Dislocation bias factors in fcc copper derived from atomistic calculations

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Abstract

Atomistic calculations were employed in order to calculate the interaction energy of an edge dislocation with different point defects. The bias factor was calculated by applying a finite element method on the interaction energy landscapes obtained from the atomistic calculations. A comparison of the calculated bias factor with a model based on elasticity theory reveals around 30% discrepancy under conditions representative for electron irradiation at 600°C. Possible reasons are discussed. The bias factor dependence on dislocation density and ambient temperature is presented and discussed.

Keywords: Dislocation bias, Atomistic calculation, Interaction energy

1. Introduction

Austenitic steels are used in nuclear reactors as structural materials since they have excellent mechanical properties. Swelling under irradiation, how-

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4 ever, is a significant drawback that is especially pronounced in austenitic
5 steels [?]. The swelling of structural materials limits the lifetime of internal
6 reactor components and also the maximal fuel burn up [?]. Therefore, great
7 efforts have been devoted to improve the swelling resistance of austenitic
8 steels [? ? ?].

9 The 316 type stainless steel (SS316) is a typical austenitic steel that is
10 widely used for internal parts of light water reactors and has therefore been
11 comprehensively studied. Improvements of SS316 led to the development of
12 15-15Ti, which today is one of the best performing steels for nuclear reactor
13 applications [? ? ?]. Its incubation dose for swelling is greater than 100 dpa
14 [?]. The current design of fast reactors, however, requires resistance to even
15 higher doses. In the ASTRID sodium fast reactor, the cladding material
16 is expected to work for 8-10 years under temperatures above 700 K and
17 accumulate an irradiation dose of over 150 dpa [?]. Should a new, even
18 more durable steel be developed, it is of use to have an improved fundamental
19 understanding of the mechanisms of irradiation swelling.

20 Austenitic steels are face-centered cubic (fcc) alloys and in order to ad-
21 dress phenomena in such an alloy, it is convenient to begin by modeling an
22 fcc metal. Given the fact that nickel is the fcc stabilizing element in most
23 austenitic alloys, one would preferably use it as a model metal. Nickel, how-
24 ever, has not been extensively studied and there is a scarcity of experimental
25 data from electron irradiation. Copper, another fcc metal, on the other hand,
26 boasts a large database of irradiation experiments and is thus more suitable

27 for computational modeling of irradiation induced swelling. In addition, the
28 stacking-fault energy (SFE) plays an important role for dislocation proper-
29 ties. Copper has an SFE rather close to that of austenitic steels, while the
30 SFE of nickel is much higher.

31 One of the most popular models used for studies of dimensional changes
32 due to irradiation, is the standard rate theory (SRT) [? ? ?]. It is formulated
33 within the framework of the mean field type chemical reaction rate theory.
34 In this model, irradiation damage produces only Frenkel pairs and they are
35 created randomly in space and time. The concept of sink bias is introduced
36 in the model. One example of the sink bias, is the dislocation bias (B_d),
37 which implies different absorption rates of interstitials and vacancies at sinks
38 [?]. Another example is void bias, which is significantly smaller compared
39 to the dislocation bias [?]. For simplicity, the void bias is neglected in
40 the standard rate theory model [?], which implies that the dislocation bias
41 becomes the only driving force for micro structure evolution in the model.
42 The swelling rate at steady state depends on density and size of voids and
43 on dislocation bias.

44 The description of defect production in this model is accurate only for
45 the case when the energy of the injected particles is close to the displace-
46 ment threshold [?]. Neutron irradiation generates recoils with much higher
47 energy hence it induces clusters of point defects formed directly in cascades.
48 Therefore, the simplified SRT model is not suitable for modeling the effects
49 of neutron irradiation. For that purpose, a more sophisticated model like

50 the production bias (PBM) [? ? ?] model has been developed. Still, the
51 dislocation bias factor is still an integral part of the model.

