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A variable-gap model for helium bubbles in nickel

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Abstract

In nuclear fission reactors, the amount of helium produced in materials by transmutation reactions at the end of the lifetime may reach several thousands of atomic parts per million (appm). Such high levels of helium production can impact the evolution of microstructures, particularly by forming helium bubbles. To better understand the role of helium on the stability of bubbles, a “variable-gap model” was parametrized with molecular dynamics (MD) calculations performed in nickel. This model predicts binding energies in a good agreement with MD values, especially for large bubbles. For very small bubbles, the influence of magic number sizes and faceting is more complex than can be described with the model. For these cases, it is proposed to use MD values directly.

Keywords: helium, bubbles, nickel, binding energy

1. Introduction

Helium production and formation of bubbles play an important role in microstructure evolution under neutron irradiation [1]. In fission reactors, helium is produced by transmutation reactions mostly from nickel interacting with thermal neutrons. Under such conditions, material is subjected to a neutron

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6 flux which causes substantial changes in the microstructure. High energy neu-
7 trons, so-called *fast* neutrons, cause displacements of atoms, creating vacancy-
8 interstitial (or Frenkel) pairs (FPs). Point-defects from FPs cluster into self-
9 interstitial atoms (SIAs) clusters and voids. Lower energy neutrons, so-called
10 *thermal* neutrons, may interact with nickel atoms and form helium by trans-
11 mutation of nickel into iron. Another less significant source of He is boron,
12 contained in steels in small amounts as impurities. The amount of helium pro-
13 duced is usually expressed in terms of a helium-to-displacement per atom ratio
14 (He/dpa). Helium production ranges in ASS from 0.1-0.8 appm He/dpa in
15 fast breed reactors [2, 3], to about 10 appm He/dpa [4, 5, 6, 2] in pressurized
16 water reactors (PWR), and about 70 He appm/dpa in HFIR [7, 8]. In nickel
17 based alloys, He production can reach more than 300 appm/dpa in CANDU
18 reactors [9].

19 As a noble gas, helium is insoluble in the material, and in an interstitial
20 position it can move easily in the bulk [10]. It is therefore quickly captured
21 by sinks - mostly bubbles, but also other defects such as dislocations and grain
22 boundaries [11, 12, 13, 14]. Theoretical calculations showed that small voids
23 in metals are metastable [15], and that the presence of gaseous atoms would
24 explain their experimental evidence [16]. Crucial ideas on modelling helium
25 accumulation in metals were reviewed by Trinkaus et al. [17]. Helium in bubbles
26 increases internal pressure, and therefore thermal stability, by reducing vacancy
27 emission. This should favour bubble nucleation, but the reality is more complex
28 and under irradiation, factors favouring nucleation may lower the growth of
29 bubbles.

30 Growth kinetics of bubbles must be modelled more accurately and quanti-
31 tatively with kinetic Monte-Carlo [18, 19] or with a rate equations approach
32 such as cluster dynamics (CD) [20, 21, 22, 23, 24, 25, 26]. In a CD approach,
33 emission coefficients are calculated using binding energies, and they describe at
34 which rates vacancies, SIAs and helium atoms are emitted from bubbles.

35 Such methods require precise parametrization of binding energies at arbi-
36 trary large sizes, and it is thus particularly important to have a model [27, 28, 29]

37 that calculates binding energy over a wide range of sizes and He densities, specifi-
 38 cally in terms of the helium-to-vacancy (He/vac) ratio, which significantly varies
 39 under different experimental conditions. The He production rate varies in dif-
 40 ferent materials under different neutron spectra, and the stability of bubbles
 41 can be quite different for different He/vac ratios.

42 In this work, a variable-gap model predicting binding energies to helium
 43 bubbles, initially developed for body-centered cubic (BCC) iron [27], is adapted
 44 based on data from molecular dynamics (MD) simulations in nickel. Nickel can
 45 provide an important foundation on the behaviour of helium bubbles in Ni-based
 46 alloys [30], but also Fe-based FCC alloys, as behavior of He is expected to be
 47 similar to Ni [31, 32]. A particular challenge arises from using nickel to simulate
 48 defect clusters at finite temperatures, as a recent combined density functional
 49 theory and MD simulation study shows that voids are unstable [33].

50 Section 2 reviews the description and assumptions of the energy model. The
 51 description of simulation settings is presented in section 3, followed by the adap-
 52 tation of the model on obtained MD data in section 4, commenting on several
 53 differences and limitations compared to the Fe model. Then, equilibrium helium
 54 density predicted by the model is compared with available experimental data.

55 2. Energy model

56 A bubble that contains m vacancies and n helium atoms is labeled in paren-
 57 theses (m, n) . The main assumption is that a void has a spherical shape [34, 35]
 58 and that repulsion between helium and metal atoms is modeled as a variable-
 59 size gap between them [27]. The free energy of a bubble (m, n) is a sum of
 60 three different contributions: elastic energy of Ni atoms around the bubble
 61 $F_{\text{Ni-Ni}}(m, n)$, interaction of He and Ni atoms $F_{\text{Ni-He}}(m, n)$, and energy of he-
 62 lium atoms $F_{\text{He-He}}(m, n)$ described with an equation of state (EOS):

$$F^{\text{f}}(m, n) = F_{\text{He-He}}(m, n) + F_{\text{Ni-He}}(m, n) + F_{\text{Ni-Ni}}(m, n). \quad (1)$$

63 We can define

$$\Delta F^f(m, n) = F^f(m, n) - F^f(m, 0), \quad (2)$$

64 where $F^f(m, 0)$ is the surface free energy, so

$$\Delta F^f(m, n) = F_{\text{He-He}}(m, n) + F_{\text{Ni-He}}(m, n) + \Delta F_{\text{Ni-Ni}}(m, n), \quad (3)$$

65 where $\Delta F_{\text{Ni-Ni}}(m, n)$ is due to the surface relaxation. We assume that because
 66 of the gap, the helium content will have no effect on the surface energy itself,
 67 but the gap will induce stretching of surface atoms arising from an elastic effect
 68 accounted in $\Delta F_{\text{Ni-Ni}}(m, n)$. Then, $\Delta F^f(m, n)$ can be defined as the formation
 69 free energy of the bubble.

