Measuring neutron capture rates on ILL-produced unstable isotopes for nucleosynthesis studies

J. Lerendegui-Marco\textsuperscript{1}, C. Guerrero\textsuperscript{1}, C. Domingo-Pardo\textsuperscript{2}, A. Casanovas\textsuperscript{3}, S. Halfon\textsuperscript{4}, S. Heinitz\textsuperscript{5}, N. Kivel\textsuperscript{5}, U. Köster\textsuperscript{6}, M. Paul\textsuperscript{7}, R. Dressler\textsuperscript{5}, D. Schumann\textsuperscript{5}, M. Tessler\textsuperscript{7} and The n_TOF Collaboration\textsuperscript{8}

\textsuperscript{1) Universidad de Sevilla, Sevilla, Spain}
\textsuperscript{2) Instituto de Física Corpuscular, Paterna, Spain}
\textsuperscript{3) Universitat Politècnica de Catalunya, Barcelona, Spain}
\textsuperscript{4) Soreq NRC, Yavne, Israel}
\textsuperscript{5) Paul Scherrer Institut, Villigen, Switzerland}
\textsuperscript{6) Institut Laue-Langevin, Grenoble, France}
\textsuperscript{7) Racah Institute of Physics, Hebrew University, Jerusalem, Israel}
\textsuperscript{8) European Center for Nuclear Research (CERN), Geneva, Switzerland}

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Interest for neutron capture measurements

- **Nuclear Astrophysics: s-process**

  - **Site:** He-intershell in AGB stars (red giant)
  - **Branching point:** $\beta^-$ decay competes with $(n,\gamma)$

  Suff. long $\tau_{1/2} ||$ Suff. High $\Phi_n$

\[ \begin{array}{ccc}
(n,\gamma) & (n,\gamma) & (n,\gamma) \\
(n,\gamma) & (n,\gamma) & (n,\gamma) \\
(n,\gamma) & \beta^- & (n,\gamma) \\
\end{array} \]

Branching point s-process

$\sigma(n,\gamma)$ input from nuclear data + ratio of chemical abundances $\rightarrow$ bounds the temperature and neutron flux in stellar models
(n,γ) measurements on unstable isotopes

Main limitation of current facilities and techniques: Need for massive (~mg) and isotopically pure samples

Successful collaboration to produce relevant unstable isotopes (ILL) and prepare high quality targets (PSI)

After this big effort to produce the samples: Complementary (n,γ) measurements using different beams and techniques available (n_TOF, LILIT, TRIGA, BRR)
$^{171}$Tm (n,γ):
thermal, resonance and MACS cross sections
**$^{171}$Tm(n,γ): Preparation of radioactive targets**

Production via (n, γ) or (n, γ)+β⁻ at the ILL research reactor [Contact: Ulli Koester]
Neutron flux: 1.5x10^{15} n/cm²/s
Irradiation time: 55 days

$^{171}$Tm: $^{170}$Er(n,γ)$^{171}$Er(β⁻, 7.5h)$^{171}$Tm (enrichment 1.8%)

3.81 mg of $^{171}$Tm (1.9 y) [1.3 x 10^{19} atoms]

Do not exist in nature → Small masses produced
Activity challenge to handle and measure

Chemical separation and targets @ PSI [Stephan Heinitz’s Talk]

Assembly mounted –

$^{147}$Pm deposit (20 mm diameter)
Aluminum (7 μm) backing
PCB frame (50 mm diameter)
Mylar (5 μm)
$^{171}\text{Tm (n,}\gamma\text{): Energy ranges and beams}$

MACS: Maxwellian Averaged Cross Section

NEUTRON CROSS SECTION MEASUREMENTS:
How to produce of neutrons in each energy range
How to measure neutron energy and detect the reaction products
$^{171}_{\text{Tm}}(n,\gamma)$ MACS: LiLiT facility @ SARAF

Liquid lithium: $^7\text{Li}(p,n)$

$\sim 1.935 \text{ MeV protons}$

1-2 mA

$^{197}_{\text{Au}}(n,\gamma)^{198}_{\text{Au}}$ \(\rightarrow\) $\beta^-$ decay with $t_{1/2} = 2.69 \text{ d}$ \(\rightarrow\) $E_\gamma = 412 \text{ keV}$

