Energy spectra of primary knock-on atoms under neutron irradiation

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Abstract

Materials subjected to neutron irradiation will suffer from a build-up of damage caused by the displacement cascades initiated by nuclear reactions. Previously, the main “measure” of this damage accumulation has been through the displacements per atom (dpa) index, which has known limitations. This paper describes a rigorous methodology to calculate the primary atomic recoil events (often called the primary knock-on atoms or PKAs) that lead to cascade damage events as a function of energy and recoiling species. A new processing code SPECTRA-PKA combines a neutron irradiation spectrum with nuclear recoil data obtained from the latest nuclear data libraries to produce PKA spectra for any material composition. Via examples of fusion relevant materials, it is shown that these PKA spectra can be complex, involving many different recoiling species, potentially differing in both proton and neutron number from the original target nuclei, including high energy recoils of light emitted particles such as α -particles and protons. The variations in PKA spectra as a function of time, neutron field, and material are explored. The application of PKA spectra to the quantification of radiation damage is exemplified using two approaches: the binary collision approximation and stochastic cluster dynamics, and the results from these different models are discussed and compared.

Keywords: radiation damage quantification, primary knock-on atoms (PKAs), nuclear data processing, neutron irradiation, recoil energy spectrum

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27 1. Introduction

28 Understanding through modelling of the damage accumulated in materials irradi-
29 ated by neutrons remains a primary goal for computational simulation of materials
30 for advanced nuclear energy systems. Several different approaches exist for predict-
31 ing the formation, evolution and behaviour of this damage, including: computationally
32 demanding molecular dynamics simulations of damage cascades with full atomic in-
33 teractions; rate theory models, where defects are described as objects with defined
34 behaviour; and kinetic Monte-Carlo (kMC) simulations. However, without exception,
35 all of these techniques need, as input at some level, information about the initial pri-
36 mary disruptions, in the form of the type, energy and spatial distribution of the primary
37 knock-on atoms (PKAs). In particular, this kind of data goes far beyond the limited
38 information provided by the traditional and ubiquitous defect index of initial damage
39 formation known as “displacements per atom” (dpa). The so-called Norgett, Robin-
40 son and Torrens NRT-dpa [1] reduces the predicted irradiation environment, perhaps
41 obtained from a Monte-Carlo neutron transport simulation on a reactor geometry, to a
42 single number that converts the energy deposited into a material by the irradiation into
43 an estimate of the number of atomic displacements that could be generated. The dpa
44 measure has proven to correlate well with various radiation damage phenomena, but
45 is not well suited to distinguishing between different types of irradiation, and, more
46 importantly, it does not provide information about damage evolution. It is also the case
47 that it should not be used as a basis for comparing irradiation behaviour in different
48 materials [2]. A more complete picture of radiation damage evolution, that may be
49 afforded by modern computational techniques, requires the spatial and energy distri-
50 bution of all the initial displacement events, including both emitted and residual nuclei
51 from nuclear interactions.

52 In modern nuclear data evaluations, for a single target species such as the major
53 ^{56}Fe isotope in Fe, there are many nuclear reaction channels that produce recoiling
54 species, including elastic, inelastic, and nonelastic nuclear reactions. In nuclear fu-
55 sion in particular, the generally higher energy of the incident neutrons, when compared
56 to fission, leads to many more channels becoming relevant, which in turn produces a

57 more complex distribution of PKAs in both energy and type. These PKAs lead to cas-
58 cades of atomic displacements, which can subsequently evolve and collapse to produce
59 extended defects such as dislocation loops and voids.

60 In this paper a modern computational methodology is described to produce, for a
61 given target species or distribution of targets, the instantaneous picture of PKA rates
62 as a function of energy (“PKA spectra”). This includes a new processing code, called
63 SPECTRA-PKA, that takes as input nuclear recoil cross-section matrices and combines
64 (collapses) these with an incident neutron spectrum to produce the PKA distributions.
65 PKA spectra for all the primary (heavy) nuclear reaction products, also known as the
66 residuals, which may be a different elemental species than the target nuclide, and any
67 secondary (light) emitted products are considered. Results for selected fusion-relevant
68 materials under various neutron irradiation fields are presented. In the final section
69 outputs from the above are used as inputs to two applications for radiation damage
70 characterisation and quantification: the binary collision approximation (BCA) calcula-
71 tions and stochastic cluster dynamics (SCD) simulations.

72 2. Methodology and Results

73 The nuclear data processing code NJOY [3], and, in particular, the GROUPE module
74 within it, can calculate group-to-group recoil cross-section matrices due to many
75 types of nuclear reactions. Using neutron-interaction data for a given target nuclide
76 x , such as ^{56}Fe , NJOY-12 [4] has been used in the present work to provide matrices
77 $M^{x \rightarrow y}$ for every $x \rightarrow y$ reaction channel. Group-to-group cross-section matrices for
78 neutron scattering, light charged particles, as well as recoils of the residual nuclei can
79 be generated from modern nuclear data libraries, following the ENDF [5] data format.
80 The evaluations include both energy transfer and angular recoil distributions for all en-
81 ergetically possible nuclear reactions on a wide range of target nuclides, which NJOY
82 reads and processes into a pre-defined group structure.

83 Two-body elastic and discrete inelastic neutron scattering, charge-particles elastic
84 scattering, continuum scattering and fission can be treated in different ways depending
85 on the completeness and accuracy of the content of the original evaluation. Previously,
86 simple theoretical models had to be employed to plug the gaps in earlier nuclear data

87 libraries, but this is becoming less necessary with the latest libraries [6], which use
88 detailed and well-validated theoretical models to derive data where no experimental
89 information exists. The resulting $m_{ij}^{x \rightarrow y}$ elements of each $M^{x \rightarrow y}$ matrix are the cross-
90 sections in barns for a recoil in energy group i produced by an incident neutron in
91 energy group j .

92 A fine 709-group [7] spectrum has been used here for both the incident and re-
93 coil energy distributions, and all the results were calculated using input data from the
94 TENDL-2014 [6] nuclear library, which was selected in part because it contains the
95 extended database of nuclides (compared to other libraries) required for complex ma-
96 terial compositions (see Section 2.1), whilst providing the complete and often intricate
97 energy-angle distributions necessary for this type of simulation.

98 Note that for the present work the detailed angular distributions of the recoiling
99 nuclei and scattered particles, which are also calculated by NJOY, are not explicitly re-
100 tained, although the angular dependence is implicitly considered in as far as it impacts
101 on the recoil energy distribution. It is assumed that any neutron-incident spectrum
102 used in conjunction with the recoil cross-section matrices will be average fluxes over
103 relatively large (on the atomic scale) volumes. In this case both the lack of direc-
104 tional information in the neutron field and the absence of structural information about
105 the irradiated materials, including the distribution of grain orientations, means that in-
106 cluding direction information in a PKA spectrum is not relevant. An isotropic recoil
107 distribution is considered a valid approximation for this level of simulation. If, in the
108 future, it is possible to accurately define the orientation of the atomic lattice relative
109 to the neutron-irradiation source, perhaps in a well-qualified fusion reactor first wall
110 (FW), then it may be necessary to revisit this approximation and consider the angular
111 dependence of recoils.