52 A large number of experiments [? ? ?] and theoretical studies [? ?
53 ? ?] have been performed in order to assess factors controlling the bias.
54 In these studies, the SRT model was applied to fit the experimental data in
55 order to obtain B_d [?] and elasticity theory was used to derive the analytical
56 expression of sink strengths. The validity of such approaches can, however,
57 be questioned. The analytical interaction from elasticity theory, which is
58 derived from the first order size interaction, is not valid in the vicinity of
59 the dislocation core due to the mathematical divergence in approaching the
60 core. Furthermore, it has been long recognized that theoretically estimated
61 values of the dislocation bias generally exceed those based on fitting to ex-
62 perimental data. This indicates that the role of dislocation – point defect
63 (PD) interaction is not completely understood. In general, all calculations of
64 the bias factor rely on an elastic description of the dislocation – point defect
65 interaction. This would be sufficient if the dominating energy term ruling
66 the capture process stems from the interaction far from dislocation core. On
67 the other hand, the detailed processes of point defect diffusion close to the
68 core will at some point be of importance. It is therefore natural to ask if
69 deviations from elasticity theory and atomistic effects are important in de-
70 termining the dislocation bias. Edge dislocation – point defect interaction
71 energies have previously been calculated in Fe and Ni using an atomistic
72 approach[?]. In Cu and Mo elasticity theory has been applied in order to

73 calculate the point defect interaction energy close to the dislocation core[?
 74]. However, in these studies, the interaction energy maps were not used to
 75 obtain estimates of sink strengths or the dislocation bias.

76 In the present work, large scale atomistic simulations with empirical po-
 77 tentials were applied in order to map the dislocation – point defect interaction
 78 energy in an fcc copper lattice. Furthermore, the results were used in order
 79 to derive the dislocation bias and the swelling rate as a function of temper-
 80 ature and dislocation density. Thus the importance of the core region of the
 81 dislocation is addressed in terms of determining the bias and ultimately the
 82 swelling rate. More precisely, the ranges of temperatures and dislocation den-
 83 sities in which the core region is of importance are discussed. Consequently,
 84 the ranges in which it can be neglected are deduced.

85 **2. Theory and Methods**

86 *2.1. Bias factor*

87 The main assumptions of the simplest dislocation bias model are as follows
 88 [? ?]: 1) the incident particles only produce isolated Frenkel pairs (FPs), 2)
 89 both self-interstitial atoms (SIAs) and vacancies migrate in three dimensions,
 90 and 3) dislocations preferentially absorb SIAs, rather than vacancies, due to
 91 the stronger dislocation–SIA interaction. In this model, dislocations are the
 92 only sinks with a bias factor different than unity, and consequently the only
 93 driving force for the evolution of the micro-structure after electron irradiation.
 94 The dislocation bias is then the reason for the excess of vacancies in

the bulk and consequently for void swelling. The swelling rate is hence given by [?]:

$$\frac{dS}{d\phi} = k_v^2(Z_v^v D_v C_v - Z_i^v D_i C_i) \approx B_d \frac{k_c^2 Z_v^d \rho_d}{(k_c^2 + Z_v^d \rho_d)(k_c^2 + Z_i^d \rho_d)} \quad (1)$$

where $k_c^2 = 4\pi\langle R \rangle N$ and $B_d = \frac{Z_i^d - Z_v^d}{Z_v^d}$ with ϕ the irradiation dose in dpa, k_v^2 the sink strength of voids, Z^d the sink strength of dislocations (sometimes referred as capture efficiency), D the diffusion coefficient, and C the point defect concentration. The subscripts i and v represent interstitial and vacancy respectively and superscripts v and d mean void and dislocation, respectively. B_d is the dislocation bias factor, ρ_d is the dislocation density, $\langle R \rangle$ and N are the mean void radius and the void number density. The approximation in the equation assumes that the capture efficiency of voids is the same for both vacancy and interstitial, i.e. $Z_v^v = Z_i^v$.

