

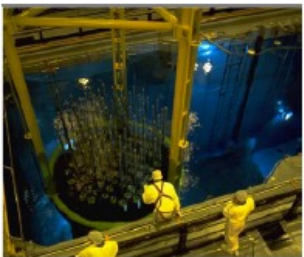
Decay Heat calculations

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On behalf on Molten Salt Reactors team (CNRS LPSC & Subatech)

Challenges associated to Decay Heat

- Safety/Radiation protection
- Economic interests for the complete cycle (Gen II, Gen III)
- Key issue for new concepts: Gen IV, innovative reactor design, innovative fuels, most of the concepts with fast neutrons => not so many data, limited reactor operation feedback
- Important design parameter for a spent fuel repository



Industrial stakes	Cooling time
Safety systems of cooling	0.1s to 8 days
Unloading of assemblies from core	5 to 25 days
Spent Fuel transport	1 to 10 years
Reprocessing, vitrification, storage	4 to 300 years
Disposal	50 to 300 000 years



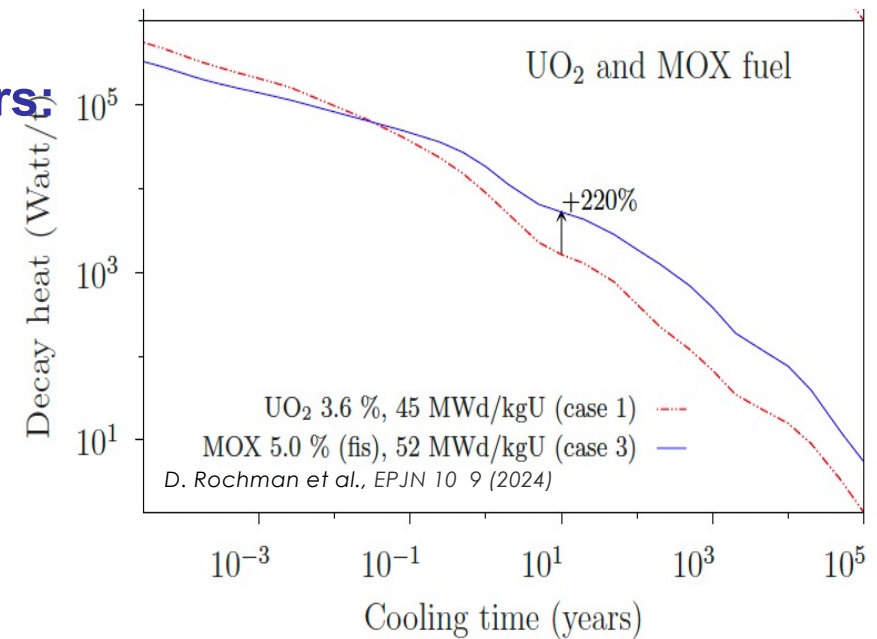
Challenges associated to Decay Heat

- Different applications => different main contributors,

- Large time range : few seconds -> 10^6 years

Dependency:

- cooling time,
- studied concept,
- irradiation history ...



- Increasing will of safety authorities => better understanding of uncertainty sources : technological, operation, calculation methods & nuclear data

Industrial interest: improved control of uncertainty calculations & safety margins



Calculations with evaluated codes (experimental measurements, other codes)

Identifying biases in calculations (ex: nuclear data) for improvement

Decay heat calculations

■ Summation method

$$DH(t_c) = f(t_c) = \sum_i^n N_i(t_c) \lambda_i \bar{E}_i$$

N_i : Number of nuclei i at cooling t_c

λ_i : Decay constant

\bar{E}_i : Mean decay energy of nucleus i

■ Atomic Densities N_i : solving Bateman's equations

Transport and depletion calculations : reactor model + neutronic code

$$\frac{\partial N_i(t)}{\partial t} = \sum_{j \neq i} (b_{j \rightarrow i} \lambda_j + \langle \sigma_{j \rightarrow i} \phi \rangle) N_j(t) - (\lambda_i + \langle \sigma_i \phi \rangle) N_i(t)$$

production & disappearance per radioactive decays & nuclear reactions

~ 40 000 nuclear data: σ , \bar{E} , Branching Ratio, λ , Fission Yields, $\bar{\nu}$

=> Complex calculation (reactor model + depletion)

=> Quality of the code but also of the input data !

Decay heat calculations

■ Summation method

$$DH(t_c) = f(t_c) = \sum_i^n N_i(t_c) \lambda_i \bar{E}_i$$

■ \bar{E}_i usually divided in 3 parts in evaluated libraries(e.g ENDF, JEFF, JENDL) :

$$\bar{E}_{LP} = \bar{E}_{\beta^-} + \bar{E}_{\beta^+} + \bar{E}_{e^-} + \dots \quad \text{Light particles component}$$

$$\bar{E}_{EM} = \bar{E}_{\gamma} + \bar{E}_{x\text{-ray}} + \bar{E}_{\text{anni.rad.}} + \dots \quad \text{Electromagnetic component}$$

$$\bar{E}_{HP} = \bar{E}_{\alpha} + \bar{E}_{SF} + \bar{E}_p + \bar{E}_n + \dots \quad \text{Heavy particles component}$$

■ \bar{E}_i measurements & Pandemonium effect

Before the 90s, conventional detection techniques: high resolution γ -ray spectroscopy

Excellent resolution but efficiency which strongly decreases with increasing energy

Risk of overlooking the existence of β^- feeding into the high energy nuclear levels of daughter nuclei

Incomplete decay schemes: overestimate \bar{E}_{beta} , underestimate \bar{E}_{gamma}

=> Most suitable technique to re-measure key nuclei:

Total Absorption Gamma Spectroscopy using 4π detectors (ex: NaI, LaBr3)

Decay Heat / Fission pulses

- Decay heat fission pulses measurements:



- No need of XS for short irradiation times
- To disentangle FY vs DD data impact (ex JENDL5 FY)
- To identify Pandemonium FP to re-measure
- To develop/test uncertainty propagation calc.

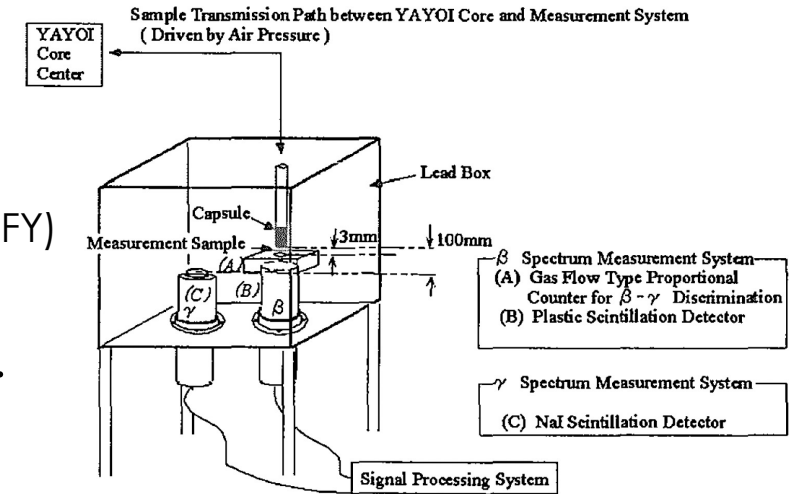
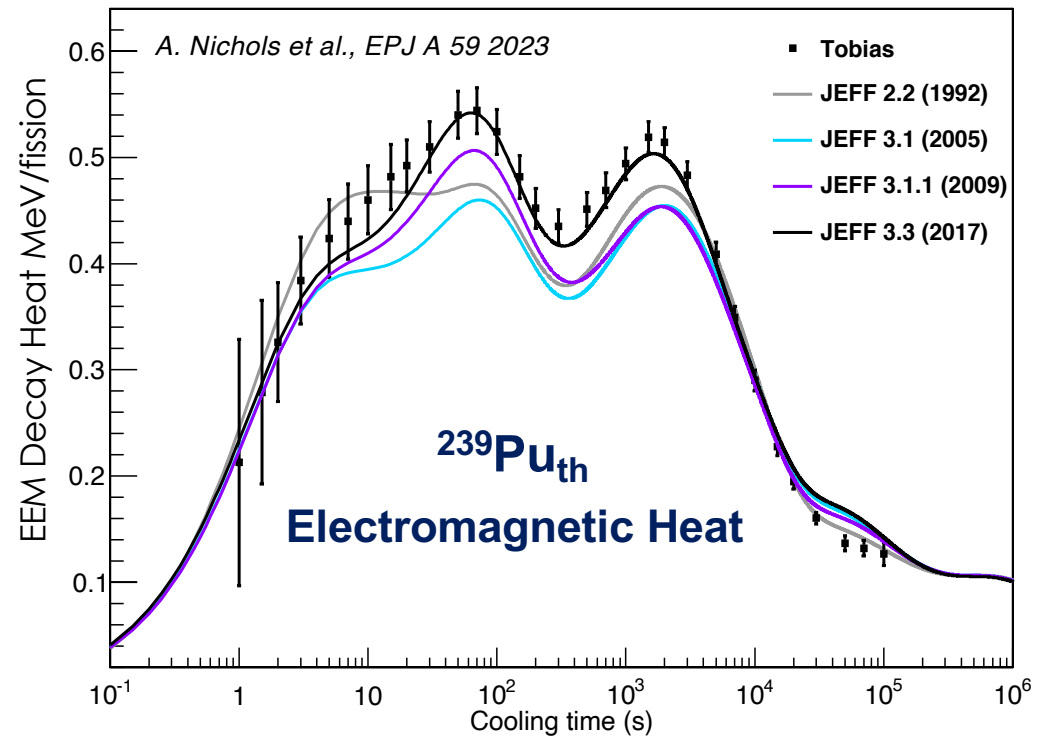
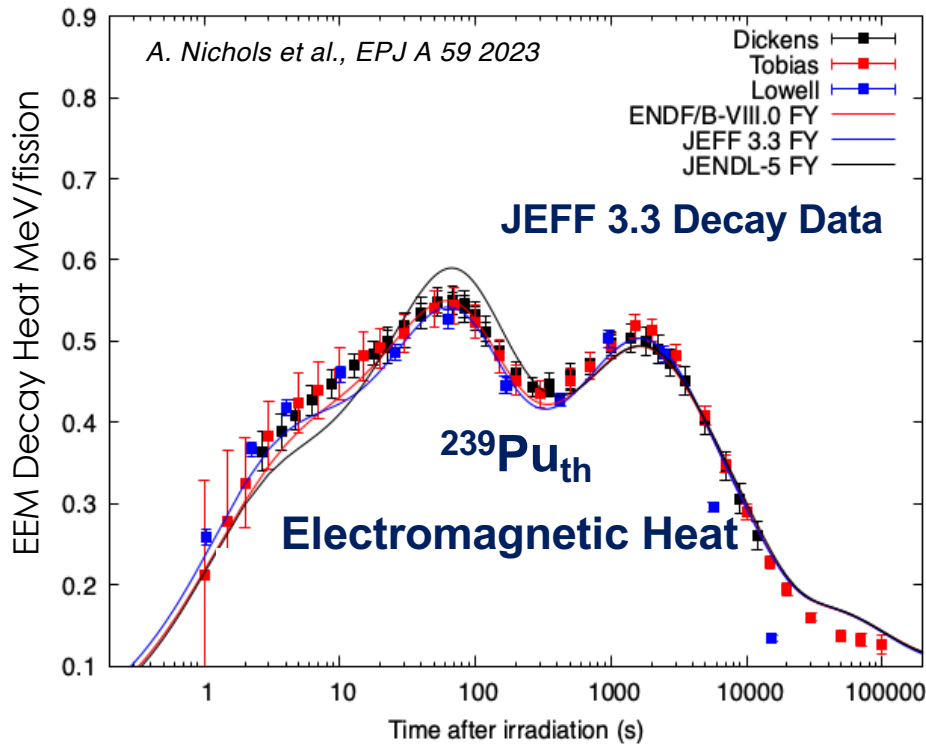


Fig.2 Conceptual View of Decay Heat Measurement System
Y. Ohkawachi et al., JNST 39 (2002)



Decay Heat / Fission pulses

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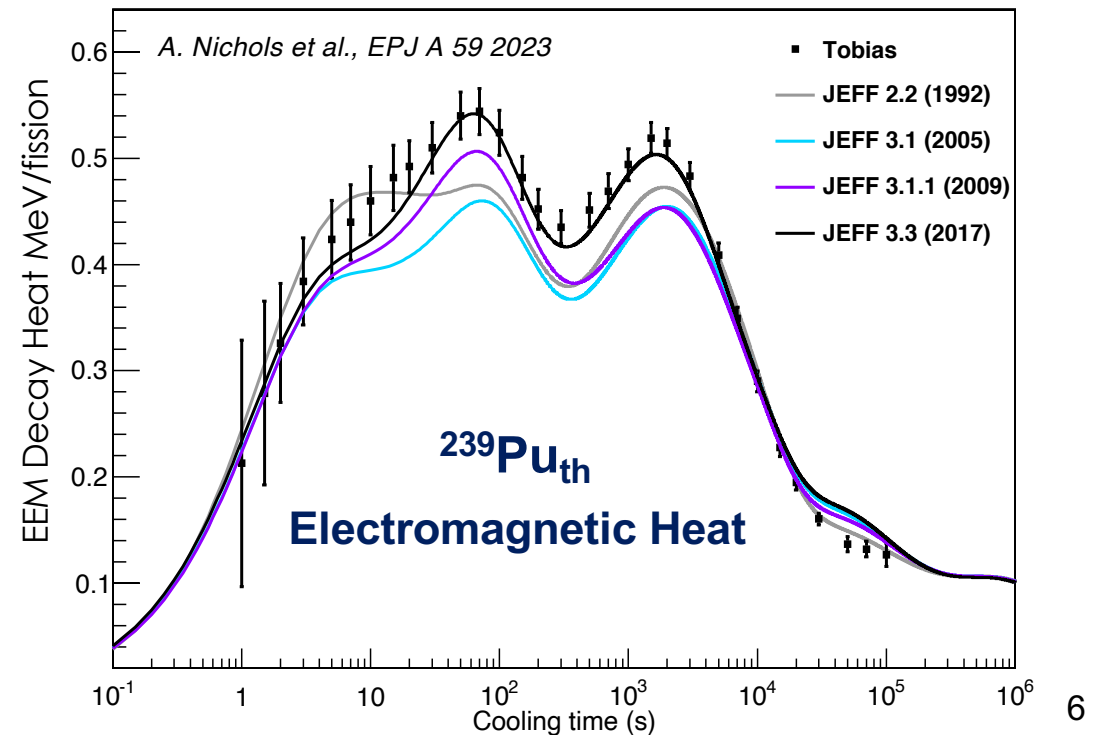
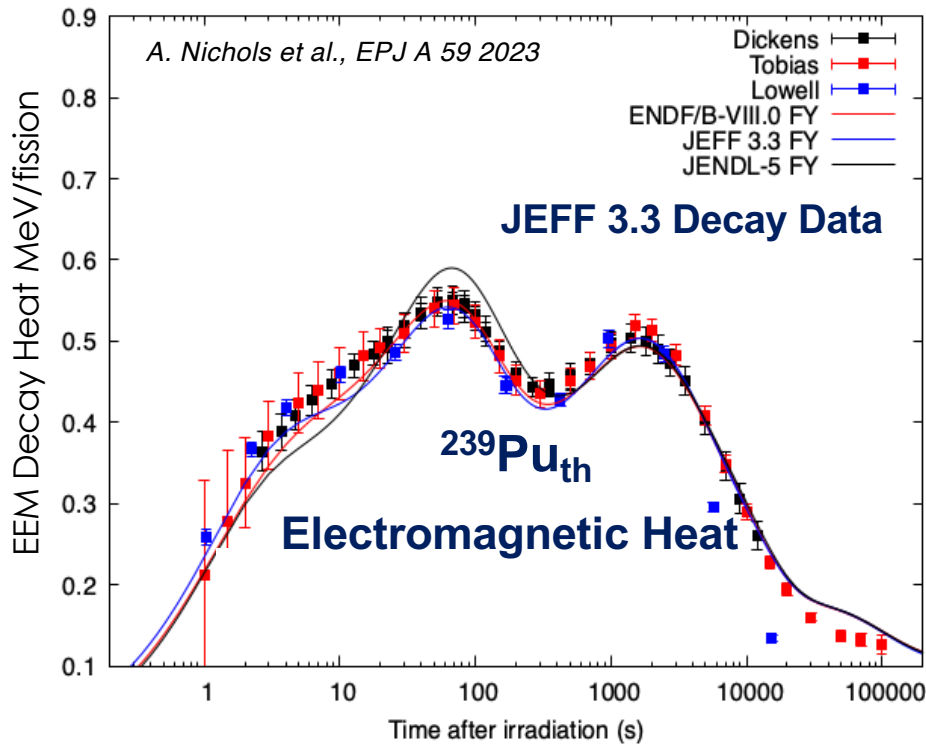
Limited set of data, uncertainty ...



