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Production and Validation of Isotope Production Cross Section Libraries for Neutrons and Protons to 1.7 GeV

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Abstract

For validation and development of codes and for modeling isotope production in high power accelerators and APT Materials studies, we have produced experimental, calculated, and evaluated activation libraries for interaction of nucleons with nuclides covering about a third of all natural elements. For targets considered here, our compilation of experimental data is the most complete we are aware of, since it contains all data available on the Web, in journal papers, laboratory reports, theses, and books, as well as all data included in the large compilation by Sobolevsky with co-authors (NUCLEX) published recently by Springer-Verlag in 4 volumes. Our evaluated library was produced using all available experimental cross sections together with calculations by the CEM95, LAHET, and HMS-ALICE codes and with the European Activation File EAF-97 and LANL Update II of the ECNAF Neutron Activation Cross-Section Library.

1. Introduction

Data on isotope production yields from various reactions are necessary to validate and develop models of nuclear reactions and are a decisive input for many applications, e.g., for accelerator transmutation of waste (ATW), accelerator-based conversion (ABC), accelerator-driven energy production (ADEP), accelerator production of tritium (APT), for the optimization of commercial production of radioisotopes used in medicine, mining, and industry, for solving problems of radiation protection of cosmonauts, aviators, workers at nuclear facilities, and for modeling radiation damage to computer chips, etc. (see details and references in [1]). Also, residual product nuclide yields in thin targets irradiated by medium- and high-energy projectiles are extensively used in cosmochemistry and cosmophysics to interpret the production of cosmogenic nuclides in meteorites by primary galactic particles.

Ideally, it would be desirable to have a universal library that includes data for all nuclides, projectiles, and incident energies. At present, neither the measurements nor the available codes permit one to create a reliable library covering all data needed at intermediate energies. First, experiments are costly and it is impossible to measure all data, in principle. Second, predictions by the best of available models, codes, and phenomenological systematics may differ for yields of certain isotopes at energies above 100 MeV by a factor of 100 or more, so that current models have to be further developed before they become reliable predictive tools (see, e.g., [1]). Construction of a universal comprehensive library would be very time consuming and costly, and is beyond the scope of the present work. Instead, we have collected experimental data, performed calculations with the most reliable codes, and evaluated cross sections for nucleon-nucleus interactions at intermediate energies for a number of targets of interest. Our progress in this activity is briefly described in this paper.

2. Experimental Data Library

We compile nucleon-induced isotope production experimental cross sections for two reasons. First, for validation [1] and further development [2] of the Cascade-Exciton Model (CEM) [3] code CEM95 [4] and to investigate the applicability of the recent ALICE code with the new Hybrid Monte Carlo Simulation model (HMS-ALICE) [5] to produce activation libraries up to 150 MeV [6, 7]. Second, we need these data to evaluate reliable libraries for our medical isotope production [8, 9] and material studies [10] at APT. For these tasks, we need data only up to several GeV. But realizing that such a compilation will be useful in the future for other problems, we do not limit ourselves to these energies, but compile all available data at any energy above several MeV. Since there are very few data on neutron-induced isotope production cross sections at energies above 100 MeV, we focus mainly on compiling proton-induced reaction data.

Many efforts have been previously made to compile experimental production yields from proton-induced reactions at intermediate energies. To the best of our knowledge, the most complete compilation was performed by Sobolevsky and co-authors and was published recently by Springer-Verlag in eight separate subvolumes [11]. Sobolevsky and co-authors have performed a major work and compiled all data available to them for target elements from Helium to transuranics for the entire energy range from thresholds up to the highest energy measured. For proton-induced reactions, this compilation contains about 37,000 data points published in the first four Subvolumes **I/13a-d** [11] (the following Subvolumes **I/13e-h** concern pion, deutron, triton, ^{3}He, and alpha induced reactions). This valuable compilation is also currently available in an electronic version as an IBM PC code named NUCLEX [11]. Unfortunately, since this compilation is very expensive (Springer Verlag sells a single new subvolume for $1647.00 or $2020.00; interested buyers may find information at: **http://www.springer-ny.com/catalog/np/nov96np/DATA/3-540-61045-6.html**) it is not easily available to individual researchers or to small libraries. Of more immediate concern is the fact that NUCLEX does not contain a large volume of data obtained during recent years, especially for proton-induced reactions.

