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Technical note

Generation of low-energy neutrons cross-sections for the Monte Carlo code FLUKA and the deterministic code ActiWiz

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ABSTRACT

Activation of material is of interest for waste treatment and hazard assessment. In particular, activation of printed circuit can lead to the production of radionuclides at an isomeric state, for example, coming from silver. In particle accelerators, the production of silver isomeric states mainly come from low energy neutrons, below 20 MeV. The quantification of activation and associated doses at CERN is based on the FLUKA and ActiWiz codes. In the FLUKA release 2011.2c, all branching ratios for isomer production were set at 50% by default. The present work provides a set of nuclide- and energy-dependent branching ratios, extracted from the library EAF-2010. In the ActiWiz release 3.3, the library JEFF3.1.1 was used for low energy neutron cross-sections. This study provides a new set of neutron cross-sections extracted from JEFF3.3, ENDFB/VIII.0 and EAF-2010 for future update of ActiWiz.

1. Introduction

Radionuclides produced via nuclear interactions are of strong interest for radiation protection in nuclear power plants and particle accelerators, because they are responsible for the phenomenon of induced radioactivity and for the associated radiological risks.

Estimates of induced radioactivity can be performed with Monte Carlo codes like FLUKA (Battistoni et al., 2015; Ferrari et al., 2005) and analytical codes like ActiWiz (Theis and Vincke, 2018). These codes rely on cross-sections for the computation of reaction rates, at least in the case of low energy neutrons as incident particles. Cross-sections are typically extracted from different nuclear libraries, like for-example EAF-2010 (The European Activation F, 2010).

FLUKA is a Monte-Carlo calculation code that simulates particle transport and interaction with matter. It includes about 60 different particles and can be used for different purposes such as shielding, design, activation, radiotherapy. ActiWiz has been developed to compare and assess radiological hazards for materials used in the partcile accelerators. It allows the computation of radionuclide inventories and associated radiological hazard according to irradiation scenarios.

The first part of this paper gives an overview of the ENDF-6 format (ENDF, 2010). which is widely used to store information in the nuclear libraries. We then describe the process used to extract cross-sections and relevant data from the libraries, and perform folding with neutron spectra in order to obtain reaction rates. Finally, we present the cases of FLUKA and ActiWiz as examples of application.

2. The ENDF-6 format

The Nuclear Energy Agency NEA[1] provides nuclear data libraries in the form of files in a readable format, typically ENDF-6 (ENDF, 2010). These libraries include values from experimental results, as well as values estimated via nuclear models. For every radionuclide of interest, the information is presented in the form of data blocks called "files" (MF), with a special identifier for each reaction type (MT).

2.1. Information on radionuclide production and reaction types

The most significant information for radionuclide production can be found in the ENDF files labelled MF2, MF3, MF9 and MF10.

MF2 provides information in the resonance domain, where cross-sections are characterized by peak values corresponding to the quantum excitation states of the nuclei.

MF3 gives the cross-sections as a function of energy. In this file, one can also find descriptive data or resonance parameters which are not of interest for this work.

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[1] https://www.oecd-nea.org/dbdata/.

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Table 1
Reactions types and their MT label.

MT Number	Reaction Type	MT Number	Reaction Type	MT Number	Reaction Type
4	(n,n')	111	(n,2p)	173	(n,4nt)
11	(n,2nd)	112	(n,palpha)	174	(n,5nt)
16	(n,2n)	113	(n,t2alpha)	175	(n,6nt)
17	(n,3n)	114	(n,d2alpha)	176	(n,2nHe3)
22	(n,nalpha)	115	(n,pd)	177	(n,3nHe3)
23	(n,n'3alpha)	116	(n,pt)	178	(n,4nHe3)
24	(n,2nalpha)	117	(n,dalpha)	179	(n,3n2p)
25	(n,3nalpha)	152	(n,5n)	180	(n,3n2alpha)
28	(n,np)	153	(n,6n)	181	(n,3npalpha)
29	(n,n2alpha)	154	(n,2nt)	182	(n,dt)
30	(n,2n2alpha)	155	(n,talpha)	183	(n,n'pd)
32	(n,nd)	156	(n,4np)	184	(n,n'pt)
33	(n,nt)	157	(n,3rd)	185	(n,n'dt)
34	(n,nHe3)	158	(n,n'dalpha)	186	(n,n'pHe3)
35	(n, n'd2alpha)	159	(n,2npalpha)	187	(n,n'dHe3)
36	(n, n't2alpha)	160	(n,7n)	188	(n,n'tHe3)
37	(n,4n)	161	(n,8n)	189	(n,n'talpha)
41	(n,2np)	162	(n,5np)	190	(n,2n2p)
42	(n,3np)	163	(n,6np)	191	(n,pHe3)
44	(n,n'2p)	164	(n,7np)	192	(n,dHe3)
45	(n,n'palpha)	165	(n,4nalpha)	193	(n, alphaHe3)
102	(n,g)	166	(n,5nalpha)	194	(n,4n2p)
103	(n,p)	167	(n,6nalpha)	195	(n,4n2alpha)
104	(n,d)	168	(n,7nalpha)	196	(n,4npalpha)
105	(n,t)	169	(n,4th)	197	(n,3p)
106	(n,nHe3)	170	(n,5th)	198	(n,n'3p)
107	(n,alpha)	171	(n,6th)	199	(n,3n2palpha)
108	(n,2alpha)	172	(n,3nt)	200	(n,5n2p)
109	(n,3alpha)				