Case 2. $^{171}_{\text{Tm}}(n,\gamma)^{172}_{\text{Tm}}$ \(\rightarrow\) $\beta^-$ decay with $t_{1/2} = 2.65 \text{ d}$ \(\rightarrow\) $E_\gamma = 1093, 1387, 1529, 1608 \text{ keV}$
$^{171}\text{Tm}(n,\gamma)$ MACS: Results

$$\sigma_{\text{MACS}}\left(^{171}\text{Tm}, \kappa T=30 \text{ keV}\right) = 198 (22) \text{ mb}$$

Significant reduction of the MACS at 30 keV

JEFF 3.2 (x6) and also KADoNiS overestimate the MACS

Evolution of calculations also tends to lower the cross-section
$^{171}$Tm(n,γ) Thermal Cross-section

- **Thermal cross-section:**
  - Maxwellian spectra @ KT= 25meV
- **Prompt Gamma Activation Analysis** using the n_TOF targets

**MACS** and thermal provide just two points: TOF measurement needed for pointwise cross-section
CERN-n_TOF: Time-of-flight technique

**BEAM LINE EAR1**

Spallation target

**Pb**

PS Proton pulses

(20 GeV/c) $\sigma = 7$ ns

**Pb**

Spallation

MeV-GeV Neutrons

**C_6D_6**

Scintillators

Reaction products

Transmission

Scattering

Flux ($\Phi$) (neutrons/cm$^2$)

Time-of-Flight to $E_n$ relation (non-rel.):

$$ToF = t - t_0 \propto \frac{L}{\sqrt{E_n}}$$

5cm water moderator

5cm water moderator

**L = 184 m**

$E_n$ = 12 MeV to 1 GeV

$\gamma$-rays

$\sigma = 7$ ns

$\nu$-MCAS

Capture setup

SILL

ToF Monitors

FLUX Monitors

<table>
<thead>
<tr>
<th>Spallation target</th>
<th>Shielding</th>
<th>Filter station</th>
<th>First collimator</th>
<th>Shielding</th>
<th>Sweeping magnet</th>
<th>Second collimator</th>
<th>Experimental area</th>
<th>Beam dump</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pb</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

0 0.35 70.2 134.9 134.9 136.7 145.4 178.0 182.3 190.3 200
\[ \sigma_{n,\gamma}(E_n) = \frac{C(E_n) - B(E_n)}{n_{\text{act}} \epsilon_{\text{det}} \Phi_n(E_n)} \]

- Counts vs En
- Resonance parameters: \( E, J, \Gamma_n, \Gamma_{\gamma} \)
- Efficiency to detect a cascade
- Counts -> Pointwise cross section
- Challenge: Sample activity
- Resonance analysis 1 - 700 eV
$^{171}$Tm (n,γ) at n_TOF: Resonance region

Before: Just TENDL-2012 (TALYS calculation)

n_TOF data: Overestimation of density and strength in TENDL $S_0$, $D_0$, $<\Gamma_\gamma>$ significantly smaller than systematics (Mughabghab)
Other s-process isotopes produced at ILL

**147Pm**

Branching at A= 147/148

\[ ^{146}\text{Nd}(n,\gamma)^{147}\text{Nd} (\beta^-, 10\text{d})^{147}\text{Pm} \rightarrow \text{Final mass 83 ug!} \]

**n_TOF - EAR2**:  
First time ever (n, γ)  
Few resonances (smallest mass ever!)

**MACS @ LiLiT**: **1200 ± 120** mb  
Significantly larger than previous measurement

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**204Tl**

**205Pb/205Tl clock**  
Early universe

\[ ^{203}\text{Tl}(n,\gamma)^{204}\text{Tl} \rightarrow 6 \text{ mg of }^{204}\text{Tl} (3.78 \text{ y}) \text{ [3.25e19 atoms]} \]

**n_TOF - EAR1**: First time ever (n, γ)  
- 200 GBq + high Q_β decay  
-Pellet disaggregation during irradiation  
-Resonances (KeV) → Direct MACS  
No activation possible (**205Tl** stable)
Relevant branching $A=80$ to constrain $s$-process $T$, density and role of main/weak

$^{78}\text{Se}(n,\gamma)^{79}\text{Se} (327\text{ky}) \rightarrow \sim 8\text{mg}$

-Difficulty in the past: Impurities in the Pb-Se alloy (Melts @ 217 ºC)
-This time: Pure $^{208}\text{Pb-}^{78}\text{Se}$ seed

Planned to be measured at n_TOF - EAR2: First time ever $(n, \gamma)$
Direct MACS from resonances

No activation possible ($^{80}\text{Se}$ stable)

Branching $A=163$, beta decay rate $\leftrightarrow$ mass density

HOLMES Project: neutrino mass with a calorimeter measuring the endpoint of the $^{163}\text{Ho}$ $\beta^+$ spectrum.

