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New SG Proposal for Revalidation for FP Decay Data for Decay Heat Calculations

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Summation calculation is a versatile tool used to predict various aggregate behaviors of fission products including the FP decay heat. In this method the decay heat is calculated as the total sum of the energy releases from all the individual β-decaying nuclides existing at a relevant cooling-time. The data describing the characteristics of the β-decay of individual FP nuclides, therefore, are the indispensable prerequisite for summation calculations as well as the fission yield data. Once the set of decay data, primarily the average β- and γ-ray energy releases per decay (hereafter E_β and E_γ), are fixed, the calculated decay heat does not change decisively depending on the selection of yield data used herein. In this sense it seems more important to use a reliable decay data in the summation calculation of the decay heat.

As for the decay data are concerned, the E_β and E_γ values are usually obtained from the published decay schemes like those given in Tables of Isotopes. In the end of 1970's, it was argued by Hardy *et al.*[1] that the conventional decay schemes are overlooking or missing the proper β-feedings into the high-energy region of the daughter nucleus. They named it "*Pandemonium*" problem. Yoshida and Nakasima[2] pointed out that the FP decay heat calculations at that time were inevitably suffering from this problem through the use of average β- and γ-ray energies per decay, or E_β and E_γ derived from these decay schemes. And, then, they proposed a method to compensate these lost β-strengths on the basis of a nuclear-model calculation. By virtue of this method the calculated decay heat became very consistent with the measured ones except the cooling-time range 300 – 3000 s. The JENDL results taken from Ref 3) are exemplified in Appendix. In spite of this success, the introduction of the theory into the summation calculations has been considered as a kind of a temporary circumvention in a view of very long time-span. It was, then, a common recognition that the theoretically calculated values of E_β and E_γ should be replaced by the values having more solid basis, namely the experimentally obtained values, in the eventual course of time. Any alternative method which reproduce the measured decay heat as good as the theory-based method, however, has not appeared since then for more than two decades of years.

In 1990's, a total absorption γ-ray spectrometer (TAGS) had been developed [4] and was applied to the short-lived FP nuclides[5] by the INEL (Idaho National Engineering Laboratory) group in late 1990s. They measured the β-feeding as a function of the excitation energy of the daughter nuclide for 44 isotopes in the FP region. The TAGS data are, in principle, thought to be free from the pandemonium problem if the measurement is carried out in an ideal way. In addition, a European group recently started a series of the TAGS experiments at the Jyvaskyla on-line mass separator for several important FP nuclides[6].

The E_β and E_γ values calculated from the INEL data[5] were substituted into the FP decay data files included in JENDL, JEF2.2 and ENDF/B-VI. Using these modified and the original (or unsubstituted) files, the decay heats after a burst fission were calculated with the summation method and the results were compared with the integral measurement from the University of Tokyo[7]. In Figs.1 through 6 all the results for Pu-239 are displayed. The most sensitive results are seen in the γ-ray component and, they are enlarged in Fig.s 7 and 8 for detail inspection. In the case of JEF2.2, where any theoretical correction is not made for the missing of the β-strengths, improvement is remarkable. This possibly implies that the TAGS detects the high-energy β-strengths as is expected and that the E_β and E_γ values derived therefrom correctly reflects the contribution from the high-excitation β-strengths which tend to be missed in the published decay schemes.

On the other hand, in the case of JENDL (solid curves), where the correction is applied on the basis of the gross theory, the very good agreement is no longer maintained. The γ-ray component is overestimated in the cooling time range from 3 to 300 seconds suggesting a kind of an over-correction. In this cooling time range, the dominant nuclides which increase the γ-ray component (and correspondingly decreases the β-ray component) from the JENDL original are Cs-141 and La-144 as is seen from Fig 9, which shows the effect of the nuclide-wise replacement of JENDL E_γ by TAGS energy E_γ. These 11 isotopes shown in Fig. 7 are those which have relative contributions more than 0.5 % in the cooling time range less than 100s.

On the basis of the above observations it can be inferred that the experimental decay-data, specifically the β-feeding data, have been accumulated since 20 years so as to do summation calculation with minimal help from the nuclear theory calculation. Followings should be carried out in order to make full use of the accumulated decay data to improve the reliability of the summation calculations.

(1) Identify the nuclides which contribute largely to the FP decay heat in the cooling time range from 10 to 5000s from summation calculation.

(2) Find out the nuclides whose decay data are crucially suffering from the pandemonium problem among the nuclides identified in the procedure (1).

(3) Validate the necessity of further β-feeding measurement including the TAGS for the nuclides found out in the procedure (2). In doing this we make full use of the information from the theoretical calculation of the β-feeding.

References

1) J.C. Hardy, L.C.Carrez, B.Jonson, P.G.Hansen: *Phys. Lett.,* **71B**, 307(1977)
2) T.Yoshida, R. Nakashima: *J.Nucl. Sci. Technol,* **18**[6], 393 (1981)
3) J.Katakura, T.Yoshida, K.Oyamatsu, T.Tachibana, *JENDL FP Decay Data File 2000*, JAERI 1343 Japan Atomic Energy Research Institute (2001)
4) R.C.Greenwood, R.G.Hwlmer, M.A.Lee, *et al.*: *Nucl. Instr. and Meth.*, **A314,** 514 (1992)

5) R.C.Greenwood, R.G.Helmer, M.H.Putnam, K.D.Watts: *Nucl. Instr. and Meth*., **A390**, 95(1997)

6) J.L.Tain, Private communication, (2004)

7) Akiyama, M., An,S.: *Proc. Int. Conf. on Nuclear Data for Science and Technology*, Antwerp, p.237 (1982)

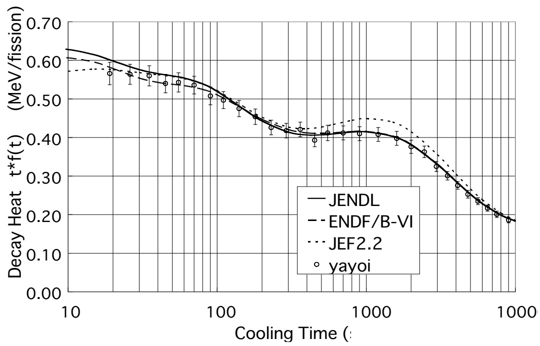


Fig.1 Decay Heat after a Burst Fission in Pu-239(β -ray Component)
Before Substitution of the TAGS E_β

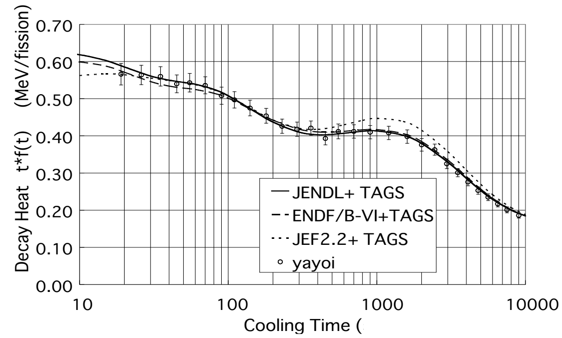


Fig.2 Decay Heat after a Burst Fission in Pu-239(β -ray Component)
After Substitution of the TAGS E_β