A nuclear reaction can produce a radionuclide in the ground or in an excited nuclear state (isomer). If isomers can be produced, the associated information can be found in the files MF9 or MF10.

MF9 tabulates the multiplicities for production of radionuclide. Multiplicities represent the fraction of the reaction cross-section specified in MF3, which produces the Level Final State (LFS) of the residual. These values are given as a function of energy but do not have the same mesh as in MF3. A linear interpolation (as recommended by the interpolation scheme for multiplicities) is then performed to get the multiplicity at the same energy as in the reaction cross-section. Finally, the reaction cross-section $\sigma(E)$ from MF3 is multiplied by the multiplicity of the LSF $\mu_{LFS}(E)$ from MF9 to get the specific cross-section of LFS (Equation (1)).

$$\sigma_{LFS}(E) = \sigma(E)^{*}\mu_{LFS}(E) \quad (1)$$

Equation 1. Cross-section of LFS isomer calculated with multiplicities from MF9 and total cross-section from MF3.

MF10 contains directly the specific cross-section of the LFS, $\sigma_{LFS}(E)$. In principle, the information in MF10 would be redundant with respect to the one in MF9 (once complemented with MF3). However, in practice the information is either provided in MF9 or in MF10, but never in both files at the same time.

Table 1 shows the MT labels used in ENDF-6 to identify reaction types.

2.2. Nuclear data libraries

The present study covers 3 different nuclear data libraries: JEFF3.3,[2] ENDFBVIII.0 (ENDF/B-VIII.0, 2018) and EAF-2010 (The European Activation F, 2010).

[2] http://www.oecd-nea.org/dbdata/jeff/jeff33/.

Table 2
Comparison between different nuclear data libraries.

	EAF	JEFF	ENDF
Number of target nuclides	816	562	557
Number of reactions	~66 000	~31 000	~28 000

JEFF3.3 and ENDFBVIII.0 are the latest evaluations from Europe and the United States, respectively. EAF-2010 is a European library which reports activation reactions from different evaluations produced worldwide (Table 2). EAF-2010 is the most complete library of the three in terms of reaction type with a total of 66,256 reactions, including for example rare reactions like (n,8n) or (n,4n^{3}He). Most of EAF-2010 reactions come from TALYS-TENDL-2009.

3. Computation of reaction rates

The cross-sections are used to compute the reaction rate $\theta_{k\leftarrow m}$ of a product nuclide k from a target nuclide m as expressed in Equation (2).

$$\theta_{k\leftarrow m} = \sum_{q} \int_{E} \sigma_{q,k\leftarrow m}(E)\Phi(E)dE \quad (2)$$

Equation (2). Reaction rate for the formation of the product nuclide k.

In particular, $\Phi(E)$ is the neutron flux at energy E, and $\sigma_{q,k\leftarrow m}(E)$ is the microscopic cross-section of the reaction q at energy E producing the nuclide k from target nuclide m.

In the typical case of a radionuclide with a relatively high mass number, the absorption cross-section over the energy range of incident neutrons usually shows three distinct domains (Fig. 1):

- Low energy domain, with the so-called 1/v region. In this domain, the cross-section is the proportional-inverse of the velocity v;
- Resonance domain, with characteristic resonance peaks. In this domain, cross-section varies deeply to high value and decreases again. These resonance peaks are due to the quantum energy levels of the nucleus;
- Fast neutrons domain.

