Improvement of Prediction Power of FP Summation Calculations by Use of the TAGS Experimental Data

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The average β- and γ-ray energies par decay were calculated from the Total Absorption γ-ray Spectrometer (TAGS) measurements carried out at Idaho for 45 isotopes to replace the original JEFF-3.1 or JENDL values. As a result, the JEFF-3.1 summation calculation became fairly consistent with the results of the sample-irradiation decay-heat measurements both in the β- and the γ-ray components. This fact implies that the TAGS measurement is free from the so-called pandemonium problem as is expected. We propose a list of the important nuclides to be measured by the TAGS technique in near future.

1. Introduction

When we use the decay-scheme information for the radioactive fission products (FP) in the decay-heat summation calculations as their basis, we have to pay attention to the problem that the β-transitions to the highly-excited levels are apt to be lost from the them1). This problem is known as the pandemonium problem2). The calculated results based on JEFF-3.1, which was released in May 2005, could not reproduce the sample-irradiation experiments performed world wide, where the β- and the γ-ray components are measured separately. On the other hand, the result with JENDL FP Decay Data File 2000 (Hereafter JENDL) is quite consistent with the integral measurements. It is because JEFF-3.1 is generated exclusively based on the decay-schemes constructed from the experimental data. On the contrary, JENDL is made up of experimental data with theoretical supplementation of the gross theory of beta decay, to attain good consistency.

In the early 1990's, a series of Total Absorption γ-ray Spectrometer (TAGS) measurements was carried out at INEL (Idaho National Engineering Laboratory) for 45 FP nuclides3). One of the most important properties of the TAGS measurement is expected to be pandemonium-problem free. In this respect, the TAGS measurement is considered that it may provide a solid basis of the summation calculations4). The INEL group, however, terminated their TAGS activity in 1990's and, then, we can no longer expect the relevant new data from the U.S. nowadays. On the contrary, a European group recently started a new collaboration5), in which the TAGS technique is fully employed in measuring the β-strength functions of FP region nuclides.

We plan to propose them a list of the nuclides to be measured by the TAGS technique in the framework of the WPEC (Working Party on International Evaluation Cooperation of the NEA Nuclear Science Committee). For this purpose we select important FP nuclides which are assumed to be suffered from the pandemonium problem among the nuclides contributing largely to the FP decay heat in this paper.

2. TAGS Measurements

In the TAGS measurement, a NaI(Tl) scintillator is used as the γ-ray detector installed at a on-line mass separator. In principle all of the *γ*-rays emitted in a cascade accompanied by a de-excitation of a certain level deposit all of their energies into the scintillator giving the level (or a group of levels) energy into which the preceding *β*-transition have taken place. In this way the TAGS gives the level energy as the pulse energy and the *β*-feeding rate as the pulse height at the same time. These are exactly the data required to calculate the average *β*- and *γ*-ray energy releases per one *β*-decay of the parent nucleus, or E_β and E_γ. Therefore, if the TAGS measurements are carried out in an ideal way, the values of E_β and E_γ obtained from them are free from the pandemonium problem.

3. Calculation Results

Figure 1 shows the *γ*-ray component of the Pu-239 decay heat after a fission burst calculated with JEFF-3.1 (solid curve) and with JENDL (dotted curve). This figure indicates that JEFF-3.1 underestimates largely the integral measurements between 2 and 3,000 seconds. They are the pandemonium nuclides that cause the pulling down of the JEFF-3.1 result. As the next step, we introduced the TAGS values that were measured by Idaho group into the decay data of JEFF-3.1 and JENDL, respectively (Fig.2). In introducing the TAGS data, E_β and E_γ values were replaced by the TAGS-origin values for the 45 nuclides for which the Idaho-group made measurement. As a result, JEFF-3.1 became fairly consistent with the sample-irradiation measurements. Namely, the JEFF-3.1 curve is pulled up between 10 and 300 seconds and, as a result, part of the curve of JEFF-3.1 is caught in the error bar of the experimental data.