The binding energy of a vacancy (V), an SIA (I), or a helium (He) atom is then calculated using formation energy $F^f(m, n)$ as

$$F_V^b(m, n) = F_V^f + F^f(m - 1, n) - F^f(m, n), \quad (4)$$

$$F_I^b(m, n) = F_I^f + F^f(m + 1, n) - F^f(m, n), \quad (5)$$

$$F_{\text{He}}^b(m, n) = F_{\text{He}}^f + F^f(m, n - 1) - F^f(m, n), \quad (6)$$

70 where F_V^f , F_I^f , and F_{He}^f are the formation energies of vacancy, SIA, and helium
 71 atom in a tetrahedral position, respectively. Parameters for these three terms
 72 were fitted on data extracted from MD simulations performed in face cubic
 73 centered (FCC) nickel using LAMMPS code [36], and the potentials to describe
 74 each interaction were: Bonny [37] for Ni-Ni, Torres for Ni-He [10], and Beck
 75 potential for He-He [38].

For fitting purposes, we assume that the function that sums up three different free energy contributions of the bubble is found as the minimum of the following function:

$$\Delta \Phi^f(m, n, r_{\text{He}}, r_V) = \Phi_{\text{He-He}}(n, r_{\text{He}}) + \Phi_{\text{Ni-He}}(n, r_V - r_{\text{He}}) + \Delta \Phi_{\text{Ni-Ni}}(m, r_V), \quad (7)$$

76 where r_{He} and r_V are helium's and the void's radii, respectively (Fig. 1). He-He
 77 interactions tend to maximize r_{He} while the $\Phi_{\text{Ni-Ni}}$ opposes to the increase of r_V .

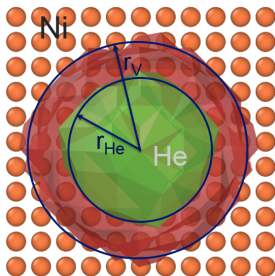


Figure 1: A typical snapshot of actual MD simulations. For visibility, helium atoms are hidden and replaced by the location of the surface of helium atoms (in green). The surface Ni atoms are also replaced by a calculated surface (in red). The structure is visualized using OVITO software [39] and *Construct surface mesh* modifier. The gap $r_{\text{Ni-He}}$ is the difference between the void's radius r_V and helium's radius r_{He} .

78 The minimum of the function $\Delta\Phi^f(m, n, r_{\text{He}}, r_V)$ (Eq. 7) with respect to r_{He}
 79 and r_V under the constraint $r_{\text{He}} < r_V$ will be equal to the formation free energy
 80 of a bubble $\Delta F^f(m, n)$ defined in Eq. 3. In this work we adapt this model
 81 to bubbles in Ni, using MD simulations to parametrize each term of Eq. 7, as
 82 described in the following sections.

83 2.1. He-He interaction

84 A possible way to describe the interaction energy of helium atoms in a bubble
 85 is with an EOS in terms of bulk atoms with a surface correction. An alternative
 86 approach used by Jelea [40] alters a bulk helium virial EOS by introducing a
 87 spherical confinement volume inside the bubble that accounts for the (repulsive)
 88 effects of the surface.

89 In this model, He-He interaction energy is described by Vinet EOS [41]. It
 90 was noted that alternative equations of state might be used for specific cases,
 91 but the fundamental behaviour doesn't change dramatically [42].

92 Bulk He atoms are identified using Voronoi volumes. For each helium atom,
 93 its Voronoi volume is calculated with Voronoi's tessellation for two cases - with
 94 and without Ni atoms. If the change of Voronoi's volume is negligible, an atom
 95 is considered a bulk atom. The Voronoi volume of helium can be computed for

96 a bubble with a minimum of 5 helium atoms, and for a bubble containing a bulk
 97 atom with a minimum of 15 helium atoms. The mean volume of helium v_{He} is
 98 computed as an average Voronoi volume of He bulk atoms. The total energy of
 99 He-He interactions extracted from MD runs at 0K is plotted in Figure 2 as a
 100 function of mean helium volume and compared with expression

$$e_{\text{He-He}}^{\text{bulk}}(v_{\text{He}}) = \int_{v_{\text{He}}}^{v_{\infty}} p_0 dv, \quad (8)$$

which describes helium bulk energy as an integral of the pressure term p_0 at

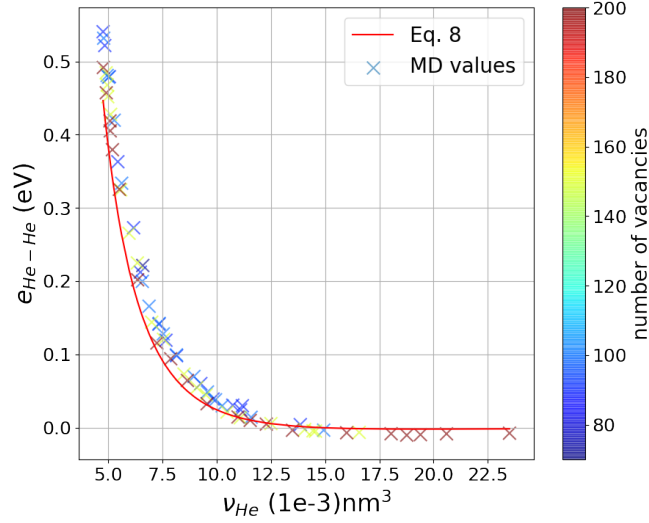


Figure 2: Energy of helium bulk atoms due to He-He interactions of different bubbles sizes and He to vacancy ratios, as a function of the mean He volume. MD values (crosses) are compared to Eq. 8 (solid line). Coloring corresponds to the number of vacancies in the bubble. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

