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HEIR: A High-Energy Intra-Nuclear Cascade Liège-based Residual nuclear data library for simulation with FISPACT-II

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ABSTRACT

FISPACT-II is an advanced activation/transmutation and inventory code that employs the most up-to-date and sophisticated nuclear data forms, including the full TENDL-2017 library. TENDL includes residual nuclide production for incident particle energies up to 200 MeV. To address the needs of activation–transmutation simulation in incident-particle fields with energies up to and above 1 GeV, FISPACT-II has been extended to splice TENDL standard ENDF-6 nuclear data with extended nuclear data forms. Comparisons between JENDL-2007/HE, HEAD-2009, and the most recent intra-nuclear cascade models found within Geant4 led to an updated method for residual nuclide nuclear data production. This has been achieved with an automated INCL++/ABLA process that produces complete residual cross section format (MF=10) for activation/transmutation and inventory calculations. The current High-Energy Intra-Nuclear Cascade Liège-based Residual (HEIR-0.1) library includes proton-induced cross sections up to 1 GeV for 2095 targets. These data are available with the most recent release 4.0 of FISPACT-II.

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 increases, the number of channels expands rapidly and nuclear data libraries utilise a residual product based data structure which can accommodate arbitrarily many products.

The TENDL-2017 [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 libraries, 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.

At energies of a few tens or hundreds of MeV and higher, intra-nuclear cascade models are commonly utilised to simulate the reaction process. These generate a set of directly emitted particles and residuals that undergo evaporation, fission and fragmentation processes, which are typically handled in secondary de-excitation models. Many physics codes (see [7] 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*

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Table 1
Summary of high-energy nuclear data libraries.

Library	Incident	Targets	Energy range (MeV)
HEIR-0.1	p	Li6-Bk251 (2095 files)	up to 1000
HEAD-2009	n, p	H1-Bi210 (629 files)	150–1000
JENDL-2007/HE	n, p	H1-Am242m (106 files)	up to 3000
TENDL-2017	n, p, d, γ, α,t,h	H1-Mc291 (2804 files)	up to 200

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, for each target isotope, in every region of interest.

In the simulation of the production of activation/transmutation products and other post-irradiation quantities, such as decay heat and activity, nuclear data libraries benefit from several notable advantages:

- calculation of all reaction rates for millions of reaction channels can take cpu-**seconds**, based on one set of (relatively) lengthy calculations that generated the data library,
- results can be more easily **reproduced**, as there is no need to run Monte-Carlo calculations while considering model versions and random number seeds,
- databases of results may be more easily **validated** than models,[1] and
- calculation results may be easily **traced** to data library versions, which enjoy a less dynamic version control process.

These advantages have resulted in ubiquitous use of nuclear data libraries for activation/transmutation simulation for most nuclear applications, even (or especially) where nuclear data are the product of semi-empirical models, rather than experimental data. The simulation of higher-energy residual nuclide production cross sections requires a reproducible and traceable process, similar to that of TENDL, that is as automated as is practical.

To generate the required data, we have surveyed the existing nuclear data libraries with residual product cross sections and tested a set of intra-nuclear cascade and de-excitation models. The Intra-Nuclear Cascade Liège (INCL++) and ABLA models [8,9] were selected for generation of a library that is the primary focus of this work.

2. HEIR data

The current High-Energy Intra-Nuclear Cascade Liège-based Residual (HEIR-0.1) Library has been generated using the `Hadr03` hadronic physics case of Geant4 version 10.3 with the `QGSP_INCLXX` physics list coupled to the in-built ABLA V3 (10.3) [10]. This selection was based upon consideration of comparisons between model predictions with full mass and isotope distributions of cross sections from the IAEA spallation benchmark studies [11]. Results are post-processed using bespoke scripts and the PREPRO-2017 code [12] is used to create fully-compliant ENDF-6 files using the `MF=3` and `MF=10` file formats with the 'other' `MT=5` reaction channel. This provides total production cross sections, *i.e.* for each product P, the HEIR-0.1 value is the sum over all channels:

$$\sigma(P) = \sum_{c \in \{channels\}} \sigma(c) N(P, c), \tag{1}$$

where $N(P, c)$ is the number of product P atoms made in that reaction channel. For example, the neutron production cross section in the library is:

$$\sigma(n) = \sigma(p, n) + 2\sigma(p, 2n) + 3\sigma(p, 3n) + \cdots + \sigma(p, np) + \cdots \tag{2}$$

[1] Databases are relatively easy to interrogate and compare, whilst models require event generators, convergence tests and post-processing to compare with experiment. This also requires non-negligible computer resources, whereas database comparison can occur with seconds of single computer core time.