The distortions caused by the formation of defects in crystal structure are described as stress fields in elasticity theory. The interactions of different defects originate from the overlap of stress fields. The flux of point defects due to diffusion and to the stress field interaction can be described by Fick's law including a drift term:

$$J = -\nabla(DC) - \beta DC \nabla E \quad (2)$$

where E is the interaction energy of dislocations with point defects.

In order to solve this equation, it is most convenient to reformulate it as:

$$J = -e^{-\beta E(r,\theta)} \nabla \Psi \quad (3)$$

113 where $\Psi = DCe^{\beta E(r,\theta)}$.

114 With the steady state condition $\nabla J = 0$, Eq.2 is reduced to the form:

$$\nabla^2 \Psi = \beta \nabla E \cdot \nabla \Psi \quad (4)$$

115 The assumption is that at the dislocation core, all point defects are ab-
 116 sorbed. Hence the boundary condition at the dislocation core $r = r_0$, is
 117 $\Psi_{r_0} = 0$. At the external boundary, i.e. the dislocation radius of influence,
 118 $r = R$, the defect concentration $C(r, \theta)$ is a constant and the interactions
 119 vanish. Hence, $\Psi_R = C^{\text{eq}}$ where C^{eq} is the concentration of point defects in
 120 the steady state.

121 Eq.4 is numerically solved by applying the finite element method, which
 122 is encoded in the MATLAB PDEtoolbox. The sink strengths and the bias
 123 factor were obtained by integrating the total flux around the dislocation core.
 124 The sink strength is defined as the ratio of PD fluxes with and without in-
 125 teraction with the dislocation, i.e. $Z = \frac{J}{J_0}$. J is the flux of PDs including the
 126 interaction with the dislocation and J_0 is the flux excluding the interaction.

127 The effect of the FEM geometry on the sink strengths and the bias factor
 128 was investigated using different inner absorption radii r_0 . The absorption
 129 boundary was applied either as a single circle around the two partial cores,
 130 or as two separate, touching circles, one around each core. For a given

absorption length, equal to the total circumference of the core absorption boundaries, the effect of choosing one or two boundaries was seen to be small, the difference varies from 0.2% to 6% depending on the temperature. The data here presented has the single core configuration with the inner radius of 38 Å, which is the same as the splitting distance.

2.2. Atomistic calculations

In Eq.4 the interaction energy is an important input parameter. The analytical expression for the interaction energy in elasticity theory has been derived from the first order size interaction of dislocations and point defects by treating the medium as being elastically isotropic [?]:

$$E = -A \frac{\sin \theta}{r} \quad (5)$$

where

$$A = \frac{\mu b}{3\pi} \frac{1 + \nu}{1 - \nu} |\Delta| \quad (6)$$

in polar coordinates (r, θ) . μ is the shear modulus, ν is Poisson's ratio, b is the Burgers vector, and, Δ is the dilatation volume of the PD.

To obtain the atomistic interaction, a model treating a periodic array of edge dislocations was applied. Two half crystals are strained to have different lattice parameters in the $\langle 110 \rangle$ direction, along the Burgers vector b . They join along the dislocation slip plane to form a misfit of b . More extensive details can be found in a dedicated study [?].

In order to model an infinite straight edge dislocation (ED), periodic

150 boundary conditions were applied in the direction of the Burgers vector b
 151 and in the direction of the dislocation line. A fixed boundary condition was
 152 applied normal to the glide plane. A vacancy is created by removing one atom
 153 from the lattice and relaxing the crystal. Dumbbell SIAs in $\langle 100 \rangle$ directions
 154 are created by adding one atom, perturbing slightly the atomic positions of
 155 the two nearest neighbor atoms and then relaxing. An initial distance of $0.4a_0$
 156 between the two relocated atoms is used. Considering both the boundary
 157 effects and the computational cost, a simulation box of 85980 atoms with
 158 the box dimensions of $99b \times 10.4b \times 58.8b$ in the $[110]$, $[\bar{1}1\bar{2}]$, $[\bar{1}11]$ directions,
 159 was set up. This gives a dislocation density of $2.6 \cdot 10^{15} \text{ m}^{-2}$. A combination
 160 of conjugate gradient and quasi static relaxation with constant volume was
 161 applied in the ideal dislocation structure.