101

102 0K from the mean bulk helium volume v_{He} , up to infinite dilution. Vinet EOS

$$p_0 = \frac{3K_0}{X^2}(1 - X)\exp(\eta_0(1 - X)), \quad (9)$$

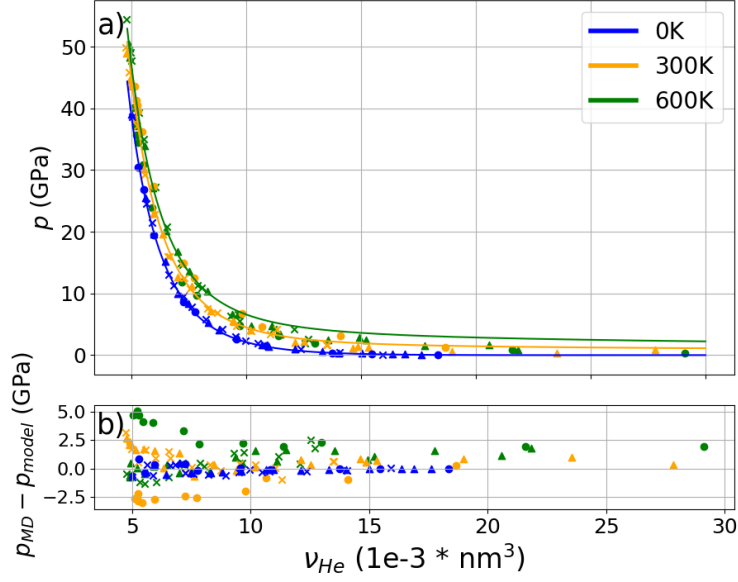


Figure 3: a) internal pressure in bulk helium with respect to average bulk helium volume, model (solid lines) compared to the values extracted from MD (points), b) the difference between the model and MD values at the following temperatures: 0 (blue), 300 (orange), and 600 K (green), for different bubble sizes (crosses: $n = 100$; triangles: $n = 150$, and circles $n = 200$). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

103 where

$$X = \left(\frac{v}{v_0}\right)^{1/3}, \eta_0 \frac{3}{2}(K'_0 - 1), \quad (10)$$

104 was used with the same parameters as in [27] ($v_0 = 1.951 \cdot 10^{-2} nm^3$, $K_0 =$
 105 $1.497 eV/nm^3$, and $K'_0 = 8.465$) and it was verified that Eq. 8 gives reasonable
 106 results compared to the MD data (Figure 2).

107 One can express the ratio between energy of all atoms and bulk helium atoms
 108 due to He-He interaction with the help of the surface correction parameter α :

$$\frac{e_{He-He}}{e_{He-He}^{bulk}} = 1 - \frac{\alpha}{n^{1/3}}. \quad (11)$$

109 To take into account the effect of temperature, a temperature-dependent term [29]
 110 is added to the 0K expression for pressure. The model was validated by checking
 111 pressure at $T = 0\text{K}$, 300K , and 600K (Fig. 3). Average pressure for He bulk
 112 atoms was calculated using LAMMPS' stress tensor described in [43]. The bulk
 113 free energy can be written as

$$f_{\text{He-He}}^{\text{bulk}}(v_{\text{He}}) = \int_{v_{\text{He}}}^{v_{\infty}} p dv + f_{\text{He-He}}^{\text{ideal}}(v_{\infty}). \quad (12)$$

114 A sufficiently high value of v_{∞} was chosen so that the ideal gas approximation
 115 is valid ($p = 10^3\text{Pa}$) and so that the free energy of an ideal gas $f_{\text{He-He}}^{\text{ideal}}(v_{\infty})$ is
 116 valid.

Helium radius is deduced from a sphere with a volume equal to the number
 of He atoms multiplied by the average helium volume v_{He}

$$nv_{\text{He}} = \frac{4}{3}\pi r_{\text{He}}^3, \quad (13)$$

117 so the final term of the free energy that corresponds to He-He interactions
 118 expressed as a function of He radius r_{He} has the following form:

$$\Phi_{\text{He-He}}(n, r_{\text{He}}) = n f_{\text{He-He}}^{\text{bulk}}(r_{\text{He}}) \left(1 - \frac{\alpha}{n^{1/3}}\right). \quad (14)$$

119 2.2. Ni-He interaction

120 In our approach, following Jourdan and Crocombette [27], Ni-He interaction
 121 energy is derived directly from the potential φ and positions of atoms over all
 122 Ni-He pairs

$$E_{\text{Ni-He}}(n, m) = \sum_{\text{Ni-He interaction pairs}} \varphi(r_i). \quad (15)$$

123 Most of the energy comes from the interaction at the surface. In this model,
 124 it is convenient to express nickel-helium interaction energy as a function of
 125 *characteristic* distance $r_{\text{Ni-He}}$ such that

$$\Phi_{\text{Ni-He}}(n, r_V - r_{\text{He}}) = n^{2/3} g(r_{\text{Ni-He}}) \varphi(r_{\text{Ni-He}}), \quad (16)$$

126 where g is a function that will be defined in section 4.2, and with $r_{\text{Ni-He}}$ being
 127 defined as the weighted radius of Ni-He pair interaction

$$r_{\text{Ni-He}} = \frac{\sum r_i \varphi(r_i)}{\sum \varphi(r_i)}. \quad (17)$$

128 2.3. Ni-Ni interaction

129 It was assumed that Ni-Ni contribution comes from matrix bulk relaxation.
 130 We generalized the formula based on Eshelby's inclusion for a spherical void
 131 in an isotropic material, to an anisotropic material (see Appendix B), leading
 132 to the following expression for the elastic energy around a void of theoretical
 133 radius r_{V}^0 ¹ due to surface relaxation $r_{\text{V}} - r_{\text{V}}^0$:

$$\Delta\Phi_{\text{Ni-Ni}}(m, r_{\text{V}}) = 8\pi\mu r_{\text{V}}^0 (r_{\text{V}} - r_{\text{V}}^0)^2, \quad (18)$$

134 where μ is an effective shear modulus (see Appendix B), and the difference
 135 $\Delta r = r_{\text{V}} - r_{\text{V}}^0$ reflects the void's radius change due to helium pressure. The Δr
 136 values were directly extracted from MD, with the algorithm that is described
 137 in Appendix A. The effect of helium is hidden in the r_{V} value, which is the
 138 actual radius of the bubble.

