Theoretical Study of decay heat and delayed neutron emission

- 4 year project of estimating decay heat and delayed neutron emission for U, Pu and minor actinides -

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Collaborate with:
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10th ASRC International Workshop “Nuclear Fission and Decay of Exotic Nuclei
March 21-22, 2013 (JAEA-Tokai)

Understanding global properties of nuclei

Nuclear mass region

- (Extremely) Superheavy：Decay modes, Structure of superheavy double magic nuclei ^{298}[114] and its neighboring, and beyond
- Proton-rich：N=126 neutron-deficient nuclei (Unknown peninsula) : enhancement of existence due to the closed shell
- Neutron-rich：Change of closed shell, Fission in the superheavy, r-process nucleosynthesis

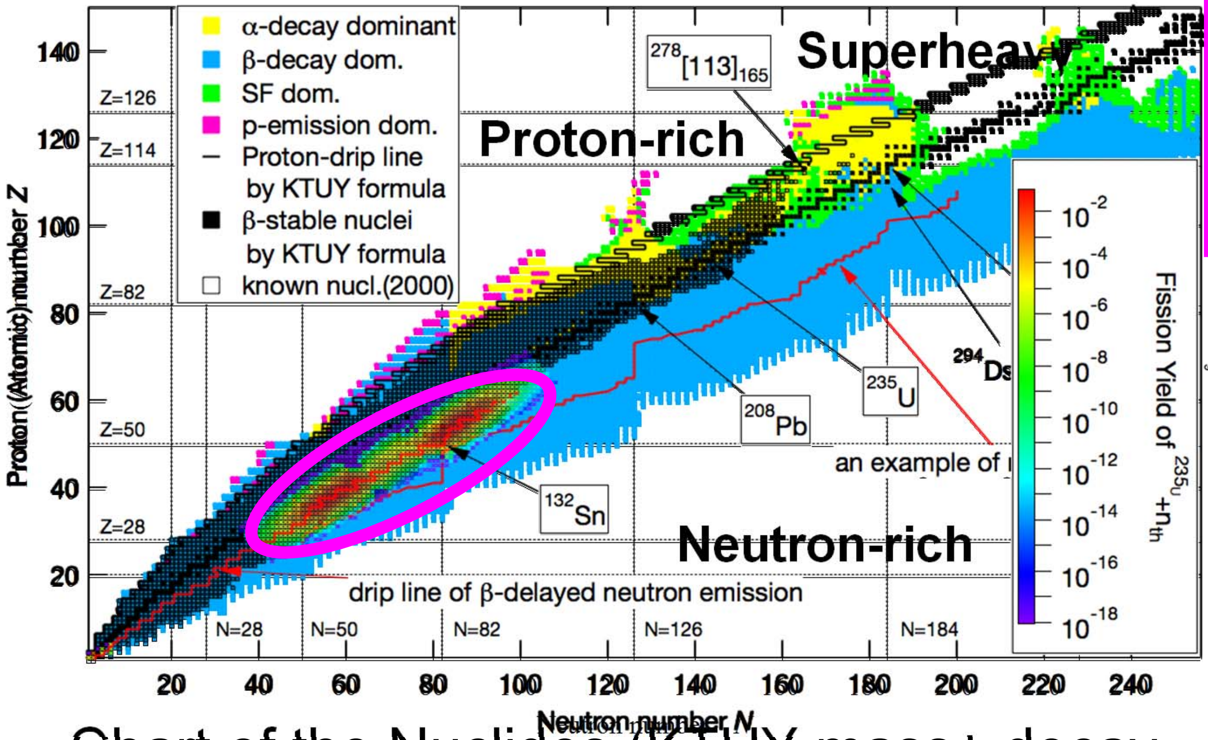
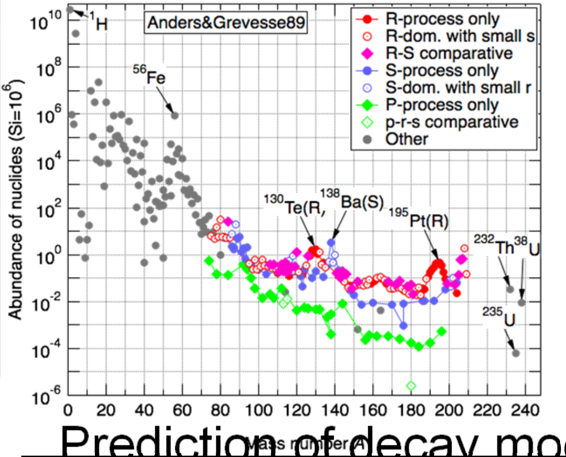


Chart of the Nuclides (KTUY mass+ decay model)

Medium-heavy n-rich region:
Astrophysics: r-process
Atomic Energy: Fission product from actinides



Prediction of decay modes of nuclei

(How far does the area of superheavy elements extend? - Decay modes of heavy and superheavy nuclei -

H. K and T. Tachibana, B. Phys.Soc. Jpn. 60, 717 (2005)

‘Development of High Precision of Delayed Neutron Rate for Evaluation of Operating Characteristic Properties of the Advanced Fast Reactors’

Entrusted project by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT)

Project Leader: **S. Chiba** (Tokyo Inst. Tech.),

Term: 2012.11-2016.3, Total budget: **200M yen (2M USD (1USD=100yen))**

Purpose: High-precision prediction of operating characteristic properties of highly-burn-up nuclear reactor and innovative nuclear reactor where minor actinides accumulate from the following way:

(1) Measurement of Fission Yield (FY) data via Surrogate reaction

(2) Construction of method for obtaining delayed neutron rate and decay heat with gross theory of beta decay

(3)Construction of theoretical method for obtaining Independent FY with Dynamical model (Two-center shell model + Langevin eq.)

(4) Nuclear Data (including verification on reactor system)

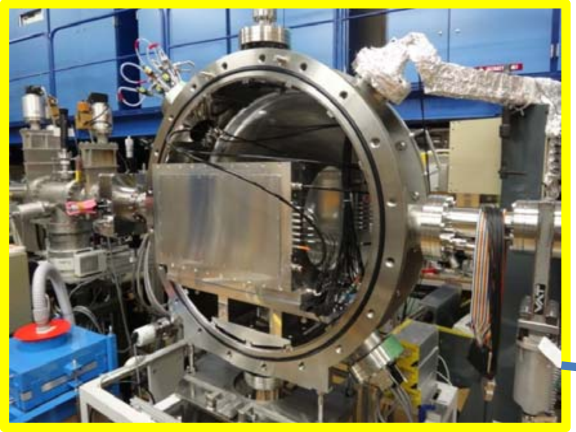
Today’s Talk

Collaborate with:
Experiment: K. Nishio, I. Nishinaka, H. Makii, T. Ishii, K. Tsukada, M. Asai, K. Furutaka
Beta-decay theory: H. Koura, Y. Utsuno
Fission theory: S. Chiba (TIT) , Y. Aritomo (TIT)
Nuclear Data and Reactor: T. Kugo, O. Iwamoto, F. Minato

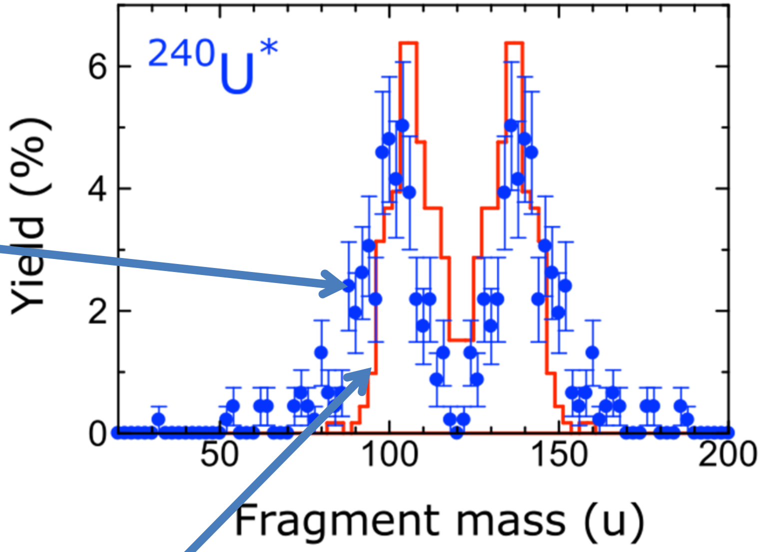
Name: Topic leader

Procedure

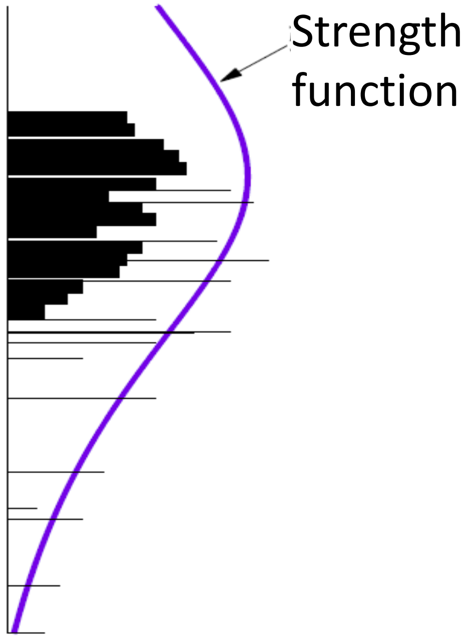
1. Surrogate reaction exp.