162 Large scale molecular statics calculations were performed using the DYMOKA
 163 code [?]. Full interaction energy landscapes around the dislocation core for
 164 PDs were obtained using a semi-empirical embedded atom method (EAM)
 165 potential for Cu [?]. This potential reproduces the properties of defects in
 166 good agreement with experimental data. The stacking fault energy obtained
 167 from the potential is 44 mJ/m^2 , the vacancy dilatation volume is -0.3Ω and
 168 the interstitial dilatation volume is 1.8Ω , where Ω is the atomic volume.

169 Different dislocation densities were generated by expanding the region
 170 described by the atomistic model and matching it to the analytical solution
 171 in the outskirts. In this manner the near core region is described as accurately
 172 as possible while at the same time one can obtain dislocation densities on

173 the same order of magnitude as in technological materials. The effect of the
174 fixed boundary and that of the periodic image interaction along the glide
175 direction were corrected for in the atomistic simulation.

176 **3. Results and discussion**

177 *3.1. Interaction energy*

178 After the relaxation of a perfect dislocation, a splitting distance of 38
179 Å was obtained for the partial dislocation cores. The interaction energies
180 of SIAs and vacancies with the dislocation are depicted in Fig.1 both from
181 the atomistic model and the elastic model. The SIA interaction map is
182 produced by averaging the results for three different dumbbell orientations,
183 namely [100], [010] and [001]. Given that the original analytical expression
184 is derived from a single core dislocation, a model of two partial cores, each
185 with a Burgers vector $b/2$ has been constructed. The superposition of the
186 interaction energy Eq.5 due to the two cores forms the analytical elastic
187 interaction model.

188 An assessment of the interaction between an SIA and the dislocation core
189 reveals a significant discrepancy between the analytical solution, treating
190 SIAs as isotropic objects, and results of the atomistic calculations, as can be
191 seen in Fig.1. The atomistic description of the SIA–dislocation interaction in
192 the core and stacking fault (binding the two partial cores) regions is essential
193 given the anisotropy of Cu, considered here, and for metals in general.

Moreover, in the attractive side of the dislocation, in the case of interstitials, the interaction is stronger in the atomistic case compared to the analytical expression. Qualitatively, this can be understood because it is energetically easier to expand the lattice than to compress it far beyond the linear regime. For the vacancy, the difference is generally smaller. This leads to a larger dislocation bias in comparison with the analytical results.

3.2. Bias factor

By applying the atomistic interaction energy maps in Eq.4, numerical solutions of Ψ were obtained on the dislocation plane. The calculated sink strengths and bias factor without the expansion at 873 K are demonstrated in Tab.1. The major difference in sink strength and bias factor, of about 30%, from the two approaches stems from the difference in the SIA–dislocation interactions as shown in Fig.1.

Although the crystal size, used in the molecular static calculations, is large enough to accurately describe the dislocation core region, the implied dislocation density is 2–3 orders of magnitude higher than the one typical for real steels. By merging the energy landscape near the dislocation core, obtained through the atomistic simulation, with the elastic solution far away from the core, which has been proven to be accurate enough, the effect of the dislocation density on the value of B_d is characterized. It is shown in Fig.4 that B_d rises steeply if dislocation density exceeds 10^{14} m^{-2} . Such a dislocation density can be reached by, for example, cold work. However, in

