New fission-product decay data measurements to improve decay heat calculations

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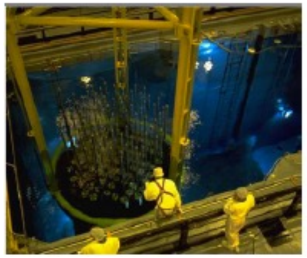
In collaboration with the TAS Collaboration (IFIC-Valencia, Univ. of Surrey, Subatech)

Part of this work is also included in an IAEA coordinated paper in preparation, submission foreseen this year
Exchanges with P. Dimitriou and T. Yoshida are acknowledged

Motivations on 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



Nuclear stage impacted	Time of cooling
Safety systems of cooling	0.1s to 8 days
Unloading of assemblies from core	5 to 25 days
Fuel transport	1 to 10 years
Reprocessing, vitrification, storage	4 to 3000 years
Storage	50 to 300 000 years and more

Decay Heat calculations

Summation Formula

$$DH(t) = f(t) = \sum_{i}^{n} N_i(t_c)\, \lambda_i\, \overline{E}_i$$

N_i : Number of nuclei i at the cooling time t_c

λ_i : Decay constant of the nucleus i

$\overline{E}_i$: Total decay energy of the nucleus i

- Large time range: 10^{-1} to 10^{6} years
- **Important quantity to design the size/capacity of safety systems**

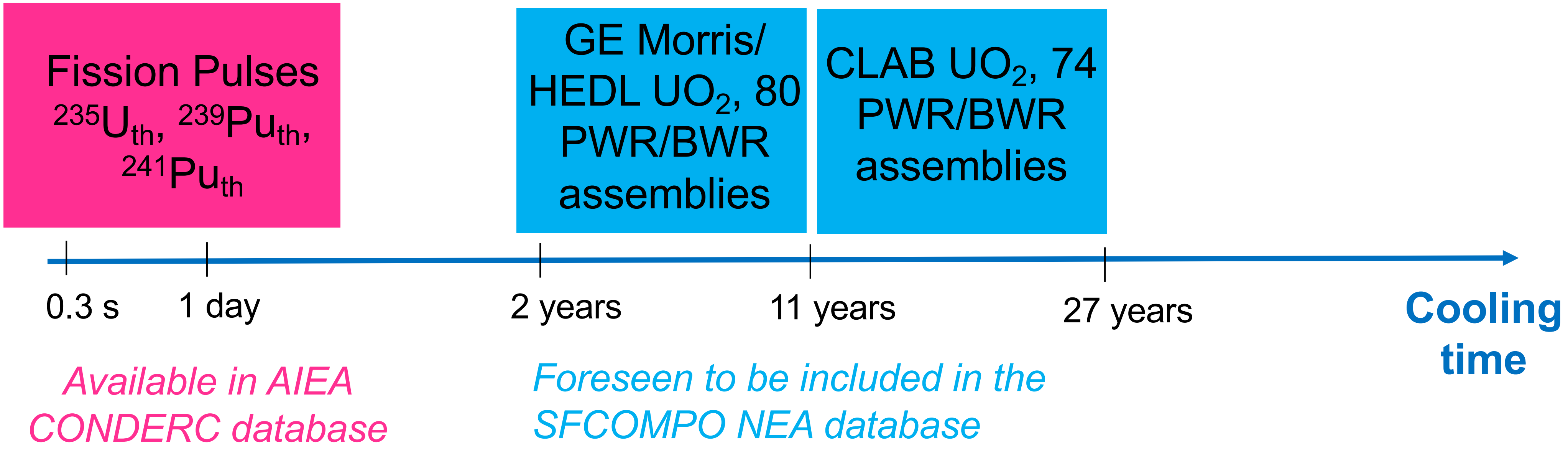
- Complex calculation (reactor modeling + depletion): quality of the code but also of the data !
- **~ 40 000 nuclear data: σ, $\overline{E}$, Branching Ratio, λ, Fission Yields, $\overline{\nu}$**

- Increasing will of safety authorities to ask for a precise calculation & detailed uncertainty quantification
- Interest of industry to reduce the uncertainty for economic reasons, with keeping the same level of safety
- For Gen IV reactors, most of codes developed/benchmarked for/on LWR reactors

➡ **Rigorous calculation with evaluated codes associated to experimental validation**
but also identification of biases in the calculation/data to improve them ….

Available Decay Heat Measurements for U/Pu cycle

Available decay heat measurements = Possible to get/use them ☺



- 2022 new Data@CLAB : DH of 5 assemblies PWR/BWR UO_2 cooling 4-21 years

Calculations performed in a blind way with same inputs: geometry, materials, reactor operation + cooling time, DH measurements given after the calculations..

Blind benchmark exercise for spent nuclear fuel decay heat, P. Jansson et al., Nucl. Sc. & Eng., 2022

- 60 new CLAB DH measurements foreseen (EPRI Report published soon)

Available Decay Heat Measurements for U/Pu cycle

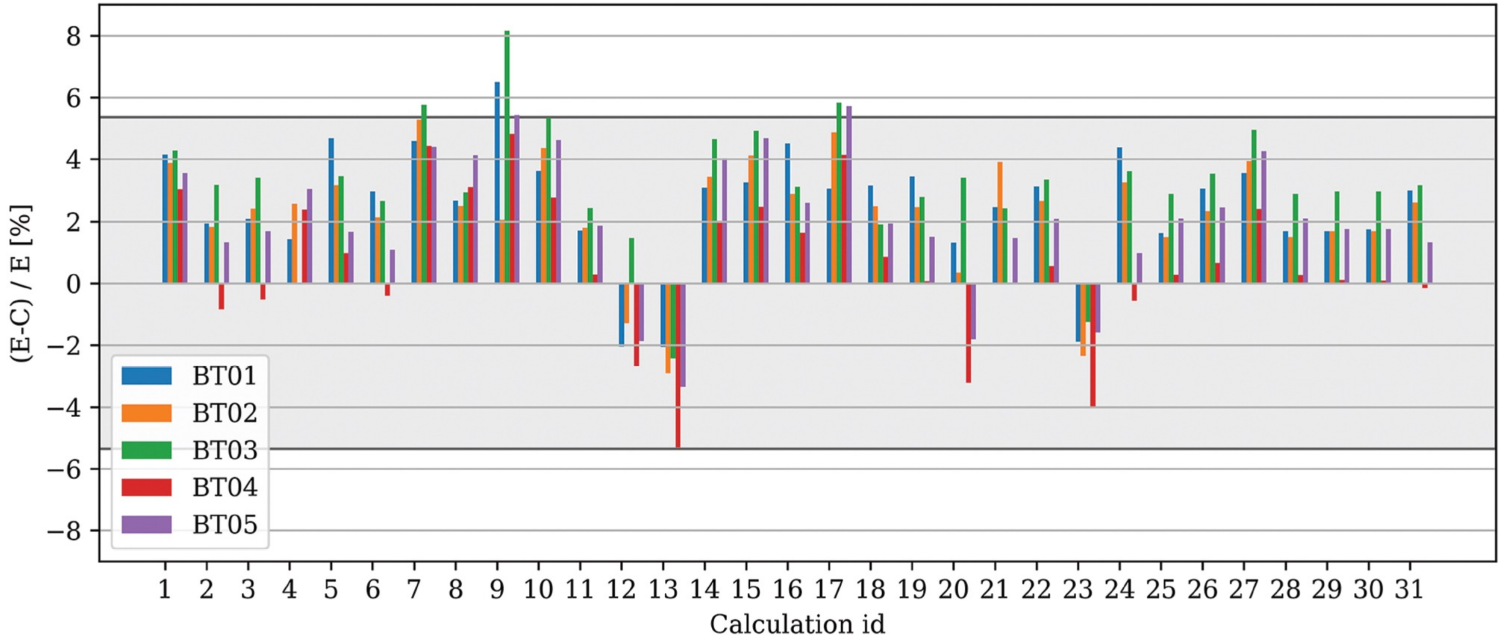


TABLE I

Fuel Assemblies Included in the Exercise and Their Basic Parameters with Data from Ref. 1

