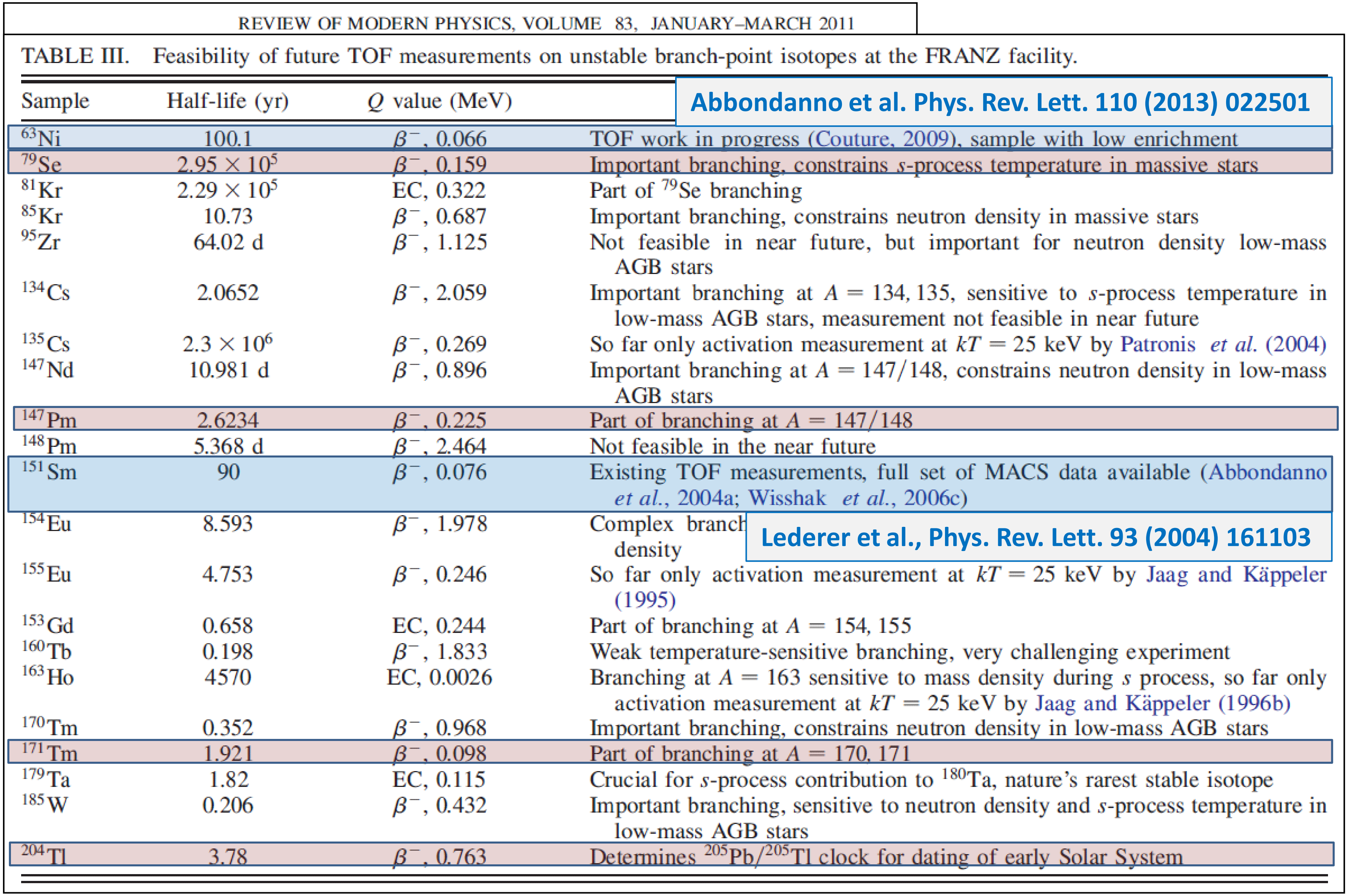
Resonance and Maxwellian cross sections of the s-process branchings ^{147}Pm, ^{171}Tm and ^{204}Tl (& ^{79}Se)

Spokespersons: C. Guerrero (US/CERN) and C. Domingo-Pardo (IFIC)
Close Collaborators: D. Schumann (PSI), R. Dresler, S. Heinitz (PSI), U. Koester (ILL)
Everyone else is welcome to join!