112 Furthermore, the uncertainties associated with the basic nuclear library evaluations
113 are not propagated into the recoil cross-section matrices derived by NJOY. Besides this,
114 there are no error estimates available for the neutron spectra that will be combined with
115 these matrices (see below) to produce PKA distributions, and so uncertainties are not
116 included in any of the results presented in this paper. In the future, it may be possible to
117 consider uncertainties using statistical methods such as the Total Monte-Carlo (TMC)

118 approach [8, 9], which can be applied to both input ingredients of a PKA spectrum –
119 the neutron spectrum and the recoil cross-section matrix.

120 Fig. 1 is a 3D plot of the elastic scattering (n, n) recoil cross-section matrix for
121 ^{56}Fe , while Fig. 2 shows 2D snapshots at several incident energy groups for the main
122 reaction channels of the same nuclide, including elastic scattering. For each snapshot
123 in Fig. 2 at incident energy group j (indicated by a vertical line in the group mid-point
124 in each graph and by the straight lines on the xy-plane of Fig. 1) the curves effectively
125 represent the j th column vectors of the $M^{x \rightarrow y}$ matrices. The figure shows that there
126 is significant variation as a function of neutron energy for this ^{56}Fe case, in terms
127 of both the reaction channels represented and the cross-sections versus recoil-energy
128 distributions of the different channels. At low neutron energies around 102 keV, only a
129 single reaction channel is open – that of the simple elastic scattering, where the same
130 neutron is emitted as was incident (hence the (n, n) representation in the figure). As the
131 neutron energy increases, other channels become important, from inelastic reactions
132 (here only the inelastic channel to the ground state (n, n'_1) is shown) and then more
133 complex non-elastic reactions, where a variety of different particles are emitted.

134 Fig. 2 also demonstrates that NJOY produces the cross-sections for the secondary
135 light particle emissions associated with some of the reaction channels. For example,
136 in addition to the recoils of ^{53}Cr from the (n, α) reaction, there are also cross-section
137 curves for the recoil of the ^4He α -particles from the same reaction. Note that in Fig. 2
138 there are no curves associated with the main (n, γ) neutron-capture channel because
139 the version of NJOY used (2012-032), does not directly output the matrices for the
140 recoils of this channel. The method by which the recoils from this important channel
141 are calculated in the present work is discussed later.

142 To calculate PKA spectra, these recoil cross-section matrices must now be folded
143 with a neutron flux versus energy spectrum for the irradiation environment of interest.
144 The code SPECTER and its sister code SPECOMP have previously been developed at
145 Argonne National Laboratory by Greenwood and Smitther [10] to perform this fold-
146 ing or collapsing for both pure elements and compounds, respectively. However, both
147 codes are relatively old, having been written in the 1980s, and are limited in their
148 ability to handle modern nuclear data and complex material compositions. For this

Figure 1: The recoil cross-section matrix for elastic scattering of neutrons on ^{56}Fe . The matrix is plotted as a separate recoil-energy vs. cross-section distribution for each incident energy group (plotted at the midpoint of the group). Note that only a subset of the possible 709 incident-energy-group distributions are shown. The coloured lines on the base xy-plane indicate the four incident energies considered in Fig. 2 to display snapshots of the recoil cross-sections for multiple reaction channels. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

reason a new code has been developed, hereafter called SPECTRA-PKA, which can take group-wise recoil matrices generated directly from NJOY (with a slight modification to the GROUPT routine therein to separate the recoil matrices from other NJOY data) and collapse them with a user-defined neutron-irradiation spectrum. In particular, the new code can take both recoil matrices and neutron spectrum in any user-defined energy group structure, a feature not available in SPECTER/SPECOMP, allowing the use of the fine 709-group structure mentioned previously. The new code also performs interpolations and averaging, as necessary, between different spectrum energy-group formats – for example if the neutron irradiation spectrum has been computed with a different format to that used for the recoil cross-section matrices – although we recommend using the same high-resolution 709-group (see [7] for details) structure to calculate irradiation spectra, particularly when using statistical Monte-Carlo methods.

A further advance over the previous code is the ability, in SPECTRA-PKA, to consider multiple reaction channels on the same target. Typically, NJOY will process the

163 entire data file for a given target nuclide and output recoil matrices for every reaction
 164 channel in the same file. Our new code will read this file and process each channel one-
 165 by-one, outputting the PKA matrix that results from each folding. For a given $x \rightarrow y$
 166 reaction channel, the PKA spectrum $R^{x \rightarrow y}$ is computed via:

$$R^{x \rightarrow y}(E) \equiv \{r_i^{x \rightarrow y}\} = \left\{ \sum_j m_{ij}^{x \rightarrow y} \phi_j \right\}, \quad (1)$$

167 where $r_i^{x \rightarrow y}$ is the PKA rate in recoil energy-group i , computed by folding the i th row
 168 of $M^{x \rightarrow y}$ in cross-section units of barns ($1 \times 10^{-24} \text{ cm}^2$) with the ϕ_j flux values of the
 169 incident neutron spectrum in units of neutrons $\text{cm}^{-2} \text{ s}^{-1}$.

170 Fig. 3, shows the PKA spectra computed for the main reaction channels on ^{56}Fe
 171 under a neutron-irradiation flux-spectrum predicted for the equatorial first wall (FW)
 172 of a typical conceptual design for a demonstration fusion power plant “DEMO”. This
 173 spectrum, shown in Fig. 4, was computed using the MCNP [11, 12] Monte-Carlo
 174 neutron transport code for a 2013 DEMO design with helium-cooling and a tritium-
 175 breeding blanket made up of a bed of Li+Be pebbles, abbreviated as hcpb in the figure
 176 for helium-cooled pebble-bed – see [13] for further details of the model and calcula-
 177 tions. Fig. 3a shows the PKA rates in units of PKAs per second per target ^{56}Fe atom
 178 for the main heavy nuclide recoil channels, while Fig. 3b shows the equivalent light
 179 particle (gas) emission PKA spectra, where they exist.