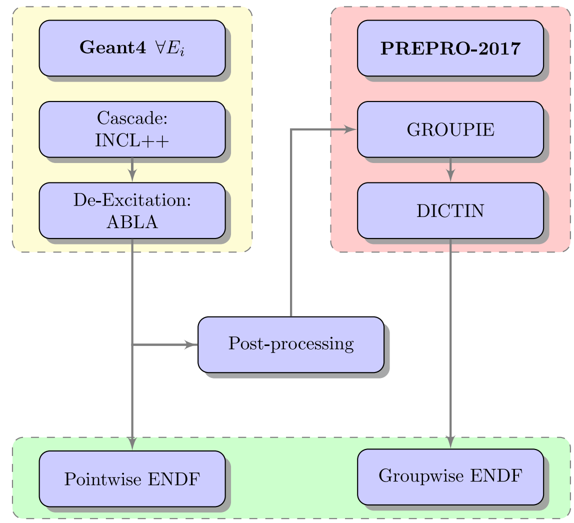


Fig. 1. Schematic for HEIR data generation, using Monte-Carlo Geant4 simulations for each incident particle energy E_i (yellow), post processing and PREPRO-2017 library processing (red) to generate ENDF nuclear data files (green). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

`GROUPIE` is used to generate 162-group[2] cross section data for use with FISPACT-II and `DICTIN` to document the file contents. This is shown schematically in Fig. 1, where each Geant4 calculation produces one set of cross sections for a given incident particle energy.

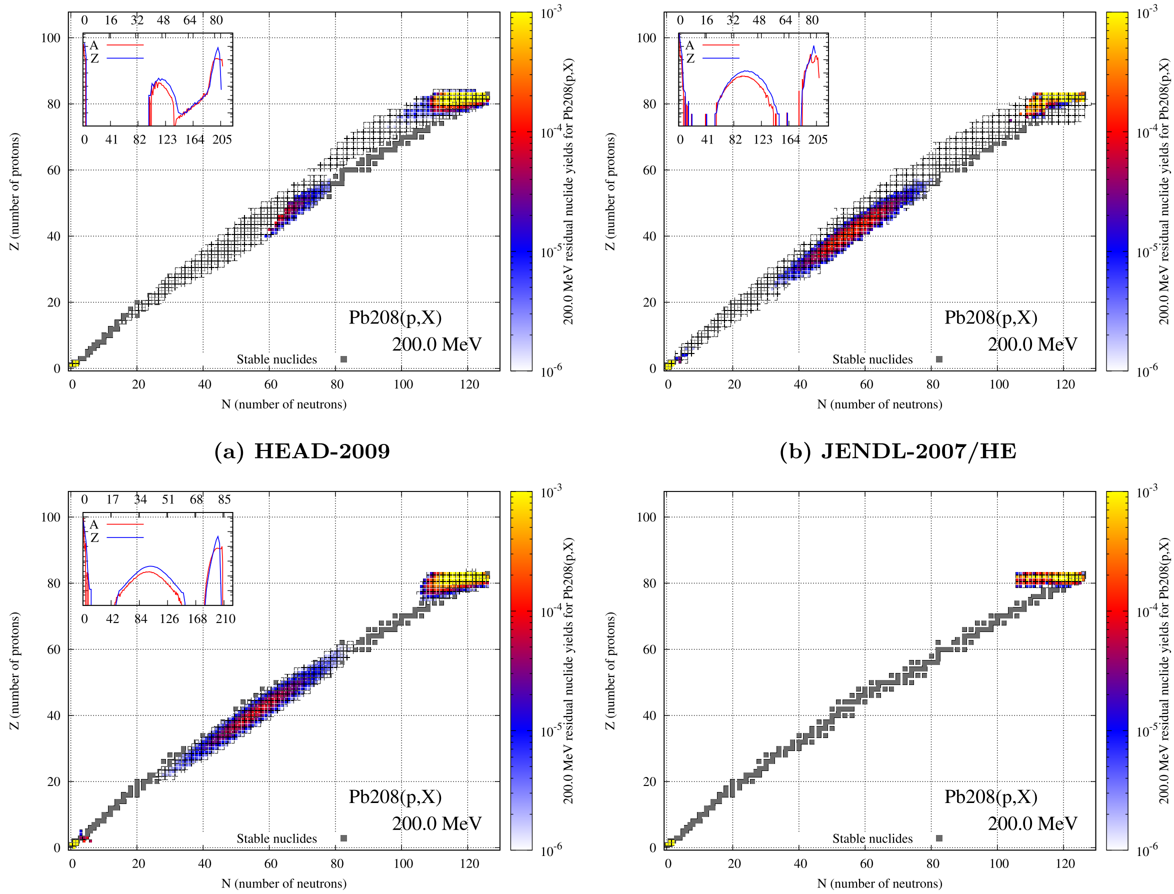
The results contained in the library are based on $\mathbf{10^6}$ events per target per incident energy for **stable isotopes** (250 targets), and $\mathbf{10^5}$ events per target per incident energy for **radioactive isotopes** up to ^{251}Bk with a half-life longer than 1 s (1845 targets). To limit the impact of statistical fluctuations due to the Monte-Carlo nature of the Geant4 calculations, a minimum of 10 events are required for the results to be retained and included in the HEIR data files.