Identification	BU (GWd/tU)	CT (a)	IE (%)
BT01	53	4.5	3.95
BT02	55	8.6	3.95
BT03	50	9.8	3.95
BT04	51	13.5	3.70
BT05	50	21.4	3.60

Code	Library
ALEPH 2.7.2	ENDF/B-VII.1
APOLLO2.8/DARWIN2.3	JEFF-3.1.1
CASMO-4E + ORIGEN-S	JEF-2.2
CASMO-5 (2.03)	ENDF/B-VII.1
CASMO-5 (2.12.00) + SNF (1.07.02)	ENDF/B-VII.1
DRAGON 4.0.5	ENDF/B-VII.1
EVOLCODE (MCNP + ACAB)	JEFF-3.3
MCNP-CINDER + Nukleonika (2D)	ENDF/B-VII.1
Monteburnsv3 + CINDER	ENDF/B-VII.1
MOTIVE (KENO-VI + VENTINA)	ENDF/B-VII.1
MOTIVE (OpenMC + VENTINA)	ENDF/B-VIII
MVP 3	ENDF/B-VII.1
MVP 3	JEFF-3.2
MVP 3	JENDL-4.0
OREST	JEF-2.2 + ENDF/B-VI
SCALE 6.0: ORIGEN-ARP	ENDF/B-V
SCALE 6.1.3: ORIGEN-ARP	ENDF/B-V
SCALE 6.2.3: ORIGAMI	ENDF/B-VII.1
SCALE 6.2.3: Polaris	ENDF/B-VII.1
SCALE 6.2.3: ORIGEN	ENDF/B-VII.1
SCALE 6.2.3: TRITON/KENO	ENDF/B-VII.1
SCALE 6.2.3: TRITON/NEWT	ENDF/B-VII.1
SEADEP	JEFF-3.1.1
Serpent 2.1.29	ENDF/B-VII.1
Serpent 2.1.29	JEFF-3.1.1
Serpent 2.1.31	JEFF-3.2 + JEFF-3.1.1

Blind benchmark exercise for spent nuclear fuel decay heat, P. Jansson et al., Nucl. Sc. & Eng., 2022

Decay Heat calculations

$$DH(t) = f(t) = \sum_{i}^{n} N_i(t_c)\, \lambda_i\, \overline{E}_i$$

- **Bateman equations** solved to get Atomic Densities N_i at the cooling time

  Depletion calculation within a reactor model + code (e.g with SERPENT)

$$\frac{dN_i(t)}{dt} = \sum_j \left(b_{j\to i}\, \lambda_j + \phi\, \sigma_{j\to i} \right) N_j(t) - \left(\lambda_i + \phi\, \sigma_i \right) N_i(t)$$

$b_{j\to i}$: branching ratio

ϕ : neutron flux

- $\overline{E}_i$ is usually divided in evaluated librairies(e.g ENDF, JEFF, JENDL) in 3 parts :

$$\overline{E}_{LP} = \overline{E}_{\beta-} + \overline{E}_{\beta+} + \overline{E}_{e-} + \cdots$$

Light particles component

$$\overline{E}_{EM} = \overline{E}_{\gamma} + \overline{E}_{x-ray} + \overline{E}_{anni.rad.} + \cdots$$

Electromagnetic component

$$\overline{E}_{HP} = \overline{E}_{\alpha} + \overline{E}_{SF} + \overline{E}_{p} + \overline{E}_{n} + \cdots$$

Heavy particles component

Decay Energy and Pandemonium effect

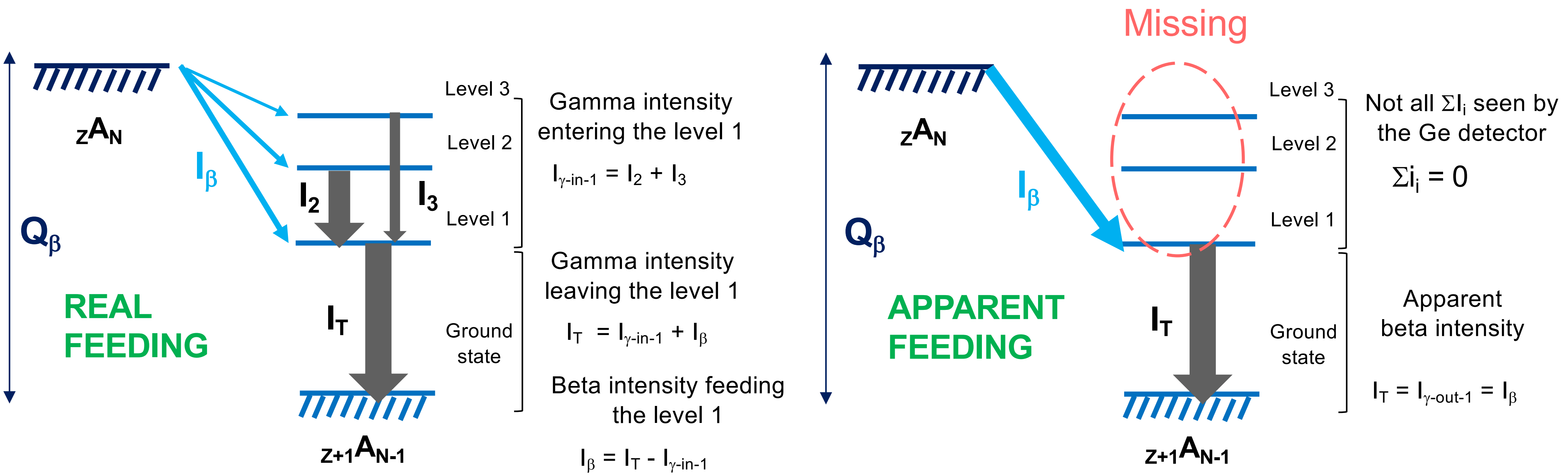
- Total Decay energy (E_i) measurements

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 E_{beta}, underestimate E_{gamma}



⇒ Bias in nuclear data bases for some key FP nuclei and all their applications (safeguards, DH, antineutrinos experiments)

=> Known as the « Pandemonium effect »

J. Hardy et al., PLB 71 (2) 307, 1977

Decay Energy and Pandemonium effect

Pandemonium (The Capital of Hell)

introduced by John Milton (XVII) in his epic poem Paradise Lost

John Martin (~ 1825) Hardy et al., Phys. Lett. 71B (1977) 307

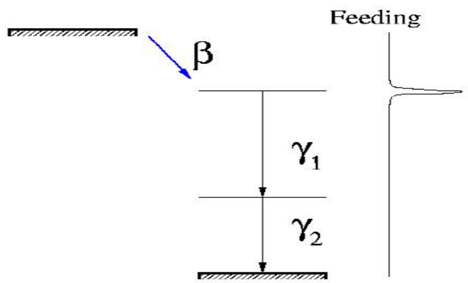
Courtesy of A. Algora

Total Absorption Gamma spectroscopy technique

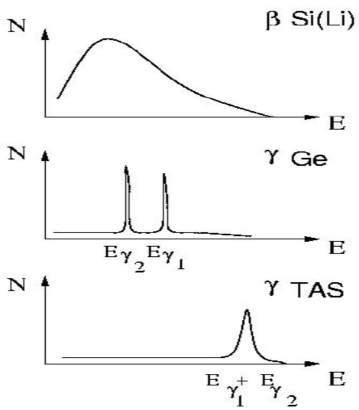
Most suitable technique to re-measure key nuclei: Total Absorption Spectroscopy

IFIC Valencia/Subatech/Surrey TAGS collaboration

Experiments @ Jyväskylä, Finland to high precision penning trap (Pure beams)



The decay seen by diff. detectors



Since the gamma detection is the only reasonable way to solve the problem, we need a highly efficient device:

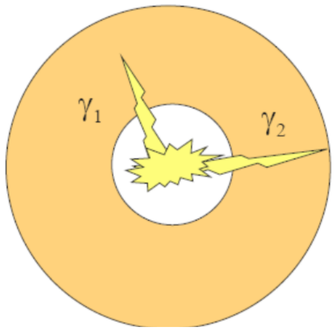
A TOTAL ABSORTION SPECTROMETER

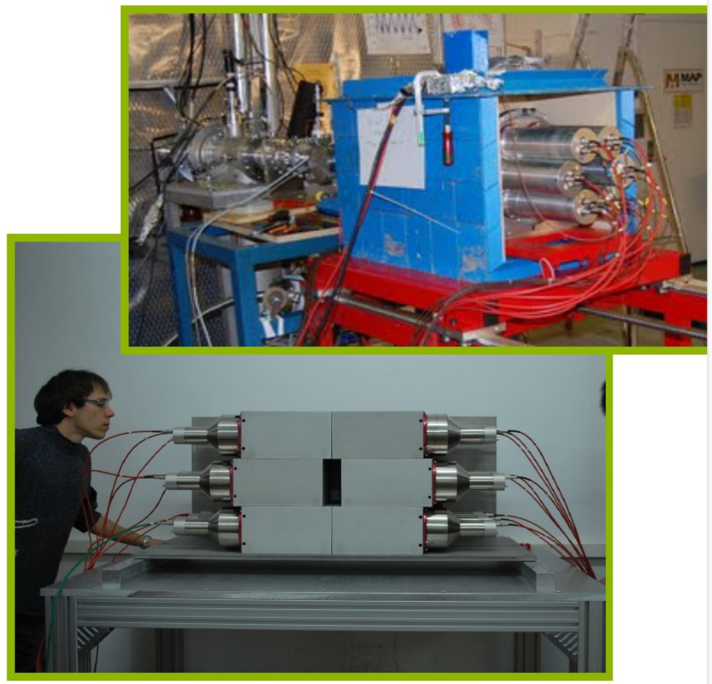
But we need a change in philosophy. Instead of detecting the individual gamma rays we sum the energy deposited by the gamma cascades in the detector.

A TAS is like a calorimeter!

Big crystal, 4π

$$d = R(B) \cdot f$$





TAGS Arrays, Valencia

From TAS collaboration: contacts A. Algora & J. L. Tain @Valencia, W. Gelletly@Surrey, M. Fallot@Subatech

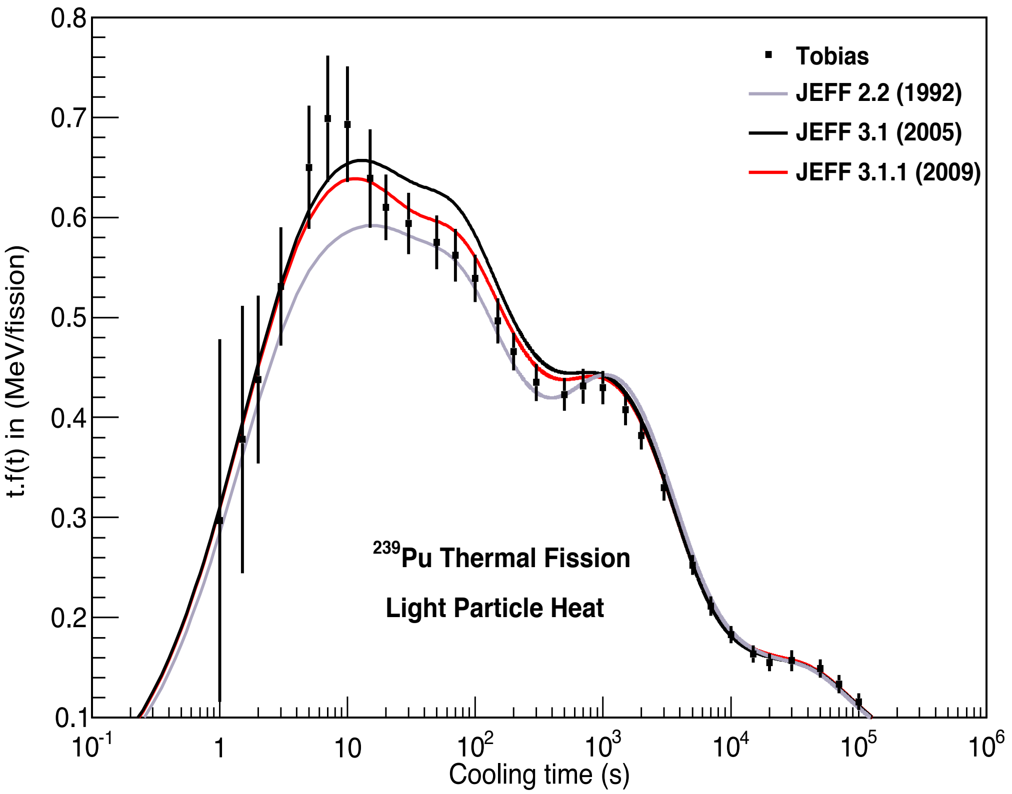
TAGS Measurements in decay libraries

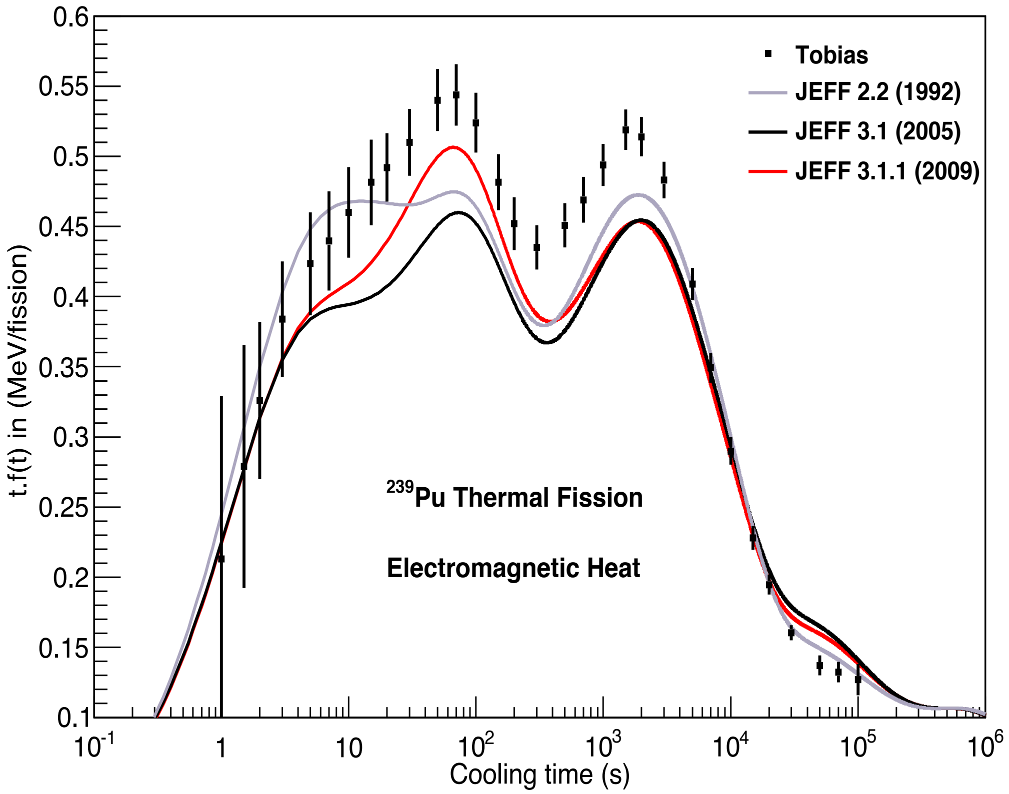
- 1990s: 50 TAGS measurements per Greenwood et al @ IDAHO National Laboratory

R. Greenwood et al., NIM A 390, 95, 1997

First inclusion of 29 nuclei in JEFF-3.1.1, *M. Kellett & O. Bersillon, EPJ Web Conf 146 0209 (2017)*

Also taken into account in the release of ENDF/B-VII.1 (2011)



