Measurements for Reactor Decay Heat

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Fission process

- Kinetic energy of fission products (FP) and neutrons
- Prompt $\gamma$ radiation from FP
- $\gamma$ and $\beta$ decay energy through the natural decay of fission products
**Decay heat: definition**

\[ f(t) = \sum_i E_i \lambda_i N_i(t) \]

- \( E_i \)  
  Decay energy of the nucleus \( i \)

- \( \lambda_i \)  
  Decay constant of the nucleus \( i \)

- \( N_i \)  
  Number of nuclei \( i \) at the cooling time \( t \)

Requirements for the calculations: large databases that contain all the required information (nuclides, lifetimes, mean \( \gamma \)- and \( \beta \)-energy released in the decay, n-capture cross sections, etc, etc …)
**Example of database: JENDL FP decay data file 2000**

<table>
<thead>
<tr>
<th>No. of Nuclides</th>
<th>Data types, comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>581</td>
<td>With theor. estimated average $\gamma$-decay energy</td>
</tr>
<tr>
<td>506</td>
<td>With measured average $\gamma$-decay energy</td>
</tr>
<tr>
<td>543</td>
<td>With theor. estimated average $\beta$-decay energy</td>
</tr>
<tr>
<td>506</td>
<td>With measured average $\beta$-decay energy</td>
</tr>
<tr>
<td>197</td>
<td>First isomeric states</td>
</tr>
<tr>
<td>8</td>
<td>Second isomeric states</td>
</tr>
<tr>
<td>1229</td>
<td>Tot. num. of nuclides (142 stable, 1087 unstable)</td>
</tr>
</tbody>
</table>
Pandemonium effect

Introduced by the work of Hardy et al (Phys. Lett 71B (1977) 307). Their study questions the possibility of building correctly a level scheme from a beta decay experiment using conventional techniques.

Several factors can contribute to this problem:

• if the feeding occurs at a place where there is a high density of levels, there is a large fragmentation of the strength among different levels and there is a large number of decay paths, which makes the detection of the weak gamma rays difficult

• we can have gamma rays of high energy, which are hard to detect
Since the gamma detection is the only reasonable way to solve the problem, we need a highly efficient device:

A TOTAL ABSORPTION SPECTROMETER

Instead of detecting the individual gamma rays we sum the energy deposited by the gamma cascades in the detector.
Problems associated with TAS

• Analysis
• Contaminants
• Technique not well known: what can be expected from a TAS measurement?
$S_i = \frac{I_i}{f(Q_\beta - E_i)T_{1/2}}$

$d_i = \sum_j R_{ij}f_j \quad or \quad d = R \cdot f$

$R$ is the response function of the spectrometer, $R_{ij}$ means the probability that feeding at a level $j$ gives counts in data channel $i$. 
Contaminants: TAZ measurements

TAS-manian devil: "Taz" for short, is described as: "A strong murderous beast, jaws as powerful as a steel trap, has ravenous appetite, eats tigers, lions, elephants, buffaloes, donkeys, giraffes, octopuses, rhinoceroses, and moose." Similar to our TAS detector
Contaminants: background, isobaric contaminants

Source of systematic uncertainty. In the neutron rich side it is not possible to use the EC process to clean the spectra.

Possible solutions:

- Separation using cycles that exploit half-life knowledge of the nucleus of interest and contaminants
- Use of chemical selectivity at the ion source
- Use of laser ionization schemes, to ionize only the species of interest
Example: measurement of the beta decay of $^{104,105}\text{Tc}$

The main motivation of this work was the study of Yoshida and co-workers (Journ. of Nucl. Sc. and Tech. 36 (1999) 135).

See $^{239}\text{Pu}$ example, similar situation for $^{235,238}\text{U}$. 
Motivations, original plans

In their work (detective work) Yoshida et al. identified some nuclei that may be responsible for the underestimation of the $E_{\nu}$ component.