2013 n_TOF Annual Collaboration Meeting
Bologna, 26-29 November 2013

Motivation: branching points@nTOF and PRL



REVIEW OF MODERN PHYSICS, VOLUME 83, JANUARY–MARCH 2011

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

Abbondanno et al. Phys. Rev. Lett. 110 (2013) 022501

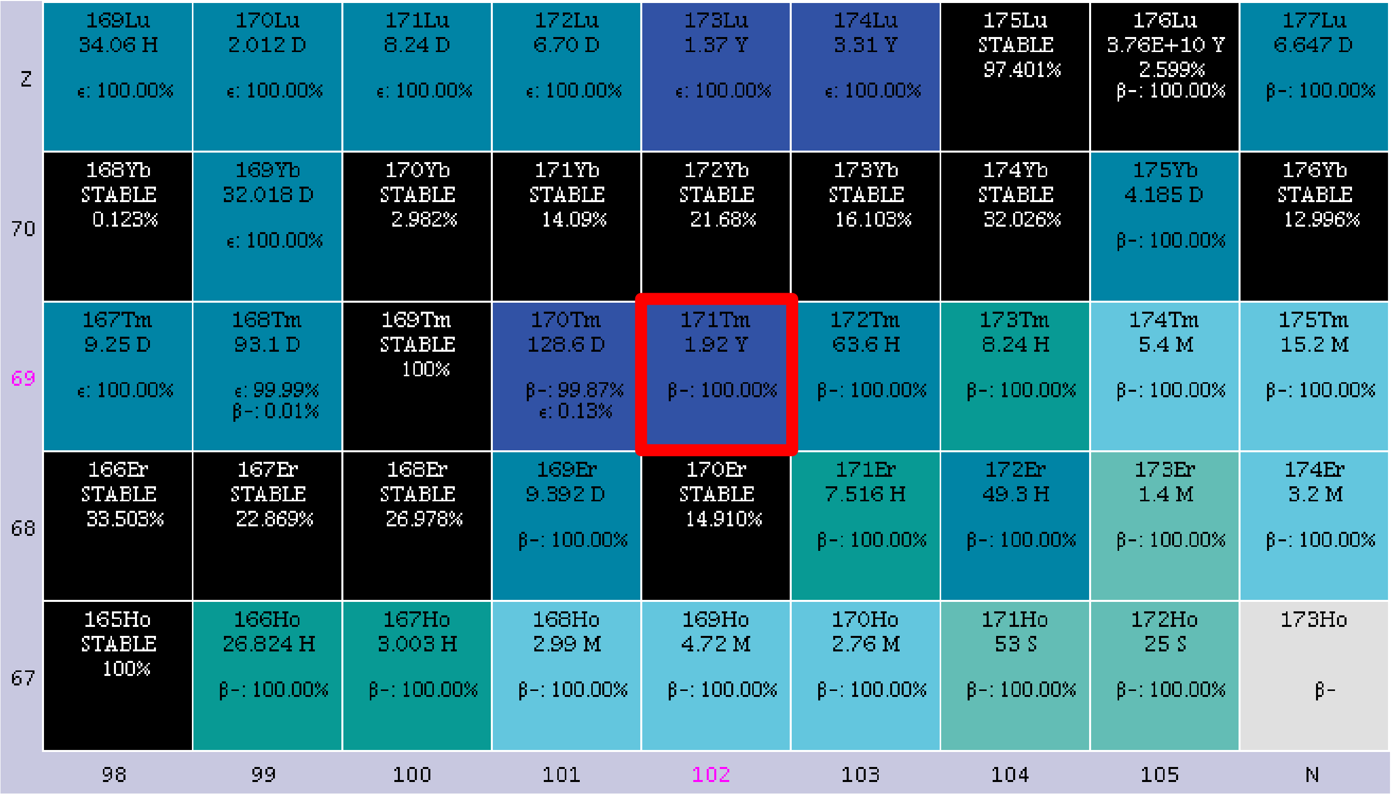
Lederer et al., Phys. Rev. Lett. 93 (2004) 161103