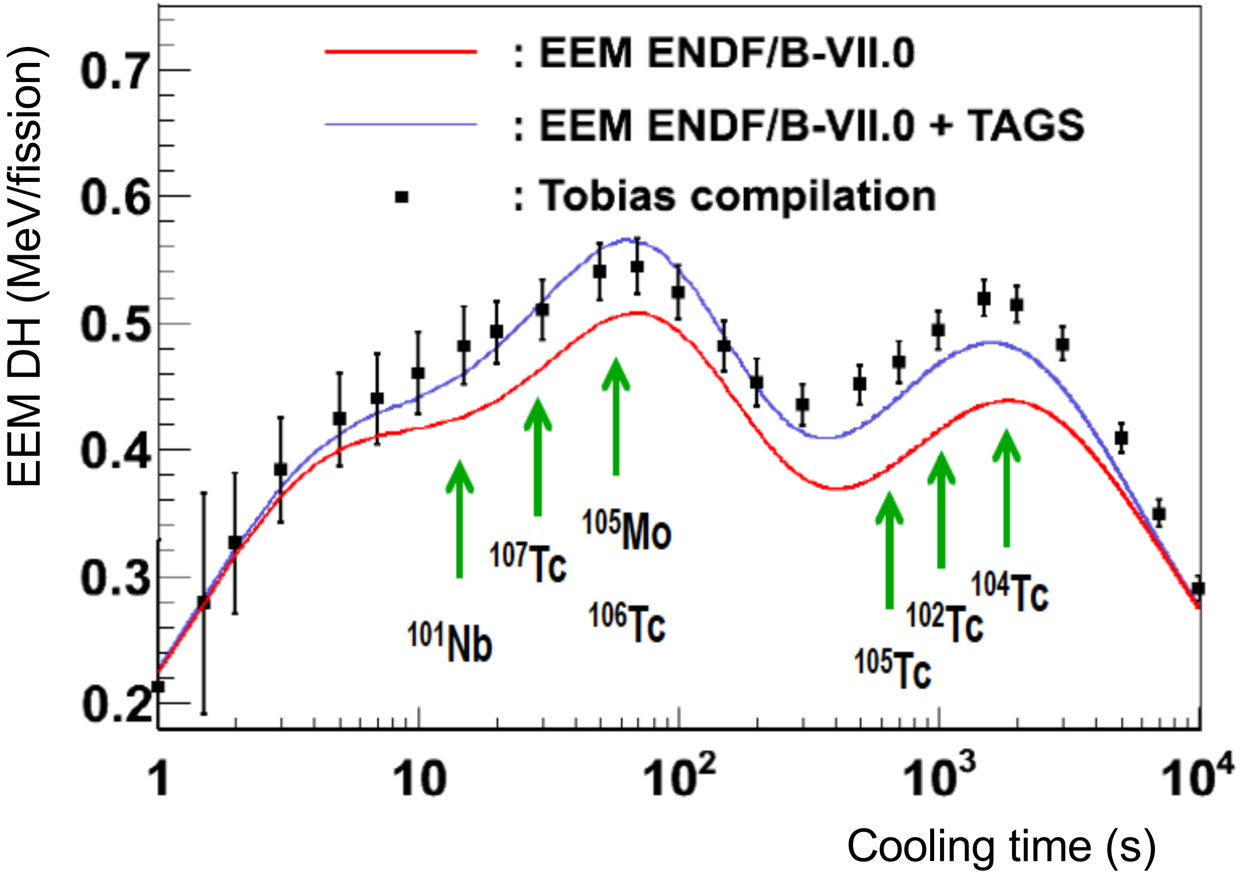


TAGS Measurements in decay libraries

Selected for a Viewpoint in *Physics*
PRL **105,** 202501 (2010) PHYSICAL REVIEW LETTERS week ending 12 NOVEMBER 2010

Reactor Decay Heat in ^{239}Pu: Solving the γ Discrepancy in the 4–3000-s Cooling Period

A. Algora,[1,2,*] D. Jordan,[1] J. L. Taín,[1] B. Rubio,[1] J. Agramunt,[1] A. B. Perez-Cerdan,[1] F. Molina,[1] L. Caballero,[1]



TAGS Measurements from the TAS collaboration

Strong impact on Electromagnetic component of Decay Heat for ^{239}Pu thermal fission

^{105}Mo, ^{104,105,106,107}Tc
Suffered from Pandemonium
In JEFF 3.3 & ENDF/B-VII.1

Same results obtained with SERPENT + JEFF 3.1.1

Published TAGS Measurements so far …

TAS Collaboration : IFIC Valencia, Univ. of Surrey, Subatech

3 experimental campaigns (2007, 2009, 2014) + Experiment 09/2022 @Jyvaskyla

MTAS Collaboration : Univ. of Warsaw, ORNL, Univ of Tennessee

Experiments @ Argonne National Laboratory's CARIBU facility

Isotope	Rel.	Isotope	Rel.	Isotope	Rel.
35-Br-86†*	1	41-Nb-99†	1	52-Te-135†	2
35-Br-87†*	1	41-Nb-100†*	1	53-I-136†	1
35-Br-88†*	1	41-Nb-101†*	1	53-I-136m†	1
36-Kr-89†	1	41-Nb-102†*	2	53-I-137†*	1
36-Kr-90†	1	42-Mo-103†*	1	54-Xe-137†	1
37-Rb-90m	2	42-Mo-105*	1	54-Xe-139†	1
37-Rb-92†*	2	43-Tc-102†*	1	54-Xe-140†	1
38-Sr-89	2	43-Tc-103†*	1	55-Cs-142*	3
38-Sr-97	2	43-Tc-104†*	1	56-Ba-145	2
39-Y-96†	2	43-Tc-105*	1	57-La-143	2
40-Zr-99†	3	43-Tc-106*	1	57-La-145	2
40-Zr-100†	2	43-Tc-107*	2		
41-Nb-98†*	1	51-Sb-132†	1		

† *: also relevant for ^{232}Th/^{233}U cycle*

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

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

+ ^{89,90}Rb

In total, 29 published nuclei

A. Algora et al., EPJ A 57, 2021

Impact of TAGS data on Decay Heat calculations

- Impact of the 28 published TAGS (wo ^{96m}Y) nuclei on Decay Heat calculations for 15 systems were studied

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

- Results presented here are part of a Review paper coordinated per IAEA (P. Dimitriou) on TAGS measurements, in completion phase

Improving Fission-product Decay Data For Reactor Applications: Decay Heat

A.L. Nichols[1,2], A. Algora[3], P. Dimitriou[a,4], M. Fallot[5], L. Giot[5], F.G. Kondev[6], T. Yoshida[7], G. Mukherjee[8], K. Rykaczewski[9], A.A. Sonzogni[10], J.L. Tain[3]

Serpent used for DH with JEFF libraries + TAGS data

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

Impact of TAGS data on Decay Heat calculations

- **For each fissioning system:**

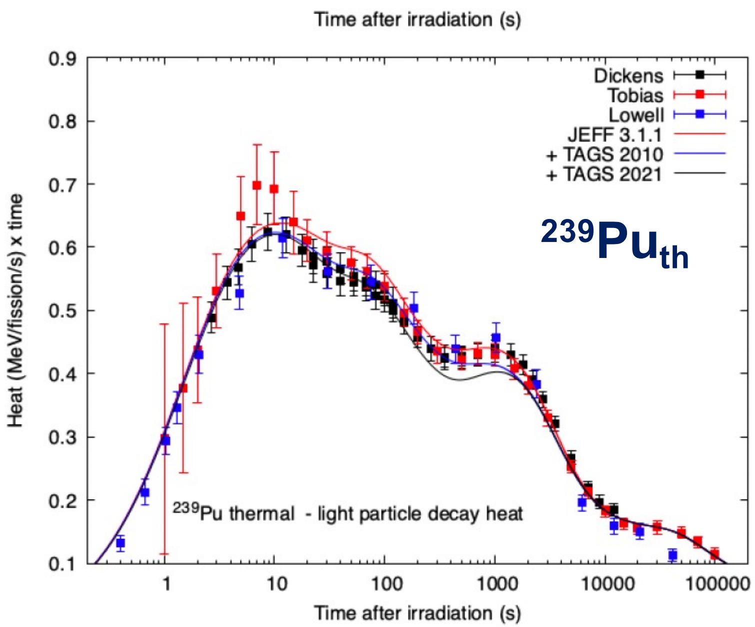
  3 sets of DH calculations combining the same FY library each time with :

