Update of the nuclear data for the neutron emissions for actinides of interest in safeguards

S. Simakov, M. Verpelli, N. Otsuka
Nuclear Data Section, IAEA

1. Spontaneous Fission Neutron Yields

The SG/IAEA requested update of Table 11-1 "Spontaneous fission neutron yields" on the page 339 of Los Alamos Report [1] published in 1991. This Table gives the recommended Total $T_{1/2}$ and Spontaneous $T(SF)_{1/2}$ Fission Half-lives, Spontaneous Fission Neutron Yields *n-Yield*, Spontaneous Fission Multiplicities $\nu(SF)$ for 18 isotopes of trans-actinides from ^{232}Th to ^{252}Cf (the numbers from the Table 11-1 are also reproduced in our Table 1 for comparison).

In this document the source of information were: for Total Half-lives $T_{1/2}$ - [2], for Spontaneous Fission Half-lives $T(SF)_{1/2}$ - [3], for Spontaneous Fission Multiplicities ν(SF) - [3] and [4]. The authors of Report [3] dated by 1981, in turn, have taken the values for $\nu(SF)$ from evaluation made by Manero and Konshin in 1972 [5]. Thus the nuclear data used in Table 11-1 of LASL Report and issued in 1991 indeed originate from the documents published in 1972 - 1981.

The currently available nuclear data relevant to the spontaneous fission neutron yields are presented in Table 1.

The **Total Half-lives** were recently evaluated in the frame of the IAEA CRP "Updated Decay Data Library for Actinides" [6, 7]. The results of evaluation were also included in the Decay Data Evaluation Project (DDEP) [8]. Since isotope ^{249}Bk was not considered in the recent IAEA CRP, its $T_{1/2}$ was taken from Evaluated Nuclear Structure Data Files (ENSDF) [9].

The **Spontaneous Fission Half-lives** for the most of isotopes we also selected from IAEA CRP results or DDEP except ^{232}Th, ^{241}Pu and ^{249}Bk, which we borrow from recommendations made by N. Holden [10]. For three isotopes ^{233}U, ^{237}Np and ^{241}Pu, which have the largest half lives, only the low limit is given.

Recently the new measurements of the spontaneous fission half-life of ^{240,242}Pu were published [11]. These results support within uncertainties the previous evaluations [10, 8].

The prompt **Spontaneous Fission Multiplicities** or nu-bar $\nu_p(SF)$ are presently available for the most of isotopes in the JEFF-3.1 evaluation [12] (these values are identical to ones in ENDF/B-VII.1).

The JEFF-3.1 nu-bars are sourced from evaluation made by Nichols and James in 1981 [13, 14]. Later in 1985 Holden published evaluations for ^{242,244}Cm and ^{249}Bk [15, 16, 17]. The nu-bars for ^{233}U, ^{237}Np and ^{241}Pu were not updated yet since the pioneer evaluation made by Manero and Konshin in 1972 [5].

Neutron Yields *n-Yield* from spontaneous fission was calculated employing formulas:

$$n - Yield = \nu(SF)_t \frac{T_{\frac{1}{2}}}{T(SF)_{\frac{1}{2}}} \qquad [\frac{n}{decay}]$$

and

$$n - Yield \ = \nu(SF)_t \ \frac{\ln(2)}{T(SF)_{\frac{1}{2}}} M \quad [\frac{n}{sec\ gram}]$$

where *M* is isotope mass taken from the Atomic Mass Evaluation NUBASE 2012 [18, 19].

Conclusion: Updated SF Neutron Yields and Comparison with Evaluation [1] **made in 1991**. The results of re-calculation of n-Yields and their uncertainties employing the updated nuclear data are presented in Table1. The comparison with values of Table 11-1 [1] shows an agreement within 1-3% for ^{238}U, ^{237}Np, ^{238,240,242} Pu, ^{241}Am, ^{244}Cm, ^{249}Bk and ^{252}Cf, however large differences for others.

The main reason for such essential differences is updated spontaneous fission half-lives became available during last 30-40 years. Thus for ^{235}U the value of $T(SF)_{1/2}$ has changed from 3.5 10^{+17} to (9.8 ± 0.28) 10^{+18} years or practically 30 times!