Possible nuclei that may be blamed for the anomaly were $^{102,104,105}$Tc

Explanation: certainly suffer from the Pandemonium effect, their half lives are in the range needed, and their fission yields are also correlated in the way required to solve the discrepancy.
The IGISOL technique

Fission ion guide: 2700 ions/s per mb, eff. of $1.6 \times 10^{-4}$ relative to the production in the target

Details of our experiment:
Beam: 30 MeV proton (5 microA)
Target: natural U
Target thickness: 15 mg/cm$^2$
Target dimensions: 10x50 mm, tilted 7 degrees
Yield of $^{112}$Rh: 3500 atoms/microC

Tight collimation scheme to avoid contamination of neighbour mases (losses of 25%)
Experimental setup at Jyväskylä

Rad. beam
Ge det.
Tape station
Si det.
TAS det
(Det 1 & det 2)
$^{104}$Tc TAS spectrum

- Last known level: 4268 keV
- $Q_{\beta} = 5600$ keV
$^{105}$Tc TAS spectrum

Last known level: 2404 keV

$Q_\beta = 3640$ keV
Analysis of $^{104}$Tc

Expectation Maximization (EM) method:
• modify knowledge on causes from effects

Algorithm:

$$f_j^{(s+1)} = \frac{1}{\sum_i R_{ij}} \sum_i R_{ij} f_j^{(s)} d_i$$

$$P(f_j | d_i) = \frac{P(d_i | f_j) P(f_j)}{\sum_j P(d_i | f_j) P(f_j)}$$

Some details ( d=Rf )

Known levels up to: 1515 keV excitation

From that level up to the $Q_\beta$ value we use an statistical model

(Back Shifted Fermi formula for the level density with parameters taken from the RIPL database ($^{102}$Ru, $^{106}$Pd)

Branching ratios
Monte Carlo simulations of the setup: geometry
Results of the analysis for $^{104}\text{Tc}$
Results of the analysis for $^{105}\text{Tc}$
Impact of the results for $^{104,105}$Tc

ENDF/B-VII Decay Data Library

$^{239}$Pu EM

105Tc+104Tc (Valencia) and INEL TAGS
104Tc (Valencia) and INEL TAGS
INEL TAGS only
No Tags
Data: Tobias 1989
Impact of the results for $^{104,105}$Tc
Possible measurements at ALTO

There are several advantages of having a stable setup for these kind of measurements:

- The possibility of doing systematic studies in a controlled way, provided on the availability of beamtime

- Very cost effective, since we are not forced to mount and dismount the setup, with a large amount of effort. There is also the advantage of the reduction of the time required for the analysis.

- The possibility of instructing people (students, and not only students) in the use of the TAS technique
<table>
<thead>
<tr>
<th>Nucl</th>
<th>$T_{1/2}$</th>
<th>$Q_\beta$</th>
<th>$E_{\text{last}}$</th>
<th>$S_n$</th>
<th>N%</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{92}\text{Rb}$</td>
<td>4.5s</td>
<td>8105</td>
<td>7363</td>
<td>7342</td>
<td>0.0107</td>
<td>Diff. sep. with $T_{1/2}$</td>
</tr>
<tr>
<td>$^{89}\text{Sr}$</td>
<td>50.5d</td>
<td>1497</td>
<td>909</td>
<td>-</td>
<td>-</td>
<td>Why in the list?</td>
</tr>
<tr>
<td>$^{97}\text{Sr}$</td>
<td>426ms</td>
<td>7467</td>
<td>2558</td>
<td>5979</td>
<td>0.005</td>
<td>Diff. sep. with $T_{1/2}$</td>
</tr>
<tr>
<td>$^{96}\text{Y}$</td>
<td>5.3s</td>
<td>7087</td>
<td>6231</td>
<td>7854</td>
<td>-</td>
<td>Diff. sep. with $T_{1/2}$, the two isomers are sim.</td>
</tr>
<tr>
<td></td>
<td>9.6s</td>
<td>+X</td>
<td>5899</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$^{100}\text{Zr}$</td>
<td>7.1s</td>
<td>3335</td>
<td>703</td>
<td>5680</td>
<td>-</td>
<td>Daugther $T_{1/2}$=1.5 s</td>
</tr>
<tr>
<td>$^{99}\text{Nb}$</td>
<td>15s</td>
<td>3639</td>
<td>235</td>
<td>5925</td>
<td>-</td>
<td>Looks ok</td>
</tr>
<tr>
<td></td>
<td>2.6m</td>
<td>3974</td>
<td>2944</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>$^{102}\text{Nb}$</td>
<td>4.3s</td>
<td>7210</td>
<td>2480</td>
<td>8117</td>
<td>-</td>
<td>High resol. meas. needed, clean beam needed</td>
</tr>
<tr>
<td></td>
<td>1.3s</td>
<td>+X</td>
<td>???</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>
### Possible cases: Yoshida’s list II