Extra fission pulses measurements needed !

CoNDERC IAEA Database

	Nucleus	Measurements	Cooling time	Uncertainty Range %	Year	Authors
Fission induced per thermal neutrons	^{235}U	β, γ	$0.4 - 10^5 \text{ s}$	3.2% - 9.5 % 2.5% - 24.1% 2.5% - 8.6%	1989, 1980,1997	Dickens, Tobias, Lowell, others
	^{239}Pu	β, γ	$1 - 10^5 \text{ s}$	2.3% - 6.9% 2.7% - 60.9% 2% - 9.9%	1989, 1980,1997	Dickens, Tobias, Lowell, others
	^{241}Pu	β, γ	$3 - 1.2 \cdot 10^4 \text{ s}$	4% - 11.1%	1980, 1989	Dickens
Fission induced per fast neutrons	^{239}Pu	β, γ	$19 - 2.4 \cdot 10^5 \text{ s}$	2.4% - 5.7%	1982	Akiyama
	^{233}U	β, γ	$19 - 2.4 \cdot 10^5 \text{ s}$	3.2% - 7.5%	1982, 2002	Akiyama, Ohkawachi
	^{235}U	β, γ	$19 - 2.4 \cdot 10^5 \text{ s}$	2.1% - 6.1%	1982	Akiyama
	^{238}U	β, γ	$0.4 - 2.4 \cdot 10^5 \text{ s}$	3.8% - 20% 1.8% - 5.3%	1997, 1998	Akiyama, Lowell
	^{232}Th	γ	$19 - 2.4 \cdot 10^5 \text{ s}$	3.7% - 9.2%	1997	Akiyama
	^{237}Np	γ	$264 - 2 \cdot 10^4 \text{ s}$	not given	2002	Ohkawachi



Published TAGS measurements so far ...

TAS Collaboration : IFIC Valencia, Univ. of Surrey, Subatech/SEN team

4 measurement campaigns (2007, 2009, 2014, 2022) @Jyväskylä

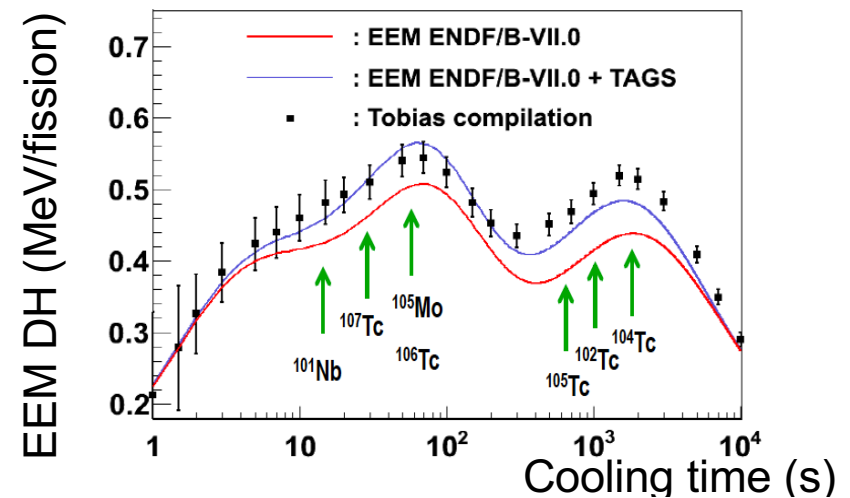
Isotope	Isotope	Isotope
35-Br-86 ^{†*}	41-Nb-99 [†]	52-Te-135 [†]
35-Br-87 ^{†*}	41-Nb-100 ^{†*}	53-I-136 [†]
35-Br-88 ^{†*}	41-Nb-101 ^{†*}	53-I-136m [†]
36-Kr-89 [†]	41-Nb-102 ^{†*}	53-I-137 ^{†*}
36-Kr-90 [†]	42-Mo-103 ^{†*}	54-Xe-137 [†]
37-Rb-90m	42-Mo-105 [*]	54-Xe-139 [†]
37-Rb-92 ^{†*}	43-Tc-102 ^{†*}	54-Xe-140 [†]
38-Sr-89	43-Tc-103 ^{†*}	55-Cs-142 [*]
38-Sr-97	43-Tc-104 ^{†*}	56-Ba-145
39-Y-96 [†]	43-Tc-105 [*]	57-La-143
40-Zr-99 [†]	43-Tc-106 [*]	57-La-145
40-Zr-100 [†]	43-Tc-107 [*]	
41-Nb-98 ^{†*}	51-Sb-132 [†]	

[†] : also important for ²³²Th/²³³U cycle

A. Algora et al., EPJA 57 (2021)

Parent nuclides identified per WPEC-25 for TAGS meas. for ²³⁵U/²³⁹Pu reactors, (NEA, T. Yoshida/ A. Nichols, 2007)

Algora et al., TAGS PRL 2010



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Algora et al., TAGS PRL 2010

+ ^{91,94,95}Rb, ^{100m,102m}Nb, ^{96m}Y

[†] : also important for ²³²Th/²³³U cycle

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TAS Collaboration : IFIC Valencia, Univ. of Surrey, Subatech/SEN team
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MTAS Collaboration : Univ. of Warsaw, ORNL, Univ of Tennessee
Experiences @ Argonne National Laboratory's CARIBU facility

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+ ^{91,94,95}Rb, ^{100m,102m}Nb, ^{96m}Y

+ ^{89,90}Rb

[†] : also important for ²³²Th/²³³U cycle

Impact of TAGS data on Decay heat calculations

Selected examples

- Review paper coordinated per IAEA (*Nichols et al., EPJ A 59 2023*)

Table 1 Irradiated fuel inventories and decay-heat calculations [38,39,40].

thermal neutron pulse (0.0253 eV)	^{235}U , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am , ^{242m}Am , ^{243}Am , ^{243}Cm , ^{245}Cm
fast neutron pulse (400 keV or 500 keV)	^{232}Th , ^{233}U , ^{238}U , ^{237}Np

Systems chosen to compare to FISPACT-II
DH calc. with classical libraries (ENDF/B-
VII.1, JEFF3.1.1, JENDL4-0)

M. Fleming, J. C. Sublet, 2015, CCFE-R15-28

DH experimental meas. available
in IAEA CONDERC database

- For each fissioning system:

3 sets of DH calculations combining the same FY library each time with :

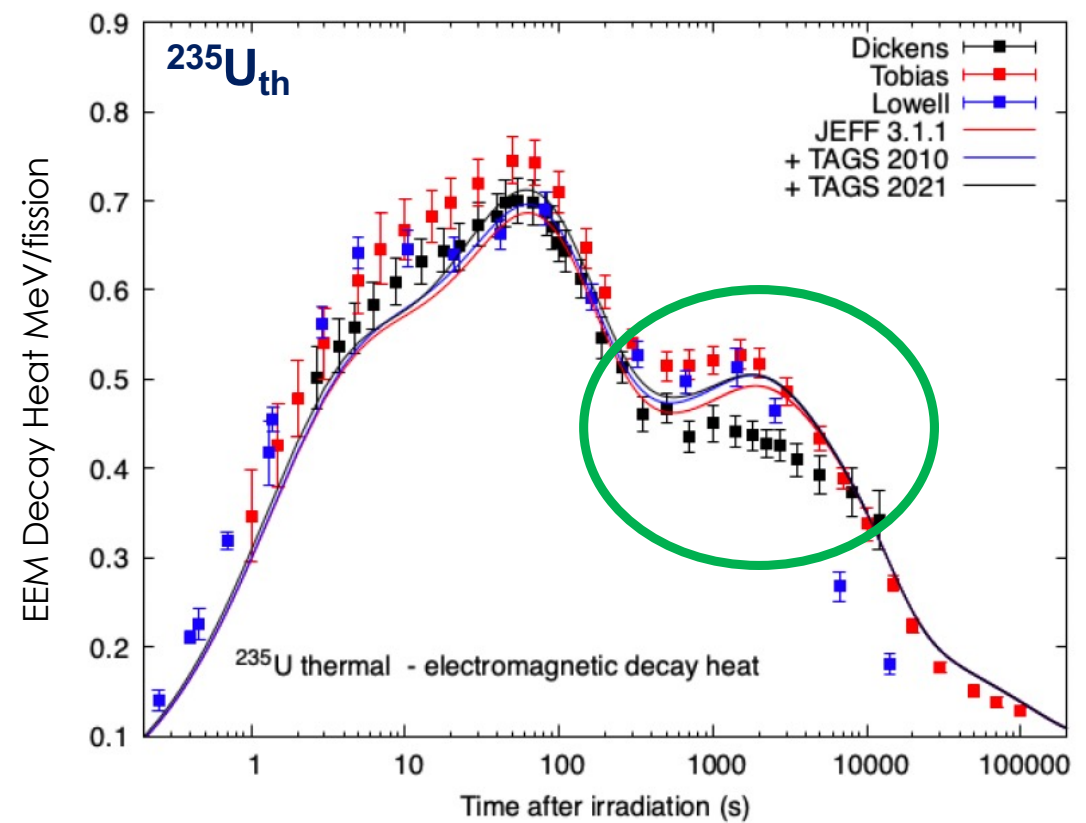
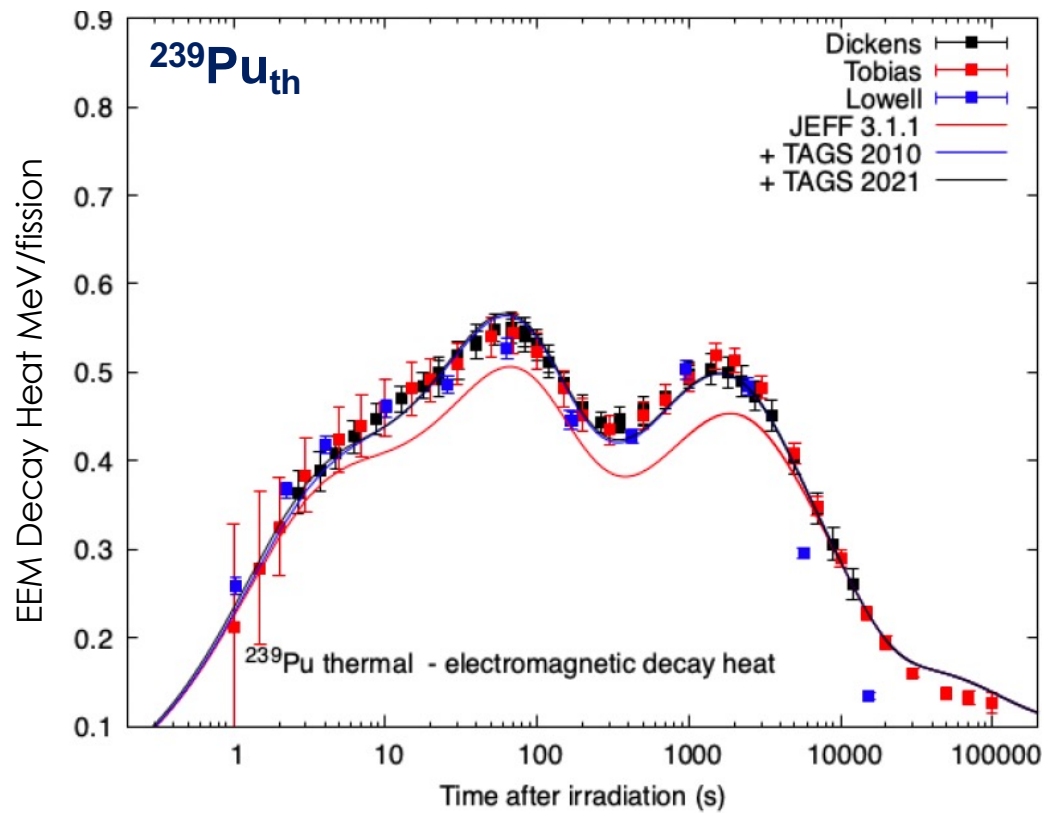
- Decay Data **without the Algora 2010 TAGS data: reference library or baseline**
- Decay Data **with the Algora 2010 TAGS data : + TAGS 2010**
- Decay Data **with the 2021 TAGS published data : + TAGS 2021**

Serpent2 used for DH with JEFF libraries

*But also used for cross-checks on DH with ENDF (FISPACT-II/P. Dimitriou)
& JENDL (OYAK98/ T. Yoshida & F. Minato)*