For three isotopes (^{233}U, ^{237}Np and ^{241}Pu) only low limit of spontaneous fission half-lives is known. For the several others (^{232}Th, ^{232,234,235}U, ^{239}Pu, ^{241}Am) the $T(SF)_{1/2}$ values have uncertainties between 15 and 33%. Obviously, the new and precise measurements are needed.

Table 1. Updated Half-Lives, Spontaneous Fission (SF) Half-Lives, SF prompt nu-bars ν_p and calculated SF neutron Yields. For comparison the values from Table 11-1 of [1] are given (*italic*) as well as Ratio of the updated neutron Yields to the previous ones [1] (highlighted as green are the ratios close to the unity within uncertainty, red - outside).

El	A	Half-Life y	SF Half-Life y	SF ν_p	New n-Yield n/s/g	*n-Yield*[a] *n/s/g*	Ratio of New to Old
Th	**232**	1.402E+10 ± 6.000E+07	1.200E+21[b] ± 4.000E+20	1.500	**7.126E-08 ± 2.375E-08**	*6.000E-08*	1.188 ± 0.396
U	**232**	7.060E+01 ± 1.100E+00	2.521E+15 ± 5.417E+14	1.710	**3.866E-02 ± 8.328E-03**	*1.300E+00*	0.030 ± 0.006
U	**233**	1.591E+05 ± 2.000E+02	> 2.700E+17	1.750[c]	< **3.679E-04**	*8.600E-04*	0.428 ± 0.000
U	**234**	2.455E+05 ± 6.000E+02	1.500E+16 ± 2.000E+15	1.800	**6.782E-03 ± 9.044E-04**	*5.020E-03*	1.351 ± 0.180
U	**235**	7.040E+08 ± 1.000E+06	9.800E+18 ± 2.800E+18	1.870	**1.074E-05 ± 3.068E-06**	*2.990E-04*	0.036 ± 0.010
U	**236**	2.343E+07 ± 6.000E+04	2.490E+16 ± 1.300E+15	1.900	**4.276E-03 ± 2.235E-04**	*5.490E-03*	0.779 ± 0.041
U	**238**	4.468E+09 ± 5.000E+06	8.202E+15 ± 6.000E+13	2.000	**1.355E-02 ± 1.003E-04**	*1.360E-02*	0.996 ± 0.007
Np	**237**	2.144E+06 ± 7.000E+03	> 1.000E+18	2.040[c]	< **1.138E-04**	*1.140E-04*	0.999 ± 0.000
Pu	**238**	8.774E+01 ± 3.000E-02	4.740E+10 ± 1.200E+09	2.210	**2.591E+03 ± 6.560E+01**	*2.590E+03*	1.000 ± 0.025
Pu	**239**	2.410E+04 ± 1.100E+01	8.000E+15 ± 2.000E+15	2.320	**1.605E-02 ± 4.012E-03**	*2.180E-02*	0.736 ± 0.184
Pu	**240**	6.561E+03 ± 7.000E+00	1.150E+11 ± 2.000E+09	2.151	**1.031E+03 ± 1.796E+01**	*1.020E+03*	1.010 ± 0.018
Pu	**241**	1.433E+01 ± 4.000E-02	> 6.000E+16[b]	2.250[c]	< **2.058E-03**	*5.000E-02*	0.041 ± 0.000
Pu	**242**	3.730E+05 ± 3.000E+03	6.790E+10 ± 1.000E+09	2.141	**1.723E+03 ± 2.891E+01**	*1.720E+03*	1.002 ± 0.017
Am	**241**	4.326E+02 ± 6.000E-01	1.200E+14 ± 3.000E+13	2.500	**1.143E+00 ± 2.858E-01**	*1.180E+00*	0.969 ± 0.242
Cm	**242**	4.459E-01 ± 2.190E-04	7.010E+06 ± 1.500E+05	2.540[d] ± 0.020	**1.980E+07 ± 4.516E+05**	*2.100E+07*	0.943 ± 0.020
Cm	**244**	1.811E+01 ± 3.000E-02	1.340E+07 ± 8.000E+05	2.720[d] ± 0.020	**1.100E+07 ± 6.620E+05**	*1.080E+07*	1.019 ± 0.061
Bk	**249**	9.035E-01[e] ± 1.095E-02	1.800E+09[b] ± 1.000E+08	3.400[d] ± 0.050	**1.003E+05 ± 5.892E+03**	*1.000E+05*	1.003 ± 0.057
Cf	**252**	2.647E+00 ± 2.600E-03	8.576E+01 ± 2.300E-01	3.759	**2.305E+12 ± 7.185E+09**	*2.340E+12*	0.985 ± 0.003

Comments to Table 1:

Half-lives data are from IAEA-CRP [6, 7] and DDEP [8], except where indicated by a superscript.