216 service conditions at elevated temperatures, recovery would occur to lower the
 217 dislocation density. Nevertheless, neutron irradiation generates dislocation
 218 loops whose contribution to the total dislocation density at a certain dose
 219 becomes comparable and then even higher than the initial dislocation density.
 220 Importantly, dislocation loops contain a different type of Burgers vector (i.e.
 221 $\langle 100 \rangle$ in bcc Fe-based steels and $1/3 \langle 111 \rangle$ in fcc FeNiCr-based steels) and
 222 character than the pre-existing dislocations. That is, most of the observed
 223 irradiation induced loops are of edge type. Whereas initial dislocations are
 224 usually of screw character with $b=1/2 \langle 111 \rangle$ in bcc and $1/6 \langle 112 \rangle$ in fcc metals.
 225 Hence, it is essential to make an appropriate evaluation of B_d for dislocation
 226 loops, since they become the predominant objects controlling the swelling
 227 process.

228 To assess to which extent the superposition of the analytical interac-
 229 tion would be in line with the correct description of a split dislocation core,
 230 B_d is calculated for different configurations. Fig.2 shows the results. The
 231 fully analytical solution, which depends only on the geometry of the dislo-
 232 cation system and the dilatation volumes of the point defects, is presented
 233 by Dubinsko et al [?]. This analytical solution of the sink strength has
 234 approximated boundary conditions. The approximation that interactions on
 235 the outer boundary is negligible is not valid for high dislocation densities.
 236 The FEM approach overestimates the B_d by 1% for densities lower than 10^{14}
 237 m^{-2} . The geometrical shape of the outer boundary does not seem to play a
 238 role, while the core configuration, namely one single full core or two partial

239 half cores, does make a difference of around 0.5% in B_d values. Thus the
240 superposed analytical model represents the essence of the original analytical
241 model.

242 Sink strengths were calculated under different temperatures and dislo-
243 cation densities by applying both the atomistic interaction energy and the
244 elastic analytical interaction formula. The results are shown in Fig.3 and
245 Fig.4. Both the atomistic and the analytical B_d decrease as the tempera-
246 ture increases. They tend to converge at the high temperature limit. The
247 reason for that could be the contribution from the dislocation cores. As
248 temperature increases, diffusion and thermal fluctuations become the domi-
249 nant driving force for the flow of PDs. Therefore, in Eq.2, the contribution
250 from the drift term decreases. The dislocation core region does not play as
251 important a role as it does at lower temperatures. Thus the atomistic and
252 analytical approaches converge in the high temperature limit. On the con-
253 trary, at low temperatures, segregation and nucleation plays an important
254 role for the bias, hence the simplified model does not apply for an accurate
255 bias calculation.

256 It worth noticing that a strong temperature dependence of B_d is demon-
257 strated, which is important to account for when considering real structural or
258 functional reactor components, which exhibit strong temperature gradients
259 upon operation.

260 Fig.4 shows that B_d increases as the dislocation density increases. At
261 higher temperatures, B_d is more weakly dependent on dislocation density

262 than at lower temperatures. The atomistic model, again, has a higher deriva-
 263 tive than the analytical model. At the low density limit, the calculated bias
 264 factor from atomistic and analytical interaction energy models tend to con-
 265 verge. For high dislocation densities the difference is significant. The possible
 266 reasons are that, with low dislocation density, diffusion of point defects are
 267 prioritized. On the other hand, at high dislocation densities, the dislocation
 268 core – point defect interactions are dominating, hence the precise core de-
 269 scription becomes more important in this case. In fact, in real fast reactor
 270 conditions, high irradiation doses generates high dislocation densities. There-
 271 fore, the high density B_d results are of technological interest. The approach
 272 which has been developed here can readily be applied to dislocation loops,
 273 which have high generation rates in cascade inducing fast neutron spectra.

274 In order to compare these results with measurements, experimental void
 275 density and void size data were selected. Together with the sink strengths
 276 obtained from the atomistic approach, the swelling rate is calculated in three
 277 different irradiation dose conditions. The experimental data is from Singh
 278 et al. [?]. The electron- and proton irradiation data were selected for the
 279 reason that the fraction of defects produced in clustered configurations in
 280 those cases were sufficiently small for the dislocation bias model to be valid.