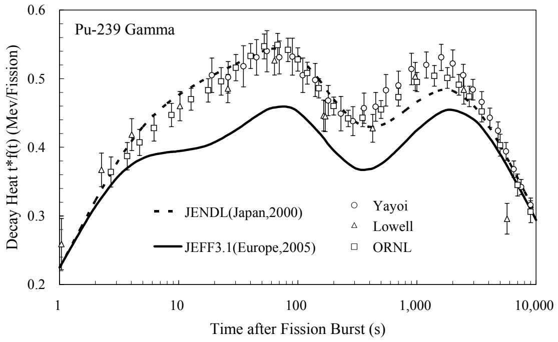

Fig. 1 Decay heat after a burst fission in Pu-239 before the TAGS-correction (γ-ray component)

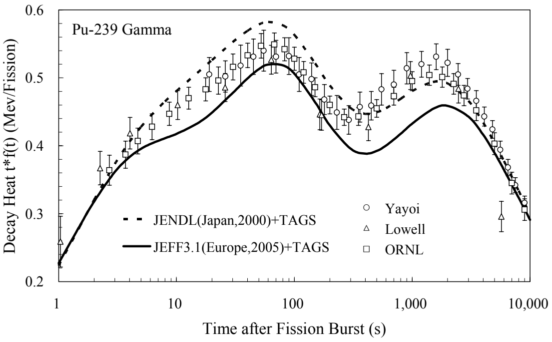

Fig. 2 Decay heat after a burst fission in Pu-239 after the TAGS correction (γ-ray component)

On the contrary, by introducing the TAGS data into the summation calculation, the JENDL curve deviated from the integral measurements. Figures 3 and 4 show the nuclide-wise contributions to the difference between the JENDL and the JEFF-3.1 curves both after the introduction of the TAGS data. Here the nuclides Tc-102, Mo-103, Mo-105, and Xe-139 are big contributors (Fig. 4). These isotopes are important candidates for the nuclides included in the list of nuclide to be measured by TAGS. Recently a series of isotopes of technetium was measured by the European group5). The results will be released sooner or later.

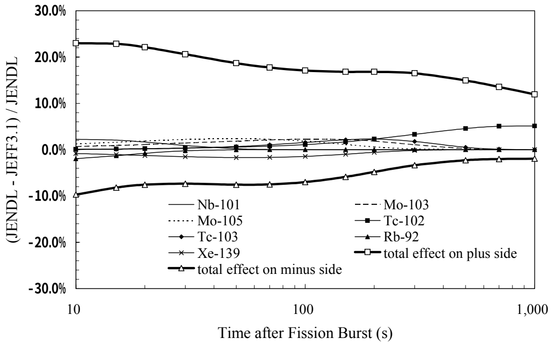

Fig. 3 Nuclide-wise contributions to the fractional difference between JENDL and JEFF-3.1 after introduction of the TAGS energies (γ-ray component of Pu-239 decay heat)

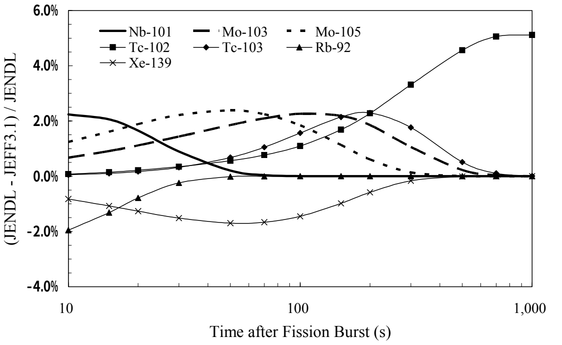

Fig. 4 Nuclide-wise contributions to the fractional difference between JENDL and JEFF-3.1 after introduction of the TAGS energies (the close-up of a few percent regions)

Figure 5 exhibits the contributions from the nuclides on the heavy (A>120, black dotted curve) and the light (A≦120, dotted curve) humps of the double-humped mass-yield curve to the γ-ray component of the Pu-239 decay heat. This figure suggests that the light group dominates the short-cooling time range and the heavy group does the long cooling-time range. The effect of the introduction of TAGS data (thin curve) into the heavy mass nuclides is bigger than the light group. Therefore, in the future TAGS measurement, the group of the light-mass nuclide should be measured with a higher priority, in the future.

Our present task is to make a high priority request list for the future TAGS measurements. For the same purpose Bersillon listed6) Br-87, Rb-92, Sr-89, Sr-97, Y-96, Nb-98, Nb-101, Nb-102, Tc-102, Tc-104, Tc-105, Te-135, Cs-142, Ba-145, La-143, and La-145 as the important nuclides to be studied (Hereafter Bersillon's list).

Table 1 through 4 list the nuclides which contribute appreciably to the difference between JEFF-3.1 and JENDL by more than 0.5% of the total fractional difference. We select the nuclides by the following three criteria:

a) If its contribution to the difference between JENDL and JEFF-3.1 is over 1.0% of the total fractional difference in the *β*-ray and *γ*-ray component of the decay heat often a burst fission in ^{235}U or ^{239}Pu or not,

b) If Appearing on the Bersillon's list or not,

c) If the highest known level is smaller than 70% of the Q-value or not.