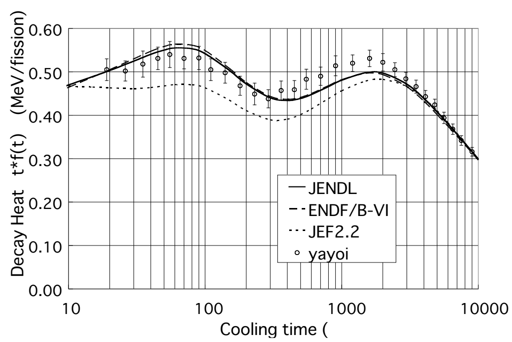


Fig.3 Decay Heat after a Burst Fission in Pu-239(γ-ray Component)
Before Substitution of the TAGS E_γ

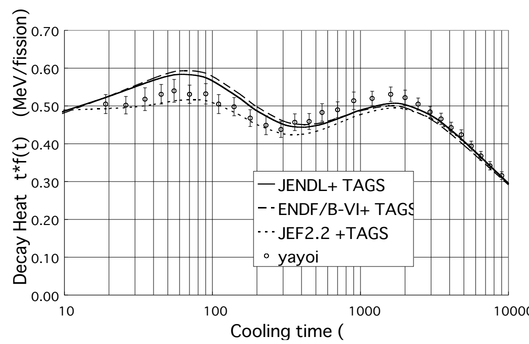


Fig.4 Decay Heat after a Burst fission in Pu-239(γ -ray Component)
After the TAGS Correction

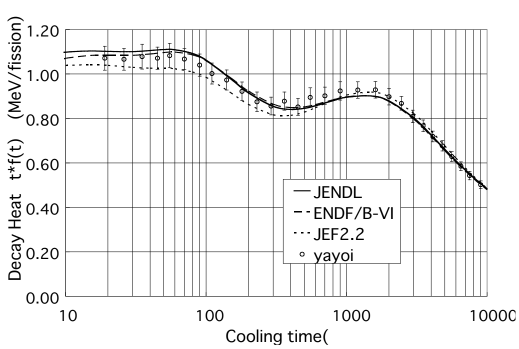


Fig.5 Decay Heat after a Burst Fission in Pu-239 (Total)
Before Substitution of the TAGS E_β and E_γ

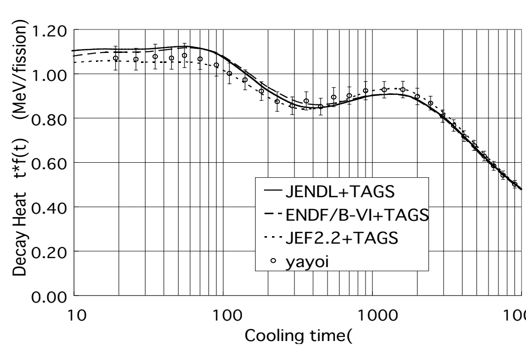


Fig.6 Decay Heat after a Burst Fission in Pu-239 (Total)
After Substitution of the TAGS E_β and E_γ

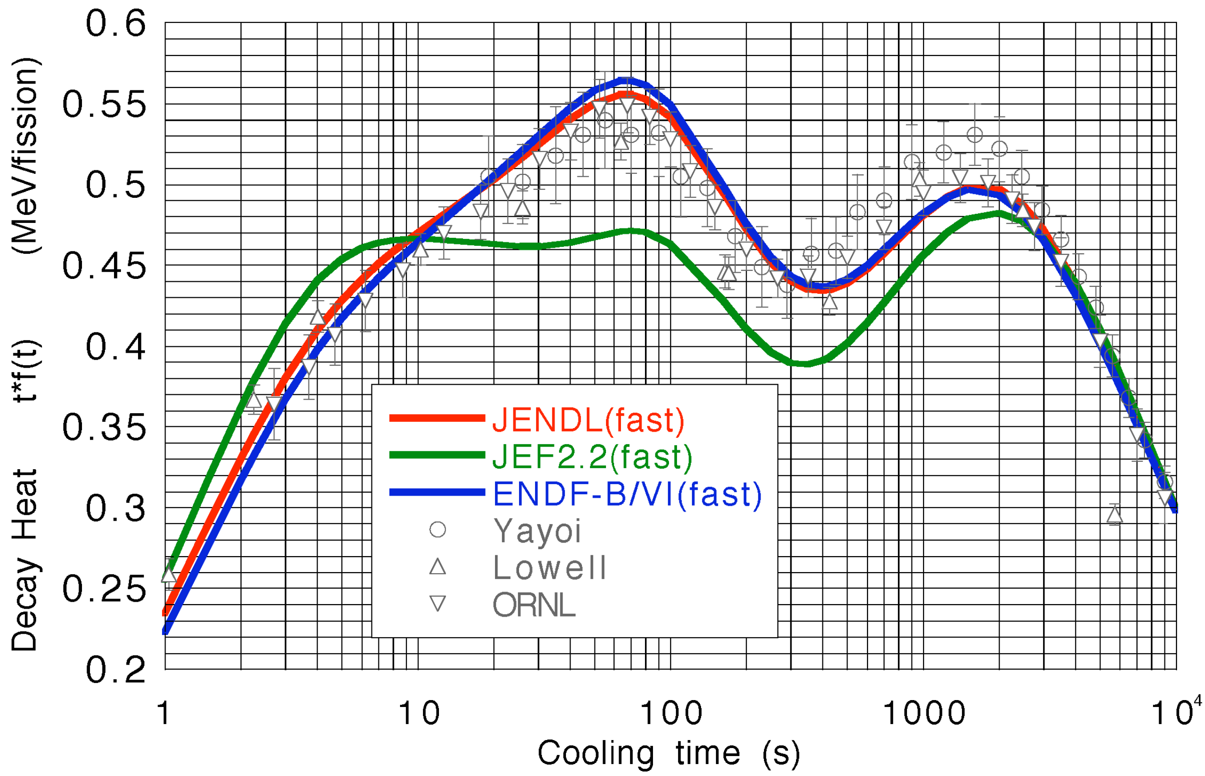


Fig. 7 Gamma-ray Component of FP Decay Heat after a Burst Fission in Pu239 Before Substitution of $E\gamma$ data from INEL-TAGS (Original Files)