281 In Table.2 the reference swelling is the one obtained from experiments
 282 [?]. The calculated swelling is obtained by applying the sink strengths
 283 from the atomistic calculation to the SRT model at the same temperature
 284 and dislocation density as in the experiments. The discrepancies are within

285 expected range considering that the SRT model here applied is a simple
286 steady state model while transient period has been taken into account in
287 the experimental fitting. Furthermore, the mean radius of voids is used here
288 while in experimental fitting the distribution of voids sizes are used.

289 Due to the feasibility of combining atomistic calculations with numeri-
290 cal solutions of the diffusion equations, the applied method here can also be
291 used in order to estimate the bias not only for dislocation-like objects, but in
292 general for other objects that are considered as sinks for point defects, such
293 as grain boundaries or non-coherent precipitates (e.g. ODS particles). More-
294 over, in technological steels, impurity segregation to the dislocation core can
295 further modify the interaction energy landscape. The atomistic approach, as
296 applied here, can readily be a solution to evaluate the impact of segregation
297 effects on B_d , given an appropriate cohesive model.

298 4. Conclusions

299 In this work, a atomistic calculation of the edge dislocation – point defect
300 interaction energy map in fcc copper was performed and applied to calcu-
301 late the bias factor. A combined method, of atomistic calculations imposed
302 on a finite element method based numerical solution of Fick’s law with a
303 drift term, was elaborated in order to determine the sink strengths of the
304 dislocation. In general, the sink strength is higher for interstitials than for
305 vacancies. The same result applies for the interaction energies, that is, inter-
306 stitials have higher interaction energies with the dislocation than vacancies

do. Furthermore, the bias factor, as well as the interaction energies, obtained from the atomistic approach are compared with the ones obtained using an analytical model derived from elasticity theory. The dislocation bias predicted by the atomistic approach is higher than that predicted by the elastic approach, especially for high dislocation densities. This implies that an atomistic description of the dislocation – point defect interaction in fcc materials is important for high dislocation densities. It is also shown that the deviation, with respect to the atomistic approach, of the interaction energies obtained from elasticity theory is larger for interstitials than for vacancies. Furthermore, the strongest deviation is at the attractive (tensile) side of the dislocation in the case of the interstitials. This is the reason the dislocation bias factor predicted by the atomistic approach is higher than that predicted by elasticity theory.

Figure 1: Edge dislocation – point defect interaction energy maps for the different approaches, A) Atomistic SIA; B) Analytical SIA; C) Atomistic vacancy; D) Analytical vacancy; E) Difference between A and B; F) Difference between C and D.

Figure 2: Analytical solution of B_d at 873 K as a function on dislocation density considering different geometries and methods. Illustration of different boundaries are shown in inner figures. A. one core with circle boundary; B. two cores with square boundary; C. one core with square boundary. Red circles represent the inner core, black circle and squares represent the outer boundaries.

Figure 3: Temperature dependence of B_d for the atomistic and analytical cases.

Figure 4: Dislocation density dependence of B_d for the atomistic and analytical cases at two temperatures.

Table 1: SIA (Z_i^d) and vacancy (Z_v^d) sink strengths calculated at 873 K and a dislocation density of $2.6 \cdot 10^{15} \text{ m}^{-2}$.

	Z_i^d	Z_v^d	B_d
Atomistic	1.34	1.00	0.34
Analytical	1.25	1.00	0.25

Table 2: Experimental swelling induced by electron– (1st row) and proton (2nd, 3rd rows) irradiation compared to steady state evaluation using the atomistic approach.

dose (dpa)	Reference swelling (%) [?]	Calculated swelling (%)
0.013	$0.2\text{--}1\cdot 10^{-3}$	$6.7\cdot 10^{-3}$
0.002	$2.8\cdot 10^{-4}$	$5.3\cdot 10^{-4}$
0.008	$1.9\cdot 10^{-3}$	$0.95\cdot 10^{-3}$