Fission fragment mass distr.

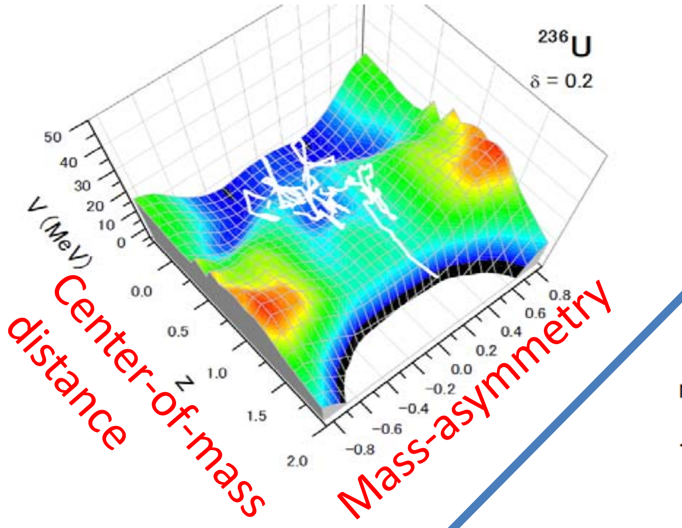


2. Gross theory of β decay



- β-ray spectra
- γ −ray spectra
- delayed neutron Yield, χ_d
- anti-neutrino spectra

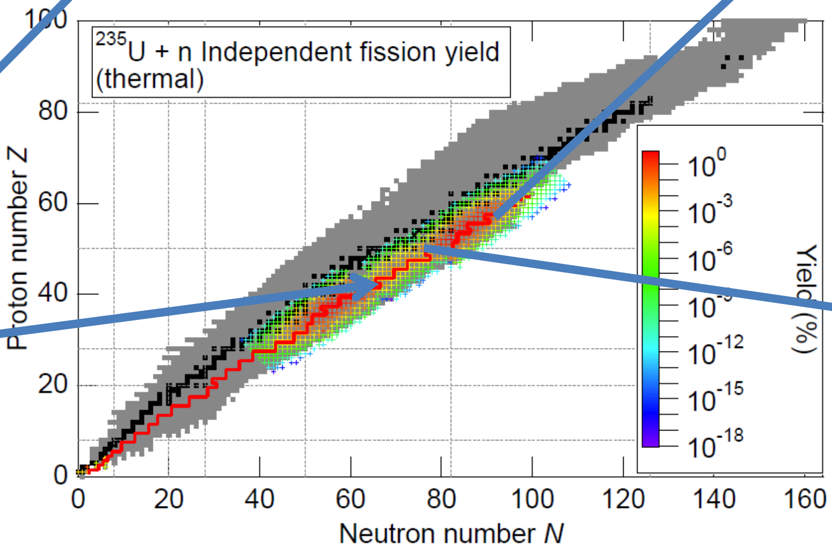
3. Multi-dimensional dynamics of Fission (Langevin) model



- FP mass-number distr.
- FP independent Yield
- prompt neutron yield

courtesy of S. Chiba

Independent Fission Fragment(N-Z distr.)



4. Library, Summation Calculation

- Analysis of decay heat data
- β_{eff} Analysis of Reactor Physics data
- Neutrino production

Nuclear β-decay

$$T_{1/2} = \frac{\ln 2}{\lambda}$$

$$\lambda = \lambda_{\mathrm{F}} + \lambda_{\mathrm{GT}} + \lambda_1^{(0)} + \lambda_1^{(1)} + \lambda_1^{(2)}$$

(up to 1st forbidden)

Trans.	Type	ΔL	Parity ch.
Allowed	Fermi	0	+
	Gamow-Teller	0,±1 (0 -×-> 0)	+
1st forbid.	non-unique 1st	0,±1	-
	unique 1st	±2	-
2nd forbid.	non-unique 2nd	±2	+
	unique 2nd	±3	+
3rd forbid.	non-unique 3rd	±3	-
	unique 3rd	±4	-

$$\lambda_{\mathrm{F}} = \frac{{m_{\mathrm{e}}}^5 c^4}{2\pi^3 \hbar^7} |g_{\mathrm{V}}|^2 \int_{-Q}^{0} |M_{\mathrm{F}}(E)|^2 f(-E) dE$$

$$\lambda_{\mathrm{GT}} = \frac{{m_{\mathrm{e}}}^5 c^4}{2\pi^3 \hbar^7} |g_{\mathrm{A}}|^2 3 \int_{-Q}^{0} |M_{\mathrm{GT}}(E)|^2 f(-E) dE$$

$$\lambda_1^{(2)} = \frac{{m_{\mathrm{e}}}^5 c^4}{2\pi^3 \hbar^7} \left(\frac{m_{\mathrm{e}} c}{\hbar}\right)^2 |g_{\mathrm{A}}|^2 \int_{-Q}^{0} \sum_{ij} |M_{ij}(E)|^2 f_1(-E) dE$$

<-- unique 1st

$$\lambda_1^{(1)} = \frac{{m_{\mathrm{e}}}^5 c^4}{2\pi^3 \hbar^7} \left(\frac{m_{\mathrm{e}} c}{\hbar}\right)^2 \left[|g_{\mathrm{V}}|^2 \int_{-Q}^{0} |M_{\boldsymbol{r}}(E)|^2 f_{1\mathrm{V}}^{(1)}(-E) dE + |g_{\mathrm{A}}|^2 \int_{-Q}^{0} |M_{\boldsymbol{\sigma} \times \boldsymbol{r}}(E)|^2 f_{1\mathrm{A}}^{(1)}(-E) dE \right]$$

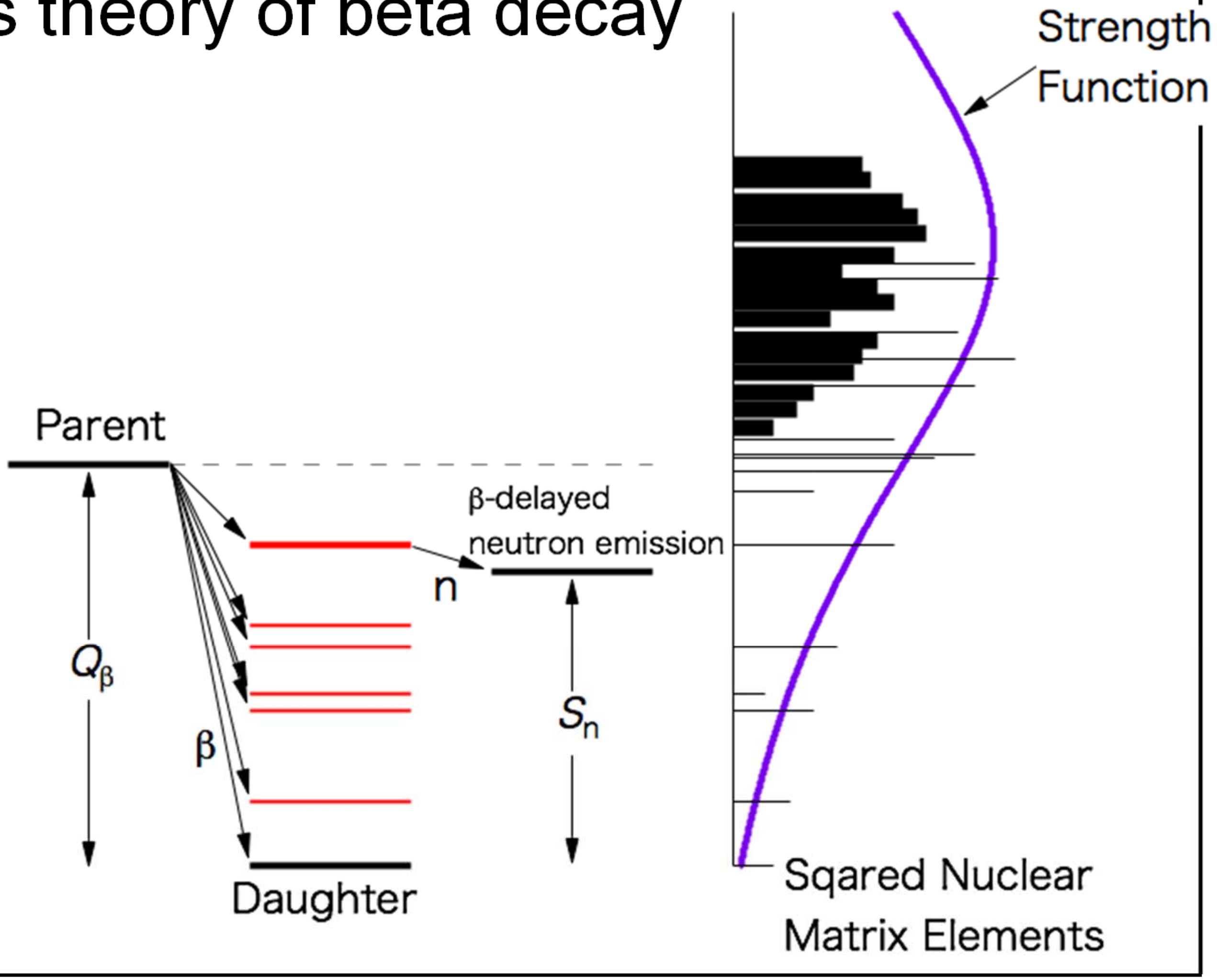
$$\lambda_1^{(0)} = \frac{{m_{\mathrm{e}}}^5 c^4}{2\pi^3 \hbar^7} \left(\frac{m_{\mathrm{e}} c}{\hbar}\right)^2 |g_{\mathrm{A}}|^2 \int_{-Q}^{0} |M_{\boldsymbol{\sigma} \cdot \boldsymbol{r}}(E)|^2 f_{1\mathrm{A}}^{(0)}(-E) dE$$