3. High-energy 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 with energies up to 1 GeV, known to the authors, are the JENDL-2007/HE [13] and HEAD-2009 [14] 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. It should be noted that JENDL-2007/HE is not designed for activation–transmutation calculations, hence the restriction to 106 isotopes.

While the JENDL-2007/HE files only cover 106 targets, they cover more product nuclides. The mixed choice between these two libraries

[2] This is taken from the FISPACT-II 162 group structure [4].



(a) **HEAD-2009** (b) **JENDL-2007/HE**

(c) **HEIR-0.1** (d) **TENDL-2017**

Fig. 2. Comparison of the residual product cross sections (barns) from a 200 MeV incident proton within the four considered nuclear data libraries; (a) HEAD-2009, (b) JENDL-2007/HE, (c) HEIR-0.1 [this work] and (d) TENDL-2017. Mass and charge distributions are shown in the top-left of each subfigure and values are in units of barns. Note that TENDL does not contain fission products and is limited to a bounded region of mass and charge loss.

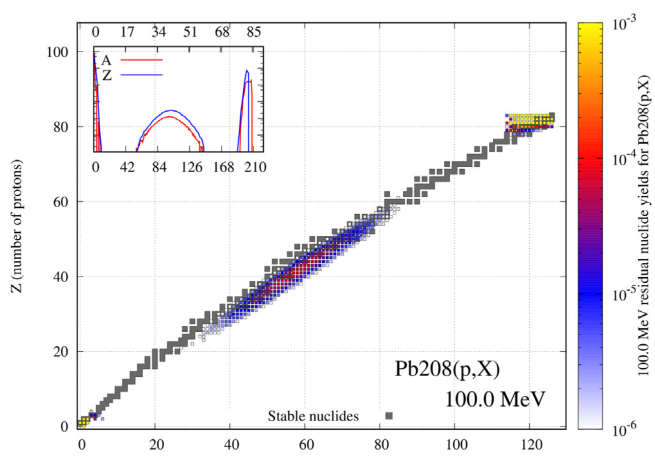


Fig. 3. Energy-dependent residual product cross sections for incident protons on Pb208 within the HEIR-0.1 nuclear data library. The energy range shown is between 100 MeV and 1 GeV. Mass and charge distributions are shown in the top-left subfigure.