Figure 2: Incident energy-group snapshots of the group-to-group recoil matrices generated using NJOY for reactions on ^{56}Fe . The four plots show the cross-sections as a function of recoil energy for different incident neutron energies groups, whose midpoint energies are indicated by the solid vertical line (blue) and the label in the top right-hand corner of each plot. The colouring of the text labels is equivalent to that used to indicate these same incident energies on Fig. 1. Note that here, and in the other figures in this paper, y-values are plotted against the midpoints of the energy group to which they correspond. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

Figure 3: The collapsed PKA spectra for the dominant reaction channels in ^{56}Fe under DEMO hcpb FW conditions. (a) the (heavy) residual recoils, and (b) the secondary light particle emissions. Note the change in energy scale in (b) reflecting the fact that the light particles are emitted with a higher proportion of the recoil energy than the associated heavy residuals produced via the same reaction channels. **Note that the PKA s⁻¹ fluxes given in these plots and elsewhere in this paper are the absolute total fluxes in each recoil energy group, plotted at the midpoint of the energy group.**

Fig. 3a shows that for much of the PKA energy range (on a logarithmic scale), the simple elastic scattering of ^{56}Fe atoms dominates, which is the typical result for most nuclides, with PKA rates of the order of 2×10^{-11} PKAs s⁻¹ per target atom at all energies below a few tenths of a keV. Only at energies above around 100 keV do other reaction channels produce statistically significant numbers of PKAs. Most notably the inelastic scattering channel dominates the elastic scattering above ~ 100 keV, while ^{53}Cr PKAs from the (n, α) reaction are the only significant heavy recoils above 1 MeV. Note in this latter case that the recoil nuclei is of a different chemical nature to the target nuclei because transmutation has occurred.

For the light particle emissions (Fig. 3b) note that these are predominantly at energies above 1 MeV, reflecting the fact that, via conservation of momentum, they receive a higher proportion of the energy from the 14 MeV neutrons that dominate the ir-

Figure 4: Neutron irradiation spectra for the equatorial (outboard) FW of several demonstration fusion reactor (DEMO) concepts. The abbreviations refer to helium-cooled lithium-lead (hcll), helium-cooled pebble-bed (hcpb), water-cooled lithium-lead (wcll), and water-cooled ceramic-breeder (wccb) reactors. Further details are given in the main text and also in [13]. Also shown, for comparison, is the fuel-assembly averaged spectra for a typical fission reactor – in this case for a pressurised water reactor or PWR – and also the spectrum predicted for the HFIR experimental fission reactor [14]. The neutron flux is plotted per lethargy interval, which is equal to the natural logarithm of the upper divided by lower energy bounds of each of energy group used to bin the spectrum.

radiation spectrum (see Fig. 4) than the heavy recoils generated in the same nuclear reactions. The PKA rates for these light particles are also relatively low compared to the rates observed for the heavy-particle scattering, but they are in agreement with the rates for the corresponding heavy recoils for the same reaction channels.

Fig. 3a also includes the PKA spectrum associated with the recoil of ^{57}Fe , which is produced by neutron-capture (n, γ) followed by photon (γ) emission. As mentioned earlier, the recoil matrix for this channel is not an automatic output from GROUPR. However, GROUPR produces the (n, γ) reaction cross-section vector from the same nuclear data and in the correct group format. There is no energetic cost associated with this type of reaction, and furthermore if we assume that no energy is lost through excitation of the residual, then a simple momentum conservation argument gives the energy range of the target atom once it has captured a neutron from a specific energy group. The resulting compound nucleus will still be in an excited state due to extra mass and must decay down the energy levels by emitting photons (γ -rays), whose total energy can be found by comparing the mass of the final ground state of the daughter atom, in the Fig. 3 case ^{57}Fe , with the combined mass of target (^{56}Fe) and neutron (the “compound nucleus”). Then, as a conservative (in the sense that the highest possible recoil energy is attained) approximation, it is assumed that all of this energy, via Newton’s Third law, results in additional momentum. An alternative to this maximum energy transfer approximation (see [15, Chap. 1]), is to take the average and use half of this energy (also suggested in [15]). Continuing with the conservative approach, for $^{56}\text{Fe}(n, \gamma)^{57}\text{Fe}$ this results in approximately 551 eV extra for every recoil energy, which, once collapsed with a neutron spectrum, results in a PKA spectrum that has no recoils below this.

In the analysis and calculations presented above and hereafter we do not consider any additional recoils that might be produced through radioactive decay. Some of the residual nuclides, even in their ground-state, will be unstable (e.g. the ^{55}Fe produced via $(n, 2n)$ reactions on ^{56}Fe) and will decay via the emission of further energetic species, such as β particles. This will, of course, via momentum conservation, cause the residual decayed nuclei to recoil. Furthermore, depending on the half-life, the decay may happen on the same timescale of the primary recoil from the nuclear reaction

and would thus increase the overall recoil energy. Alternatively, the decay may happen long after the radioactive species has come to rest and thus would initiate a new, independent recoil. While it is expected that the recoil energies associated with these decays will generally be lower in energy than that created by the nuclear interaction and significantly less frequent, they should still be included for completeness and we are investigating how this can be done most effectively. Note, that in the earlier code, SPECTER [10], the (n, γ) reaction channel was treated with subsequent β decay of the residual included in the kinematics, and this is an important channel in the nuclear fission industry, but for more general purposes, including fusion, we would need to consider all decay channels.

2.1. Calculations for real materials

For a real material, even a single element with multiple isotopes, the results for a single target nuclide are not sufficient. Thus, SPECTRA-PKA has been written to process multiple sets of recoil cross-section matrices and perform various summing to produce PKA spectra in the most appropriate format for damage modelling. For each target species, each with its own separate input file of recoil cross-section matrices, the code folds the matrix for every channel with the neutron flux-spectrum, but then weights the resulting spectrum of PKA rates by a user-defined atomic fraction of the current target in the material composition. Subsequently, when the PKA rates for all targets have been processed in this way, the spectra are summed according to whether they are for the same PKA daughter. So, for example, in an irradiation of pure W, where ^{182}W , ^{183}W and ^{184}W all form part of the natural composition, the total PKA spectrum for ^{183}W , would include contributions from scattering (both elastic and inelastic) on ^{183}W itself, but also from the neutron-capture (n, γ) reaction on ^{182}W , and the $(n, 2n)$ reaction channel on ^{184}W . Numerically, the total PKA spectrum for a given recoil species y is:

$$R^y(E) = \sum_{i,j} w_i R^{x_i \xrightarrow{c_j} y}(E), \quad (2)$$

where w_i is the atomic fraction of target x_i in the material of interest, and the contribution to the y PKAs from x_i is the sum over all possible reaction channels c_j that

251 produce y from x_i .

252 For Fe, four isotopes make up its natural composition, with ^{56}Fe the most common
253 at 91.75 atomic % abundance then ^{54}Fe , ^{57}Fe , and ^{58}Fe at 5.85, 2.12, and 0.28 atomic
254 %, respectively. The resulting summed nuclide PKA spectra for the hcpb DEMO irra-
255 diation scenario are shown in Fig. 5, where the isotopes of Fe, Cr, and Mn are shown in
256 three separate plots to allow easy identification, together with a fourth plot showing the
257 summed PKA spectra for α particles and protons. Note that while the output from the
258 folding of recoil cross-section matrices with neutron flux spectra produces PKA rates
259 with units of PKAs s^{-1} per target atom, in this figure and subsequent ones in this paper
260 the PKA rates are given for unit volumes of target material. This avoids the ambiguity
261 associated with “per target” when there is a complex material composition, and is more
262 useful for modelling where a defined volume of material is considered and hence the
263 number of atoms is known, for example in a molecular dynamics simulation of a colli-
264 sion cascade. Note that this cm^{-3} measure is not related to the cm^{-2} employed when
265 defining the neutron flux field that every target atom sees, but is a more convenient unit
266 for the present results.

