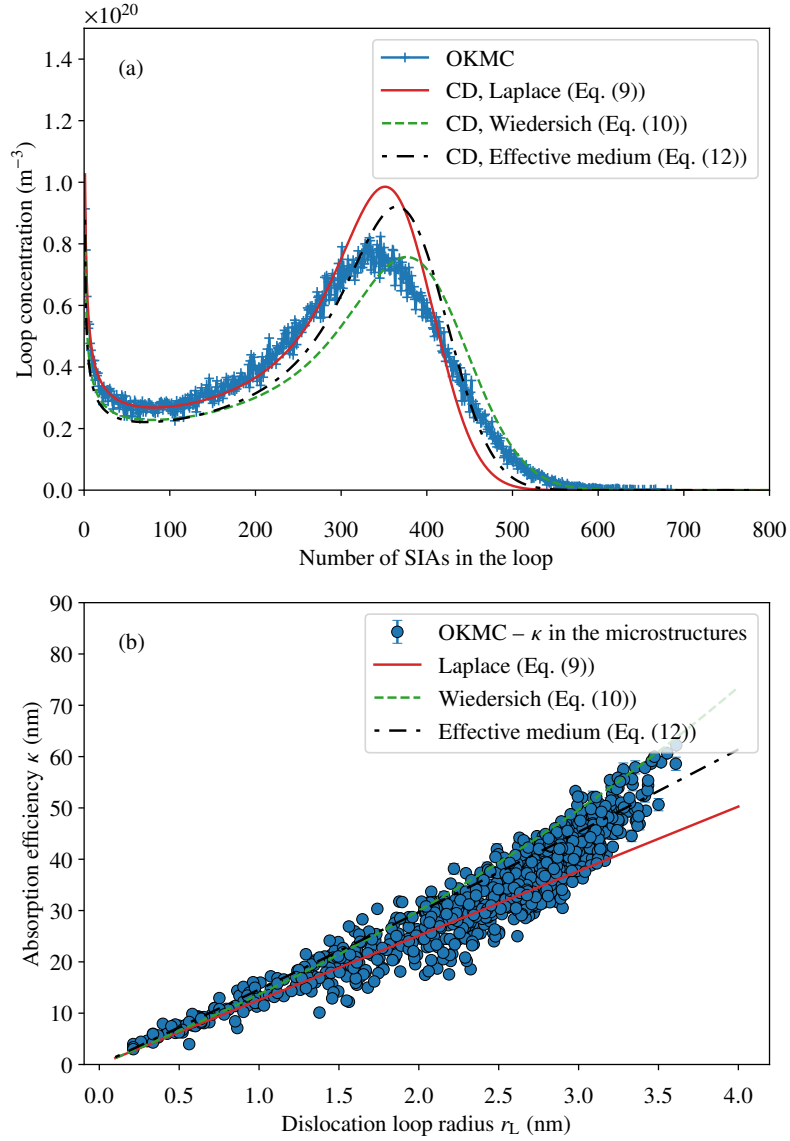


Figure 2: (a) Cluster size distributions obtained from OKMC simulations and equivalent CD simulations, using various sink strength models (see text), obtained with $G = 10^{-1} \text{ dpa.s}^{-1}$ and $t = 10^{-3} \text{ s}$. (b) Absorption efficiencies calculated for the loops in the OKMC microstructures and sink strength models used in CD.

214 this figure that the dispersion increases with the loop density.

215 **3. A model to represent sink strength dispersion**

216 To analyze the correlation between the sink strength value and the environ-
217 nment of loops, we determine the Voronoi cells of all loops in OKMC microstruc-
218 tures, using the QHULL program [23]. The Voronoi cell of a loop contains all
219 points which are nearer to the loop than to any other loop. It gives a first ap-
220 praisal of the local environment around a loop: the sink density around a loop
221 increases as the volume of the Voronoi cell of the loop decreases.

222 Results for the microstructures obtained in OKMC with $G = 10^{-1}$ dpa.s⁻¹
223 and $t = 10^{-3}$ s are shown in Fig. 4. They unveil a correlation between the
224 volumes and the sink strength values: loops with higher sink strengths tend to
225 have bigger Voronoi volumes. Similar results are obtained for other simulated
226 doses, especially for lower values of G [18]. One can also see in Fig. 4 that the
227 bigger loops tend to be in bigger cells. This can be understood by the mechanism
228 of loop growth: a loop in a big cell has no loop in its close neighborhood.
229 Hence, it can grow more rapidly. Inversely, a big loop easily absorbs neighboring
230 mobile point defects, preventing any loop nucleation in its neighborhood, so
231 its Voronoi cell remains large. The correlation between the sink strength and
232 the Voronoi volume is however not perfect. Indeed, we checked that the sink
233 strength values are also influenced by the shape of Voronoi cells and the loop
234 position in its cell [18]. However, the cell size remains the prevalent effect in the
235 cases considered here.

236 The Voronoi volumes therefore seem to be key elements to understand the
237 sink strength dispersion. In order to be able to use these parameters as input
238 data for CD calculations, the distributions of normalized Voronoi volumes in
239 the OKMC microstructures are calculated. The results are shown in Fig. 5
240 at different times, for $G = 10^{-1}$ dpa.s⁻¹. They are compared to the so-called
241 “Poisson-Voronoi distribution”, which refers here to the distribution of volumes
242 obtained for a Voronoi tessellation of points randomly distributed in space. An

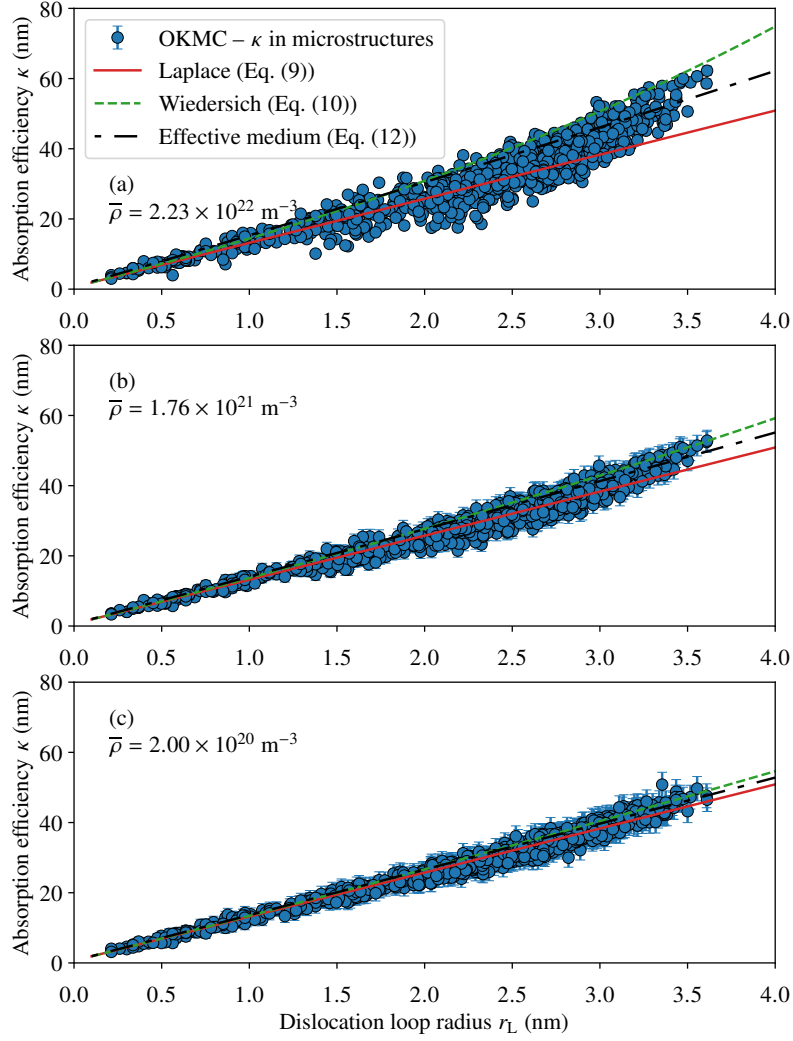


Figure 3: (a) Absorption efficiencies calculated in the OKMC microstructures obtained with $G = 10^{-1} \text{ dpa.s}^{-1}$ and $t = 10^{-3} \text{ s}$, loop density of $\bar{\rho} = 2.23 \cdot 10^{22} \text{ m}^{-3}$ ($l = 200 \text{ nm}$) (b) Absorption efficiencies obtained in dilated boxes ensuring a loop density of $\bar{\rho} = 1.76 \cdot 10^{21} \text{ m}^{-3}$ ($l = 466 \text{ nm}$) (c) Absorption efficiencies obtained in dilated boxes ensuring a loop density of $\bar{\rho} = 2.00 \cdot 10^{20} \text{ m}^{-3}$ ($l = 962 \text{ nm}$). The error bars represent the statistical error [18], they are displayed on all data.

