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PKA distributions: contributions from transmutation products and from radioactive decay

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Abstract

The neutrons generated in fusion plasmas interact with materials via nuclear reactions. The resulting transmutations and atomic displacements have life-limiting consequences for fusion reactor components. A detailed understanding of the production, evolution and material consequences of the damage created by cascades of atomic displacements requires, as a vital primary input, a complete description of the energy-spectrum of initial (prompt) atomic displacement events (the primary knock on atoms or PKAs) produced by direct neutron nuclear interactions. There is also the possibility that the radionuclides produced under transmutation will create further PKAs as they decay, and so the rate of these must also be quantified. This paper presents the latest results from the analysis of PKA spectra under neutron irradiation, focussing particularly on the variation in PKA distributions due to changes in composition under transmutation, but also on the PKA contributions from radioactive decay of materials that become activated under irradiation.

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

1. Introduction

It is well established that the neutrons generated by fusion reactions in the plasma of experimental devices and in future fusion power plants will interact with the surrounding materials via nuclear reactions (elastic, inelastic, and non-elastic). Furthermore, the generally high-energy, in the MeV range, of the interactions will induce cascades of atomic displacements, and the resulting accumulation of crystal lattice defects will have life-limiting consequences for reactor structural components. A proper assessment, at the engineering level, of the viability of a reactor, including the maintenance schedule and overall costs of components and their replacements, requires a full understanding of the creation, evolution, and materials consequences of this radiation-induced material damage. This can only be achieved by computational and theoretical modelling of materials, backed-up by experimental validation. The modelling must integrate across

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neutron transport simulations for reactor designs, through nuclear interaction calculations, including transmutation, then to defect production and evolution at all necessary length and time scales, to finally produce engineering relevant predictions.

A full description of the initial, prompt atomic displacement events (the primary knock-on atoms or PKAs) produced when neutrons interact with the atoms of a material is a necessary input to atomic modelling of damage production. Along with predictions of compositional changes due to transmutation, such PKA data transfers the maximum amount of information garnered from neutron transport simulations. In particular, it goes well-beyond the level of detail provided by integrated quantities such as the ubiquitous dpa (displacements per atom) measure. Furthermore, it can be used to guide the planning and design of ion-irradiation experiments, where the ions are used as a surrogate for neutrons, which avoid the radioactivity issues of neutron irradiation and can also implant damage more rapidly, allowing experimental results on project timescales.

Complete evaluation of PKA events requires all possible nuclear reaction channels to be considered, including not-only elastic and inelastic scattering, but also non-elastic reactions where the incident neutron is captured followed by the emission of one or more light particles (protons, α-particles, γ-rays, *etc.*). Complex and complete nuclear data libraries must therefore be read and processed, before being combined with predictions of the neutron fields, to produce the energy spectrum of PKA fluxes.

In this paper we present some of the latest results obtained using the SPECTRA-PKA [1] code developed for this purpose, which show, in particular, how the transmutation-induced changes of material composition under neutron irradiation can also produce non-negligible changes to the energy distribution of nuclear recoils (PKAs) as the material is irradiated further. Furthermore, some of the transmutation products, which may be of different elemental nature to the original material, may be radioactive, leading to additional recoils via decay. Here, we present a conservative (worst-case) approximation of the contributions to PKAs from this radioactive decay, and hence illustrate that they could be important for some materials.

2. Methodology

The full computational methodology used to calculate PKA spectra for a given composition of input nuclides has been fully described in [1]. Briefly, the nuclear data processing code NJOY-12 [2] is used to derive, from the raw nuclear reaction cross section data (for the present work from the TENDL-2014 [3] library), energy dependent recoil cross-section matrices for every possible reaction channel of a given target nuclide under neutron irradiation. The newly developed SPECTRA-PKA code then reads the set of matrices for every nuclide (target) x in the composition, and folds each $\mathbf{M}^{x\to y}$ matrix with the neutron energy spectrum $\mathbf{\Phi}$ (a vector) to produce the $\mathbf{R}^{x\to y}$ vector for the $x \to y$ reaction channel, viz:

$$\mathbf{R}^{x\to y} = \mathbf{M}^{x\to y} \cdot \mathbf{\Phi}, \tag{1}$$

or, equivalently

$$\{r_i^{x\to y}\} = \sum_j m_{ij}^{x\to y}\phi_j, \tag{2}$$

where $r_i^{x\to y}$ is the PKA rate at recoil energy i, computed by folding the ith row $m_{ij}^{x\to y}$ of $\mathbf{M}^{x\to y}$ in cross-section units of barns (1×10^{-24} cm^2) with the ϕ_j flux values of the incident neutron spectrum $\mathbf{\Phi}$ in units of neutrons cm^{-2} s^{-1}. Note that for the present work, both the incident and recoil energies use a high-resolution group structure containing 709 bins covering the range 1×10^{-5} eV to 1 GeV.

SPECTRA-PKA then computes, further, the total PKA spectrum $\mathbf{R}^y$ for each nuclide y, by summing, via

$$\mathbf{R}^y = \sum_{ij} w_i \mathbf{R}^{x_i \xrightarrow{c_j} y}, \tag{3}$$

where w_i is the atomic fraction of target nuclide x_i in the material composition, and the contribution from each x_i to the PKAs of y is the sum over all possible reaction channels c_j that produce y from x_i. The program also calculates elemental PKA distributions (*i.e.* by summing over all relevant y nuclides), and the total PKA distribution for the material.

3. PKA distributions as a function of irradiation time

An important aspect of PKA spectra, is how they vary with material isotopic composition. In particular, as a material, perhaps initially composed of a single, pure element, is irradiated by a continuous flux of neutrons its composition will change by nuclear-reaction transmutations. The simulation code FISPACT-II [4] evolves in time a material composition under neutron irradiation, and then the composition at a given time can be fed into SPECTRA-PKA to compute the PKA spectra that would be experienced by the material at that instant in time. For example, Fig. 1 shows the variation with irradiation time of the PKA distributions, summed as a function of recoiling element, in initially pure tungsten exposed to the continuous neutron flux spectrum predicted for the first wall (FW) of the proposed demonstration fusion power plant DEMO. The DEMO neutron spectrum was calculated using the Monte-Carlo neutron transport code MCNP [5] using a finite element conceptual model for a DEMO reactor with a helium cooled pebble-bed (HCPB) tritium-breeding blanket – see [6] for more details, including the plotted FW spectrum (figure 4) –, and has a total flux (see figure 3 in [6]) of 6.6×10^{14} n cm^{-2} s^{-1}.

At $t = 0$ in Fig. 1a, we see that the PKAs are dominated by the recoils of the primary host atoms of W, with only minor contributions from transmutation products. Since there are now impurities in the composition initially, the only way to produce a non-W recoil is directly in a transmutation nuclear reaction. For example, the tantalum recoils are created mainly by (n, p) (neutron capture followed by proton emission) reactions, which also produce the proton distributions. Similarly, the hafnium and α distributions are created mainly by (n, α) (neutron capture followed by α-particle emission) reactions. There are no direct, neutron-induced reactions that can produce