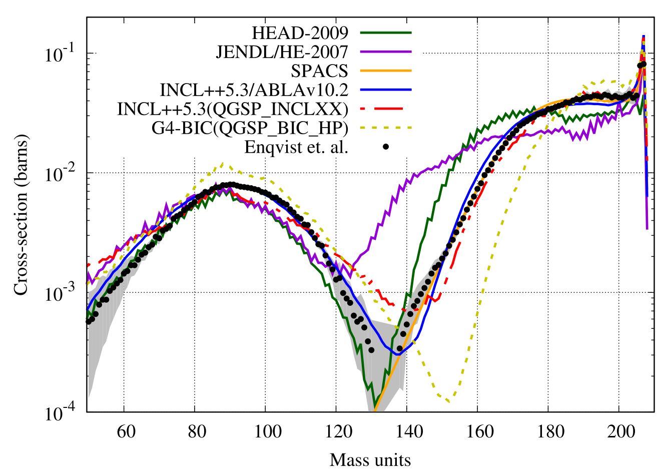
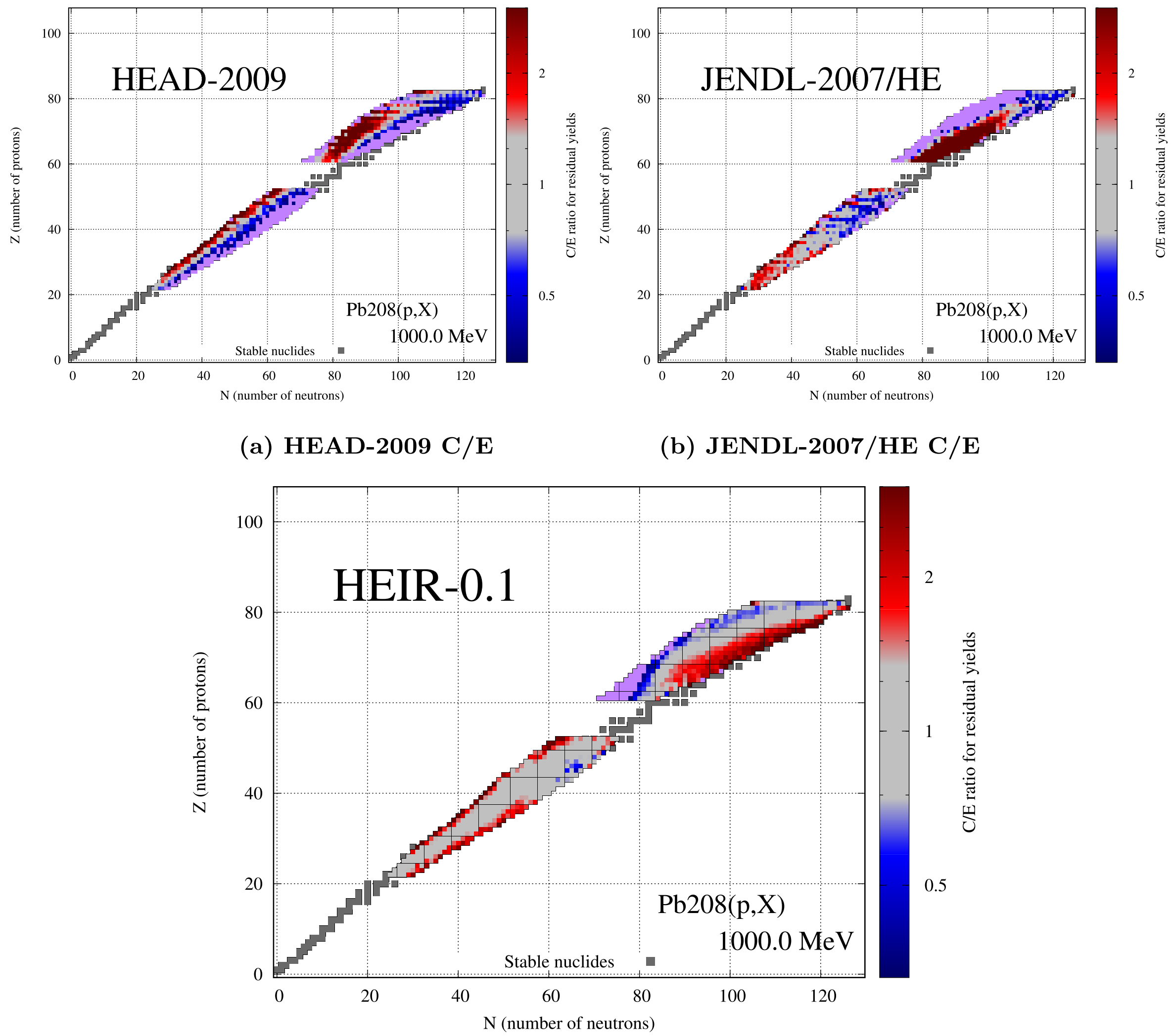