267 As expected, the highest PKA rates are calculated for the four naturally-occurring
268 isotopes of Fe (Fig. 5a), with ^{56}Fe dominating overall due to its high abundance in
269 pure Fe combined with the large scattering cross-sections. PKAs associated with heavy
270 Mn or Cr recoils (Figs. 5c and 5b, respectively) are less frequent and generally only
271 at higher energy due to the lower cross-section and threshold nature of the charged
272 particle reactions ((n, α) , (n, p) , etc), that produce them, although, for Cr in particular,
273 the maximum PKA energy is higher than that of Fe PKAs. Finally, the recoils of light
274 particles (Fig. 5d) are at even higher energies, in agreement with the earlier results for
275 ^{56}Fe , and with similar lower production rates as for Mn and Cr.

276 Two further merges of the results, which are also built into the SPECTRA-PKA
277 processing code, are also relevant to materials modelling. Noting that simulating dif-
278 ferent isotopes of the same element is not normally considered in atomic simulations,
279 it is more useful to consider the summed PKAs for each daughter element produced
280 in the nuclear reaction, whether these are different from the original target elements
281 or not. Furthermore, taking a more pessimistic view of simulations, unless the mate-

Figure 5: The set of PKA-rate spectra for pure Fe irradiated in the FW hcpb DEMO neutron-field (Fig. 4). (a) shows the PKA spectra for the isotopes (both naturally occurring and those produced by nuclear reactions) of Fe, while (b) and (c) show the PKA spectra for the isotopes of Cr and Mn, respectively, generated by neutron capture followed by proton (for Mn) or α (Cr) emission reactions on Fe. (d) shows the summed PKA spectra for the light particles themselves. The conversion from the original “per target isotope” units (as used in Fig. 3) to $\text{PKAs s}^{-1} \text{cm}^{-3}$ was performed using the density and mass of pure Fe given in Table 1.

282 rial under investigation is a genuine mixed alloy or compound, it is unlikely that the
 283 models used to describe atomic interactions will include parameters for every possible
 284 atom species generated under neutron irradiation, or even be able to consider the spon-
 285 taneous transformation of one atom type into another. In this sense we are primarily
 286 limited to considering PKAs that are identical to the host lattice and so the code also
 287 outputs the total PKA spectrum for all recoils, by summing over all daughter elements.
 288 It is expected that this would be most useful in situations where the target material is a
 289 single element or simple alloy.

290 Fig. 6a shows the summed elemental PKA spectra for pure Fe under the DEMO
 291 hcpb FW neutron spectrum, while Fig. 6b has results for pure W, which has five natural
 292 isotopes (180, 182, 183, 184, and 186 at 0.12, 26.56, 14.31, 30.64, and 28.43 atomic
 293 %, respectively), under the same conditions. Note that these PKA spectra, and those
 294 shown in previous figures, represent a snapshot at $t = 0$, i.e. before any neutron-
 295 induced transmutation has taken place. For Fe, where transmutation rates are relatively
 296 low under fusion neutron conditions, this is a reasonable representation of the picture
 297 throughout the lifetime of an irradiated sample. However, in a material such as W
 298 the transmutation rates are much higher and there could be a significant change in
 299 chemical composition during reactor and component lifetimes. In such circumstances,
 300 the change in chemical composition might lead, as a side-effect, to a discernable change
 301 in PKA spectra profiles, leading to a corresponding change in damage accumulation.
 302 This will be investigated in a later section, but note that a consequence of the $t = 0$
 303 assumption in these figures, and elsewhere where no transmutation is considered, there
 304 are no PKA contributions from elements with a higher atomic number Z than the parent
 305 target atoms – in this case Fe and W, respectively for Figs. 6a and b. This is a natural
 306 consequence of the fact that there is no direct reaction channel with a neutron as the
 307 projectile that leads to an increase in Z – some kind of decay process, such as β^-
 308 decay, is needed for that. As suggested earlier, the recoils of such decays may give a
 309 non-negligible contribution to the total PKA picture.

310 Fig. 6, shows, for W in particular, a very startling result for the recoil energies of
 311 the α particles, namely that the energies at which they can be emitted is significantly
 312 above the maximum neutron energy, in this case around 14 MeV. Indeed, the maximum

Figure 6: The set of summed elemental PKA spectra in pure (a) Fe and (b) W under the DEMO hc pb FW irradiation scenario (see Fig. 4). The conversion to PKAs $\text{s}^{-1} \text{cm}^{-3}$ was performed using the densities and masses of pure Fe and W given in Table 1.

energy of the emitted α particles under this fusion spectrum is above 20 MeV, with the distribution peaking at around 18 MeV. The reason for the very high emission energy is that the Q-values for the (n, α) reaction channels on *all* of the isotopes of W are positive and in the 5-10 MeV range. The extra energy produced by these exothermic reactions is available to the α particle when it is emitted. This is combined with the energy it already gains from the momentum of the absorbed neutron, which for a 14 MeV neutron on ^{184}W , for example, could be as much as 13.7 MeV by a simple mass distribution argument (with the ^{181}Hf residual taking the remainder). With a Q-value of 7.3 MeV for the $^{184}\text{W}(n, \alpha)^{181}\text{Hf}$ in the TENDL-2014 library, this then explains how the α particle can have emission energies of 20 MeV or more, especially given the fact the maximum Q-value for (n, α) on W is 9.1 MeV on ^{183}W .

This important observation about the energy of light particle emissions, which is not well known or appreciated in the literature, is also observed to a lesser extent in Fe. While the Q-value for the (n, α) on ^{56}Fe is only 325.9 keV, for the heavier ^{57}Fe it is 2.4 MeV, and for the lighter ^{54}Fe 0.8 MeV, explaining why there is a small chance of producing α particles with energies around 14 MeV.

Even though the probability of production for these high-energy α particles, and to

330 a lesser extent protons, is low compared to the dominant scattering channels (particu-
331 larly in W), the extreme nature of the energies they are produced at suggests that they
332 cannot be ignored when considering the accumulation of radiation damage.

333 Total PKA spectra for several materials under the same DEMO hcpb FW neutron
334 spectrum are shown in Fig. 7a, illustrating one of the benefits of such a simplification
335 – the ability to compare the distribution of initial damage events between different
336 materials (or indeed different irradiation scenarios – see later). Note that for the carbon
337 in SiC, the nuclear data file used in NJOY is that of natural C rather than a combination
338 of ^{12}C and ^{13}C . In TENDL-2014 [6] this is the only data for C and comes directly
339 from JENDL-4.0 [16], which is not as complete – with missing or erroneously merged
340 reaction channels and energy-angular distributions – compared to the data for other
341 nuclides.