Gross theory of beta decay

$$|M_\Omega(E)|^2 = \int_{\epsilon_{\mathrm{min}}}^{\epsilon_{\mathrm{max}}} D(E,\epsilon)W(E,\epsilon)\frac{dn_1}{d\epsilon}d\epsilon$$

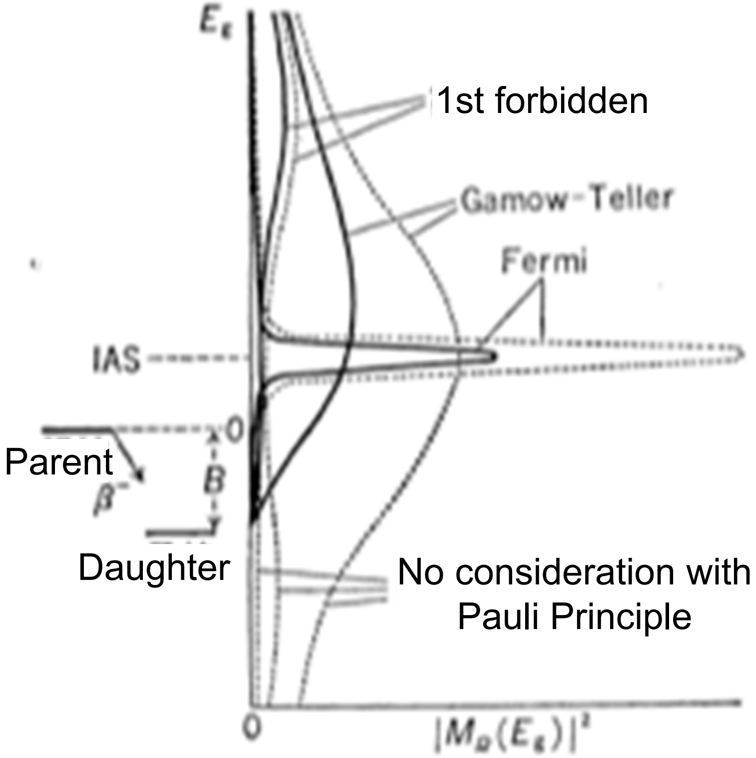
$D(E,\epsilon)$: one particle strength function

half-live, energy distribution,
delayed neutron probability

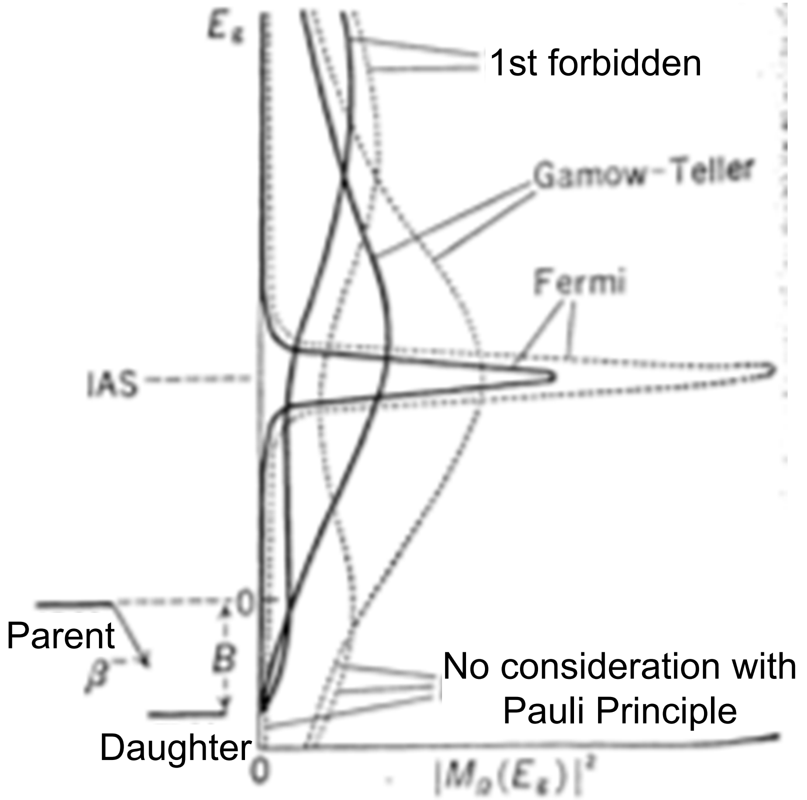


β-decay strength function

Neutron-rich side



Schematic view of β^- decay for light nuclei



Schematic view of β^- decay for heavy nuclei nuclei

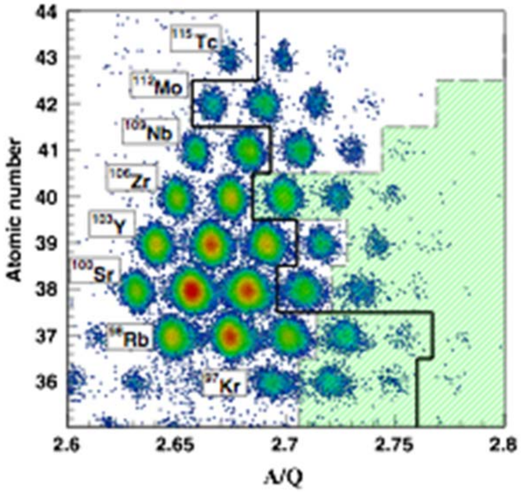
Light nuclei : G-T dominance

Heavy nuclei : Competition between GT and 1st forbidden

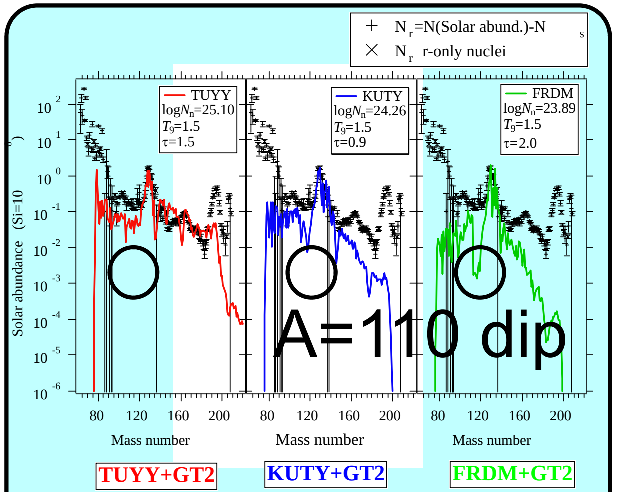
No consideration with Pauli Principle <—> $W(E,\epsilon)=0$

Half-life measurement in the n-rich nuclei at RIBF: Gross theory vs QRPA

S. Nishimura, et al. PRL106(2011)



Nuclide Identification
Left from Black line: nuclei with known half-lives
Filled green: part of the r-process path (WP Approx.)