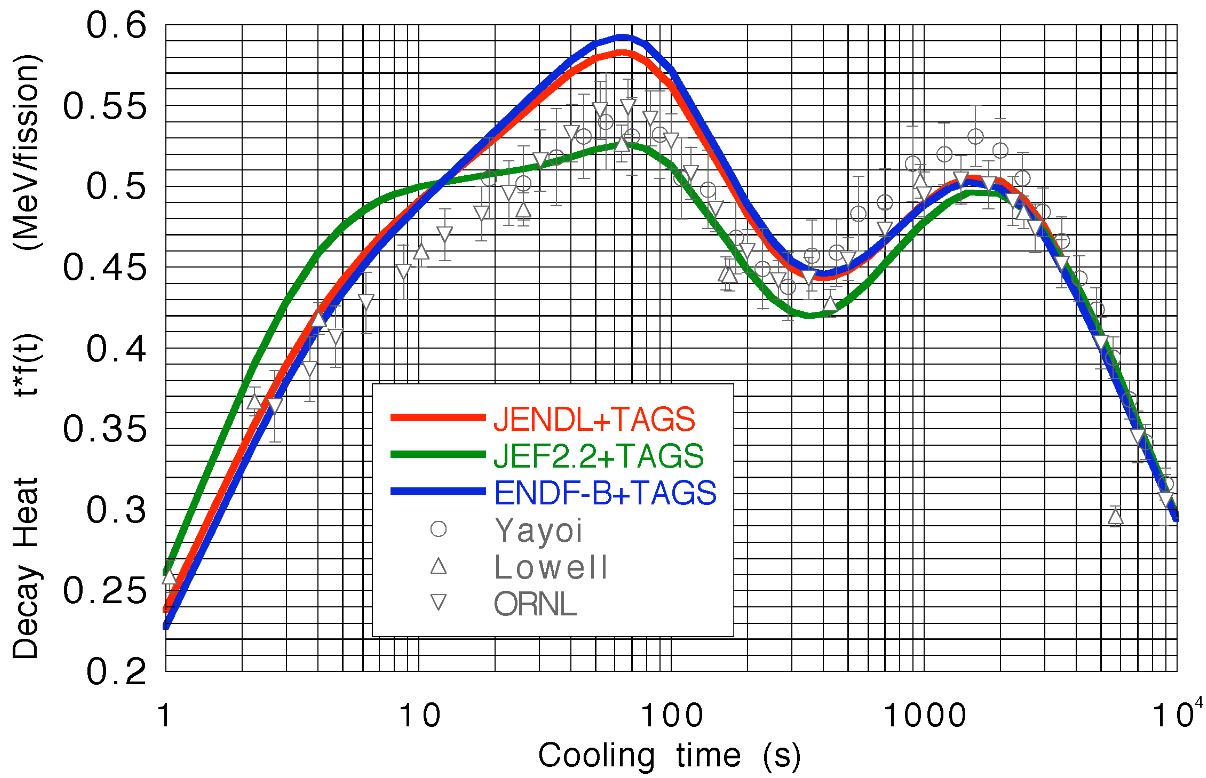


Fig. 8 Gamma-ray Component of FP Decay Heat after a Burst Fission in Pu239 After Substitution of $E\gamma$ data from INEL-TAGS

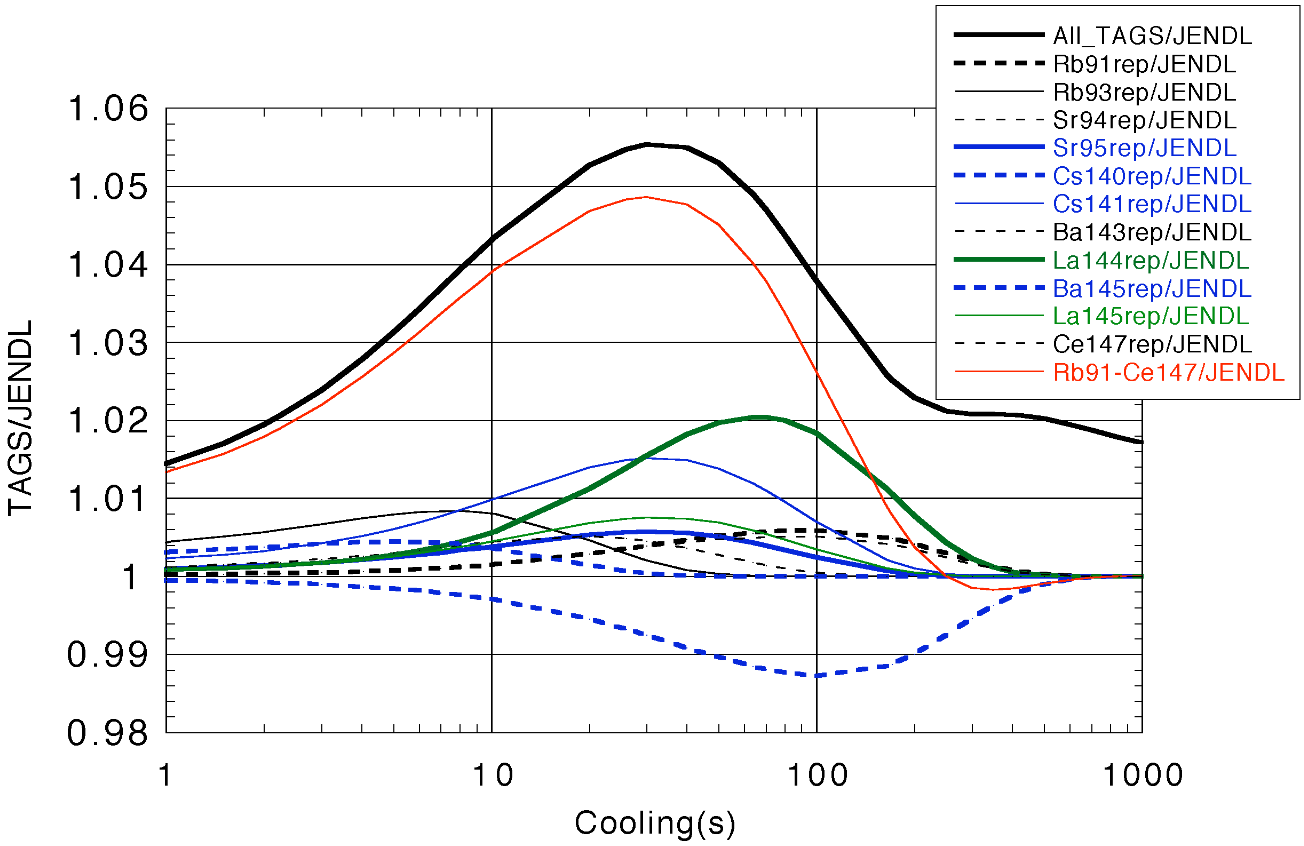
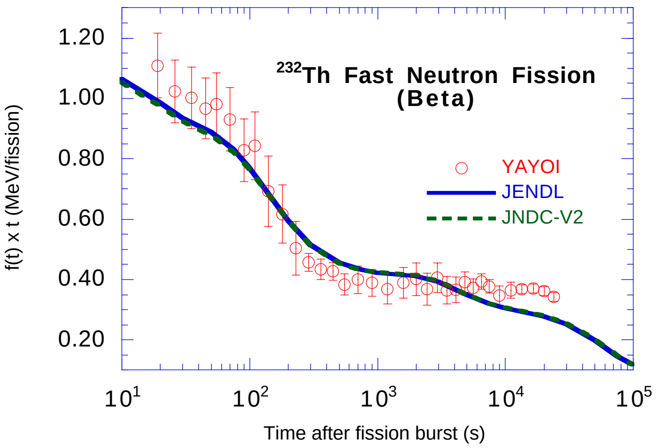