Sample	Half-life (yr)	Q value (MeV)	
^{63}Ni	100.1	β^-, 0.066	TOF work in progress (Couture, 2009), sample with low enrichment
^{79}Se	2.95×10^5	β^-, 0.159	Important branching, constrains *s*-process temperature in massive stars
^{81}Kr	2.29×10^5	EC, 0.322	Part of ^{79}Se branching
^{85}Kr	10.73	β^-, 0.687	Important branching, constrains neutron density in massive stars
^{95}Zr	64.02 d	β^-, 1.125	Not feasible in near future, but important for neutron density low-mass AGB stars
^{134}Cs	2.0652	β^-, 2.059	Important branching at $A = 134, 135$, sensitive to *s*-process temperature in low-mass AGB stars, measurement not feasible in near future
^{135}Cs	2.3×10^6	β^-, 0.269	So far only activation measurement at $kT = 25$ keV by Patronis *et al.* (2004)
^{147}Nd	10.981 d	β^-, 0.896	Important branching at $A = 147/148$, constrains neutron density in low-mass AGB stars
^{147}Pm	2.6234	β^-, 0.225	Part of branching at $A = 147/148$
^{148}Pm	5.368 d	β^-, 2.464	Not feasible in the near future
^{151}Sm	90	β^-, 0.076	Existing TOF measurements, full set of MACS data available (Abbondanno *et al.*, 2004a; Wisshak *et al.*, 2006c)
^{154}Eu	8.593	β^-, 1.978	Complex branch density
^{155}Eu	4.753	β^-, 0.246	So far only activation measurement at $kT = 25$ keV by Jaag and Käppeler (1995)
^{153}Gd	0.658	EC, 0.244	Part of branching at $A = 154, 155$
^{160}Tb	0.198	β^-, 1.833	Weak temperature-sensitive branching, very challenging experiment
^{163}Ho	4570	EC, 0.0026	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)
^{170}Tm	0.352	β^-, 0.968	Important branching, constrains neutron density in low-mass AGB stars
^{171}Tm	1.921	β^-, 0.098	Part of branching at $A = 170, 171$
^{179}Ta	1.82	EC, 0.115	Crucial for *s*-process contribution to ^{180}Ta, nature's rarest stable isotope
^{185}W	0.206	β^-, 0.432	Important branching, sensitive to neutron density and *s*-process temperature in low-mass AGB stars
^{204}Tl	3.78	β^-, 0.763	Determines ^{205}Pb/^{205}Tl clock for dating of early Solar System