The information on the resonance domain is provided in MF2. The NEA website provides the tool PREPRO (PREPRO, 2015), which contains different codes that are designed to process specific aspects of nuclear data, from cross-section linearization to consistency checks and Doppler Broadening.

The PREPRO codes shall be used in the following sequence:

1) ENDF2C, which transforms the ENDF format into a FORTRAN/C++ readable format;
2) LINEAR, which linearizes cross-sections following the interpolations laws described in ENDF format. The result is a cross-section that can be linearly interpolated;
3) RECENT, which constructs the contribution of resonances to the cross-section. Reich-Moore theory and the formalism of R matrix are used for this purpose;
4) SIGMA1, which allows to take into account the Doppler Broadening at a temperature T defined by the user;
5) ACTIVATE, which combines neutron cross-sections from MF3 with the multiplicities of MF9 to produce MF10;
6) FIXUP, which among other corrections ensures that the total cross-section is equal to the sum of the cross-sections for the different reaction types.

The actual solution of Equation (2) may require multi-group approximation depending on the field of application. In the next section we provide two examples of applications at CERN.

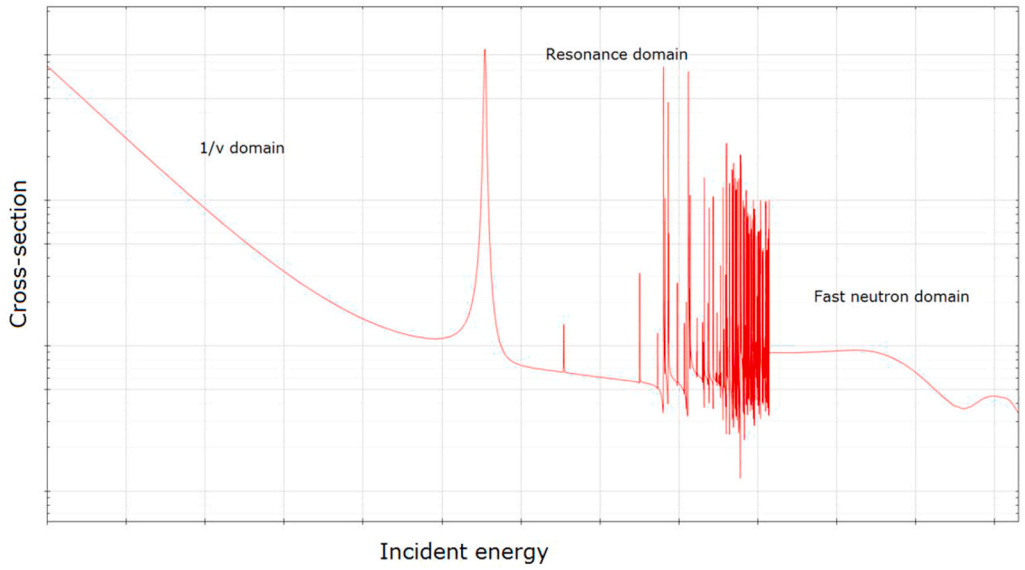

Fig. 1. Cross-section domains over energies of incident neutrons (from Janis4.0 https://www.oecd-nea.org/janis/).

4. Examples of application

4.1. Generation of branching ratios for isomer production in FLUKA

As a first example of application, we here describe how we generated the branching ratios between isomeric state and ground state at a given temperature for a given energy mesh, in the frame of the development of the Monte Carlo code FLUKA. More specifically, the generation of branching ratios for isomers in FLUKA covers the reaction types up to MT117 (Table 1) from the library EAF-2010, for all isomeric states tabulated in MF10.

The branching ratio will depend on the temperature of the media containing the nuclide, the excitation energy of the isomeric state, and the energy group number of the output.

The temperature is used for two purposes:

To take into account Doppler Broadening generated from PREPRO;

The generation of a reference neutron flux for thermal neutrons, following a specific distribution.