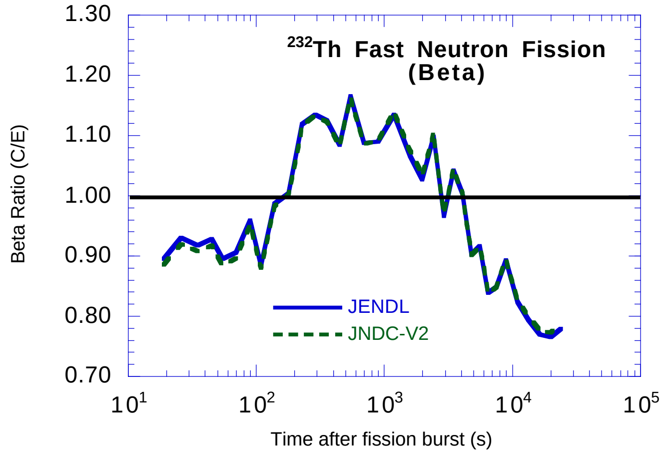
Fig. 9 Nuclide-wise Breakdown of the Differences in Calculated Decay Heat before and after the Substitution of INEL-TAGS E_γ Values into JENDL
(Only the Nuclides with Half-lives Greater than 100s are Taken into Account)

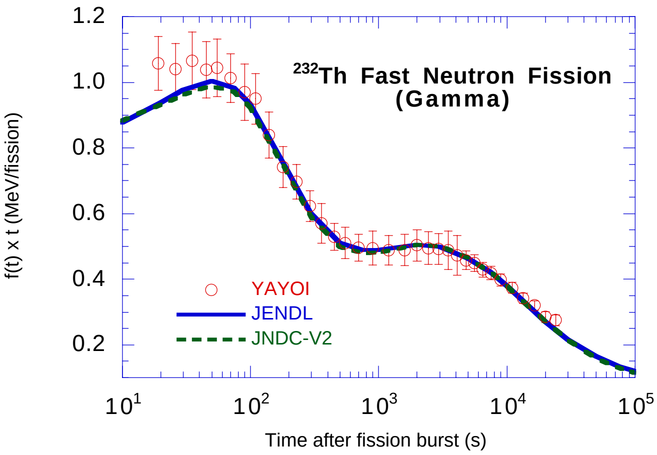
APPENDIX

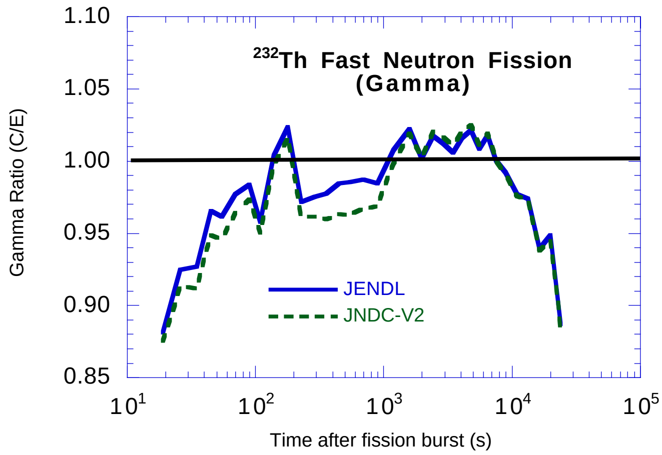
Comparison of the Summation Calculations with the YAYOI Experiments

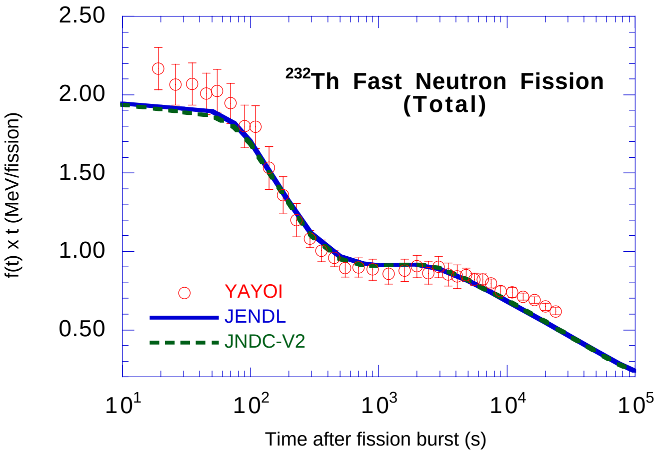
JENDL: JENDL FP Decay Data File 2000 (JAERI 1343, 2001)
JNDC: JNDC FP Decay Data File Version 2 (JAERI 1320,1990)

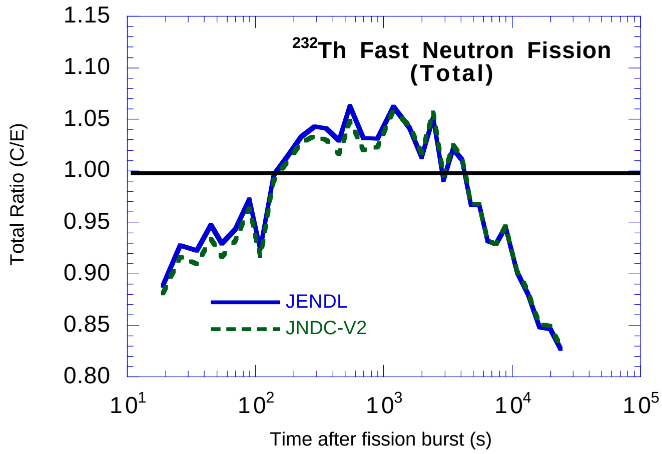

232Th Fast Neutron Fission
(Beta)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)

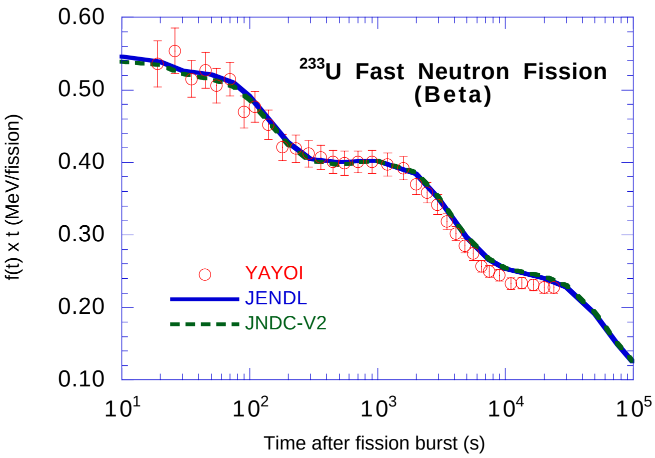

232Th Fast Neutron Fission
(Beta)
JENDL
JNDC-V2
Beta Ratio (C/E)
Time after fission burst (s)