^{171}Tm as part of branching at A=170/171

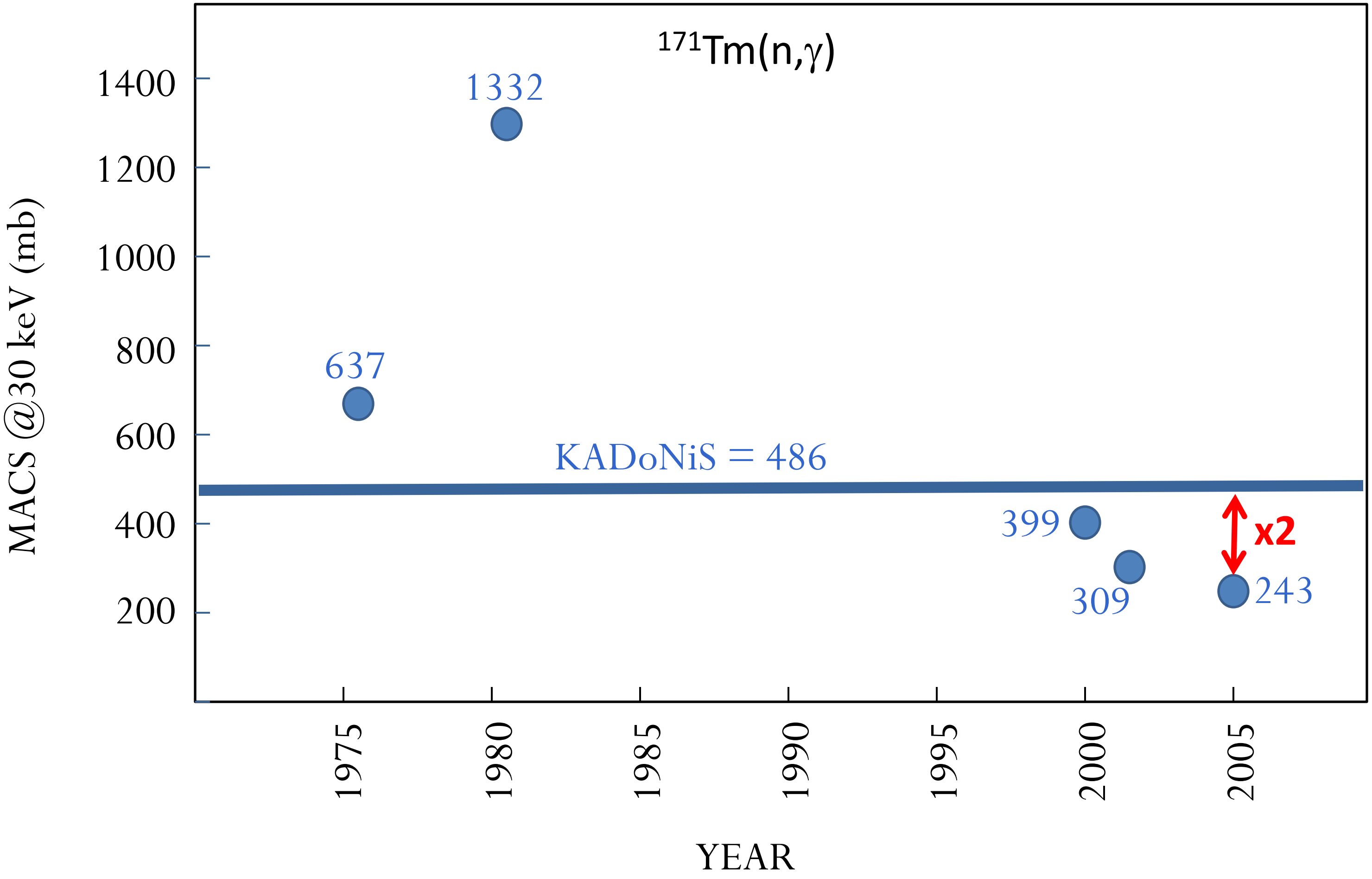
The A=170/171 branching point is one of the branchings that **is independent of stellar temperature** and therefore **suited to constrain explicitly the s-process neutron density**. The branching analysis again benefits from the fact that Tm is a rare earth element with a very well known relative abundance.

In view of the difficulties related to production of ^{170}Tm and based on the arguments outlined before, experimental information on ^{171}Tm becomes important as part of the branching, but also as the more important for improved HF predictions of the ^{170}Tm cross section.



Z	169Lu 34.06 H ϵ: 100.00%	170Lu 2.012 D ϵ: 100.00%	171Lu 8.24 D ϵ: 100.00%	172Lu 6.70 D ϵ: 100.00%	173Lu 1.37 Y ϵ: 100.00%	174Lu 3.31 Y ϵ: 100.00%	175Lu STABLE 97.401%	176Lu 3.76E+10 Y 2.599% β-: 100.00%	177Lu 6.647 D β-: 100.00%
70	168Yb STABLE 0.123%	169Yb 32.018 D ϵ: 100.00%	170Yb STABLE 2.982%	171Yb STABLE 14.09%	172Yb STABLE 21.68%	173Yb STABLE 16.103%	174Yb STABLE 32.026%	175Yb 4.185 D β-: 100.00%	176Yb STABLE 12.996%
69	167Tm 9.25 D ϵ: 100.00%	168Tm 93.1 D ϵ: 99.99% β-: 0.01%	169Tm STABLE 100%	170Tm 128.6 D β-: 99.87% ϵ: 0.13%	171Tm 1.92 Y β-: 100.00%	172Tm 63.6 H β-: 100.00%	173Tm 8.24 H β-: 100.00%	174Tm 5.4 M β-: 100.00%	175Tm 15.2 M β-: 100.00%
68	166Er STABLE 33.503%	167Er STABLE 22.869%	168Er STABLE 26.978%	169Er 9.392 D β-: 100.00%	170Er STABLE 14.910%	171Er 7.516 H β-: 100.00%	172Er 49.3 H β-: 100.00%	173Er 1.4 M β-: 100.00%	174Er 3.2 M β-: 100.00%
67	165Ho STABLE 100%	166Ho 26.824 H β-: 100.00%	167Ho 3.003 H β-: 100.00%	168Ho 2.99 M β-: 100.00%	169Ho 4.72 M β-: 100.00%	170Ho 2.76 M β-: 100.00%	171Ho 53 S β-: 100.00%	172Ho 25 S β-: 100.00%	173Ho β-
	98	99	100	101	102	103	104	105	N