139 3. Setting up the simulation

140 Simulations were performed in a simulation box of a size $30a_0 \times 30a_0 \times 30a_0$
 141 (with $a_0 = 0.352$ nm). It was verified that the box size is sufficient to avoid
 142 periodic boundary condition interaction for defects up to 200 vacancies in size.
 143 Potentials used are mentioned in section 2.

144 Initially, MD should have been used to explore energy landscape of bubbles,
 145 but it was found that in the case of nickel, bubbles are not thermodynamically
 146 stable below a given He/vac ratio, and transform into stacking fault tetrahedra

¹Theoretical value r_{V}^0 is deduced from the void's volume as the number of vacancies n multiplied by atomic volume V_{at} .

147 (SFT)-like structures. This transformation was observed in FCC Cu during ac-
148 celerated MD (parallel-replica dynamics) of vacancy clusters [44]. It was also
149 observed using first-principles calculations based on density-functional theory
150 (even after much shorter times - ps) at temperatures up to 1000K in Ni, whereas
151 at 0K voids are more stable than SFTs [33]. These results suggest the impor-
152 tance of possible temperature effects in concluding thermodynamic stability of
153 vacancy clusters in FCC metals. Although quite general in irradiated pure fcc
154 metals, the presence of SFTs is much more controversial for more complex alloys
155 such as 304 and 316 ASS [45], which are used in typical applications.

156 As our goal is to determine formation energies of bubbles with helium, we
157 want to simulate spherically shaped bubbles, and thus be ensured that trans-
158 formation into SFT due to thermal activation did not occur.

159 Firstly, spherical voids (up to the size of 200 vacancies) were created, by a
160 sequence of removing atoms and relaxing structures with the conjugate gradient
161 (CG) algorithm. For voids up to 20 vacancies, atoms with the highest potential
162 energy were removed. For bigger voids, to keep the spherical shape of voids,
163 atoms closest to the center were removed. We are aware of the fact that this
164 particular process of creating void structures can place potential energy at some
165 point (local minimum) of energy landscape that does not have to be the global
166 minimum, but it was considered sufficient, as thermostating at some higher
167 temperature would lead to transformation towards SFT. Afterwards, helium
168 atoms were inserted into voids, filling them with various helium content, from 0
169 to a He/vac ratio equal to 2.5, followed by CG relaxation. This was the starting
170 configuration for the actual MD. For bubbles with more than 150 vacancies and
171 a He/vac ratio bigger than 2.5, emission of SIAs was observed.

172 The conditions were then set to allow for thermostating at moderate tem-
173 peratures. After the atoms' velocity scaling reached the temperature of 100K²
174 (50 000 steps by 0.2 fs in NVT ensemble), the system switched to NVE ensemble

²Higher temperatures 300K and 600K indicated structure changes, and so some effect of thermal stability

175 for the same number of steps³. Every 500 time steps, positions of atoms were
176 saved and later relaxed with CG. Out of all of the relaxed structures from the
177 MD run, the one with the lowest energy was chosen.

178 The effect of helium on the bubble’s radius was extracted from the lowest
179 energy configuration, comparing it with the radius of the bubble that had He
180 atoms removed and subsequently was relaxed.

181 4. Adaptation of the model to the FCC case

182 4.1. He-He interaction

183 It was verified that the bulk part of helium energy agrees with the values
184 from MD. However, it was found that the value of α (surface correction term)
185 is equal to 2.038 (Fig. 4), which is higher than in the previous studies in BCC
186 Fe by Jourdan [27] ($\alpha = 1.354$) and by Morishita [18] ($\alpha = 1.37$). The value
187 1.37 was derived from the number of deficit bonds for a spherical FCC cluster
188 in the limit of a large cluster size [46]. Since the bubble’s surface is faceted,
189 the different value of α may point to an effect of the bubble’s shape on Ni-He
190 interaction and thus on the surface correction term.

191 4.2. Ni-He interaction

192 A function that accounts for the change in the number of Ni-He bonds as
193 $r_{\text{Ni-He}}$ changes,

$$g(r_{\text{Ni-He}}) = \frac{g_0}{\exp\left(\frac{r_{\text{Ni-He}} - r_0}{\Delta r}\right) + 1}, \quad (19)$$

194 was fitted on the MD data (Fig. 5), yielding $g_0 = 12.53$, $r_0 = 3.7$ nm, and
195 $\Delta r = 0.22$ nm. Compared to α -Fe, Ni-He interaction energy is lower for bubbles
196 with a gap smaller than 0.28 nm, and higher for bubbles with a gap bigger than
197 0.28 nm. Standard deviation of the distribution $P(r)\varphi(r)$ (radial distribution
198 function $P(r)$ represents the density of bonds, $\varphi(r)$ is the interatomic potential

³No transformation to SFT was observed

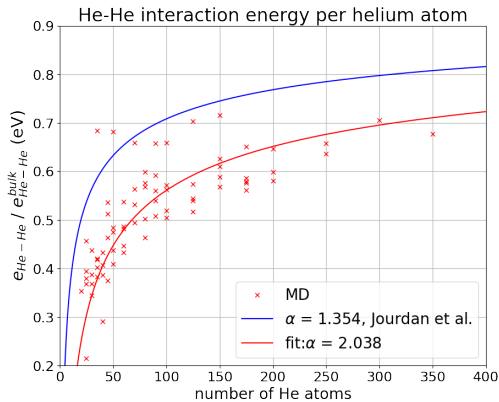


Figure 4: Ratio of energy of all helium and bulk helium atoms due to He-He interactions of different bubbles sizes as a function of number of the helium atoms n in the bubble, with its fit for surface correction in the current study (red line) compared to a value in ref [27] (blue line).