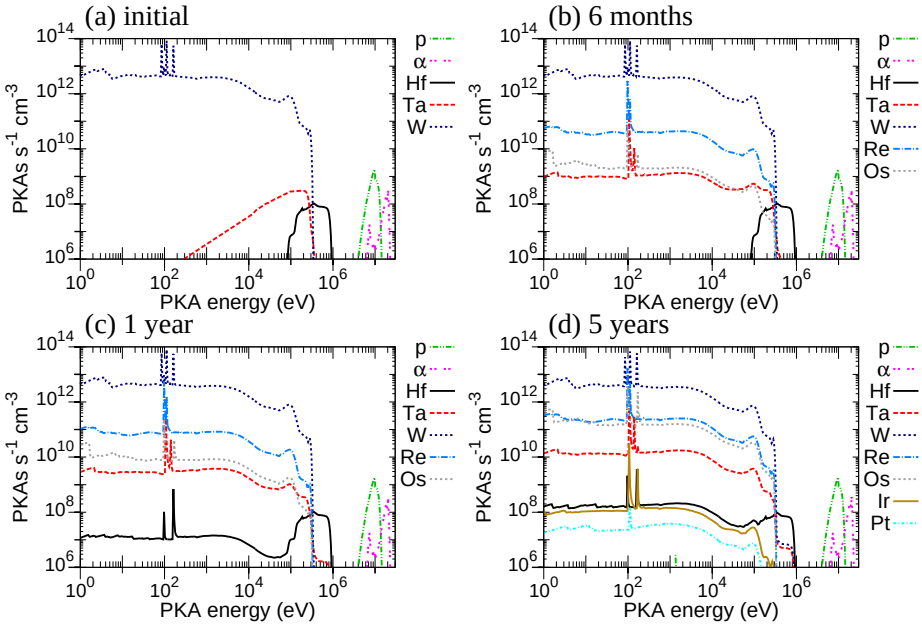

Figure 1: Variation in time of the instantaneous distributions of elemental PKA spectra in initially pure W exposed to a DEMO first wall neutron spectrum. (a) shows the PKA distributions before irradiation, at $t = 0$, with the distributions in the remaining pictures representing the PKA distributions for the material composition after the full power irradiation time given in the top left of each plot. Note that an isotope is included in the list of possible targets in SPECTRA-PKA if it contributes more than 1E-4 atomic % of the composition. The PKAs s^{-1} cm^{-3} flux units are computed from the raw PKAs s^{-1} per target results from SPECTRA-PKA using a standard W density of 19.3 g cm^{-3} and the molar mass of the composition at each irradiation time.

atomic species with a higher proton number than W, and so there are PKAs of heavier elements, such as rhenium or osmium.

However, as the irradiation proceeds the combined occurrence of pure neutron capture (n, γ) reactions and subsequent β^- decay of unstable W isotopes (mainly ^{185}W and ^{187}W) produces Re isotopes, which can capture further neutrons and then decay to produce Os, and so on. At this point, the newly created Re, Os, *etc.* , atoms (in fact isotopes), as well as those of Ta, Hf, *etc.* , from other pathways involving nuclear break-up reactions, become new "targets" for the PKA calculations with SPECTRA-PKA. This results in a significant change in the elemental PKA spectra as the irradiation time increases. After 6 months (Fig. 1b) there is a measurable distribution of Re PKAs, contributing 0.7% of the total number of PKAs. This Re distribution continues to increase as the irradiation time increases, and is later joined by significant spectra of Os and Ta PKAs, and together, after 5 years of continuous irradiation, these three spectra contribute more than 8% of the total number of PKAs (1.5×10^{14} PKAs s^{-1} cm^{-3} out of a total of 1.8×10^{15}).

Note that the probabilistic nature of nuclear reactions and decays means that the pure W composition immediately becomes a complex mixture of many elements and

isotopes as soon as the irradiation begins, and so, in Fig. 1, there are minor distributions of Os, Ta, *etc.* , even at 6 months. As indicated in the figure caption, for the purposes of the present calculations, the list of target nuclides input into SPECTRA-PKA only included those nuclides that contributed more that 1E-4 atomic % at the particular irradiation time. This limit was mainly chosen for computational reasons to avoid reading and processing the recoil cross section matrices for very many unimportant nuclides. For example, after 5 years of irradiation, the composition of W contains 225 different nuclides but only 38 are at concentrations above 1E-4 atomic %.