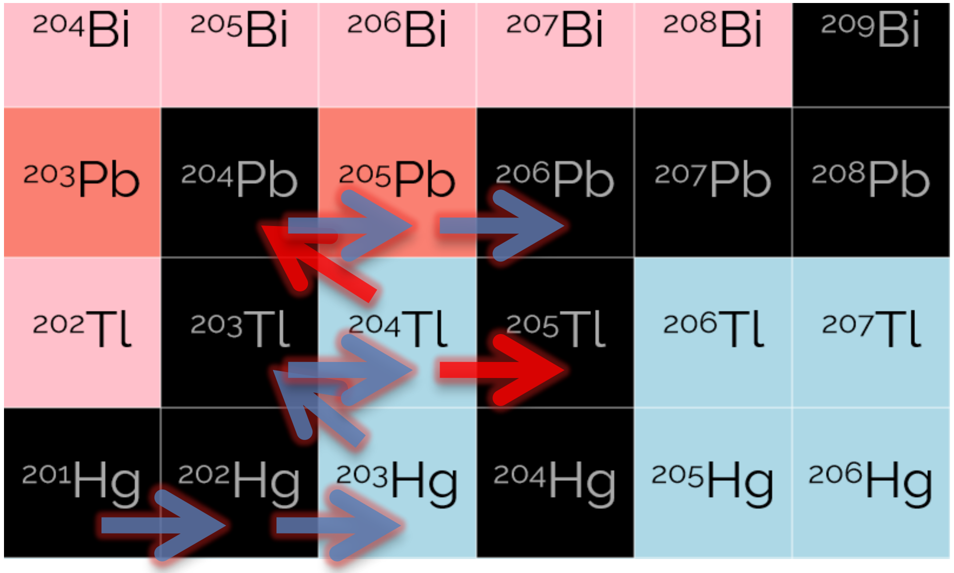
Status of ^{171}Tm(n,g) MACS calculations



^{204}Tl determines the ^{205}Pb/^{205}Tl clock for dating of early Solar System

^{205}Pb ($t_{1/2} = 1.5\times10^{7}$ a) is produced only by the s-process:
If the ^{204}Tl β-decay competes effectively versus n-capture, then the SS abundances of **^{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, upper limit for the ^{205}Pb/^{204}Pb abundance ratio of 9×10^{-5} in meteorites]