H. Koura et al.

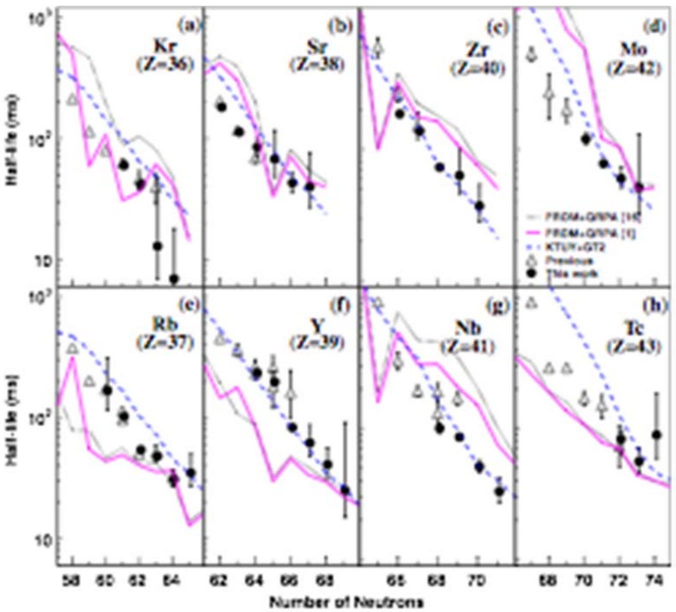


FIG. 3 (color online). Neutron number dependence of β-decay half-lives for (top) even-Z (a) Kr, (b) Sr, (c) Zr, and (d) Mo, and (bottom) odd-Z (e) Rb, (f) Y, (g) Nb, and (h) Tc. Filled circles and open triangles represent results from the present work and previous studies, respectively. The respective solid and dotted lines are predictions from the FRDM + QRPA models, while the dashed lines are from the KTUY + GT2.

Absolute Comparison of $T_{1/2}$

Pink : FRDM+QRPA
Dashed blue : KTUY+GT2

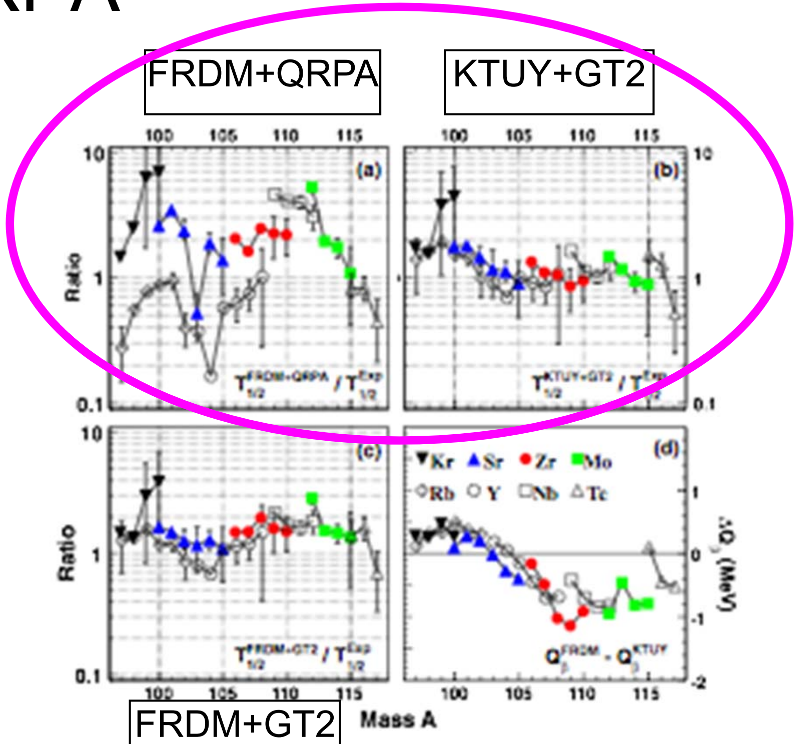


FIG. 4 (color online). Mass number dependence of the ratio of theoretical $T_{1/2}$ values from (a) FRDM + QRPA [16], (b) KTUY + GT2 [17,18], and (c) FRDM + GT2, to the experimental values deduced in the present work. (d) The difference between Q_β values predicted by the FRDM and KTUY mass formulas.

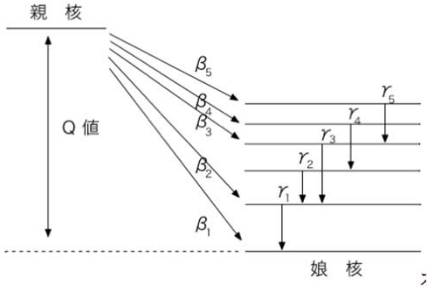
Ratio of theoretical $T_{1/2}$ to exp.

FRDM+QRPA : rather large discrepancy due to QRPA
KTUY+GT2 : rather good reproduction

Decay Heat and Delayed neutron emission

Decay heat :

Fission products (FP) decays β-decay and γ decay. These sum of each heats is decay heat.

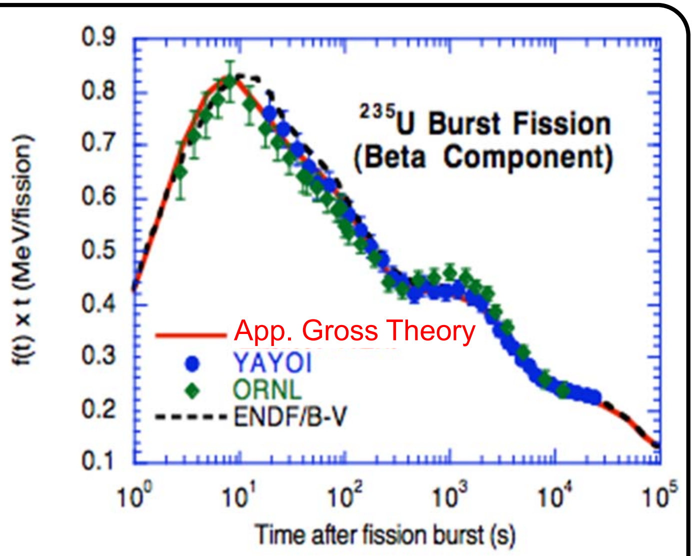


$$f(t) = \sum_i \lambda_i \cdot \overline{E_t^i} \cdot N_i(t)$$
$$= \sum_i \lambda_i \cdot (\overline{E_\beta^i} + \overline{E_\gamma^i}) \cdot N_i(t)$$

$$\overline{E_\gamma} = \sum_j E_\gamma^j I_\gamma^j / \text{number}$$

$$\overline{E_\beta} = \sum_j E_\beta^j I_\beta^j / \text{number}$$

$N_i(t)$:Production Yield at time t



Delayed neutron rate: From fission theory and exp.

Thoery: $\lambda_n = \sum_\Omega \int_{-Q+S_n}^{0} \frac{\Gamma_n}{\Gamma_n + \Gamma_\gamma} \lambda_\Omega(E) dE$,

Ω:Type of β decay (Fermi, Gamow-Teller, forbidden,...)

Exp.: $\lambda_n^{\exp} = \ln 2 \, P_n^{\exp} / t_{1/2}^{\exp}$,

P_n:Delayed neutron probability

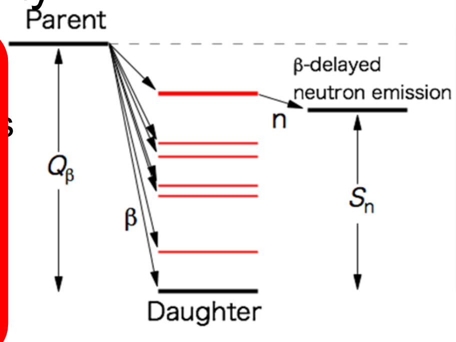
Delayed neutron yield: $\overline{\nu_\mathrm{d}} = \sum_i P_{\mathrm{n}i} Y_i$

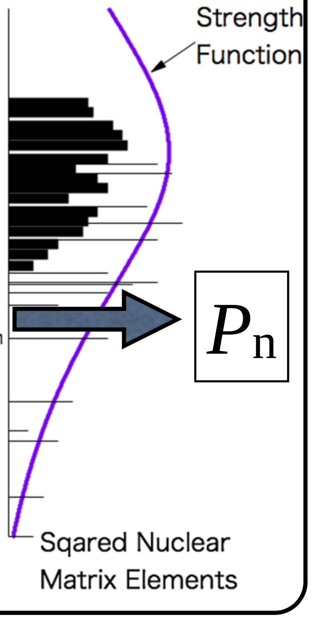
Y:Cumulative Fission Yield

i:all delayed neutron precursors

Time-dependency of delayed neutron emission : $n_\mathrm{d}(t) = \sum_i P_{\mathrm{n}i} \lambda_i Y_i(t)$

(6-Group Approx.: $n_{\mathrm{d}6}(t) = \overline{\nu_\mathrm{d}} \sum_{i=1}^{6} \alpha_i \lambda_i \exp(-\lambda_i t)$





Both phenomena accompany β decay.