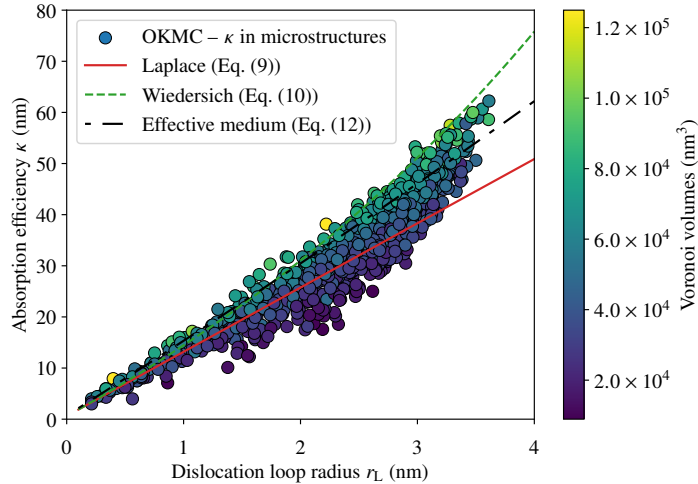


Figure 4: Correlations between the absorption efficiencies and Voronoi volumes (colorbar). Values are obtained in OKMC microstructures with a creation rate of $G = 10^{-1}$ dpa.s $^{-1}$ and for a simulated time of $t = 10^{-3}$ s.

243 accurate approximation of the Poisson-Voronoi distribution is given by the ex-
 244 pression proposed by Kumar *et al.* [24], whose parameters have been determined
 245 by Lazar *et al.* [25]. It can be seen that the distribution of normalized Voronoi
 246 volumes obtained in OKMC remains unchanged with time and that it is in
 247 very good agreement with the Poisson-Voronoi distribution. Therefore, it can
 248 be inferred that the loops are randomly distributed in the box. In addition,
 249 the expression of Kumar *et al.* can be used to give the distribution of Voronoi
 250 volumes, which is the main physical ingredient for sink strength dispersion.

251 The correlation of sink strengths with Voronoi volumes can be understood
 252 the following way. A cluster i in a small Voronoi volume V_i is surrounded by
 253 a density of clusters larger than the average density. The local sink strength
 254 around i , $k_{i,\text{loc}}^2$, is therefore higher than the average total sink strength k_{tot}^2 .
 255 Since as a first approximation, the local monomer concentration around i is
 256 related to the local sink strength through

$$C_{i,\text{loc}} = \frac{\Gamma}{V_{\text{at}} D_1 k_{i,\text{loc}}^2}, \quad (17)$$

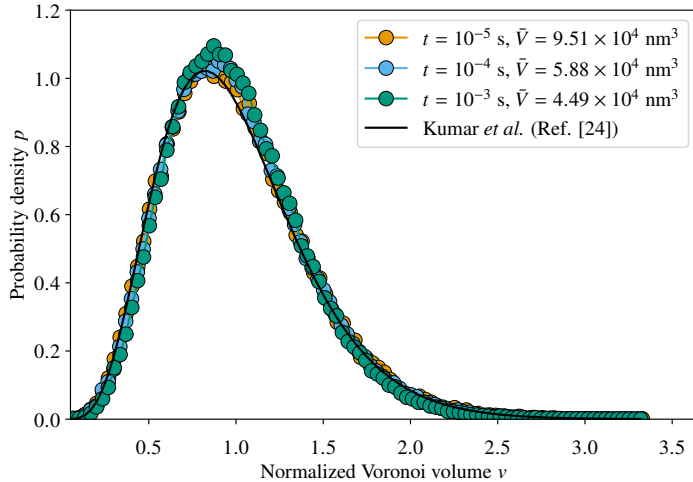


Figure 5: Distribution of normalized volumes of Voronoi cells $v = V/\bar{V}$, where \bar{V} is the average Voronoi volume, at different times, in OKMC microstructures with a creation rate of $G = 10^{-1}$ dpa.s $^{-1}$.

257 we see that it is lower than the average SIA concentration \bar{C} , given by

$$\bar{C} = \frac{\Gamma}{V_{\text{at}} D_1 k_{\text{tot}}^2}. \quad (18)$$

258 In other words, the local concentration of point defects around sink i is lower
 259 than the average concentration because there is a larger number of sinks in its
 260 vicinity to absorb point defects. Therefore the loss rate to the sink, which is re-
 261 lated to the local concentration of point defects available for absorption, appears
 262 to decrease as the Voronoi volume decreases. In the CD mean field formalism,
 263 the sink strength relates the loss rate to a sink to the point defect concentration
 264 averaged over the whole system (\bar{C} , or equivalently, \bar{N}_1 , see Eq. (15)). It means
 265 that for a small Voronoi volume, the low loss rate is equivalent to a low sink
 266 strength.

267 A simple calculation permits to relate more precisely the sink strength to
 268 Voronoi volumes. Let φ_i be the loss rate of SIAs to sink i . For a spherical sink,
 269 it can be written, to lowest order, as

$$\varphi_i = 4\pi r_i D_1 C_{i,\text{loc}}. \quad (19)$$

270 Using Eqs. (17) and (18) in Eq. (19) yields

$$\varphi_i = 4\pi r_i D_1 \frac{k_{\text{tot}}^2}{k_{i,\text{loc}}^2} \bar{C}. \quad (20)$$

271 Assuming that the average radius of clusters surrounding i is \bar{r} and the local
272 volume is $V_{i,\text{loc}}$, the local sink strength is approximately

$$k_{i,\text{loc}}^2 = 4\pi \bar{r} \frac{1}{V_{i,\text{loc}}}. \quad (21)$$

273 Since $k_{\text{tot}}^2 = 4\pi \bar{r} / \bar{V}$, we finally obtain

$$\varphi_i = 4\pi r_i D_1 \frac{V_{i,\text{loc}}}{\bar{V}} \bar{C}. \quad (22)$$

274 The local volume V_{loc} associated with each loop remains to be determined.
275 A simple approach is to use the average Voronoi volume of its nearest neighbors.
276 To determine an expression of this volume, a Poisson-Voronoi tessellation of 10^5
277 points is generated. For each Voronoi cell of volume V_i , the average volume of
278 nearest neighbors is calculated (Fig. 6). Nearest neighbor cells are defined as
279 neighbors which share a face. The average volume of neighbors is well fitted by
280 the following expression:

$$\frac{V_{\text{loc}}}{\bar{V}} = \left(\frac{V}{\bar{V}} \right)^\alpha + \beta, \quad (23)$$

281 with $\alpha = 0.25$ and $\beta = 0.07$. It is not clear if the volume itself should be included
282 in the average. In principle it affects the local concentration, so taking it into
283 account may be more correct. As shown in the figure, including it marginally
284 impacts the average volume, due to the large number of neighbors [26]. In the
285 following this contribution will not be considered.

286 The normalized local volumes V_{loc}/\bar{V} have also been extracted from the
287 OKMC simulations, using Eq. (22). They are displayed in Fig. 7 as a function
288 of normalized Voronoi volumes $v = V/\bar{V}$, for $G = 10^{-1} \text{ dpa.s}^{-1}$. These OKMC
289 results are in good agreement with Eq. (23). OKMC values are still dispersed
290 around the analytical formula. This dispersion comes from the fact that sink
291 strengths not only depend on Voronoi volume, but also on the shape of the
292 Voronoi cell and the sink position in its cell [18].

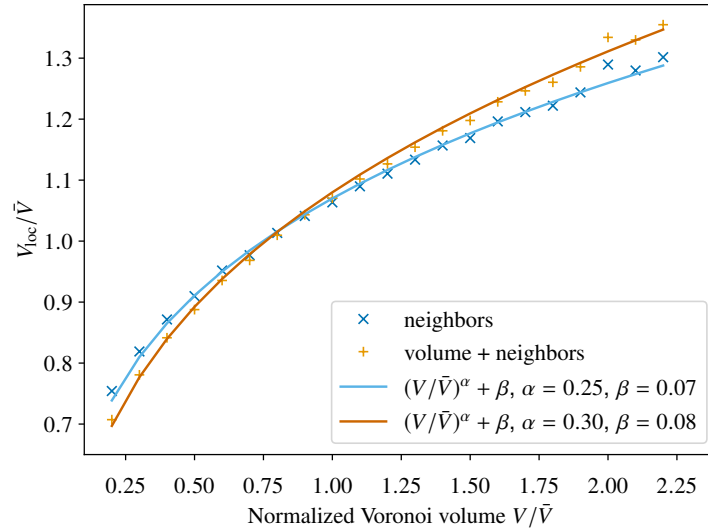


Figure 6: Average Voronoi volume of neighbors in a Poisson-Voronoi tessellation (blue symbols). For the orange symbols, the cell itself is included in the average. The distribution contains 10^5 Voronoi volumes.