SF ν_p data from JEFF 3.1.1 [12], which takes them from Nichols (1981) [13] except where indicated by a superscript:

a) Values from Reilly (1991) [1]

b) Values from Holden (2000) [10]

c) Values from Manero and Konshin (1972) [5]

d) Values from Holden (1985) [15, 16]

e) Values from ENSDF [4]

Bibliography

[1] D. Reilly, N. Ensslin and H. Smith, "Passive Nondestructive Assay of Nuclear Materials," Report NUREG/CR-5550, Los Alamos Laboratory, 1991.

[2] C. Lederer and V. S. Shirley, Table of Isotopes, 7th ed., New York: John Wiley & Sons, 1978.

[3] R. Perry and W. W. B., "Neutron Production from (a,n) Reactions and Spontaneous Fission in ThO2, UO2, and (U,Pu)O2 Fuels," Report LA-8869-MS, Los Alamos, 1981.

[4] "Evaluated Nuclear Data File ENDF/B-V".

[5] F. Manero and V. Konshin, "Status of the Energy-Dependent v-Values for the Heavy Isotopes (Z>90) from Thermal to 15 MeV and of v-Values for Spontaneous Fission," *Atomic Energy Review*, p. 637, 1972.

[6] M. Kellett and N. A. (Edtrs), "Library of Recommended Actinide Decay Data, 2011," IAEA, Vienna, STI/PUB/1618, 2013.

[7] "UPDATED DECAY DATA LIBRARY FOR ACTINIDES: 2012," IAEA, [Online]. Available: https://www-nds.iaea.org/act_ddl/index.htmlx.

[8] "Table of Radionuclides," [Online]. Available: http://www.nucleide.org/DDEP_WG/DDEPdata.htm.

[9] "Evaluated Nuclear Structure Data File (ENSDF)," [Online]. Available: http://www.nndc.bnl.gov/ensdf/.

[10] N. Holden and H. D.C., "Spontaneous fission half-lives for ground-state nuclides," *Pure Appl. Chem.*, vol. 72, no. 8, p. 1525–1562, 2000.

[11] P. Salvador-Castineira and T. Brys, "Highly accurate measurements of the spontaneous fission half-life of 240,242Pu," *Phys. Rev.*, vol. C88, p. 064611, 2013.

[12] "JEFF-3.1," [Online]. Available: http://www.oecd-nea.org/dbdata/jeff/.

[13] A. Nichols and M. James, "Radiocative Heavy Element Decay Data for Reactor Calculations," AEEW-R 1407, Winfrith, 1981.

[14] A. Nichols and M. James, "UKHEDD-1 Radiocative Heavy Element Decay Data for Reactor Calculations," INDC(NDS)-0060, IAEA, 1981.

[15] N. Holden and M. Zucker, "Prompt Neutron Multiplicities for the Transuranium Nuclides," BNL-NCS-36379, Brookhaven, 1985.

[16] N. Holden and M. Zucker, "Neutron Multiplicities for the Transuranium Nuclides," BNL 36467, Brookhaven, 1985.

[17] N. Holden and M. Zucker, "A re-evaluation of the average prompt neutron emission multiplicity (nubar) values from fission of uraniium and transuranium nuclides," *Nuclear Standard Reference Data*, p. 248, IAEA-TECDOC-335, 1985.

[18] G. Audi, M. Wang and e. al., "The AME2012 atomic mass evaluation," *Chinese Physics C*, vol. 36, no. 12, p. 1287, 2012.

[19] "Atomic Mass Data Cente," 2012. [Online]. Available: https://www-nds.iaea.org/amdc/.