Fig. 4. Comparison of nuclear data libraries, SPACS and intra-nuclear cascade model results for 1 GeV proton-induced Pb208 residual product cross sections, compared with experimental data from Enqvist et al. [15] with experimental uncertainties in grey. Geant4 simulations were used with multiple physics lists and using an ABLA [9] optional de-excitation mode coupled with INCL++ [8].

are between target coverage and product coverage. However, 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 have follow-on reactions. To accommodate a true inventory calculation for high-energy reactions, a complete set of reactions are required, including the radionuclide products.



(a) **HEAD-2009 C/E**

(b) **JENDL-2007/HE C/E**

(c) **HEIR-0.1 C/E**

Fig. 5. Comparison against EXFOR data of Enqvist et al. [15] of the residual product cross sections from a 1 GeV incident proton within the three considered nuclear data libraries; (a) HEAD-2009, (b) JENDL-2007/HE, and (c) HEIR-0.1 [this work]. Ratios are provided for all documented experimental nuclide product values with a logarithmic scale. Values shown as violet are absent in the nuclear data files. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

To compare between all of these data libraries, the 200 MeV incident proton residual cross sections are shown in Fig. 2. These demonstrate the various limitations of each library, notably the limited set of products in TENDL-2017, inconsistent fission treatment and lack of fragmentation for HEAD-2009 and nonphysical discontinuities in JENDL-2007/HE. The full contents of the HEIR-0.1 Pb208 file are shown in Fig. 3.

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 results from some of the standard physics list options within Geant4 are in much better agreement with experimental data, as shown in Fig. 4. The isotopic distributions are shown in Fig. 5, where violet entries identify measurements for which the library lacks any data. While this occurs for all libraries, with HEIR-0.1 it occurs exclusively for very neutron-poor isotopes near the end of the spallation 'tail', due to the artificial boundary set at $Z = 60$. HEIR possesses data for each of the associated masses, but these particular isotopes are beyond the statistical reach of the current HEIR library — less than 10 events (on average) per million incident particle simulations.

To generate the full tabulated data required for all targets in activation simulations, the semi-empirical isotopic spallation simulation code SPACS [16] was also utilised to build complete isotopic yield data files for incident neutrons and protons with energies from 100 MeV to a few GeV [17]. These can be read directly into FISPACT-II and used with the complete nuclear reaction data of TENDL to provide a consistent simulation framework. This includes high-energy activation residual products down to approximately half of the target mass, but lacks methods to predict residual products due to fission and fragmentation processes.

Of particular interest, given the radiological importance of the long-lived α-emitter ^{148}Gd, is the mass yield for 148. In this case, the 11 mb JENDL-2007/HE and 5.6 mb HEAD-2009 values are significantly larger than the 1.2 mb HEIR-0.1 value, which is within 3 σ of the 1.6 mb value from Enqvist et al. [15]. 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 Fig. 4 demonstrates, INCL-based calculations provide significant improvement in the calculation of this mass yield, as well as many others.