199 for Ni-He) can be used to account for spread of the density of bonds in terms
 200 of distance around $r_{\text{Ni-He}}$:

$$\sigma(r_{\text{Ni-He}}) = \sigma_1 r_{\text{Ni-He}} + \sigma_0, \quad (20)$$

201 with fitted parameters $\sigma_1 = -0.721$ and $\sigma_0 = 2.931$ nm (see section 4.4).

202 4.3. Ni-Ni interaction

203 Eq. 18 describing matrix bulk relaxation was compared to energies from MD.
 204 In practice, two quantities can be extracted from MD - atomic strain tensor
 205 (comparing positions of Ni atoms with a case where He atoms were removed
 206 and bubble was relaxed using CG, later used to compute elastic energy), or the
 207 change in total Ni-Ni interaction energy. The former estimates the elastic energy
 208 and has the advantage of having information on the contribution of each atom
 209 (relaxed elastic energy due to He atoms in the bubble). The latter collapses
 210 into one macroscopic value. Unfortunately, none of these were able to capture
 211 the exact trend of the energy with respect to the Δr value, as shown in Fig. 6.

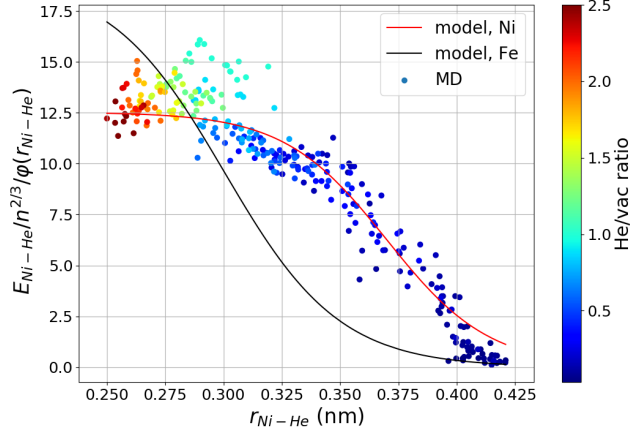


Figure 5: Normalized Ni-He interaction energy. Values from MD (circles) are fitted to Eq. 19 for nickel (red solid line), compared with α -Fe (black solid line) [27]. Coloring refers to helium to vacancy ratio. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

212 Further analysis of bubbles' surfaces revealed non-negligible faceting typical
 213 of octahedral-like shapes, as experimentally observed in FCC alloys [1, 47, 48].
 214 This shows that the assumption of sphericity in this study was not precisely
 215 met. To be more quantitative, we may correlate this with the difference between
 216 equilibrium crystal shapes given by the Wulff construction at $T = 0\text{K}$ of Fe and
 217 Ni, using a ratio between surface and volume $\eta = A/V^{2/3}$. A reference value
 218 for a sphere is $\eta_{\text{sphere}} = (36\pi)^{1/3} = 4.83$, while for α -Fe it is $\eta_{\text{Fe}} = 4.94$, and
 219 $\eta_{\text{Ni}} = 5.18$ for nickel [49]. In BCC, equilibrium shape consists of more higher
 220 order planes (more spherical), whereas in FCC it mostly consists of $\{111\}$ and
 221 $\{100\}$ planes, having less spherical shape due to the low surface energy of these
 222 planes [50].

223 An algorithm described in Appendix A was used to detect the surface's
 224 orientation by finding the closest crystallographic direction to its normal. A
 225 majority of surfaces were detected as $\{111\}$ and $\{100\}$ planes. Small voids
 226 were mostly composed of $\{111\}$ planes/facets, while with increasing size, the

227 surface grew in complexity. The shape looks like an experimentally observed
 228 truncated octahedron. It is expected that with increasing size, the surface will
 229 be composed of numerous facets of higher order planes, as shown by the Wulff
 230 construction for equilibrium crystal shape of Ni using values for surface energies
 231 calculated by DFT [33]. Figure 6 (b) shows that the discrepancy is linked to the
 232 surface orientation and thus to the faceting. Although the discrepancy for some
 233 configurations seems to be in the order of tens of eV, it should be noted that the
 234 model is used to calculate the binding energies (Eq. 4- 6), and therefore only
 235 the increment of the Ni-Ni part (Eq. 18) of the formation energy is relevant, as
 236 r_V varies due to the emission of a vacancy or a helium atom. The error of this
 237 quantity is much lower.

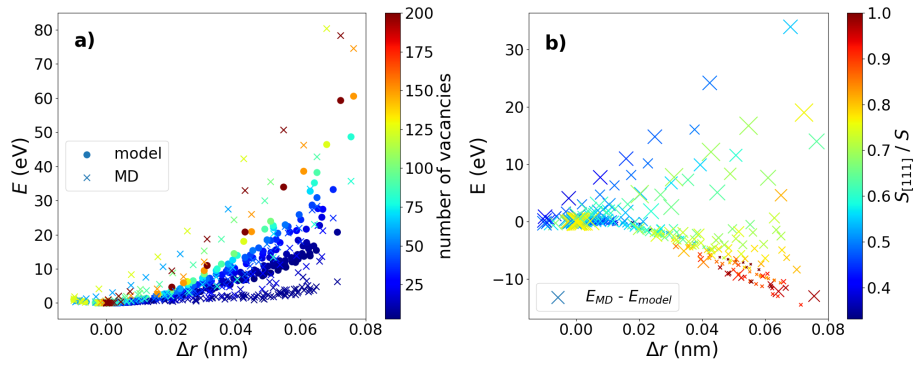


Figure 6: (a) Energy due to the elastic relaxation (spheres) and change in the energy extracted from MD (crosses) with respect to an increment of radius (coloring: bubble size), and (b) its difference (coloring: ratio of bubbles' surfaces oriented in [111] direction with respect to overall surface, marker size: number of vacancies). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