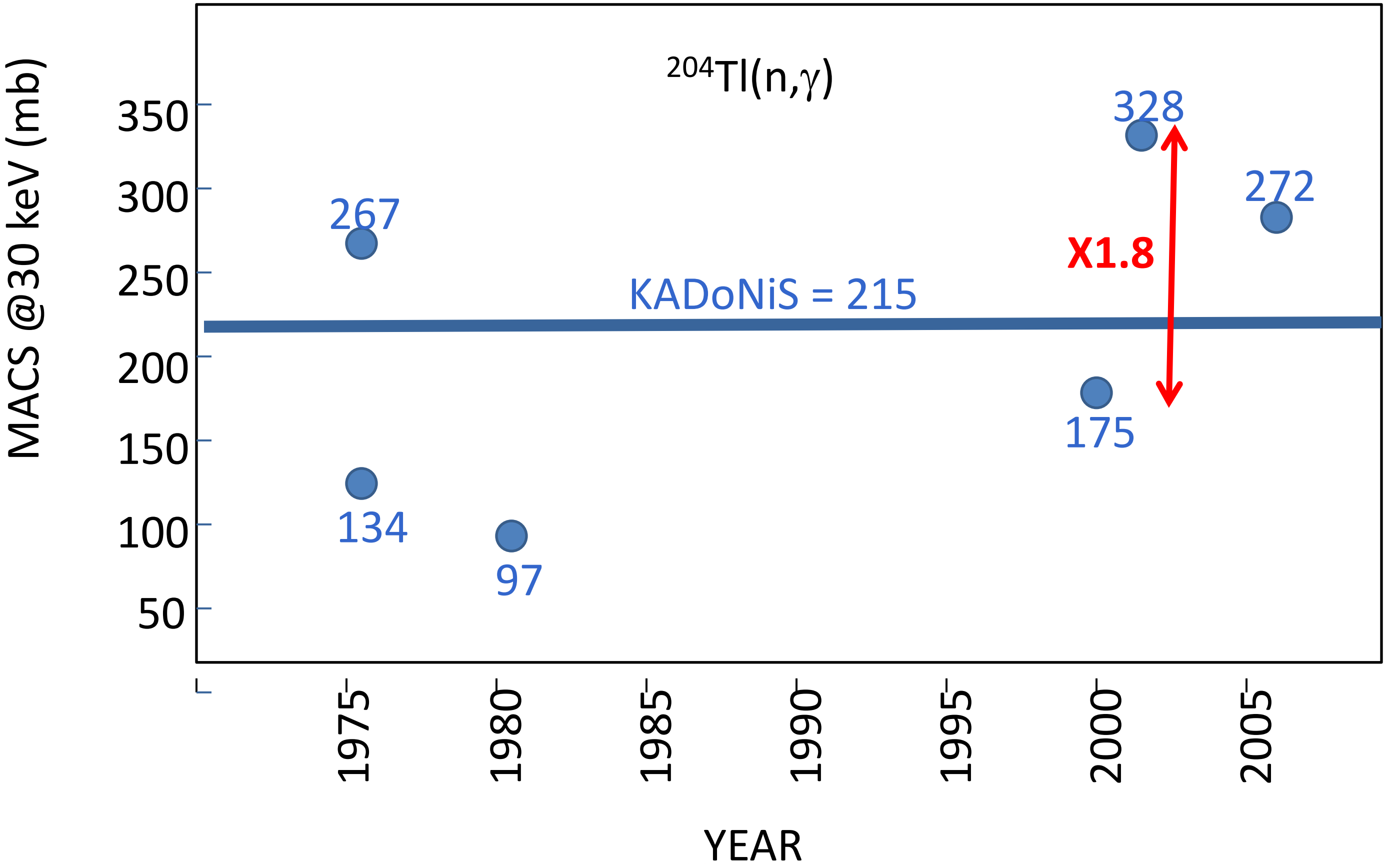
^{204}Bi	^{205}Bi	^{206}Bi	^{207}Bi	^{208}Bi	^{209}Bi
^{203}Pb	^{204}Pb	^{205}Pb	^{206}Pb	^{207}Pb	^{208}Pb
^{202}Tl	^{203}Tl	^{204}Tl	^{205}Tl	^{206}Tl	^{207}Tl
^{201}Hg	^{202}Hg	^{203}Hg	^{204}Hg	^{205}Hg	^{206}Hg

Blake et al., Nature, 1973
Blake and Schramm, ApJ, 1975
Macklin &Winters, ApJ, 1976

Status of ^{204}Tl(n,γ) MACS calculations

… such astrophysical studies require a reliable MACS for ^{204}Tl(n,g), however:

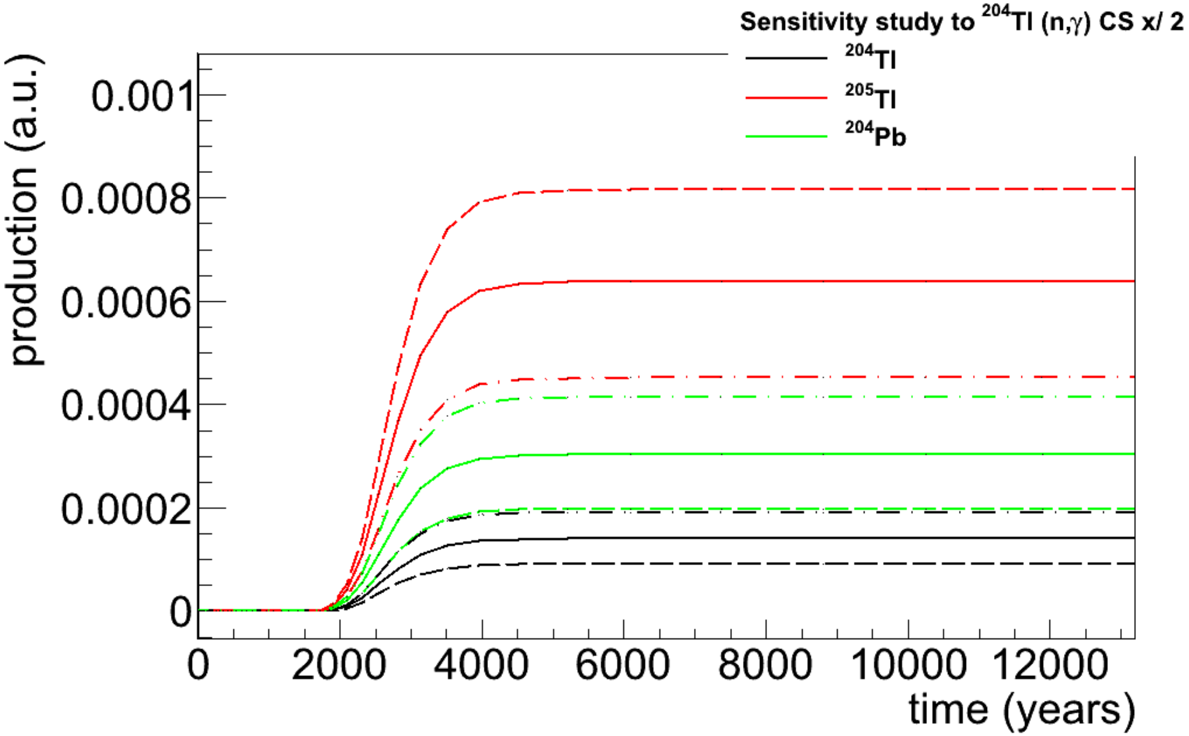
- **No experimental information available at all.**
- **Present MACS based on theoretical HF-calculations.**



A factor of ~2 uncertainty in the ^{204}Tl(n,γ) MACS is currently hindering a reliable and detailed analysis of the s-branching at ^{204}Tl

^{204}Tl determines the ^{205}Pb/^{205}Tl clock for dating of early Solar System

Indeed, a factor of 2 uncertainty only in the ^{204}Tl(n,g) CS makes a very large impact in the production of the two branching products ^{205}Tl and ^{204}Pb

