IAEA
International Atomic Energy Agency
Atoms for Peace and Development

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Radioactive Decay Calculations from Independent Fission Product Yields

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2. Tokyo Institute of Technology
3. Uppsala University

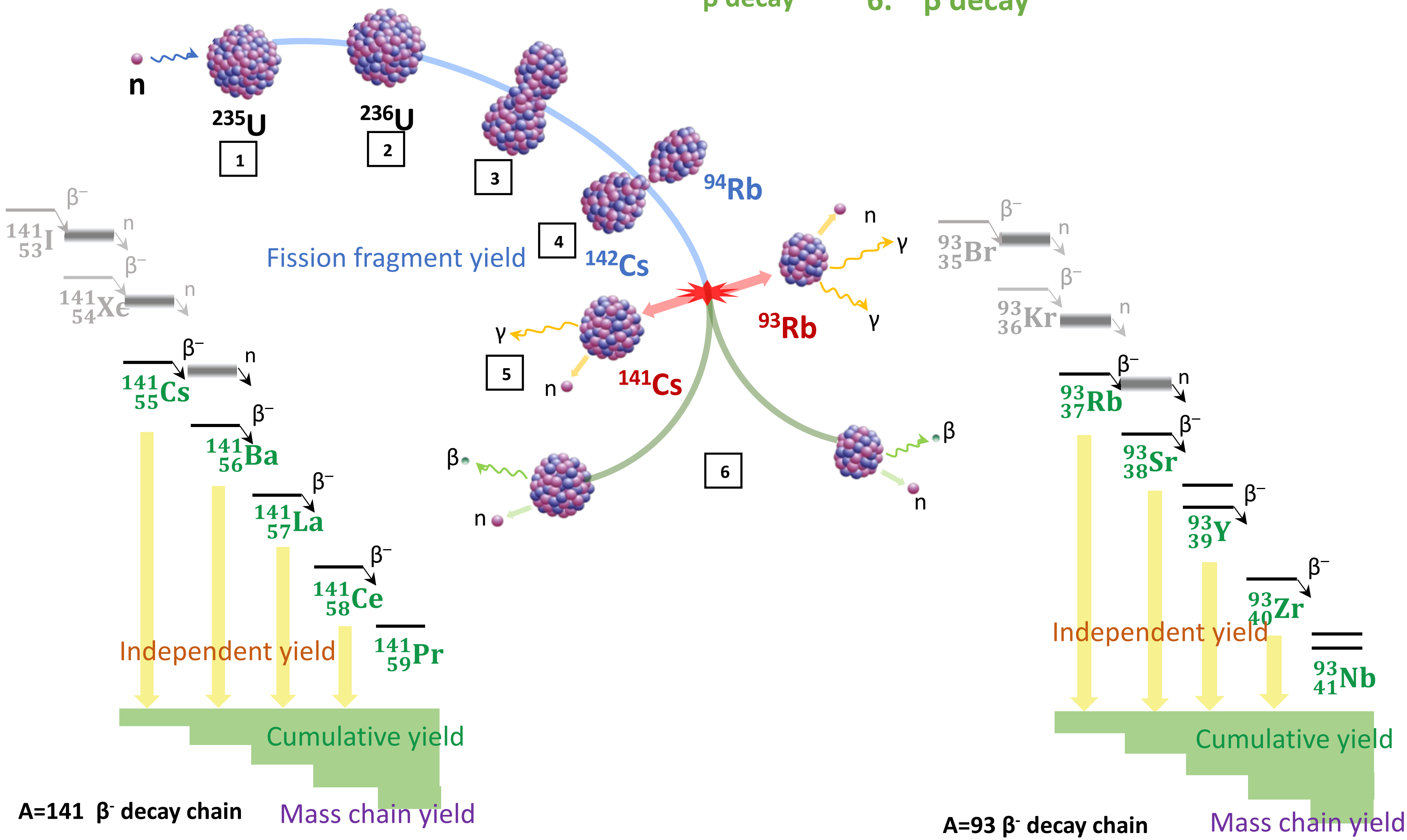
Introduction

- Nuclear fission, 3 different physics

Fission physics
1. Capture neutron by ^{235}U
2. Compound nucleus formation of ^{236}U
3. Deformation and elongation
4. Scission

Prompt decay
5. **Prompt n, γ release**

β decay
6. **β decay**



Contents

Required consistencies among fission observables

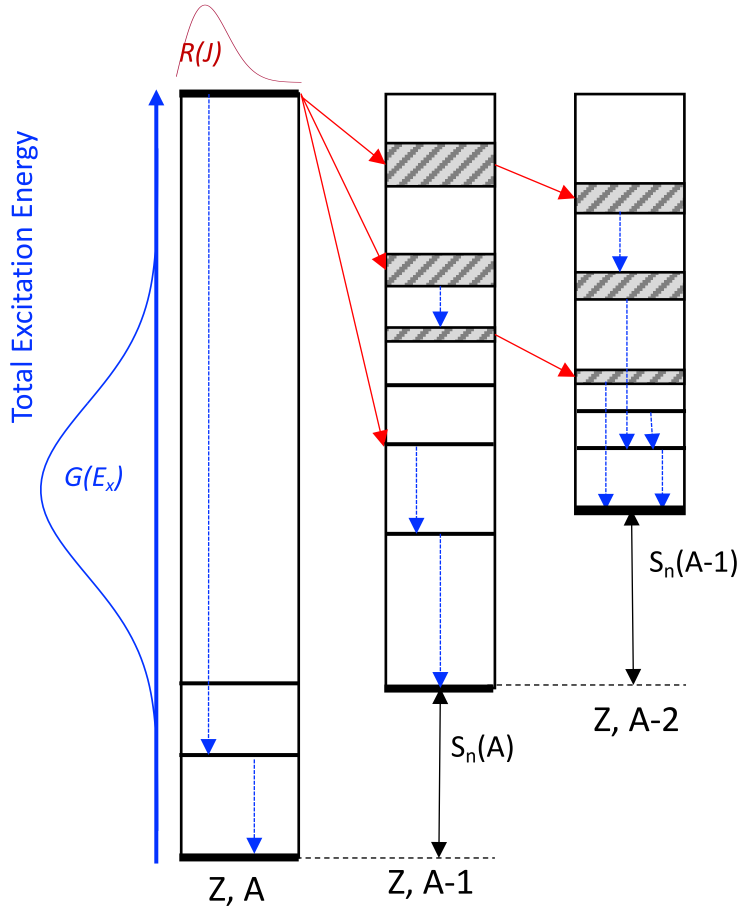
- Independent fission product yield (MF8 MT454)
  - Number of beta-delayed neutron emissions (MF1 MT455) via Decay data
  - Cumulative fission product yield via Decay data
  - Decay heat via Decay data
- Cumulative fission product yield(MF8 MT459)
  - Decay data
  - Independent fission product yield (MF8 MT454)
  - Number of beta-delayed neutron emissions (MF1 MT455)
- Number of total neutron emissions (MF1 MT452)
  - Number of prompt neutron emissions (MF1 MT456)
  - Prompt neutron spectrum (MF5 MT18)
  - Number of beta-delayed neutron emissions (MF1 MT455)
    - Beta-delayed neutron emission probabilities (branching ratio) in the decay data

To ensure consistencies among fission observables, it is important to calculate from fission fragment to beta decay in one flow.

Identifying Inconsistencies in Fission Product Yield Evaluations with Prompt Neutron Emission
P. Jaffke, *J. Nucl. Sci. Eng.*,190,258-270 (2018) .

Deterministic way of Hauser-Feshbach statistical decay of fission fragments in HF³D model

- Treating a primary fission fragment as a compound nucleus.
- Generating distributions of primary fission fragment characterized by $Y(Z, A, E_{ex}, J^{\Pi})$ and integrating deterministically for all primary fission fragment pairs (no Monte Carlo sampling required).