FLUKA uses 260 groups of low-energy neutrons up to 20 MeV. In order to create a correspondence between energy-dependent cross-sections in EAF-2010 and group-dependent fluxes in FLUKA, each of the 260 groups i is divided into 100 bins j with an average energy $\overline{E}_j = \sqrt{E_j E_{j+1}}$. The neutron flux inside a group i is then approximated with a weight function $w(\overline{E}_j)$ according to Equation (3). If the minimum energy of the FLUKA group is below $5kT$ where k is Boltzmann constant and T the temperature at which cross-sections are computed, then we assume a Maxwell distribution for the weight function. Otherwise, we assume an inverse distribution in energy.

$$\begin{cases} w_j = \dfrac{\overline{E}_j}{kT} e^{-\frac{\overline{E}_j}{kT}} \; if \; E_j < 5kT \\ w_j = \dfrac{1}{\overline{E}_j} \quad otherwise \end{cases} \tag{3}$$

Equation (3). Weight function, approximating the neutron flux.

The microscopic reaction rate $\widehat{\sigma}_{q,k\leftarrow m,i}$ in the group i for the reaction q producing radionuclide k from target m is then calculated with the multigroup approximation formalism (Equation (4)). It is the sum of the cross-sections at average energy $\overline{E}_j$ in subgroup j weighted by the flux distribution w_j.

$$\widehat{\sigma}_{q,k\leftarrow m,i} = \frac{\sum_{j\in sub(i)} w_j \sigma_{q,k\leftarrow m}\left(\overline{E}_j\right)\left(E_{j+1} - E_j\right)}{\sum_{j\in sub(i)} w_j \left(E_{j+1} - E_j\right)}, i \in [1, 260] \tag{4}$$

Equation (4). Multigroup approximation formalism for cross-section in the group i.

The branching ratio $\widehat{BR}_{q,k\leftarrow m,i}$ in the group i for the reaction q producing radionuclide k from target m is then computed following Equation (5).

$$\widehat{BR}_{q,k\leftarrow m,i} = \frac{\widehat{\sigma}_{q,k\leftarrow m,i}}{\sum_j \widehat{\sigma}_{q,j\leftarrow m,i}} \tag{5}$$

Equation (5). Branching ratio in the group i.

In the nuclear database used in FLUKA, isomeric states are identified via their excitation energies. In order to match isomeric states in FLUKA with the ones in the library EAF-2010, we calculated the excitation energy of each isomer from the file MF10.

Table 3
Comparison of excitation energies between FLUKA and EAF-2010.

			FLUKA		EAF-2010 (MF10)		Relative difference (%)	Absolute difference (MeV)
Symbol	A	Z	LFS	E (MeV)	LFS	E (MeV)		
Mn	50	25	1	0.23	1	0.23	***1.65%***	3.72E-03
Mn	52	25	1	0.38	1	0.38	0.00%	1.00E-06
Fe	53	26	1	3.04	1	3.04	0.00%	3.00E-05
In	120	49	1	0.00	1	0.00		4.00E-05
In	120	49	2	0.07	2	0.00	***97.09%***	***6.80E-02***
Sn	120	50	1	2.48	–	–		
Sn	120	50	2	2.90	–	–		
I	120	53	–	–	1	0.90		

