Journal of Physics: Conference Series

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To cite this article: Michael Fleming *et al* 2018 *J. Phys.: Conf. Ser.* **1046** 012002

High-Energy Activation Simulation Coupling TENDL and SPACS with FISPACT-II

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Abstract. To address the needs of activation-transmutation simulation in incident-particle fields with energies above a few hundred MeV, the FISPACT-II code has been extended to splice TENDL standard ENDF-6 nuclear data with extended nuclear data forms. The JENDL-2007/HE and HEAD-2009 libraries were processed for FISPACT-II and used to demonstrate the capabilities of the new code version. Tests of the libraries and comparisons against both experimental yield data and the most recent intra-nuclear cascade model results demonstrate that there is need for improved nuclear data libraries up to and above 1 GeV. Simulations on lead targets show that important radionuclides, such as ^{148}Gd, can vary by more than an order of magnitude where more advanced models find agreement within the experimental uncertainties.

1. Introduction

The simulation of activation-transmutation processes in nuclear environments requires knowledge of all reaction cross sections that are energetically allowable. This is often handled through the use of nuclear data libraries, which contain cross sections for every unique channel, for example, with incident neutrons, (n,p), (n,2n), (n,α), etc. Below 30 MeV, the number of reaction channels are sufficiently small, and the channels sufficiently distinguishable, that this method has remained in standard use. However, as the incident energy climbs, the number of channels rapidly expands and nuclear data libraries utilise a residual product based data structure which can accommodate arbitrarily many products.

The TENDL-2015 [1, 2] neutron and charged-particle nuclear data files cover some 2809 targets with complete reaction data including all emitted particles for incident energies up to 200 MeV. These data are contained within the international standard ENDF-6 format [3], with full product energy-angle data (in `MF=6`) that may be processed into production cross-section data (in `MF=10`) and utilised by codes such as FISPACT-II [4, 5] for activation simulations. The driving engine behind TENDL, the TALYS code [6], performs model calculations to predict the cross sections for each of these yields. This incurs significant computational expense and, even with the most recent, advanced data, does not provide data for the full spallation yield 'tail' including nucleon differences in excess of a few dozen. This is analogous to the case of fission, where each individual product from the fission process is not considered, but a separate data set of fission yields is coupled with the total fission cross section to determine creation rates for every product. It is standard practice to use other models and software (e.g. GEF [7]) to complement the data.

At energies of a few tens or hundreds of MeV and higher, intra-nuclear cascade models are commonly utilised to simulate the reaction process, generating a set of directly emitted particles and residuals that undergo evaporation, fission and fragmentation processes. Many models (see [8] for a survey and references therein) have been engineered to be used in Monte-Carlo transport simulations, providing an intra-nuclear Monte-Carlo process within the inter-nuclear transport calculation. This method has been demonstrated to be highly effective for transport calculations, but all outputs must be converged in these Monte-Carlo simulations. While for low-mass particle transport this is often a reasonable computational problem, for the production of *all residual nuclei*, it remains a challenge. Individual products of radiological importance may be produced in only one of several thousand or fewer events, requiring (potentially many) millions of reaction simulations in every region of interest.

While in the well-studied fission and fusion applications the nuclear reactions above 30 MeV do not play an essential role, in a large number of related research, such as materials damage studies, nuclear medicine, accelerator-driven research or space exploration, the nuclide inventories and resulting observables from such high-energy reactions are of significant importance.

2. Activation-transmutation simulation

While in some simulation systems (c.f. [9]) the record of each residual nuclide is retained for post-processing and activation-transmutation calculation, an alternative is to rely completely upon nuclear data libraries for this secondary calculation. With this alternative, the incident particle fluxes and spectra are calculated in each region and coupled with complete nuclear data for all relevant energies and reaction channels. This is shown schematically in Figure 1. The 'low-energy' nuclear data referenced in Figure 1 may range from 10^{-5} eV to 30 MeV or run as high as 200 MeV, but the principle is the same: results from Monte-Carlo intra-nuclear model calculations are used to determine *reaction rates* for radionuclide production. 'All-energy' nuclear data is required to compensate for the lack of residual tally data, providing reaction rates for all products.