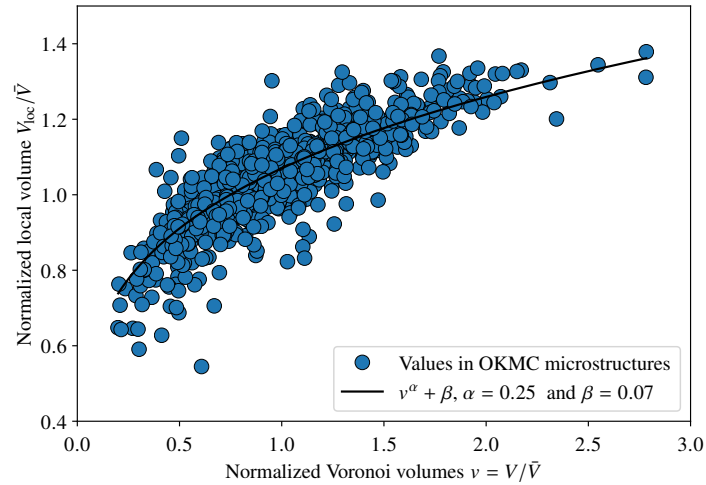


Figure 7: Values of normalized local volumes $V_{i,\text{loc}}/\bar{V}$ (see text) obtained in OKMC microstructures, as a function of the normalized Voronoi volume.

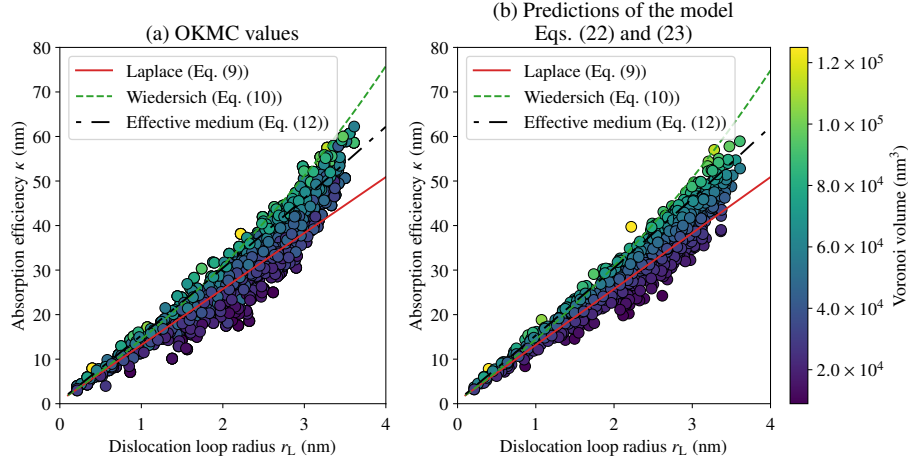


Figure 8: (a) Absorption efficiencies calculated in the OKMC microstructures obtained with $G = 10^{-1}$ dpa.s⁻¹ and $t = 10^{-3}$ s. (b) Absorption efficiencies calculated from the loop radii and Voronoi volumes using Eqs. (22) and (23).

293 Finally, for the conditions $G = 10^{-1}$ dpa.s⁻¹ and $t = 10^{-3}$ s, absorption
 294 efficiencies calculated by OKMC and predicted by Eqs. (22) and (23), using the
 295 Voronoi volumes from OKMC simulations, exhibit almost identical distributions
 296 (Fig. 8). This means that Voronoi cell shape and sink position in Voronoi cell
 297 are probably second order effects compared to the volume of the Voronoi cell
 298 itself, when we focus on the sink strength. We note, however, that since the sink
 299 strength dispersion is observed to increase with density, the proposed approach
 300 tends to overestimate the dispersion for low sink densities and to underestimate
 301 it for large sink densities.

302 To summarize, the absorption efficiencies in the microstructures can be re-
 303 produced by the expression

$$\kappa = 4\pi r \frac{V_{\text{loc}}}{\bar{V}}, \quad (24)$$

304 where V_{loc} is the average Voronoi volume of loops in the neighborhood. It
 305 depends on the Voronoi volume of the loop according to Eq. (23), with $\alpha = 0.25$
 306 and $\beta = 0.07$. The normalized Voronoi volume V/\bar{V} follows the distribution
 307 corresponding to a Poisson-Voronoi tessellation. An analytical form of this

308 distribution is given by Kumar *et al.* [24], and the parameters can be found in
309 Ref. [25].

310 **4. Introducing sink strength dispersion in cluster dynamics simula-** 311 **tions**

312 In previous sections we characterized the sink strength dispersion and de-
313 rived an expression to reproduce this dispersion, based on the distribution of
314 Voronoi volumes. Now we introduce the sink strength dispersion in the classical
315 CD equations (4) and (5). To do so, the absorption coefficient β_n is assumed
316 to depend on the normalized Voronoi volume v for a size larger than n^* , with
317 $n^* \geq 2$:

$$\beta_n(v) = 4\pi r_n (v^\alpha + \beta) D_1. \quad (25)$$

318 The cluster concentration of a given class n now also depends on v , so it is
319 noted $C_n(v)$. As reported in Section 2, large clusters are present more fre-
320 quently in large Voronoi cells than in small ones. This means that the change
321 of neighborhood of a cluster, or in other words the change of its Voronoi cell,
322 due to the creation of a cluster nearby, must happen over timescales which are
323 sufficiently large with respect to the growth process. Accordingly, we do not
324 include any coupling term between $C_n(v)$ and $C_n(v')$: the neighborhood of a
325 cluster is assumed to remain the same. Therefore a cluster in a large Voronoi
326 volume, whose sink strength is large, remains in a large Voronoi volume. In
327 reality, depending on the irradiation conditions, some clusters may nucleate in
328 its vicinity, leading to a reduction of its Voronoi volume and of its sink strength.
329 This approximation can be checked a posteriori on cluster distributions.

330 Equations (4)–(5) become

$$\frac{dC_n}{dt} = \beta_{n-1}C_{n-1}C_1 - \beta_n C_n C_1 \quad 2 \leq n \leq n^* - 1 \quad (26)$$

$$\frac{dC_n(v)}{dt} = P(v)\beta_{n-1}C_{n-1}C_1 - \beta_n(v)C_n(v)C_1 \quad n = n^*, v \in]0, \infty[\quad (27)$$

$$\frac{dC_n(v)}{dt} = \beta_{n-1}(v)C_{n-1}(v)C_1 - \beta_n(v)C_n(v)C_1 \quad n > n^*, v \in]0, \infty[\quad (28)$$

$$\frac{dC_1}{dt} = -\beta_1 C_1 C_1 - \sum_{1 \leq n \leq n^* - 1} \beta_n C_n C_1 - \int_0^\infty \sum_{n \geq n^*} \beta_n(v)C_n(v)C_1 P(v) dv. \quad (29)$$

331 In these equations, $P(v)$ is the Poisson-Voronoi distribution in normalized Voronoi
332 volumes.

333 To solve these equations numerically, two methods can be used. The first
334 one consists in discretizing the values of v , so concentrations can be noted
335 $C_{n,i} = C_n(v_i)$, where $i = 1, \dots, N$ and N is the number of equally spaced
336 possible normalized Voronoi volumes (see Fig. A.14 in Appendix A). The nu-
337 merical cost can increase substantially, since the number of equations is roughly
338 multiplied by N compared to a classical calculation involving a single popula-
339 tion. A more elegant way to solve equations (26)–(29) is to resort to a hybrid
340 deterministic-stochastic scheme recently developed for cluster dynamics equa-
341 tions [27]. In this method, small clusters ($n < n^*$) are treated deterministically,
342 while cluster dynamics equations are solved stochastically for larger sizes. The
343 deterministic and stochastic regions are separated by a buffer region where the
344 transfer between deterministic cluster density and stochastic particles is per-
345 formed. Sink strength dispersion can be naturally introduced in this method.
346 Each time a stochastic particle is created, due to the flux of clusters from the de-
347 terministic region to the stochastic region, a normalized Voronoi volume, drawn
348 in the Poisson-Voronoi distribution, is associated to this particle. The particle
349 then evolves according to the value of the absorption coefficient corresponding
350 to the normalized Voronoi volume. To ensure good performance and accuracy
351 of the hybrid algorithm, the deterministic region must contain at least a few
352 tens of classes. Here, n^* is set to 20.

353 Results are compared to OKMC simulations for $t = 10^{-3}$ s, $t = 2 \cdot 10^{-3}$ s and
 354 $t = 10^{-2}$ s for the highest dose rate ($G = 10^{-1}$ dpa.s $^{-1}$), which corresponds to
 355 the highest cluster density (Figs. 9 and 10; see Supplementary Material for the
 356 case $G = 10^{-3}$ dpa.s $^{-1}$ and $t = 10^{-2}$ s). The fully deterministic calculations
 357 with dispersion were performed with $N = 50$. It was checked, by varying this
 358 value, that in the conditions considered the cluster distributions are accurately
 359 simulated. For too low values of N ($N \lesssim 30$), cluster distributions become
 360 distorted at the largest dose. Two values for n^* were considered in these calcu-
 361 lations: $n^* = 2$ and $n^* = 20$. The latter corresponds to the value used in the
 362 hybrid method.