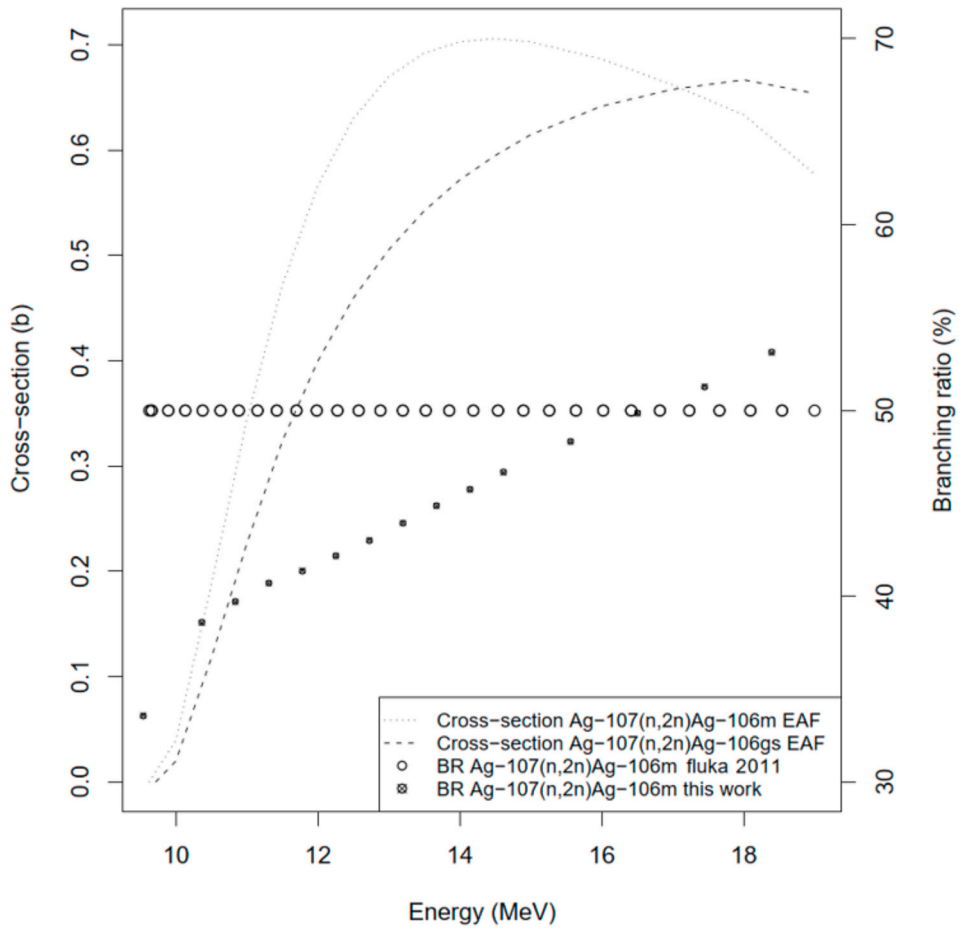

Fig. 2. Example of isomer branching ratios produced for FLUKA in the case of Ag-107 (n,2n)Ag-106m reaction. Cross-sections are taken from EAF-2010. Acronyms stand for ground state ("gs") and branching ratio ("BR").

Automated matching among isomers is straightforward for the vast majority of cases, but there a few exceptions (Table 3) which required manual intervention.

For example, Sn-120-LFS1 and Sn-120-LFS2 exist in the 2011 FLUKA database (with 50% branching ratio) but not in EAF-2010. Conversely, I-120 is present in EAF-2010 but not in the FLUKA database. As a last example, Mn-50-LFS1 and In-120-LFS2 show significant differences in terms of relative or absolute values.

An example of branching ratios is presented in Fig. 2. From the cross-sections for the ground state and the isomeric state of Ag-108 we calculated the associated branching ratios, which are then compared with the default value of 50% of the 2011 FLUKA version. The difference between default and recommended branching ratios can be in the order of a factor of two in this specific case.

4.2. Generation of cross-sections for ActiWiz

The neutron cross-sections of the current version of ActiWiz are based on the JEFF3.1.1 library. The purpose of this work is to upgrade and extend such cross-sections by extracting data from JEFF3.3, ENDFBVIII.0 and EAF-2010.

To generate the new set of cross-sections, we start from the list of target and product nuclides which are currently implemented in ActiWiz, including all possible isomeric states. For these nuclides we then produce the cross-sections per reaction, per energy and per element with the same energy mesh as in the nuclear data library. The study performed for ActiWiz covers all the reaction types given in Table 1.

The cross sections are extracted and then converted to 260 groups as described in paragraph 3.2, according to Equation (3) and Equation (4).

Then the macroscopic cross-sections $\widehat{\sigma}_{k\leftarrow compound,i}$ in the energy group i for a specific compound is calculated by summing all reactions q from target nuclides j of this compound leading to the creation of a product nuclide k. Target nuclides of the compound are weighted by their abundances[3] A_j.

$$\widehat{\sigma}_{k\leftarrow compound,i} = \sum_{j\in compound} \sum_{q} A_j \widehat{\sigma}_{q,k\leftarrow j,i} \tag{6}$$

Equation (6). Macroscopic cross-section of a compound.

As shown in Table 2, JEFF3.3/ENDFBVII.0 cover fewer reactions than EAF-2010. We decided to use the nuclear library with the following order of priority: JEFF3.3, ENDFBVIII.0 and EAF-2010. Whenever applicable, for a given target and product nuclides we give priority to the library with the highest number of reactions or isomers available.

