2nd EFNUDAT workshop

Determination of thermal radiative capture cross section

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• PGAA-NIPS experimental facilities at the Budapest
• Precision of internal calibration of partial $\gamma$-ray Xsections
• Crossing Intensity Sums (CIS)
  – Intensities of the $^{14}$N(n,$\gamma$) high energy standard
• Determination of thermal neutron capture Xsections with $\gamma$-ray spectroscopy and its limitations
• Completeness from experimental point of view
  – An example
• Conclusion
PGAA-NIPS facilities
Partial $\gamma$-ray production cross section (standardization)

- Comparator method for prompt activation
  - Standardization (table) or dedicated standardization experiment
  - Provides thermal Xsection values for $1/v$ nuclei

\[
\sigma_{\gamma_x} = \sigma_{\gamma_c} \frac{n_c}{n_x} \frac{A_{\gamma_x}}{A_{\gamma_c}} \frac{\varepsilon(E_{\gamma_c})}{\varepsilon(E_{\gamma_x})} \frac{f(E_{\gamma_c})}{f(E_{\gamma_x})}, \quad \sigma_{\gamma} = \theta P_{\gamma} \sigma_{th}
\]

<table>
<thead>
<tr>
<th>Partial $\gamma$-ray Xsection</th>
<th>$\sigma_{\gamma}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of atoms</td>
<td>$n_x$ and $n_c$</td>
</tr>
<tr>
<td>(integer for chemical compounds)</td>
<td></td>
</tr>
<tr>
<td>Peak area</td>
<td>$A$</td>
</tr>
<tr>
<td>Detector efficiency</td>
<td>$\varepsilon$</td>
</tr>
<tr>
<td>Absorption correction</td>
<td>$f$</td>
</tr>
<tr>
<td>Enrichment</td>
<td>$\theta$</td>
</tr>
<tr>
<td>Absolute $\gamma$-ray decay probability</td>
<td>$P_{\gamma}$</td>
</tr>
</tbody>
</table>
**Precision of internal $\sigma_\gamma$ calibration for enriched samples**

$$
\sigma_{\gamma x} = \sigma_{\gamma c} \frac{n_c}{n_x} \frac{A_{\gamma x}}{A_{\gamma c}} \frac{\varepsilon (E_{\gamma c})}{\varepsilon (E_{\gamma x})} \frac{f (E_{\gamma c})}{f (E_{\gamma x})}
$$

<table>
<thead>
<tr>
<th>Factor number</th>
<th>Type of error</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Systematic (~ 1%)</td>
</tr>
<tr>
<td>Derived from primary (H) or secondary (Cl, N…) standards (table or dedicated experiment)</td>
<td></td>
</tr>
<tr>
<td>2.</td>
<td>0</td>
</tr>
<tr>
<td>Exactly 1</td>
<td></td>
</tr>
<tr>
<td>3.</td>
<td>Statistical</td>
</tr>
<tr>
<td>Uncertainty can be decreased by time or by count rate</td>
<td></td>
</tr>
<tr>
<td>4.</td>
<td>Statistical + systematic, 0 at pivot ($\equiv E_{\gamma c}$) point, (~ 1%)</td>
</tr>
<tr>
<td>Uncertainty can be decreased to systematic level (Most important term which can be minimized by improving standards)</td>
<td></td>
</tr>
<tr>
<td>5.</td>
<td>Statistical + systematic (~ 1%)</td>
</tr>
<tr>
<td>Can be measured or calculated using attenuation models (thin sample minimizes the uncertainty)</td>
<td></td>
</tr>
</tbody>
</table>
Improvement of photo peak efficiency standards

- High energy radioactive $\gamma$-ray sources
  - $^{56}$Co, $^{66}$Ga and $^{226}$Ra