Impact of TAGS data on Decay heat calculations

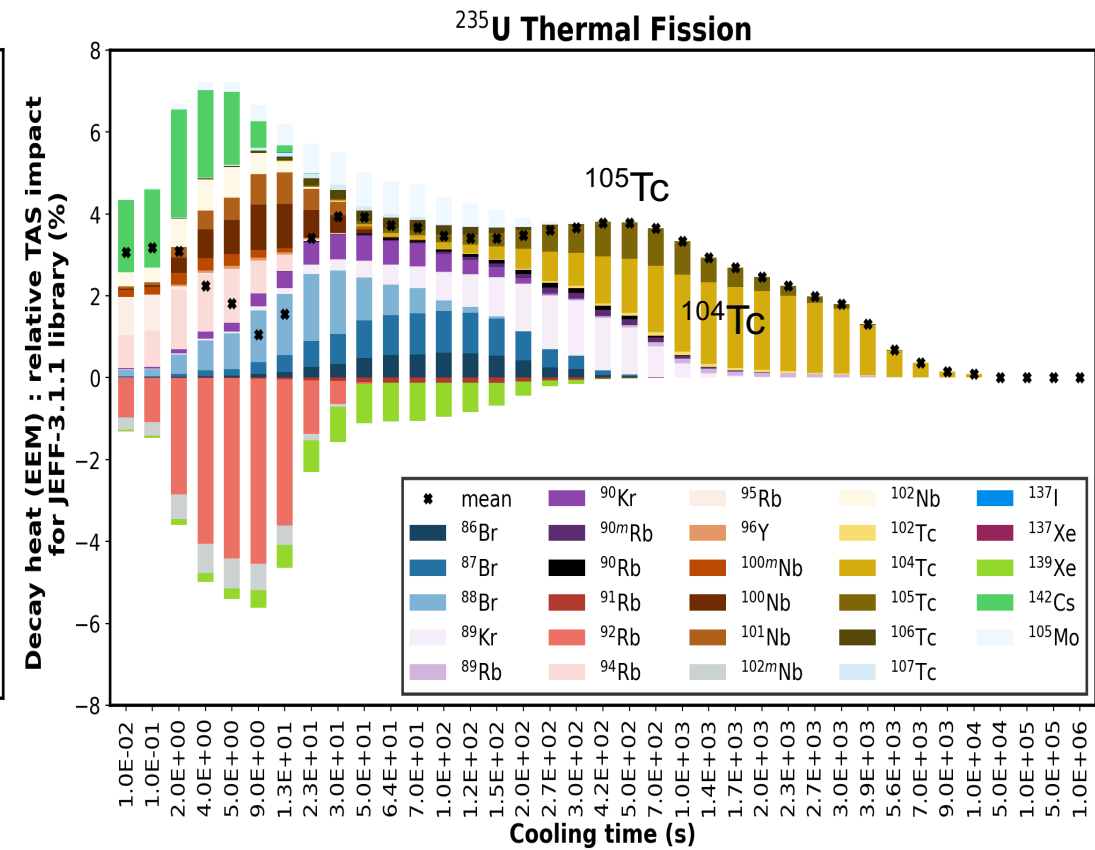
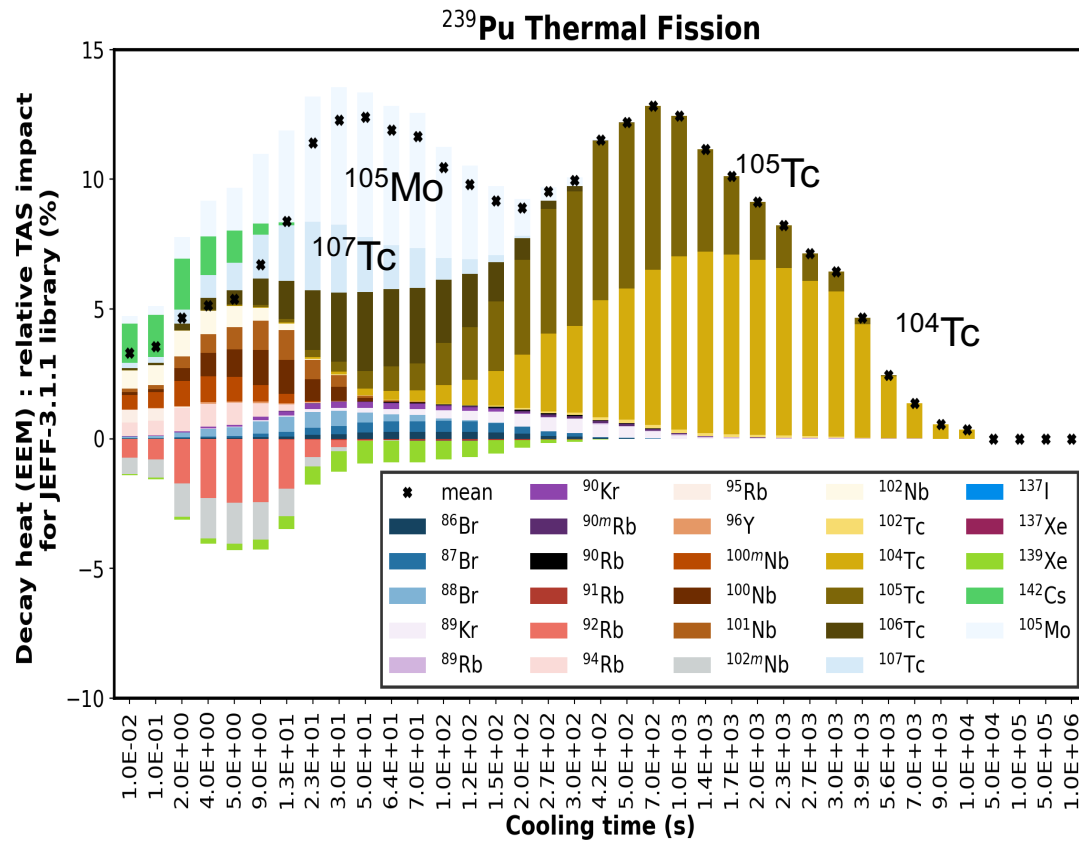
Selected examples



Same conclusions with ENDF library

Impact of TAGS data on Decay heat calculations

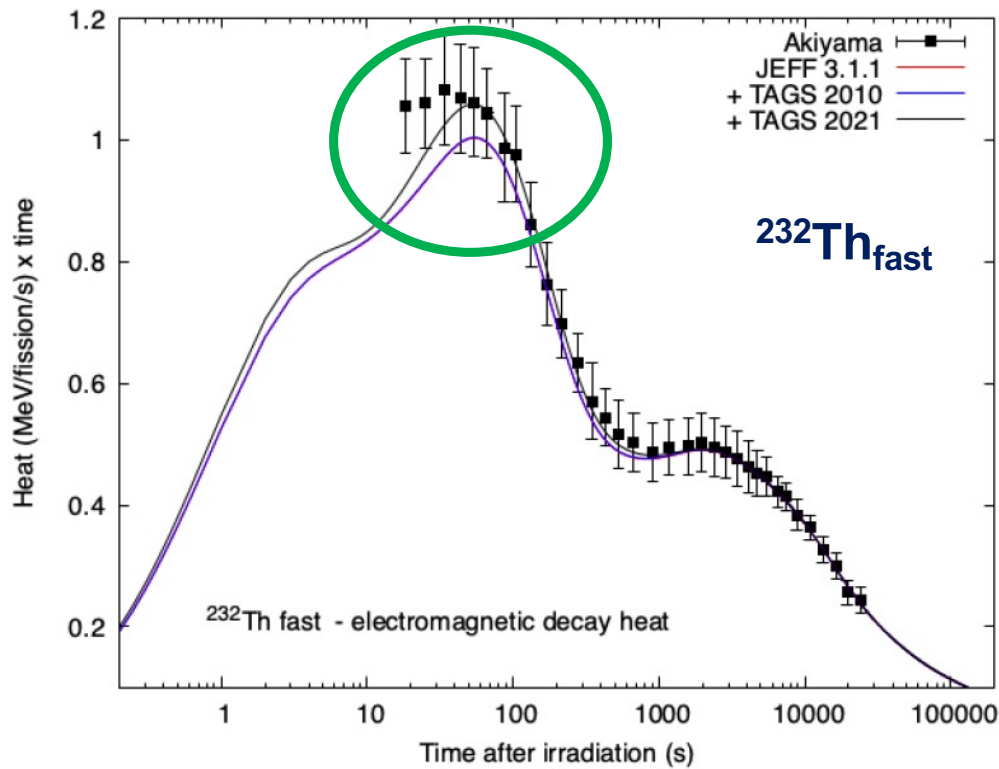
Selected examples



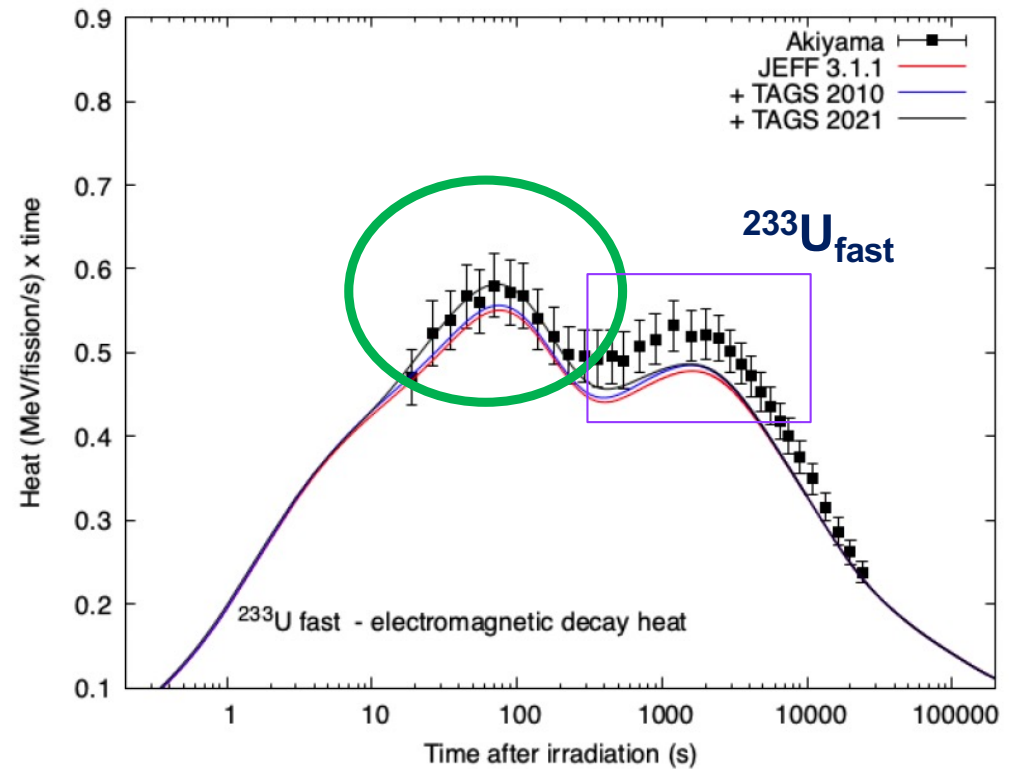
Impact of TAGS data on Decay heat calculations

Selected examples

+ TAGS 2021 : improved agreement of EEM component for $^{233}\text{U}_{\text{fast}}$, $^{238}\text{U}_{\text{fast}}$, $^{232}\text{Th}_{\text{fast}}$, for cooling time below 100s



But also need of new DH fission pulse experiments 😊



Need to investigate key FPs for cooling range > 100s

Decay heat for the U/Pu industrial cycle

- « Validation of the existing codes with measured SNF decay heat is currently considered as a necessity by most of existing national and international regulations » *

IAEA Safety Standards

for protecting people and the environment

Design of the
Reactor Core for
Nuclear Power Plants

Specific Safety Guide

No. SSG-52



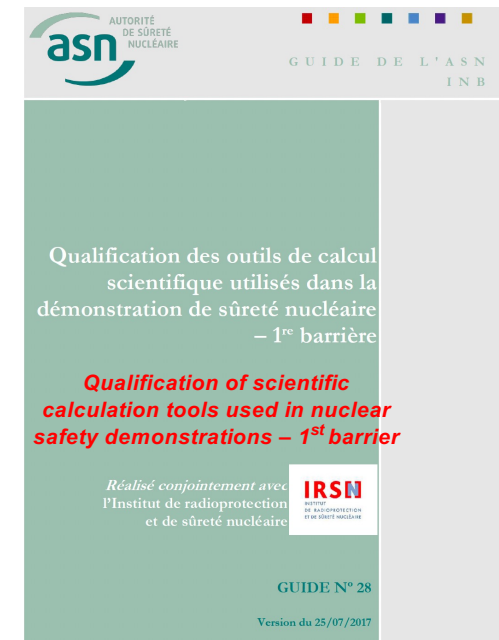
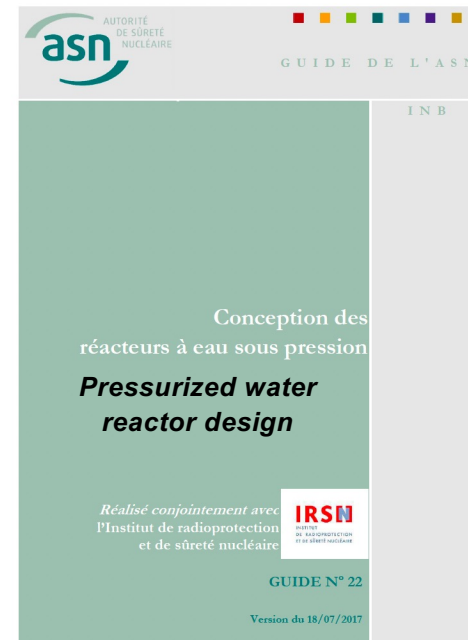
IAEA Safety Standards

for protecting people and the environment

Design of the Reactor
Coolant System and
Associated Systems for
Nuclear Power Plants

Specific Safety Guide

No. SSG-56

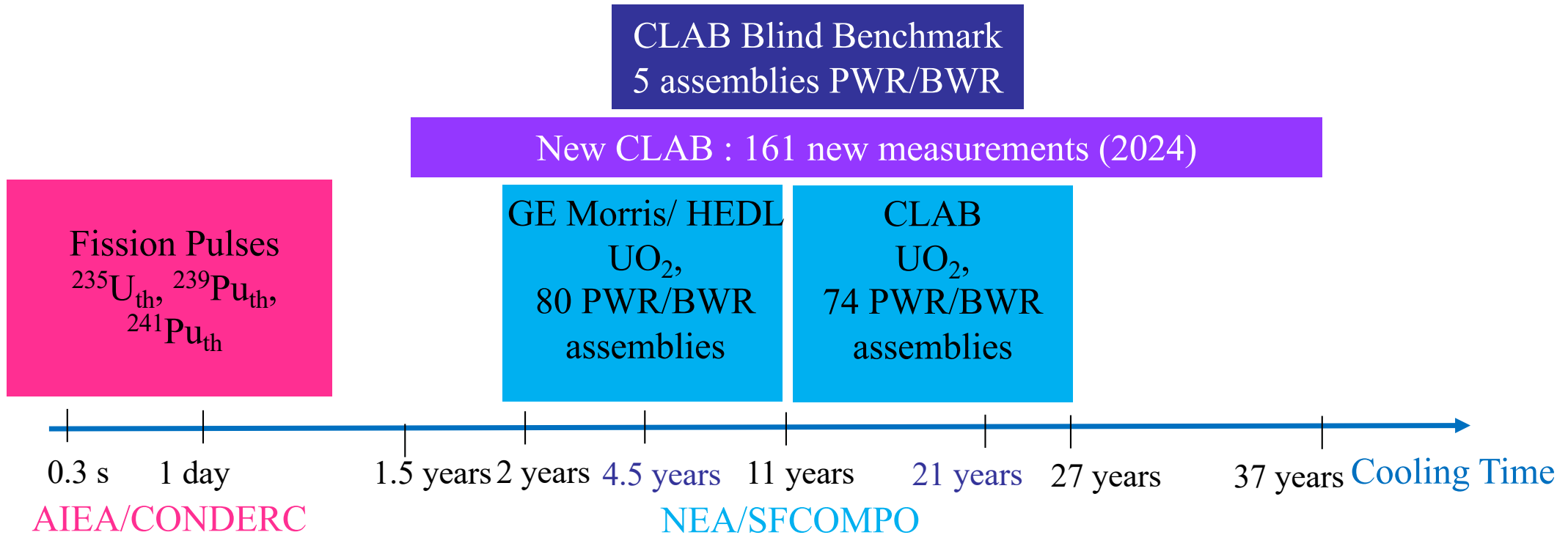


Validation => Code + given nuclear data library

*: An introduction to Spent Nuclear Fuel decay heat for Light Water Reactors: a review from the NEA WPNCs, D. Rochman et al., EPJN 10 9 (2024), Review paper of the NEA/SG12 on decay heat