4. Recoils from radioactive decay

As discussed above, under neutron irradiation, a material's composition may change due to transmutation nuclear reactions and this can change the distributions of PKA spectra. Furthermore, some of the nuclides created by nuclear interactions may be unstable. As these radionuclides decay, the energy released will induce further recoil of the decayed atomic nucleus. Here we assess, using a rough approximation, whether these additional PKAs could contribute non-negligibly to the total PKA population during irradiation. At the same time, it is important to note that even if this is not the case, these "decay-PKAs" will be the only PKAs in the material after shutdown, or during a period of scheduled maintenance, when the neutron field is no longer incident, and so their evaluation is necessary in order to understand how a damage may accumulate in a material post-irradiation.

In the present work, we restrict the analysis to simple two-body decays, where the decay occurs by the emission of a neutron, proton, or α-particle, and β^- or β^+ decay. These five decay modes comprise 80% [7] of the total decay reactions, across all radionuclides, for which data is available in the decay library used with the FISPACT-II [4] inventory code. More than 10% of the other decay reactions are either isomeric transitions, for which there are no additional recoils, or spontaneous fission, while the remaining, more complex break-up reactions, where two or more decay products are emitted, are rare, particularly for the main materials of interest for fusion applications.

To approximate the possible PKA energy associated with a decay recoil, we calculate, for each $x \rightarrow y$ decay reaction, the mass difference between the initial radionuclide "parent" and the total mass of the residual "daughter" nuclide combined with the mass of the emitted particle. Then the energy of the residual, recoiling atom is calculated via conservation of momentum. For β decays this is a conservative maximum because, in reality, there is a spectrum of β emission energies, since an emitted neutrino or antineutrino, which we neglect here, will also take some of the decay energy.

For two-body decay, we can calculate the recoil energy using the laws of conservation of both energy and momentum. Where the rest mass of the emitted particle is the same order of magnitude as the residual, as in, for example, α decay, its velocity will be much less than c, and so the kinematic momentum applies. In this case the recoil energy E_r^y of residual y is calculated via:

$$E_r^y = \frac{Q^{x \rightarrow y}}{1 + \frac{m_y}{m_z}}, \tag{4}$$

where $Q^{x\to y}$ is the energy (or "Q-value") calculated from the mass difference for the $x \to y$ decay reaction, and m_i is the rest mass (in kg) for the daughter residual y or emitted particle z.

For β decay, where the mass of the emitted particle is low, the velocities may approach c, and so we must consider its relativistic momentum. The energy of the residual, which is still treated kinematically, is then calculated from:

$$E_r^y = Q^{x\to y} + (m_e + m_y)c^2 - \sqrt{(m_e + m_y)^2c^4 + 2m_yQ^{x\to y}c^2}, \tag{5}$$

where, for β decay, the rest mass of the emitted particle is m_e, the mass of an electron. Note that in the case of positron emission (β^+) decay, the mass difference used to calculate the Q-value includes the subtraction of $2m_e$ to account for the negative charge on the daughter atom and thus extra electron in the system, compared to β^- decay where the charge on the daughter atom is positive and cancels with the emitted electron and this the mass difference is simply that between the parent and daughter. Furthermore, in the FISPACT-II decay file there is no distinction between positron emission and electron capture – they are both identified as β^+ – but only proper positron emission reactions would result in a positive Q-value.

Fig. 2 shows (as curves) the elemental PKA spectra computed for (a) Al, (b) Fe, and (c) W under the same DEMO HCPB FW neutron spectrum discussed already. For each element, the input composition to SPECTRA-PKA is that predicted by FISPACT-II for each material after 1-year of continuous irradiation in the neutron spectrum. For the radionuclides present in the composition at this time, the Becquerel activity per unit mass – computed using the nuclide's half-life – is part of the output produced by FISPACT-II. A particular radionuclide may undergo several different decay reactions, each with a different recoil energy, but the branching ratio (also available from the decay file) allows the correct number of Becquerels (decays per second) to be attributed to each decay mode, thus giving the number of recoils per second per unit mass of the material produced by that reaction. Numerically, the procedure to calculate the number of y recoils for a particular $x \to y$ decay reaction is:

$$N_r^{x\to y} = A_x b_{\text{ratio}}^{x\to y} \equiv c_x \lambda_x b_{\text{ratio}}^{x\to y}, \tag{6}$$

where $N_r^{x\to y}$ is the number of y recoils (PKAs) produced by the $x \to y$ decay per second per unit mass, A_x is the Becquerel activity per unit mass for the concentration c_x of radionuclide x with decay constant $\lambda_x (= \ln 2/T_{1/2}^x)$, and $b_{\text{ratio}}^{x\to y}$ is the fractional branching ratio of the decays of x that follow the $x \to y$ path.