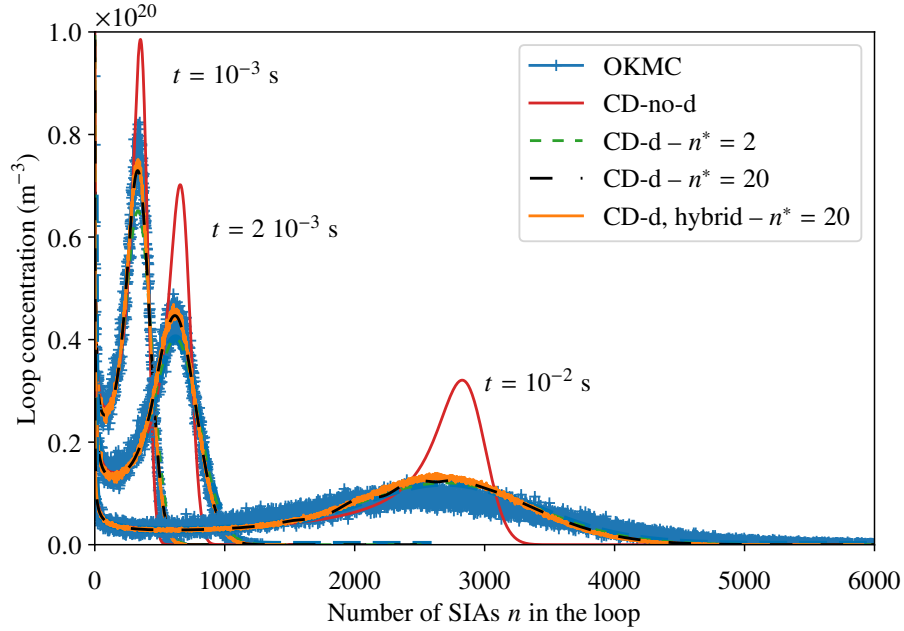


Figure 9: Cluster distributions at $t = 10^{-3}$ s, $t = 2 \cdot 10^{-3}$ s and $t = 10^{-2}$ s, obtained with OKMC and different CD models: deterministic calculation without dispersion and Laplace expression for sink strengths (CD-no-d), deterministic calculation with sink strength dispersion based on Eqs. (A.1)–(A.4) with $N = 25$ and two values for n^* (CD-d), hybrid deterministic-stochastic calculation using sink strength dispersion in the stochastic region (CD-d, hybrid). Hybrid calculations were performed with 2 million stochastic particles.

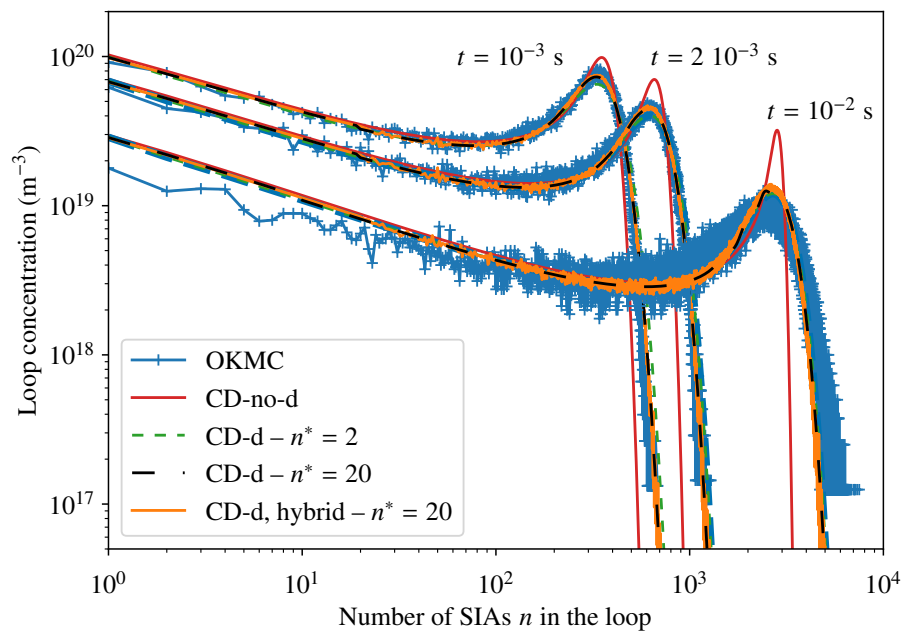


Figure 10: Cluster distributions at $t = 10^{-3}$ s, $t = 2 \cdot 10^{-3}$ s and $t = 10^{-2}$ s, obtained with OKMC and different CD models, in logarithmic scale. See Fig. 9 for more information.

363 We first see that the results obtained with the hybrid and the deterministic
 364 solving, for $n^* = 20$, perfectly match, which validates the two numerical meth-
 365 ods. However, the large number of equations to solve deterministically at high
 366 doses ($\sim 5 \times 10^5$) makes the deterministic method computationally intensive, so
 367 the hybrid method should be preferred. CD calculations including sink strength
 368 dispersion are all in much better agreement with OKMC than is the classical
 369 CD calculation using Laplace expression for sink strengths, which leads to ex-
 370 cessively peaked distributions. To compare more quantitatively the agreement
 371 between CD and OKMC distributions, we calculate the “distance” between CD
 372 and OKMC distributions \tilde{C}^{CD} and \tilde{C}^{OKMC} by using the L^2 norm:

$$\left\| \tilde{C}^{\text{CD}} - \tilde{C}^{\text{OKMC}} \right\| = \sqrt{\sum_{n \geq 2} (C_n^{\text{CD}} - C_n^{\text{OKMC}})^2}. \quad (30)$$

373 The ratio of the distance involving the CD distributions with sink strength dis-
 374 persion to that involving the standard CD distributions without dispersion is
 375 0.32, 0.21 and 0.33 at times $t = 10^{-3}$ s, $t = 2 \cdot 10^{-3}$ s and $t = 10^{-2}$ s, respec-
 376 tively. This shows that at all times the improvement is significant. Distributions
 377 obtained with $n^* = 2$ are slightly broader than with $n^* = 20$, due to the larger
 378 number of classes where dispersion is present. However, this difference tends
 379 to decrease with time, since the flux of clusters in the region of cluster space
 380 containing small clusters becomes lower (the nucleation rate decreases).

381 At short time ($t = 10^{-3}$ s), especially for $n^* = 2$, the CD cluster distribu-
 382 tions are broader than the OKMC distributions. As discussed previously, sink
 383 strength dispersion increases with cluster density (so with time) and Eq. (23)
 384 with $\alpha = 0.25$ and $\beta = 0.07$ reproduces well the dispersion for $t = 10^{-3}$ s,
 385 but overestimates the dispersion for shorter times. This overestimation can ex-
 386 plain why the distributions are slightly too broad. The choice $n^* = 20$, due
 387 to numerical constraints in the hybrid method, improves the agreement with
 388 OKMC results. This is due to the fact that no dispersion is taken into account
 389 for $n < n^*$, which compensates for the overestimation of dispersion at short
 390 times. At $t = 2 \cdot 10^{-3}$ s, the discrepancy with OKMC increases if no dispersion
 391 is taken into account, whereas CD simulations including dispersion remain very

392 close to OKMC. Finally, if the calculation is continued up to $t = 10^{-2}$ s, we
 393 see that CD cluster distributions without sink strength dispersion are far too
 394 peaked, whereas distributions with sink strength dispersion tend to be slightly
 395 narrower than OKMC distributions. Here again, this is due to the absence of
 396 dependence of Eq. (23) on cluster density. Despite this limitation, the overall
 397 good agreement for times $t = 2 \cdot 10^{-3}$ s and $t = 10^{-2}$ s is rather encouraging
 398 concerning the generality of the approach, inasmuch as no data were fitted at
 399 these times. In the following, we use the hybrid approach and thus we consider
 400 only calculations with $n^* = 20$.

401 The absorption efficiencies of the stochastic particles κ is shown in Fig. 11
 402 as a function of the cluster radius, for $t = 10^{-3}$ s. The overall shape of the
 403 distribution is very similar to the one in Fig. 2 (b), with large clusters mostly
 404 present in large Voronoi cells. This is consistent with the fact that clusters with
 405 large Voronoi cells grow faster and that the environment of particles does not
 406 change with time in CD calculations. The similarity of sink strength distribu-
 407 tions in CD and OKMC validates this approximation. A more refined model
 408 could be envisaged, by resampling some of the absorption rates of stochastic
 409 particles depending on the nucleation rate of clusters. For the present case, this
 410 additional complexity proved to be unessential.