Decay heat measurements for the U/Pu cycle

Available decay heat measurements



Limits :

- max. Burnup: 51 GWd/t, low enrichment
- Missing cooling time ranges
- Uncertainty measurements :
HEDL: 10%, GE-Morris : 3-4%, CLAB : 1-2%
- No measurements for MOX assemblies

New measurements :

- Calorimeter @CLAB still operating, unique
- **161 new Decay Heat measurements**
EPRI report (Electric Power Research Institute), October 2024
- New calorimeter NAGRA, Gösgen powerplant

Objectives

- Characterize the state of the art of calculations ... without an access to the measurements before ... & same initial informations
- 27 laboratories/companies, codes Monte-Carlo & deterministic codes, many libraries

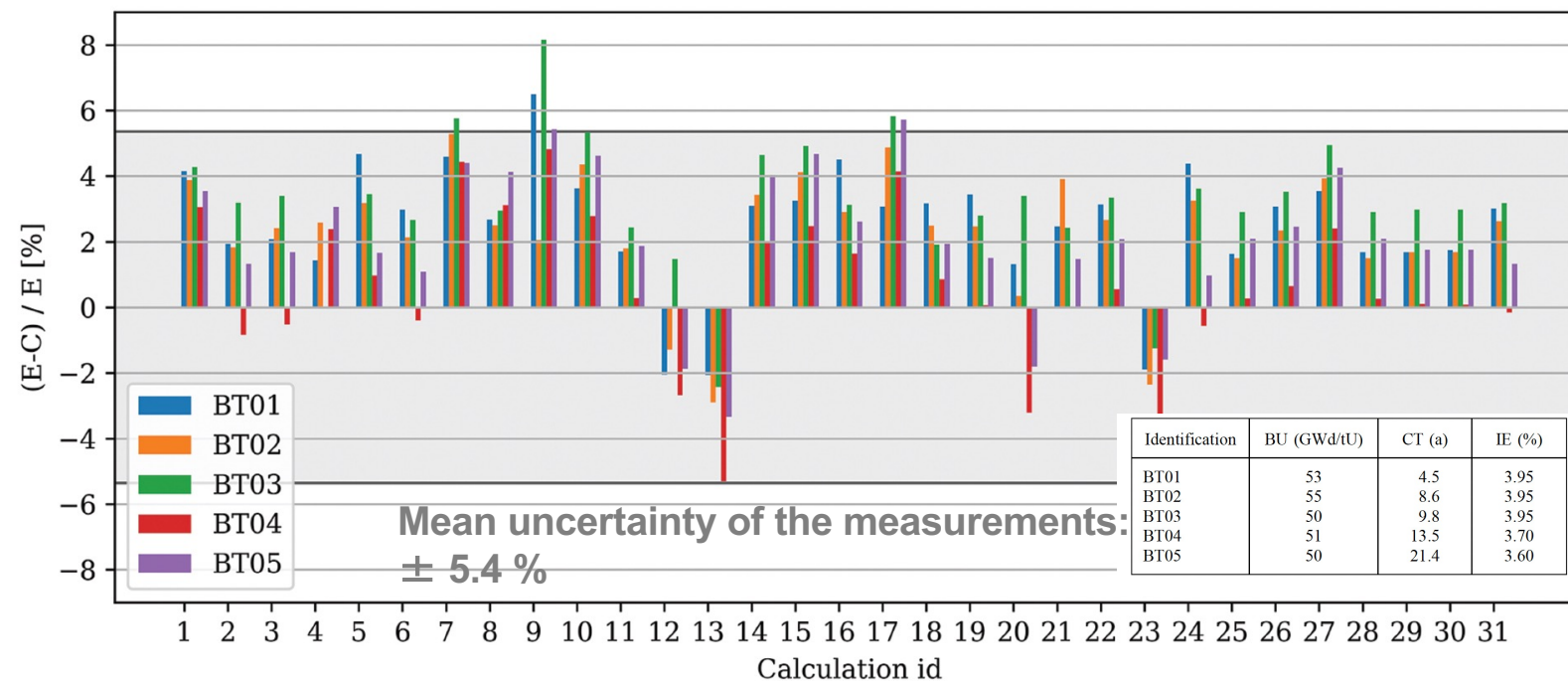
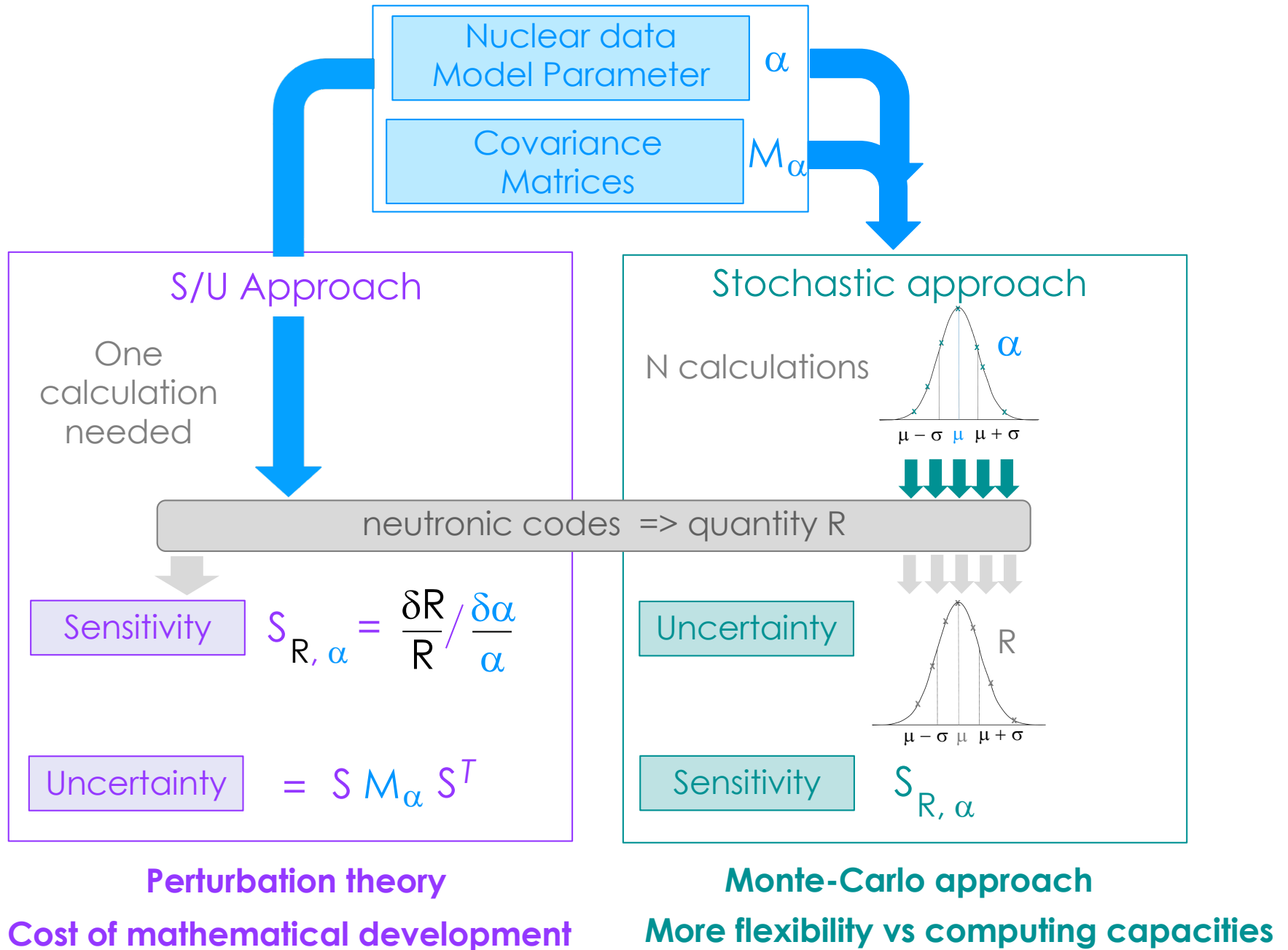


Fig. 8. Relative difference between measured (E) and calculated (C) decay heat rate values for the five different assemblies studied.

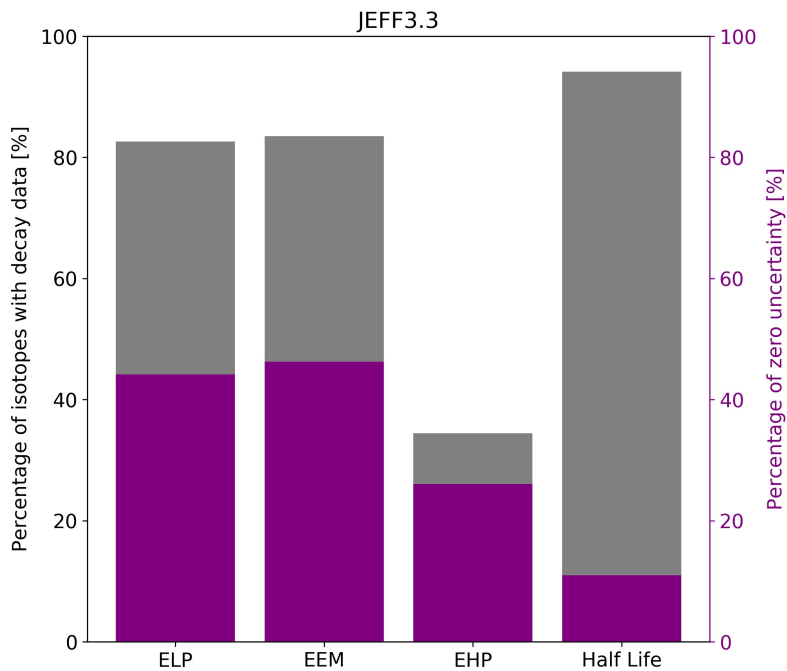
- First exercise at the international level !
- Importance of open source measurements & with lower uncertainties !

Uncertainty propagation



DD/FY inputs for uncertainty propagation

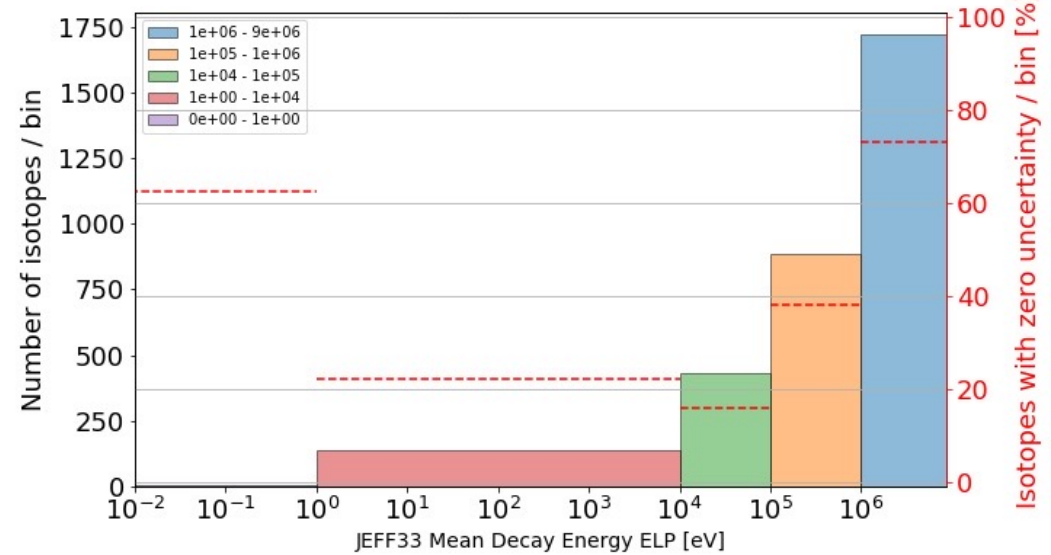
Decay Data



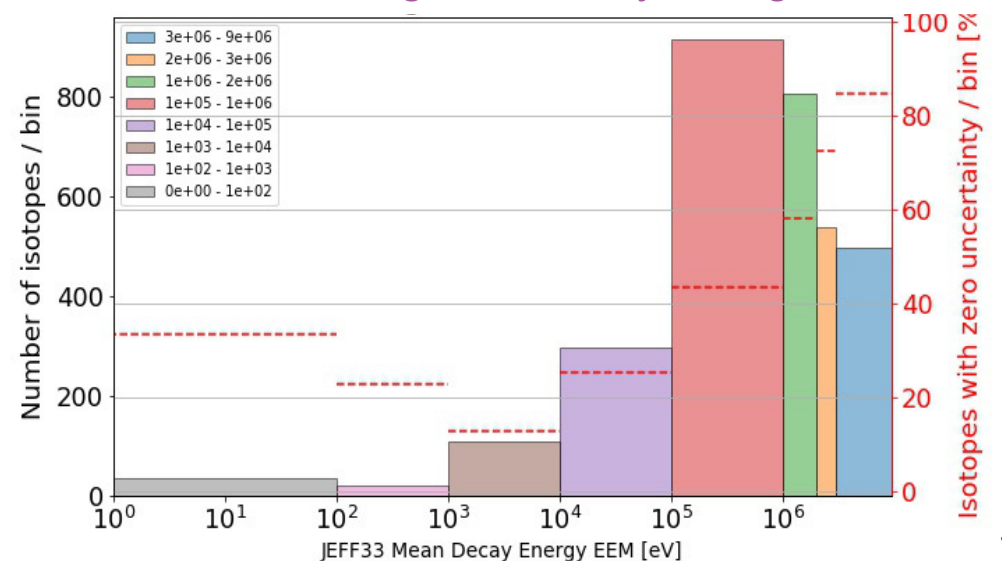
- Almost half of decay energies without uncertainty values

- Correlation matrices not available

ELP Decay energies

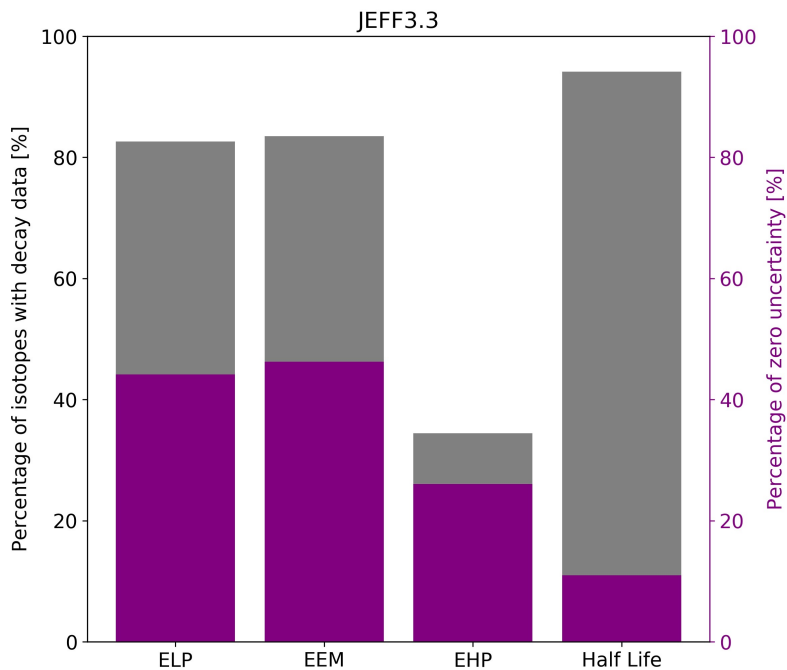


Electromagnetic Decay Energies



DD/FY inputs for uncertainty propagation

Decay Data



- Almost half of decay energies without uncertainty values

- Correlation matrices not available

Fission yields

Complete correlation matrices are very scarce

In evaluated libraries => mean values & std, but not correlation matrices !