Application of Gross theory of β-decay to atomic energy

1. Delayed neutron summation calculation

$$\nu_{\mathrm{d}} = \sum_{\mathrm{allFP}} P_{\mathrm{n}} Y_{\mathrm{Cum}}$$

M.C. Brady and T.R. England, NST **103**,

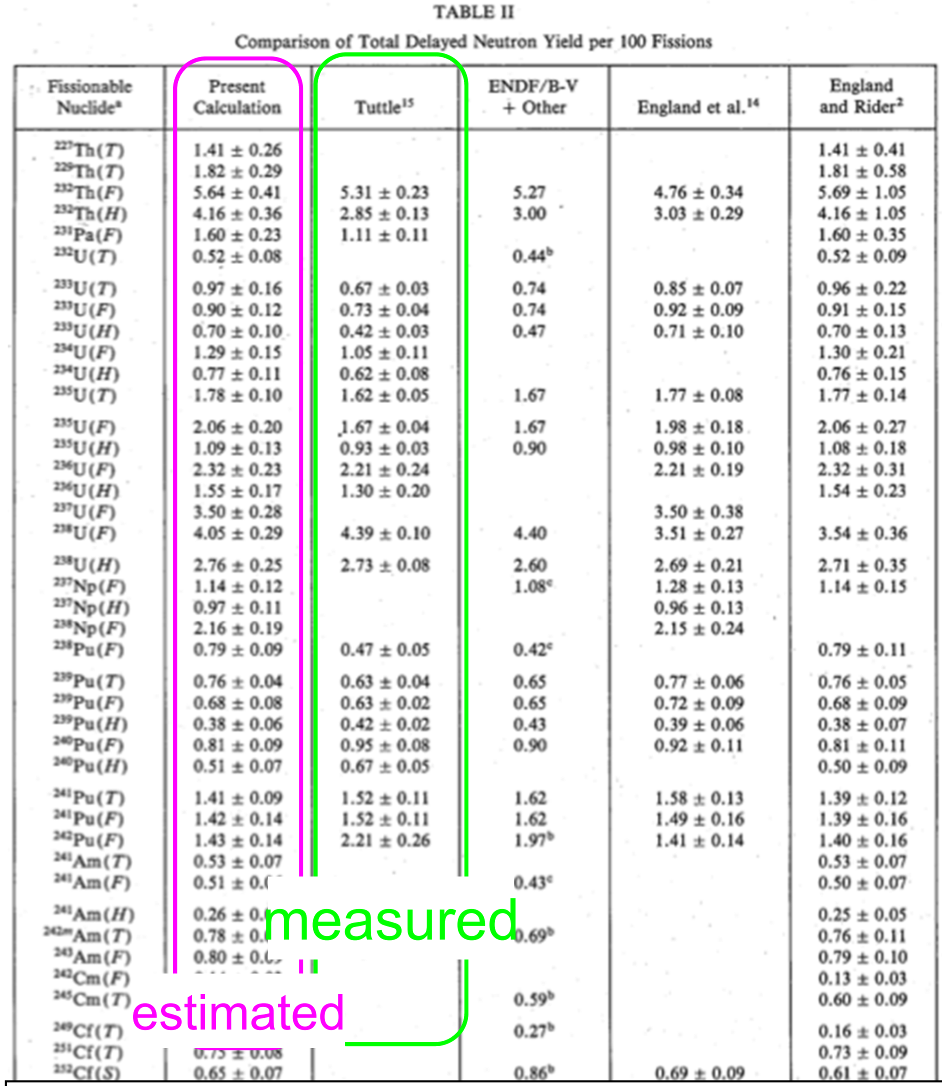


TABLE II

Comparison of Total Delayed Neutron Yield per 100 Fissions

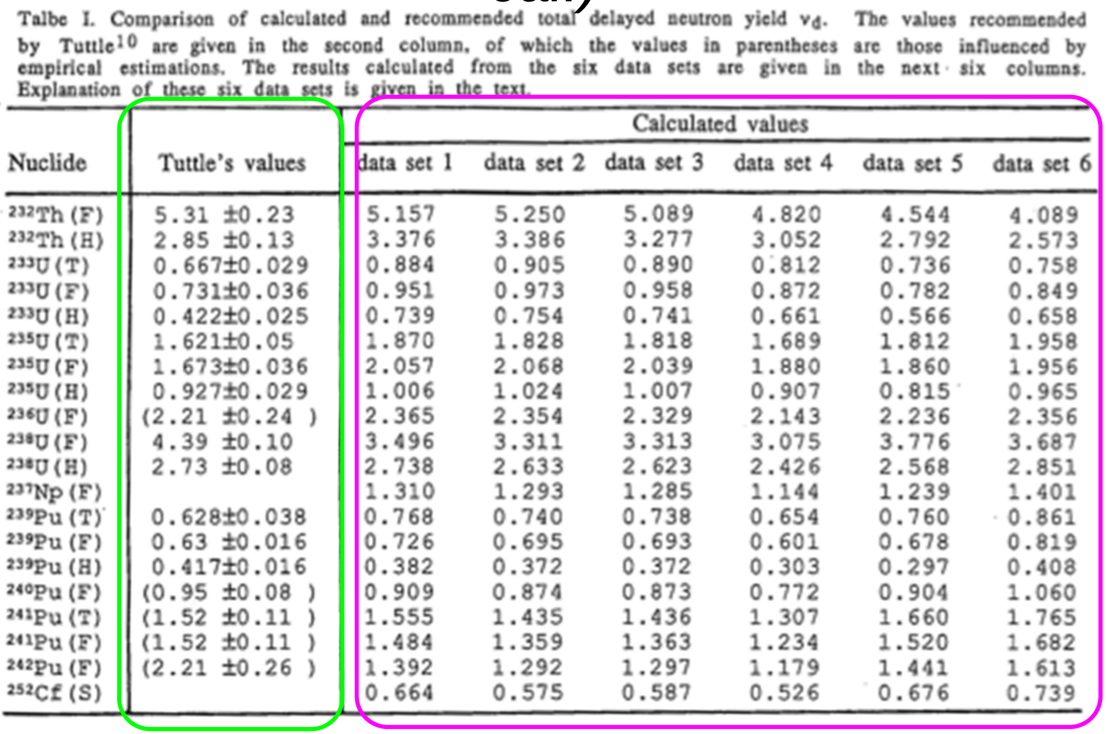
Fissionable Nuclide[a]	Present Calculation	Tuttle[15]	ENDF/B-V + Other	England et al.[14]	England and Rider[2]
^{227}Th(T)	1.41 ± 0.26				1.41 ± 0.41
^{229}Th(T)	1.82 ± 0.29				1.81 ± 0.58
^{232}Th(F)	5.64 ± 0.41	5.31 ± 0.23	5.27	4.76 ± 0.34	5.69 ± 1.05
^{232}Th(H)	4.16 ± 0.36	2.85 ± 0.13	3.00	3.03 ± 0.29	4.16 ± 1.05
^{231}Pa(F)	1.60 ± 0.23	1.11 ± 0.11			1.60 ± 0.35
^{232}U(T)	0.52 ± 0.08		0.44[b]		0.52 ± 0.09
^{233}U(T)	0.97 ± 0.16	0.67 ± 0.03	0.74	0.85 ± 0.07	0.96 ± 0.22
^{233}U(F)	0.90 ± 0.12	0.73 ± 0.04	0.74	0.92 ± 0.09	0.91 ± 0.15
^{233}U(H)	0.70 ± 0.10	0.42 ± 0.03	0.47	0.71 ± 0.10	0.70 ± 0.13
^{234}U(F)	1.29 ± 0.15	1.05 ± 0.11			1.30 ± 0.21
^{234}U(H)	0.77 ± 0.11	0.62 ± 0.08			0.76 ± 0.15
^{235}U(T)	1.78 ± 0.10	1.62 ± 0.05	1.67	1.77 ± 0.08	1.77 ± 0.14
^{235}U(F)	2.06 ± 0.20	1.67 ± 0.04	1.67	1.98 ± 0.18	2.06 ± 0.27
^{235}U(H)	1.09 ± 0.13	0.93 ± 0.03	0.90	0.98 ± 0.10	1.08 ± 0.18
^{236}U(F)	2.32 ± 0.23	2.21 ± 0.24		2.21 ± 0.19	2.32 ± 0.31
^{236}U(H)	1.55 ± 0.17	1.30 ± 0.20			1.54 ± 0.23
^{237}U(F)	3.50 ± 0.28			3.50 ± 0.38	
^{238}U(F)	4.05 ± 0.29	4.39 ± 0.10	4.40	3.51 ± 0.27	3.54 ± 0.36
^{238}U(H)	2.76 ± 0.25	2.73 ± 0.08	2.60	2.69 ± 0.21	2.71 ± 0.35
^{237}Np(F)	1.14 ± 0.12		1.08[c]	1.28 ± 0.13	1.14 ± 0.15
^{237}Np(H)	0.97 ± 0.11			0.96 ± 0.13	
^{238}Np(F)	2.16 ± 0.19			2.15 ± 0.24	
^{238}Pu(F)	0.79 ± 0.09	0.47 ± 0.05	0.42[c]		0.79 ± 0.11
^{239}Pu(T)	0.76 ± 0.04	0.63 ± 0.04	0.65	0.77 ± 0.06	0.76 ± 0.05
^{239}Pu(F)	0.68 ± 0.08	0.63 ± 0.02	0.65	0.72 ± 0.09	0.68 ± 0.09
^{239}Pu(H)	0.38 ± 0.06	0.42 ± 0.02	0.43	0.39 ± 0.06	0.38 ± 0.07
^{240}Pu(F)	0.81 ± 0.09	0.95 ± 0.08	0.90	0.92 ± 0.11	0.81 ± 0.11
^{240}Pu(H)	0.51 ± 0.07	0.67 ± 0.05			0.50 ± 0.09
^{241}Pu(T)	1.41 ± 0.09	1.52 ± 0.11	1.62	1.58 ± 0.13	1.39 ± 0.12
^{241}Pu(F)	1.42 ± 0.14	1.52 ± 0.11	1.62	1.49 ± 0.16	1.39 ± 0.16
^{242}Pu(F)	1.43 ± 0.14	2.21 ± 0.26	1.97[b]	1.41 ± 0.14	1.40 ± 0.16
^{241}Am(T)	0.53 ± 0.07				0.53 ± 0.07
^{241}Am(F)	0.51 ± 0.0		0.43[c]		0.50 ± 0.07
^{241}Am(H)	0.26 ± 0.				0.25 ± 0.05
^{242m}Am(T)	0.78 ± 0.		0.69[b]		0.76 ± 0.11
^{243}Am(F)	0.80 ± 0.				0.79 ± 0.10
^{242}Cm(F)	[illegible]				0.13 ± 0.03
^{245}Cm(T)	[illegible]		0.59[b]		0.60 ± 0.09
^{249}Cf(T)	[illegible]		0.27[b]		0.16 ± 0.03
^{251}Cf(T)	0.75 ± 0.08				0.73 ± 0.09
^{252}Cf(S)	0.65 ± 0.07		0.86[b]	0.69 ± 0.09	0.61 ± 0.07