$^{162}\text{Er}(n, \gamma)^{163}\text{Er} (b^+)^{163}\text{Ho} (4570 \gamma) : \sim 6\text{ mg}$

Proposed at n_TOF - EAR1+EAR2: First time ever $(n, \gamma)$
Resonance parameters + MACS
Summary and conclusions

- **s-process: (n, γ) cross-section** certain isotopes key input for stellar evolution models

- \(^{171}\text{Tm},^{147}\text{Pm},^{204}\text{Th},^{79}\text{Se},^{163}\text{Ho}\): s-process branching points:
  Benefitted from the **collaboration** with ILL for the production + PSI for target preparation of radioactive targets, our main interest for both astrophysics and reactor physics.

- \(^{171}\text{Tm}\) (n,γ): different neutron beams & techniques in complementary energy ranges

- **n_TOF**: White neutron beam + Time-of-flight technique
  - Pointwise cross section \(\rightarrow\) Resonances and average parameters for the keV region

- **LiLiT @ SARAF**: Quasi-stellar spectra (\(^7\text{Li}(p,n)^7\text{Be}\) reaction)
  - MACS measurement via activation \(\rightarrow\) Decay of the produced \(^{172}\text{Tm}\) nuclei

- **TRIGA** (analysis ongoing) and **BRR** (near future): **Maxwellian spectra at thermal**
  - Measurement via activation and PGAA

- **n_TOF** and **SARAF** indicate a significant reduction of x-section compared to models
THANKS FOR YOUR ATTENTION!
### s-process branching points