The FISPACT-II implementation[1] requires the standard 'low-energy' ENDF-6 nuclear data libraries, as well as an extended data library of residual production `MF=10` format. The use of a new keyword, **USESPALLATION**, causes data from both of these sources to be spliced in the reading of energy-dependent cross sections. The user supplies two keyword options: an energy cutoff (in MeV) E_{cut} and a number of isotopes N_{iso} that this should be treated. This is followed by a list of isotopes, typically the stable isotopes present in some irradiated material. Inputting $N_{iso} = -1$ causes FISPACT-II to splice data for all isotopes that possess data. The one group cross sections are then calculated as;

$$\overline{\sigma} = \frac{\left[\sum_{\{i|E_i<E_{cut}\}} \phi_i \, \sigma_{LE,i}\right] + \left[\sum_{\{j|E_j \geq E_{cut}\}} \phi_j \, \sigma_{HE,j}\right]}{\sum_k \phi_k}, \tag{1}$$

where the low- and high-energy library cross sections are subscripted as LE and HE, respectively.

Once the rate equation matrix elements have been calculated, the full machinery of FISPACT-II may be used to calculate all nuclide inventories and responses, as shown in Figure 2 for a one year, 1 GeV proton irradiation of natural tungsten. The production of each nuclide is followed by complete tracking of all decay processes, showing the relative importance of all possible radionuclides. In this case, the presence of ^{148}Gd, with a half-life of approximately 75 years, will remain a dominant contributor and, as an α-emitting nuclide, one of radiological importance [10].

[1] As of `FISPACT-II` v4.00

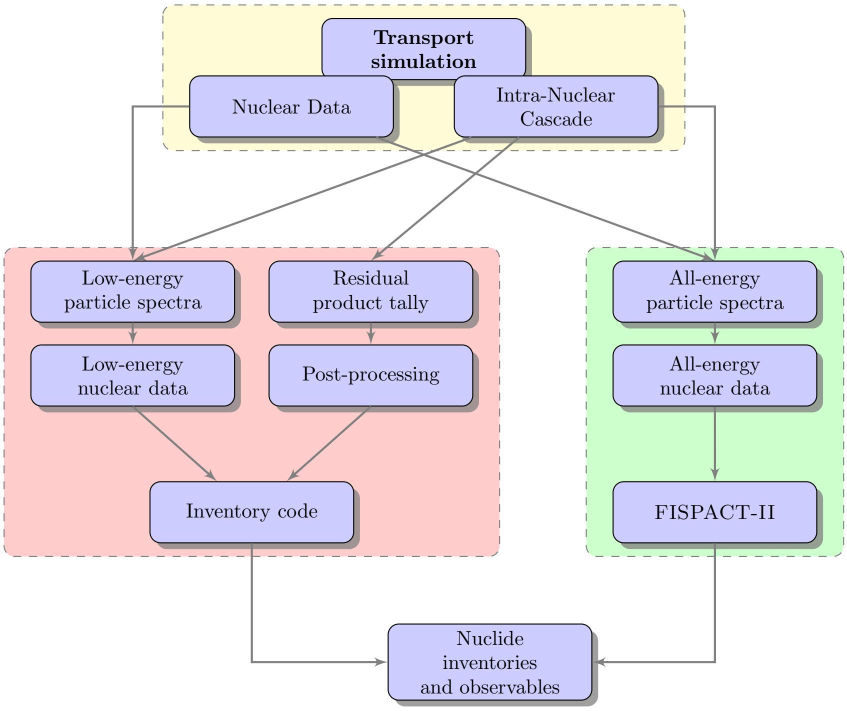

Figure 1. Schematic for activation-transmutation inventory calculations, using a Monte-Carlo transport simulation (yellow), coupled with either (red) separate nuclear data and residual history data or (green) pure nuclear data library calculations.