Due to the increasing interest in intermediate-energy data for ATW, ABC, ADEP, APT, astrophysics, and other applications, precise and voluminous measurements of proton-induced spallation cross sections have been performed recently, and are presently in progress, by R. Michel et al. from Hannover University [12], Yu. E. Titarenko et al. at ITEP, Moscow [13], Yu. V. Aleksandrov et al. at JINR, Dubna [14], B. N. Belyaev et al. at B. P. Konstantinov St. Petersburg Institute of Nuclear Physics [15], N. I. Venikov et al. at Kurchatov Institute, Moscow [16] A. S. Danagulyan et al. at JINR, Dubna [17], H. Vonach et al. at LANL, Los Alamos [18], S. Sudar and S. M. Qaim at KFA, Jülich [19], D. W. Bardayan et al. at LBNL, Berkeley [20], J. M. Sisterson et al. at TRIUMF and other accelerators [21], etc. Finally, we note another, "new" type of nuclear reaction intensively studied in recent years, which provides irreplacable data for our needs. These are from reactions using reverse kinematics, when relativistic ions interact with hydrogen targets and they often provide the only way to obtain reliable data for interaction of intermediate energy protons with separate isotopes of an element with a complex natural isotopic composition. Much data from this type of reaction have been recently obtained, e.g., by W. R. Webber et al. at the LBL Bevalac [22, 23] and L. Tassan-Got et al. at GSI, Darmstadt [24]. These new data, as well as a number of other new and old measurements have not been covered by NUCLEX. Therefore, we do not confine ourselves solely to NUCLEX as a source of experimental cross sections; instead, we compile all available data for the targets in which we are interested, searching first the World Wide Web, then any other sources available to us, including the compilation from NUCLEX.

Table 1 shows the whole list of isotope production cross sections we have so far included in our experimental library file. Actually, our experimental library consists at present of 32 files, a separate file for each element, stored in a simple and easy to read format, and a README file with the description of

Table 1

Experimental Data Library for Proton-Induced Isotope Production

Target Z	Target #	Target	No. of reactions	The same, in NUCLEX [11]	No. of data points	The same, in NUCLEX [11]
6					652	487
	1	^{13}C	1	1		
	2	^{12}C	7	0		
	3	^{nat}C	13	8		
7					493	419
	4	^{14}N	11	0		
	5	^{13}N	5	5		
	6	^{nat}N	9	8		
8					400	347
	7	^{18}O	5	5		
	8	^{16}O	14	0		
	9	^{nat}O	16	11		
9					153	136
	10	^{19}F	9	9		
10					28	7
	11	^{22}Ne	1	1		
	12	^{20}Ne	21	0		
	13	^{nat}Ne	4	4		
11					96	91
	14	^{23}Na	8	8		
12					596	497
	15	^{26}Mg	3	3		
	16	^{25}Mg	3	3		
	17	^{24}Mg	26	3		
	18	^{nat}Mg	17	14		
13					1170	816
	19	^{27}Al	45	23		
15					42	37
	20	^{31}P	5	4		
16					93	49
	21	^{34}S	1	1		
	22	^{32}S	33	0		
	23	^{nat}S	10	9		
17					25	25
	24	^{nat}Cl	4	4		
18					102	66
	25	^{40}Ar	36	0		
	26	^{nat}Ar	17	17		
19					20	20
	27	^{nat}K	3	3		

Table 1 (continued)

Target Z	Target #	Target	No. of reactions	The same, in NUCLEX [11]	No. of data points	The same, in NUCLEX [11]
20					660	258
	28	^{48}Ca	1	1		
	29	^{44}Ca	4	4		
	30	^{43}Ca	2	2		
	31	^{42}Ca	1	1		
	32	^{40}Ca	57	0		
	33	^{nat}Ca	26	12		
26					2638	1189
	34	^{58}Fe	4	4		
	35	^{57}Fe	5	5		
	36	^{56}Fe	99	7		
	37	^{44}Fe	2	2		
	38	^{nat}Fe	92	58		
27					2213	871
	39	^{59}Co	54	48		
30					997	996
	40	^{70}Zn	2	2		
	41	^{68}Zn	6	6		
	42	^{67}Zn	3	3		
	43	^{66}Zn	3	3		
	44	^{64}Zn	6	6		
	45	^{nat}Zn	43	42		
31					356	356
	46	^{71}Ga	12	12		
	47	^{69}Ga	13	13		
	48	^{nat}Ga	6	6		
32					651	651
	49	^{76}Ge	31	31		
	50	^{74}Ge	3	3		
	51	^{73}Ge	2	2		
	52	^{72}Ge	6	6		
	53	^{70}Ge	29	29		
	54	^{nat}Ge	15	15		
33					198	180
	55	^{75}As	62	59		
39					1509	614
	56	^{89}Y	65	50		
40					2351	812
	57	^{96}Zr	14	14		
	58	^{94}Zr	30	30		
	59	^{92}Zr	4	4		