411 5. Extension to toroidal geometry and effect of elastic interactions

412 The aim of this part is to show that the new CD formulation can be ex-
 413 tended to a more realistic situation in which absorption by a loop is realized on
 414 a torus and in which elastic effects are taken into account. SIAs are absorbed
 415 on the torus of radius $r_{L,n}$ (see section 2.1). Using the same method as previ-
 416 ously described, we determine the cluster size distributions, with or without the
 417 strain fields generated by the loops. The strain field of loops can be taken into
 418 account in OKMC simulations using the method described in [17], and the an-
 419 alytical strain field expression given in [28, 29]. Values of elastic dipole tensors
 420 determined in Ref. [17] by density functional theory calculations at stable and

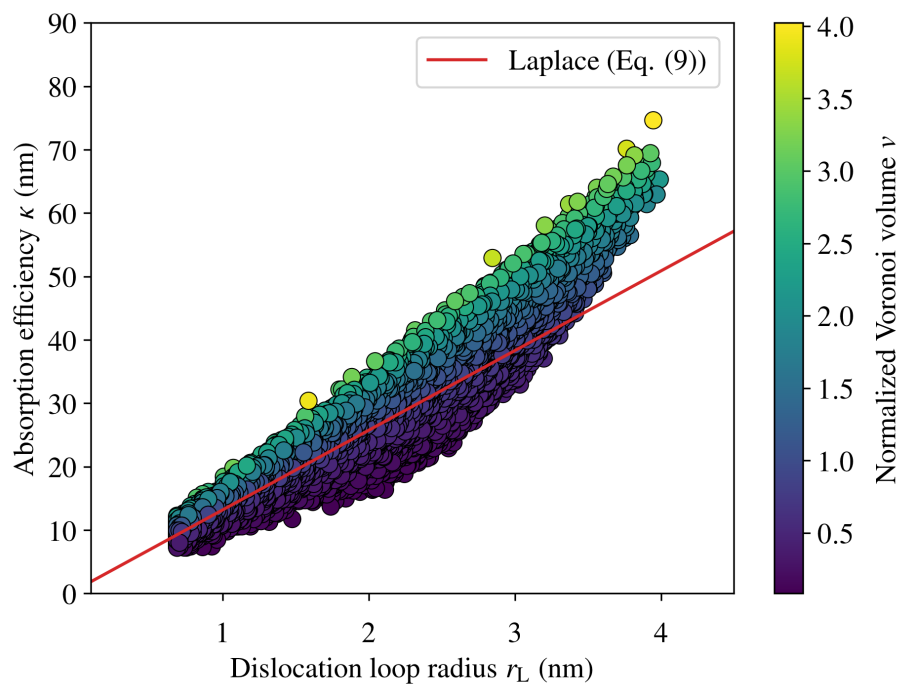


Figure 11: Normalized sink strengths at $t = 10^{-3}$ s, extracted from the values ascribed to stochastic particles in CD hybrid calculations.

421 saddle positions are used in the present OKMC simulations.

422 The corresponding CD models used here are described in Ref. [15]. When the
423 strain fields are neglected, the absorption can be represented by expression (18)
424 of Ref. [15], combined with expression (17) of Ref. [15] to include the effect of
425 non-zero volume fraction. To take into account the strain fields, the pipe radius
426 is replaced by an effective radius r_p^{eff} depending on the material and point defect
427 properties, according to expressions (19) and (20) of Ref. [15]. In this expression
428 SIAs are considered to be isotropic defects with the same relaxation volume at
429 stable and saddle points (see Table 1).

430 The cluster size distributions obtained in OKMC are compared to the corre-
431 sponding CD distributions in Fig. 12, for an SIA creation rate of $G = 10^{-1} \text{ dpa.s}^{-1}$
432 and a simulated time of $t = 10^{-3} \text{ s}$. The first thing to note is the large influence
433 of elastic interactions on the cluster distributions. As for the spherical clusters
434 studied above, one can see that the OKMC distributions are wider than the CD
435 ones when no dispersion is taken into account.

436 The same analysis of the sink strengths in OKMC microstructures has been
437 performed, and similar results were obtained regarding the sink strength disper-
438 sion and Voronoi volume distributions. To reproduce the sink strength disper-
439 sion, we use the same approach as the one described above. In agreement with
440 what was done for the absorption on spheres, we choose to disperse the values
441 of sink strengths around the value for the infinite medium. The absorption
442 efficiency reads:

$$\kappa = 2\pi r_L Z^{\text{loop}} \frac{V_{\text{loc}}}{V}, \quad (31)$$

443 where V_{loc} is given by Eq. (23), and Z^{loop} is defined in Ref. [15] and depends
444 on r_p if strain fields are neglected or r_p^{eff} if they are taken into account.

445 Following section 4, the dispersion is introduced in CD using the absorption
446 coefficient β_n , which reads

$$\beta_n = 2\pi r_{L,n} Z^{\text{loop}} (v^\alpha + \beta) D_1. \quad (32)$$

447 The results obtained using this model are shown in Fig. 12. As for the case
448 of spherical clusters, the introduction of dispersion in CD leads to a much better

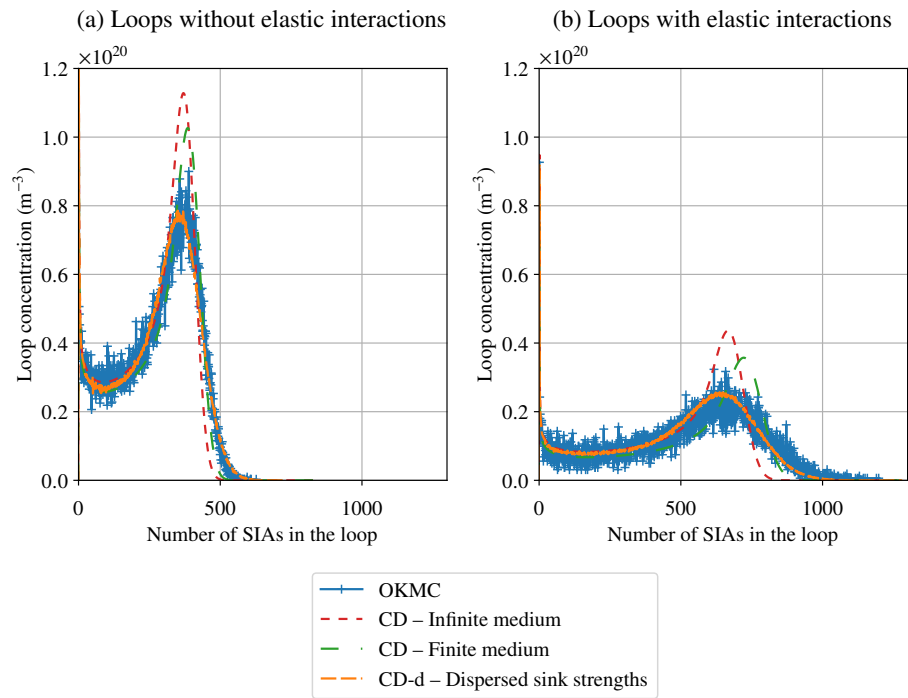


Figure 12: Cluster size distributions obtained when the absorption occurs on the dislocation line (a) without elastic interactions and (b) with elastic interactions. The CD models without dispersion are given in Ref. [15] (see text).

449 agreement with OKMC distributions. Therefore the proposed method can be
450 extended to more realistic systems, assuming that the absorption occurs on the
451 torus and taking into account the effect of elastic interactions.

452 **6. Comparison between experimental and simulated loop distribu-** 453 **tions**

454 The effect of sink strength dispersion on cluster distributions has been tested
455 up to limited doses (10^{-3} dpa) because we wanted to compare the CD results to
456 reference cluster distributions obtained by OKMC simulations. In this section,
457 we describe CD simulations to much higher doses (1 dpa). Since no comparison
458 can be made with OKMC in this case, simulations are compared to experimen-
459 tal measurements of cluster distributions. Results obtained by Chen *et al.* [30]
460 for dislocation loops in aluminum thin foils were chosen for several reasons. In
461 this experiment, the damage was produced by electron irradiation. Under these
462 conditions, only Frenkel pairs are generated, so point defect clusters are created
463 by the successive agglomeration of point defects. Frenkel pair accumulation is
464 known to be more easily simulated than displacement cascade conditions with
465 mean field CD [31, 32], especially at high doses where cascade overlap occurs.
466 The foils considered in the experiment by Chen *et al.* are relatively thick (around
467 500 nm), which, here again, is relatively favorable for mean field approaches.
468 Finally, cluster distributions are provided at several times. This permits to de-
469 termine some simulation parameters with more confidence. While comparing
470 simulations to experiments, it should be kept in mind that rather large bins (5
471 nm on loop diameter) were used to determine the cluster distributions experi-
472 mentally.