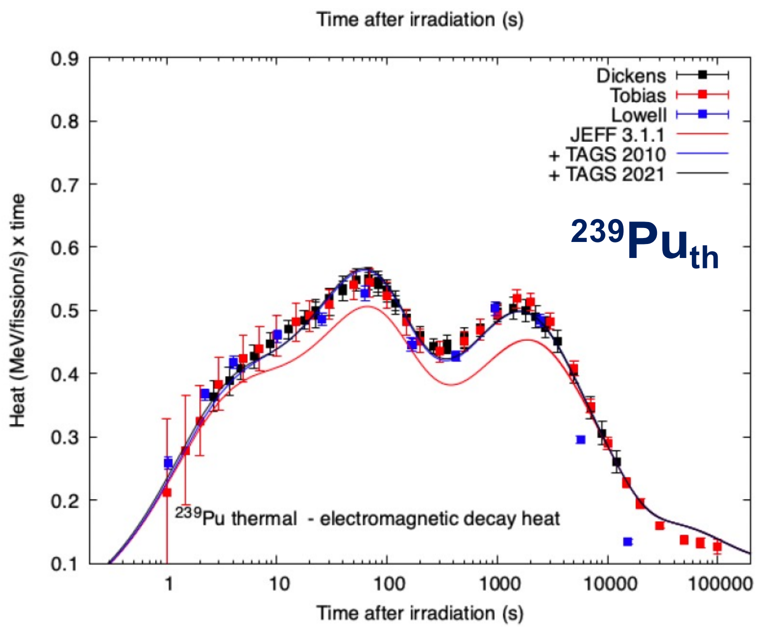
  - Decay Data without the Algora 2010 TAGS data: reference library or baseline

  - 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 $^{239}Pu_{th}$, $^{241}Pu_{th}$ & $^{238}U_{fast}$
small impact for $^{232}Th_{fast}$ & $^{233}U_{fast}$





Same conclusions with ENDF library

Impact of TAGS data on Decay Heat calculations

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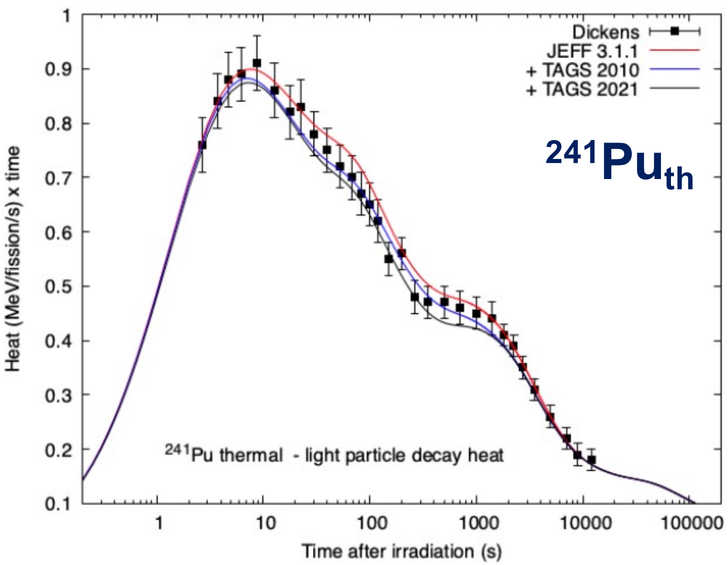

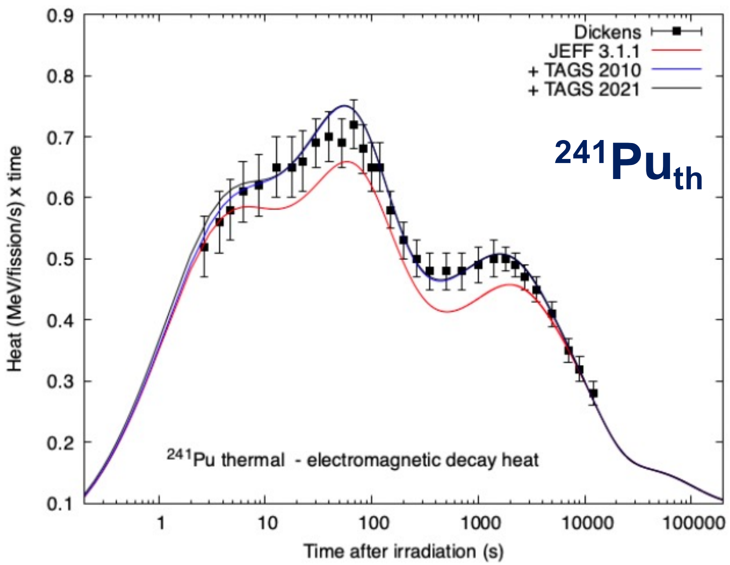

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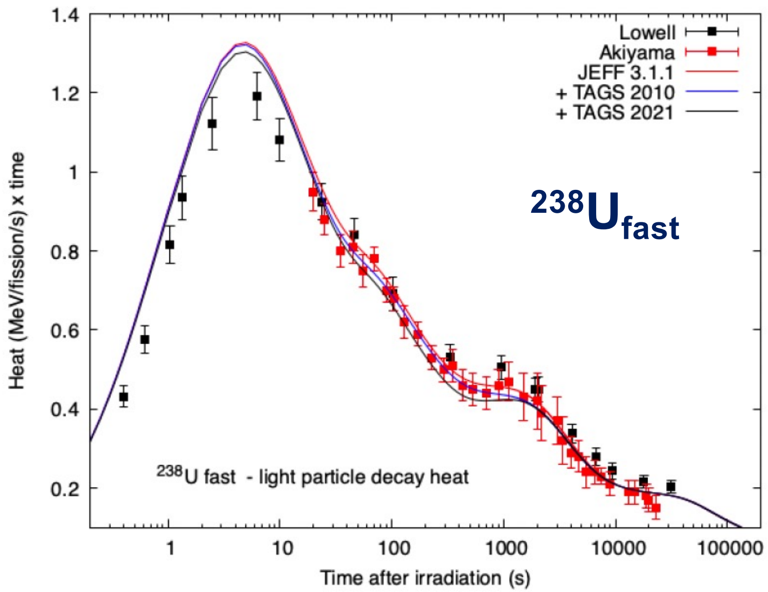
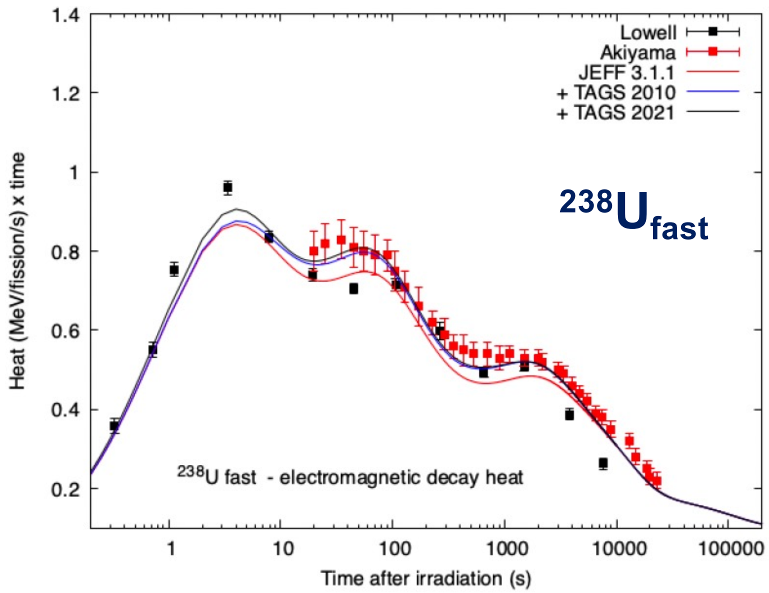




