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THE REAC-ECN-3 DATA LIBRARY WITH ACTIVATION AND TRANSMUTATION CROSS-SECTIONS FOR USE IN FUSION REACTOR TECHNOLOGY

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Abstract: This paper describes the work performed to revise and extend the REAC data file of Mann. The revisions were made by means of renormalizations of the cross-sections to experimental data at 14.5 MeV or to data from 14.5 MeV systematics. Uncertainty estimates are given for the systematics. Furthermore, a number of reactions has been added. The file essentially contains cross-sections for almost all stable and unstable nuclides with half-lifes exceeding 1 day. If a reaction can produce one or two isomers the cross-sections for producing the ground and isomeric states are given separately. In most cases these cross-sections were obtained by a simple scaling using isomer ratios at 14.5 MeV, based upon experimental data or recently developed systematics. For about 50 reactions leading to long-lived states a special treatment was followed including a detailed uncertainty analysis. The revised file is called REAC-ECN-3. A version with multi-group cross-sections has also been generated (GREAC-ECN-3).

(Neutron reactions, fast neutrons, cross-sections, evaluated and experimental data)

The data library REAC-ECN

In 1985 Mann /1/ prepared a data base called the REAC library. This file contains more than 6000 reactions and over 300 target nuclei. It contains evaluated data from the relatively recent ENDF/B-V library and other evaluations, however the bulk of the data is based upon calculations with the very simple code THRES /2/. Therefore the REAC file should be checked with experimental data or data from reliable systematics. In /3/ a first attempt is described to improve the REAC library by performing renormalizations to 14.5 MeV data and by other mofidifications and extensions (addition of unstable targets). The revised library was called REAC-ECN-1 (version 1986). The work described in the present paper is a further update of the REAC-ECN-1 library which was executed in two steps. Firstly by introducing revised systematics for cross-sections and isomer ratios (REAC-ECN-2) /4/, while in the second stage the work included a check and possible improvement of the cross-sections for the 51 important reactions leading to long-lived ground and isomeric states. Furthermore, six short-lived isotopes were added and some minor modifications were applied. The results are described in ref. /6/ and the corresponding updated data file is called REAC-ECN-3. This work has been performed in the framework of a cooperation with the Joint Research Centre at Ispra and AERE-Harwell.

Nuclear models for excitation functions

The THRES-F code calculates most reactions (except radiative capture) based on Pearlstein's empirical approach /2/ using simple nuclear models and systematics. The shape of excitation functions was found in reasonably good agreement with experimental data except for the (n,n'γ) reaction, for which the low energy part of the calculated shape overestimates the data.

The FISPRO-ECN code is based upon a relatively simple statistical-model to calculate (n,γ) cross-sections at energies above the resolved-resonance range, including a direct/semi-direct component important at energies above 10 MeV. For several (n,γ) cross-sections, regarded as important reactions, this model was applied. At low energies an 1/v-component normalized to the experimental 2200 m/s cross-sections was added or a full resolved resonance range was supplied if available.

Renormalizations at 14.5 MeV

Experimental data

For the experimental data our main source of 14.5 MeV data is still the extensive compilation of Qaim /5/. This compilation contains activation data and branching ratios for the reactions: (n,n'γ) to metastable states, (n,2n), (n,p), (n,α), (n,n'α) and (n,d)+(n,np). In addition to these references we have scanned some other compilations, e.g. for (n,t)+(n,nd), (n,He-3) etc. Furthermore, we have used many other references to renormalize the data, see /6/.

Systematics of cross-sections

For most nuclides with A > 20 the systematics of Forrest /6/ have been used. This author has studied various formulae for the systematics of σ(np), σ(nα), σ(nd)+σ(nnp), σ(nt)+σ(nnd) and σ(n³He). The new systematics gives improved χ^2-values and in addition the new analysis contains error estimates (standard deviations) which are expressed as

$\sigma^{+f\sigma}_{-\sigma/f}$ (see table 1).

Other cross-sections

The systematics for (n,n'γ) data is taken from Vonach /8/. The uncertainty factor was estimated to be about f = 1.5. Note that the results of THRES calculations are way below the systematics. The following systematics is proposed for 14.5 MeV (n,γ) cross-sections, based on the data from Ref. /9/:

$$\sigma_{n,\gamma} = 1.18 - 1.13 \exp [-0.01338\ A]\ \text{mb}.$$

A simplified error treatment has been applied and resulted in values of χ^2 = 3.0 and f = 1.5.