473 Equations used to simulate the experiments are based on Eqs. (4) and (5)
474 (Eqs. (26)-(29) if the dispersion of sink strengths is taken into account). In
475 addition, vacancies and vacancy clusters are taken into account. Single vacancies
476 can be absorbed by point defect clusters and recombine with SIAs. Point defects
477 can also be absorbed by surfaces. Emission of point defects by point defect

478 clusters is taken into account in the model. However, the binding energies of
479 point defects to dislocation loops are so high that at the temperature considered,
480 emission can be neglected. Although vacancy clusters are included in the model
481 for the sake of generality, they do not form since the di-vacancy is not stable [33],
482 so they do not play any role in the simulations. A more detailed description of
483 the equations can be found, for example, in Ref. [6].

484 In the simulations, surfaces are taken into account by a specific sink term [34],
485 for the sake of simplicity and computational efficiency. Such an approach leads
486 to similar results as spatialized calculations [35], but avoids to perform averages
487 of cluster distributions over the depth to make a comparison with experiments,
488 which could introduce artificial oscillations in the distributions' shape if the
489 number of slices was too low. The initial dislocation density is set to zero, since
490 micrographs show the absence of dislocations in the region of observation before
491 irradiation [30]. The temperature is set to 300 K. Based on a displacement cross-
492 section of 15 barns [36], the damage rate is estimated to 3×10^{-4} dpa.s⁻¹. It
493 has been shown by Chen *et al.* that the production of defects depends on the
494 orientation of the foil, the $\langle 111 \rangle$ direction being one where it is the highest.
495 In addition, the fraction of freely migrating defects (FMDs) is not precisely
496 known. Only FMDs must be taken into account in CD models, since other
497 defects quickly recombine with each other and do not participate to loop growth.
498 Therefore, there is some uncertainty on the production rate of defects to use in
499 CD. We chose a fraction of FMDs equal to one, to reflect the high production
500 rate of defects along $\langle 111 \rangle$ direction and the probably small proportion of close
501 pairs in this material [30]. This means that the actual creation rate of SIAs and
502 vacancies in the model is 3×10^{-4} dpa.s⁻¹. More work at atomic scale would be
503 necessary to determine the damage rate precisely, but this is beyond the scope
504 of this application.

505 Experiments were first simulated with the deterministic CD model described
506 in Ref. [15] (Eqs. 18 – 20) and used in the previous section, without sink
507 strength dispersion. Some parameters were adjusted to obtain a reasonable
508 agreement with the experimental densities and average sizes of loops (Tab. 2).

509 The saturation in loop density observed after 10 min of irradiation indicates that
 510 loop nucleation has stopped at this time. This behavior is not observed if the
 511 recommended value for the vacancy migration energy (0.61 eV, see Ref. [19])
 512 is taken. A slightly higher value must be adopted to increase the vacancy
 513 concentration and suppress loop nucleation from a certain time, *i. e.* the system
 514 must enter the recombination regime. The value 0.7 eV has been adopted. The
 515 migration energy of SIAs has also been increased to 0.19 eV, to produce loop
 516 densities similar to the experimental values. The recommended value (0.11
 517 eV) produces far too low cluster densities. These two parameter modifications
 518 may point to an effect of impurities, as suggested in the experimental study.
 519 Finally, the relaxation volume of the vacancy was changed from $-0.4 V_{\text{at}}$ [19]
 520 to $-0.8 V_{\text{at}}$. The resulting increase of the elastic interactions between vacancies
 521 and loops may reflect the effect of saddle point anisotropy of vacancies on the
 522 sink strength [17]. This anisotropy has been shown to increase significantly the
 523 sink strength of loops [18].

524 Loop distributions are shown at three different times in Fig. 13. As discussed
 525 previously, although the densities and average sizes are correctly reproduced,
 526 cluster distributions obtained experimentally and with CD without sink strength
 527 dispersion are markedly different. Experimental distributions are much broader.

528 Simulations were also performed with the new formalism, which includes
 529 the effect of sink strength dispersion. This effect was taken into account for
 530 the absorption of both SIAs and vacancies. Since the sink strength dispersion
 531 only depends on Voronoi volumes, which are not related to the properties of the
 532 absorbed defect, the approach is the same for vacancies as for SIAs.

533 Introducing the sink strength dispersion slightly shifts the peak position to
 534 larger sizes, since the average value of V_{loc}/\bar{V} in Eq. (23) does not equal 1 (it
 535 is the case for $\beta = 0.01687$). It also significantly broadens the distribution and
 536 improves the agreement with experiments, which shows the importance of taking
 537 into account the dispersion. The ratio of distances between cluster distributions
 538 (see Eq. (30)) is 0.47, 0.43 and 0.31 at 15, 30 and 60 min. Although a clear
 539 improvement is obtained, the agreement with experimental results is still not

Parameter	Value	Reference
Temperature	300 K	
Damage rate	3×10^{-4} dpa s ⁻¹	
Thickness of the foil	500 nm	
Network dislocation density	0	
Lattice parameter	0.405 nm	
Diffusion prefactor for vacancies	10^{-5} m ² s ⁻¹	[19]
Diffusion prefactor for SIAs	5×10^{-6} m ² s ⁻¹	[19]
Migration energy of vacancies	0.7 eV	adjusted
Migration energy of SIAs	0.19 eV	adjusted
Relaxation volume of SIAs	$1.9 V_{\text{at}}$	[19]
Relaxation volume of vacancies	$-0.8 V_{\text{at}}$	adjusted
Recombination radius	$2a_0$	[37]
Binding energy of two vacancies	0	[33]
Binding energy of two SIAs	0.8 eV	

Table 2: Parameters for CD simulations of electron irradiation of aluminum thin foils.

540 perfect. Among the possible sources of discrepancy, the absence of dependence
541 of Eq. (23) on cluster density is a probable one. Trends are consistent with
542 the fact that CD distributions with sink strength dispersion tend to be slightly
543 narrower than OKMC distributions at the largest simulated dose ($t = 10^{-2}$ s, see
544 Figs. 9 and 10). Other sources of discrepancy include the simplified treatment
545 of the effect of vacancy anisotropy at saddle position on the sink strength,
546 effects of impurities which have been taken into account in an effective way
547 and surface effects. Even if the foil thickness is large, surfaces may have some
548 effects on the loop distributions which are not properly handled in mean field
549 calculations. In particular, our analysis on Voronoi volume implicitly assumes
550 that the considered sink is surrounded by other sinks, which is not the case when
551 it is close to a surface. Larger spreading of growth rates may arise because of
552 the presence of this additional sink.

553 7. Discussion

554 In the previous sections we have shown that it is possible to obtain a good
555 agreement between CD results and OKMC results at high volume fraction, pro-
556 vided that the sink strength dispersion is introduced in CD. This dispersion is
557 primarily due to the variation of local sink density around sinks. To our knowl-
558 edge, sink strength dispersion has not been much studied for microstructures
559 obtained under irradiation; however, there is a vast amount of theoretical and
560 experimental results for the effect of local surroundings in Ostwald ripening. In
561 this context, most of the works have focused on much higher volume fractions
562 (of the order of percent or more) and theoretical works have introduced a de-
563 pendency of the particle's average growth rate on volume fraction (for a review,
564 see Ref. [10]). Although some laws can be transposed to the microstructures
565 under irradiation (see for example Refs. [13] and [14]), the simulation of Ostwald
566 ripening has its own peculiarities. Evolution of particles is due to the Gibbs-
567 Thomson effect, so the dependency of the growth rate on local surroundings can
568 result not only from different local densities, but also from different particle's

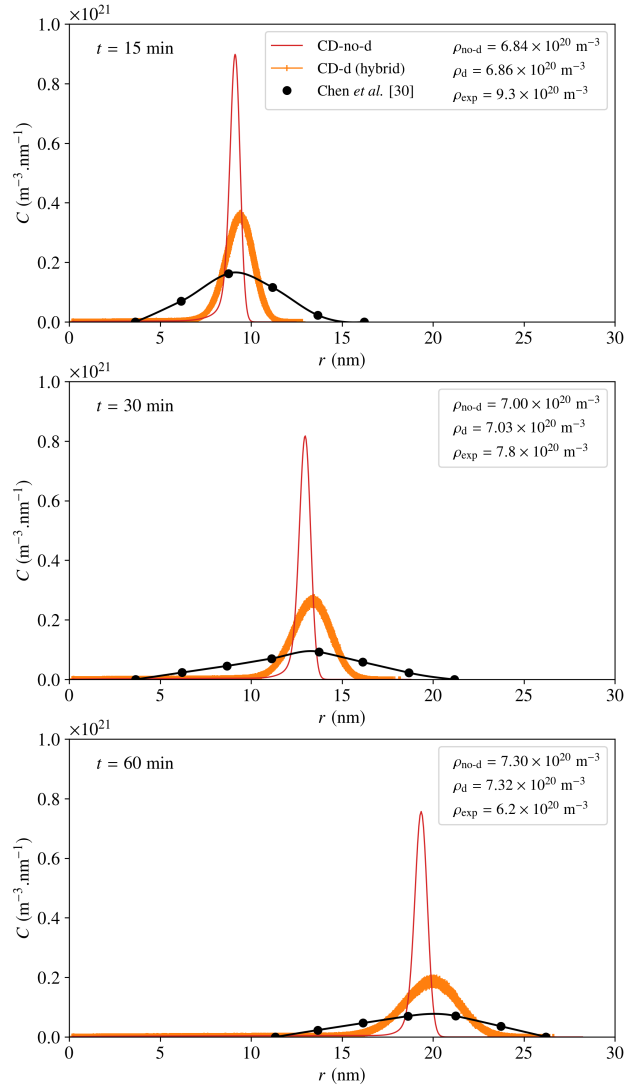


Figure 13: Experimental and simulated loop distributions for electron irradiated aluminum thin foils at different times. CD simulations are performed without (CD-no-d) and with (CD-d) sink strength dispersion. Loop densities $\rho_{\text{no-d}}$, ρ_{d} and ρ_{exp} are shown at each time.