HEIR-0.1 does not provide superior agreement for all targets, incident energies and calculated values, as demonstrated in Fig. 6. While the most probable mass yields are in better agreement with those

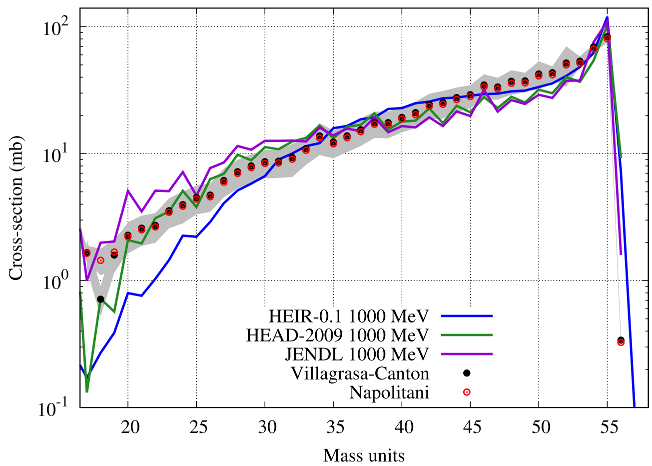


Fig. 6. Comparison of nuclear data libraries for 1 GeV proton-induced Fe56 residual product cross sections, compared with experimental data from Villagrasa-Canton et al. [19] and Napolitani et al. [20], with experimental uncertainties in grey.

measurements, the yields for reactions with the smallest measured masses show under-prediction that is consistent across all incident energies between 300 MeV and 1 GeV. Model improvements are still required and the implementation of more sophisticated de-excitation models [18] should offer more accurate simulations for future library versions.

4. Simulations with FISPACT-II

While in some simulation systems (c.f. [21,22]) 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 Fig. 7. The 'low-energy' nuclear data referenced in Fig. 7 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[3] 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;

$$\bar{\sigma} = \frac{\left[\sum_{\{i|E_i<E_{cut}\}} \phi_i\, \sigma_{LE,i}\right] + \left[\sum_{\{j|E_j\geq E_{cut}\}} \phi_j\, \sigma_{HE,j}\right]}{\sum_k \phi_k}, \tag{3}$$

where ϕ and σ represent the particle flux and cross sections, and the low- and high-energy library cross sections are subscripted as *LE* and *HE*, respectively.

An example simulation is shown in Fig. 8, where a 1 year, 10^{13} protons cm^{-2} s^{-1}, 1 GeV proton irradiation on natural lead has been calculated. The post-irradiation activity is shown, where a significant difference occurs between a few years and a few decades. During this time period, tritium activity[4] is the dominant contribution for all

[3] As of `FISPACT-II` release 4.0.

[4] For this example we consider total activity generated, irrespective of mobility. Tritium production remains an important topic, regardless.

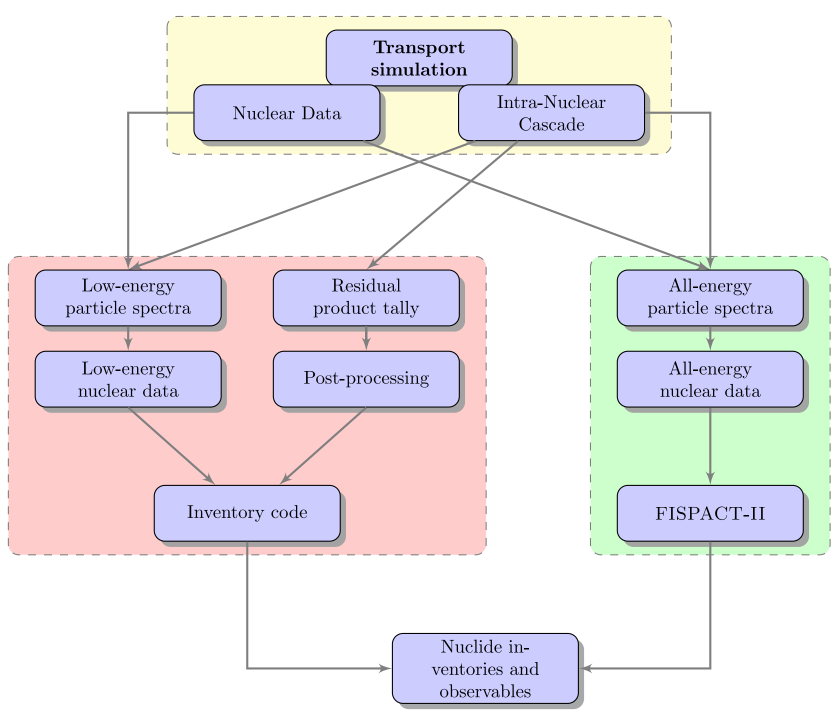


Fig. 7. Schematic for activation–transmutation inventory calculations, using a Monte-Carlo transport simulation (yellow), coupled with either (red) separate nuclear data and residual history data or (green) pure nuclear data library calculations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