<table>
<thead>
<tr>
<th>Nucl</th>
<th>$T_{1/2}$</th>
<th>$Q_\beta$</th>
<th>$E_{\text{last}}$</th>
<th>$S_n$</th>
<th>$N%$</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{135}\text{Te}$</td>
<td>19s</td>
<td>5960</td>
<td>4773</td>
<td>7900</td>
<td>-</td>
<td>☺</td>
</tr>
<tr>
<td>$^{145}\text{Ba}$</td>
<td>4.31s</td>
<td>4930</td>
<td>2566</td>
<td>6150</td>
<td>-</td>
<td>Greenwood case</td>
</tr>
<tr>
<td>$^{145}\text{La}$</td>
<td>24.8s</td>
<td>4120</td>
<td>2607</td>
<td>4730</td>
<td>-</td>
<td>Greenwood case</td>
</tr>
<tr>
<td>$^{87}\text{Br}$</td>
<td>55.6s</td>
<td>6853</td>
<td>5821</td>
<td>5514</td>
<td>2.57</td>
<td>Case study, Nichols list</td>
</tr>
<tr>
<td>$^{142}\text{Cs}$</td>
<td>1.7s</td>
<td>7306</td>
<td>5280</td>
<td>6170</td>
<td>0.091</td>
<td>Diff. $T_{1/2}$ cleaning.</td>
</tr>
<tr>
<td>$^{143}\text{La}$</td>
<td>14.2m</td>
<td>3425</td>
<td>2825</td>
<td>5145</td>
<td>-</td>
<td>☺</td>
</tr>
</tbody>
</table>
### Other possible cases: Nichols

<table>
<thead>
<tr>
<th>Nucl</th>
<th>$T_{1/2}$</th>
<th>$Q_\beta$</th>
<th>$E_{\text{last}}$</th>
<th>$S_n$</th>
<th>$N%$</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{87}\text{Br}$</td>
<td>55.6s</td>
<td>6853</td>
<td>5821</td>
<td>5514</td>
<td>2.57</td>
<td>Case study, good $T_{1/2}$ sep</td>
</tr>
<tr>
<td>$^{88}\text{Br}$</td>
<td>7.1s</td>
<td>8960</td>
<td>7000</td>
<td>7053</td>
<td>6.4</td>
<td>Case study, good $T_{1/2}$ sep</td>
</tr>
<tr>
<td>$^{90}\text{Br}$</td>
<td>1.92s</td>
<td>10350</td>
<td>5730</td>
<td>6310</td>
<td>24.6</td>
<td>More diff. case</td>
</tr>
<tr>
<td>$^{137}\text{I}$</td>
<td>24.5s</td>
<td>5880</td>
<td>5170</td>
<td>4025</td>
<td>6.97</td>
<td>Separable using $T_{1/2}$</td>
</tr>
<tr>
<td>$^{138}\text{I}$</td>
<td>6.49s</td>
<td>7820</td>
<td>5341</td>
<td>5810</td>
<td>5.5</td>
<td>Still possible sep. with $T_{1/2}$</td>
</tr>
</tbody>
</table>
Conclusions

- From the available information (databases) it is clear that there is a huge amount of work to be done. It requires close collaboration with the experts of the field in order to determine priorities.

- The work requires the installation of a new TAS setup, and counting on the availability of beam time. In other words large support from the laboratory.

- There are specific issues that need to be addressed for each case of interest: purity of the beam, beta delayed neutron emission, etc.