Table 1 (continued)

Target Z	Target #	Target	No. of reactions	The same, in NUCLEX [11]	No. of data points	The same, in NUCLEX [11]
40					2351	812
	60	^{91}Zr	44	42		
	61	^{90}Zr	40	40		
	62	^{nat}Zr	73	34		
41					898	205
	63	^{93}Nb	70	61		
42					1432	1339
	64	^{100}Mo	8	8		
	65	^{98}Mo	7	7		
	66	^{97}Mo	5	5		
	67	^{96}Mo	25	25		
	68	^{95}Mo	8	8		
	69	^{94}Mo	8	8		
	70	^{92}Mo	8	8		
	71	^{nat}Mo	113	38		
54					145	94
	72	^{126}Xe	2	0		
	73	^{124}Xe	6	2		
	74	^{nat}Xe	1	1		
55					370	370
	75	^{133}Cs	83	83		
56					834	238
	76	^{nat}Ba	79	30		
57					122	122
	77	^{nat}La	66	66		
77					114	94
	78	^{93}Ir	1	1		
	79	^{nat}Ir	46	28		
79					2104	935
	80	^{197}Au	278	271		
80					73	73
	81	^{202}Hg	10	10		
83					1142	995
	82	^{209}Bi	262	174		
Total:	**82**		**2272**	**1574**	**22679**	**13389**

the format and of references. Our library is still in progress and we hope to extend it, depending on our needs, and to make it available for users through the Web.

Presently, our experimental library contains 22,679 data points for 82 targets of 32 elements covering 2,272 proton-induced reactions. We also have begun to store in our library data for intermediate energy neutron-induced reactions, but so far we have only 95 data points for Bi and C targets covering 14 reactions induced by neutrons.

For comparison, we also show in Table 1 the statistics of available data for the same targets in NU-CLEX [11]. One can see, that for several targets like Cl, K, Ga, Ge, Cs, La, and Hg, no new measurements were performed in recent years, and we have not found more data than is in NUCLEX. On the other hand, for such targets like Ca, Fe, Co, Y, Zr, Ba, and Au, we have available 2-3 times more data points than in NUCLEX. Also, there are a number of targets like ^{12}C, ^{14}N, ^{16}O, ^{20}Ne, ^{32}S, ^{40}Ar, ^{40}Ca, and ^{126}Xe, for which there are no data at all in NUCLEX, while we presently have data for 181 reactions on these targets.

3. Calculated Cross Section Library

We needed to calculate a library of isotope production by nucleons at energies above 100 MeV to simulate the production of medical radioisotopes in a high-energy neutron and proton environment, e.g., at an APT Facility [8, 9], to benchmark [1] the CEM95 code [4], and to see how a similar library created by M. B. Chadwick [6] using the HMS-ALICE code [5] for energies below 150 MeV agrees with calculations at higher energies [7].

We use the CEM95 code [4] to perform most of the calculations for our activation library above 100 MeV. A number of reactions are calculated also with the LAHET code system (version 2.83) [25, 26], and just a few reactions, with the recently improved version of the CEM code [2]. For energies below 100 MeV, we use the LA150 activation library by M. B. Chadwick [6].

We store calculated cross sections for the production of all possible isotopes from 78 targets (see the list in Table 2), and then use the results we need. Most of the reactions are calculated for energies from 100 MeV to 1.7 GeV, according to the needs of our medical isotope production study [8, 9], while several reactions are calculated only up to 1.0 GeV for [10], while some others used extensively in our benchmark [1, 7], were calculated from 10 MeV to 5 GeV. Having already a tested method for production of activation cross sections at these energies, our library can be extended easily for other targets, when necessary.