All other types of cross-sections were obtained from calculations with THRES. The (n,nt), (n,n³He) and (n,2p) reactions are as yet not renormalized to experimental data or data from systematics, but their cross-sections are very small.

Due to the importance of the (n,2n) and (n,3n) reactions at 14.5 MeV we have decided to inspect the calculated data in the following ways:

1. For the (n,2n) data a new systematic has been derived, fitting directly the (n,2n) data, and not the ratio of σ_{nN}/σ_{ne} as earlier. The latter approach has been used in the THRES-code and thus we felt that an independent treatment could be of importance. For the data-base the compilation of Qaim /5/ has been used. The previously established smooth dependence of $\sigma_{n,2n}$ on the asymmetry parameter s has been adopted and resulted in a satisfactory fit of the function:

$$\sigma_{n,2n} = 2.8 - 4.117 \exp[-7.403\ s]\ \ [b],$$

with $\chi^2 = 4.2$ and f = 1.2 for values of s ≥ 0.05. For smaller values of s, i.e. s < 0.05 (targets with A < 60), an exponential function of s has been fitted:

$$\sigma_{n,2n} = 1502.7\ s^{3.428}\ \ [b],$$

with $\chi^2 = 5.1$ and f = 3.0. The comparison of the calculated cross-sections with this systematic gave us the confidence that the THRES values are reasonably accurate.

2. For the (n,3n) reaction we have compared the available experimental information (generally not used for renormalization) with the calculations. This comparison showed that the calculation exceeds in general the experimental data with a factor upto 3 to 4. Because the excitation function is rather steep at 14.5 MeV, the calculated as well as the experimental values may be subject to strong fluctuations. The overestimation of these values in THRES is related to the underestimation of (n,n'γ) cross-sections. A revision of the neutron emission cross-section in THRES is probably necessary.

The available systematics allow us for the first time to assess uncertainty estimates almost to all evaluated cross-sections at 14.5 MeV, based on a quantitative treatment. The adopted values are listed in Table 1.

Table 1.
Recommended uncertainties f for the 14.5 MeV data

Reaction	Error treatment of systematics			Qualitative treatment based on experimental trends
(n,n'γ)	1.5	/8/	a)	
(n,2n)	1.2			
(n,3n)	-		b)	$3.0^{a)}$
(n,nα)	3.0			
(n,d) } (n,np)	2.0	/7/		
(n,t) } (n,nd)	1.6	/7/		
(n,nh)	-			-
(n,2p)	-			-
(n,γ)	1.5		a)	
(n,p)	1.5	/7/		
(n,h)	1.9	/7/		
(n,α)	1.6	/7/		

a) Present study.
b) Rough guess.

Systematics of isomer ratios

In the first version of the REAC-ECN file /3/ almost all isomeric branching ratios were set to a value of b=0.5. We have decided to improve this approach and to renormalize all isomer branching ratios based on recently developed systematics in case no experimental data are available. Vonach /8/ derived the systematic for the isomer (n,n'γ) cross-section at E_n = 14.5 MeV, while Kopecky and Gruppelaar /10/ examined the systematic behaviour of the isomer ratio in the (n,n'γ), (n,p), (n,t), (n,α) and (n,2n) reactions which is partly based upon model calculations with the code GNASH.

Single- and multi-particle emission reactions, except (n,n'γ), (n,t) and (n,γ).

The energetically possible neutron-induced reactions at 14.5 MeV have been divided according to their mechanism, namely as single- and multiparticle emission reactions. This categorization is based on the comparison of calculated and experimental branching ratios in ref. /10/, which for the (n,2n) reaction showed a behaviour different from that of the studied single-particle emission reactions. We have assumed that as a general feature the isomer ratios for the (n,2n) reaction are representative for all multi-particle emission reactions. This assumption is based on the proposition that the much lower residual energy left in the product nucleus is responsible for the different behaviour, and this is approximately common to all multi-particle emission reactions.