3. High-energy nuclear data

The essential ingredient required for the full multi-group nuclear data approach is a complete nuclear data library, including all reaction channels and products. The only non-deprecated nuclear data libraries known to the authors are the JENDL-2007/HE [12] and HEAD-2009 [13] libraries, whose content is briefly summarised in Table 1. The HEAD-2009 data is a high-energy extension to complement other nuclear data below 150 MeV and has been created using a suite of model codes, as detailed in the reference report. These contain full emitted particle energy spectra in `MF=6`, but have been processed into residual production group-wise data for FISPACT-II. The JENDL-2007/HE data is a high-energy extension of JENDL data files that utilise different file formats depending on the evaluator methodology.

Table 1. Summary of high-energy nuclear data libraries.

Library	Targets	Energy range (MeV)
HEAD-2009 (neutron/proton)	H1-Bi210 (629 files)	150-1000
JENDL-2007/HE (neutron/proton)	H1-Am242m (106 files)	up to 3000

While the JENDL-2007/HE files only cover 106 targets, the graphs of the yield data in Figures 3 and 4 demonstrate that they are more complete and cover all products. The mixed choice between these two libraries are between target coverage and product coverage. However,

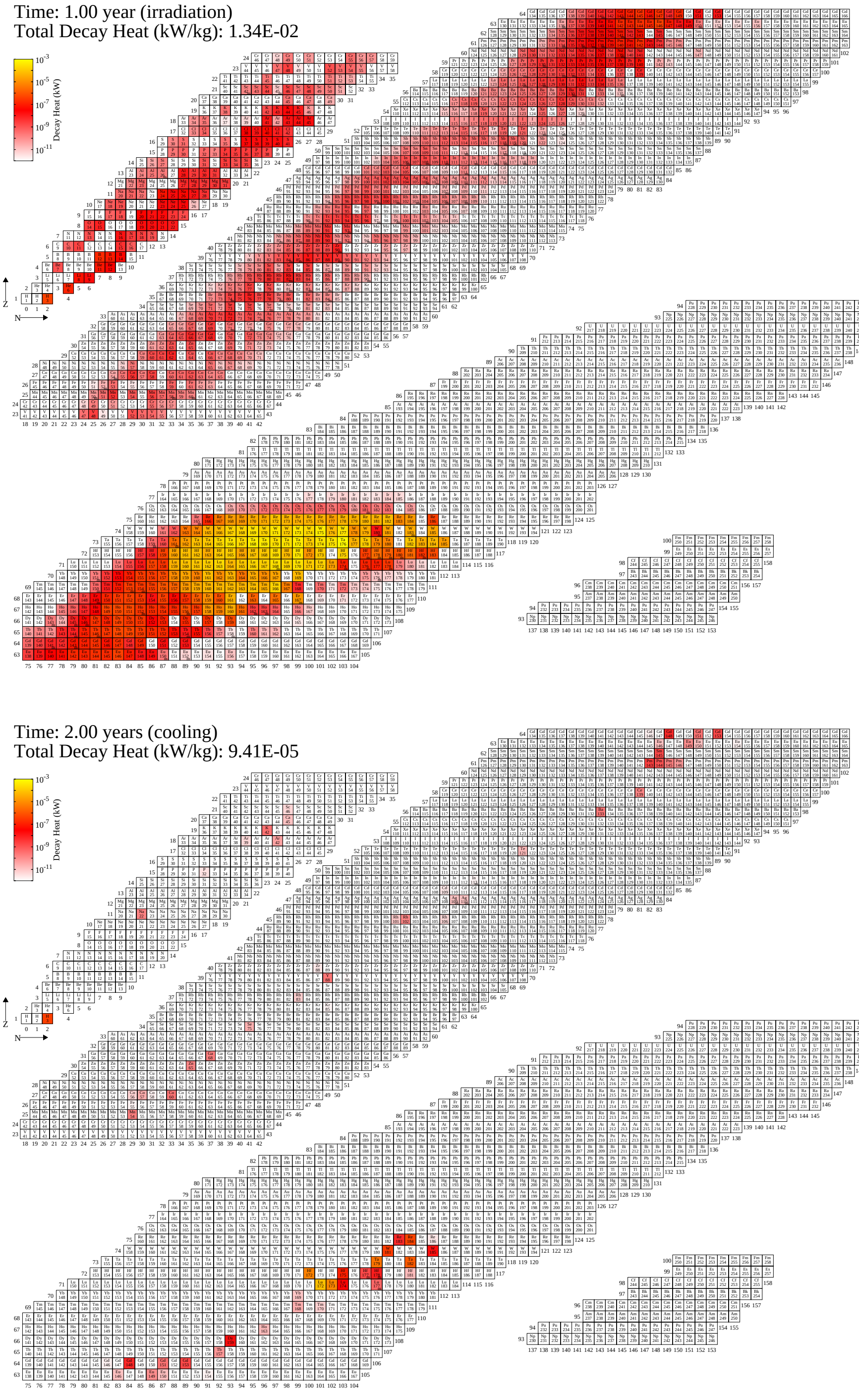