- High energy capture sources
  - $^{14}$N(n,$\gamma$)$^{15}$N primary source
    - Belgya, T., Improved accuracy of gamma-ray intensities from basic principles for the calibration reaction $^{14}$N(n,$\gamma$)$^{15}$N. Phys. Rev. C 74 (2006) 024603-1-8
  - $^{35}$Cl(n,$\gamma$) and $^{NAT}$Cr(n,$\gamma$) secondary sources (not yet fully compatible with N source)
$^{14}\text{N}(n,\gamma)$ spectrum
Determination of nitrogen intensities and detector efficiency function in one step
Crossing Intensity Sum (CIS)

\[ I_{i,j} = A_{i,j} \varepsilon^{-1}(E_{i,j}) \]

\[ T_f = \sum_{i>f}^{l>f} \sum_{j>f}^{l>f} A_{i,j} \varepsilon^{-1}(E_{i,j}) = C \quad f = 1,2,...n-1 \]

\[ Q = \sum_{1\leq f \leq n-1} (T_f - C)w_{f,s}(T_s - C) + \sum_{m} \left( \frac{\varepsilon_{m}^{-1} - \varepsilon^{-1}(E_{i,j})}{\sigma_m^2} \right)^2 \]

- Sums of crossing intensities are constant C
- Least-squares fit for inverse efficiency function \( \varepsilon^{-1} \) and C
- Input is peak areas and efficiencies at low energy
- CIS for line 1 \( \equiv \) intensity sum to the ground state
- CIS for line n-1 \( \equiv \) intensity sum for the primary transitions

New: \( \sigma_{th} = 80 - 83 \text{ mb} \)
Mughabghab: \( \sigma_{th} = 79.8(14) \text{ mb} \)
Results

- **Radioactive sources**
- **Spline fit**
- **Hypermet fit with Jurney intensities**

### Z-score vs. Efficiency
- Z-score:
  - -3
  - -2
  - -1
  - 0
  - 1
  - 2
  - 3
- Efficiency:
  - $10^{-5}$
  - $10^{-4}$
  - $10^{-3}$

### Intensity Ratio

- **Ratio** = Intensity Jurney / Intensity this work
### Total neutron capture cross section from $\gamma$-spectroscopy

<table>
<thead>
<tr>
<th>Method</th>
<th>Equation</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\sigma_{th} = \frac{\sigma_\gamma}{\theta P_\gamma}$</td>
<td>$P_\gamma$ must be known, for example from beta decay if the captured nucleus is unstable.</td>
</tr>
<tr>
<td>2</td>
<td>$\sigma_{th} = \sum_{f=1}^{n-1} \sigma_{\gamma c\rightarrow f} (1 + \alpha_f)(1 + PCC_f)$</td>
<td>The sum of all primary transitions from the capture state can be used for nuclei with relatively simple decay scheme.</td>
</tr>
<tr>
<td>3</td>
<td>$\sigma_{th} = \sum_{i=2}^{n} \sigma_{\gamma i\rightarrow g.s.} (1 + \alpha_i)(1 + PCC_i)$</td>
<td>The sum of all ground state transitions can be used for nuclei with relatively simple decay scheme. Conversion coefficients $\alpha$ must be known.</td>
</tr>
<tr>
<td>4</td>
<td>Average of CISs: $Q = \min \left{ \sum_{1\leq f \leq n-1} \left( T_f - \sigma_{th} \right) w_{f,s} \left( T_s - \sigma_{th} \right) \right}$</td>
<td>Well balanced and relatively simple decay scheme. Conversion coefficients $\alpha$ must be known.</td>
</tr>
<tr>
<td>5</td>
<td>$\sigma_{th} = \sum_i E_i \sigma_{\gamma i} (1 + \alpha_i)(1 + PCC_i) / B_n$</td>
<td>The energy weighted sum can be used for any nuclei with resolved gamma-transitions. $E_i$ is the energy of the transition, $B_n$ is the binding energy and PCC is the pair conversion.</td>
</tr>
</tbody>
</table>
For which nuclei we can use that
(Simple and complex spectra)

1.0E+00 1.0E+01 1.0E+02 1.0E+03 1.0E+04 1.0E+05 1.0E+06 1.0E+07 1.0E+08 1.0E+09 1.0E+10 1.0E+11 1.0E+12 1.0E+13 1.0E+14 1.0E+15 1.0E+16

Counts

- 14N(n,g)
- 57Fe(n,g)
- 101Ru(n,g)
- Eu(n,g)