342 For the total spectra in Fig. 7 the contribution from PKAs of light particles have
343 been omitted (via an option within the code). The damage produced by these high-
344 energy, low mass particles is completely different to similarly high-energy heavy par-
345 ticles and so it would be misleading to include them in total spectra for the materials
346 considered in the figure (even for SiC). Simulations using the binary collision approxi-
347 mation (BCA) performed with the SRIM (Stopping and Range of Ions in Matter) code
348 in full cascade mode (see [17] for more details of SRIM) for 14 MeV ^4He ions imping-
349 ing on Fe and W reveal that the average energy of the above 10 eV (primary) recoils
350 generated by the helium ions is only 76 and 118 eV, respectively. Thus the distribu-
351 tion of recoils from α particles, which are essentially the secondary knock-on atoms
352 (SKAs) relative to the original incident neutron, would produce primarily single point
353 defects or very small clusters, with very occasional extended defect regions. In com-
354 parison, the above 10 eV average energies for recoils generated by equivalent 14 MeV
355 self-ions is 960 eV and 8 keV (again calculated in SRIM), respectively, in Fe and W,
356 which would produce very much greater defect sizes and distributions. Thus it is in-
357 appropriate to have heavy ion recoils at the energies associated with the light particle
358 recoil energies – if the damage produced by such light particles is considered signif-
359 icant then the best option would be to evaluate the SKA spectrum, for example with
360 SRIM, and then add this to the total PKA spectrum from heavy particles.

Table 1: Total PKAs (above 1 eV) and average PKA energy (above 10 eV) for various materials caused by the FW conditions in the hcpb DEMO reactor (Fig. 4). The calculated molar masses (using the 2012 atomic mass evaluations [18] with the isotopic abundances for each material) and assumed material densities, used to convert to PKAs per unit volume, are also given.

Material	Total PKAs above 1 eV (PKAs s ⁻¹ cm ⁻³)	Average PKA energy above 10 eV (keV)	Molar Mass (amu)	Density (g cm ⁻³)
Fe	4.33E+14	18.8	55.85	7.9
W	1.11E+15	3.2	183.84	19.3
Ni	9.48E+14	10.4	58.69	8.9
Cu	5.77E+14	13.2	63.55	9.0
SiC	3.16E+14	76.1	20.05	3.2

Another useful representation for modelling is the cumulative distribution of PKAs, such as those shown in Fig. 7b, for the same total PKA spectra in Fig. 7a. Here PKA events with energies less than 1 eV are omitted, since these do not contribute to damage production. These cumulative distributions could be used to statistically sample PKA events, for example in a Monte-Carlo calculation, or in a stochastic cluster dynamics simulation (see Section 4.1). The sampling rate, i.e. the PKAs s⁻¹, per unit volume is obtained by integrating (above 1 eV here) the equivalent total PKA curve from Fig. 7a, and are given for these curves in Table 1. As would be predicted, the PKAs in a heavy material like W are, on average, lower in energy than in lighter materials such as Fe. Indeed, the average PKA energy can be evaluated directly, and the results for these total PKA distributions are also shown in Table 1. Since the characteristic threshold displacement energy, below which a PKA will not escape from its lattice site, is of the order of 10¹ eV, there is no point including PKA energies below this energy in the averaging. Thus all of the average PKA energies shown in Table 1 and elsewhere are for PKAs strictly above 10 eV in energy. From the table we see that W has a very low average of only 3.3 keV, with the average in Fe significantly higher at 19.0 keV.

Interestingly, these values of average PKAs are significantly lower than typical values presented in the literature, where, for example, the average PKA energy was thought to be about 150 keV for W under fusion neutron irradiation [19]. This large discrepancy comes from the assumptions made in other calculations. The present results have been calculated for a full FW fusion neutron spectrum (Fig. 4), with its significant proportion of moderated, lower energy neutrons in addition to the main

Figure 7: (a) Total PKA spectra and (b) cumulative PKA distributions for different elements under the DEMO hcpb FW conditions. Note that in (b), and in other cumulative curves presented later, the distributions begin from 1 eV. The conversion to PKAs $\text{s}^{-1} \text{cm}^{-3}$ was performed using the densities and masses given in Table 1, where the total PKAs and above 10 eV average PKA energies for each distribution are also presented.

14 MeV peak and, more importantly, the complete nuclear reaction cross-sections, including full angular dependencies, have been used to produce the recoil cross-section matrices with NJOY. The literature values, on the other hand, are obtained via an approximation of the neutron fields to a single delta-function at 14 MeV, and only the scattering channel with an isotropic angular distribution is considered. Fig. 2 demonstrates that this last approximation is not sufficient as there is significant deviation from isotropy (an isotropic response would have produced constant flat distributions for the distributions shown there). This observation has important implications for radiation damage modelling and will be discussed further in a future publication.

3. Variations in time and space

The PKA spectra for the complete set of naturally occurring elements (from hydrogen to bismuth) under DEMO hcpb FW conditions have recently been calculated for inclusion in a comprehensive nuclear physics handbook [20], which also describes the neutron-induced transmutation and activation of materials. Elemental PKA distributions, in particular, are presented in great detail, with an additional supplement [21]

398 giving the distributions, such as those in Fig. 6, in tabular form. However, with the
399 methodology developed here, it is a simple matter to consider the variation in PKA
400 spectra due to different irradiation scenarios, perhaps from different positions within a
401 nuclear reactor model.

402 Fig. 8 shows the total PKA spectra (a) and cumulative PKA distributions (b) for
403 pure Fe as a function of position into the equatorial wall of the hcpb DEMO reactor
404 vessel, while the total PKAs above 1 eV and PKA average energies (above 10 eV)
405 are given in Table 2. In order of depth the different regions considered are: the FW
406 itself (0-2.3 cm depth from plasma face), as shown in Fig. 4; the middle and rear of
407 the tritium breeding blanket (at around 14 and 77 cm depth respectively); the blanket
408 backplate (~80-110 cm); the vacuum vessel (VV) shield (115-160 cm); and the VV
409 itself (around 2 m depth). In agreement with the decrease in total neutron flux, as a
410 function of depth the total number of PKAs in pure Fe decreases dramatically, with
411 the above 1 eV PKAs falling by 4 orders of magnitude between the FW and VV, from
412 4.3×10^{14} PKAs $\text{s}^{-1} \text{cm}^{-3}$ to only 4.3×10^{10} PKAs $\text{s}^{-1} \text{cm}^{-3}$. However, from Fig. 8a
413 it is also evident that the proportion of PKAs at higher energies is decreasing faster with
414 depth than the overall PKA levels, which is confirmed by the drop in the above 10 eV
415 average PKA energy (Table 2). On the other hand, the cumulative PKA distributions
416 shown in Fig. 8b demonstrate that the picture is slightly more complex than this. While
417 it is the case that through the FW and blanket the PKA distributions shift to lower and
418 lower energies, beyond this there is not such a clear trend. Indeed, the blanket backplate
419 has a slightly higher proportion of higher-energy PKAs than the rear of the blanket
420 itself. There is a subtle interplay between moderation and overall neutron absorption
421 that produces this changing picture, with initially (near the plasma) high moderation
422 levels, but relatively little absorption, before, once a significant proportion of neutron
423 have been moderated, increased absorption of these moderated neutrons in particular.