Figure 2. FISPACT-II decay heat simulation [11] for a one year, 1 GeV proton irradiation of natural tungsten, using spliced data from TENDL-2015 up to 150 MeV and the HEAD-2009 library to 1 GeV, supplemented with SPACS data. The top figure shows the decay heat immediately at the end of irradiation and the bottom figure after two years of cooling. The color axis provides the decay heat response for each radionuclide, ranging from tritium to tungsten and above.

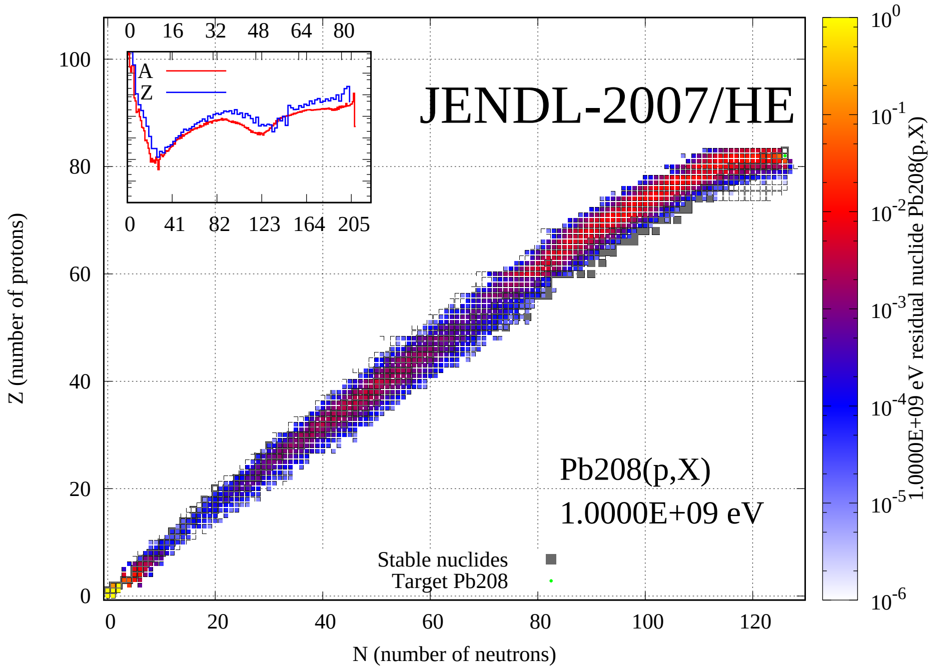

Figure 3. JENDL-2007/HE 1 GeV proton-induced residual product yields, showing a complete data set from target to proton.

the production of more than 1000 products per target highlights the need for more than simply 629 target files, since each long-lived product will itself require negative rate equation matrix elements. To accommodate a true inventory calculation for high-energy reactions, a complete set of reactions are required, including the radionuclide products.

To generate the full tabulated data required for all targets in activation simulations, the semi-empirical isotopic spallation simulation code SPACS [14] was utilised to build complete isotopic yield data files for incident neutrons and protons with energies from 100 MeV to a few GeV. These can be read directly into the FISPACT-II code and used with the complete nuclear reaction data of TENDL to provide a consistent simulation framework including high-energy activation residual products down to approximately half of the target mass.