We here put priority A, AA or AAA to each nuclide according to the number of the criteria which the nuclide in question satisfies. The results are listed in Table 5. As an exception we put a high priority to several technetium isotopes, for we have enough basis to believe that these isotopes are suffered from the pandemonium problem[7)].

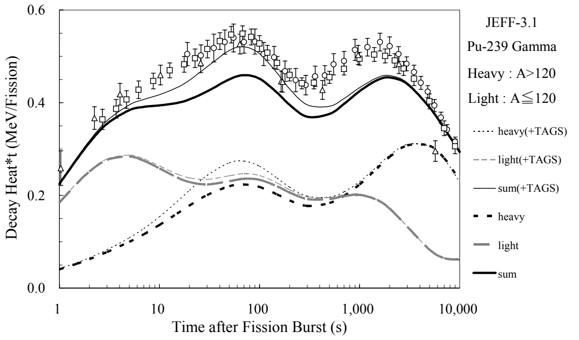

Fig. 5 The contributions from the nuclides on the heavy and the light humps of the double-humped of fission mass-yield curve nuclides to decay heat of γ-ray component of Pu-239 for JEFF-3.1

Table 5 The high priority request list for future TAGS measurement

nuclide				priority	nuclide				priority
Z		A	m		Z		A	m	
41	Nb	98	0	AAA	42	Mo	103	0	AA
41	Nb	101	0	AAA	42	Mo	105	0	AA
43	Tc	102	0	AAA	43	Tc	103	0	AA
43	Tc	104	0	AAA	43	Tc	106	0	AA
43	Tc	105	0	AAA	43	Tc	107	0	AA
37	Rb	92	0	AA	52	Te	135	0	AA
38	Sr	89	0	AA	56	Ba	145	0	AA
38	Sr	97	0	AA	57	La	145	0	AA
39	Y	96	0	AA	35	Br	87	0	A
40	Zr	100	0	AA	55	Cs	142	0	A
41	Nb	99	0	AA	57	La	143	0	A
41	Nb	102	0	AA					

4. Future Plan and Conclusion

We have to pay attention to the so-called pandemonium problem in calculating the average β- and γ-ray energies for decay-heat summation calculations. The successful introduction of the INEL-TAGS data into the decay-heat summation calculation on the basis of JEFF-3.1 decay data file suggests that TAGS data are free from pandemonium problem as has been expected. In this respect, further TAGS measurements for the FP region nuclides are highly encouraged. We selected important FP nuclides, which are assumed to be suffered from the pandemonium problem, among those contributing appreciably to the FP decay heat in rather short cooling-time range, and propose a list of the nuclides to be measured by the TAGS technique with high priority.

References

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Table 1 The nuclide contributing to the difference between JEFF-3.1 and JENDL by more than 0.5% of the total sum (U-235 Beta-ray component)

time(s)	nuclide				Q-value [keV]	last level [keV]	★1	★2	JENDL [MeV/fis.]	JEFF-3.1 [MeV/fis.]	★3	priority
	Z		A	m								
20	37	Rb	92	0	8105	7363	○	90.8%	1.49E-03	1.25E-03	0.66%	A
	41	Nb	99	0	3639	236		6.5%	1.10E-03	1.30E-03	-0.55%	A
	41	Nb	101	0	4569	811	○	17.7%	1.49E-03	1.70E-03	-0.57%	AA
	54	Xe	140	0	4060	2324		57.3%	7.23E-04	9.76E-04	-0.69%	A
	41	Nb	98	0	4586	2608	○	56.9%	1.48E-03	1.79E-03	-0.84%	AA
	39	Y	96	0	7087	6232	○	87.9%	1.34E-03	1.77E-03	-1.19%	AA
	52	Te	135	0	5960	4773	○	80.1%	1.19E-03	1.65E-03	-1.26%	AA
100	43	Tc	103	0	2660	1065		40.0%	8.85E-05	1.26E-04	-0.66%	A
	53	I	136	0	6930	6624		95.6%	2.01E-04	2.41E-04	-0.71%	
	41	Nb	98	0	4586	2608	○	56.9%	2.48E-04	3.00E-04	-0.90%	AA
1,000	43	Tc	102	0	4530	2909	○	64.2%	2.24E-05	3.10E-05	-2.07%	AAA