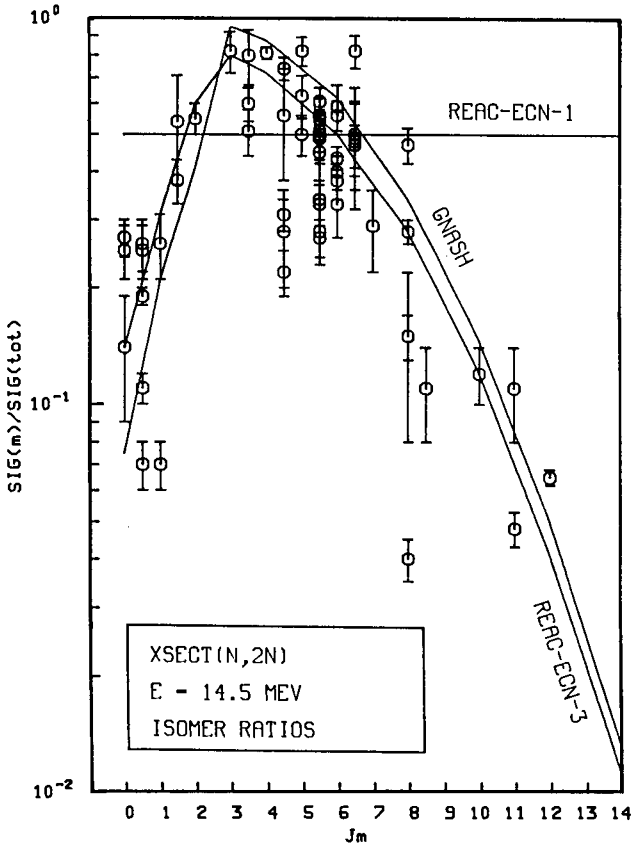

Fig. 1. Experimental isomeric cross-section ratios for the (n,2n) reaction based on the data library compiled in ref. /10/. The calculated prediction (GNASH) and the adjusted curve have been plotted.

The predicted isomer ratios have been constructed from the calculated curves and available data /10/, by shifting the curves to obtain optimum agreement with the mean values of data points. An example of such treatment is displayed in Fig. 1 for the (n,2n) reaction, which has the largest amount of data points. The recommended values of isomeric ratios have been stored in the form of tables /6/.
Based purely on the qualitative inspection of recommended values and experimental data the following uncertainties are proposed: for the (n,2n) reaction f_b = 1.6 and for the other reactions f_b = 2.0.

The (n,n'γ) reaction

For the high-spin isomers ($J_m > J_g$) the systematic derived by Vonach /8/ has been used. This systematic, however, does not apply to the branching ratio but predicts the absolute value of the isomeric cross-section σ_m(n,n'γ).

No systematic has been proposed in /8/ for low-spin isomers ($J_m < J_g$). We have thus adopted the recommended branching ratios and the expected constant total (n,n'γ) cross-section value which amounts to about 400 mb for most of the nuclei.

There is one observation to be made on the (n,n'γ) cross-sections calculated with THRES, as listed in the early REAC-ECN-1 file /3/. We have made a careful comparison between the calcuated and either the experimental or systematic entries.

Practically all calculated cross-sections were much lower, sometimes even by a factor 10 to 50. The reason for that is the way the inelastic scattering cross-section is derived in the THRES code. It is calculated as a difference between the total neutron emission cross-section $\sigma_{n,N}$ and cross-sections for (n,2n) and (n,3n) channels, and thus carries a substantial uncertainty typical for a subtraction of two large numbers.

The (n,t) reaction

The isomer ratio systematic proposed by Qaim [11] slightly deviates (is narrower) from the general trend of available data and calculations for the (n,n'γ), (n,p) and (n,α) reactions as studied in /10/.

Considering that the experimental data are preferential, we have kept the systematic of Qaim for (n,t) from the previous REAC-ECN-1 file /3/.

The (n,γ) reaction

In the absence of any systematic of isomer ratios for this reaction, the old approximative approach with b = 0.5 has been maintained in most cases.