Neutron emission multiplicity

$$\overline{\nu}^{(k)}_{l,h} = \int dE_x \sum_{J\Pi} \int d\epsilon\ R(J,\Pi) G(E_x) \phi^{(k)}_{l,h}(J,\Pi,E_x,\epsilon)$$

- Distribution of excitation energy, *G(E)*

$$G(E_x) = \frac{1}{\sqrt{2\pi}\delta_{l,h}} \exp\left\{ -\frac{(E_x - E_{l,h})^2}{2\delta^2_{l,h}} \right\}$$

$$\delta_{l,h} = \frac{\delta_{\mathrm{TXE}}}{\sqrt{E_l^2 + E_h^2}} E_{l,h}$$

- Probability of nucleus having the state of spin *J* and parity *Π*, *R(J, Π)*

$$R(J,\Pi) = \frac{J+1/2}{2f^2\sigma^2(U)} \exp\left\{ -\frac{(J+1/2)^2}{2f^2\sigma^2(U)} \right\}$$

$\sigma^2(U)$: spin cut-off parameter
U: Excitation energy
f: scaling factor

HF³D Model:
Okumura et al. *J. Nucl. Sci. Technol.*,55,1009-1023,(2018)

Hauser-Feshbach calculations implemented in TALYS

Prompt decay

1. Read fission fragment distribution file

$$Y_{ff}(Z, A, E_{ex}, J, \Pi)$$

Y_{ff}: fragment yield
Z: fragment charge
A: framgment mass

E_{ex}: Excitation energy
J: spin
Π : parity

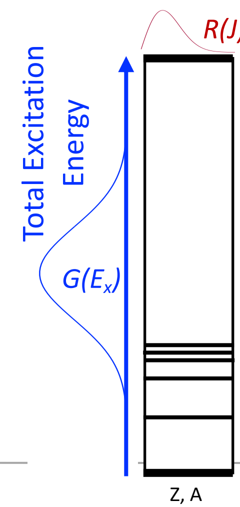
```
# Z          =   92
# A          =  236          Fissioning system
# Ex (MeV)   = 6.55e+00
```

# Zl	Al	Zh	Ah	**Yield**	TKE[MeV]	TXE[MeV]	**El[MeV]**	**Wl[MeV]**	**Eh[MeV]**	**Wh[MeV]**
30	74	62	162	**3.0000e-06**	1.4373e+02	2.5550e+01	**9.7733e+00**	**1.7875e+00**	**1.5777e+01**	**5.7377e+00**
30	75	62	161	**1.1000e-05**	1.4544e+02	2.2786e+01	**5.7400e+00**	**3.1184e+00**	**1.7046e+01**	**6.9337e+00**
31	75	61	161	**2.0000e-06**	1.4076e+02	2.6930e+01	**5.7300e+00**	**5.5013e+00**	**2.1200e+01**	**2.4042e+00**
30	76	62	160	**5.0000e-05**	1.4532e+02	2.6206e+01	**6.1234e+00**	**2.4469e+00**	**2.0082e+01**	**6.5358e+00**
				:						
46	117	46	119	**9.0000e-05**	1.6262e+02	3.4208e+01	**1.7155e+01**	**8.5389e+00**	**1.7053e+01**	**7.6726e+00**
47	117	45	119	**7.0000e-06**	1.6130e+02	3.2700e+01	**1.9750e+01**	**7.0597e+00**	**1.2950e+01**	**4.2823e+00**
45	118	47	118	**7.0000e-06**	1.6167e+02	3.1759e+01	**1.4299e+01**	**1.0776e+01**	**1.7460e+01**	**8.7686e+00**
46	118	46	118	**9.0000e-05**	1.6435e+02	3.5416e+01	**1.7522e+01**	**6.7362e+00**	**1.7893e+01**	**7.0032e+00**
47	118	45	118	**1.1000e-05**	1.6072e+02	3.2713e+01	**1.6269e+01**	**5.4600e+00**	**1.6444e+01**	**7.6689e+00**

(Z,A) of light fragment
(Z,A) of heavy fragment
Mean and width of light fragment E_{ex}
Mean and width of heavy fragment E_{ex}

2. Reconstruct fission fragment distributions (*$G(E_x)$ and $R(J^\Pi)$*)

3. Run the Hauser-Feshbach statistical decay calculation
   - neutron and gamma emissions from each fragment



$$R(J,\Pi) = \frac{J+1/2}{2f^2\sigma^2(U)} \exp\left\{-\frac{(J+1/2)^2}{2f^2\sigma^2(U)}\right\}$$

$$G(E_x) = \frac{1}{\sqrt{2\pi}\delta_{l,h}} \exp\left\{-\frac{(E_x - E_{l,h})^2}{2\delta_{l,h}^2}\right\}$$

Fission fragment yield models stored in TALYS (1.96)

Prompt decay

GEF
(ffmodel 1)

Designed with global fitting parameters based on experimental data
F. Nordström, Technical Report UPTEC ES21016, Uppsala university, 2021.
From $_{76}$Os to $_{115}$Mc, 737 nuclides

HF^{3}D
(ffmodel 2)

Designed with a fully deterministic technique with fitting functions
S. Okumura, T. Kawano, P. Jaffke, P. Talou, and S. Chiba, JNST, 55(9),1009–1023, 2018.
^{236}U, ^{239}U, and ^{240}Pu, 3 nuclides

SPY
(ffmodel 3)

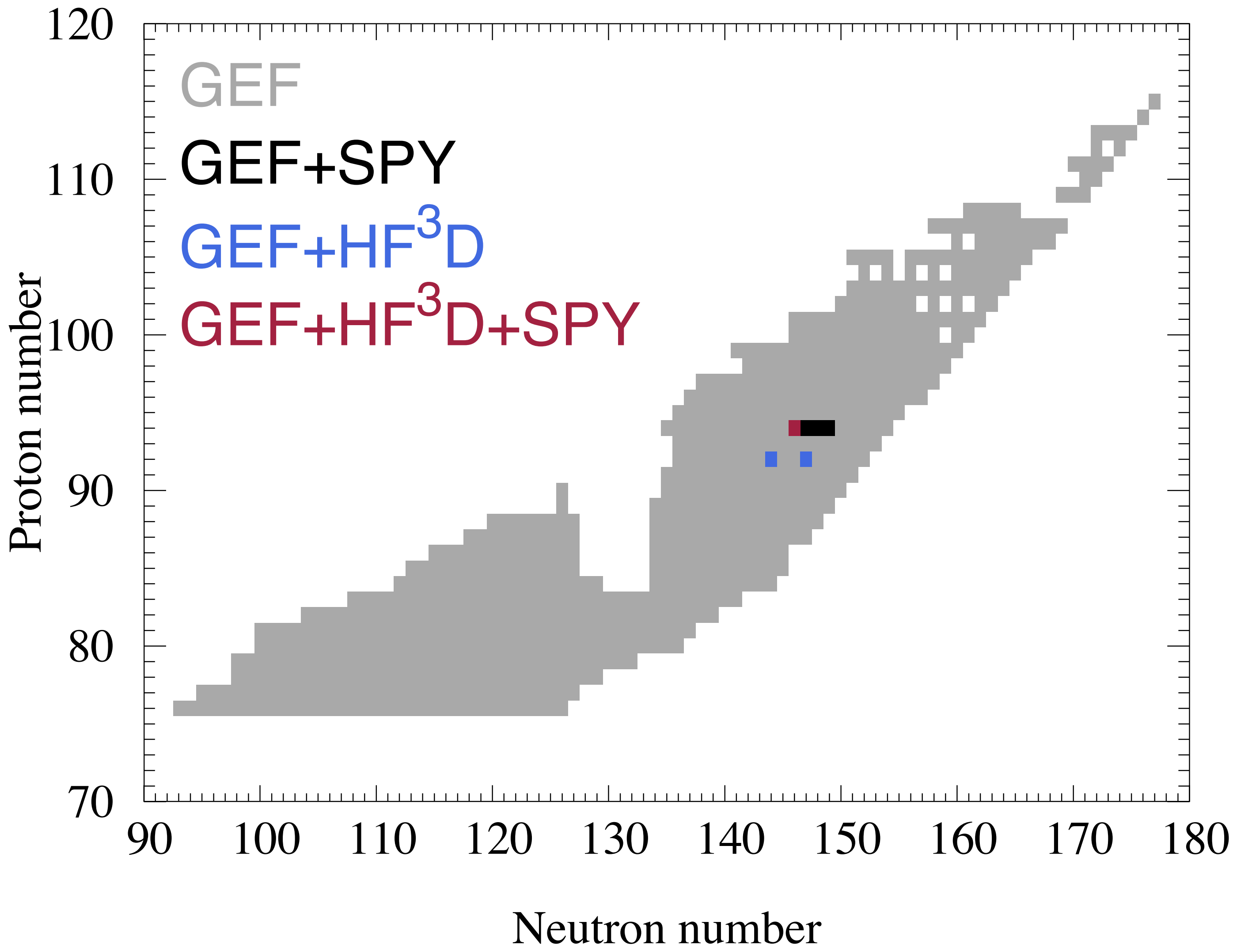
Designed with a statistical scission point model using microscopic calculation
J.-F. Lemaître, S. Goriely, S. Hilaire, and J.-L. Sida, PRC99, 034612, 2019.
^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{243}Pu, 4 nuclides

Local
(ffmodel 0)

Arbitrary fission fragment data provided by users.
(will be implemented in next update of TALYS)

Fission fragment yield models stored in TALYS (1.96)