- JEFF 4-0 :

CEA 2024: New FY evaluation

$^{235}\text{U}_{\text{th}}$ & $^{239}\text{Pu}_{\text{th}}$ with correlation matrices

- NEA/WPEC SG17 and after :

CEA:

FY covariances using Bayesian GLS, JEFF-3.1.1 data
N. Terranova et al., Nuc. Data Sheets 123 2015

GEF code:

derived from semi empirical model with parameters
K. H. Schmidt et al., Nuc. Data Sheets 131 2016

PSI:

Combination of Bayesian and Monte Carlo
D. Rochman et al., Annals of Nuc. Ener. 95 2016

SCK-CEN:

Covariances generated from JEFF-3.1.1 FY unc.
& Bayesian process integrating physical constraints
L. Fiorito et al., Annals of Nuc. Ener. 88 2016

Uncertainty propagation: codes development at CNRS



COCODRILO

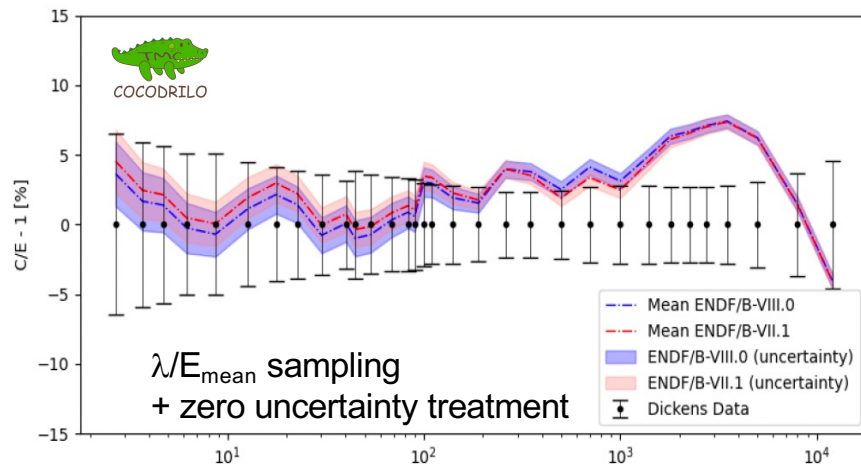
- Sampling of DD/FY : data with no-uncertainty
=> 0 vs 100%std to calc. impact
- Coupled to SERPENT2 code
- First application to fission pulses : cov FY on-going



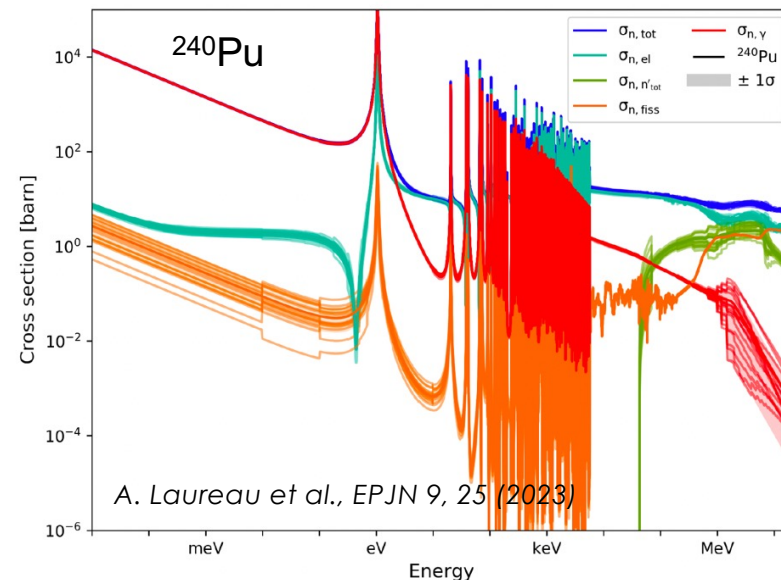
COCONUST

- Sampling of XS from covariance matrices (NJOY or others)
- Coupled to SERPENT2 or OpenMC codes
- First application to dosimetry : PETALE program @CROCUS, EPFL

$^{239}\text{Pu}_{\text{th}}$ Decay heat



Y. Molla PhD, Subatech, 2025



Uncertainty propagation: codes development at CNRS



COCODRILO

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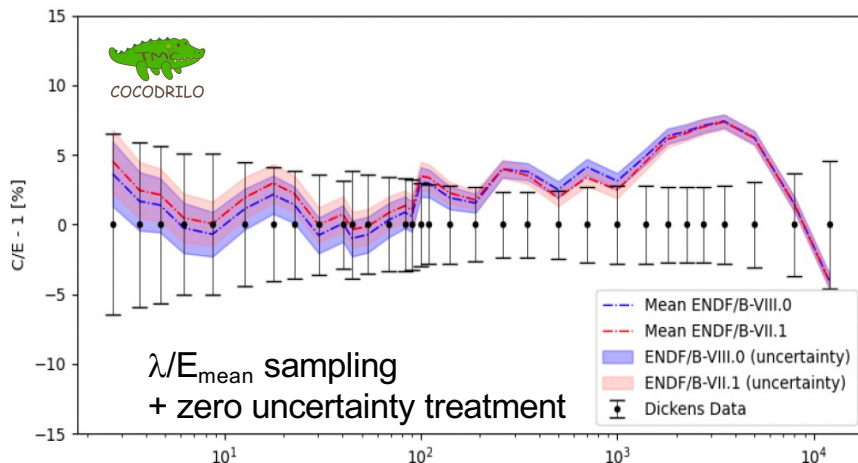


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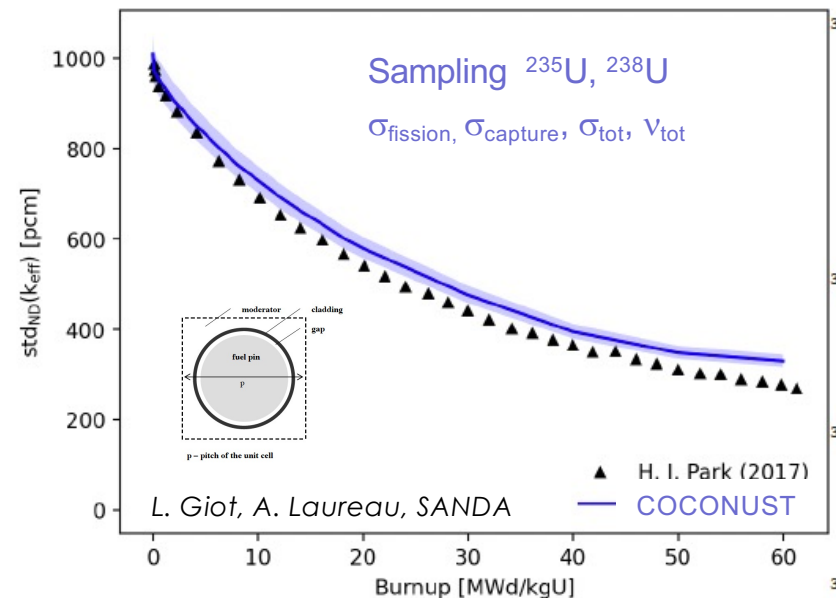
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A. Laureau et al., EPJN 9, 25 (2023)

$^{239}\text{Pu}_{\text{th}}$ Decay heat



Y. Molla PhD, Subatech, 2025

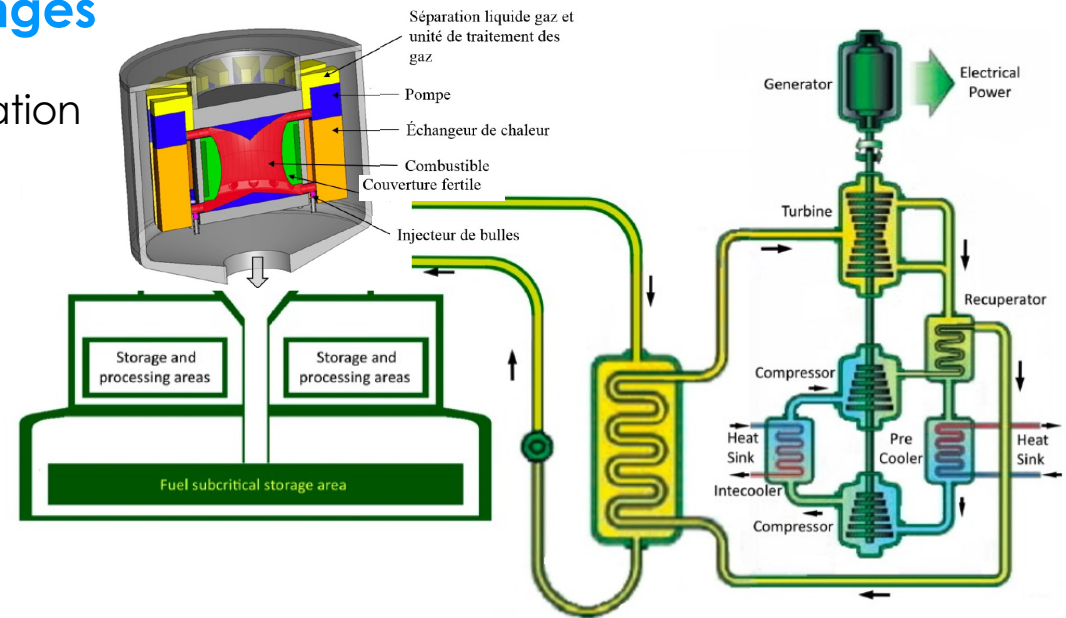


Multiple interests but also with R&D challenges

- Versatile applications, Advantages for safety & operation

- Decay heat needs/issues

- different localisations, barriers ?, EPUR systems
- DH values for scenarii
- Gen IV
 - => Feedback ?
 - => Nuclear data needs for depletion calc.
 - => Pandemonium nuclei ?



Fuel Depletion calculations

$$\frac{\partial N_i^C(t)}{\partial t} = \sum_{j \neq i} (b_{j \rightarrow i} \lambda_j + \langle \sigma_{j \rightarrow i} \phi \rangle) N_j^C(t) - (\lambda_i + \langle \sigma_i \phi \rangle) N_i^C(t) - \sum_{C \neq E} \lambda_{Treat.}^{C \rightarrow E} N_i^C(t)$$

$+\sum_{E \neq C} \lambda_{Feeding}^{E \rightarrow C} N_i^E(t)$ material supply
 (Eutectic control, criticality control per composition adjustment ...)

Fuel treatment (Chemistry & off-gas systems)



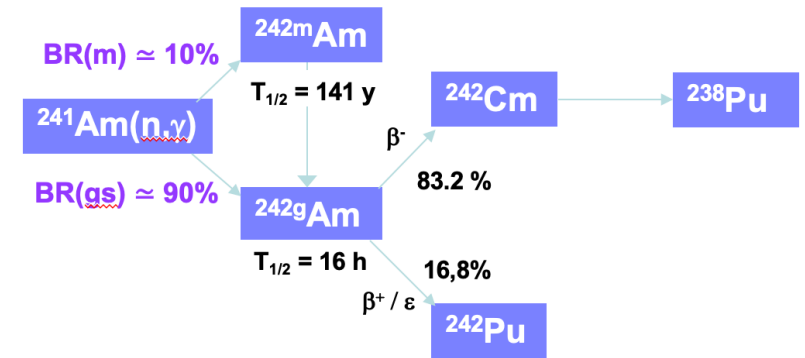
Dedicated depletion codes : REM (CNRS),
 Python code + existing depletion codes : CEREIS (CNRS), EQL0D (PSI), MOSARELA (CEA), PYMS (EDF)
 New developments : SCALE 6.3 or industrial codes based on OpenMC for example..

(n,2n), (n,g) reactions can lead to gs or isomeric states

=> isomeric branching ratio of $^{241}\text{Am}(n,\gamma)$ impact on Am, Pu and Cm inventories

=> important for core physics and fuel cycle calculations

Reactivity, spent-fuel (neutron, gamma activities, decay heat)



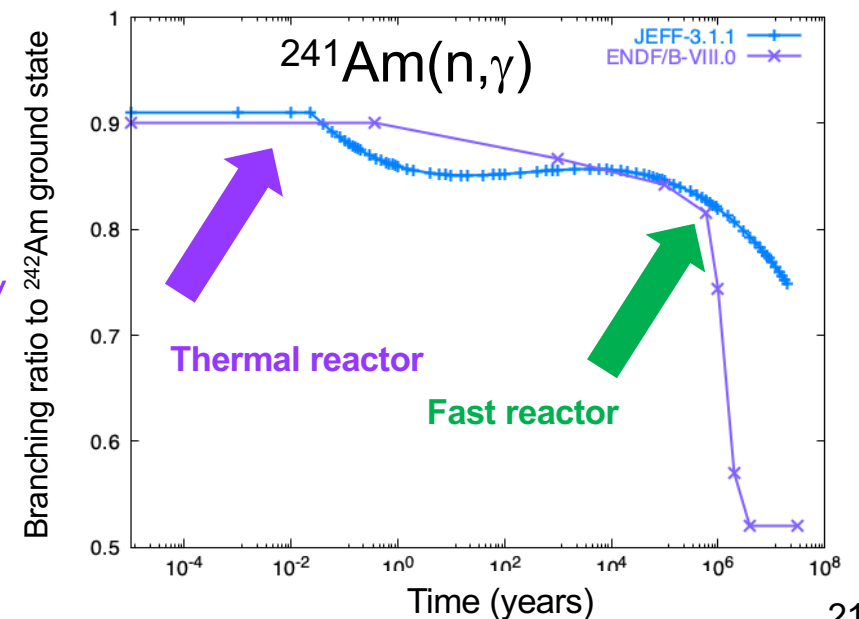
JEFF-3.1.1, Thermal data



Branching ratio (BR) depends on the neutron energy

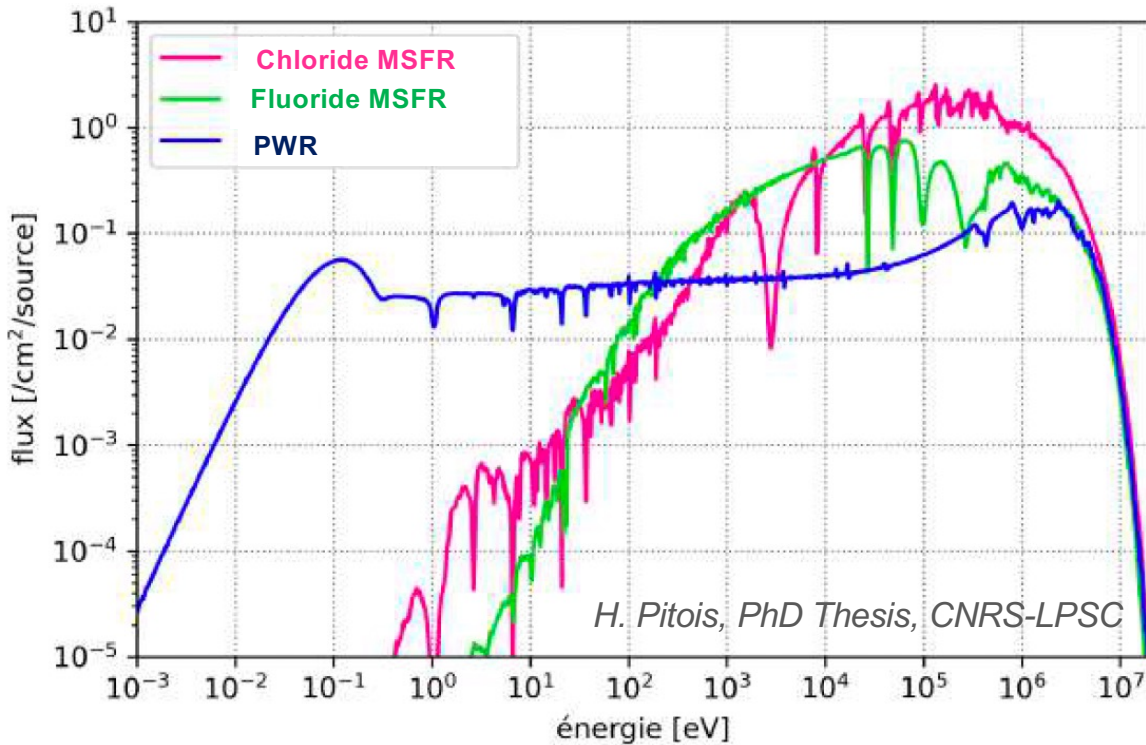
Most of the reactor codes use a single BR ratio