measured

estimated

T. Tachibana, M. Yamada et al. (NDST 1988) 885

(First apply of Gross theory for summation cal.)



Talbe I. Comparison of calculated and recommended total delayed neutron yield ν_d. The values recommended by Tuttle[10] are given in the second column, of which the values in parentheses are those influenced by empirical estimations. The results calculated from the six data sets are given in the next six columns. Explanation of these six data sets is given in the text.

Nuclide	Tuttle's values	Calculated values					
		data set 1	data set 2	data set 3	data set 4	data set 5	data set 6
^{232}Th (F)	5.31 ±0.23	5.157	5.250	5.089	4.820	4.544	4.089
^{232}Th (H)	2.85 ±0.13	3.376	3.386	3.277	3.052	2.792	2.573
^{233}U (T)	0.667±0.029	0.884	0.905	0.890	0.812	0.736	0.758
^{233}U (F)	0.731±0.036	0.951	0.973	0.958	0.872	0.782	0.849
^{233}U (H)	0.422±0.025	0.739	0.754	0.741	0.661	0.566	0.658
^{235}U (T)	1.621±0.05	1.870	1.828	1.818	1.689	1.812	1.958
^{235}U (F)	1.673±0.036	2.057	2.068	2.039	1.880	1.860	1.956
^{235}U (H)	0.927±0.029	1.006	1.024	1.007	0.907	0.815	0.965
^{236}U (F)	(2.21 ±0.24)	2.365	2.354	2.329	2.143	2.236	2.356
^{238}U (F)	4.39 ±0.10	3.496	3.311	3.313	3.075	3.776	3.687
^{238}U (H)	2.73 ±0.08	2.738	2.633	2.623	2.426	2.568	2.851
^{237}Np (F)		1.310	1.293	1.285	1.144	1.239	1.401
^{239}Pu (T)	0.628±0.038	0.768	0.740	0.738	0.654	0.760	0.861
^{239}Pu (F)	0.63 ±0.016	0.726	0.695	0.693	0.601	0.678	0.819
^{239}Pu (H)	0.417±0.016	0.382	0.372	0.372	0.303	0.297	0.408
^{240}Pu (F)	(0.95 ±0.08)	0.909	0.874	0.873	0.772	0.904	1.060
^{241}Pu (T)	(1.52 ±0.11)	1.555	1.435	1.436	1.307	1.660	1.765
^{241}Pu (F)	(1.52 ±0.11)	1.484	1.359	1.363	1.234	1.520	1.682
^{242}Pu (F)	(2.21 ±0.26)	1.392	1.292	1.297	1.179	1.441	1.613
^{252}Cf (S)		0.664	0.575	0.587	0.526	0.676	0.739

measured estimated

Error in summation cal.:order of 10%

Goal: High precise reproduction for U, Pu, and reliable prediction for minor actinides (also energy dependency)

Pandemonium Problem (Decay Heat)

2. Decay heat

$$f(t) = \sum_i \lambda_i \cdot \overline{E_{\mathrm{t}}^i} \cdot N_i(t)$$

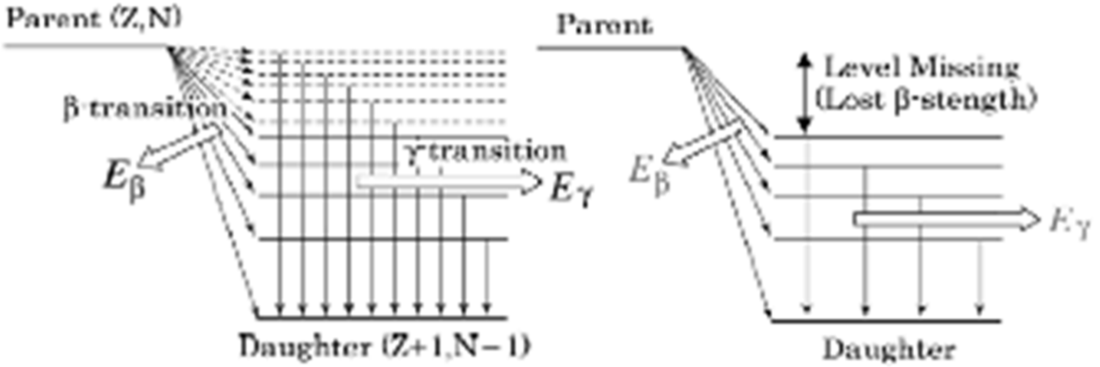
$$= \sum_i \lambda_i \cdot (\overline{E_\beta^i} + \overline{E_\gamma^i}) \cdot N_i(t)$$