Zoltán Kis

x10^3, manageable
x10^6, very complex
x10^9, extremely complex
Consequences

- Method 1 based on decay $\gamma$-rays or X-rays can be used for any complexity
  - Precision depends on the $P_\gamma$ value
- Method 2-5 is applicable for nuclei with manageable decay scheme (~500 $\gamma$-rays)
  - Limitation can be the unobserved conversion electron intensities
- Method 5 is applicable for spectra with resolvable $\gamma$-lines (~700-800 $\gamma$-rays)
  - Limitation can be the unobserved conversion electron intensities
  - Similar to the weighting function method used with C$_6$D$_6$ detectors
- Above that quasi continuum of $\gamma$-rays appears in the spectra
  - A possible approach is stripping or deconvolution of spectra on the few percent level accuracy
  - C$_6$D$_6$ detector or total absorption detector (needs highly enriched sample, unobserved conversion electron can also be a problem)
Question of completeness from the experimental point of view

• The Q-value test (equivalent to method 5) was applied in the past to estimate completeness

\[ B_n = \sum \frac{E_i \sigma_{\gamma_i}}{\sigma_{\text{th}}} ; \quad \sigma_{\text{th}} = \sum \frac{E_i \sigma_{\gamma_i}}{B_n} \]

• This is still the best way to find out the degree of completeness, however we propose to use the inverse Q-value instead and compare it directly to other independent Xsection values

• There are experimental methods that do not depend on observing all of the \( \gamma \)-rays, they provide the independent Xsections (they usually have other problems)
  - Pile oscillator, activation, transmission and calorimeter
An example

$^{101}\text{Ru}(n,\gamma)$ reaction (proposed by ILL)

Earlier data:
- No $\gamma$-rays in the ENSDF database
- EXFOR

<table>
<thead>
<tr>
<th>$\sigma_{th}$ (b)</th>
<th>Facility</th>
<th>Method</th>
<th>Author</th>
<th>year</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.4(9)</td>
<td>Reactor Internal McMaster</td>
<td>PGAA</td>
<td>Islam</td>
<td>1991</td>
</tr>
<tr>
<td>3.1(9)</td>
<td>Reactor Oakridge</td>
<td>Activation ??</td>
<td>Halperin</td>
<td>1964</td>
</tr>
<tr>
<td>5.5(1.4)</td>
<td>Reactor Oakridge</td>
<td>Mass spectrometry</td>
<td>Halperin</td>
<td>1965</td>
</tr>
</tbody>
</table>

The only independent or different method is the mass spectrometry
$^{101}\text{Ru}(n,\gamma)$ reaction studied at our PGAA facility

Unobserved continuum is at least 40%  
$^{101}\text{Ru}$ g.s. spin $5/2^+$ → capture state spins $2^+$ and $3^+$  
$^{102}\text{Ru}$: final state cumulative level number for spins 2,3,4,5 at 9.3 MeV $\sim 1.5 \times 10^5$  
Minimum observed $\sigma_\gamma$ in the 2-5 MeV range is 0.001 b  
Estimate: maximum missing intensity = $1.5 \times 10^5 \times 0.001/4 = 37.5$ b ! → useless limit → We need to use better model (e.g. DICEBOX Frantisek Becvar)
Conclusions

• The PGAA-NIPS facilities provide excellent means for determination of $\sigma_{\text{th}}$ and $\sigma_\gamma$
  – Good background conditions
  – Chopped neutron beam option
  – Firmly established methods
• Present methods can provide accurate cross sections for nuclei with moderated level density with PGAA
• Activation works for higher level density nuclei, but accuracy is limited by the accuracy of decay branching ratio
• Method outlined by Frantisek Becvar helps for nuclei with higher level density to determine capture cross sections
• Other experimental methods need to be worked out for high level density nuclei
Thank you for your attention!
Test of nitrogen new intensities $^{27}$Al(n,γ) reaction inverted Q-value

Test with $^{27}\text{Al}(n, \gamma)$

Method 4
$$\sigma_{th} = \sum E_i \sigma_{\gamma_i} c_i / B_n$$

Method 1
$$\sigma_{th} = \sigma_{\gamma 1779}$$

Al-27
\( ^{206}\text{Pb}(n,\gamma) \) spectrum

\( ^{206}\text{Pb}(n,\gamma) \) Compton-suppression

Counts

counts

\( E_\gamma \) (keV)

counts

\( E_\gamma \) (keV)

\( ^{207}\text{Pb} 6738 \)

\( ^{207}\text{Pb} 6738 \) SE

\( ^{207}\text{Pb} 898 \)

Annih.