Check and improvement of cross-sections for 51 important reactions

In this section we briefly describe the checks and improvements made for the 51 important reactions, listed in Table 2. They lead to longlived unstable isotopes or isomers, either directly or via intermediate steps.
The following methods were used:
- a search for possibly better evaluations in existing libraries ENDF/B-V, JENDL-2, JEF-1 or EFF-1 or from literature;
- a search of experimental data for the above-mentioned reactions from literature;
- an evaluation of isomeric branching ratios at 14.5 MeV from experimental data, from systematics, or both;
- if this was impossible, a new evaluation or calculation was made;
- renormalization of total cross-section mainly to fit the selected 14.5 MeV value, either from evaluated experimental data or from systematics;
- application of the evaluated isomer ratios at 14.5 MeV to obtain the cross-sections for production of ground and metastable states;
- application of a second isomer ratio in the thermal and resolved-resonance range for (n,γ) reactions, based upon experimental data at 0.0253 eV.

Table 2. Review of modifications on important reactions

Reaction	Shape	Renormalization to 14.5 MeV data
N - 14(n,p) C - 14	ENDF/B-5	RN to exp.
Al- 27(n,2n)Al- 26g,m	ENDF/B-5	RN to exp.
K - 39(n,p) Ar- 39	ACTL	RN to exp.
Ca- 42(n,α) Ar- 39	THRES	RN to syst.
Ca- 40(n,d) K - 39	THRES	No RN
Ca- 40(n,np)K - 39	ACTL	No RN
Ti- 48(n,α) Ca- 45	ENDF/B-5	RN to exp.
Ca- 45(n,α) Ar- 42	THRES	RN to syst.
Co- 59(n,γ) Co- 60g,m	ENDF/B-5	No RN
Ni- 60(n,p) Co- 60g,m	ENDF/B-5	RN to exp., b from syst.
Cu- 63(n,α) Co- 60g,m	ENDF/B-5	RN to exp.
Ni- 58(n,γ) Ni- 59	ENDF/B-5	No RN
Ni- 60(n,2n)Ni- 59	ENDF/B-5	RN to syst.
Cu- 63(n,p) Ni- 63	ENDF/B-5	RN to exp.
Nb- 93(n,γ) Nb- 94g,m	ENDF/B-5	b from syst.
Mo- 94(n,p) Nb- 94g,m	JENDL-2	RN to exp.
Mo- 95(n,np)Nb- 94g,m	THRES	RN to exp.
Mo- 95(n,d) Nb- 94g,m	THRES	RN to exp.
Mo- 94(n,2n)Mo- 93g,m	JENDL-2 *	RN to syst., b from exp.
Ag-107(n,γ) Ag-108g,m	ENDF/B-5	b from syst.
Ag-109(n,2n)Ag-108g,m	ACTL	RN to syst., b from exp.
Eu-151(n,2n)Eu-150g,m	ECN (TAT)	RN to exp.
Eu-151(n,γ) Eu-152g,m1,m2	ENDF/B-5	b from syst.
Eu-153(n,2n)Eu-152g,m1,m2	THRES	RN to exp.
Tb-159(n,2n)Tb-158g,m	THRES	RN to exp.
Gd-158(n,γ) Gd-159	ENDF/B-4	No RN
Gd-160(n,2n)Gd-159	ECN (BRC)	RN to exp.
Ho-165(n,γ) Ho-166g,m	ENDF/B-5	b from syst.
Ho-166(n,n'γ)Ho-166m	THRES	RN to syst.
Hf-178(n,n'γ)Hf-178m1,m2	JENDL-2 *	RN to syst., b from syst.
Hf-179(n,2n)Hf-178g,m1,m2	ACTL	RN to syst., b from exp. and syst.
Hf-177(n,γ) Hf-178g,m1,m2	JENDL-2	b from syst.
Ta-181(n,t) Hf-179g,m1,m2	THRES *	RN to syst., b from syst.
Ta-181(n,nd)Hf-179g,m1,m2	THRES	RN to syst., b from syst.
W -182(n,α) Hf-179g,m1,m2	ACTL	RN to syst., b from syst.
W -182(n,nα)Hf-178g,m1,m2	THRES	Ibid
W -184(n,γ) W -185g,m	ENDF/B-4	b from syst.
W -186(n,2n)W -185g,m	ECN (BRC)	RN to exp.
W -186(n,γ) W -187	ENDF/B-4	No RN
Re-185(n,γ) Re-186g,m	ENDF/B-4	b from syst.