Usually thermal data are used per default



Selected example: Fission yields depend on the neutron energy

Neutron spectrum



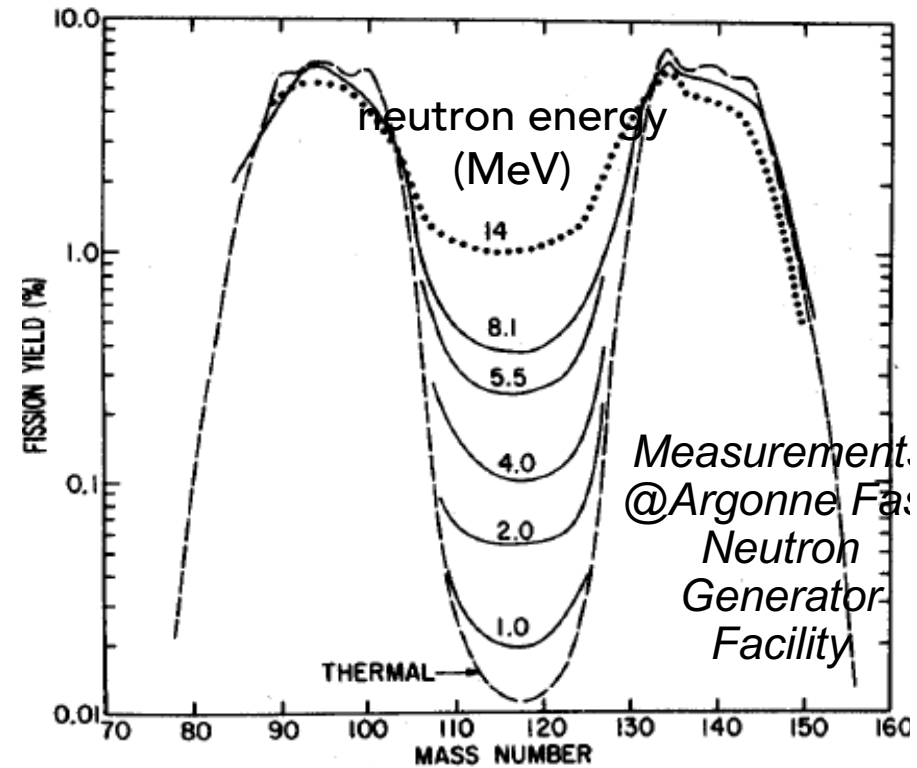
In evaluated libraries (JEFF, ENDF, JENDL),
divided in 3 groups of energy ...

Thermal
0.025 eV

Fast
400-500 keV

High
14-15 MeV

²³⁵U(n,fission)

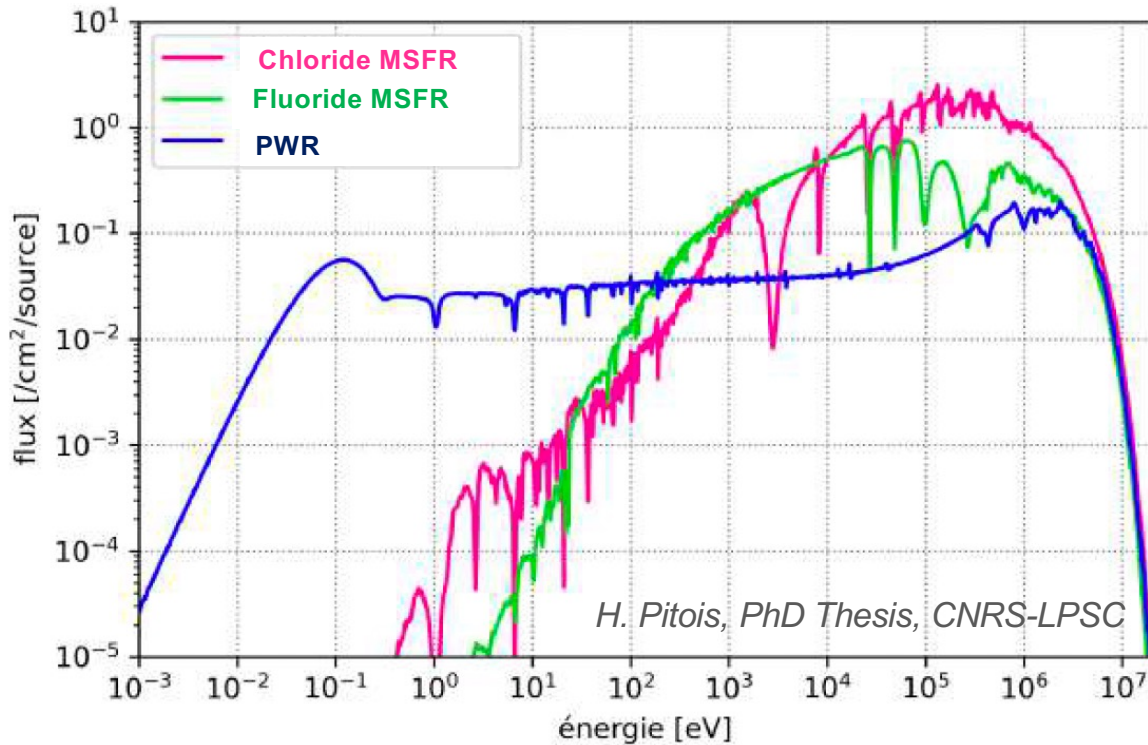


Glendenin et al., Phys. Rev. C24 6 (1981)

=> different E_n to handle (data & codes implementation)

Selected example: Fission yields depend on the neutron energy

Neutron spectrum



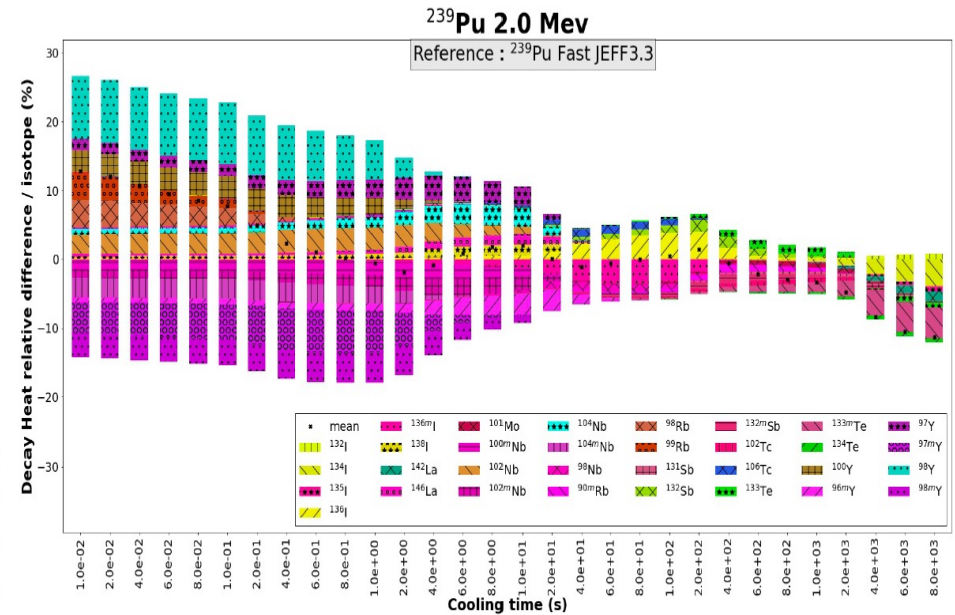
In evaluated libraries (JEFF, ENDF, JENDL),
divided in 3 groups of energy ...

Thermal
0.025 eV

Fast
400-500 keV

High
14-15 MeV

Also impact on decay heat



On- going work with GANIL
FY from D. Ramos et al., Phys. Rev. Lett 123 (2019)
F. Hervy' internship, Subatech, 2024

Conclusions & Outlooks

- Validation of Code + nuclear data library mandatory at the industrial level

Decay heat calculations needed at “different system sizes”

fission pulses, SNF assemblies, core level ...

new type of measurements to disentangle the effects of nuclear data ?

Increasing demand to assess uncertainties

tools and methods under development, needs of extra inputs for nuclear data

R&D drive for new reactors concepts & associated fuels open new needs

nuclear data, new decay heat measurements



**Importance of enhancing exchanges between the communities
(experimentalists, evaluators, reactor physicists & industrials)**

Acknowledgments

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- **IAEA:** J. C. Sublet, P. Dimitriou **Tokyo Institute of Technology:** T. Yoshida
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- **TAS collaboration** (IFIC Valencia, SEN team@Subatech, Univ. of Surrey)

For collaborative work or scientific discussions !

Thanks for your attention

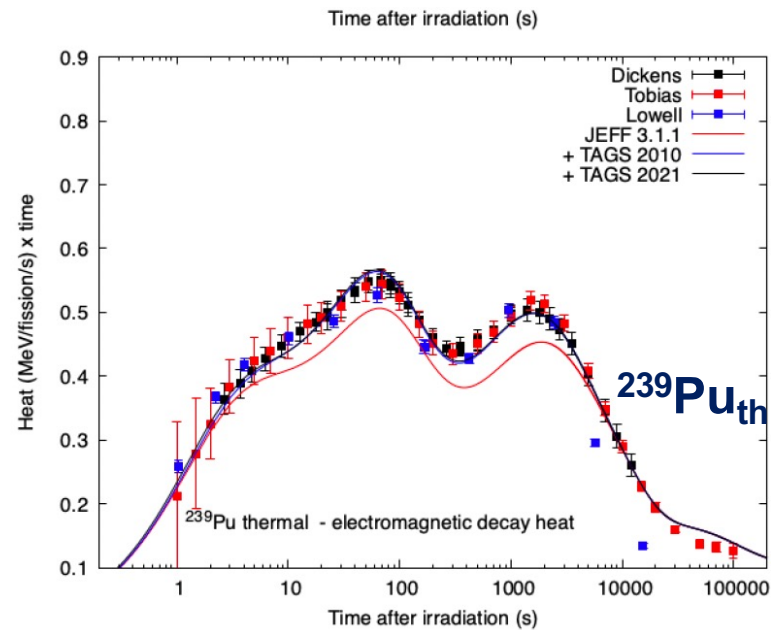
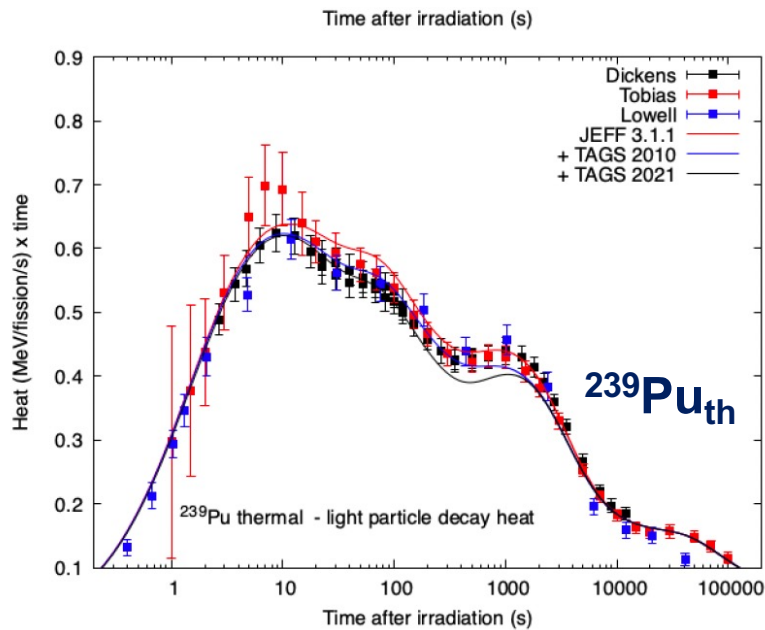
Backup

Impact of TAGS data on Decay Heat calculations

- For each fissioning system:

3 sets of DH calculations combining the same FY library each time with :

- Decay Data **without the Algora 2010 TAGS data: reference library or baseline**
- Decay Data **with the Algora 2010 TAGS data : + TAGS 2010**
- Decay Data **with the 2021 TAGS published data : + TAGS 2021**



Same conclusions
with ENDF library

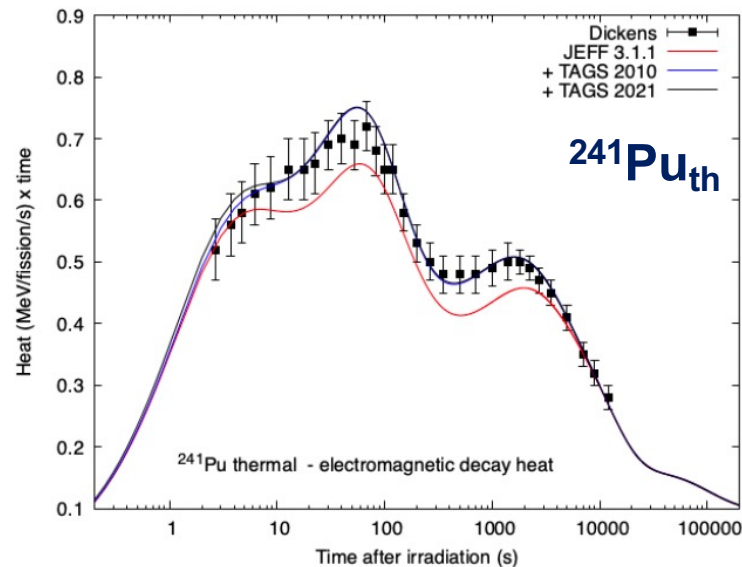
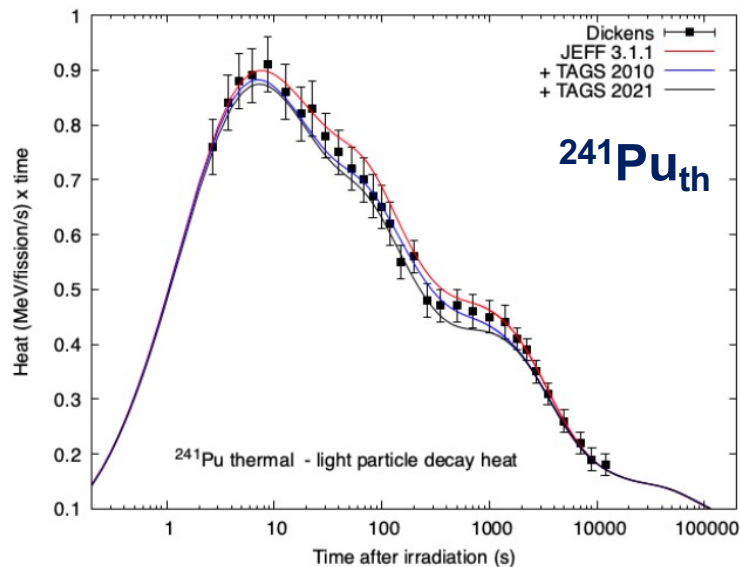
Impact of TAGS data on Decay Heat calculations