424 We can also compare the PKA spectra between different reactors, as shown in
425 Fig. 9 for the equatorial FW of the four different DEMO concepts discussed in [13]:
426 the helium-cooled reactor with a tritium-breeding blanket of Li+Be pebbles already in-
427 troduced (hcpb); a helium-cooled reactor with a self-cooling liquid LiPb blanket (hell),
428 a water-cooled reactor with the liquid blanket (wcll), and a water-cooled Li+Be ce-

Figure 8: (a) Total PKA spectra and (b) cumulative PKA distributions for pure Fe as a function of position (depth) within the outboard equatorial wall of the hcpb DEMO concept. The conversion to PKAs per unit volume was performed using the density and molar mass of pure Fe given in Table 1. The total PKAs and above 10 eV average PKA energies for each distribution are given in Table 2.

429 ramic breeder concept (wccb). The neutron irradiation field for FW position within
 430 each concept is shown in Fig. 4. The results here indicate that although for the wccb
 431 concept the total PKA rate is the lowest (see Table 2), the average PKA energy above
 432 10 eV is actually significantly higher at 32.0 keV compared to the other three concepts
 433 that all have averages around 19-21 keV.

434 Finally, the present methodology can be combined with inventory calculations,
 435 to investigate how the PKA spectra for a material might change with time due to
 436 transmutation-induced changes in chemical composition. Using the inventory code
 437 FISPACT-II [7], together with the modern TENDL [6] nuclear data libraries we can
 438 compute the variation in composition in pure W under the fission spectrum evaluated
 439 for the High Flux Isotopes Reactor (HFIR) at Oakridge National Laboratory. The neu-
 440 tron spectrum, reproduced from [14], is shown in Fig. 4. This spectrum, as calculated
 441 and presented in the literature, has a very high proportion of low energy neutrons,
 442 leading to very high transmutation rates (particularly in W), which is useful here to
 443 illustrate the concept. Indeed after only 5 years of exposure to the HFIR neutron field,
 444 the initially pure W material contains more than 50 atomic % of transmutation products
 445 different from W, including Os, Pt, Re, and Ir.

Figure 9: (a) Total PKA spectra and (b) cumulative PKA distributions for pure Fe in the equatorial FW of a DEMO fusion reactor as a function of the cooling and tritium breeding concept. See the main text for details of the designations of the different concepts. The total PKAs and above 10 eV average PKA energies for each distribution are given in Table 2.

446 Fig. 10 shows how the total PKA spectrum and cumulative distribution varies
 447 in the material as a function of time. These curves have been obtained from the
 448 SPECTRA-PKA code with recoil cross-section matrices calculated by NJOY using the
 449 same TENDL libraries as used in the inventory calculations. For computational rea-
 450 sons, and also because low concentration nuclides do not contribute significantly, only
 451 nuclides making up more than 0.1 atomic % of the composition at each time were in-
 452 cluded in the PKA calculations. The total PKA rates and energy averages at each time
 453 are also given in Table 2.

454 At first glance, the total PKA spectra in Fig. 10a do not look all that different.
 455 However, Table 2 shows that the total PKAs (above 1 eV) is varying quite significantly,
 456 dropping by almost 40% between $t = 0$ and $t = 5$ years, while, at the same time,
 457 the average PKA energy increases by roughly the same fraction. Note that the mate-
 458 rial density and molar mass assumed for all compositions, and used to calculate the
 459 PKAs $\text{s}^{-1} \text{cm}^{-3}$, was the same as that in pure W (see Table 1). In reality, both the
 460 density and molar mass change slightly with time, but the difference is not significant
 461 enough to produce a visible change in the PKA distributions.

Figure 10: (a) Total PKA spectra and (b) cumulative PKA distributions for initially pure W as a function of total exposure time to a typical neutron spectrum for the HFIR test reactor. The total PKAs and above 10 eV average PKA energies for each distribution are given in Table 2.

Table 2: Total PKAs (above 1 eV) and average PKA energy (above 10 eV) for Fe in different fusion DEMO irradiation fields, and for W as a function of time in the HFIR experimental reactor.

Element	Conditions	Total PKAs above 1 eV (PKAs s ⁻¹ cm ⁻³)	Average PKA energy above 10 eV (keV)
Fe	FW hcpb	4.33E+14	18.8
Fe	blanket (middle) hcpb	3.00E+14	10.3
Fe	blanket (rear) hcpb	2.58E+12	6.0
Fe	blanket backplate hcpb	7.83E+11	5.5
Fe	Shield hcpb	2.03E+11	2.8
Fe	Vacuum Vessel hcpb	4.31E+10	1.6
Fe	FW hcll	4.48E+14	20.5
Fe	FW wcll	4.21E+14	20.8
Fe	FW wccb	2.21E+14	31.5
W	HFIR t=0	8.82E+15	1.0
W	HFIR t=6 months	7.30E+15	1.2
W	HFIR t=1 year	6.43E+15	1.5
W	HFIR t=2 years	6.00E+15	1.6
W	HFIR t=5 years	5.68E+15	1.8

4. Application to radiation damage

Ideally, PKA distributions such as those presented here would be used in combination with detailed atomistic simulations of displacement cascades to predict the production of damage, perhaps followed by Monte-Carlo approaches to trace the long-term evolution of large distributions of defects. Describing the defect formation in displacement cascades and subsequent evolution is currently receiving significant attention, with several groups working on using a combination of experimental observations and atomistic simulations, particularly for W [19, 22], to define defect clustering and size-scaling laws. However, while those works are still maturing, here we consider simpler approximations that can make use of the PKA spectra presented in this work.

Firstly, the Frenkel pair (FP) production rate can be approximated by combining elemental PKA spectra, such as those in Fig. 6 for the DEMO FW, with distributions as a function of ion energy (E_{ion}) of the FPs-per-ion (N_{FPs}) calculated using a suitable binary collision approximation (BCA) code – and in present work the well-known Monte-Carlo based SRIM[17] program has been used. For each PKA species predicted for neutron irradiated Fe and W (i.e. Fe, Cr, Mn, He, and H into Fe, and W, Ta, Hf, He, and H into W - see Fig. 6) a range of E_{ion} values were simulated, covering the necessary range of predicted PKA energies for each ion type. For a given E_{ion} , 1000 ions were simulated in SRIM's full cascade mode to produce a statistically reliable average number of vacancies per ion, which is equivalent, in this approximation, to N_{FPs} . For the threshold displacement energy E_d , which controls the minimum energy required to produce a stable FP, standard literature values (see, for example, Table II in [23]) of 40 eV and 90 eV were used for Fe and W, respectively. The resulting N_{FPs} versus E_{ion} curves have been fitted to appropriate mixed polynomial and logarithmic functions using a nonlinear least-squares Marquardt-Levenberg algorithm built into the GNUPLOT [24] data plotting program.