Prompt decay



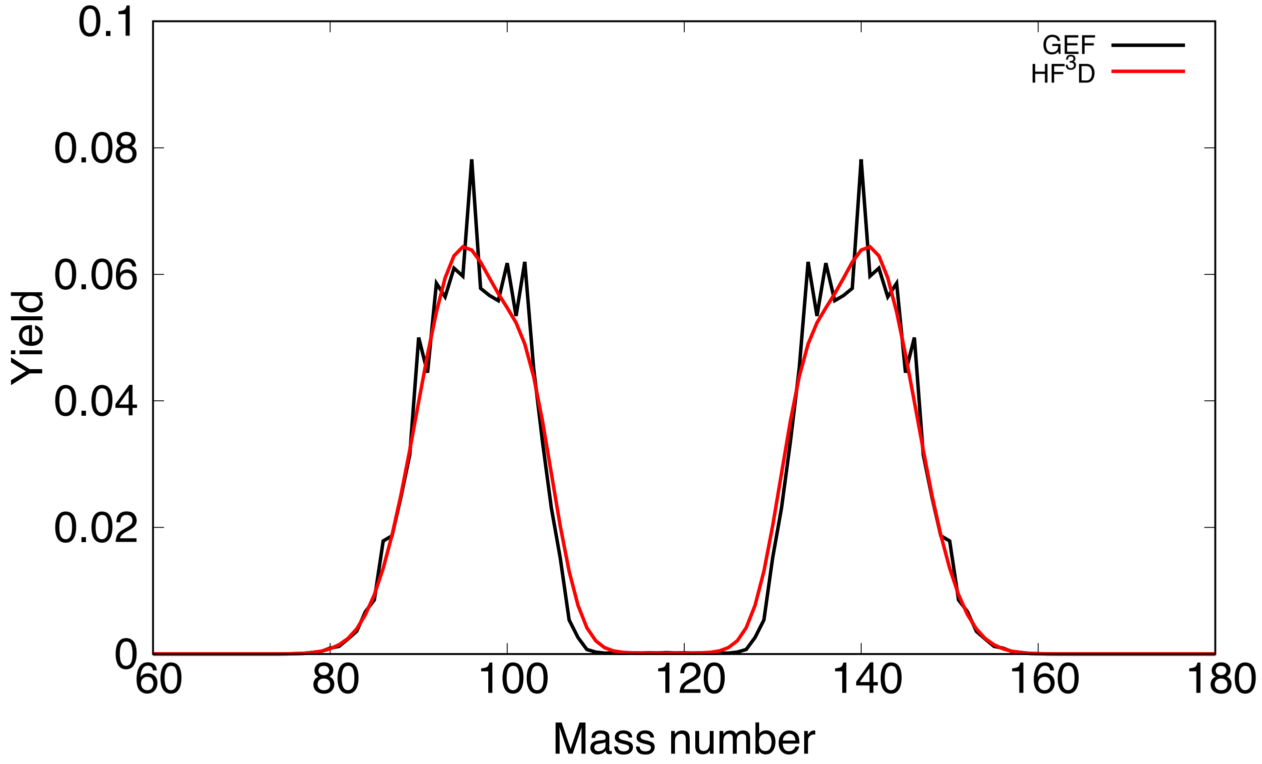
Calculatable physical quantities

Prompt decay

Table 1: Representative fission quantities comparable to experimental data.

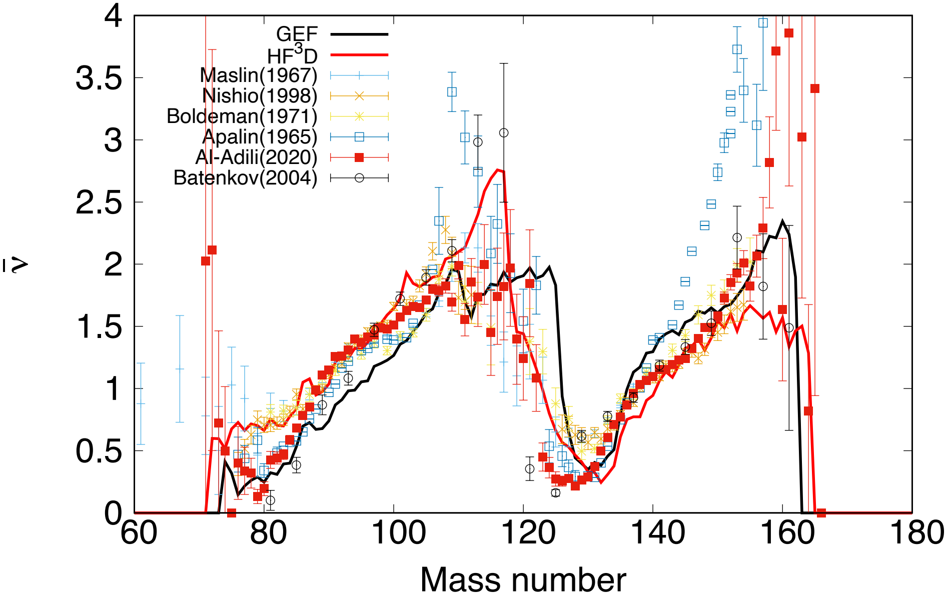
Type	Description
$Y_i(A)$	Independent fission yield as a function of mass number
$Y_i(Z, A, M)$	Independent fission yield of all isotopes including meta-stable state
$\overline{\nu}$	Average number of neutrons per fission
$\overline{\gamma}$	Average number of γ-rays per fission
$\overline{\nu}(A)$	Average neutron multiplicity as a function of fission product mass
$\overline{\gamma}(A)$	Average γ-ray multiplicity as a function of fission product mass
$\langle E_n \rangle$	Average prompt neutron energy
$\langle E_\gamma \rangle$	Average prompt γ-ray energy
$\langle E_n \rangle(A)$	Average neutron energy as a function of product mass
$\langle E_\gamma \rangle(A)$	Average γ-ray energy as a function of product mass
$\mathrm{P}(\nu)$	Neutron multiplicity distribution
$\chi(\nu)$	Prompt fission neutron energy spectrum (PFNS)
$\phi(\gamma)$	γ-ray energy spectrum

Fission fragment mass distribution of $^{235}U+n_{th}$

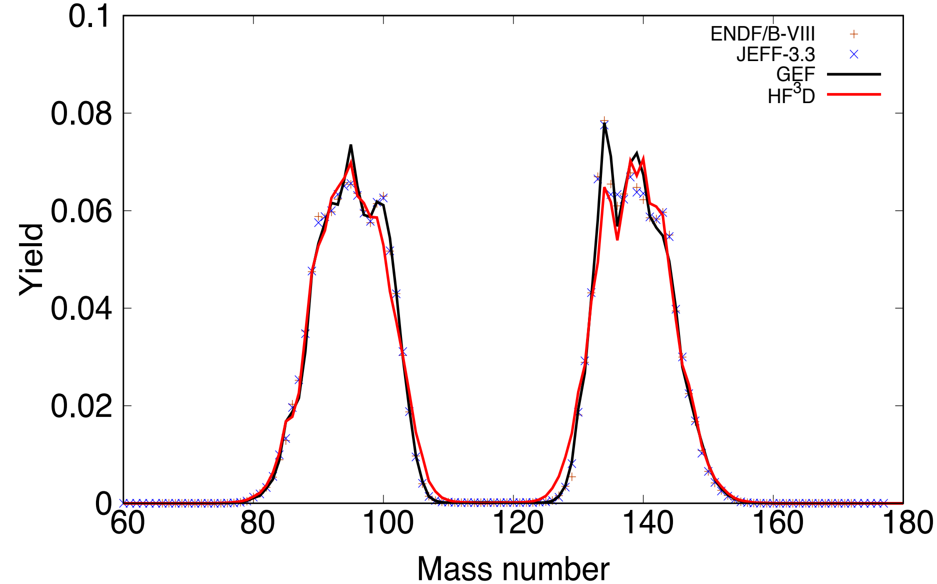


Fission fragment mass distribution generated by GEF and HF3D model stored in TALYS fission fragment database

Calculated ν(A) and Y(A) for $^{235}U+n_{th}$



Calculated ν(A) from GEF and HF³D model FF distributions and Hauser-Feshbach statistical decay by TALYS



Calculated independent FPY from GEF and HF³D model FF distributions and Hauser-Feshbach statistical decay by TALYS

