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

Lydie.Giot@subatech.in2p3.fr

SUBATECH,
CNRS-IN2P3, France

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

<table>
<thead>
<tr>
<th>Nuclear stage impacted</th>
<th>Time of cooling</th>
</tr>
</thead>
<tbody>
<tr>
<td>Safety systems of cooling</td>
<td>0.1s to 8 days</td>
</tr>
<tr>
<td>Unloading of assemblies from core</td>
<td>5 to 25 days</td>
</tr>
<tr>
<td>Fuel transport</td>
<td>1 to 10 years</td>
</tr>
<tr>
<td>Reprocessing, vitrification, storage</td>
<td>4 to 3000 years</td>
</tr>
<tr>
<td>Storage</td>
<td>50 to 300 000 years and more</td>
</tr>
</tbody>
</table>
Decay Heat calculations

**Summation Formula**

\[ DH(t) = f(t) = \sum_{i}^{n} N_i(t_c) \lambda_i \bar{E}_i \]

- 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: $\sigma$, $\bar{E}$, Branching Ratio, $\lambda$, Fission Yields, $\bar{\nu}$

- 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 = Possible to get/use them 😊

Fission Pulses
$^{235}\text{U}_{th}, ^{239}\text{Pu}_{th}, ^{241}\text{Pu}_{th}$

GE Morris/HEDL UO$_2$, 80 PWR/BWR assemblies

CLAB UO$_2$, 74 PWR/BWR assemblies

Cooling time

0.3 s 1 day 2 years 11 years 27 years

Available in AIEA CONDERC database

Foreseen to be included in the SFCOMPO NEA database

- 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

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

\[ \text{DH}(t) = f(t) = \sum_{i}^{n} N_i(t_c) \lambda_i \overline{E}_i \]

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

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

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

- \(b_{j \rightarrow i}\): branching ratio
- \(\lambda_i\), \(\sigma_i\), \(\phi\): neutron flux

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

- \(\overline{E}_{LP}\) = \(\overline{E}_{\beta^-} + \overline{E}_{\beta^+} + \overline{E}_{e^-} + \cdots\) \(\text{Light particles component}\)
- \(\overline{E}_{EM}\) = \(\overline{E}_{\gamma} + \overline{E}_{x-ray} + \overline{E}_{anni.rad.} + \cdots\) \(\text{Electromagnetic component}\)
- \(\overline{E}_{HP}\) = \(\overline{E}_{\alpha} + \overline{E}_{SF} + \overline{E}_{p} + \overline{E}_{n} + \cdots\) \(\text{Heavy particles component}\)
Decay Energy and Pandemonium effect

- Total Decay energy ($E_i$) measurements
Before the 90s, conventional detection techniques: high resolution $\gamma$-ray spectroscopy
Excellent resolution but efficiency which strongly decreases with increasing energy
Risk of overlooking the existence of $\beta^-$ 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


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)

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

A TOTAL ABSORPTION 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, $4\pi$

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

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

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


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
II. Decay Heat Calculation

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

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

†: 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, $^{96}$mY

+ $^{89,90}$Rb

In total, 29 published nuclei

A. Algara et al., EPJ A 57, 2021
Impact of TAGS data on Decay Heat calculations

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

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

<table>
<thead>
<tr>
<th>thermal neutron pulse (0.0253 eV)</th>
<th>$^{235}\text{U}$, $^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$, $^{242}\text{Pu}$, $^{241}\text{Am}$, $^{242m}\text{Am}$, $^{243}\text{Am}$, $^{243}\text{Cm}$, $^{245}\text{Cm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>fast neutron pulse (400 keV or 500 keV)</td>
<td>$^{232}\text{Th}$, $^{233}\text{U}$, $^{238}\text{U}$, $^{237}\text{Np}$</td>
</tr>
</tbody>
</table>

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


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


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
II. Decay Heat Calculation

**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

- 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

- 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

\[ ^{235}\text{U}_{\text{th}} \]

+ 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 underestimation of ELP component for \(^{235}\text{U}_{\text{th}}\), \(^{239}\text{Pu}_{\text{th}}\), \(^{241}\text{Pu}_{\text{th}}\), \(^{233}\text{U}_{\text{fast}}\), and \(^{238}\text{U}_{\text{fast}}\) at cooling times ranging from 30s to 1000s.

**Results:**

- 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?

- Dickerson
- Tobias
- Lowell
- ENDF/B-VII.0
- JEFF-3.3
- JENDL-5 FY

$^{239}\text{Pu}_{\text{th}}$ thermal - electromagnetic decay heat

JEFF 3.3 Decay Data
Final remarks and Outlooks

- Further extensive assessments need to accommodate 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, $^{130}$m, $^{132}$Sb, $^{138}$Cs, $^{143}$La
    priority 2 nuclei:
    $^{84}$As, $^{85}$Se, $^{84,89}$Br, $^{91}$Kr, $^{92-95}$Sr, $^{97}$Y, $^{105}$Nb, $^{104,107}$Mo, $^{108}$Tc, $^{133}$Sb,
    $^{136,137}$Te, $^{139,141,143,144}$Ba, $^{144-147}$La, $^{146}$Pr, $^{139-141}$Cs, $^{136,136}$m, $^{140}$Xe

- 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