H 2223

\( ^{208}\text{Pb} 7368 \)

\( ^{207}\text{Pb} 6738 \)

\( ^{207}\text{Pb} 6738 \) SE

\( ^{207}\text{Pb} 6738 \) SE

\( ^{207}\text{Pb} 898 \)

Annih.

H 2223

\( ^{208}\text{Pb} 7368 \)
New neutron capture decay scheme of $^{207}$Pb

Configurations

$4s_{1/2}$
$3d_{5/2}$

$3^{-}_{208Pb} \otimes 3p_{1/2}^{-1}$

$3p_{3/2}^{-1}$

$3p_{1/2}^{-1}$

E. Radermacher et al., NP A620 (1997) 151-170; incollaboration with IRMM, P. Schillebeeckx
Crossing Intensity Sums for the decay scheme of $^{207}\text{Pb}$

\[
\text{average}=1.73(2)
\]
## Results for $^{204,206,207}$Pb Xsec

<table>
<thead>
<tr>
<th>Isotope</th>
<th>This work (mb)</th>
<th>Mugabgab (mb)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{204}$Pb</td>
<td>482(20)</td>
<td>661(70)</td>
<td>preliminary</td>
</tr>
<tr>
<td>$^{206}$Pb</td>
<td>28.7(7)</td>
<td>26.6(12)</td>
<td>Increase is due to the N source</td>
</tr>
<tr>
<td>$^{207}$Pb</td>
<td>649(14)</td>
<td>625(30)</td>
<td>Increase is due to the N source</td>
</tr>
</tbody>
</table>
$^{127,129}$I chopped beam (n,γ) spectra

![Graph showing gamma spectra with peaks at 417, 536, 669, 739, and 1063 keV, and beta decay of $^{128}$I to $^{128}$Xe at 443 keV.]
Simplified decay scheme of $^{130}$I from literature

$T_{1/2} = 8.8 \text{ m}$

$T_{1/2} = 12.4 \text{ h}$

$^{130}$I

- $\beta^-$ 100%
- $129$I
- $1.6 \times 10^7 \text{ a}$
- $(n,\gamma)$

- $536 \text{ keV}$

- $^{130}$Xe

$^{130}$Xe

- $536 \text{ keV}$
## Results for $^{129}\text{I}$ Xsec

<table>
<thead>
<tr>
<th>Year</th>
<th>Author</th>
<th>Method</th>
<th>$\sigma_{th}$ (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1956</td>
<td>Purkayastha et al.</td>
<td>Activation reactor</td>
<td>35</td>
</tr>
<tr>
<td>1958</td>
<td>Roy et al.</td>
<td>Activation reactor</td>
<td>26.7(20)</td>
</tr>
<tr>
<td>1963</td>
<td>Pattenden et al.</td>
<td>TOF</td>
<td>28(2)</td>
</tr>
<tr>
<td>1969</td>
<td>Block et al.</td>
<td>TOF</td>
<td>31(4)</td>
</tr>
<tr>
<td>1983</td>
<td>Friedmann et al.</td>
<td>Activation reactor</td>
<td>33.9(19)</td>
</tr>
<tr>
<td>1996</td>
<td>Nakamura et al.</td>
<td>Activation reactor</td>
<td>30.3(12)</td>
</tr>
<tr>
<td>2007</td>
<td>Belgya et al.</td>
<td>Chopped cycl. act.</td>
<td>30.6(11)</td>
</tr>
</tbody>
</table>
Neutron energy is mostly below the first resonance

Wavelength spectra of thermal and cold beams

Energy is mostly below the first resonance at 1.8 Å = 25 meV.

\[ \lambda \sim \frac{1}{\sqrt{E}} \]
Total neutron capture cross section
another way

• Total energy detector concept, \( \mathcal{E} = E_\gamma \) or inverse Q value;
\[
\sigma_{\text{th}} = \sum_i E_i \sigma_{\gamma_i} (1 + \alpha_i)(1 + PCC_i) / B_n
\]

  – \( i \) = all transitions, (no decay scheme is needed)
  – Good identification is necessary, eg. \(^{209}\text{Bi},^{206}\text{Pb},^{27}\text{Al}, \) etc.
Total neutron capture cross section
another way

• From decay gamma rays: \( \sigma_{th} = \frac{\sigma_\gamma}{\theta P_\gamma} \)