Same conclusions with ENDF library

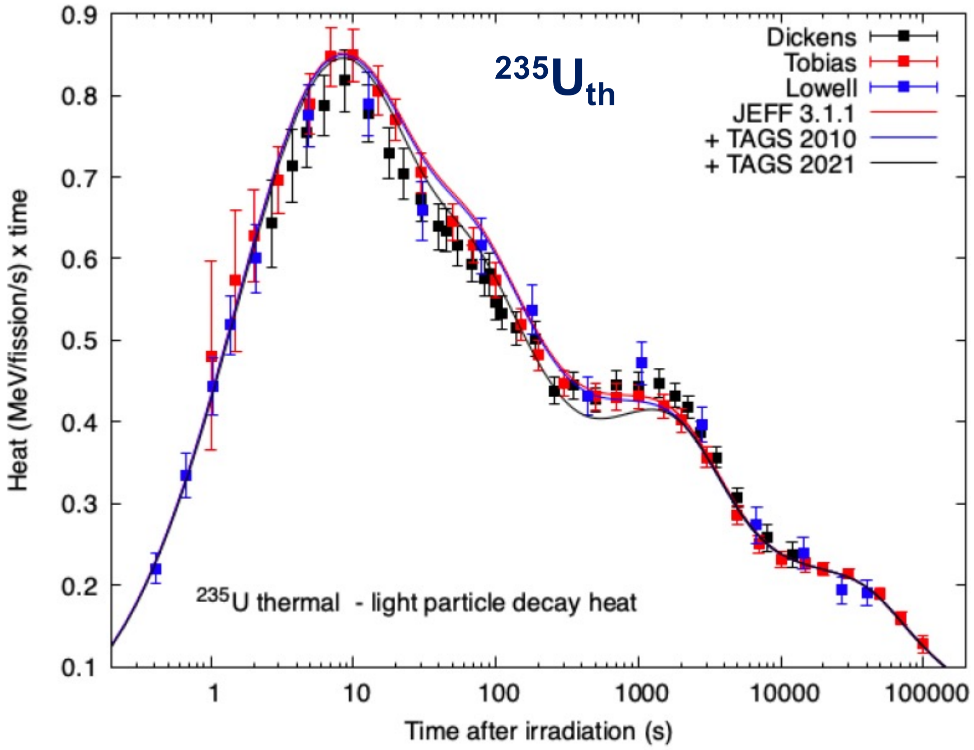
Impact of TAGS data on Decay Heat calculations

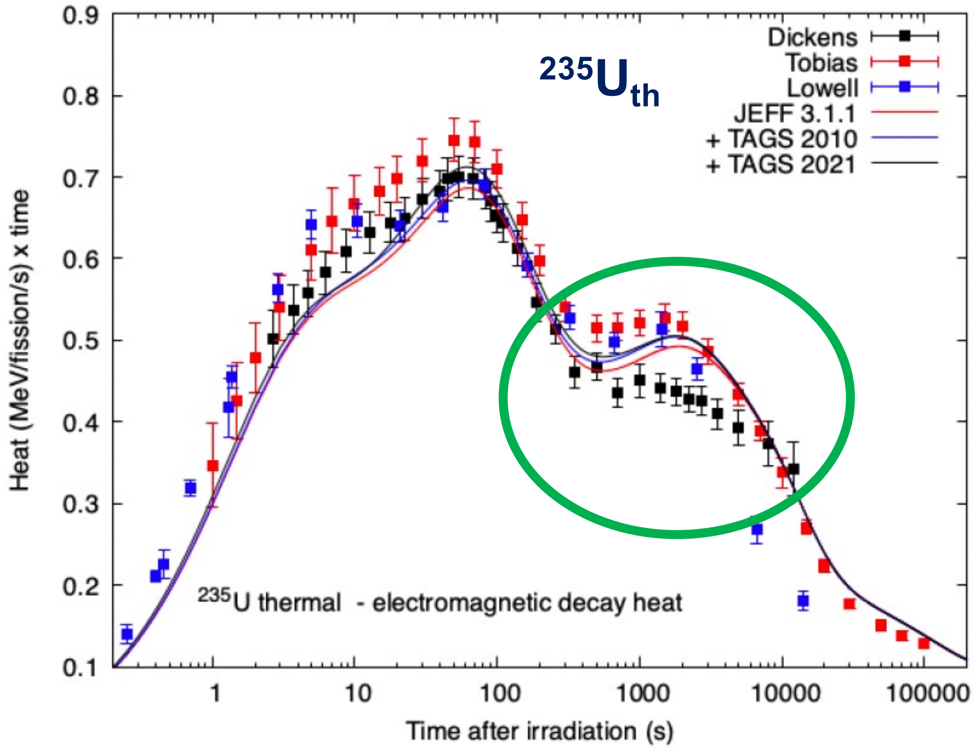
$^{235}U_{th}$

+ 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 !