As an example, we preferred ENDFBVIII.0 over JEFF3.3 for the carbon element, because product nuclides are discriminated by mass number only in ENDFBVIII.0.

More generally, we preferred JEFF3.3 over ENDFBVIII.0 and EAF-2010 for reason of coherences. JEFF3.3 is the standard library in Europe for power plants and seemed the logical choice, in particular, as previous decay library implemented in ActiWiz is JEFF3.1.1.

The isomeric states names are implemented sequentially when they are found in the library. The first LFS found will become the first metastable state (with suffix "-m"), and the second one will be the

[3] Abundances taken from NIST: https://www.nist.gov/pml/atomic-weights-and-isotopic-compositions-relative-atomic-masses.

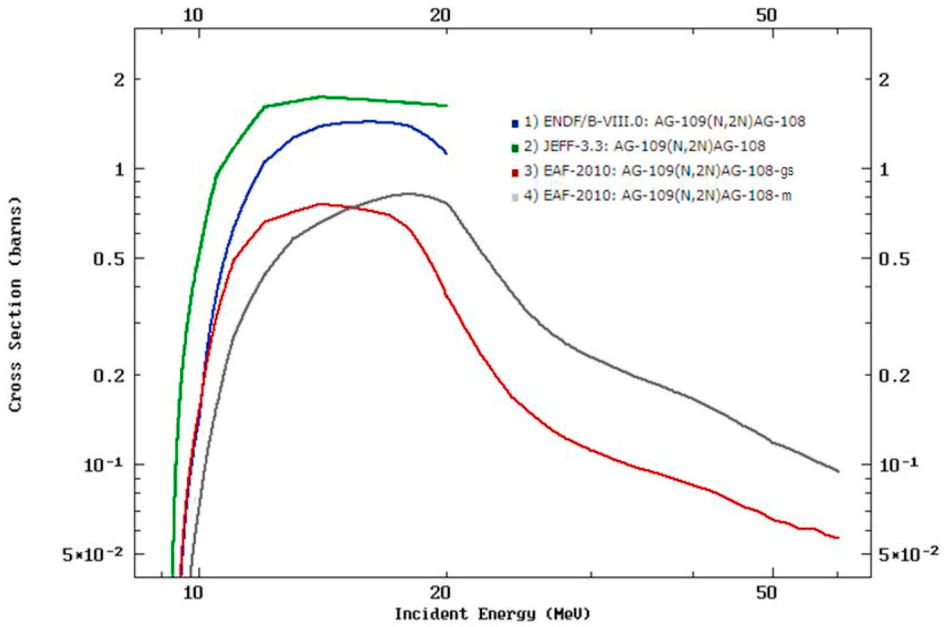

Fig. 3. Ag-109 (n,2n)Ag-108 reaction for ENDFBVIII.0, JEFF3.3 and EAF2010 (from EXFOR https://www-nds.iaea.org/exfor/endf.htm).