  – Continuous beam activation (traditional)
  – Chopped beam activation (new)
  – Absolute decay probability \( P_\gamma \) is needed, \(^{238,235}\text{U},^{232}\text{Th},^{129}\text{I},^{99}\text{Tc},^{27}\text{Al},\) Te isotopes etc.,
Long decay of $^{130}\text{I}$ ground state

<table>
<thead>
<tr>
<th>E (keV)</th>
<th>$T_1/2$ (h)</th>
<th>Uncertainty</th>
<th>mean</th>
<th>deviation</th>
<th>ENSDF</th>
<th>uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>417</td>
<td>12.375</td>
<td>0.031</td>
<td></td>
<td></td>
<td>12.390</td>
<td>0.035</td>
</tr>
<tr>
<td>536</td>
<td>12.439</td>
<td>0.019</td>
<td></td>
<td></td>
<td>12.36</td>
<td>0.01</td>
</tr>
<tr>
<td>668</td>
<td>12.360</td>
<td>0.022</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>740</td>
<td>12.387</td>
<td>0.020</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1158</td>
<td>12.253</td>
<td>0.039</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Peak area of 536 and 668 keV $^{130}$I($\beta^-$)$^{130}$Xe decay gamma rays during activation and decay

$F=71\%, \, R_m=36\%, \, \lambda_m=1.279E-3, \chi^2=1.25$ proposed new
## New fit results and uncertainties

<table>
<thead>
<tr>
<th></th>
<th>Present work</th>
<th>Unc.</th>
<th>Rel. U.</th>
<th>Literature</th>
</tr>
</thead>
<tbody>
<tr>
<td>d</td>
<td>516.9</td>
<td>8.0</td>
<td>1.55</td>
<td>-</td>
</tr>
<tr>
<td>$R_m$</td>
<td>0.355</td>
<td>0.008</td>
<td>2.14</td>
<td>0.60(9) a)</td>
</tr>
<tr>
<td>$\lambda_m$</td>
<td>1.279E-3</td>
<td>1.6E-05</td>
<td>1.27</td>
<td>1.316E-3(3E-6) b)</td>
</tr>
<tr>
<td>$\lambda_g$</td>
<td>1.558E-05</td>
<td>2.3E-07</td>
<td>1.49</td>
<td>fixed</td>
</tr>
<tr>
<td>F</td>
<td>0.709</td>
<td>0.009</td>
<td>1.26</td>
<td>0.83(3) a)</td>
</tr>
<tr>
<td>$b_g$</td>
<td>0.997</td>
<td>0.016</td>
<td>1.58</td>
<td>fixed</td>
</tr>
<tr>
<td>$b_m$</td>
<td>0.987</td>
<td>0.034</td>
<td>3.39</td>
<td>fixed</td>
</tr>
<tr>
<td>deadt_corr</td>
<td>0.0218</td>
<td>0.0013</td>
<td>6.10</td>
<td>-</td>
</tr>
<tr>
<td>b668g</td>
<td>0.832</td>
<td>0.012</td>
<td>1.39</td>
<td>-</td>
</tr>
</tbody>
</table>


Peak area of 536 and 668 keV $^{130}$I($\beta$-)\textsuperscript{130}Xe decay gamma rays during activation and decay

$F=83\%$ (fixed), $R_m=0.6$, $\lambda_m=1.344E-3$ $\chi^2=4.5$ Literature
Uncertainty budget

ISO Guide to the expression of Uncertainty in Measurement (GUM)

\[\sigma_{\gamma x} = \sigma_{\gamma c} \frac{n_c}{n_x} \frac{A_{\gamma x}}{A_{\gamma c}} / \varepsilon(E_{\gamma x}) / f(E_{\gamma x}) \]

- Advantages of the relative internal calibration method
  - Absolute flux, inhomogeneity of sample and flux, multiple scattering (build up effect), dead time, energy distribution of flux, sample weight cancel out

- Uncertainty components for partial gamma ray cross sections
  - Uncertainty \((1\sigma, \delta A)\) of the area \(A\) is obtained from peak fitting with Hypermet PC
  - Uncertainty efficiency ratio can be obtained from the correlation matrix of the efficiency fit. Relative uncertainty of the efficiency is about 0.5-1% in the 0.1-10 MeV range.
  - Gamma and neutron self absorption is calculated with numerical integration over simple shape of samples. For thin samples they are close to 1 and the estimated uncertainty is 5% of the difference from 1