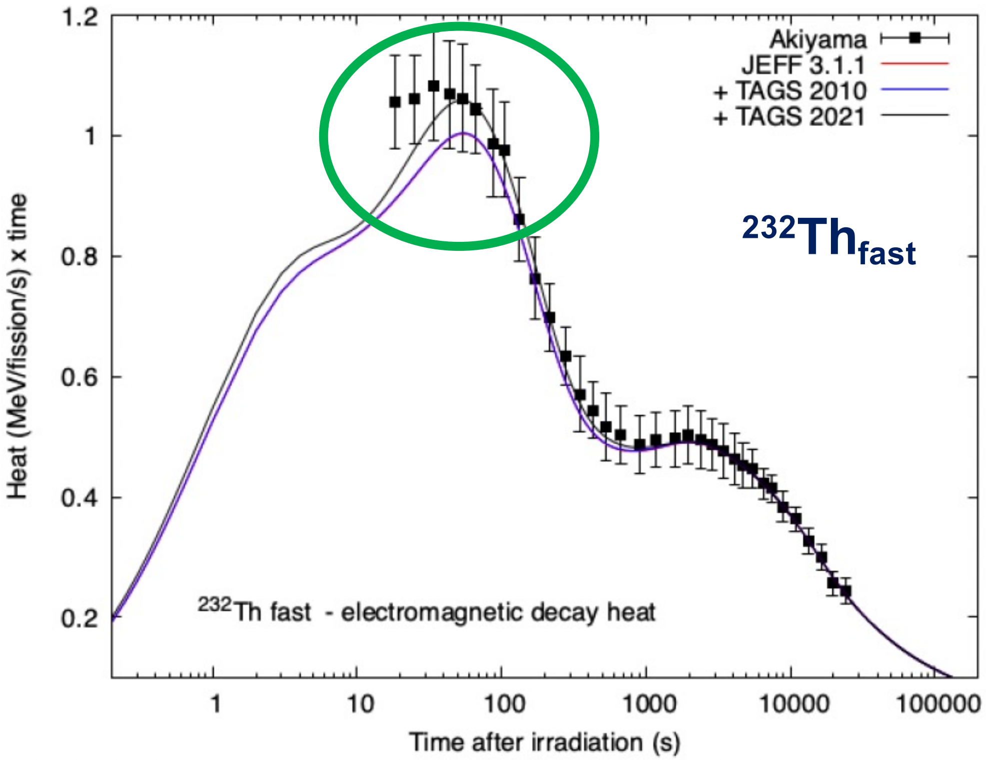


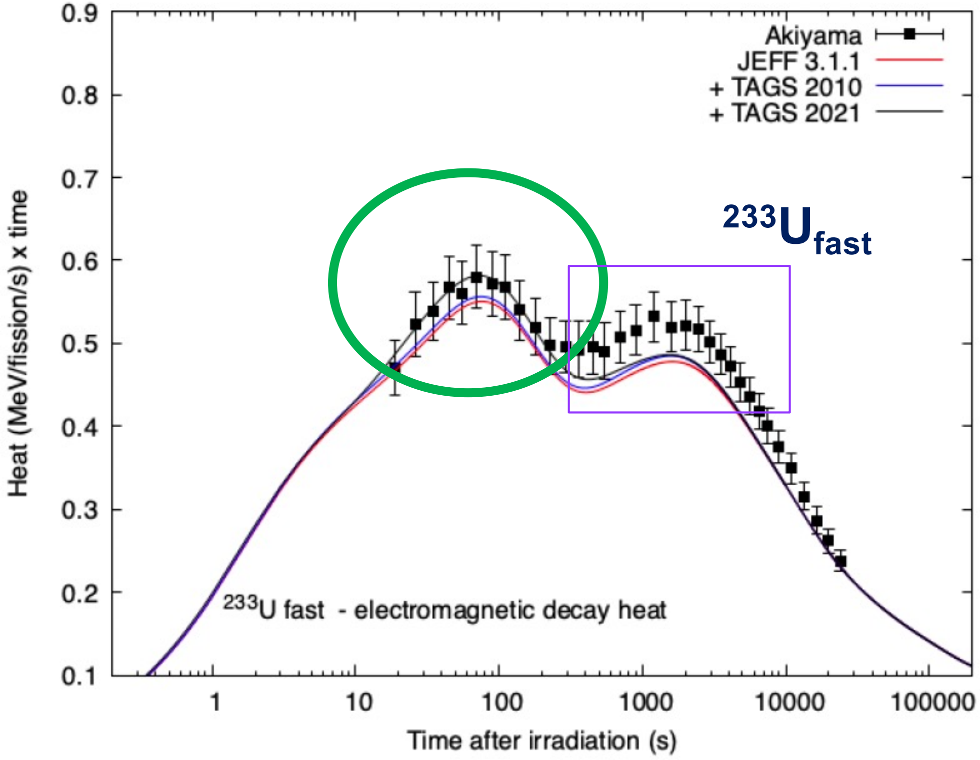
Same conclusions with ENDF library

Need of new DH fission pulse experiments ☺

Impact of TAGS data on Decay Heat calculations

+ TAGS 2021 : improved agreement of EEM component for $^{233}U_{fast}$, $^{238}U_{fast}$, $^{232}Th_{fast}$, for cooling time below 100s



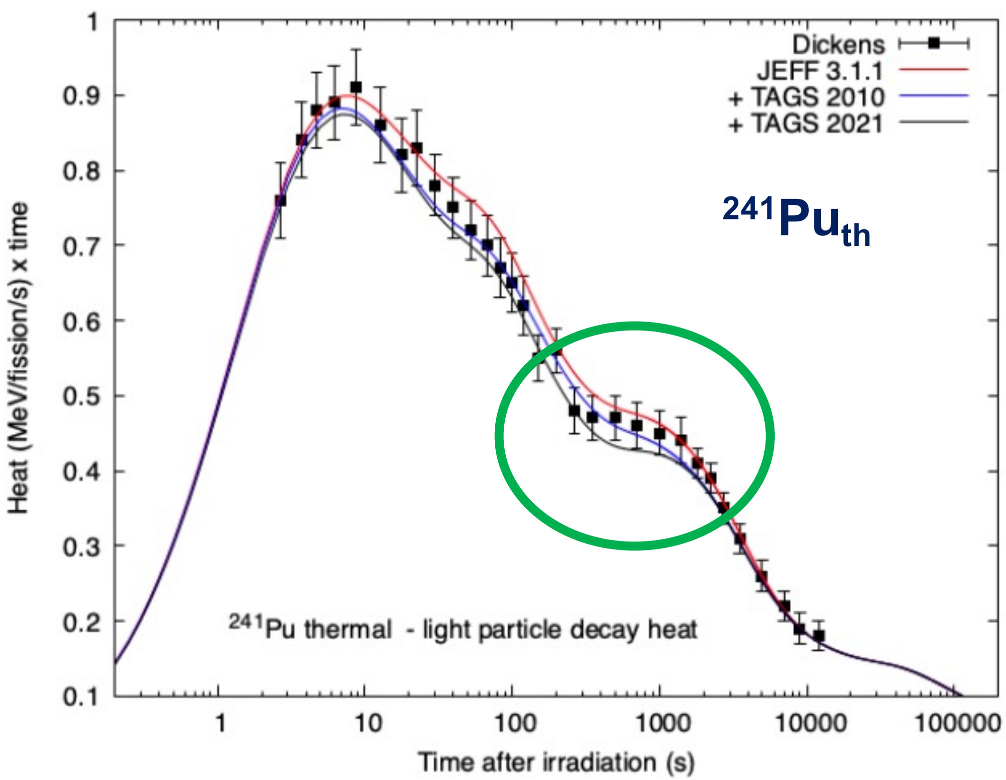


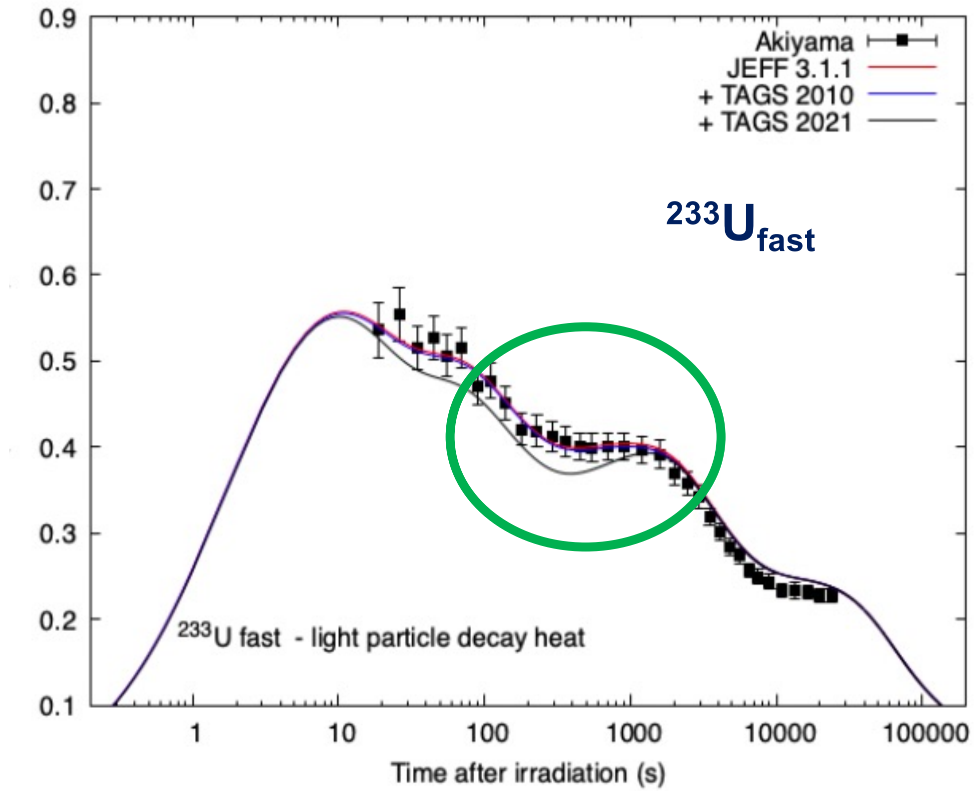
But also need of new DH fission pulse experiments ☺

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 $^{235}U_{th}$, $^{239}Pu_{th}$, $^{241}Pu_{th}$, $^{233}U_{fast}$ & $^{238}U_{fast}$ at cooling times ranging from 30s to 1000s





=> Only one set of experimental data, till in the errors bars for $^{241}Pu_{th}$

=> Needs for extra experimental data but also extra investigation on key FP suffering of Pandemonium effect