569 radii. Under irradiation, at sufficiently low temperature, the large supersatu-
570 ration makes Gibbs-Thomson effect negligible. Both effects (local density and
571 particule radius) are included in the sink strength dispersion highlighted in the
572 simulation of Ostwald ripening (see Fig. 4 in Ref. [11]), but their respective
573 contribution remains unknown. In the case of microstructures produced under
574 irradiation, we have seen that introducing only the average effect of volume
575 fraction, as it is done classically for Ostwald ripening, is not sufficient: the sink
576 strength dispersion must also be included. The large effect of local density on
577 the particle's growth rate seems rather surprising, compared to the simulations
578 of Ostwald ripening. More in-depth investigation would be needed to explain
579 these seemingly different results.

580 The inability of models based only on volume fraction-dependent average
581 growth rate to reproduce OKMC cluster distributions is visible in Fig. 2. One
582 of these models (effective medium), proposed by Brailsford *et al.* [14], is shown in
583 this figure. Although the average values measured by OKMC reasonably agree
584 with this law, the associated cluster distributions are markedly different. Sink
585 strengths obtained by OKMC can even be lower than the Laplace solution, which
586 is below the values given by the volume fraction-dependent laws. The inability
587 of these laws to reproduce such dispersed data explains why cluster distributions
588 obtained with OKMC are much broader than those obtained using standard CD.
589 We have chosen to introduce dispersion around the Laplace law, since it fairly
590 represents the average value of the sink strengths for all cases considered in this
591 study and it can be efficiently implemented in CD. Additional data should be
592 collected, especially at larger doses, to see if this choice is always relevant.

593 To ensure that the correction is general, more complex cases should be con-
594 sidered. For example, the dispersion is assumed to be independent of the volume
595 fraction. Fig. 3 shows, however, that the dispersion tends to increase with vol-
596 ume fraction. In addition, we have only considered one type of defect in our
597 simulations where OKMC and CD are compared. A more realistic case would
598 be to consider vacancies and SIAs at the same time. Concerning vacancies, our
599 model including elastic interactions remains to be tested: analytical expressions

600 for sink strengths all rely on the description of defects as isotropic defects with
601 the same relaxation volume at stable and saddle points [38, 39, 15]. Although
602 this approximation is deemed correct for SIAs, it may be too crude for vacancies
603 which are highly anisotropic at saddle position. It may be one of the reasons of
604 the remaining discrepancy between our simulations and experiments by Chen *et*
605 *al.* [30] reported in Section 6. As discussed in this section, additional sinks such
606 as surfaces and grain boundaries may also change the sink strength dispersion
607 of clusters if their density is appreciable.

608 8. Conclusion

609 Using a combination of CD and OKMC simulations, we have studied the
610 effect of local surroundings on particle size distributions for microstructures
611 under irradiation. Distributions obtained by OKMC are shown to be broader
612 than those simulated by CD, even if the average effect of finite volume fraction
613 is introduced in the sink strengths used in CD. We have shown that the sink
614 strength dispersion must be included in CD in order to reproduce OKMC results.
615 To come to this conclusion, we used a two-step procedure:

- 616 • Simple laws for the sink strengths were proposed to reproduce the sink
617 strength dispersion measured with OKMC in realistic microstructures.
618 These laws depend on the solution in infinite medium (Laplace solution),
619 multiplied by a factor which depends on the Voronoi volume associated
620 with the sink.
- 621 • A new formalism of CD was developed in order to integrate the sink
622 strength dispersion. Cluster classes not only depend on the cluster size,
623 but also on the normalized Voronoi volume of the cluster.

624 Using this new approach, we showed that it is possible to obtain a satisfactory
625 agreement between the cluster distributions simulated by CD and OKMC in
626 all cases considered, although CD distributions become slightly narrower at
627 the largest doses. A comparison was then made with experimental dislocation

628 loop distributions obtained by electron irradiation of aluminum thin foils up
629 to 1 dpa. It was shown that the agreement is much better with sink strength
630 dispersion than without, but that differences still exist. Sources for this residual
631 discrepancy were discussed. More complex cases, involving for example two
632 kinds of point defects and the presence of other sinks, should now be considered.

633 **Acknowledgments**

634 This study was partially funded by the SIMMAT project from the I3P insti-
635 tute (CEA-EDF R&D). It has received funding from the Euratom research and
636 training program 2014-2018 under Grant Agreement N°661913. CRESCENDO
637 code is co-developed in collaboration with EDF R&D within the framework of
638 I3P institute.

639 **Data availability**

640 The relevant data are available within the article or from the authors upon
641 reasonable request.

642 **Appendix A. Deterministic solving of the CD equations including** 643 **sink strength dispersion**

644 The deterministic solving of Eqs. (26)–(29) can be done by discretizing the
645 values of v (Fig. A.14), noted v_i , where $i = 1, \dots, N$ and N is the number of
646 equally spaced possible normalized Voronoi volumes. Values v_i between 0 and 5
647 are sufficient to accurately sample the Poisson-Voronoi distribution (Fig. 5). The
648 spacing between two values of v is noted Δv . We use the notations $\beta_{n,i} = \beta_n(v_i)$

649 and $P_i = P(v_i)$. This leads to the set of equations to solve:

$$\frac{dC_n}{dt} = \beta_{n-1}C_{n-1}C_1 - \beta_n C_n C_1 \quad 2 \leq n \leq n^* - 1 \quad (\text{A.1})$$

$$\frac{dC_{n,i}}{dt} = \Delta v P_i \beta_{n-1} C_{n-1} C_1 - \beta_{n,i} C_{n,i} C_1 \quad n = n^*, i \in [1, N] \quad (\text{A.2})$$

$$\frac{dC_{n,i}}{dt} = \beta_{n-1,i} C_{n-1,i} C_1 - \beta_{n,i} C_{n,i} C_1 \quad n > n^*, i \in [1, N] \quad (\text{A.3})$$

$$\frac{dC_1}{dt} = -\beta_1 C_1 C_1 - \sum_{1 \leq n \leq n^*-1} \beta_n C_n C_1 - \sum_{i=1}^N \Delta v P_i \sum_{n \geq n^*} \beta_{n,i} C_{n,i} C_1. \quad (\text{A.4})$$

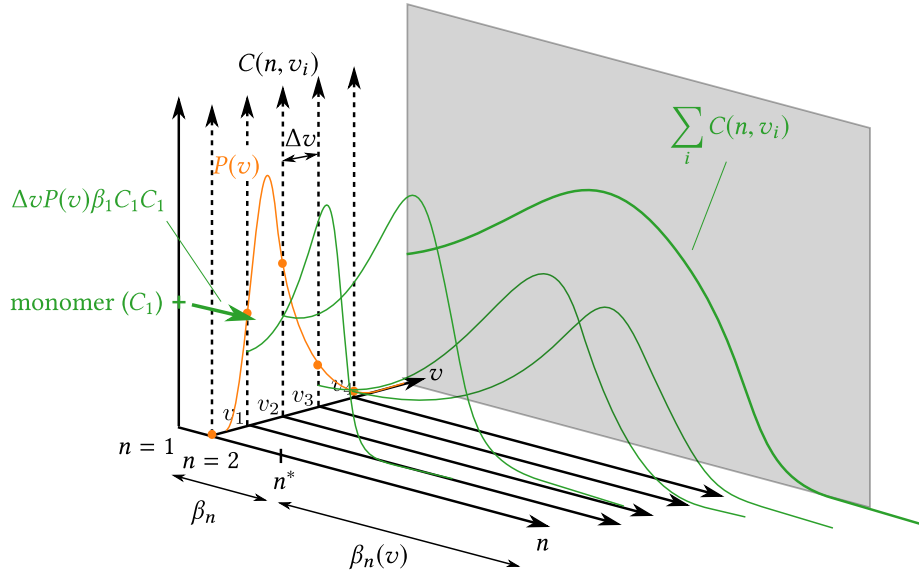


Figure A.14: Schematics of the deterministic solving of CD equations including sink strength dispersion.