Table 2 The nuclide contributing to the difference between JEFF-3.1 and JENDL by more than 0.5% of the total sum (U-235 Gamma-ray component)

time(s)	nuclide				Q-value [keV]	last level [keV]	★1	★2	JENDL [MeV/fis.]	JEFF-3.1 [MeV/fis.]	★3	priority
	Z		A	m								
20	52	Te	135	0	5960	4773	○	80.1%	8.46E-04	2.60E-04	1.72%	AA
	39	Y	96	0	7087	6232	○	87.9%	6.06E-04	4.43E-05	1.65%	AA
	41	Nb	98	0	4586	2608	○	56.9%	7.77E-04	2.95E-04	1.42%	AAA
	41	Nb	101	0	4569	811	○	17.7%	6.34E-04	2.23E-04	1.21%	AAA
	41	Nb	99	0	3639	236		6.5%	5.36E-04	1.50E-04	1.14%	AA
	40	Zr	100	0	3335	704		21.1%	5.64E-04	1.86E-04	1.11%	AA
	39	Y	96	1	7087	5899	○	83.2%	1.33E-03	1.01E-03	0.95%	A
	35	Br	88	0	8960	7000		78.1%	1.22E-03	1.05E-03	0.53%	
	54	Xe	139	0	5057	4228		83.6%	6.50E-04	1.07E-03	-1.24%	A
	37	Rb	92	0	8105	7363	○	90.8%	2.21E-04	7.59E-04	-1.58%	AA
100	41	Nb	98	0	4586	2608	○	56.9%	1.30E-04	4.95E-05	1.15%	AAA
	41	Nb	99	1	4004	2944		73.5%	1.05E-04	4.15E-05	0.90%	
	42	Mo	103	0	3750	1621		43.2%	1.25E-04	7.33E-05	0.74%	A
	43	Tc	102	0	4530	2909	○	64.2%	4.72E-05	3.23E-06	0.63%	AA
	43	Tc	103	0	2660	1065		40.0%	6.95E-05	3.20E-05	0.53%	A
	51	Sb	133	0	4003	2756		68.8%	1.89E-04	1.54E-04	0.51%	A
	53	I	136	0	6930	6624		95.6%	2.37E-04	2.86E-04	-0.69%	
	54	Xe	139	0	5057	4228		83.6%	1.61E-04	2.64E-04	-1.48%	A
1,000	43	Tc	102	0	4530	2909	○	64.2%	1.88E-05	1.29E-06	3.40%	AAA
	51	Sb	130	1	4960	3413		68.8%	7.67E-06	3.97E-06	0.72%	A

★1: *Bersillon's list* $\bigstar 2:\dfrac{last\quad level}{Q-value}$ $\bigstar 3:\dfrac{JENDL-JEFF\text{ - }3.1}{JENDL}$

Table 3 The nuclide contributing to the difference between JEFF-3.1 and JENDL by more than 0.5% of the total sum (Pu-239 Beta-ray component)

time(s)	nuclide				Q-value [keV]	last level [keV]	★1	★2	JENDL [MeV/fis.]	JEFF-3.1 [MeV/fis.]	★3	priority
	Z		A	m								
20	53	I	138	0	7820	5342		68.3%	4.12E-04	2.32E-04	0.63%	A
	43	Tc	106	0	6547	3930		60.0%	8.00E-04	9.52E-04	-0.53%	A
	41	Nb	101	0	4569	811	○	17.7%	1.55E-03	1.76E-03	-0.72%	AA
	41	Nb	99	0	3639	236		6.5%	1.10E-03	1.31E-03	-0.73%	A
	39	Y	96	0	7087	6232	○	87.9%	7.47E-04	1.05E-03	-1.06%	AA
	41	Nb	98	0	4586	2608	○	56.9%	1.46E-03	1.77E-03	-1.07%	AAA
	43	Tc	107	0	4820	2680		55.6%	5.44E-04	8.82E-04	-1.17%	AA
100	43	Tc	102	0	4530	2909	○	64.2%	7.88E-05	1.10E-04	-0.61%	AA
	43	Tc	106	0	6547	3930		60.0%	2.07E-04	2.46E-04	-0.76%	A
	42	Mo	103	0	3750	1621		43.2%	2.84E-04	3.30E-04	-0.89%	A
	41	Nb	98	0	4586	2608	○	56.9%	2.44E-04	2.96E-04	-1.02%	AAA
	43	Tc	103	0	2660	1065		40.0%	2.03E-04	2.82E-04	-1.54%	AA
1,000	55	Cs	139	0	4213	3951		93.8%	3.34E-05	3.58E-05	-0.59%	
	51	Sb	130	1	4960	3413		68.8%	2.88E-06	6.04E-06	-0.77%	A
	43	Tc	104	0	5600	4268	○	76.2%	2.93E-05	3.42E-05	-1.19%	AA
	43	Tc	102	0	4530	2909	○	64.2%	3.14E-05	4.37E-05	-3.02%	AAA