The $N_r^{x\to y}$ values (converted to per unit volume using the appropriate material densities) obtained for irradiated Al, Fe, and W after the 1-year irradiation are shown as points in Fig. 2, with the element type of each decay recoil indicated by the type of point used. Note that in the key of each plot, some elements are represented by both a line and point – indicating that they are present as both PKAs from direct neutron interactions and also as the recoiling species following radioactive decay. Meanwhile, some elements only appear as points, meaning that they are only produced as PKAs by decay and not in significant quantities by direct nuclear reactions (there may be a neutron-induced PKA spectrum for these elements, but the rates are too low to be

seen in the plots). Furthermore, none of the radionuclides produced in Al, Fe, or W under neutron irradiation undergo decay by proton or α emission that produce recoiling protons or αs in the plot range shown, and so these entries in each key are lines only.

Fig. 2 demonstrates an obvious point; that for the assumed two-body decays the energy of the residual recoils is inversely proportional to the mass of the decaying radionuclides. Thus, in Al, where the nuclide masses are relatively light, the decay-PKA energies are generally higher than those in the heavier Fe, which itself has more energetic decay-PKAs than in W – the heaviest of the three elements. This means that the damage induced by decay PKAs may be more significant in lighter materials. Indeed, in W, none of the decay-PKA energies calculated in the current methodology and appearing in the plot (Fig. 2c) are above the standard atomic threshold displacement energy E_d for W of 90 eV [8] (although there are a small number of decay PKAs above the threshold, but at too low a rate to appear in the plot – see table 1).

Table 1 quantifies the relative comparison between decay recoils and recoils from nuclear reactions. For each of the materials and compositions considered in Fig. 2, the total number of PKAs due to direct neutron-induced reactions or from the decay of radionuclides are computed. Secondly, the average PKA energy (in keV) in these totals is also calculated. In both cases, only the PKAs with energies greater than the standard [9, 8] threshold displacement energies (also given in the table) for the three elements are included in the calculation. Note also that for the neutron-induced PKAs, the total and averages do not include the contributions from α-particles or protons because, as was discussed elsewhere [1], these produce very different types and amounts of damage, and so it is inappropriate to include them in the global quantities with the PKAs of the other heavier elements.

For Al table 1 shows that there are a significant number of PKAs from decay, although the total number is still more than two orders of magnitude lower than that from neutron-induced PKAs, and the average energy is much less – only 500 eV compared to an average of 56 keV from the irradiation. In Fe there are even less decay-PKAs, and the average energy has dropped by a factor of five (compared to only a factor of 2 decrease in the average PKA energy from the neutron irradiation between Al and Fe). In W, we have already seen in Fig. 2c that there are essentially no decay-PKAs above $E_d = 90$ eV, although the total in the table is non-zero because of some rare, but relatively energetic, α decays (mainly) from the very long-lived naturally occurring isotopes of W, which also lead to a higher than expected average decay-PKA energy of 36.8 keV.

5. Summary

This paper has presented some of the latest findings from work to analyze and quantify the initial atomic displacement (the PKAs) produced in materials under exposure to a neutron irradiation field. In particular, we have shown that the compositional changes brought about by transmutation reactions under neutron irradiation can also change the predicted spectra of PKAs – most notably in highly transmuting materials such as tungsten, where the high rate of production of transmutant Re, Os, and Ta, causes their contributions to the PKA distributions to become non-negligible as the irradiation time increases.

