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Energy spectra of primary knock-on atoms under neutron irradiation

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HIGHLIGHTS

- Recoil cross-section matrices under neutron irradiation are generated.
- Primary knock-on atoms (PKA) spectra are calculated for fusion relevant materials.
- Variation in PKA spectra due to changes in geometry are considered.
- Inventory simulations to consider time-evolution in PKA spectra.
- Damage quantification using damage functions from different approximations.

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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.

1. Introduction

Understanding through modelling of the damage accumulated in materials irradiated by neutrons remains a primary goal for computational simulation of materials for advanced nuclear energy systems. Several different approaches exist for predicting the formation, evolution and behaviour of this damage, including: computationally demanding molecular dynamics simulations of damage cascades with full atomic interactions; rate theory models, where defects are described as objects with defined behaviour; and kinetic Monte-Carlo (kMC) simulations. However, without exception, all of these techniques need, as input at some level, information about the initial primary disruptions, in the form of the type, energy and spatial distribution of the primary knock-on atoms (PKAs). In particular, this kind of data goes far beyond the limited information provided by the traditional and ubiquitous defect index of initial damage formation known as “displacements per atom” (dpa). The so-called Norgett, Robinson and Torrens NRT-dpa [1] reduces the predicted irradiation environment, perhaps obtained from a Monte-Carlo neutron transport simulation on a reactor geometry, to a single number that converts the energy deposited into a material by the irradiation into an estimate of the number of atomic displacements that could be generated. The dpa measure has proven to correlate well with various radiation damage phenomena, but is not well suited to distinguishing between

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different types of irradiation, and, more importantly, it does not provide information about damage evolution. It is also the case that it should not be used as a basis for comparing irradiation behaviour in different materials [2]. A more complete picture of radiation damage evolution, that may be afforded by modern computational techniques, requires the spatial and energy distribution of all the initial displacement events, including both emitted and residual nuclei from nuclear interactions.

In modern nuclear data evaluations, for a single target species, such as the major ^{56}Fe isotope in Fe, there are many nuclear reaction channels that produce recoiling species, including elastic, inelastic, and nonelastic nuclear reactions. In nuclear fusion, in particular, the generally higher energy of the incident neutrons, when compared to fission, leads to many more channels becoming relevant, which in turn produces a more complex distribution of PKAs in both energy and type. These PKAs lead to cascades of atomic displacements, which can subsequently evolve and collapse to produce extended defects such as dislocation loops and voids.

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

2. Methodology and results

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

Two-body elastic and discrete inelastic neutron scattering, charge-particles elastic scattering, continuum scattering and fission can be treated in different ways depending on the completeness and accuracy of the content of the original evaluation. Previously, simple theoretical models had to be employed to plug the gaps in earlier nuclear data libraries, but this is becoming less necessary with the latest libraries [6], which use detailed and well-validated theoretical models to derive data where no experimental information exists. The resulting $m_{ij}^{x\rightarrow y}$ elements of each $M^{x\rightarrow y}$ matrix are the cross-sections in barns for a recoil in energy group i produced by an incident neutron in energy group j.

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

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

Furthermore, the uncertainties associated with the basic nuclear library evaluations are not propagated into the recoil cross-section matrices derived by NJOY. Besides this, there are no error estimates available for the neutron spectra that will be combined with these matrices (see below) to produce PKA distributions, and so uncertainties are not included in any of the results presented in this paper. In the future, it may be possible to consider uncertainties using statistical methods such as the Total Monte-Carlo (TMC) approach [8,9], which can be applied to both input ingredients of a PKA spectrum – the neutron spectrum and the recoil cross-section matrix.

Fig. 1 is a 3D plot of the elastic scattering (n, n) recoil cross-section matrix for ^{56}Fe, while Fig. 2 shows 2D snapshots at several incident energy groups for the main reaction channels of the same nuclide, including elastic scattering. For each snapshot in Fig. 2 at incident energy group j (indicated by a vertical line in the group mid-point in each graph and by the straight lines on the xy-plane of Fig. 1) the curves effectively represent the jth column vectors of the $M^{x\rightarrow y}$ matrices. The figure shows that there is

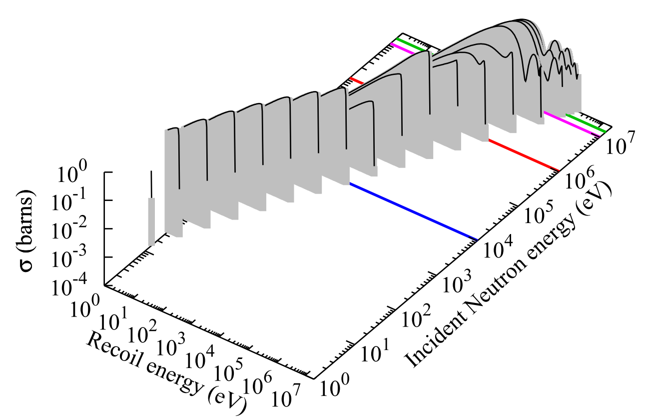

Fig. 1. The recoil cross-section matrix for elastic scattering of neutrons on ^{56}Fe. The matrix is plotted as a separate recoil-energy vs. cross-section distribution for each incident energy group (plotted at the midpoint of the group). Note that only a subset of the possible 709 incident-energy-group 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.)