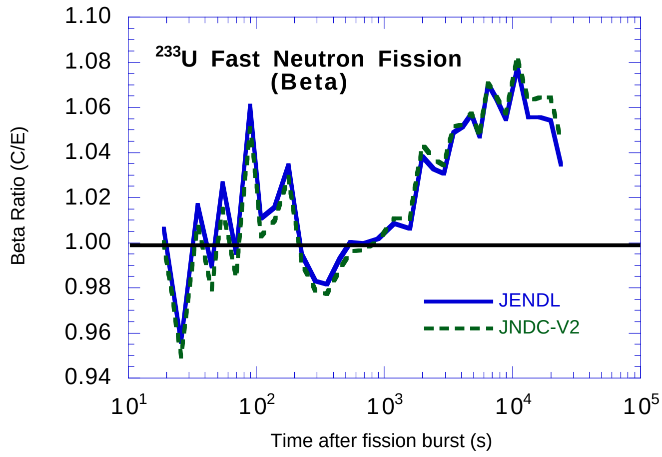

232Th Fast Neutron Fission
(Gamma)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)

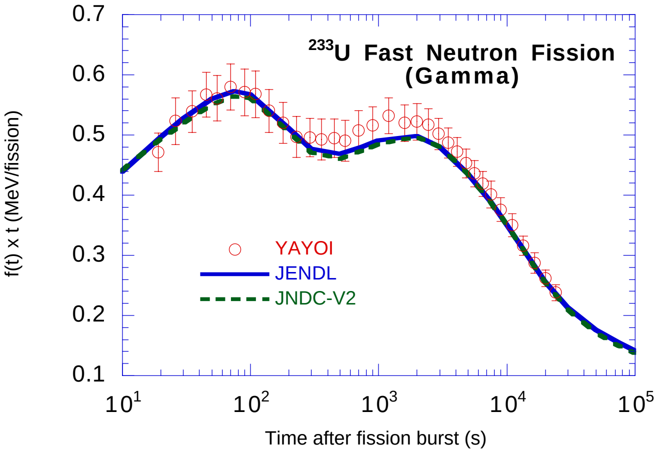

232Th Fast Neutron Fission
(Gamma)
JENDL
JNDC-V2
Gamma Ratio (C/E)
Time after fission burst (s)

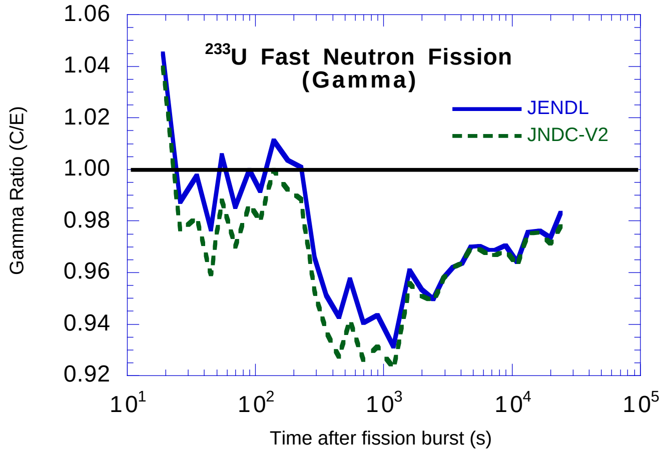

232Th Fast Neutron Fission
(Total)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)

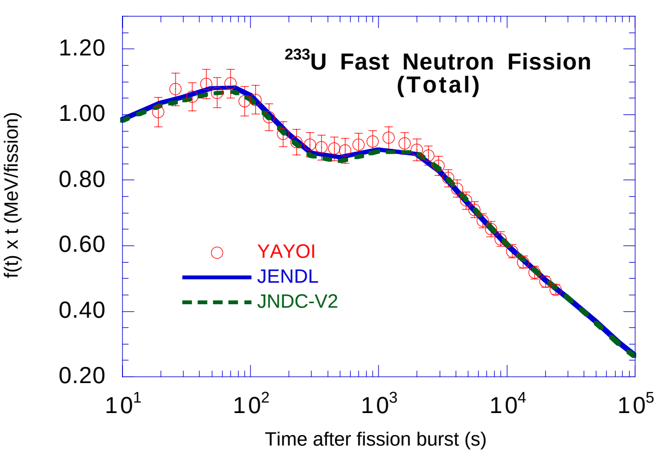

232Th Fast Neutron Fission
(Total)
JENDL
JNDC-V2
Total Ratio (C/E)
Time after fission burst (s)

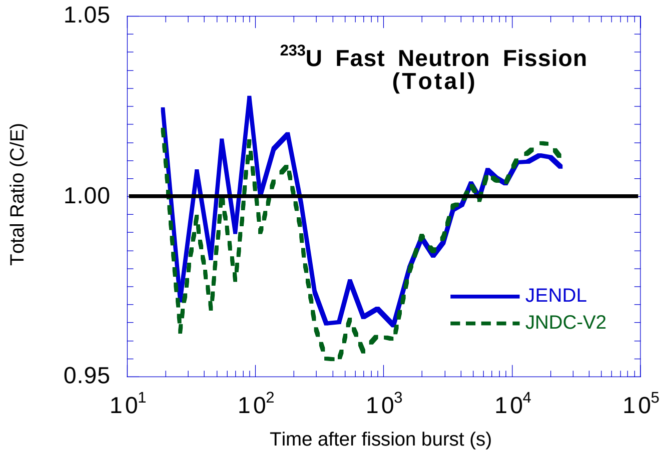

233U Fast Neutron Fission
(Beta)
f(t) x t (MeV/fission)
0.60
0.50
0.40
0.30
0.20
0.10
YAYOI
JENDL
JNDC-V2
10^1
10^2
10^3
10^4
10^5
Time after fission burst (s)

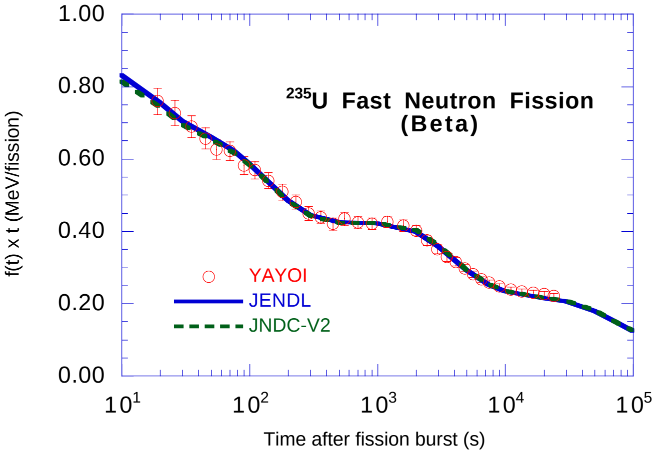

233U Fast Neutron Fission
(Beta)
Beta Ratio (C/E)
1.10
1.08
1.06
1.04
1.02
1.00
0.98
0.96
0.94
JENDL
JNDC-V2
10^1
10^2
10^3
10^4
10^5
Time after fission burst (s)


233U Fast Neutron Fission
(Gamma)
f(t) x t (MeV/fission)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
YAYOI
JENDL
JNDC-V2
10^1
10^2
10^3
10^4
10^5
Time after fission burst (s)