650 This set of equations can be readily introduced in the CD code CRESCENDO,
 651 which enables the use of different cluster populations coupled together only
 652 through the mobile species [6]. Here, the different cluster populations actually
 653 correspond to the same clusters, but in different environments.

654 References

- 655 [1] M. H. Mathon, A. Barbu, F. Dunstetter, F. Maury, N. Lorenzelli, C. H.
 656 de Novion, J. Nucl. Mater. 245 (1997) 224.

- 657 [2] E. Clouet, A. Barbu, L. Lae, G. Martin, *Acta Mater.* 53 (2005) 2313.
- 658 [3] T. Jourdan, F. Soisson, E. Clouet, A. Barbu, *Acta Mater.* 58 (2010) 3400.
- 659 [4] S. I. Golubov, B. N. Singh, H. Trinkaus, *Philos. Mag. A* 81 (2001) 2533.
- 660 [5] A. Hardouin-Duparc, C. Moingeon, N. Smetniansky-de-Grande, A. Barbu,
661 *J. Nucl. Mater.* 302 (2002) 143.
- 662 [6] T. Jourdan, G. Bencteux, G. Adjanor, *J. Nucl. Mater.* 444 (2014) 298.
- 663 [7] J. Lépinoux, *Philos. Mag.* 86 (2006) 5053.
- 664 [8] J. Lépinoux, C. Sigli, *Philos. Mag.* 93 (2013) 3194.
- 665 [9] T. Jourdan, J.-P. Crocombette, *Comput. Mater. Sci.* 145 (2018) 235.
- 666 [10] P. W. Voorhees, *J. Stat. Phys.* 38 (1985) 231.
- 667 [11] P. W. Voorhees, M. E. Glicksman, *Acta Metall.* 32 (1984) 2013.
- 668 [12] I. Rovelli, S. L. Dudarev, A. P. Sutton, *J. Mech. Phys. Solids* 103 (2017)
669 121.
- 670 [13] A. D. Brailsford, P. Wynblatt, *Acta Metall.* 27 (1979) 489.
- 671 [14] A. D. Brailsford, R. Bullough, M. R. Hayns, *J. Nucl. Mater.* 60 (1976)
672 246–256.
- 673 [15] T. Jourdan, *J. Nucl. Mater.* 467 (2015) 286.
- 674 [16] C. Domain, C. S. Becquart, L. Malerba, *J. Nucl. Mater.* 335 (2004) 121.
- 675 [17] D. Carpentier, T. Jourdan, Y. Le Bouar, M.-C. Marinica, *Acta Mater.* 136
676 (2017) 323.
- 677 [18] D. Carpentier, Simulation of the absorption kinetics of point defects by
678 dislocations and defect clusters, Ph.D. thesis, Université Paris-Saclay, 2018.

- 679 [19] P. Ehrhart, Atomic defects in metals · Al: Datasheet from Landolt-
680 Börnstein - Group III Condensed Matter · Volume 25: “Atomic Defects
681 in Metals” in SpringerMaterials (http://dx.doi.org/10.1007/10011948_58),
682 1991.
- 683 [20] F. A. Nichols, J. Nucl. Mater. 75 (1978) 32.
- 684 [21] L. Malerba, C. S. Becquart, C. Domain, J. Nucl. Mater. 360 (2007) 159.
- 685 [22] T. Ahlgren, L. Bukonte, J. Nucl. Mater. 496 (2017) 66.
- 686 [23] C. B. Barber, D. P. Dobkin, H. T. Huhdanpaa, ACM Trans. Math. Softw.
687 22 (1996) 469.
- 688 [24] S. Kumar, S. K. Kurtz, J. R. Banavarn, M. G. Sharma, J. Stat. Phys. 67
689 (1992) 523.
- 690 [25] E. A. Lazar, J. K. Mason, R. D. MacPherson, J. D. Srolovitz, Phys. Rev.
691 E 88 (2013) 063309.
- 692 [26] A. Thorvaldsen, Mater. Sci. Forum 94-96 (1992) 307.
- 693 [27] P. Terrier, M. Athènes, T. Jourdan, G. Adjanor, G. Stoltz, J. Comput.
694 Phys. 350 (2017) 280.
- 695 [28] T. A. Khraishi, J. P. Hirth, H. M. Zbib, M. A. Khaleel, Int. J. Eng. Sci. 38
696 (2000) 251.
- 697 [29] F. Kroupa, Czech. J. Phys. B 10 (1960) 284.
- 698 [30] J. H. Chen, P. Rao, P. S. Ho, Radiat. Eff. 18 (1973) 157.
- 699 [31] T. Jourdan, J.-P. Crocombette, Phys. Rev. B 86 (2012) 054113.
- 700 [32] A. Y. Dunn, L. Capolungo, Comp. Mater. Sci. 102 (2015) 314.
- 701 [33] H. Wang, D. Rodney, D. Xu, R. Yang, P. Veyssière, Phys. Rev. B 84 (2011)
702 220103.

- 703 [34] R. Bullough, M. R. Hayns, M. H. Wood, *J. Nucl. Mater.* 90 (1980) 44.
- 704 [35] A. Barbu, C. S. Becquart, J.-L. Bocquet, J. Dalla Torre, C. Domain, *Philos.*
705 *Mag.* 85 (2005) 541.
- 706 [36] O. S. Oen, Cross sections for atomic displacements in solids by fast elec-
707 trons, Technical Report ORNL-4897, ORNL, 1973.
- 708 [37] W. G. Wolfer, A. Si-Ahmed, *J. Nucl. Mater.* 99 (1981) 117.
- 709 [38] W. A. Coghlan, M. H. Yoo, in: M. F. Ashby, R. Bullough, C. S. Hartley,
710 J. P. Hirth (Eds.), *Dislocation Modelling of Physical Systems*, Pergamon,
711 1981, pp. 152–157.
- 712 [39] V. I. Dubinko, A. S. Abyzov, A. A. Turkin, *J. Nucl. Mater.* 336 (2005) 11.

Supplementary material for “Effect of sink strength dispersion on cluster size distributions simulated by cluster dynamics”

D. Carpentier¹, T. Jourdan¹, P. Terrier^{1,2}, M. Athènes¹, and Y. Le Bouar³

¹*DEN-Service de Recherches de Métallurgie Physique, CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France*

²*Université Paris-Est, CERMICS (ENPC), INRIA, F-77455 Marne-la-Vallée, France*

³*LEM, Université Paris-Saclay, ONERA, CNRS, 29 av. de la division Leclerc, 92322 Châtillon, France*

In this supplementary material we present the cluster distributions obtained with a lower damage rate ($G = 10^{-3} \text{ dpa.s}^{-1}$), in the case of the absorption on spheres without elastic interactions. The reference distribution could not be obtained with OKMC due to CPU time constraints, so event-based kinetic Monte Carlo (EKMC) was used instead. This method does not permit to take into account elastic interactions but in the conditions considered here, it is faster than OKMC and yields identical results.

The same results as in the article are obtained (Fig. S1). The cluster distribution without sink strength dispersion (CD-no-d) is too peaked. Introducing sink strength dispersion through our hybrid approach (CD-d) leads to a much better agreement with the reference calculation.

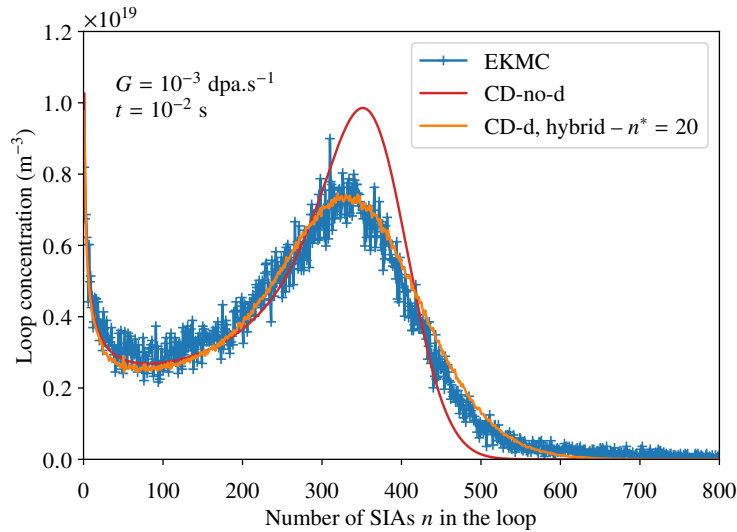


FIG. S1: Cluster distributions at $t = 10^{-2} \text{ s}$, obtained with EKMC and two different CD models: deterministic calculation without dispersion and Laplace expression for sink strengths (CD-no-d), hybrid deterministic-stochastic calculation using sink strength dispersion in the stochastic region (CD-d).