- For each fissioning system:

3 sets of DH calculations combining the same FY library each time with :

- Decay Data **without the Algora 2010 TAGS data: reference library or baseline**
- Decay Data **with the Algora 2010 TAGS data : + TAGS 2010**
- Decay Data **with the 2021 TAGS published data : + TAGS 2021**

+ TAGS 2010 : improved agreement for $^{239}\text{Pu}_{\text{th}}$, $^{241}\text{Pu}_{\text{th}}$ & $^{238}\text{U}_{\text{fast}}$
small impact for $^{232}\text{Th}_{\text{fast}}$ & $^{233}\text{U}_{\text{fast}}$



Same conclusions
with ENDF library

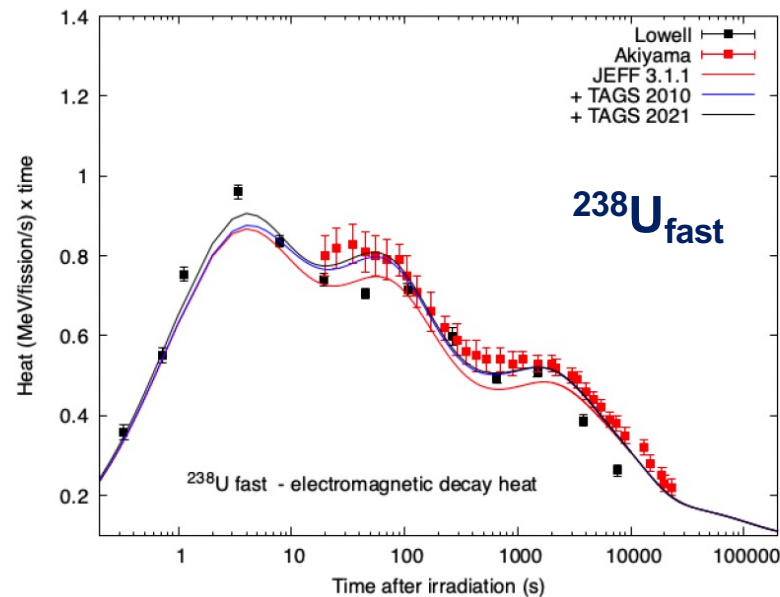
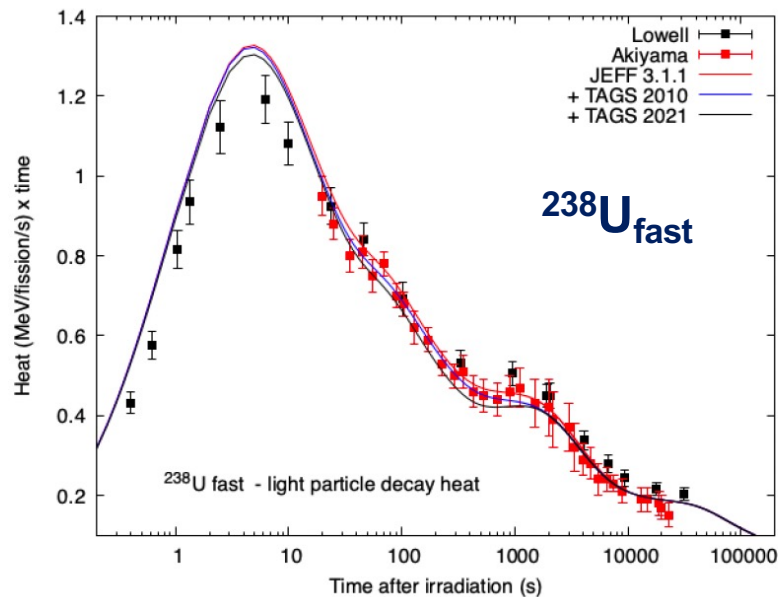
Impact of TAGS data on Decay Heat calculations

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Same conclusions
with ENDF library

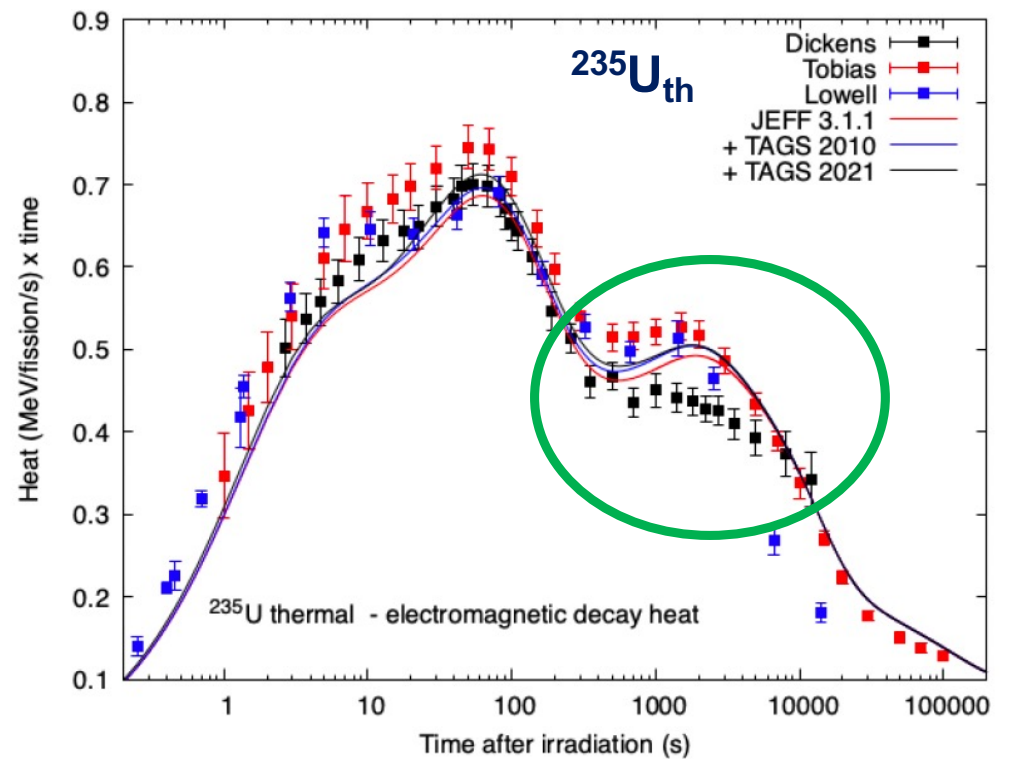
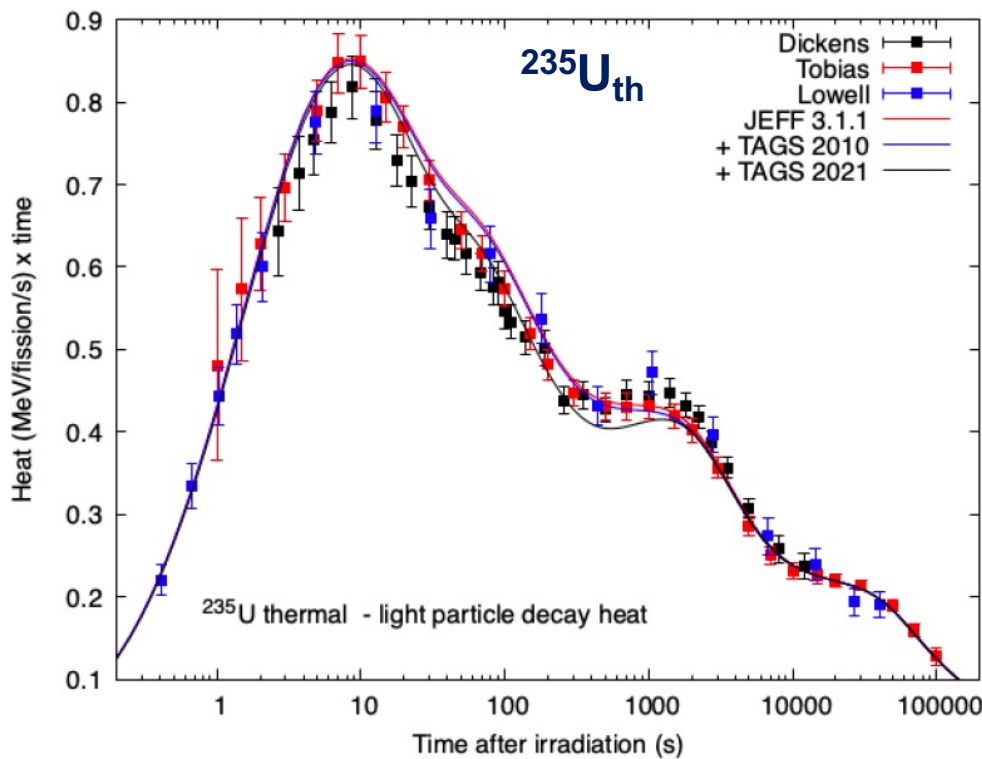
Impact of TAGS data on Decay Heat calculations

$^{235}\text{U}_{\text{th}}$

+ TAGS 2010 : no impact on ELP component

+ TAGS 2021 : ELP slightly improved in 10-400s but underestimation in 400-1000s

Hard to say on EEM wrt differences between the 3 experimental sets !

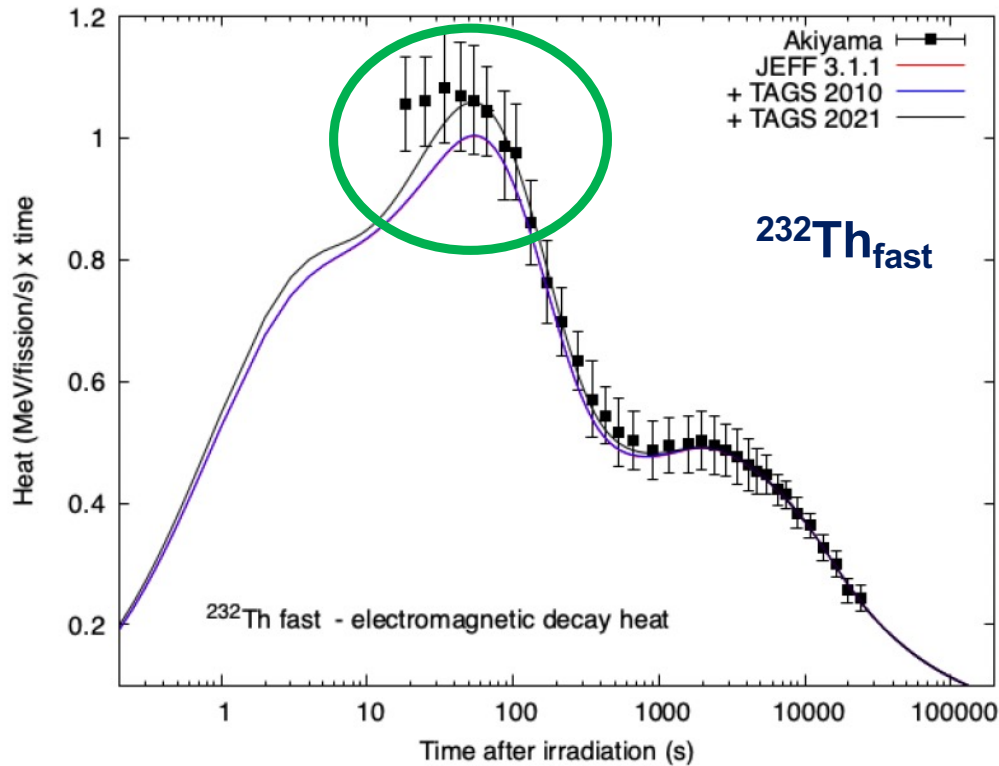


Same conclusions with ENDF library

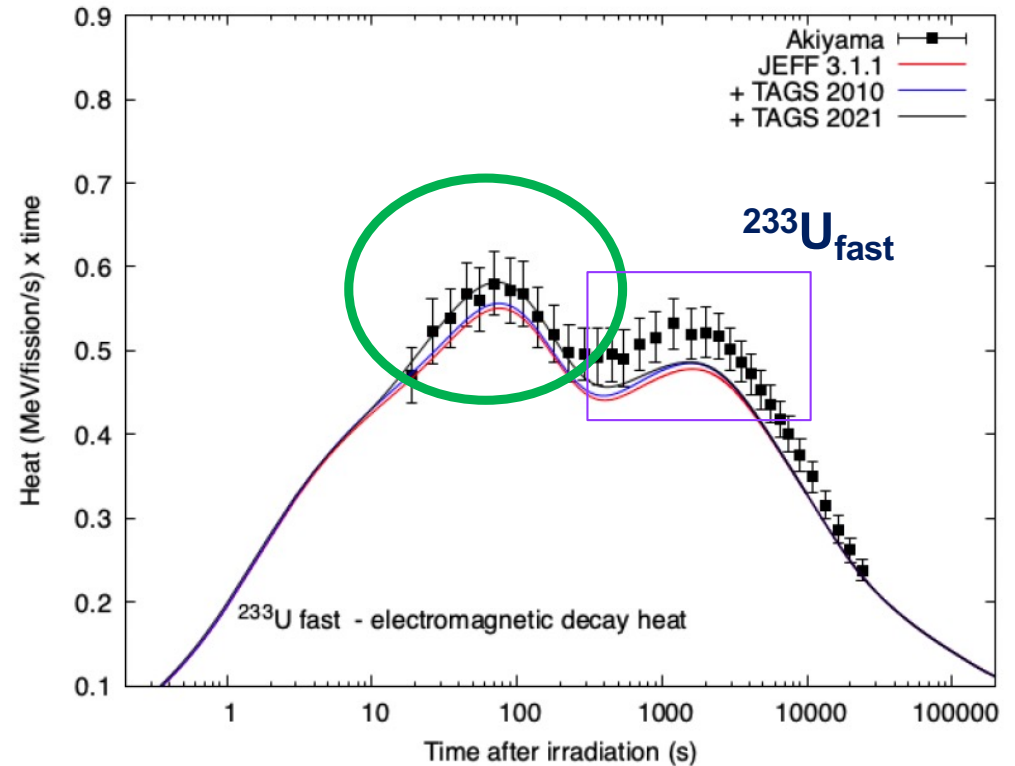
Need of new DH fission pulse experiments ☺

Impact of TAGS data on Decay Heat calculations

+ TAGS 2021 : improved agreement of EEM component for $^{233}\text{U}_{\text{fast}}$, $^{238}\text{U}_{\text{fast}}$, $^{232}\text{Th}_{\text{fast}}$, for cooling time below 100s