238 In order to better understand the effect of He on a bubble's surface orien-
 239 tation, further investigation is necessary. The sizes of bubbles in this study
 240 are at the resolution limit of transmission electron microscope (TEM), and it is
 241 challenging to identify bubbles below 1nm. Furthermore, at such small sizes,
 242 specimens might not be suitable for further analysis to determine helium density
 243 in the bubble with electron energy loss spectroscopy (EELS) technique [51], and
 244 in experiments it is assumed that pressure induced by helium cause spherical

245 shapes of cavities [52]. To verify this, it would be necessary to introduce more
 246 helium in bubbles exceeding a He/vac ratio of 2.5, but it was found that these
 247 configurations of helium bubbles in nickel using the current set of potentials are
 248 not stable at finite temperatures.

249 *4.4. Binding energy*

250 Until now, particular free energy contributions to the model have been fitted
 251 with physical values calculated from MD. To minimize the difference between
 252 binding energies predicted by the model and the values from MD, parameters β
 253 and r_s in the equation for bubble radius were introduced, adding some degrees
 254 of freedom:

$$r_V = r_{\text{He}} + r_{\text{Ni-He}} - \beta\sigma(r_{\text{Ni-He}}) - r_s. \quad (21)$$

255 The best fit of β and r_s with emphasis on bubbles containing a higher number
 256 of vacancies and a lower number of He atoms was found for values $r_s = 0$ and
 257 $\beta = 1.05$.

258 The final comparison of helium and vacancy binding energies is shown in
 259 Figure 7 (color scale indicates the number of vacancies in the bubble). The
 260 model predicts binding energies accurately, except for very small sizes (number
 261 of vacancies ≤ 15), where the model assumptions are weakly met. For small
 262 clusters, shapes are strongly faceted and energetics is non-monotonic due to
 263 magic number sizes with high symmetry. Particularly more pronounced effect of
 264 facetting is also for bubble containing 80 vacancies, and for various bubble sizes
 265 above 2 He/vac ratio, which cause higher discrepancy between binding energy
 266 values from the model and from MD. However, when the model is implemented
 267 in larger scale CD simulation codes, MD values for small clusters can be used
 268 directly.

269 **5. Equilibrium helium density**

270 In metallic materials, helium density in bubbles can be measured using differ-
 271 ent experimental techniques. Each of these techniques that are used for charac-

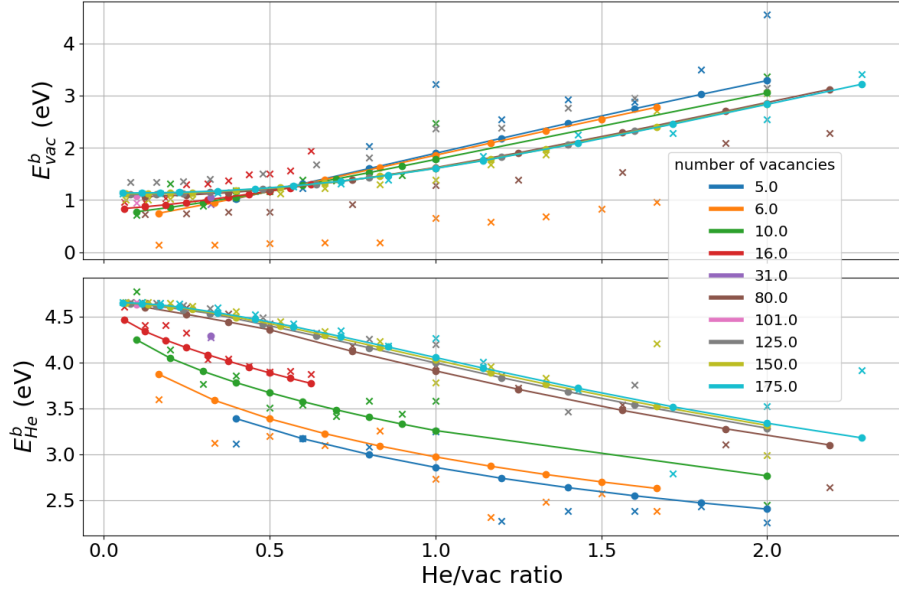


Figure 7: Comparison of vacancy (top) and helium (bottom) binding energies extracted from MD (symbols) with the model predictions (lines) as a function of helium to vacancy ratio for various bubble sizes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

272 terization of nanometric sized defects include some non-negligible uncertainty,
 273 especially for smaller radii and higher He densities.

274 Ameranda et al. [53] used positron-annihilation spectroscopy (PAS) to mea-
 275 sure helium density in pure nickel with homogeneously implanted He as a func-
 276 tion of the temperature during isochronal annealing.

277 Qiang-Li et al. [54] used a contrast variation method of small angle neutron
 278 scattering (SANS) combined with TEM to extract information on the bubble
 279 structure in Ni, implanted with 1200 appm helium at room temperature, and
 280 annealed at various temperatures between 820K and 1170K. Torres et al. [30]
 281 used electron energy loss spectroscopy (EELS) combined with scanning TEM to
 282 characterize Ni-alloy Inconel X-750, irradiated up to 80 dpa in a high thermal
 283 flux at 300-330°C and 25000 appm helium [9, 55]. Walsh et al. [56] measured He
 284 bubbles in Ni-based PE16 alloy irradiated during operation in a nuclear reactor

285 at a temperature of about 550°C with EELS and TEM.