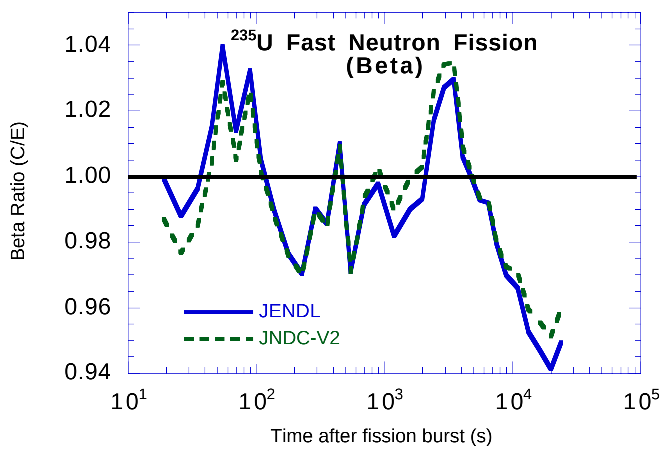

233U Fast Neutron Fission
(Gamma)
Gamma Ratio (C/E)
1.06
1.04
1.02
1.00
0.98
0.96
0.94
0.92
JENDL
JNDC-V2
10^1
10^2
10^3
10^4
10^5
Time after fission burst (s)


233U Fast Neutron Fission
(Total)
f(t) x t (MeV/fission)
1.20
1.00
0.80
0.60
0.40
0.20
YAYOI
JENDL
JNDC-V2
10^1
10^2
10^3
10^4
10^5
Time after fission burst (s)

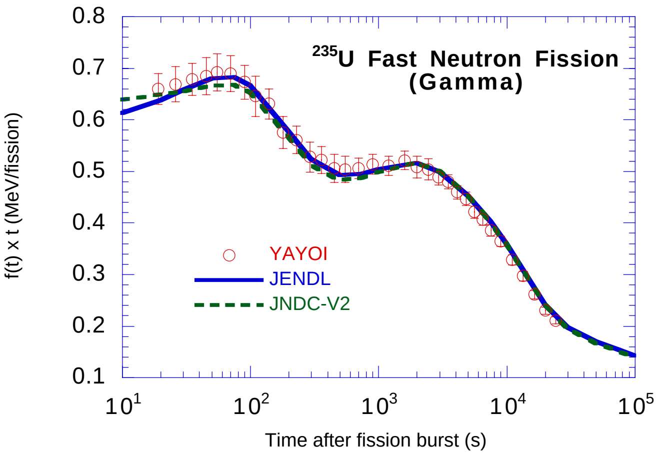

233U Fast Neutron Fission
(Total)
Total Ratio (C/E)
1.05
1.00
0.95
JENDL
JNDC-V2
10^1
10^2
10^3
10^4
10^5
Time after fission burst (s)



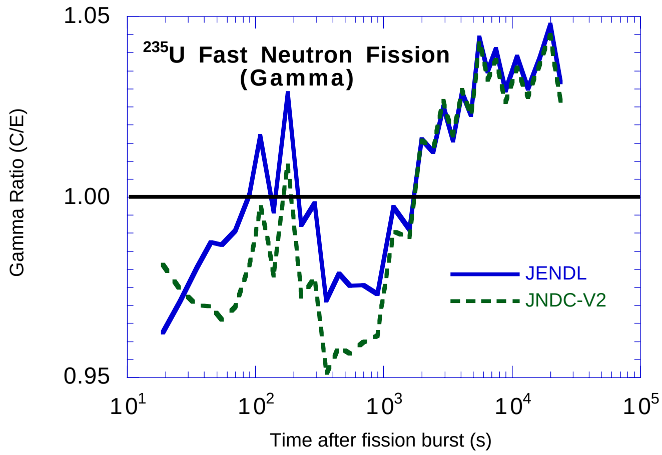
^{235}U Fast Neutron Fission
(Beta)
f(t) x t (MeV/fission)
Time after fission burst (s)
YAYOI
JENDL
JNDC-V2
1.00
0.80
0.60
0.40
0.20
0.00
10^1
10^2
10^3
10^4
10^5



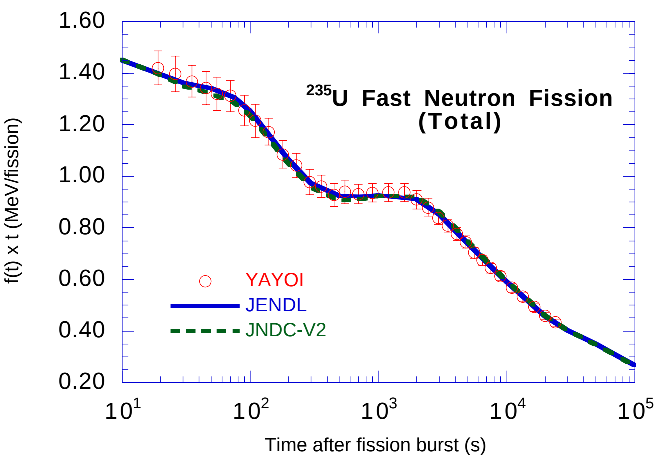
^{235}U Fast Neutron Fission
(Beta)
Beta Ratio (C/E)
Time after fission burst (s)
JENDL
JNDC-V2
1.04
1.02
1.00
0.98
0.96
0.94
10^1
10^2
10^3
10^4
10^5



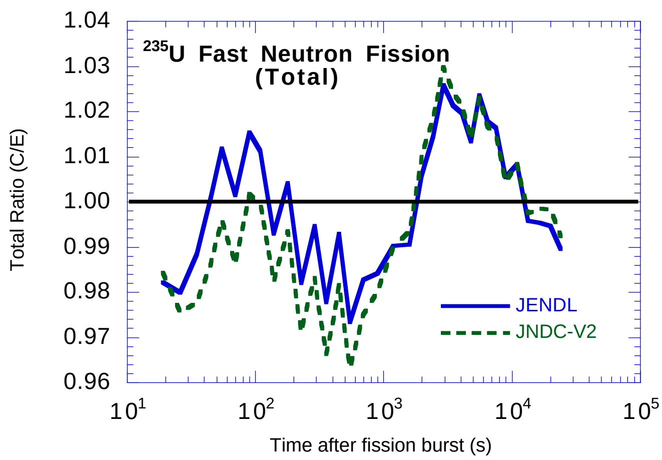
^{235}U Fast Neutron Fission
(Gamma)
f(t) x t (MeV/fission)
Time after fission burst (s)
YAYOI
JENDL
JNDC-V2
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
10^1
10^2
10^3
10^4
10^5