Fig. 11 shows both the raw SRIM results (as points) and the fitted curves (as lines) for the ten ion implantation types necessary for the simulation of PKAs in Fe and W. Note that the functions are only fitted over the necessary range of energies output from SPECTRA-PKA. The functional form used, and fitted parameters obtained, for each

Figure 11: Number of Frenkel pairs per ion N_{FPs} as a function of ion energy E_{ion} predicted using the BCA code SRIM [17] for various ion types in pure Fe and W. (a) results for the heavy ions (Fe, Cr, and Mn into Fe, and W, Hf, and Ta into W) and (b) results for light He and H ions. The plotted points are the raw results from SRIM, and the curves are the fitted functions calculated using GNUPLOT [24]. In each plot the main graph shows the results on linear axes, while the inset graph uses log-log axes to better present the results at low ion energies.

N_{FPs} vs. E_{ion} curve shown in the figure are given in the supplementary material.

Fig. 12 shows the FPs $\text{s}^{-1} \text{cm}^{-3}$ distributions obtained by combining the fitted FP curves with each of the elemental PKA curves from Fig. 6. Note that for both Fe (Fig. 6a) and W (Fig. 6b) only the FP distributions produced by PKAs of the original host element have a lower limit at the E_d values used in the SRIM calculations. While these values of E_d , chosen somewhat arbitrarily from the literature, influence the results for all PKA species as far as absolute FP numbers are concerned, it is only the self-PKA distributions that have energies in the 10s of eV range or below (due to neutron scattering) and are thus truncated by E_d . Table 3 gives the total FP production rate from each PKA species, together with overall total FP production rate for the irradiated Fe and W. The table and figure demonstrate the expected dominance of the damage produced by the host element (Fe or W), particularly in the case of W, where the FPs produced by the W PKAs represent almost 100% of the $1.65 \times 10^{16} \text{ FPs s}^{-1} \text{cm}^{-3}$.

Confirming the earlier assertion (see Section 2.1) the figure shows that the damage,

Figure 12: The Frenkel pair production rates for (a) Fe and (b) W under the DEMO hcpb FW neutron irradiation scenario predicted using the BCA code SRIM [17]. The plot shows the curves for each of the different PKA distributions shown in Fig. 6.

Table 3: Frenkel pair production rates in Fe and W from the different PKA species generated under the neutron field predicted in the FW of a conceptual design of a hcpb DEMO fusion reactor.

Element	Frenkel Pair production rates (FPs s ⁻¹ cm ⁻³)					
Fe	Frenkel Pairs from:					Total
	Fe	Mn	Cr	He	H	
	7.38E+16	4.80E+15	1.49E+15	7.14E+13	6.41E+13	8.02E+16
W	Frenkel Pairs from:					Total
	W	Ta	Hf	He	H	
	1.65E+16	8.27E+12	6.19E+12	4.73E+11	4.47E+11	1.65E+16

in this case measured as theoretical FP production, caused by the high energy light protons and α particles is indeed low compared to the heavier PKA species, even accounting for the reduced probability of production. For example, in Fig. 6a, the peak PKA rate from Mn is similar to that of the protons produced from the same reaction channels on Fe. The protons, however, are mainly generated in the 5-10 MeV range compared to the few 100s of keV range of the Mn, but nonetheless produce a peak FP rate that is at least two orders of magnitude lower than that from Mn. This observation justifies the omission of the light particle PKAs from the total PKA curves discussed earlier, and that the α /proton distributions should be used to generate appropriate SKA distributions that can then be included in the total PKA distributions.

Fig. 12, for Fe and W, also illustrates the relative importance of different PKA energies to ‘damage’ production. Earlier, it was observed that the average PKA energy for Fe and W under the DEMO FW conditions was less than 20 keV (see Table 1). However, after multiplying by a damage measure, in this case the FP production predicted by SRIM, it is clear that the important range of PKAs is much higher than this. In Fig. 12a, the dominant $\text{FP s}^{-1} \text{ cm}^{-3}$ curve from Fe PKAs has a maximum of almost $1 \times 10^{15} \text{ FP s}^{-1} \text{ cm}^{-3}$ at around 300 keV, with a broad range of PKA energies – from 10 to 900 keV – giving FP production rates above $1 \times 10^{14} \text{ FP s}^{-1} \text{ cm}^{-3}$. Thus, when performing modelling and simulation of damage creation and evolution, a large range of PKA energies must be considered.

On the other hand, in W, Fig. 12b, indicates that there is instead a very pronounced peak in damage due to PKAs at approximately 100 keV in energy, which is very different from the 3.2 keV average obtained from the raw PKA curves.

4.1. Stochastic cluster dynamics

The BCA calculations discussed above are a very rough approximation of damage produced under irradiation, and in particular do not take into account the evolution of defect populations, via migration, recombination and clustering. An improved approach that attempts to include some aspects of this behaviour was investigated by Marian *et al.* [25, 26], who have employed the stochastic cluster dynamics method to evaluate irradiation damage accumulation on pure Fe [25] and W [26] including the

536 effect of He and H.

537 Here we use SCD to explore defect clustering during the initial stages of irradiation exposure of Fe and W in the same DEMO FW spectrum considered above. The
 538 SCD computational methodology for solving the stochastic variants of the mean-field rate theory ODEs described in [25] has been modified to directly use the PKA-rate
 539 source terms calculated in the present work, **without any explicit reference to the neutron irradiation spectrum. This is instead of the original approach to modelling neutron irradiation discussed in [26], whereby neutron energies were sampled from the neutron spectrum and then individual neutrons were introduced and slowed down producing a sequence of PKA events selected from the predicted total PKA distribution.** For the
 540 present purpose, the SCD code samples the cumulative PKA distributions of heavy recoils for Fe and W shown in Fig. 7b (*i.e.* without considering the implications of
 541 different recoil species) at the total PKA rates given in Table 1. Once a PKA energy E_{PKA} is chosen, the number of FPs created, N , is governed by a non-linear damage
 542 production law that reflects the deviation from the NRT formalism as given by molecular dynamics (MD) analysis. This law is a simple power law for W:
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$$N = aE_{\text{PKA}}^b, \quad (3)$$

552 or an exponential correction on the NRT linear relationship for Fe:

$$N = \eta \frac{kE_{\text{PKA}}}{2E_{th}}, \quad (4)$$

$$\eta = \exp(-cE_{\text{PKA}})$$

553 Here a , b , and c are adjustable parameters defined via statistics extracted from MD
 554 simulations of displacement cascades. For Fe, $c = 3.57$ as given by Malerba *et al.* [27],
 555 and for W, $a = 1.49$ and $b = 0.82$ [28] (E_{PKA} in keV). Clusters of various sizes are
 556 then inserted according to a binomial distribution that obeys the global defect cluster
 557 fractions until N is exhausted. In W, these fractions are 0.5 and 0.2 for self-interstitial
 558 atoms (SIAs) and vacancies, respectively (Fikar and Schäublin [29]), while in Fe they
 559 are 0.55 and 0.25 [27]. Note also that there are hard limits of 0.62 (W) and 0.33 keV
 560 (Fe) in each case [28, 27] below which a PKA does not produce any damage.