286 In some studies [57, 58, 59], measured values are compared to the “ $p =$
 287 $2\gamma/r$ ” law, where γ is the surface energy, p is pressure in the bubble, and r
 288 its radius, which is valid for bubbles in thermal equilibrium. In refs [54, 53],
 289 measured values were found almost 3GPa larger than values from the “ $p = 2\gamma/r$ ”
 290 law, stating that the vacancy supply is too small to relax the bubbles to the
 291 equilibrium. The fact that He was (pre)implanted with α -particles certainly
 292 plays a role. The equilibrium helium density for each void size can be calculated
 293 as a He/vac ratio where the vacancy binding energy is approaching vacancy
 294 formation energy (the chemical potential of a vacancy inside a bubble is $\mu_V =$
 295 $F^f(m, n) - F^f(m-1, n) = F^f(1, 0) - F_V^b(m, n)$). The variable-gap model predicts
 296 lower He density for small bubbles than the “ $p = 2\gamma/r$ ” law, converging to the
 297 same values with increasing radius r_V^0 (Fig 8). Small bubbles, which appeared
 298 to be nearly at equilibrium in refs [54, 53], when compared to the “ $p = 2\gamma/r$ ”
 299 law, are in fact in overpressurized as other bubbles if the variable-gap model is
 300 considered.

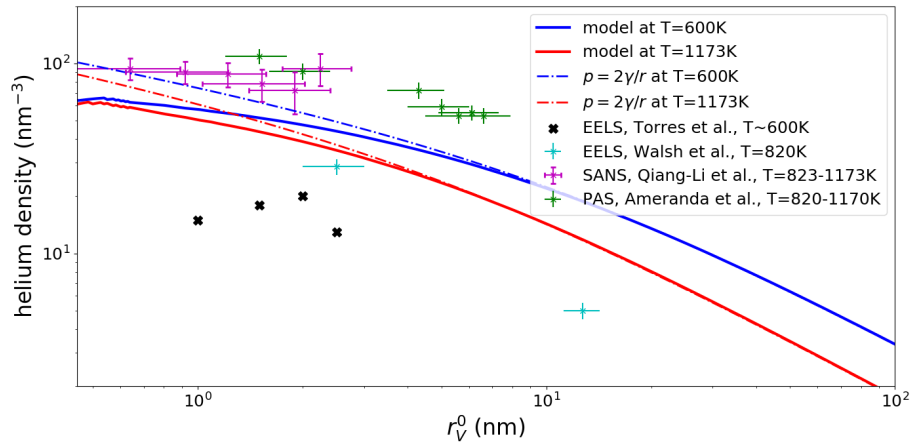


Figure 8: Helium density in bubbles at thermal equilibrium as a function of bubbles’ unrelaxed radius r_V^0 . Solid lines: the variable-gap model in Ni, dash-dot lines: “ $p = 2\gamma/r$ ” law with Trinkaus’ EOS [29] at 600K (blue) and 1173K (red). Symbols: experimental measurements in Ni and Ni-based alloys [30, 53, 54, 56].

301 On the contrary, with other experimental results, EELS measurements in [30,
302 56] indicated that bubbles were rather underpressurized. This can be explained
303 by the low He/dpa, or temperature being too low to induce sufficient vacancy
304 emission from bubbles to reach thermal equilibrium. The presence of hydrogen
305 that plays a similar role as He [60, 61, 62, 63] could also contribute to the
306 observed differences.

307 **6. Conclusion**

308 In the current work, parametrization of a variable-gap model for helium
309 bubbles in nickel was presented, highlighting the difference between its version
310 for iron. The model was then compared with experimental measurements of
311 helium densities.

312 It was shown that in a FCC case, surface effect plays an important role in
313 the Ni-Ni interaction. The observed faceting seems to be in agreement with
314 experimental observations [1, 47, 48]. The majority of surfaces were identified
315 as $\{111\}$ and $\{100\}$ planes, with surfaces of small voids having mostly $\{111\}$
316 planes, but the surface started to be more complex, exhibiting an octahedron-
317 like shape, with increasing size.

318 The model predicts helium and vacancy binding energies in a good agree-
319 ment with MD values, except for a few cases of small sizes, where size (in terms
320 of number of vacancies) is equal to their magic numbers. For these sizes, binding
321 energies exhibit non-monotonic behavior as a function of the number of vacan-
322 cies that can be hardly captured by the model, as the shape, complexity, and
323 differences in symmetry between neighbouring sizes are approximated in a sim-
324 ple way. This limitation can be eliminated by using binding energy values from
325 MD directly, as the present model is to be used in kinetic models such as CD
326 to provide insight on nucleation and growth of bubbles in Ni and FCC metals
327 under various He/dpa ratios. As a first approach, the model can be adopted
328 to more complex alloys (e.g. 300 series ASS) by simply modifying elastic con-
329 stants. It is expected that He-He and He-metal parts of the model would remain

330 globally unchanged.

331 **CRedit authorship contribution statement**

332 **Miroslav Fokt:** Writing - original draft, Investigation, Formal analysis.

333 **Gilles Adjanor:** Writing - review editing, Supervision, Conceptualization.

334 **Thomas Jourdan:** Writing - review editing, Supervision, Conceptualization,
335 Methodology.

336 **Declaration of Competing Interest**

337 The authors declare that they have no known competing financial interests or
338 personal relationships that could have appeared to influence the work reported
339 in this paper.

340 **Data availability**

341 The raw/processed data required to reproduce these findings cannot be
342 shared at this time due to technical or time limitations.

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348 **Appendix A. Shape analysis of bubbles' surface**

349 The aim was to develop a method to consistently analyze shape and the
350 surface of a bubble at the atomic scale (FCC nickel in our case). This can be
351 a challenging problem, because a bubble - from a geometrical point of view - is
352 a hole in the bulk. So the use of concave or convex hull algorithms could omit

353 some of the surface atoms, and thus underestimate or overestimate the overall
354 volume of the bubble, respectively.