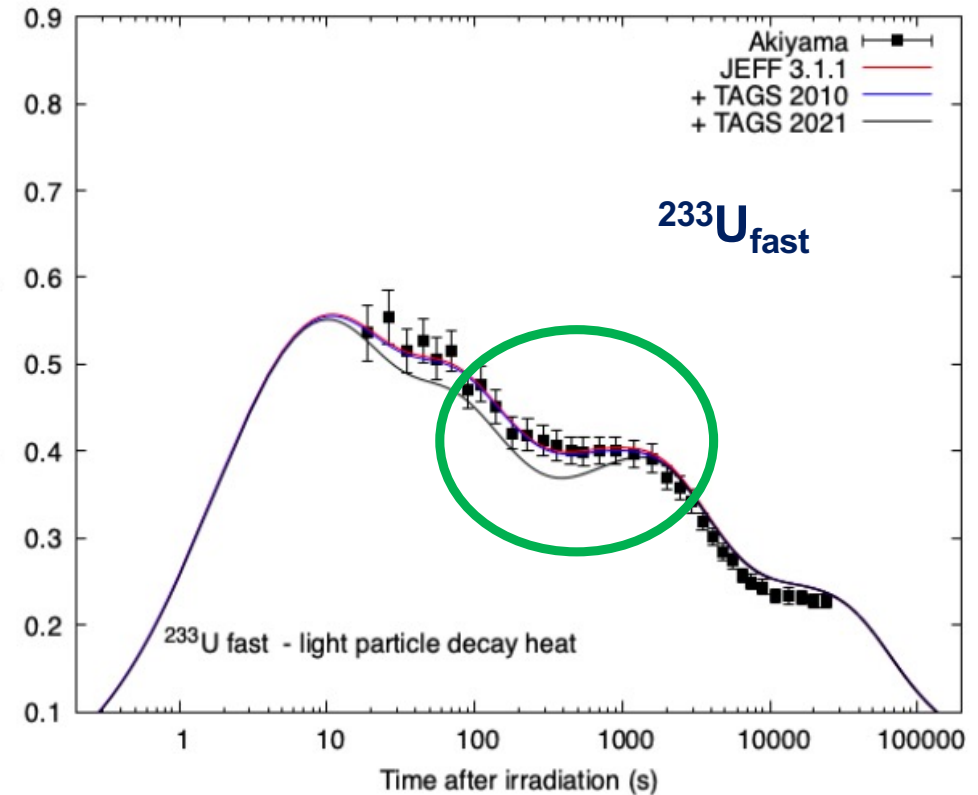
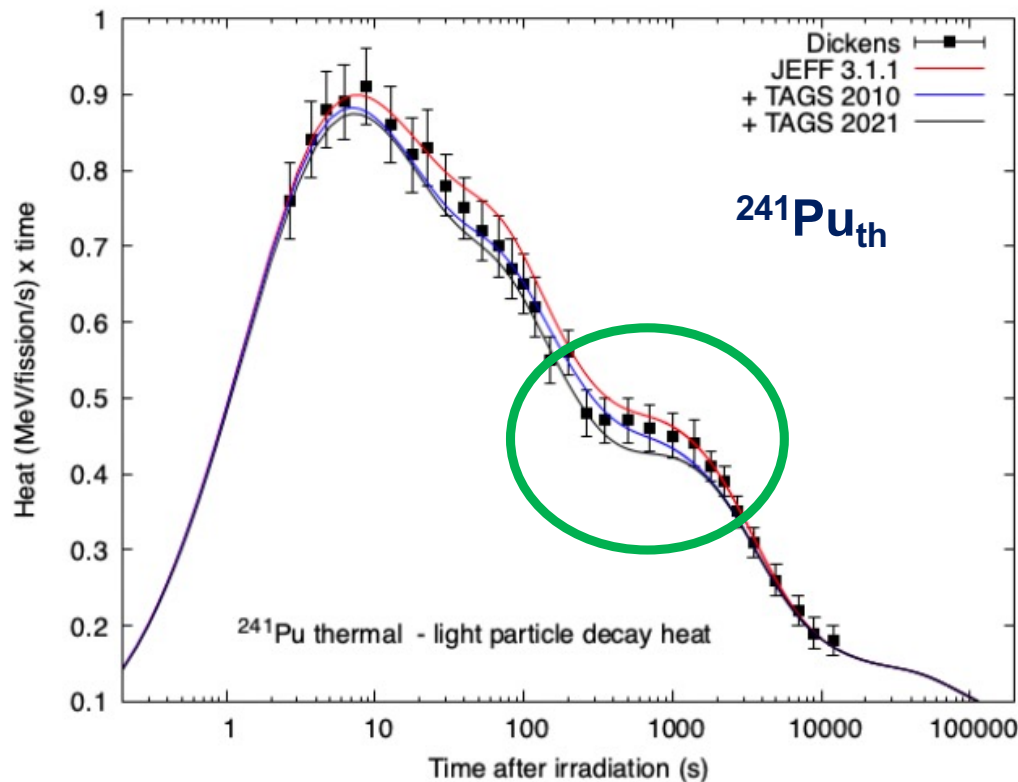
But also need of new DH fission pulse experiments 😊



Need to investigate key FPs for cooling range > 100s

Impact of TAGS data on Decay Heat calculations

+ TAGS 2021 : small under estimation of ELP component for $^{235}\text{U}_{\text{th}}$, $^{239}\text{Pu}_{\text{th}}$, $^{241}\text{Pu}_{\text{th}}$, $^{233}\text{U}_{\text{fast}}$ & $^{238}\text{U}_{\text{fast}}$ at cooling times ranging from 30s to 1000s



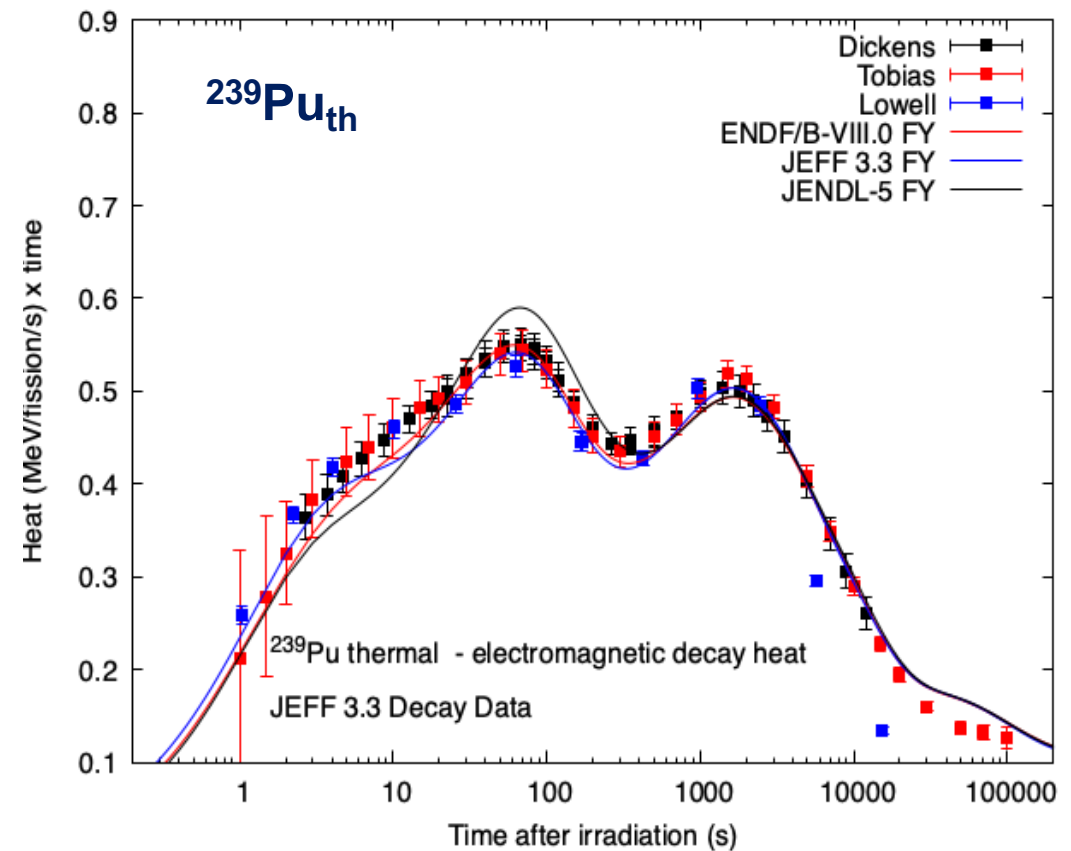
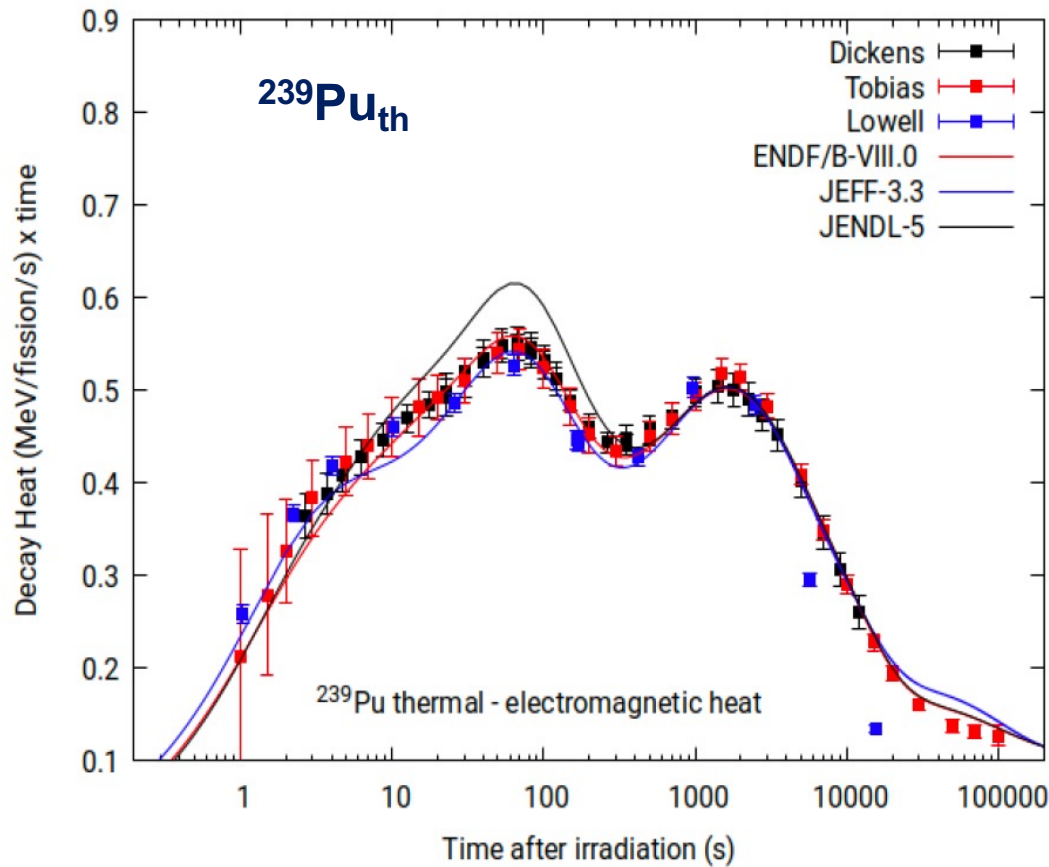
- => Only one set of experimental data, till in the errors bars for $^{241}\text{Pu}_{\text{th}}$
- => Needs for extra experimental data but also extra investigation on key FP suffering of Pandemonium effect
- => Also on going work to take into account FY and DD uncertainties through MC sampling

Same conclusions with ENDF library

Impact of TAGS data on DH calculations

Overestimation of EEM component for JENDL5 for $^{239}\text{Pu}_{\text{th}}$

Impact of Fission Yields ?



Decay heat measurements for the U/Pu cycle

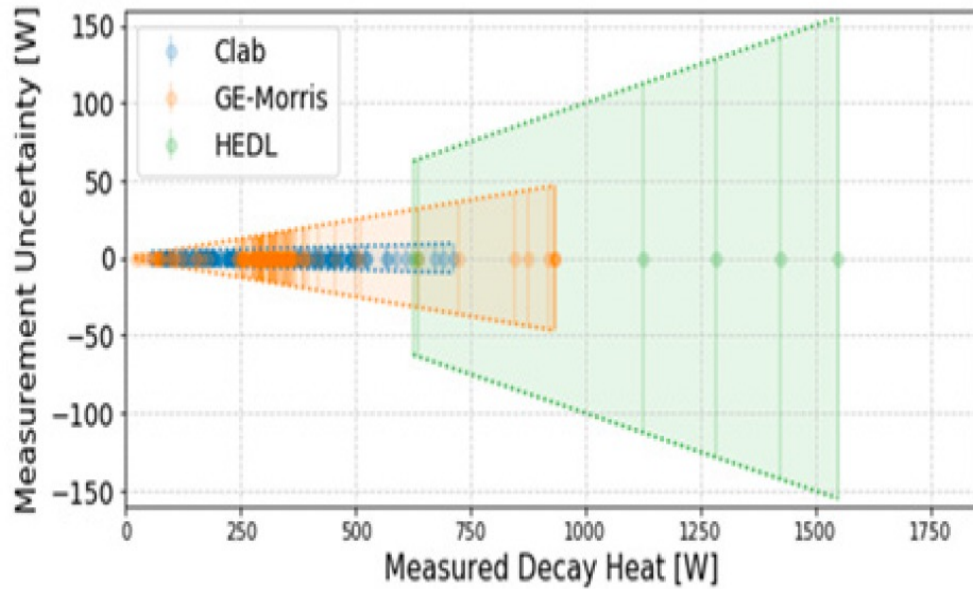


Figure 1. Decay heat measurement uncertainty for the available data sets [1]

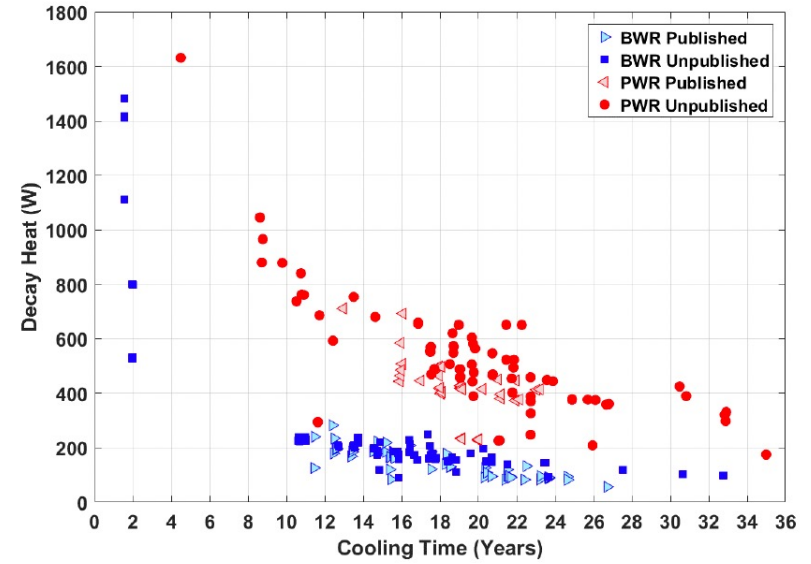


Figure 12. Cooling time versus measured decay heat for published and unpublished Clab decay heat measurements