λ_i : Decay constant of nuclide i

$\overline{E_{\mathrm{x}}^i}$: Average energy /one decay

$x = \beta, \gamma, \mathrm{total}$

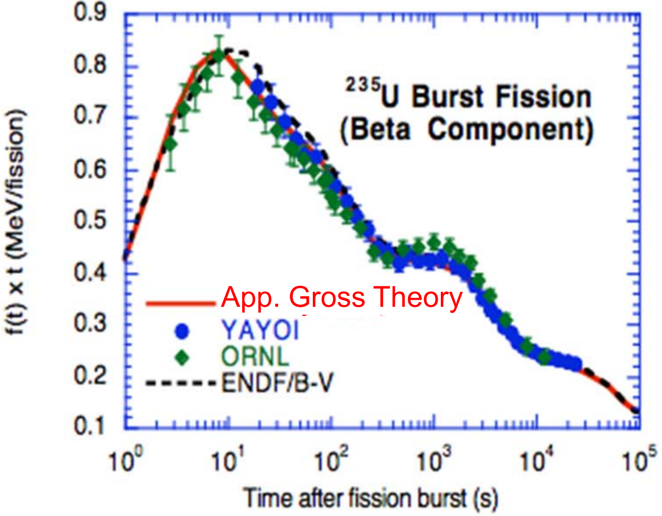
$N_i(t)$: Production Yield at time t

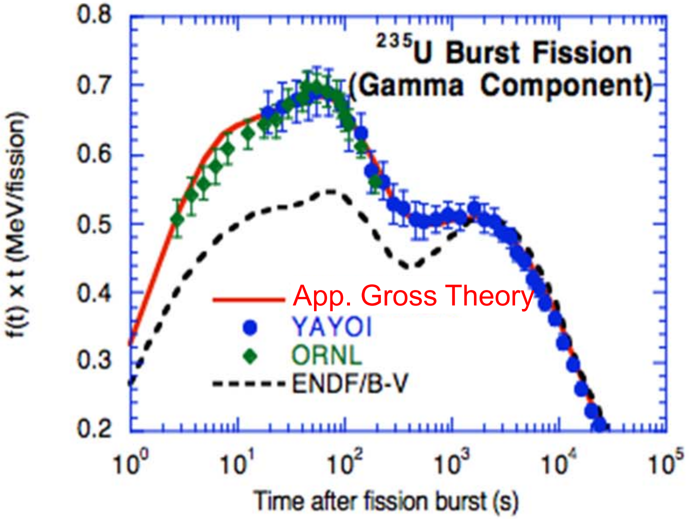


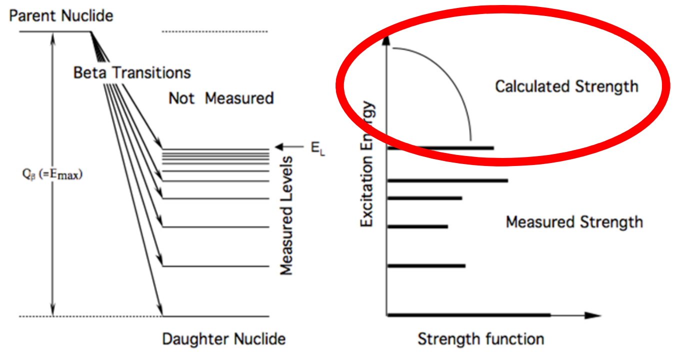
Real (correct) Eβ, Eγ

Actual measurements of Eβ, Eγ
(Eβ: overestimated, Eγ:underestimated)

'Virtual' nuclide with such a incomplete decay scheme is referred to as 'Pandemonium.'







'ENDF' in Fig. :Theory is applied for only unmeasured nuclides

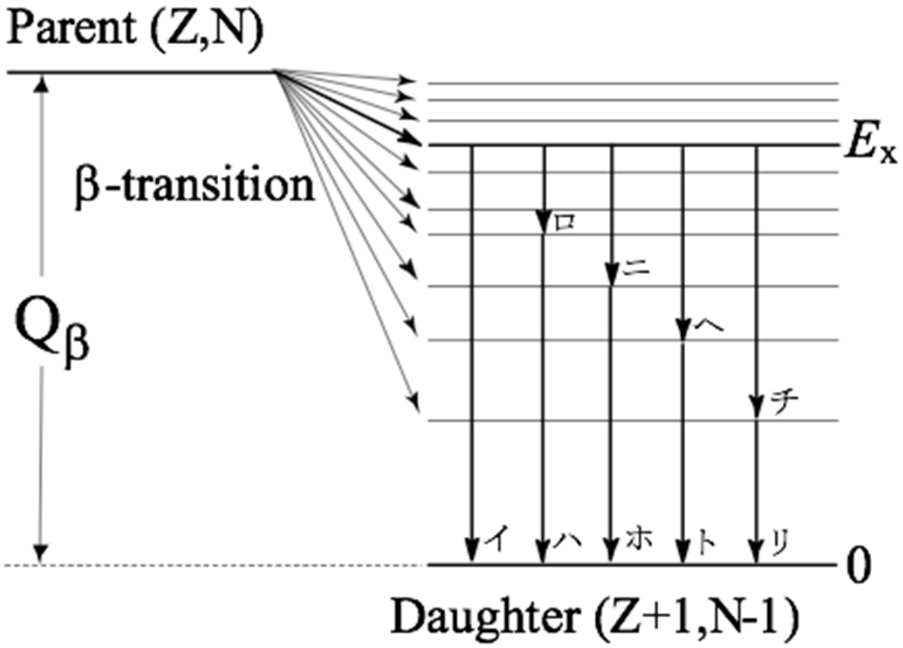
'App. Gross Theory' in Fig. : Gross Theory is applied for not only unmeasured nuclides but also measured nuclides

J. Katakura

- Lack of energy levels effects results of summation calc. in decay heat : Importance of nuclear structure

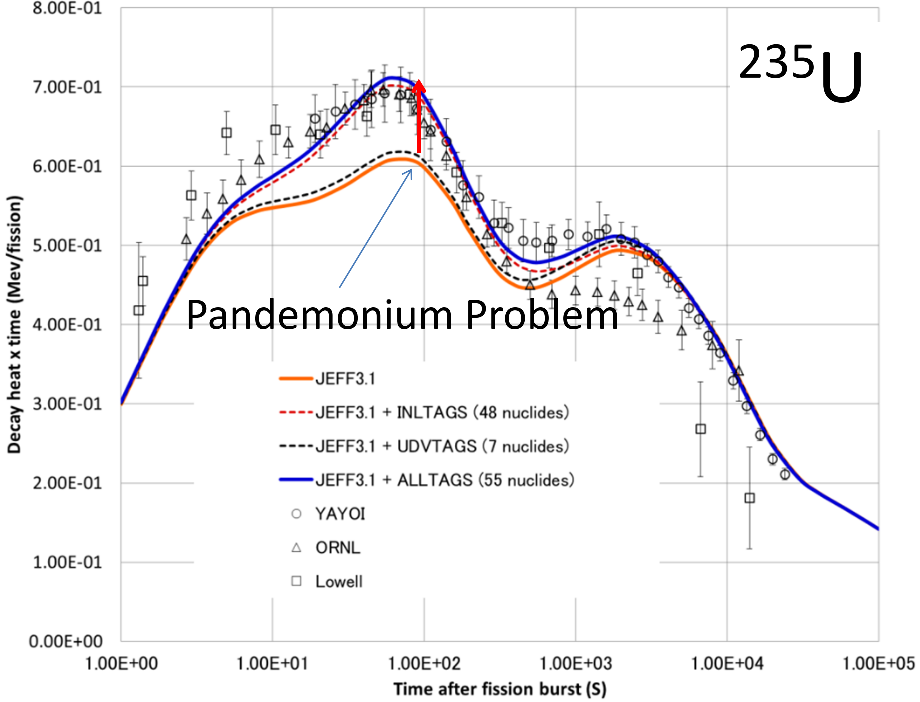
TAGS: Total Absorption Gamma-ray Spectrometer