Re-187(n,2n)Re-186g,m	ACTL	RN to syst., b from exp.
Os-190(n,γ) Os-191g,m	ECN(FISPRO)	b from syst.
Os-192(n,γ) Os-193g,m	ECN(FISPRO)	b from syst.
Os-192(n,2n)Os-191g,m	ECN (IBJ)	RN to exp.
Ir-191(n,γ) Ir-192g,m1,m2	ECN(FISPRO)	b from syst.
Ir-193(n,2n)Ir-192g,m1,m2	ECN (IBJ)	RN to syst., b from syst. and exp.
Pb-206(n,2n)Pb-205	ECN (BRC)	RN to exp.
Pb-208(n,γ) Pb-209	JENDL-2	No RN
Bi-209(n,2n)Bi-208	BRC(EFF-1)	RN to exp.
Bi-208(n,2n)Bi-207	THRES	RN to exp.
Bi-209(n,γ) Bi-210g,m	BRC(EFF-1)	b from syst.

* Different shapes for ground and metastable state production.

For all reactions mentioned above we have made uncertainty estimates based upon the information available at 14.5 MeV. These uncertainties are valid only at 14.5 MeV, but may also be representative for threshold reactions in a flux spectrum with a fusion peak at 14.1 MeV. For (n,γ) reactions the data at lower energies are probably more relevant and therefore individual treatment for different energy ranges was performed.

For a number of long-lived metastable targets the cross-sections are of interest, because it is possible that by neutron-induced reactions these (radio-active) isomers transform to a stable or short-lived nuclide. Therefore cross-sections were included for the following six isomers: Ag-108m, Ho-166m, Hf-178m2, Re-186m, Ir-192m2 and Bi-210m. Of special interest are the so-called "superelastic" (n,n') cross-sections that transform the metastable state by inelastic scattering into a ground state which is stable or has a shorter half life than the metastable target.

For cross-sections of metastable targets the same values as for the targets in the ground state have been assumed in REAC-ECN-3. This is a very rough assumption, but the systematics used for the 14.5 MeV cross-section values are independent of the target spin and thus the same values at 14.5 MeV result. For the isomer ratios there is only a dependence of the spin of the product nuclei (except for inelastic scattering), so also for the isomer ratios the same values result. For the (n,n'γ) reaction the systematics of Vonach /8/ is used for the g --> m transitions, whereas the systematics of ref. /10/ is used for the m --> g transitions. Two cases were distinguished:

1. One metastable-state targets - These have been treated as the low-spin isomers /10/ and the cross-section was estimated from $\sigma(n,n'\gamma) = 400 \cdot b(J_g)$ in mb.
2. Second metastable-state targets - The same procedure has been applied again, ignoring the presence of the first metastable state. It has to be noted, however, that especially for the Hf-178m2 target this approach is only very approximative due to the rather high energies of both isomeric states.

This approach results in appreciable superelastic scattering cross-sections, reducing long-lived activity.

The excitation functions for reactions starting from a metastable state have been assumed equal to those starting from the ground state. This is a crude approximation, certainly if the energy of the metastable state is large. It is noted for instance that for superelastic scattering there is no threshold, i.e. even at thermal energies the reaction is possible! Therefore, threshold corrections are required, in particular for Hf-178m2. These corrections have not been applied as yet.

Conclusions

We have produced a new data base REAC-ECN-3 for nuclear activation and transmutation calculations containing 8486 reactions with cross-sections between 0 and 20 MeV.

It is at present probably the largest source of data that is available for activation and transmutation calculations of fusion reactors based upon the (d,t) reaction. It is possibly the best data base for activation calculations of the first wall and inner blanket, since all 14.5 MeV data have been checked rather carefully. However, further improvements are needed, in particular for a selected number of important reactions replacing the THRES data by better estimates.

The next version of the current REAC-ECN-3 file will be issued as an 'European Activation File' (EAF) as a part of the EC Fusion Technology Programme. The number of reactions will be extended, including also important short-lived targets. The main emphasis, however, will be in the completeness and quality of the (n,γ) and (n,n'γ) data. A simplified statistical model code will be used for mass production of capture data, while for (n,n'γ) reaction the shape of the excitation curve will be improved including Qvalue adjustments. These reactions are important at relatively low energies and therefore the thermal and resonance ranges will be considered too.

In this way EAF will become a general activation file applicable for both fission and fusion reactor technology.

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