We have benchmarked our calculations against all available experimental data. Figures with comparison of more than 1000 excitation functions calculated with CEM95, LAHET, and HMS-ALICE codes with the available experimental data and predictions of other well-known models can be found in [1, 7, 8, 9]. From this comparison we see that, for the majority of nuclides the results agree quite well with each other and with the data. We can therefore conclude that the calculated activation cross sections are reasonably reliable and can be used together with available experimental data to produce an evaluated activation library, which we describe in the next section.

4. Evaluated Library

As we mentioned previously, neither available experimental data nor any of the current models or phenomenological systematics can be used alone to produce a reliable evaluated activation library covering a large area of target nuclides and incident energies. Therefore, we choose to create our evaluated library by approximating by hand smoothly, wherever possible, excitation functions using all available experimental data along with calculations using some more reliable codes, employing each of them in the corresponding regions of targets and incident energies where they work better. When we have reliable experimental data, they are taken as the highest priority for our approximation as compared to model results.

Table 2
List of targets covered by the CEM95 Activation Library: $p(n) + A \rightarrow$ any isotope
($10/100\,\text{MeV} \le T_0 \le 1.7/5.0\,\text{GeV}$)

^{12}C	^{58}Ni, ^{60}Ni, ^{61}Ni, ^{62}Ni, ^{64}Ni, ^{nat}Ni
^{14}N	^{63}Cu, ^{65}Cu, ^{nat}Cu
^{16}O, ^{18}O	^{67}Zn, ^{68}Zn, ^{70}Zn
^{19}F	^{69}Ga, ^{71}Ga
^{21}Ne, ^{22}Ne	^{70}Ge, ^{73}Ge, ^{74}Ge, ^{76}Ge
^{23}Na	^{75}As
^{24}Mg, ^{25}Mg, ^{26}Mg , ^{nat}Mg	^{89}Y
^{27}Al	^{90}Zr, ^{91}Zr, ^{92}Zr, ^{94}Zr, ^{96}Zr, ^{nat}Zr
^{32}S, ^{33}S, ^{36}S , ^{nat}S	^{93}Nb
^{35}Cl, ^{37}Cl	^{92}Mo, ^{94}Mo, ^{95}Mo, ^{96}Mo, ^{97}Mo, ^{98}Mo, ^{100}Mo
^{36}Ar, ^{38}Ar, ^{40}Ar	^{132}Xe, ^{134}Xe
^{39}K, ^{40}K, ^{41}K	^{133}Cs
^{40}Ca	^{134}Ba, ^{135}Ba, ^{136}Ba, ^{137}Ba, ^{138}Ba , ^{nat}Ba
^{54}Fe, ^{56}Fe, ^{57}Fe, ^{58}Fe, ^{nat}Fe	^{138}La, ^{139}La
^{59}Co	^{197}Au

The recent *International Code Comparisons for Intermediate Energy Nuclear Data* organized by NEA/OECD at Paris [27, 28], our own comprehensive benchmarks [1, 7, 8, 9], and several studies by Titarenko et al. [13] have shown that CEM95 and LAHET generally have the best predictive powers for spallation reactions at energies above 100 MeV as compared to other available models. Therefore, we choose them above 100 MeV to create our evaluated library. Actually, we employ the calculated library described in Sec. 3. The same benchmarks have shown that at lower energies, the HMS-ALICE code does one of the best jobs in comparison with other models. So, we use the activation library calculated by M. Chadwick [6] with the HMS-ALICE code [5] for protons below 100 MeV and neutrons between 20 and 100 MeV. In the overlapping region, between 100 and 150 MeV, we use both HMS-ALICE and CEM95 and/or LAHET results. For neutrons below 20 MeV, data of the European Activation File EAF-97, Rev. 1 [29] with some recent improvements by M. Herman [30] seem to be the most reliable, therefore we use them as the first priority in our evaluation.

Measured cross-section data from the compilation described in Sec. 2, when available, are included together with theoretical results and are used to evaluate cross sections for our medical isotope production study [8, 9] for 70 nuclides of 25 elements. We note that when we put together all these different theoretical results and experimental data, rarely do they agree perfectly with each other, providing a smooth continuity of evaluated excitation functions. Often, the resulting compilations show significant disagreement at energies where the available data progresses from one source to another. These sets are thinned to eliminate discrepant data, providing data sets of more-or-less reasonable continuity defining our evaluated cross sections used in calculations [8, 9].