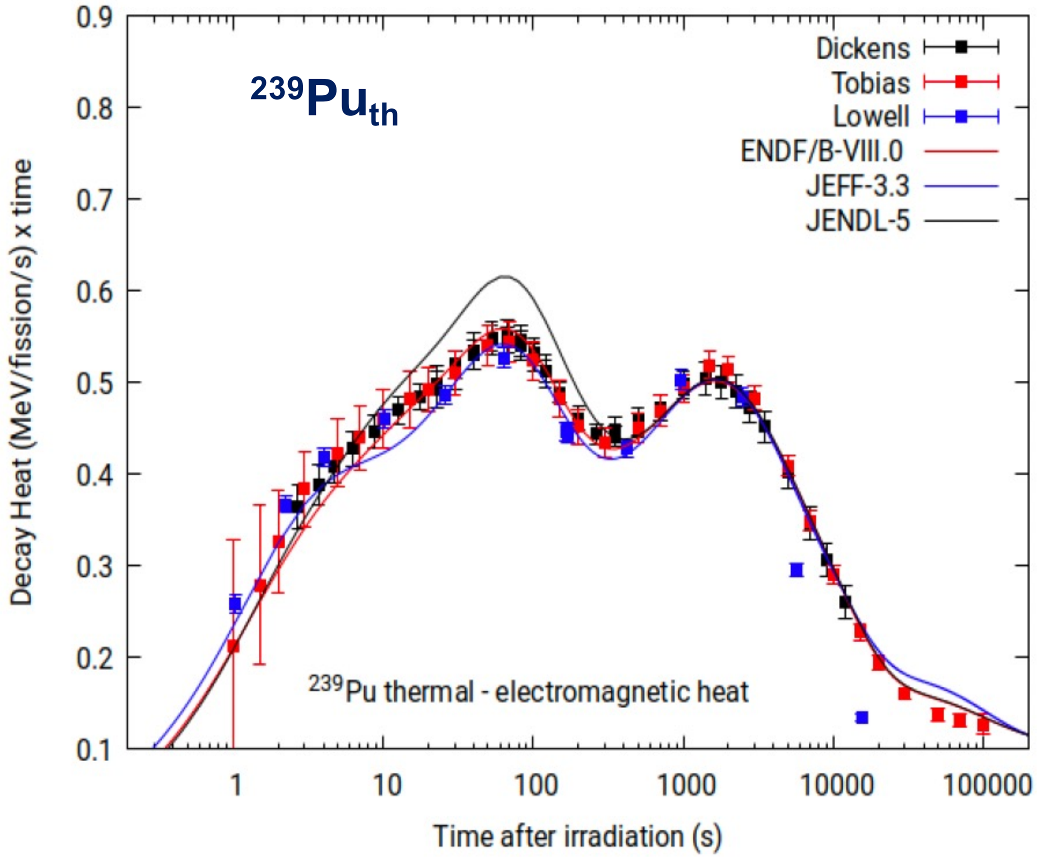
=> Also on going work to take into account FY and DD uncertainties through MC sampling

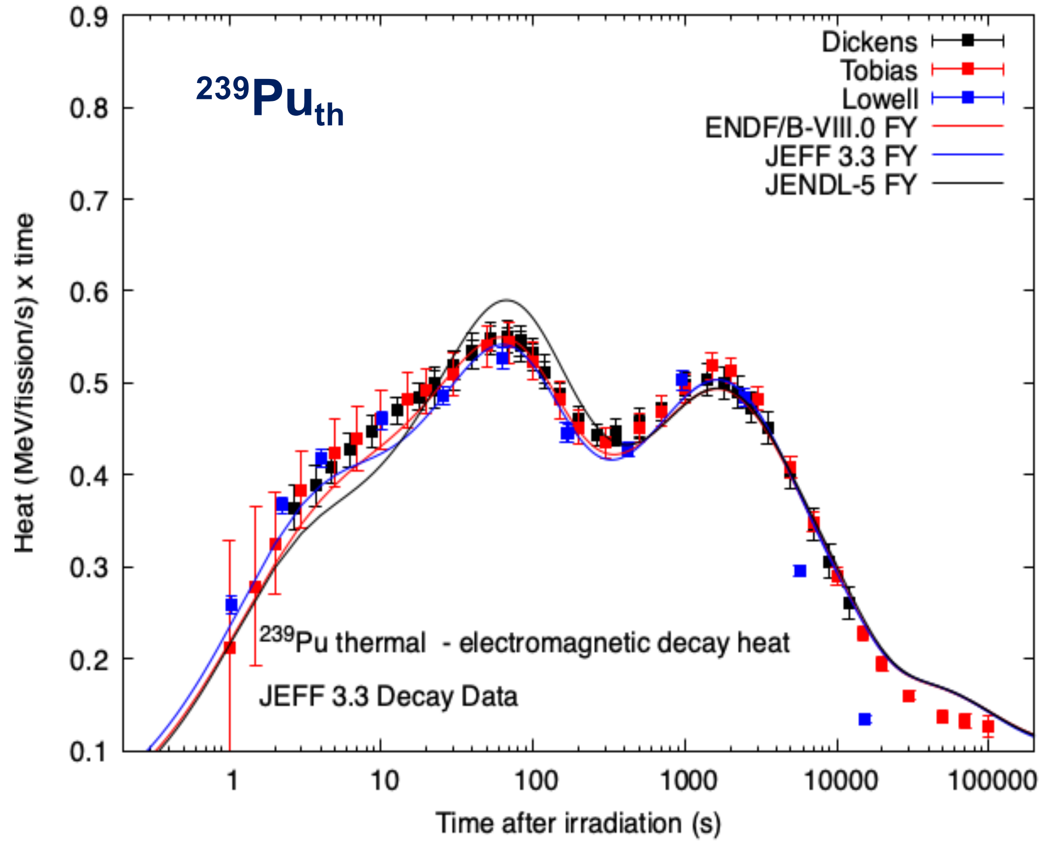
Same conclusions with ENDF library

Impact of TAGS data on DH calculations

Overestimation of EEM component for JENDL5 for $^{239}Pu_{th}$

Impact of Fission Yields ?





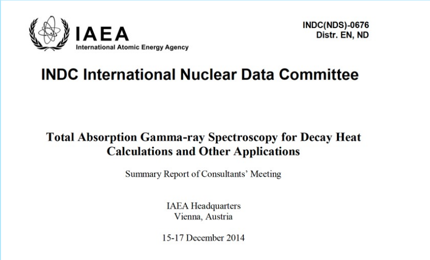
Final remarks and Outlooks

- **Further extensive assessments need to accomodate the impact of delayed neutrons on decay heat up 10s cooling time**

- **Further investigations are needed to improve DH exp data vs calculations :**

- **New Decay Heat fission pulse experiments …**

- **New TAGS measurements based on WPEC-25 list and IAEA consultants' meetings**
  priority 1 nuclei : ^{99,100}Zr, ^{98,99}Nb, $^{130m,\ 132}$Sb, ^{138}Cs, ^{143}La
  priority 2 nuclei :
  ^{84}As, ^{85}Se, ^{84,89}Br, ^{91}Kr, $^{92\text{-}95}$Sr, ^{97}Y, ^{105}Nb, ^{104,107}Mo, ^{108}Tc, ^{133}Sb, ^{136,137}Te, $^{139,\ 141,\ 143,\ 144}$Ba, $^{144\text{-}147}$La, ^{146}Pr, $^{139\text{-}141}$Cs, ^{136,136m}I, ^{140}Xe


IAEA
International Atomic Energy Agency
INDC(NDS)-0676
Distr. EN, ND

INDC International Nuclear Data Committee

Total Absorption Gamma-ray Spectroscopy for Decay Heat Calculations and Other Applications

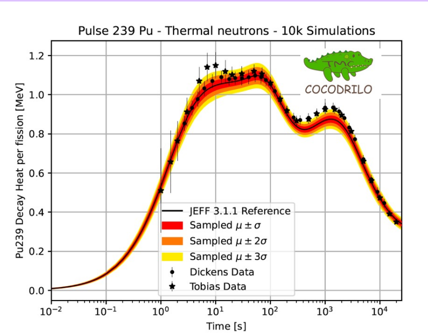
Summary Report of Consultants' Meeting

IAEA Headquarters
Vienna, Austria

15-17 December 2014

- **Extra investigations to identify key FP & Pandemonium candidates**
  -- on pulse calculations
  taking also into account uncertainties on Decay Data
  on-going PhD @Subatech Y. Molla, 2021-2024

  -- on new fuels/ reactor concepts
  ex: new PhD @Subatech&LPSC M. Tazreiter 2022-2025
  on Molten Salt Reactors



Thank you