Average number of neutrons/fission

GEF+TALYS	2.275
HF³D	2.353
JENDL	2.420

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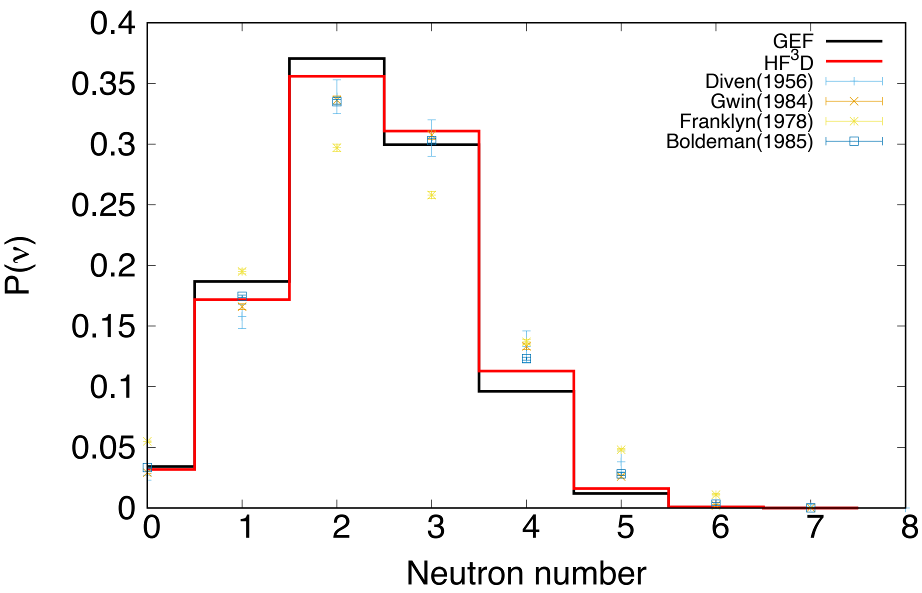
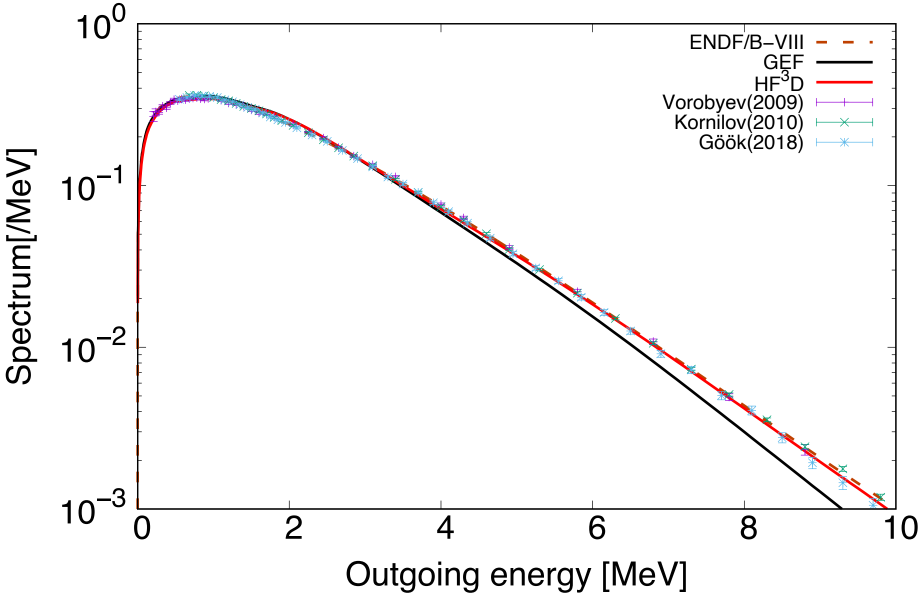
Calculated P(ν) and PFNS for $^{235}U+n_{th}$

Prompt decay

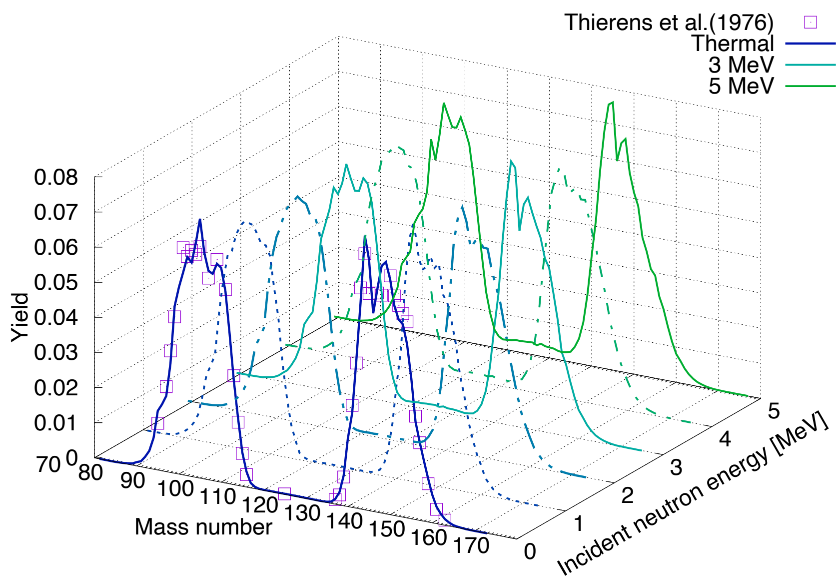


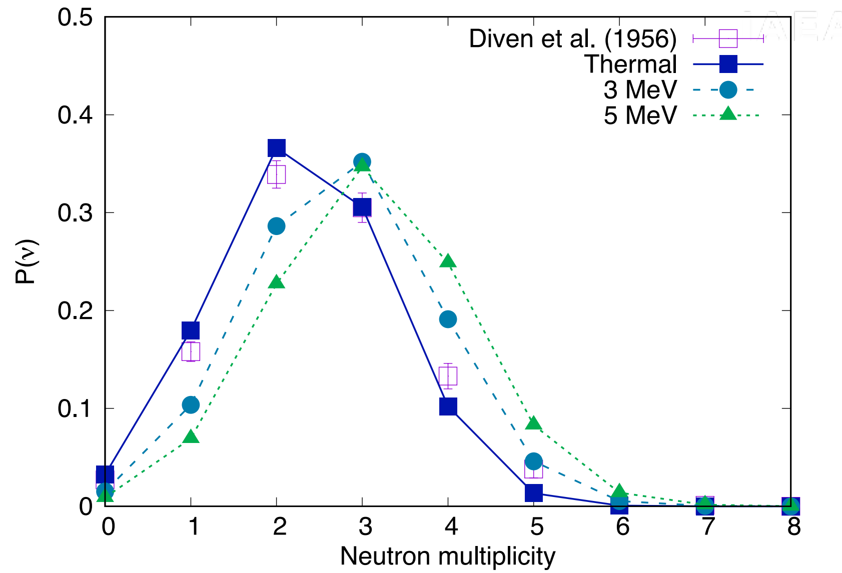
Calculated P(ν) from GEF and HF³D model FF distributions and Hauser-Feshbach statistical decay by TALYS

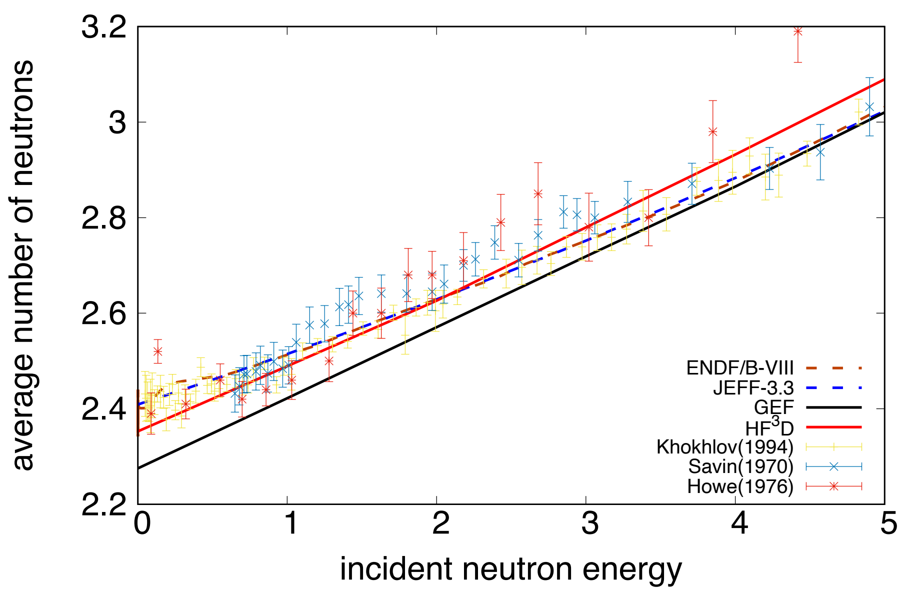


Calculated prompt fission neutron spectrum (PFNS) from GEF and HF³D model FF distributions and Hauser-Feshbach statistical decay by TALYS

Energy dependent fission observables for $^{235}U+n$







Calculated independent FPY, P(ν), and average ν from GEF FF distributions and Hauser-Feshbach statistical decay by TALYS up to 5 MeV

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Acknowledgements

- The IAEA-NDS acknowledges the internship program “The Nuclear Regulation Human Resource Development Program (ANSET: Advanced Nuclear 3S Education and Training)” entrusted to Tokyo Institute of Technology by the Nuclear Regulation Agency of Japan, for supporting this work.
- T. Kawano (Los Alamos National Lab) for his advice and intensive support on our work.
- A. Al-Adili (Uppsala University) for GEF calculations and data supply.
- S. Goriely (Université Libre de Bruxelles) and J.-F. Lemaitre (CEA) for SPY calculations and data supply.