Moving beyond the completeness of the nuclear data files, the quality should be determined by comparison with available experimental data. The 1 GeV proton-induced Pb208 data of Enqvist et al [15] provides one such comparison, where the agreement found between standard physics list options within Geant4 [16] and the experimental data is much greater than with the nuclear data libraries, as shown in Figure 5.

Of particular interest, given the radiological importance of ^{148}Gd, is the mass yield for 148 as contained within the nuclear data libraries and as predicted by model codes. The order-of-magnitude errors presented by the available nuclear data files is not surprising, given the relative spread of simulation results, but underlines the potential for improvement. As Figure 5 demonstrates, INCL-based calculations and SPACS provide significant improvement in the calculation of this mass yield, as well as many others.

4. Discussion

The recent modifications to FISPACT-II allow users to employ nuclear data libraries to cover both the energies up to 200 MeV with TENDL, as well as extended nuclear data forms up to

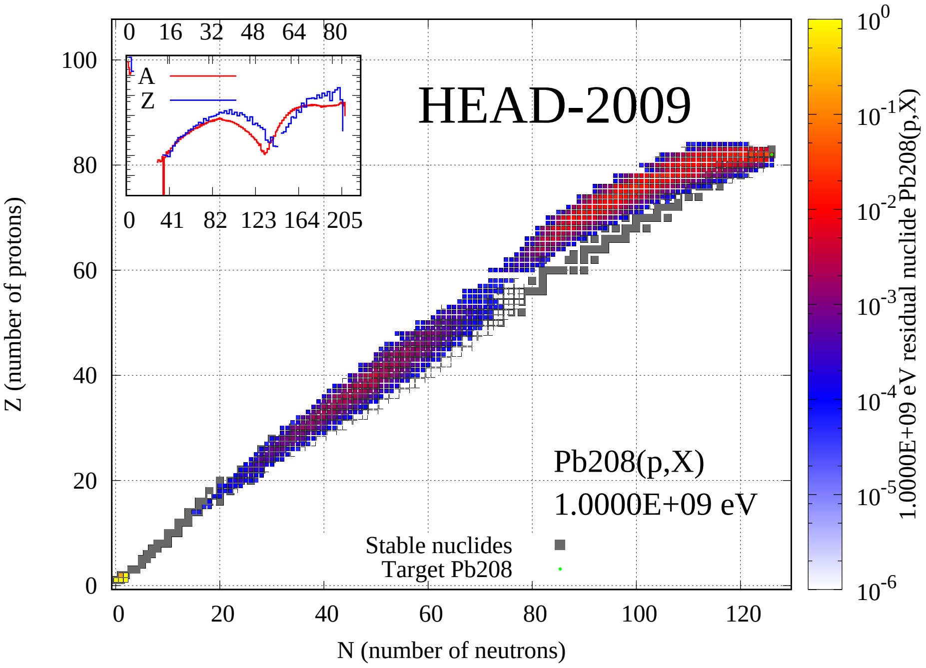

Figure 4. HEAD-2009 1 GeV proton-induced residual product yields, showing an incomplete data set from target to proton, but offering a larger number of incident targets with one reproducible evaluation methodology.

several GeV. For high-energy activation-transmutation calculations, this offers the significant advantage of using converged, reproducible residual nuclide production cross sections, rather than attempting to converge spallation residual tallies.

The remaining challenge is the production of accurate and reliable nuclear data files, although this is identical to the challenge for model codes within Monte-Carlo transport, since the same models are utilised in the production of nuclear data libraries. A survey of the available nuclear data files, including HEAD-2009 and JENDL-2007/HE, has found that both the targets and products are not fully covered in the data. The use of deprecated of intra-nuclear cascade and de-excitation models has been demonstrated with comparisons using the most recent models as available in Geant4. Systematic data generated with SPACS can offer yields for targets not considered in HEAD-2009 or JENDL-2007/HE, but without the processes for and products from fission and fragmentation.