**TABLE III. Feasibility of future TOF measurements on unstable branch-point isotopes at the FRANZ facility.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Half-life (yr)</th>
<th>$Q$ value (MeV)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{65}$Ni</td>
<td>100.1</td>
<td>$\beta^-$, 0.066</td>
<td>TOF work in progress (Couture, 2009), sample with low enrichment</td>
</tr>
<tr>
<td>$^{79}$Se</td>
<td>$2.95 \times 10^3$</td>
<td>$\beta^-$, 0.159</td>
<td>Important branching, constrains $s$-process temperature in massive stars</td>
</tr>
<tr>
<td>$^{81}$Kr</td>
<td>$2.29 \times 10^3$</td>
<td>EC, 0.322</td>
<td>Part of $^{79}$Se branching</td>
</tr>
<tr>
<td>$^{85}$Kr</td>
<td>10.73</td>
<td>$\beta^-$, 0.687</td>
<td>Important branching, constrains neutron density in massive stars</td>
</tr>
<tr>
<td>$^{95}$Zr</td>
<td>64.02 d</td>
<td>$\beta^-$, 1.125</td>
<td>Not feasible in near future, but important for neutron density low-mass AGB stars</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>2.0652</td>
<td>$\beta^-$, 2.059</td>
<td>Important branching at $A = 134, 135$, sensitive to $s$-process temperature in low-mass AGB stars, measurement not feasible in near future</td>
</tr>
<tr>
<td>$^{135}$Cs</td>
<td>$2.3 \times 10^6$</td>
<td>$\beta^-$, 0.269</td>
<td>So far only activation measurement at $kT = 25$ keV by Patronis et al. (2004)</td>
</tr>
<tr>
<td>$^{147}$Nd</td>
<td>10.981 d</td>
<td>$\beta^-$, 0.896</td>
<td>Important branching at $A = 147/148$, constrains neutron density in low-mass AGB stars</td>
</tr>
<tr>
<td>$^{147}$Pm</td>
<td>2.6234</td>
<td>$\beta^-$, 0.225</td>
<td>Part of branching at $A = 147/148$</td>
</tr>
<tr>
<td>$^{148}$Pm</td>
<td>5.368 d</td>
<td>$\beta^-$, 2.464</td>
<td>Not feasible in the near future</td>
</tr>
<tr>
<td>$^{151}$Sm</td>
<td>90</td>
<td>$\beta^-$, 0.076</td>
<td>Existing TOF measurements, full set of MACS data available (Abbondanno et al., 2004a; Wisshak et al., 2006c)</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>8.593</td>
<td>$\beta^-$, 1.978</td>
<td>Complex branching at $A = 154, 155$, sensitive to temperature and neutron density</td>
</tr>
<tr>
<td>$^{155}$Eu</td>
<td>4.753</td>
<td>$\beta^-$, 0.246</td>
<td>So far only activation measurement at $kT = 25$ keV by Jaag and Käppeler (1995)</td>
</tr>
<tr>
<td>$^{153}$Gd</td>
<td>0.658</td>
<td>EC, 0.244</td>
<td>Part of branching at $A = 154, 155$</td>
</tr>
<tr>
<td>$^{160}$Tb</td>
<td>0.198</td>
<td>$\beta^-$, 1.833</td>
<td>Weak temperature-sensitive branching, very challenging experiment</td>
</tr>
<tr>
<td>$^{162}$Ho</td>
<td>4570</td>
<td>EC, 0.0026</td>
<td>Branching at $A = 163$ sensitive to mass density during $s$ process, so far only activation measurement at $kT = 25$ keV by Jaag and Käppeler (1996b)</td>
</tr>
<tr>
<td>$^{170}$Tm</td>
<td>0.352</td>
<td>$\beta^-$, 0.968</td>
<td>Important branching, constrains neutron density in low-mass AGB stars</td>
</tr>
<tr>
<td>$^{171}$Tm</td>
<td>1.971</td>
<td>$\beta^-$, 0.098</td>
<td>Part of branching at $A = 170, 171$</td>
</tr>
<tr>
<td>$^{179}$Ta</td>
<td>1.82</td>
<td>EC, 0.115</td>
<td>Crucial for $s$-process contribution to $^{180}$Ta, nature’s rarest stable isotope</td>
</tr>
<tr>
<td>$^{185}$W</td>
<td>0.206</td>
<td>$\beta^-$, 0.432</td>
<td>Important branching, sensitive to neutron density and $s$-process temperature in low-mass AGB stars</td>
</tr>
<tr>
<td>$^{204}$Tl</td>
<td>3.78</td>
<td>$\beta^-$, 0.763</td>
<td>Determines $^{208}$Pb/$^{205}$Tl clock for dating of early Solar System</td>
</tr>
</tbody>
</table>
$^{171}$Tm (n,γ): a European trip towards the thermal, resonance and MACS cross sections

1) Irradiation of stable seeds
2) Separation and samples
3) Activation: thermal reactor
4) TOF + white n beam: Pointwise cross section
5) Activation: Quasi-stellar spectrum
6) PGAA: thermal reactor

Coordination and analysis
Nucleosynthesis through the $s$ process

Solar abundance distribution

Stellar burning

NSE

$r$-process

Weak

Main $s$-process

$p$-process

Others?

Mass number

Abundance

$10^{10}$

$10^{9}$

$10^{8}$

$10^{7}$

$10^{6}$

$10^{5}$

$10^{4}$

$10^{3}$

$10^{2}$

$10^{1}$

$10^{0}$

$10^{-1}$

$10^{-2}$

$10^{-3}$

$10^{-4}$

$10^{-5}$

$10^{-6}$

$10^{-7}$

$10^{-8}$

$10^{-9}$

$10^{-10}$
$^{171}\text{Tm}$ as part of branching at $A=170/171$

The $A=170/171$ branching point is one of the branchings that is independent of stellar temperature, therefore suited for constraining the s-process neutron density in low-mass AGB stars (i.e. main s-process component).


$$n_n = 0.7^{+4.9}_{-0.5} \cdot 10^8 \text{ cm}^{-3}$$

In view of the difficulties related to production of $^{170}\text{Tm}$, experimental information on $^{171}\text{Tm}$ becomes important as part of the branching, but also as the more important for improved HF predictions of the $^{170}\text{Tm}$ cross section.
$^{171}\text{Tm (n,}\gamma\text{): Activated nuclei to MACS}$

$$\sigma_{MACS}(Tm, kT = 30\text{keV}) = \frac{2}{\sqrt{\pi}} \frac{C_{En}(Tm, kT = 30\text{keV})}{\sigma_{\text{exp}}(Au) N_{172Tm}/N_{171Tm}}$$