Continuous beam

\[
\sigma_{\gamma\gamma}(1 \pm \delta \sigma_{\gamma\gamma}) = \sigma_{\gamma\gamma} \frac{A_{\gamma\gamma} n_c \varepsilon(E_{\gamma\gamma}) f(E_{\gamma\gamma})}{A_{\gamma\gamma} n_e \varepsilon(E_{\gamma\gamma}) f(E_{\gamma\gamma})}.
\]

\[
1 \pm \sqrt{(\delta A_{\gamma\gamma})^2 + (\delta \varepsilon(E_{\gamma\gamma}))^2 + (\delta \varepsilon(E_{\gamma\gamma}))^2 - 2 \delta \text{cov}(\varepsilon(E_{\gamma\gamma}), \varepsilon(E_{\gamma\gamma})) + (\delta \sigma_c)^2 + \left( \frac{\delta \varepsilon(E_{\gamma\gamma})}{\varepsilon(E_{\gamma\gamma})} \right)^2}
\]

• Without correlation we get overestimate of the uncertainty
• For sum of partial cross sections the uncertainty calculus must be used, if we use simple re-normalization than the correlation is neglected
• For calculus of decay partial gamma-ray cross sections and uncertainties related to chopper methodology please see

Selected recent publications

Handbook of PGAA with neutron beams (Eds. G.L. Molnár, Kluwer Academic Publisher), 2004

$^{99}\text{Tc}(n,\gamma)$:


$^{127,129}\text{I}(n,\gamma)$:


$^{238}\text{U}(n,\gamma)$:


$^{209}\text{Bi}(n,\gamma)$


Te isotopes:

Pb in progress

$^{15}\text{N}, ^{208}\text{Pb}, ^{27}\text{Al}$:


Pd:

Fig. 4. Absolute full-energy peak efficiency (left-hand scale) as a function of γ-ray energy for the Budapest detector. The residuals of the fit to the data points (experimental value minus fitted value divided by the experimental uncertainty) are shown at the top of the figure. The dashed curve represents the relative standard uncertainties for the fitted curve (in percent, right-hand scale).
$^{99}$Tc 0.5 g sample (n,\(\gamma\)) spectrum
\(^{99}\text{Tc} (n,\gamma \gamma)\) prompt-coincidence spectrum

Gate on 299 keV

Counts

\[ \begin{align*}
31 & \quad 39 & \quad 63 & \quad 75 & \quad 90 & \quad 99 & \quad 105 & \quad 128 & \quad 172 & \quad 179, 180, 181 \\
\end{align*} \]

inconsistent
consistent
unplaced
Partial $\gamma$-ray production cross sections of capture and decay lines for a $^{99}$Tc target

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>Origin</th>
<th>$P_\gamma$ ($\gamma$/100 captures or decays)</th>
<th>$\sigma_\gamma$ (b)</th>
<th>Sensitivity (cps/mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>172.1</td>
<td>$^{99}$Tc(n,$\gamma$)</td>
<td>67±6</td>
<td>16.61±0.15</td>
<td>3.0</td>
</tr>
<tr>
<td>223.4</td>
<td>$^{99}$Tc(n,$\gamma$)</td>
<td>6.1±0.6</td>
<td>1.472±0.013</td>
<td>0.24</td>
</tr>
<tr>
<td>263.5</td>
<td>$^{99}$Tc(n,$\gamma$)</td>
<td>5.9±0.5</td>
<td>1.425±0.012</td>
<td>0.21</td>
</tr>
<tr>
<td>539.5</td>
<td>$^{100}$Tc $\beta^-$</td>
<td>6.6±0.5 $^b$</td>
<td>1.604±0.014</td>
<td>0.14</td>
</tr>
<tr>
<td>590.7</td>
<td>$^{100}$Tc $\beta^-$</td>
<td>5.3±0.5</td>
<td>1.296±0.011</td>
<td>0.10</td>
</tr>
<tr>
<td>89.5</td>
<td>$^{99}$Tc $\beta^-$</td>
<td>(6.5±1.5)$ \times$10^{-4} $^c$</td>
<td></td>
<td>4.3$ \times$10^{-3}</td>
</tr>
</tbody>
</table>