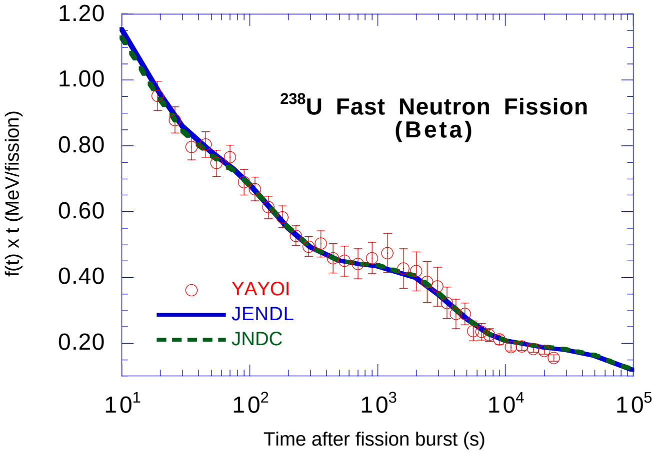
^{235}U Fast Neutron Fission
(Gamma)
Gamma Ratio (C/E)
Time after fission burst (s)
JENDL
JNDC-V2
1.05
1.00
0.95
10^1
10^2
10^3
10^4
10^5



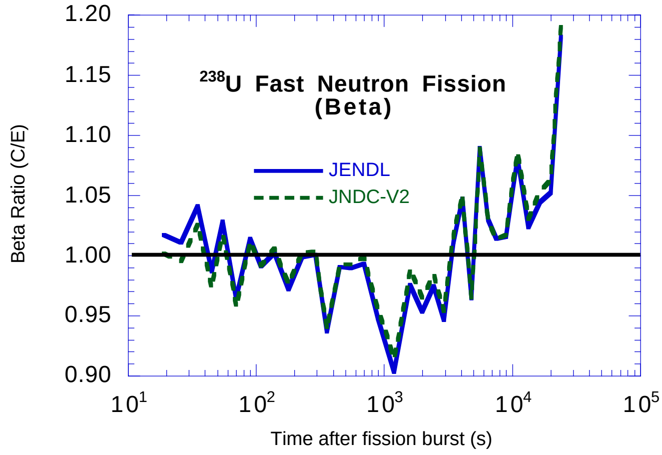
^{235}U Fast Neutron Fission
(Total)
f(t) x t (MeV/fission)
Time after fission burst (s)
YAYOI
JENDL
JNDC-V2
1.60
1.40
1.20
1.00
0.80
0.60
0.40
0.20
10^1
10^2
10^3
10^4
10^5



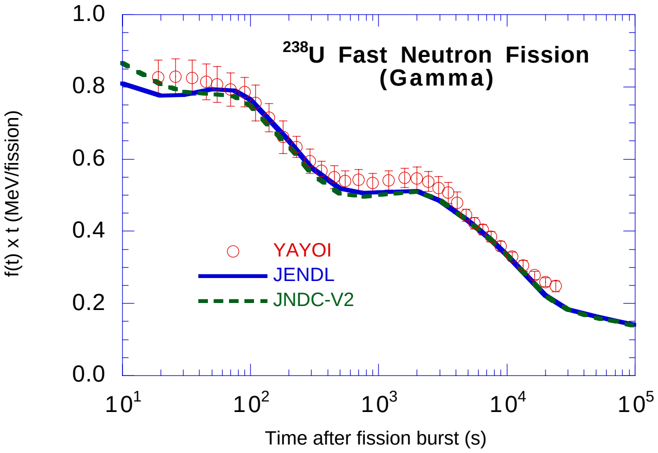
^{235}U Fast Neutron Fission
(Total)
Total Ratio (C/E)
Time after fission burst (s)
JENDL
JNDC-V2
1.04
1.03
1.02
1.01
1.00
0.99
0.98
0.97
0.96
10^1
10^2
10^3
10^4
10^5



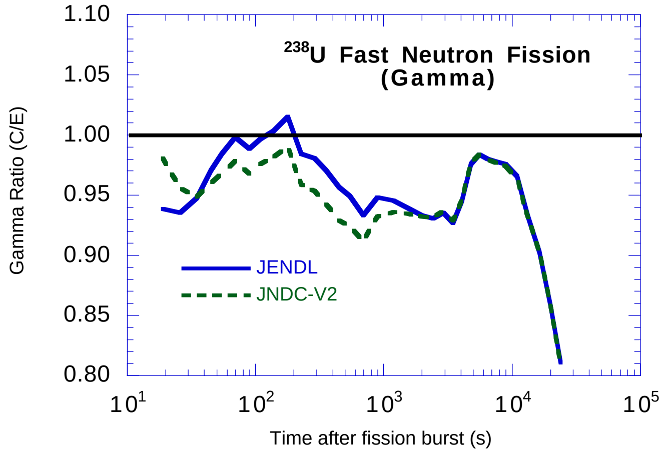
238U Fast Neutron Fission
(Beta)
YAYOI
JENDL
JNDC
f(t) x t (MeV/fission)
Time after fission burst (s)



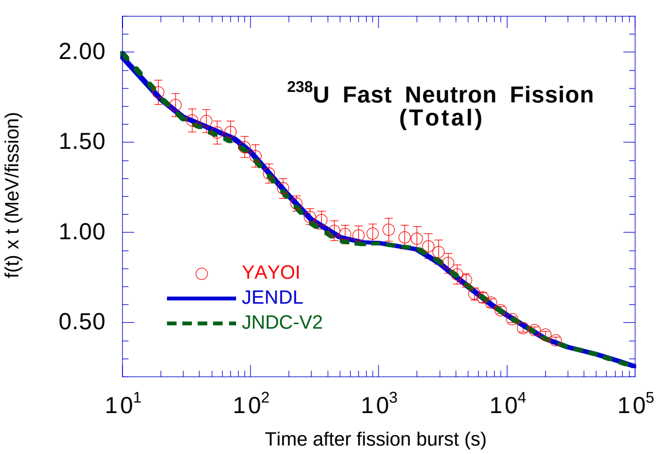
238U Fast Neutron Fission
(Beta)
JENDL
JNDC-V2
Beta Ratio (C/E)
Time after fission burst (s)



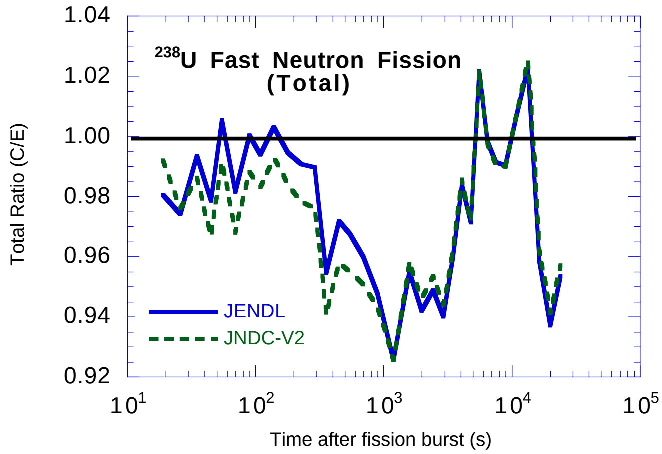
238U Fast Neutron Fission
(Gamma)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)