$C_{En}(Tm, kT = 30\text{keV})$

$$= \frac{2}{\sqrt{\pi}} \int_{0}^{\infty} \sigma_{\text{lib}}(Tm, E_n) E_n e^{-E_n/kT} dE_n \frac{\int_{0}^{\infty} dE_n dE_n}{\int_{0}^{\infty} E_n e^{-E_n/kT} dE_n}$$

Correction for the shapes of the flux and the $^{171}\text{Tm}$ X-section

Correction for the shape of the flux of the Au ref. MACS

Measured quantity: Activated nuclei
Normalization factors of *INCLXX_HPT physics lists are improved in v10.1.1 by about 15% w.r.t v10.0.3 of the Geant4 toolkit.
$^{171}\text{Tm (n,γ) at n_TOF: Resonance region}$

Average resonance parameters

$D_0 = 21(3) \text{ eV (sys: 7 eV)}$

$S_0 = 0.90(26) \text{ (sys: 1.6)}$

$<\Gamma_\gamma> = 76(9) \text{ meV (sys: 100 meV)}$

Significant reduction of x-section wrt systematic studies
Branching points: example at A=147/148

Carlos GUERRERO “Neutron capture cross sections of the s-process branching points $^{147}$Pm, $^{171}$Tm and $^{204}$Tl”
Nuclei in the Cosmos NIC-2016, Niigta, Japan (June 20th-24th 2016)
Results for $^{147}$Pm(n,$\gamma$) @n_TOF-EAR2

- Activity background not a problem in EAR2
- Statistics very limited, due to small mass
- But still, 10-15 resonances observed (1st time!)
147\text{Pm}(n,\gamma)148\text{Pm} \rightarrow \beta^- \text{ decay with } t_{1/2} = 5.4 \pm 1.41 \text{ d}

1014 \text{ keV: from } 147\text{Pm}(n,\gamma)148\text{Pm} \rightarrow 1465 \text{ keV: from } 147\text{Pm}(n,\gamma)148\text{Pm}

Preliminary results indicate that the partial cross sections are quite different, while the only available (Reifarth:2003) data suggest only a difference of only 25%.
MACS results for $^{147}$Pm

\[ \sigma_{\text{MACS}}(^{147}\text{Pm}, \text{kT}=30 \text{ keV}) = 1236 (185^*) \text{ mb} \]

\[ \sigma_{\text{MACS}}(^{147}\text{gPm}, \text{kT}=30 \text{ keV}) = 406(60^*) \text{ mb} \]

\[ \sigma_{\text{MACS}}(^{147}\text{mPm}, \text{kT}=30 \text{ keV}) = 830(125^*) \text{ mb} \]

*Preliminary 15% uncertainty
$^{204}$Tl determines the $^{205}$Pb/$^{205}$Tl clock for dating of early Solar System

$^{205}$Pb ($t_{1/2} = 1.5 \times 10^7$ a) is produced only by the s-process:

The ratio $^{205}$Pb/$^{205}$Tl provides highly interesting chronometric information about the time span between the last nucleosynthetic events that were able to modify the composition of the solar nebula and the formation of solar system solid bodies.

(At present, there is an upper limit for the $^{205}$Pb/$^{205}$Tl abundance ratio of $9 \times 10^{-5}$ from meteorites)

K. Yokoi et al., Astronomy and Astrophysics 145, 339-346 (1985)

Blake et al., Nature, 1973
**HOLMES**: The Electron Capture Decay of $^{163}$Ho to Measure the Electron Neutrino Mass with sub-eV sensitivity

The determination of the absolute neutrino mass $m_\nu$ is still an open question in particle physics. Currently, the most stringent limit of $m_{\nu e} < 2 \text{ eV}$ was achieved for the electron anti-neutrino mass from the measurement of the endpoint of the decay of $^3\text{H}$. Novel experimental approaches exploring the endpoint of the $\beta^-$-decay of $^3\text{H}$ or the electron capture of $^{163}\text{Ho}$ offer great potential to reach a sub–eV sensitivity on $m_{\nu e}$.