Alongside this introduction of clusters directly in cascades, the populations evolve via diffusion, which varies as a function of cluster size and nature, via binary reactions, by recombination, and by annihilation at sinks. In the latter case, dislocations and grain boundaries are considered inexhaustible defect sinks [26]. The sink strengths in each case depend on the grain size and dislocation densities considered. For W, these were $100\ \mu\text{m}$ and $10^{14}\ \text{m}^{-2}$, while for Fe they were $50\ \mu\text{m}$ and $1.5 \times 10^{15}\ \text{m}^{-2}$ (see [25] and [26] for details).

To promote vacancy clustering and SIA survival in this formulation we follow [26], and insert He into the system at rates calculated by the inventory simulator FISPACT-II [7] for each material under the same neutron irradiation conditions. For the DEMO FW scenario considered here the calculated rates are 3.6×10^{-6} and 6.7×10^{-6} appm He s^{-1} for Fe and W, respectively, which are the average values during 1 full power year (fpy) of exposure. Here appm is atomic parts per million. At this point, hydrogen is not considered due to a lack of a full data set, although the SCD formulation can treat another species trivially by construction.

Fig. 13a shows the defect accumulation results in Fe and W during simulation at a temperature of 300K, which was chosen to be in agreement with the effective temperature employed when calculating the nuclear recoil cross-sections with NJOY and the neutron-energy spectrum for the DEMO FW with MCNP – see Section 2. In the figure, the vacancy populations, are split into those that are essentially immobile (more than 2 vacancies per cluster) and the mobile point vacancy defects, comprising the sum of mono- and di-vacancies. Note that the simulation volume assumed in these simulations was $10^{-10}\ \text{cm}^3$ and hence the minimum concentration – when there is one defect in a given population – corresponds to the $10^{16}\ \text{m}^{-3}$ minimum in Fig. 13a (this limit has been relaxed in the Fig. 13b bar graph to give a proper sense of the concentrations at different sizes).

In contrast to the BCA results, where Fe was observed to have the highest FP production rate, the SCD simulations suggest that W will experience the greater defect production rate, with, for example, at least an order of magnitude more mobile vacancy defects after the same simulation time.

The initial annihilation and recombination rate in Fe is much higher than in W,

592 partially due to the differential mobility of defects, which – even at this modest tem-
 593 perature – can be of two orders of magnitude. Furthermore, the combined effect of the
 594 smaller grain size and higher dislocation density in Fe also increases defect annihila-
 595 tion. The overall result is a delay in the formation and accumulation of defects in Fe.
 596 This is further illustrated by comparing the size distribution of defects. For example, af-
 597 ter 48 hours (Fig. 13b) both vacancy and SIA defects in Fe have a significantly smaller
 598 size distribution than the equivalent populations after the same simulation time in W.
 599 Note that for these size distributions we have assumed that vacancies adopt a spherical
 600 shape and that SIAs are arranged as circular loops [26], with the size referring to the
 601 diameter of the corresponding spherical or circular shape.

602 Even though the defect evolution modelled here is not the complete picture, it
 603 nonetheless demonstrates the importance of including evolution when attempting to
 604 quantify and compare radiation damage in materials. Neglecting this evolution in Fe
 605 and W, as was the case in the previous section when using a damage function from BCA
 606 calculations, suggested an entirely opposite comparison between these two important
 607 fusion materials.

Figure 13: SCD simulation results. (a) the evolution in concentration of small point-like vacancy defects (mono- and di-vacancies), larger vacancy clusters, and SIA clusters of any size as a function simulation time in Fe and W. (b) The size distribution of vacancy and SIA clusters after simulating for 48 hours of real time. The dashed vertical line in (a) corresponds to this 48-hours of simulation. In (b) it is assumed that vacancy clusters adopt a spherical shape and that SIAs are arranged as circular loops [26], with the size referring to the diameter of the corresponding spherical or circular shape.

608 5. Summary

609 A detailed understanding of the initial knock-on events generated under neutron
610 irradiation is a vital component in the investigation of radiation damage accumulation.
611 The methodology described in this paper for predicting the spectra of primary knock-on
612 atoms (PKAs) produced when a material is subjected to a field of neutrons should make
613 their calculation a straightforward and routine part of materials research for nuclear
614 applications.

615 Using group-wise incident-to-recoil energy cross-section matrices processed from
616 the latest, modern and complete nuclear data libraries by the NJOY-12 [4] system, a
617 newly written code SPECTRA-PKA collapses this data with a neutron irradiation field
618 of interest, and produces the PKA spectrum for every possible nuclear reaction channel
619 on a target nuclide or sequence of targets. In particular, the unique ability to merge and
620 sum data for several target nuclides, **which goes beyond what was available previously,**
621 **enables SPECTRA-PKA to compute PKA distributions for any material composition,**
622 **including complex alloys, subjected to arbitrary non-trivial neutron fields.**

623 SPECTRA-PKA performs post-processing of the raw PKA spectra separated as a
624 function of nuclear reaction channel to provide summed nuclide and elemental PKA
625 distributions, **and the modern, efficient computational approach has already enabled**
626 **such distributions to be produced for all naturally occurring elements [20, 21].** SPECTRA-PKA
627 also provides total (heavy) PKA distributions, which are particularly suitable for use as
628 sampling distributions in atomistic modelling of radiation damage creation and evolu-
629 tion. Calculations of the summed PKA spectra as a function of material, neutron spec-
630 trum, and irradiation time reveal that there can be significant variation, which would,
631 in turn, result in very different damage production rates.

632 For example, when elemental sum PKA distributions for Fe and W under a charac-
633 teristic fusion reactor neutron spectrum are merged with damage functions – defined as
634 the number of Frenkel-pairs (FPs) for a given recoil energy – calculated using the bi-
635 nary collision approximation (BCA), the results suggest that the rate of FP production
636 is an order of magnitude higher in Fe. However, this approach, which uses the SRIM
637 BCA code, is relatively simplistic as it does not consider evolution, and in this respect

638 it is perhaps nearly equivalent to the damage quantifications based directly on the nu-
639 clear cross-section data, such as the displacements per atom (dpa) measure, which is
640 known to be unsuitable for comparisons between materials [2].

641 Therefore, we also consider another approach which samples the recoil energies
642 from the global (heavy) sum PKA distributions according to the total PKA rate, in-
643 serts the appropriate distribution of defects, and simulates the evolution of objects via
644 a stochastic cluster dynamics (SCD) model. The results for Fe and W under the same
645 conditions suggest a completely opposite picture to the results based on FP data from
646 BCA – W defect production and clustering is much higher than in Fe. This demon-
647 strates the importance of considering evolution when attempting to quantify and com-
648 pare radiation damage in materials. Hence the ability to reliably, efficiently, and accu-
649 rately calculate the complete distribution of primary damage events using SPECTRA-
650 PKA, to provide input to advanced evolution models, is vital.

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