### Inferred total thermal-neutron capture cross section of $^{99}$Tc

<table>
<thead>
<tr>
<th>Method</th>
<th>Basis</th>
<th>$\sigma$ (b)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{100}$Tc($\beta^{-}$)$^{100}$Ru</td>
<td>539 $\gamma$</td>
<td>24.7±2.3</td>
<td>with $P_\gamma$ Furutaka et al.</td>
</tr>
<tr>
<td></td>
<td>591 $\gamma$</td>
<td>23.9±1.8</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>24.3±2.2</td>
<td>unweighted average</td>
</tr>
<tr>
<td>$^{99}$Tc($n,\gamma$)$^{100}$Tc</td>
<td>$\sum \sigma_\gamma$ g.s.</td>
<td>21.21±0.17</td>
<td>lower limit</td>
</tr>
</tbody>
</table>

**Literature:** EXFOR database

- H. Pomerance 1975: 19±2 b, pile oscillator
- R.B. Tattersall 1960: 16±7 b, pile oscillator
- N.J. Pattenden 1958: 25±2 b, transmission
- M. Lucas 1977: 20±2 b, mass spectrometer
- V.V. Ovechkin 1973: 24±4 b, activation
- H. Harada 1995: 22.9±2.6 b, activation
- Mughabgab 2003: 20±1, evaluation INDC(NDS)-440
Parameters of the NIPS station

- Neutron beam cross section: \(2.5 \times 2.5\, \text{cm}^2\)
- Thermal-equivalent flux at target: \(\approx 3 \times 10^7\, \text{cm}^{-2}\text{s}^{-1}\)
- Vacuum in target chamber (optional): \(\approx 1\, \text{mbar}\)
- Form of target at room temperature: Solid, powder, liquid, gas in pressure container
- Largest target dimensions: \(1.5 \times 1.5 \times 3.5\, \text{cm}^3\)
- \(\gamma\)-ray detector No.1: n-type coax. HPGe
  - Relative efficiency: 13\% at 1332 keV
  - FWHM: 1.8 keV at 1332 keV
- \(\gamma\)-ray detector No 2.
  - Relative efficiency: 30\% at 1332 keV
  - FWHM: 1.9 keV at 1332 keV
- \(\gamma\)-ray detector No 3.
  - Planar HPGe
  - FWHM: 0.6 keV at 122 keV
Parameters of the PGAA station

- beam cross section: \( \leq 2 \times 2 \text{ cm}^2 \)
- Thermal-equivalent flux at target: \( \approx 5 \times 10^7 \text{ cm}^{-2}\text{s}^{-1} \)
- Vacuum in target chamber (optional): \( \approx 1 \text{ mbar} \)
- Form of target at room temperature: Solid, powder, liquid, gas in pressure container
- Largest target dimensions: \( 4 \times 4 \times 10 \text{ cm}^3 \)
- \( \gamma \)-ray detector: n-type coax. HPGe, with BGO shield
- Distance from target to detector: 23.5 cm
- Relative efficiency: 25% at 1332 keV
- FWHM: 1.8 keV at 1332 keV
- Compton suppression enhancement: \( \approx 5 \) (1332 keV) to \( \approx 40 \) (7000 keV)
What’s **NIPS**?

- **Neutron Induced Prompt** gamma-ray **Spectroscopy**
- **Intent:** To build a multipurpose experimental station
  - Close detector geometry (2.5 cm)
  - Place for more detectors (≥3)
  - Good shielding (\(^6\)Li-poly)
  - Multiparameter data acquisition
The $^{99}$Tc

- One of the most important LLFF
- $^{99}$Tc half-life: 210 000 years
- cumulative fission yield in reactor: 6.1%
- The $(n,\gamma)$ reaction can efficiently destroy the Tc waste
99Tc transmutation

• TARC experiments at CERN to measure transmutation rates
  • A. Abanades et al., Nucl. Instr. and Methods A 478 (2002) 577–730

Carlo Rubbia's TARC (Transmutation by Adiabatic Resonance Crossing) experiment at CERN. Accelerator-driven transmutation has emerged as a potentially complementary technology for radioactive waste handling by transmuting the longest-lived radioactive isotopes into short-lived or stable ones.
Scheme of transmuting $^{99}$Tc