Table 4 The nuclide contributing to the difference between JEFF-3.1 and JENDL by more than 0.5% of the total sum (Pu-239 Gamma-ray component)

time(s)	nuclide				Q-value [keV]	last level [keV]	★1	★2	JENDL [MeV/fis.]	JEFF-3.1 [MeV/fis.]	★3	priority
	Z		A	m								
20	42	Mo	105	0	4950	2766		55.9%	7.89E-04	2.97E-04	1.885%	AA
	52	Te	135	0	5960	4773	○	80.1%	6.34E-04	1.45E-04	1.875%	AA
	41	Nb	98	0	4586	2608	○	56.9%	7.66E-04	2.91E-04	1.82%	AAA
	43	Tc	107	0	4820	2680		55.6%	6.59E-04	2.21E-04	1.68%	AA
	41	Nb	101	0	4569	811	○	17.7%	6.62E-04	2.31E-04	1.65%	AAA
	39	Y	96	1	7087	5899	○	83.2%	1.56E-03	1.13E-03	1.65%	AA
	41	Nb	99	0	3639	236		6.5%	5.36E-04	1.51E-04	1.47%	AA
	40	Zr	100	0	3335	704		21.1%	4.80E-04	1.65E-04	1.21%	AA
	43	Tc	106	0	6547	3930		60.0%	1.39E-03	1.07E-03	1.20%	AA
	39	Y	96	0	7087	6232	○	87.9%	3.39E-04	2.63E-05	1.20%	AA
	42	Mo	103	0	3750	1621		43.2%	6.52E-04	3.61E-04	1.12%	AA
	40	Zr	98	0	2261	0		0.0%	1.36E-04	0.00E+00	0.52%	A
	53	I	136	1	7570	6624		87.5%	4.83E-04	6.54E-04	-0.66%	
	44	Ru	109	0	4160	2270		54.6%	1.46E-04	3.22E-04	-0.67%	A
	37	Rb	92	0	8105	7363	○	90.8%	7.94E-05	2.85E-04	-0.79%	A
	54	Xe	139	0	5057	4228		83.6%	3.80E-04	7.11E-04	-1.27%	A
100	42	Mo	103	0	3750	1621		43.2%	2.87E-04	1.59E-04	2.26%	AA
	42	Mo	105	0	4950	2766		55.9%	1.66E-04	6.26E-05	1.84%	AA
	43	Tc	103	0	2660	1065		40.0%	1.59E-04	7.13E-05	1.57%	AA
	43	Tc	106	0	6547	3930		60.0%	3.59E-04	2.77E-04	1.45%	AA
	41	Nb	98	0	4586	2608	○	56.9%	1.28E-04	4.88E-05	1.41%	AAA
	41	Nb	99	1	3639	2944		80.9%	1.03E-04	3.98E-05	1.12%	A
	43	Tc	102	0	4530	2909	○	64.2%	6.63E-05	4.56E-06	1.10%	AAA
	51	Sb	132	0	5290	3562		67.3%	1.35E-04	8.98E-05	0.81%	A
	45	Rh	108	0	4510	1540		34.1%	5.37E-05	1.29E-05	0.73%	A
	43	Tc	107	0	4820	2680		55.6%	4.88E-05	1.63E-05	0.58%	A
	45	Rh	109	0	2591	1318		50.9%	2.95E-05	5.93E-05	-0.53%	A
	44	Ru	109	0	4160	2270		54.6%	2.92E-05	6.45E-05	-0.63%	A
	45	Rh	110	0	5400	2805		51.9%	2.27E-06	5.20E-05	-0.88%	A
	53	I	136	1	7570	6091		80.5%	1.48E-04	2.00E-04	-0.93%	
	54	Xe	139	0	5057	4228		83.6%	9.40E-05	1.76E-04	-1.45%	A
1,000	43	Tc	102	0	4530	2909	○	64.2%	2.64E-05	1.82E-06	5.12%	AAA
	43	Tc	104	0	5600	4268	○	76.2%	4.68E-05	4.06E-05	1.31%	AA
	45	Rh	108	0	4510	1540		34.1%	5.58E-06	1.34E-06	0.88%	A
	53	I	134	0	4170	3492		83.7%	1.81E-05	1.53E-05	0.59%	