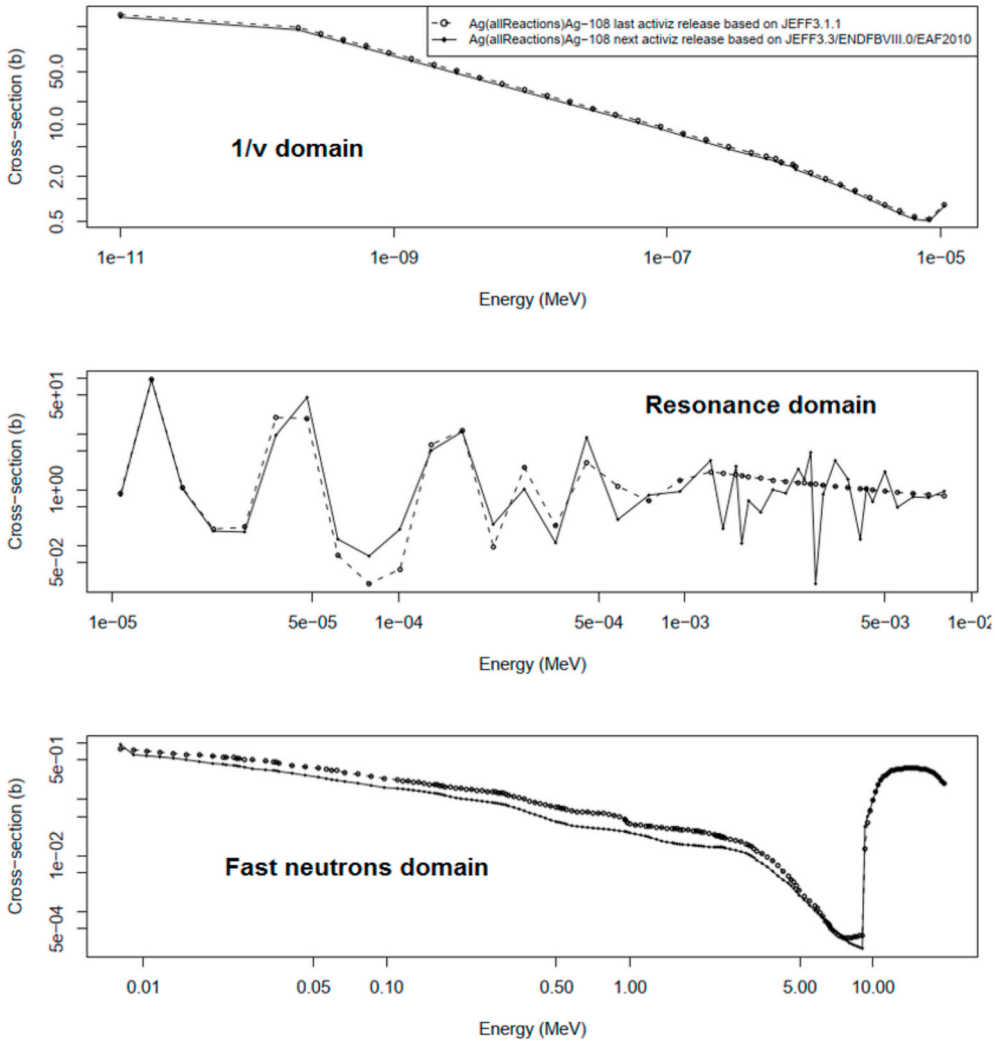

Fig. 4. Example of evolution of cross-sections with the new set. Application to target material: Silver, product nuclide: Ag-108 ground state.

second metastable state ("-n"). For example, for Ag-108 the library JEFF does not provide any data for LFS = 1, but does provide values for LFS = 2. In this case, we consider LFS = 2 to be the first isomeric state "-m".

We will now take the case of Ag-108 to illustrate differences among nuclear libraries.

The libraries ENDFBVIII.0 and JEFF3.3 do not allow discrimination between Ag-108 and Ag-108m (Fig. 3). In addition, the library EAF-2010 is the only one that contains information on the reaction of Ag-109 (n,2n)Ag-108m, whereas ENDF and JEFF only include the production of Ag-108. We notice consistency between the total cross-sections given by ENDF and JEFF, and the sum of the two reaction-specific cross-sections provided by EAF-2010 as presented in Fig. 3.

Fig. 4 represents the previous and next cross-section set of Ag (all reactions)Ag-108 for ActiWiz. At low energy, one can see that both cross-sections are similar until 1e-05 MeV in the 1/v domain. Then differences appear in the resonance domain. They are a result of the evolution of JEFF3.1.1 to JEFF3.3. We also observe these discrepancies at the beginning of the fast neutron domai

5. Conclusions

We generated new sets of branching ratios for isomers and neutron cross-sections to update the ones currently used in FLUKA and ActiWiz. A python script tool was specifically written in order to produce these data automatically from any library in ENDF format.

This new data will be included in the next releases of FLUKA and ActiWiz. It is particularly of interest for material activation and radioprotection purposes in particle accelerators. In particular, the activation process of electronic components such as printed circuit leads to the creation of silver isomer (Ag-108m for example) which mainly comes from low energy neutrons.

Regarding isomers in FLUKA, we showed that replacing default values of 50% with recent data from the library EAF-2010 leads to considerable differences in the evaluation of activity. In the case of ActiWiz, the new set of cross-sections will improve the accuracy of production rates and include new isomeric states.

In a general way, for the range of temperatures found in accelerators complexes, temperature has relatively weak impact on Doppler Broadening as well as weight function to approximate a neutron fluence.

The identification of isomers with their respective LFS tabulated in the libraries can sometimes lead to mistakes of interpretations. We then recommend to identify the isomers with their excitation energy, calculated by the difference between Mass-difference Q value and reaction Q value.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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