- $^{99}$Tc
  - 2.11 x 10^5 y
  - 1.6 x 10^{-3}\% transmutation path
  - 90 keV
  - 100% $\beta^-$
  - $^{99}$Ru
    - stable
  - $^{99}$Tc+n
    - transmutation path
  - 15.8 s
  - 5.7% 591 keV
  - 0.6% 539 keV
  - 93% 539 keV

$^{100}$Ru
- stable

$^{100}$Tc
- 5.7% 591 keV
- 0.6% 539 keV
- 93% 539 keV

±10% major source of uncertainty in TARC experiments
99Tc measurements at our PGAA & NIPS facilities

✅ NH₄TcO₄  PGAA (partial cross section)
✅ ⁹⁹Tc(n,γ)  PGAA (rel. γ intensities)
✅ ⁹⁹Tc(n,γ)  Chopped beam PGAA

(¹⁰⁰Tc β⁻ decay rel. γ intensities)

Evaluation is in progress:
• ⁹⁹Tc(n,γγ)  Coincidence (decay scheme)
• ⁹⁹Tc(d,p)¹⁰⁰Tc  High resolution proton spectrum
  (level scheme)

measured at , TU Munich
$^{100}\text{Tc}$ levels observed in (d,p) spectrum
Quantities we can measure and their role in nuclear waste transmutation

• Partial gamma-ray cross sections
  on-line transmutation yield measurement
  non-destructive assay of waste by PGAA

• Decay gamma-ray partial cross sections
  on- and off-line transmutation yield measurement

• Thermal neutron capture cross sections
  transmutation yield calculations
  normalization point for differential cross section experiments (e.g. GELINA, Geel, Belgium, n-TOF, CERN)

Our goal is to measure these quantities with high precision
$^{99}$Tc (d,p) spectrum (München tandem)

D:\munka\tcdp_wirth\tc_30deg.csv

$P_k = EMG  \ 22 \ Peaks$

$r^2 = 0.996905 \  SE = 25.2052 \  F = 4723.9$
Tc(n,γ) partial Level-scheme

Why to measure nuclear waste, structural material capture cross sections?

• Waste is generated by power production
• Safe disposal → transmutation by ADS
• Problematic wastes are the Long-Lived Fission Fragments or LLFFs ($^{99}$Tc, $^{129}$I,…) and minor actinides
• Structural materials are parts of ADS (Bi, Pb, …)
• For optimization of transmutation yields, control of transmutation and detection of various isotopes in waste:

  Accurate $\gamma$-ray yields and thermal neutron cross sections are needed
Objectives

(n,γ) reactions

PGAA

Cross sections

Nuclear Physics

Research
- Archaeology (IAEA)
- Geology
- Catalyst
- Material sciences
- In-beam Mössbauer
- Safeguard

Methodology
- PGAA library (IAEA, LBL)
- Chopped beam PGAA
- Standards (IAEA)

Research
- Xsections for ADS
- LLFF
  - 99Tc, 129I (IRMM)
- Fuel
  - 238,235U, 232Th
- Structural mat.
  - 209Bi, 204,206,207Pb (IRMM)

Methodology
- Internal comparator
- Chopped beam

Research
- Decay schemes
  - 204,206,207Pb (IRMM)
  - 99Tc
- Strength functions
  - 57Fe (Oslo, Frank L.)
  - Mo, Yb, Gd
- Modeling Decay s.
  - 99Tc

Methodology
- (n,γ), (n,γγ)
- Monte Carlo
Editor: G. L. Molnár†

1. Principles of the PGAA method
   (Zs. Révay, T. Belgya)

2. Beams and Facilities
   (R.M. Lindstrom, Zs. Révay)

3. Samples and Standards
   (R.M. Lindstrom, Ch. Yonezawa)

4. Gamma-Ray Spectrometry
   (T. Belgya, Zs. Révay)

5. Quantitative Analysis
   (Ch. Yonezawa)

6. Applications of PGAA with Neutron Beams
   (D.L. Anderson, Zs. Kasztovszky)

7. Appendices Reference Data
   (R.B. Firestone, G.L. Molnár, Zs. Révay)

+ CD supplement

General precision 1-5% for $\sigma_\gamma$
$\text{NH}_4\text{TcO}_4 (n, \gamma)$ spectrum, H comparator