Beta Decay Calculations from Independent Fission Product Yields

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Standalone beta decay program

- Python

1. Convert ENDF format decay data library files to
    - (1) Simplified format (inspired by the Oyak/Decayfinite code)

```
  55  141    0  2.4910000E+01  6.0000000E-02  1.3970950E+02    2    5 1920
1.5473620E+06  1.8786310E+05  1.7248140E+06  5.7510480E+03  7.5359650E+01  3.0848980E+00
1.0000000E+00  0.0000000E+00  5.2560000E+06  1.0000000E+04  9.9965800E-01  2.6533000E-05
1.5000000E+00  0.0000000E+00  7.2100000E+05  1.2000000E+04  3.4200000E-04  1.4000000E-05
```

    - (2) JSON format
2. Generate β-decay chains from decay data library
3. Import independent fission product yield file (TALYS output format)

```
#   Z    A iso      FP yield      FF yield        FP xs        FF xs
   30   74 -1     5.3961E-06    3.0000E-06    1.9191E+00    1.0670E+00
   30   75 -1     1.4014E-05    1.1000E-05    4.9841E+00    3.9123E+00
   31   75 -1     2.7919E-06    2.0000E-06    9.9294E-01    7.1132E-01
```

4. Solve the Bateman equations
5. Calculate beta-decay observables from independent yield
    - Cumulative fission product yield (at 1000 years time period)
    - Decay heat from β and γ rays (time dependent)
    - Delayed neutron yield (at 1000 years time period and time dependent)
    - Spectrum (Under development)

- Preview decay-chain
- Open source in Github
    - Test version: https://github.com/shinokumura/FPY_betadecay
    - Will be moved to Github/IAEA-NDS near future

Features

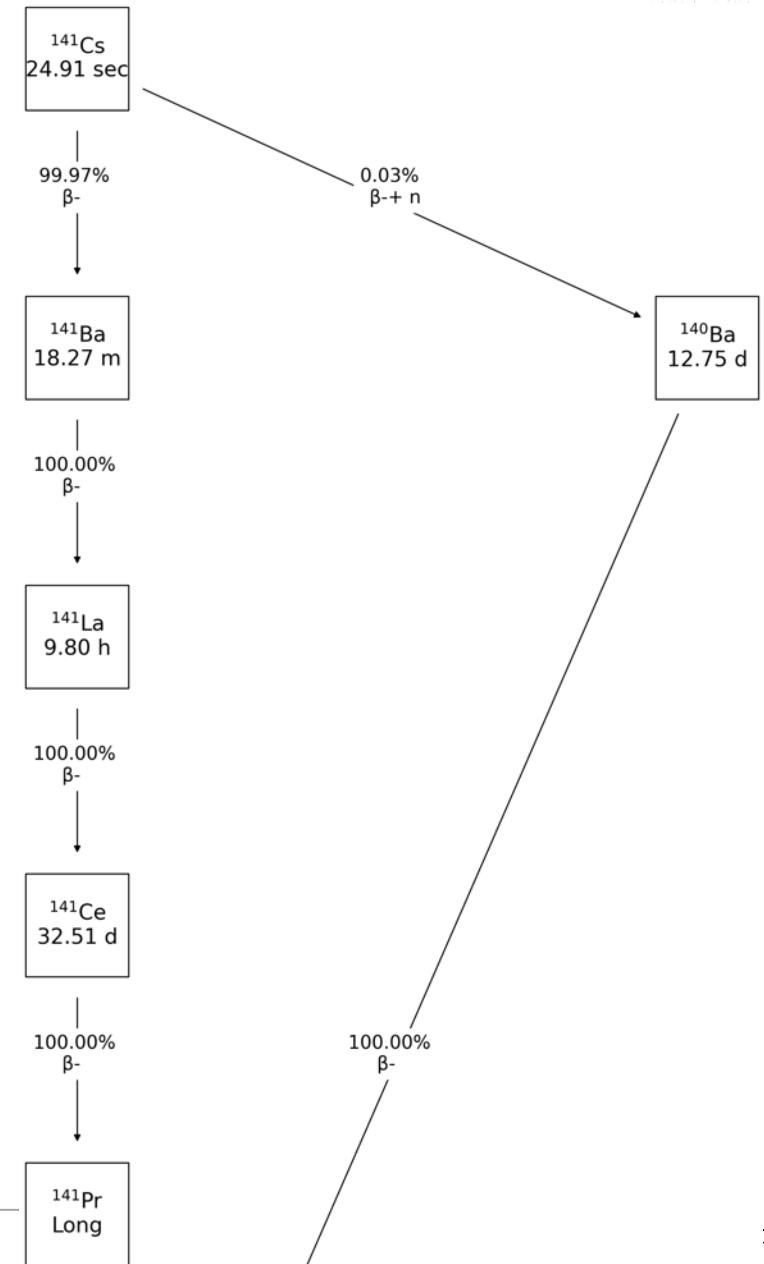
Decay data


```
"55-Cs-141-00": {
   "Z": "55",
   "ELM": "Cs",
   "MASS": "141",
   "LIS": "00",
   "HL": "2.4910000E+01",
   "LAMBDA": 0.027826,
   "En_beta": "1.5473620E+06",
   "En_gamm": "1.7248140E+06",
   "En_alpha": "7.5359650E+01",
   "DecayInfo": {
     "0": {
       "RTYP": "1.0000000E+00",
       "RFS": "0.0000000E+00",
       "Q": "5.2560000E+06",
       "BR": "9.9965800E-01",
       "DAUGHTER": "56-Ba-141-00"
     },
     "1": {
       "RTYP": "1.5000000E+00",
       "RFS": "0.0000000E+00",
       "Q": "7.2100000E+05",
       "BR": "3.4200000E-04",
       "DAUGHTER": "56-Ba-140-00"
     }
   },
   "daughters": [
     "56-Ba-141-00",
     "56-Ba-140-00"
   ]
 },
```

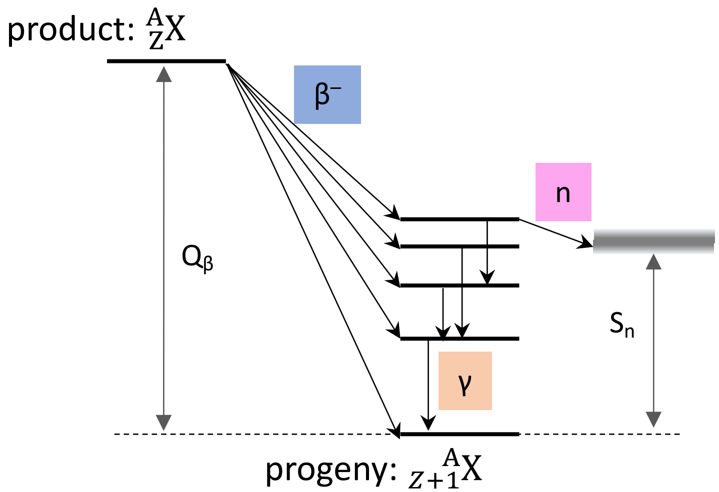

Decay chain (linearized)


```
"55-Cs-141-00": {
   "1": {
        "chain": [
        "56-Ba-141-00",
        "57-La-141-00",
        "58-Ce-141-00",
        "59-Pr-141-00"
        ],
        "branching": [
        0.999658,
        1.0,
        1.0,
        1.0
        ],
        "rtyp": [
        1.0,
        1.0,
        1.0,
        1.0
        ],
        "lmbds": [
        0.027826,
        0.00063232,
        4.9118e-05,
        2.4676e-07,
        6.9315e-51
        ],
        "en_betas": [
        "1.5473620E+06",
        "9.6628250E+05",
        "9.8713460E+05",
        "1.9438810E+05",
        0.0
        ],
        "en_gamms": [
        "1.7248140E+06",
        "9.0968270E+05",
        "2.6780420E+04",
        "7.6901970E+04",
        0.0
        ]
        },
   "2": {
        "chain": [
        "56-Ba-140-00",
        "57-La-140-00",
        "58-Ce-140-00"
        ],…
```


Decay chain (network diagram plot)



Decay heat and delayed neutron yield at E_n= thermal



Decay heat:

Sum of average energies of β and γ rays from fission products as a function of time following a single fission event.

$$DH_{\gamma,\beta}(t) = \sum \langle E_{\gamma,\beta} \rangle \lambda_{\gamma,\beta} N_i(t) \quad N_i(t)\text{: FP Yield at time } t$$

Delayed neutron:

Sum of the number of delayed neutron emissions.

$$\nu_d(t) = \sum P_n \lambda_i N_i(t) \quad N_i(t)\text{: FP Yield at time } t$$