New nuclear data libraries employing the best model codes, as employed in standard Monte-Carlo transport simulations, are required to provide the most accurate activation-transmutation simulations. To ensure reliability, traceability and reproducibility of the nuclear data libraries, precise model versions must be employed in a automated process, much as the TENDL nuclear data libraries are generated using a robust and repeatable method [19, 1].

Acknowledgements

This work was funded by the Research Councils United Kingdom Energy Programme under grant EP/P012450/1. M Fleming would like to thank J-Ch. David, D. Mancusi, A. Koning and K-H. Schmidt for helpful conversations and insights.

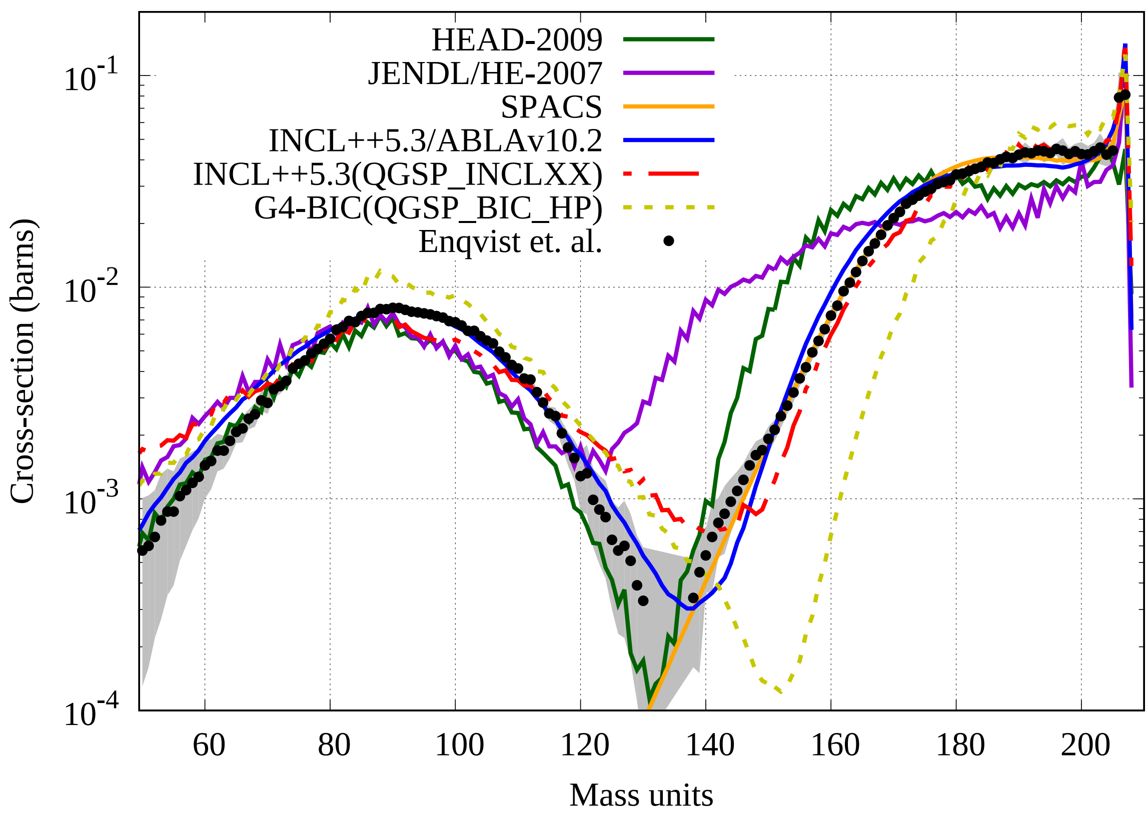

Figure 5. Comparison of nuclear data libraries, SPACS and intra-nuclear cascade model results for 1 GeV proton-induced Pb208 residual product cross sections, compared with experimental data from Enqvist et al [15] with experimental uncertainties in grey. Geant4 simulations were used with multiple physics lists and using an ABLA [17] optional de-excitation mode coupled with INCL++ [18].

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