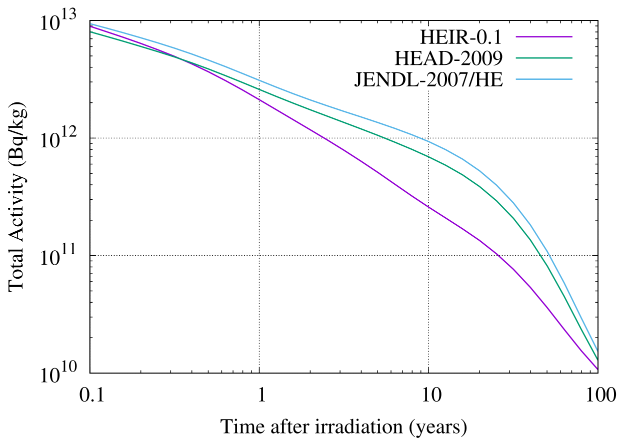


Fig. 8. Comparison of post-irradiation activity for a 1 year, 10^{13} proton per square cm per second, 1 GeV proton irradiation on natural lead, using JENDL-2007/HE, HEAD-2009 and HEIR-0.1.

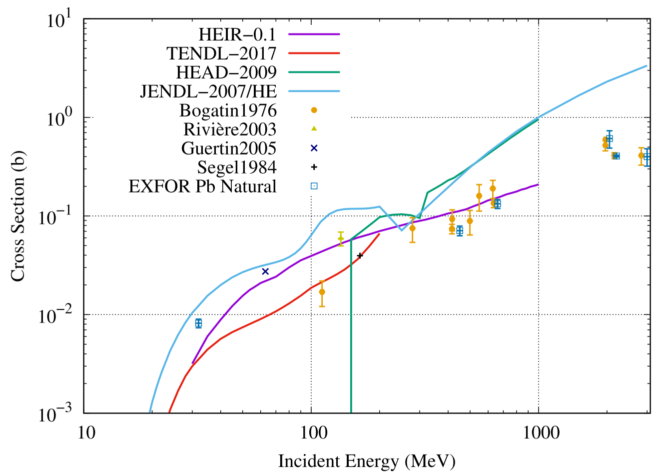


Fig. 9. Comparison total tritium production for all nuclear data libraries and EXFOR data, including the Pb208 measurements of Bogatin, et al. [23], Guertin et al. [24], Segel et al. [25], Rivière [26], and all natural lead data.

simulations, although it varies by more than a factor of five between HEIR-0.1 and the others. Examining the EXFOR entries for total tritium production on lead, Fig. 9 shows that there is a clear over-prediction for HEAD-2009 and JENDL-2007/HE.

5. 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 several GeV. For high-energy activation–transmutation calculations, this offers the significant advantage of using converged, reproducible residual nuclide production cross sections. In comparison with the requirement to converge spallation residual tallies for each region of a Monte-Carlo model calculation, only the incident particle spectra are required. The use of a pre-converged nuclear data library then effectively multiplies the statistical quality of the results by the sampling of the library (1 million per incident energy for stable isotopes in HEIR-0.1) divided by the number of events in the Monte-Carlo calculation, for each region, energy and target isotope.

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 intra-nuclear cascade and de-excitation models has been demonstrated with comparisons using the most recent models as available in Geant4, including the Intra-Nuclear Cascade Liège (INCL) and ABLA models.

A new nuclear data library, the High-Energy INCL-based Residual (HEIR-0.1) library includes proton-induced cross sections up to 1 GeV for 2095 targets. It is based on the most recent INCL++ and ABLA models available within the Geant4 simulation system, providing a reproducible and traceable library for all target isotopes and with all products that can be practically simulated.

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 [27,1]. By automating this process, new libraries can be generated as and when models are improved, for example with the new ABLA++ within Geant4 [18].

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