$$\overline{\nu_d} = \sum P_n N_i \quad N_i\text{: Cumulative yield}$$

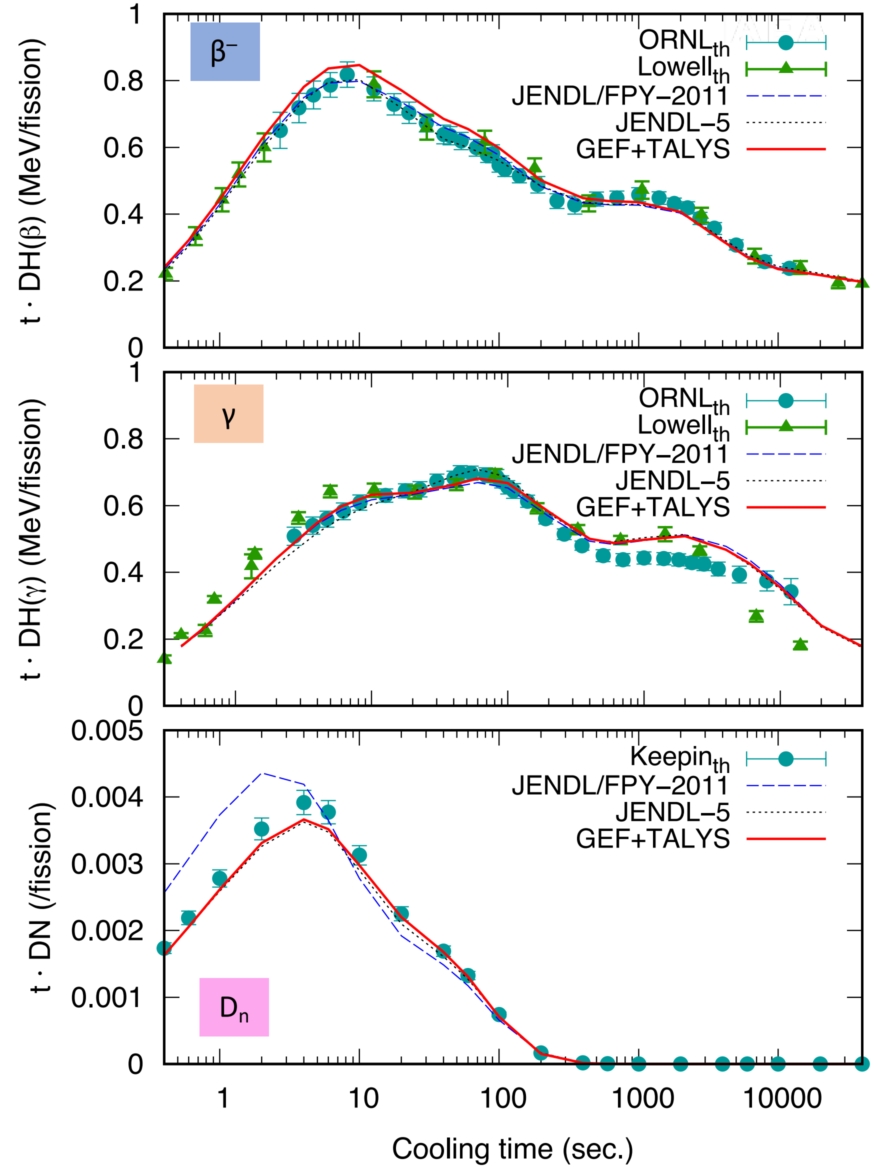


Fig 2. β and γ decay heat and delayed neutron yield burst functions of ^{235}U

Method

1. Import calculated independent fission product yields, $Y_{IND}(Z, A, M)$

yieldZA1.00E-06.fis

```
# n + 235U : Z, A Fission yields
# E-incident =  1.00000E-06
#
#
#   Z    A iso      FP yield      FF yield        FP xs         FF xs    Isom. Ratio
   30   74 -1     5.3961E-06    3.0000E-06    1.9191E+00    1.0670E+00
   30   75 -1     1.4014E-05    1.1000E-05    4.9841E+00    3.9123E+00
   31   75 -1     2.7919E-06    2.0000E-06    9.9294E-01    7.1132E-01
   30   76 -1     5.7899E-05    5.0001E-05    2.0592E+01    1.7783E+01
   31   76 -1     5.5761E-06    3.0000E-06    1.9832E+00    1.0670E+00
   32   76 -1     1.9258E-06    2.0000E-06    6.8493E-01    7.1132E-01
   30   77 -1     7.2165E-05    4.0001E-05    2.5666E+01    1.4226E+01
   30   77  0     5.3674E-05                  1.9090E+01                  7.4378E-01
   30   77  1     1.8490E-05                  6.5761E+00                  2.5622E-01
   31   77 -1     5.7907E-05    4.6001E-05    2.0595E+01    1.6360E+01
   29   78 -1     2.1861E-06    3.0000E-06    7.7751E-01    1.0670E+00
```

2. Import decay data library
3. Run the radioactive decay calculation

Energy dependent cumulative yield

β decay

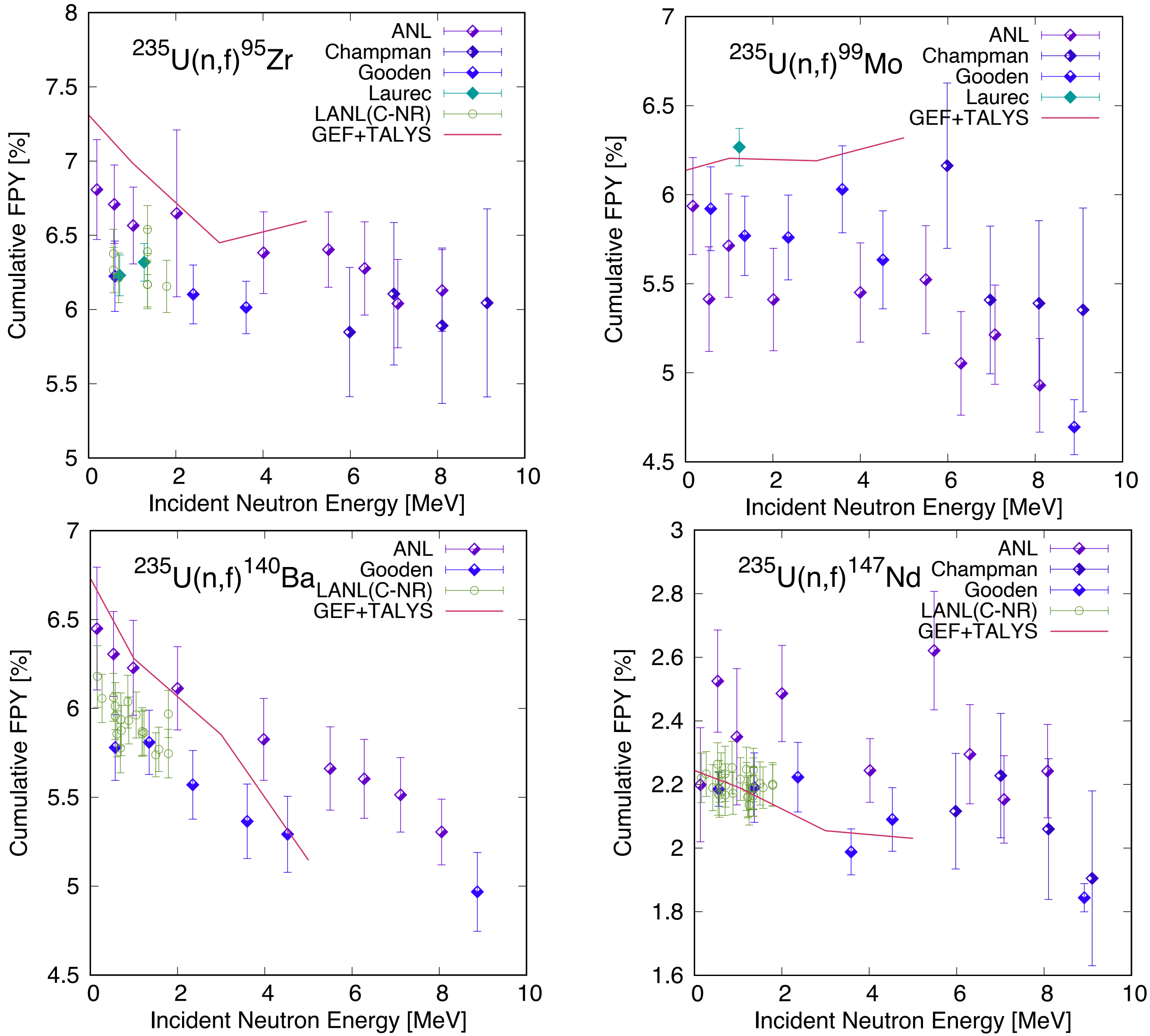


Fig 1. Comparison of cumulative yield of selected fission products showing as function neutron energy