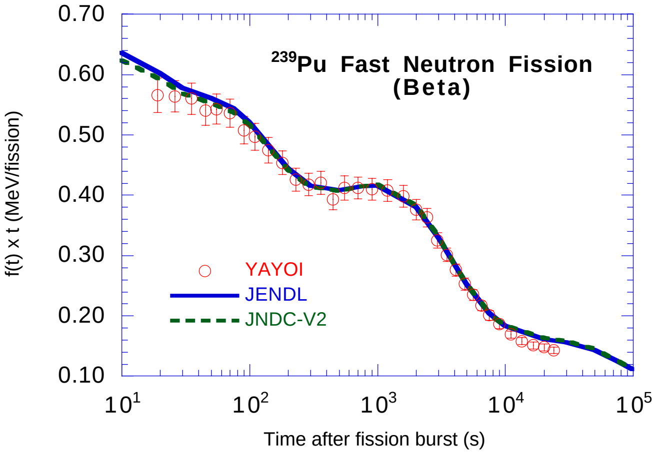
238U Fast Neutron Fission
(Gamma)
JENDL
JNDC-V2
Gamma Ratio (C/E)
Time after fission burst (s)



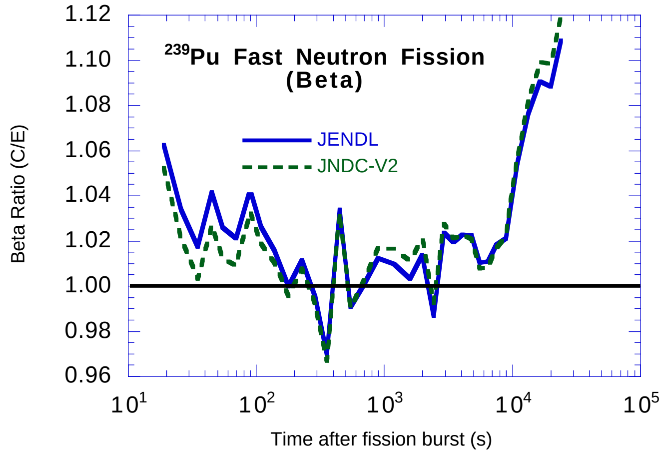
238U Fast Neutron Fission
(Total)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)



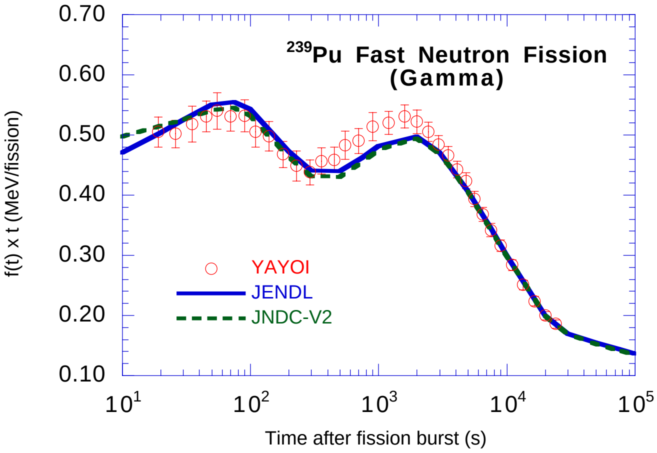
238U Fast Neutron Fission
(Total)
JENDL
JNDC-V2
Total Ratio (C/E)
Time after fission burst (s)



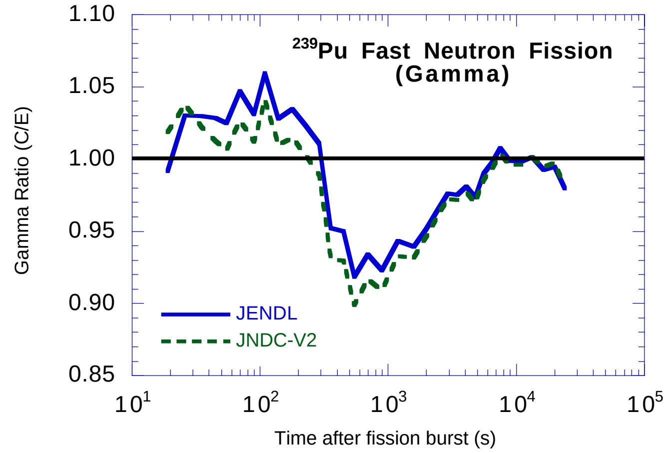
^{239}Pu Fast Neutron Fission
(Beta)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)



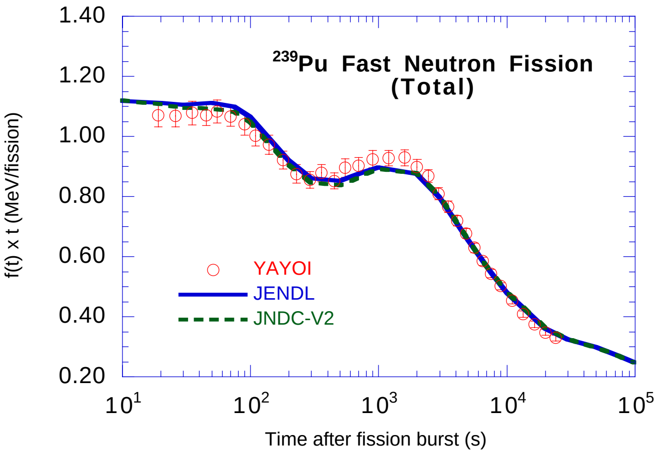
^{239}Pu Fast Neutron Fission
(Beta)
JENDL
JNDC-V2
Beta Ratio (C/E)
Time after fission burst (s)



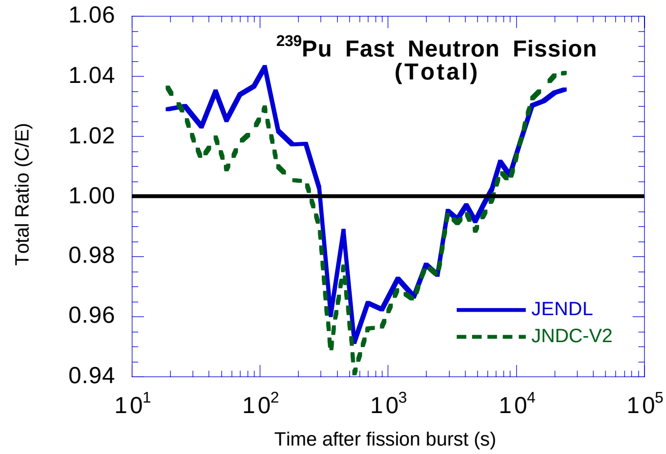
^{239}Pu Fast Neutron Fission
(Gamma)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)



^{239}Pu Fast Neutron Fission
(Gamma)
JENDL
JNDC-V2
Gamma Ratio (C/E)
Time after fission burst (s)



^{239}Pu Fast Neutron Fission
(Total)
YAYOI
JENDL
JNDC-V2
f(t) x t (MeV/fission)
Time after fission burst (s)



^{239}Pu Fast Neutron Fission
(Total)
JENDL
JNDC-V2
Total Ratio (C/E)
Time after fission burst (s)