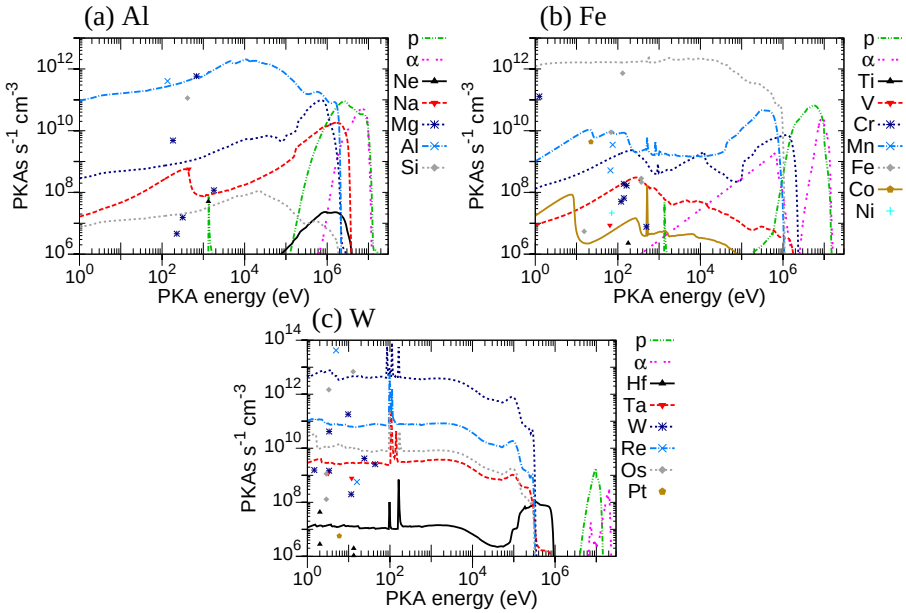

Figure 2: Neutron-induced elemental PKA spectra and decay-recoil rates as a function of recoil energy after a 1-year continuous irradiation in a fusion DEMO first wall spectrum of initially (at $t = 0$) pure (a) Al, (b) Fe, and (c) W. For the neutron-induced PKA spectra, an isotope was included in the target list given to SPECTRA-PKA if it contributed more than 1E-4 atomic % of the composition after 1 year of irradiation. The PKA rates in s^{-1} cm^{-3} units are computed from the raw PKAs s^{-1} per target from SPECTRA-PKA or from the recoils s^{-1} kg^{-1} values calculated for radioactive decay of the composition (see the main text) using standard material densities of 2.7, 7.9, and 19.3 g cm^{-3} for Al, Fe, and W, respectively.

The additional recoils due to the subsequent decay of radionuclides produced by transmutation reactions have also been considered for, as a first approximation and neglecting any massless particles, the dominant two-body decays such as β or α decay. The results demonstrate that, particularly for light materials, these decays may contribute non-negligibly to the total PKA production rate under neutron irradiation, and, furthermore, that they will produce a continuous flux of PKAs in the material long after the neutron irradiation has been turned off.

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Table 1: Total PKA rates and average PKA energies from both neutron irradiation and radioactive decay for the material compositions in initially (at $t = 0$) pure elements after a 1-year continuous irradiation in the predicted neutron field for the first wall of a HCPB DEMO fusion reactor. Each value is computed only for those PKA energies above the standard [9, 8] displacement threshold energies E_d of the elements – also given in the table. The cm^{-3} units were computed from the raw data using standard material densities (given in the main text).

Element	E_d (eV)	Total decay PKAs (PKAs s^{-1} cm^{-3})	Average decay PKA energy (keV)	Total irr. PKAs (PKAs s^{-1} cm^{-3})	Average irr. PKA energy (keV)
Al	27.0	1.10E+12	0.5	1.81E+14	56.0
Fe	40.0	7.39E+11	0.1	3.07E+14	21.9
W	90.0	3.46E-03	36.8	5.62E+14	4.7

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