Energy dependent decay heat and delayed neutron

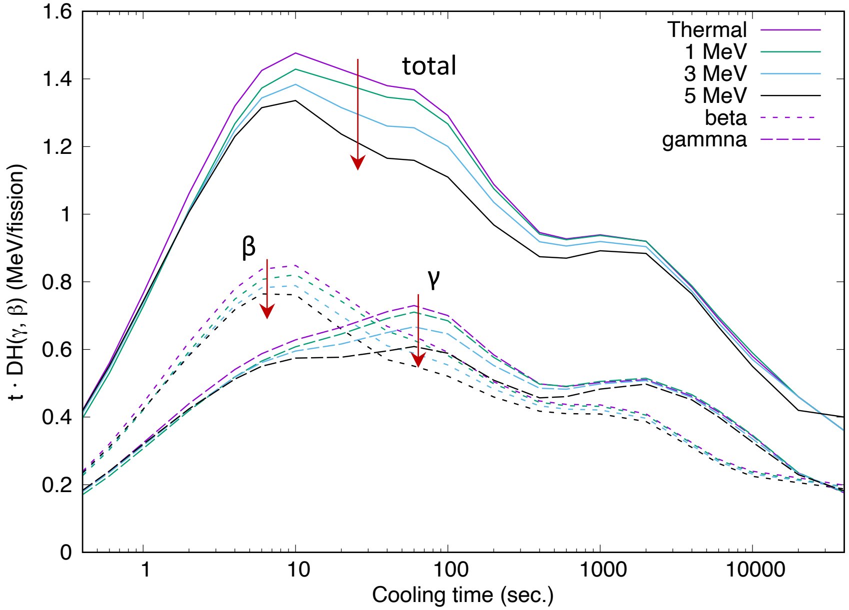
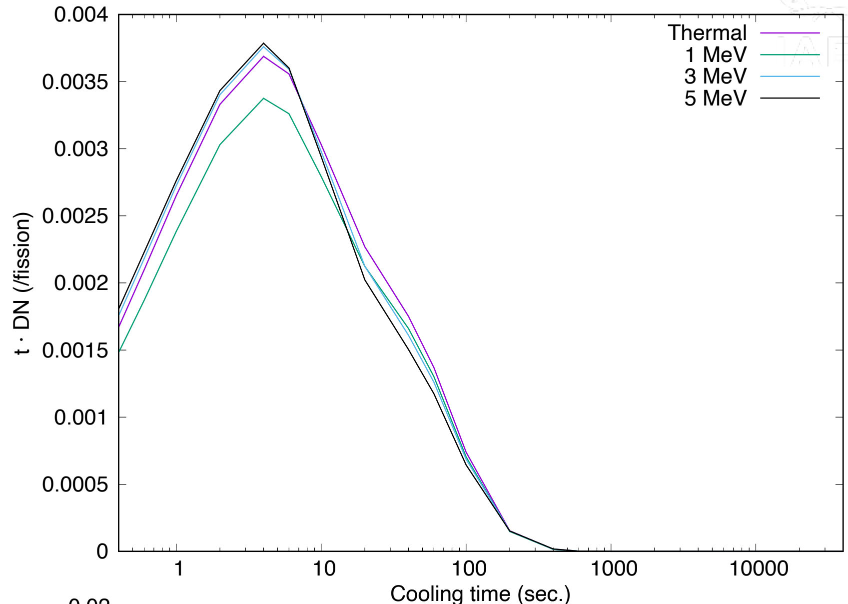


Fig 3. Comparison of total decay heat (–:total, dashed: β and γ) burst functions of ^{235}U for various neutron energies



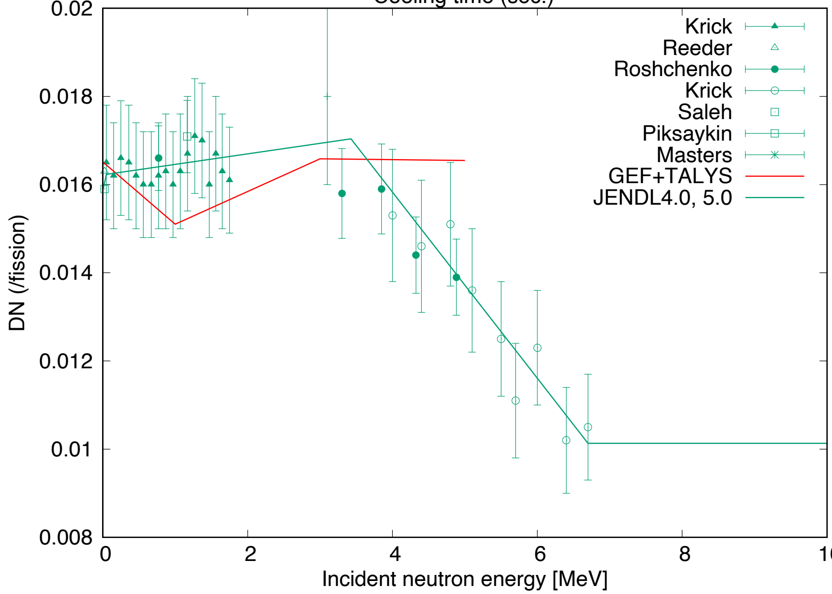


Fig 4. Incident neutron energy dependence of average delayed neutron yield

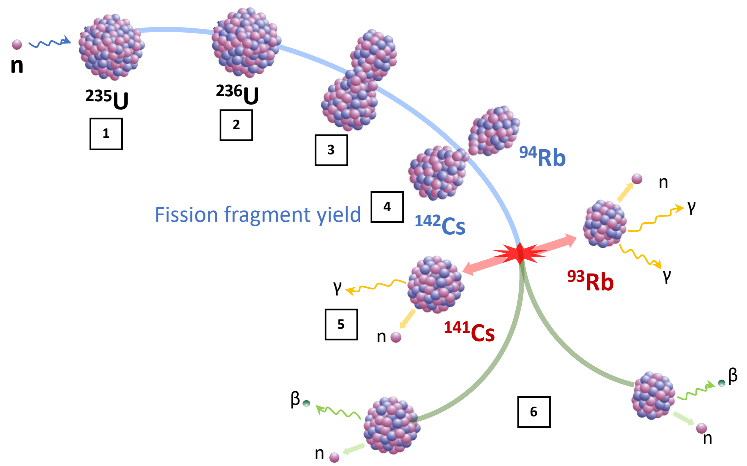
Summary

(1) TALYS[1] fission fragment de-excitation

- Fission fragment distributions by GEF[2], HF^3D[3], SPY[4], and user's own model
- Application of the Hauser-Feshbach theory to the fission fragment de-excitation process
- Calculate independent fission product yield, prompt neutron and gamma observables
- Code comparisons, multi-chance fission implementation is underway

(2) Beta-decay observables from beta-decay of fission products

- Cumulative fission product yield
- Decay heat from β and γ rays
- Delayed neutron yield



Details is published in the IAEA NDS report
https://nds.iaea.org/publications/nds/iaea-nds-0239/

[1] Koning A.J., Hilaire S., Duijvestijn M.C. AIP Conference Proceedings. 2005;769(1):1154–1159.
[2] Schmidt K.H., Jurado B., Amouroux C., Schmitt C. Nuclear Data Sheets. 2016;131:107 – 221.
[3] Okumura S., Kawano T., Jaffke P., Talou P., Chiba S. JNST. 2018;55(9):1009–1023.
[4] Lemaitre J.F., Goriely S., Hilaire S., Sida J.L., Phys Rev C. 2019 Mar;99:034612.

Some Tools and Developments

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Easy-to-use nuclear data plotter: Dataexplorer

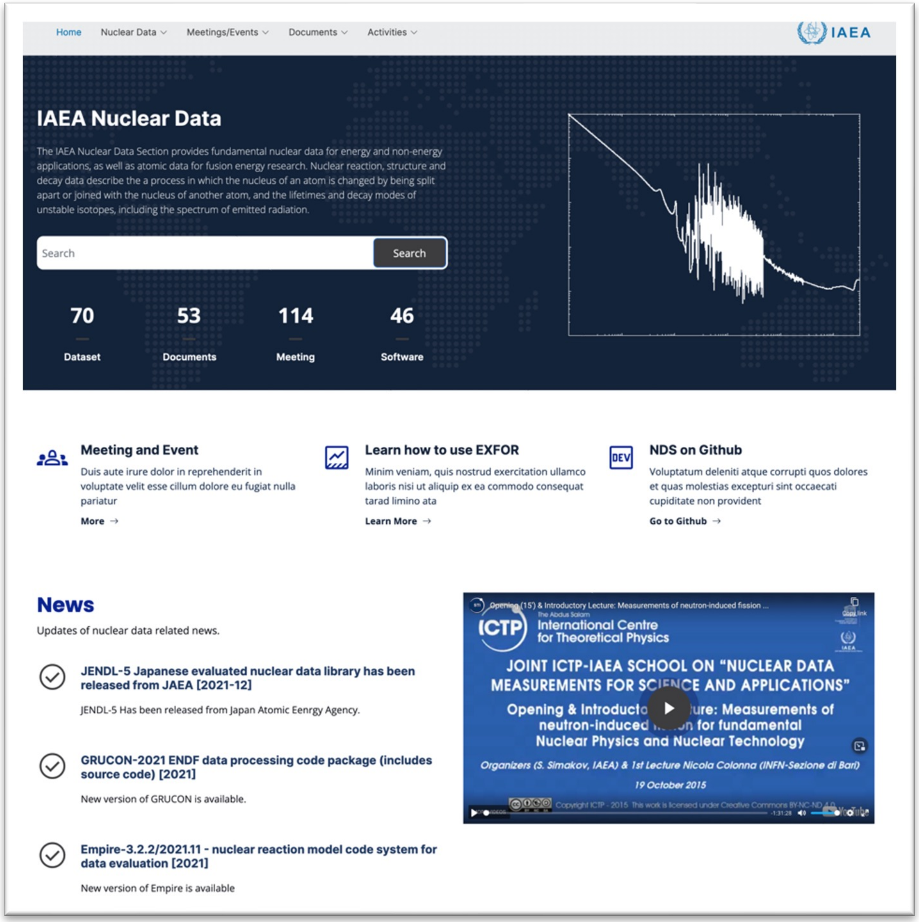
- Plot, data table view, and tabulated data download option
  - Evaluated cross section with experimental data: https://nds.iaea.org/dataexplorer/xs
  - Residual production with experimental data: https://nds.iaea.org/dataexplorer/rp
  - Fission yields: https://nds.iaea.org/dataexplorer/fy