Examples with typical results of evaluated activation cross sections for several proton and neutron reactions are shown in Figs. 1 and 2 by broad gray lines. 51 similar color figures for proton-induced reactions and 56 figures for neutrons, can be found on the Web, in our detailed report [9].
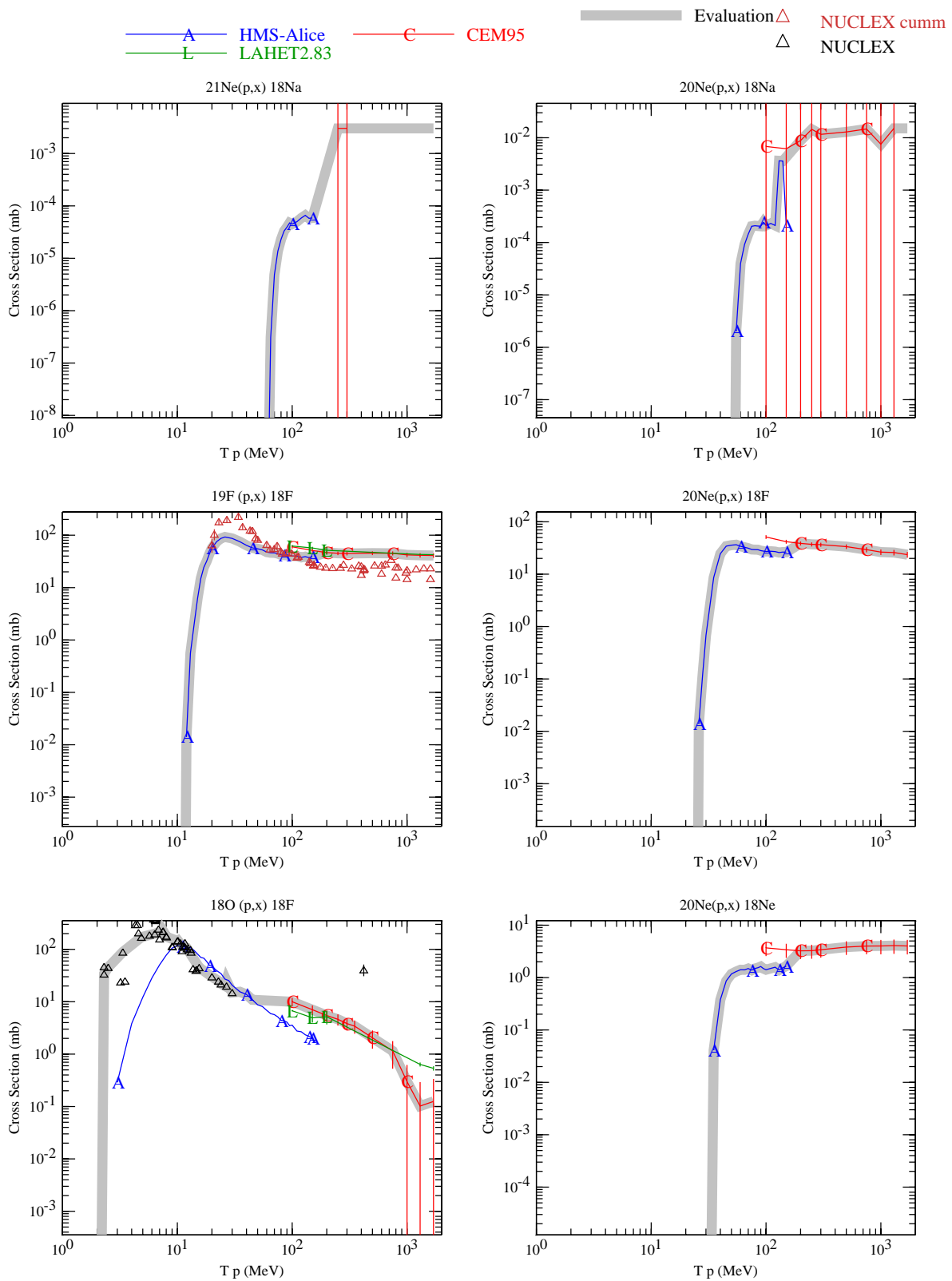
Figure 1. Example of several evaluated proton-induced activation cross sections. Evaluated cross sections are shown by broad gray lines, other notation are given in the plots and described in the text and in [9].
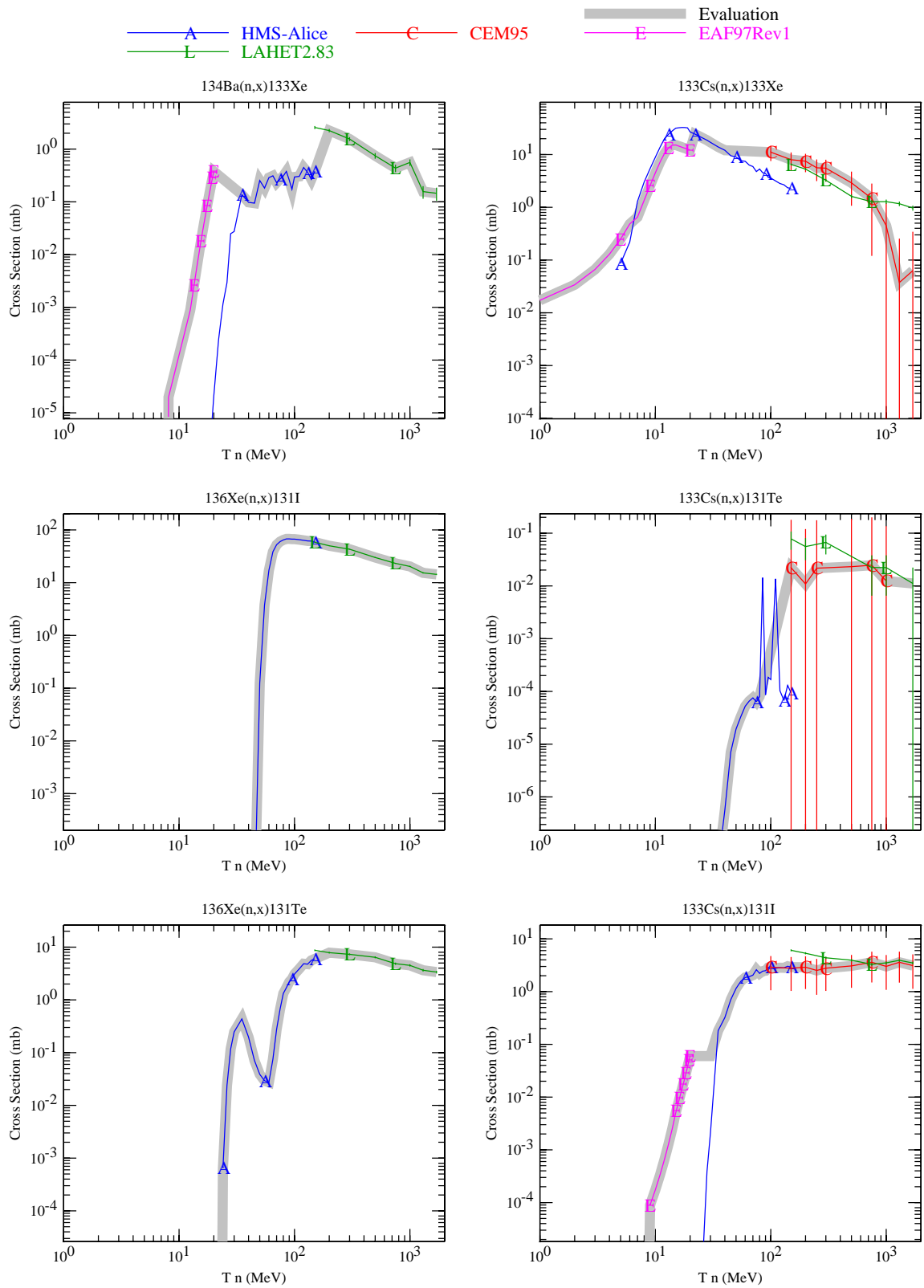
Figure 2. The same as in Fig. 1, but for neutron-induced reactions.

5. Summary

We have produced experimental, calculated, and evaluated activation libraries for the interaction of nucleons with nuclides covering about a third of all natural elements. Our compilation of experimental proton-induced cross sections contains 22,679 data points for 82 targets of 32 elements covering 2,272 proton-induced reactions and is the most complete we are aware of for these targets.

The methods developed are applicable to an extended set of reactions. We plan to extend our libraries, depending on our needs, and to make them available for users through the Web.

Acknowledgements

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