R.C. Greenwood, Idaho National Engineering and Environmental Laboratory



Taken from NDN 99, p14, T. Yoshida

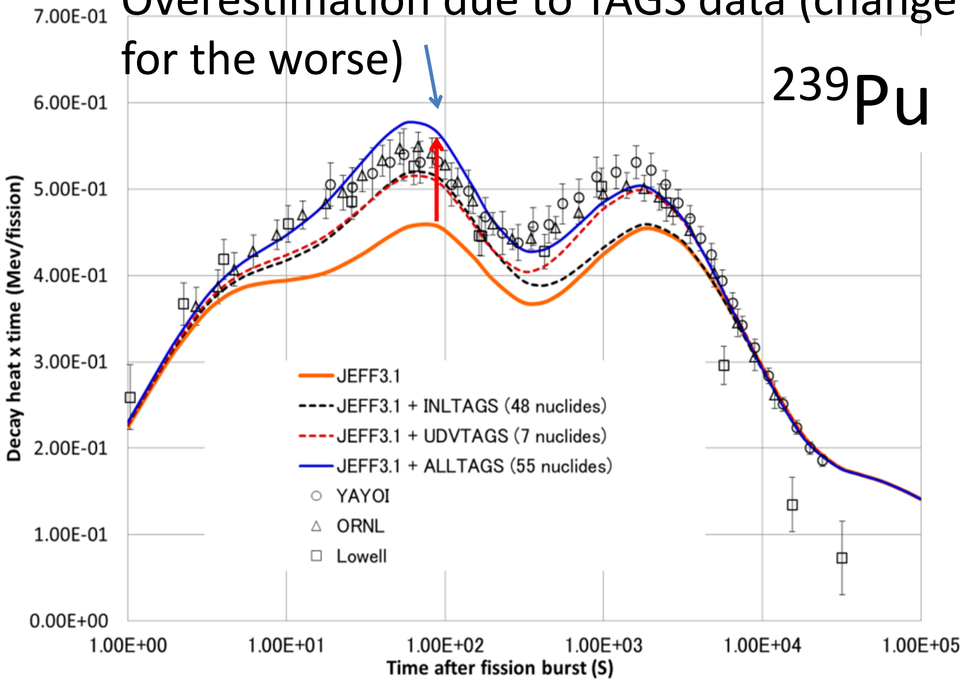
time evolution of γ-ray heat



^{235}U

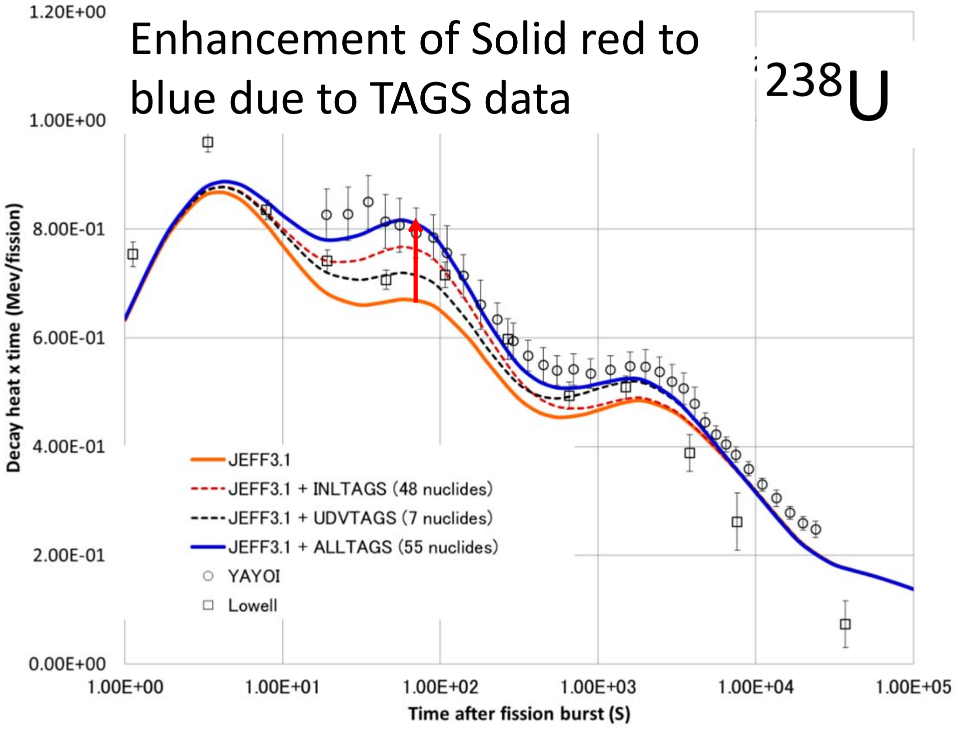
Pandemonium Problem

Overestimation due to TAGS data (change for the worse)



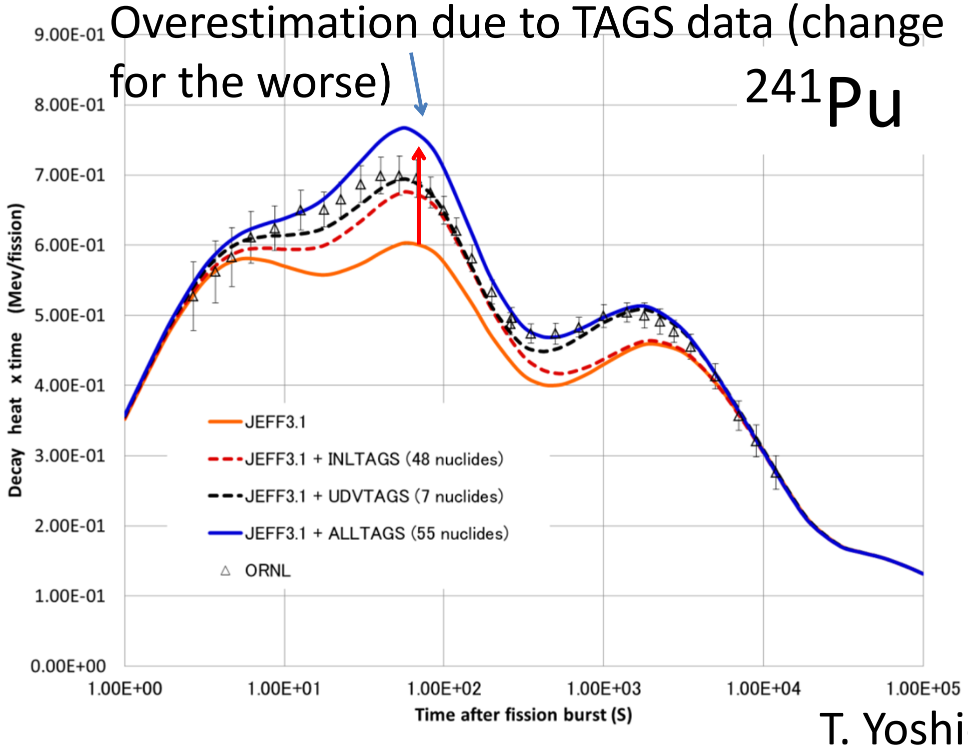
^{239}Pu

Enhancement of Solid red to blue due to TAGS data



^{238}U

Overestimation due to TAGS data (change for the worse)

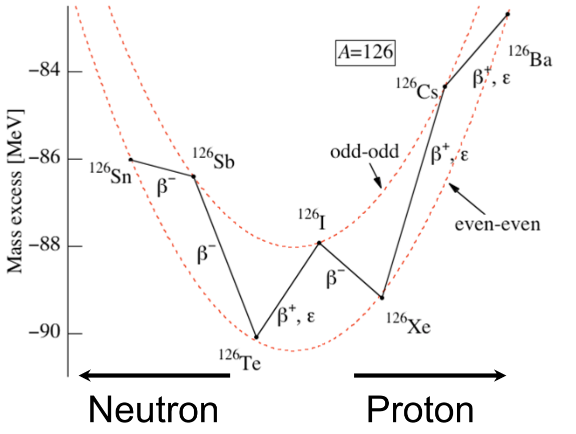


^{241}Pu

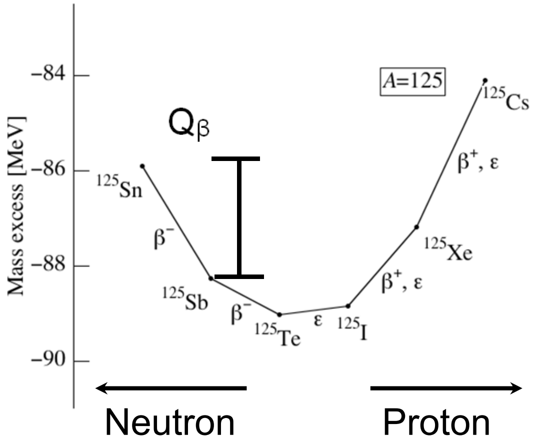
T. Yoshida

Improvement of Gross theory for half-life, neutron emission and decay heat (in progress)

- Spin-parity of g.s. odd-odd
  - (odd-odd nuclei (J^-, J is large in most case) → even-even nuclei (0^+))
  - cf. H. Nakata, T. Tachibana and M. Yamada, NPA594 (1995): only for half-lives
- Spin-parity dependency of low-energy excited states
- Consideration of sum rules of one-particle strength function in high-energy part
  - (Analysis of ^{90}Zr(p,n): broad distribution of strength function at 50MeV)
  - ...



even-even, odd-odd



odd-A

● Database

β decay half-life
delayed neutron prob.
spin-parity
← extracted ← ENSDF (May 2012 version)

Decay heat
← extracted ← Lowell(U-235, etc (3 nucl.)
Oak Ridge (3 nucl.)
Yayoi (Univ. Tokyo) (3 nucl.)

TAGS data
← extracted ← collab. with T. Yoshida (TIT)

Conclusion

- From this late fiscal year, we start a project for 3.5 year, related to delayed neutron and decay heat based on nuclear theory and experiment.
- We will develop a comprehensive code to calculate beta-decay, delayed neutron emission and decay heat, etc.
- Through this work, we will apply to understanding nuclear structure and decay, and will also apply to nuclear astrophysics as the r-process nucleosynthesis.