The HOLMES experiment is going to investigate the neutrino mass with a calorimeter measuring the endpoint of the $^{163}\text{Ho}$ spectrum.
$^{163}$Ho as part of branching at A=163

$^{163}$Dy is stable, but $\beta^-$ (47 d) to $^{163}$Ho when fully ionized (stellar plasma)

Equilibrium abundance of $^{163}$Ho from decay of $^{163}$Dy and decay back from $^{163}$Ho makes it possible to produce $^{164}$Ho via (n,g). The equilibrium abundance of $^{163}$Ho is determined by the temperature and electron density in the star, which is directly related to the mass density.

S. Jaag and F. Käppeler, ApJ 464 (1996) → $^{163}$Ho MACS$_{30\text{keV}} = 2125(95)$ mb (0.2mg target)
Measuring $\sigma(n,\gamma)$: activation & ToF

Activation with a Maxwellian ($kT=$ tens of keV) neutron beam from $^7$Li$(p,n)$ reactions

This project:
Highest intensity $^7$Li$(p,n)$ beam worldwide: LiLiT @ SARAF (Israel)

Outcome: capture cross sections at a given $kT=$ tens of keV (ONE POINT!)

Time of Flight experiments with white neutron beams

This project:
Highest inst. intensity pulsed white beam worldwide: n_TOF@CERN
(we need to measure $(n,\gamma)$ $\gamma$-rays from GBq targets)

Outcome: (point-wise) capture cross sections as function of $E_n$
$^{171}$Tm (n,γ): MACS measurement @ LiLiT

$2 \times 10^{10}$ n/s/mA
1-2 mA
~2 kW

Liquid Lithium Target
(@SARAF, Israel)
$^{171}\text{Tm} (n,\gamma)$ measurement @ n_TOF

EAR1 using Total Energy Detectors

B6D6#1
B6D6#2
B6D6#3
B6D6#4

C$_6$D$_6$ Scintillators

Nov-Dec 2014

PULSE HEIGHT WEIGHTING TECHNIQUE:
Accurate simulations required

Geant4 model of the setup
Total Energy Detection technique

- Two conditions must be fulfilled:

  I.) Low Efficiency Detectors: \( \varepsilon_{\gamma_i} \ll 1, \forall i \)

  \[
  \varepsilon_c = 1 - \prod_{i=1}^{m_r} (1 - \varepsilon_{\gamma_i}) \approx \sum_{i=1}^{m_r} \varepsilon_{\gamma_i}
  \]

  II.) Efficiency to detect a \( \gamma \)-ray is proportional to its energy: \( \varepsilon_{\gamma_i} \propto E_{\gamma_i} \)

  \[
  \varepsilon_c = k \sum_{i=1}^{m_r} E_{\gamma_i} = k E_c
  \]

  Total efficiency depends on \( E_c \) and not on the decay path

  First idea: **Moxon-Rae** detectors:
  - Using a converter: maximum depth of escaping electrons increases with \( E_{\gamma} \) ...
  - Proportionality not really fulfilled
  - Sensitive to neutrons
TED with C$_6$D$_6$ detectors

- **C$_6$D$_6$ Detectors**: Low neutron sensitivity ($10^{-4}$ $\epsilon_\gamma$)
  - Benzene: Organic scintillator
  - H replaced by D $\rightarrow$ Avoid H(n,$\gamma$)D + 2.2MeV $\gamma$-ray

- **Fulfill condition I**: Low efficiency detectors $\epsilon_{\gamma_i} << 1$

  Detecting a cascade: $\epsilon_c = 1 - \prod (1 - \epsilon_{\gamma_i}) \approx \sum \epsilon_{\gamma_i}$

- The efficiency is **NOT proportional** to $\gamma$-ray energy:

  $$\epsilon_{\gamma_i} \propto E_{\gamma_i}$$

**The proportionality between efficiency and $\gamma$-ray energy is obtained by software manipulation of the detector response ($R_{ij}$)**

Proportionality fulfilled with Weighting factors $W_j$ dependent on the Energy deposited $E_{\gamma_j}$:

$$W_j = W(E_{\gamma_j}) : \sum_j W_j R_{ij} = E_{\gamma_i}, \quad \epsilon_i \approx \sum_j R_{ij}$$