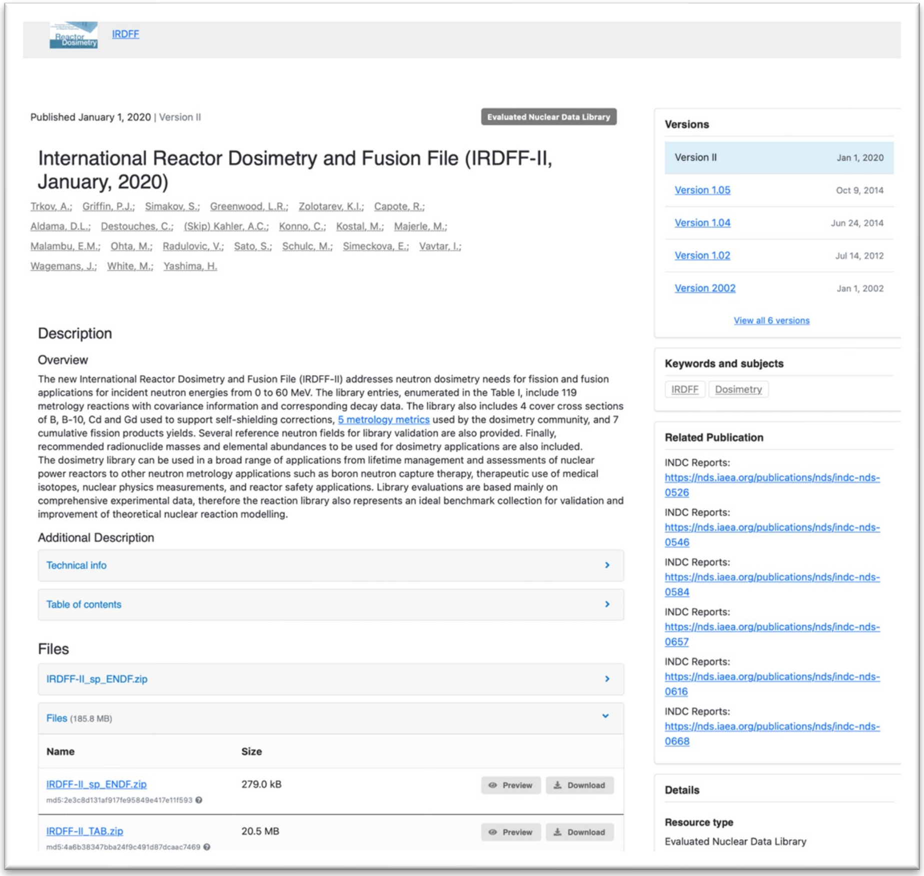
https://nds.iaea.org/dataexplorer/**fy**?fissile_elem=**U**&fissile_mass=**238**&inc_pt4=**n**&inc_energy=**MeV**&fy_type=**Cumulative**

Modernization work

- IAEA-NDS Github
    - https://github.com/IAEA-NDS
- EXFOR master files:
    - https://github.com/IAEA-NDS/exfor_master
- EXFOR dictionary in JSON:
    - https://github.com/IAEA-NDS/exfor_dictionary

IAEA-NDS website renewal





Thank you for your attention.

IAEA

International Atomic Energy Agency

Atoms for Peace and Development

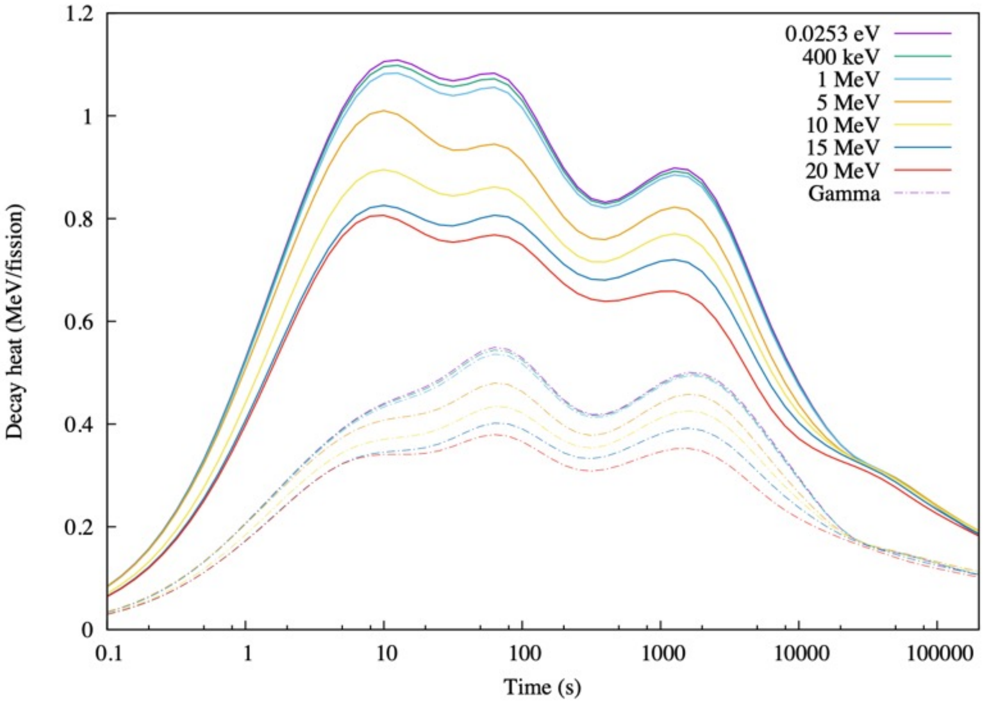
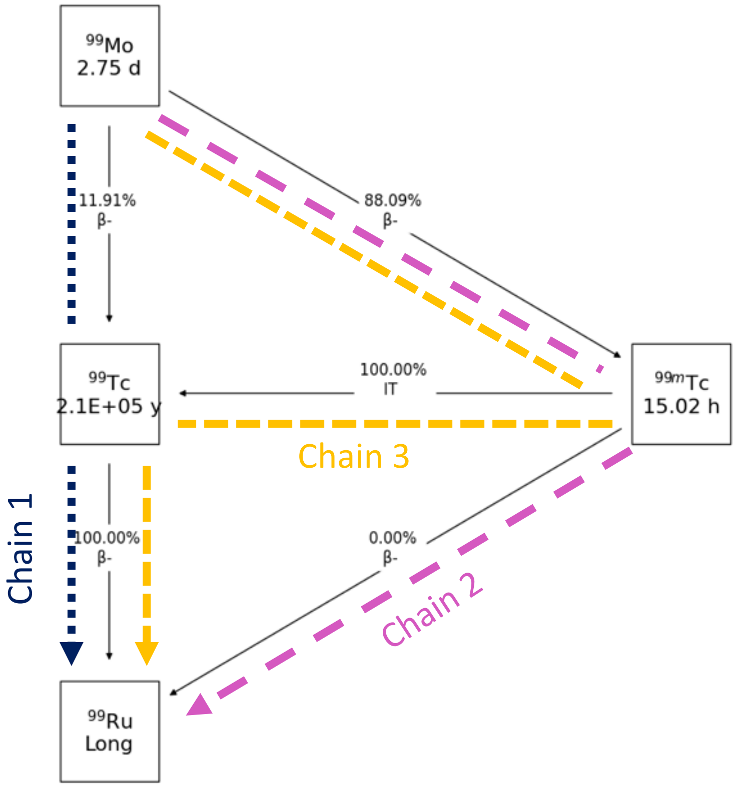


Figure 7: Comparison of total decay heat burst functions of ^{239}Pu for various neutron energies. All simulations use GEF-4.2 nFY with ENDF/B-VII.1 decay data.

- The asymmetric distributions of fission products for well-known fissiles become considerably more broad at higher neutron energy. As a result, the dominant nuclides for some thermal response function, which are the most likely fission products, will have less production in exchange for increased production from the ‘shoulders’ of the fission yield distribution.
- The general change in the response function will be a decrease across all time periods, potentially with some new response from a previously minor nuclide. This can be seen in the neutron-energy- dependent decay heat curves from fission pulses calculated with GEF-4.2 fission yields. Two examples are shown in Figures 6 and 7: 235U and 239Pu, respectively.



The time evolution of nuclide yield undergoing a linear decay chain is governed by a set of a first-order differential equation, called Bateman equations.
The Bateman equations for radioactive decay case of *i*-nuclide series in linear chain describing nuclide yield are as follows:

$$\frac{dN_1}{dt} = \lambda_1 N_1$$
$$\frac{dN_i}{dt} = \lambda_{i-1} N_{i-1} - \lambda_i N_i \quad (i = 2, n)$$

where λ_i is the decay constant of ith nuclide. By assuming zero yields of all progeny after time t, the yield of nth nuclide in the specific decay chain is given by:

$$N_n(t) = \frac{N_1(0)}{\lambda_n} \sum_{i=1}^{n} \lambda_i c_i e^{(-\lambda_i t)}$$
$$c_n = \sum_{j=1, i \neq j}^{n} \frac{\lambda_j}{\lambda_j - \lambda_i},$$