355 The method is based on the Wigner-Seitz defect analysis followed by the
356 Delaunay tessellation. The first step is to identify the vacancy positions by
357 comparing two atomic systems: a **reference state** (a perfect bulk nickel FCC
358 lattice), and a **deformed state** (a bubble, where some atoms are missing and
359 others are displaced due to relaxation). For each atom in a deformed state, the
360 closest site in a reference state is found. The number of atoms (from deformed
361 state) assigned to each atom (in reference state) is called occupancy, and can
362 be equal to zero (vacancy), one (normal atom), or two and more (interstitial
363 atom). In other words, one could say that occupancy is the number of atoms in
364 a deformed state that lie within the reference state atom’s Voronoi cell.

365 Then, in a deformed state, all helium atoms (if any) are replaced by artificial
366 ‘vacancy’ atoms (atoms placed at vacancy positions), and the new configuration
367 is tessellated using Delaunay triangulation⁴, creating simplices that have vertices
368 at atoms’ positions. Surface atoms are identified as bulk vertices of simplices
369 with one vertex as a vacancy atom. Three atoms at the bubble’s surface form
370 triangle - one face of a simplex.

371 This method was tested as the most reliable in terms of detecting all surface
372 atoms with their coordinates, and surface triangles for further analysis (shape,
373 orientation of normals). Another advantage is the detection of *true* bubble
374 volume V_{rel} as a sum of volumes of all simplices with at least one vertex as a
375 vacancy atom. A bubble radius is

$$r = \sqrt[3]{\frac{3V_{\text{rel}}}{4\pi}}. \quad (\text{A.1})$$

376 From our perspective, this is a rigorous way to calculate a bubble’s radius at
377 the atomistic scale, as we want to be able to detect changes in a void’s ra-
378 dius/shape when a point-defect is added. The convex hull of all surface atoms

⁴Python’s `scipy.spatial.Delaunay` library based on Qhull library

379 is overestimating the final volume, and it is not sensitive enough to small size
 380 changes.

381 **Appendix B. Eshelby's inclusion for a void in anisotropic material**

382 In this section, we generalize the formula giving the relaxation energy to
 383 anisotropic material. Using the equivalent inclusion approach, the effect of
 384 helium pressure (or surface tension) is modeled by an equivalent homogeneous
 385 inclusion of eigenstrain ϵ_{ij}^* . By equating the stress within the inclusion to the
 386 stress produced by helium (or surface tension), we find that

$$\epsilon_{ij}^* = \frac{-p\delta_{ij}}{(C_{11} + 2C_{12})(S_{11} + 2S_{12} - 1)} = \epsilon^* \delta_{ij}. \quad (\text{B.1})$$

387 In this equation, δ_{ij} is the Kronecker delta, p is pressure, $C_{11} = C_{1111}$ and
 388 $C_{12} = C_{1122}$ are the elastic constants and $S_{11} = S_{1111}$ and $S_{12} = S_{1122}$ are
 389 terms of the Eshelby tensor [64]. It can be shown that the relaxation volume in
 390 an infinite medium ΔV_∞ is

$$\frac{\Delta V_\infty}{V} = 3(S_{11} + 2S_{12})\epsilon^*, \quad (\text{B.2})$$

where V is the actual volume. So the eigenstrain reads

$$\epsilon_{ij}^* = \frac{\Delta V_\infty}{V} \frac{1}{3(S_{11} + 2S_{12})} \delta_{ij} = \frac{\Delta R_\infty}{R} \frac{1}{S_{11} + 2S_{12}} \delta_{ij}, \quad (\text{B.3})$$

391 with ΔR_∞ being the relaxation radius and R the actual radius in an infinite
 392 medium. The energy stored in the matrix is

$$E^M = -\frac{1}{2} \sigma_{ij}^I \epsilon_{ij}^C V, \quad (\text{B.4})$$

where $\sigma_{ij}^I = -p\delta_{ij}$ is the stress in the inclusion and ϵ_{ij}^C is the constrained deformation in the inclusion, which is equal to

$$\epsilon_{ij}^C = S_{ijkl} \epsilon_{kl}^*. \quad (\text{B.5})$$

We can deduce that

$$E^M = 2\pi \frac{1 - S_{11} - 2S_{12}}{S_{11} + 2S_{12}} (C_{11} + 2C_{12}) R (\Delta R_\infty)^2. \quad (\text{B.6})$$

The displacement of the surface can be also calculated through

$$u_i = u_i^C = S_{ijkl}\epsilon_{kl}^* x_j, \quad (\text{B.7})$$

which reads as

$$u_i = (S_{11} + 2S_{12})\epsilon^* x_i. \quad (\text{B.8})$$

393 We can see that the displacement is isotropic, even if the material is not elasti-
394 cally isotropic.

For an elastically isotropic material, we have

$$C_{11} + 2C_{12} = 2\mu \frac{1 + \nu}{1 - 2\nu}, \quad (\text{B.9})$$

$$1 - S_{11} - 2S_{12} = \frac{2}{3} \frac{1 - 2\nu}{1 - \nu}, \quad (\text{B.10})$$

$$S_{11} + 2S_{12} = \frac{1}{3} \frac{1 + \nu}{1 - \nu}, \quad (\text{B.11})$$

and so we obtain

$$E^M = 8\pi\mu R(\Delta R^\infty)^2. \quad (\text{B.12})$$

395 Comparing both isotropic and anisotropic cases, the equivalent shear modulus
396 is equal to

$$\mu = \frac{1}{4} \frac{(1 - S_{11} - 2S_{12})}{S_{11} + 2S_{12}} (C_{11} + 2C_{12}). \quad (\text{B.13})$$

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