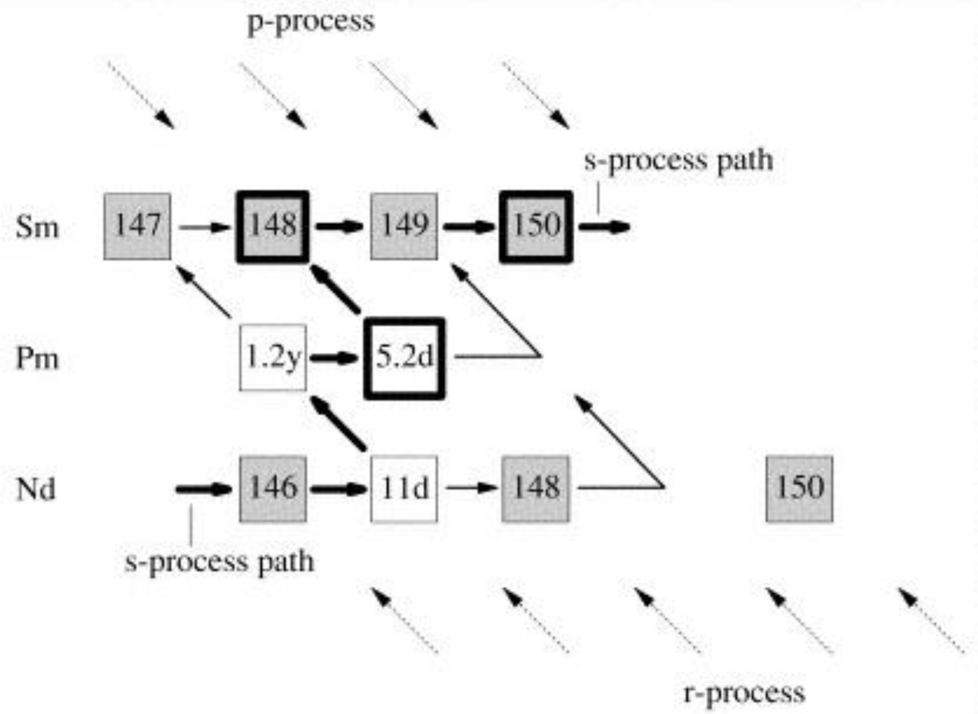


- Sensitivity study based on a simple classical s-process approach (r = 1x10^3 g/cm3, 10^8 n/cm^3, T_9 = 0.2 K). Network input uses Kadonis02 MACS, nuc-net code from B. Meyer (Clemson Univ.).
- Assuming a factor of 2 uncertainty in the CS of ^{204}Tl, impacts almost by a factor of 2 in the production of the branching nuclei ^{205}Tl and ^{204}Pb.

^{147}Pm branching and the s-process neutron density

The Nd-Pm-Sm branchings sketched are of particular interest for constraining the s-process neutron density.

- Their overall strength is well defined by the two s-only isotopes, ^{148}Sm and ^{150}Sm, both shielded against β-decays from the r-process region.
- the relative abundances of these rare earth elements are known <2%.
- The abundance ratio of ^{148}Sm and ^{150}Sm is affected by three branchings of the neutron capture chain: ^{147}Nd, ^{147}Pm, and ^{148}Pm



Samples production in three steps: step 1

Branching points are radioactive!

Production via (n,γ) or (n, γ)β^- in the ILL research reactor [Contact: Ulli Koester]

Neutron flux: $1.5\text{x}10^{15}$ n/cm^2/s (highest $F_{n,th}$ worldwide)

Irradiation time: 60 days (1.3 cycles) [March –June 2012]

Step 1: purchase of the stable isotopes and production of pellets to be irradiated.

Samples transformed into pellets at PSI
[Contact: J. Neuhausen, D. Schumann]

- Pressed 5 mm diameter pellets
- Sintered at 1150°C

Successfully completed in Spring 2013!

Samples production in three steps: step 2

Branching points are radioactive!

Production via (n,γ) or (n, γ)β^- in the ILL research reactor [Contact: Ulli Koester]

Neutron flux: 1.5x10^{15} n/cm^2/s (highest $F_{n,th}$ worldwide)

Irradiation time: 60 days (1.3 cycles) [March –June 2012]

Step 2: irradiation during 60 days at ILL

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^{147}Pm: ^{146}Nd(n,γ)^{147}Nd (β^-, 10d)^{147}Pm (enrichment 0.35%)

0.29 mg of ^{147}Pm (2.6 y) [1.2e18 atoms]

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

3.63 mg of ^{171}Tm (1.9 y) [1.3e19 atoms]

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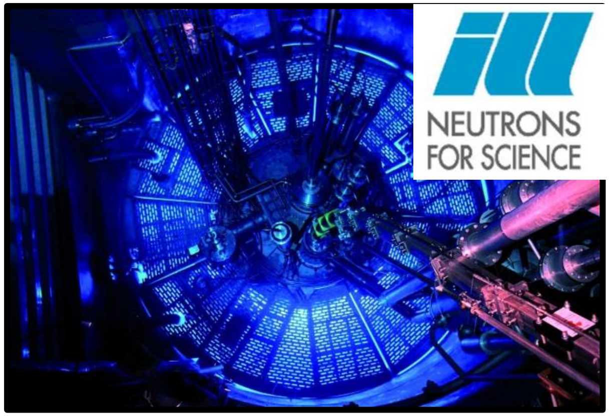
^{204}Tm: ^{203}Tl(n,γ)^{204}Tl (enrichment 5.3%)

11 mg of ^{171}Tm (3.78 y) [3.25e19 atoms]



A new source of radioactive samples for n_TOF

Successfully completed in Summer 2013!

Samples production in three steps: step 3 and 4

Branching points are radioactive!

Production via (n,γ) or (n, γ)β^- in the ILL research reactor [Contact: Ulli Koester]

Neutron flux: 1.5×10^{15} n/cm^2/s (highest $F_{n,th}$ worldwide)

Irradiation time: 60 days (1.3 cycles) [March –June 2012]

Step 3: chemical purification at PSI

We have now:

^{147}Pm: 0.35% in a ^{146}Nd pellet of 5 mm in diameter

^{171}Tm: 1.80% in a ^{170}Er pellet of 5 mm in diameter

Since October 2013 Stephan Heinitz (postdoc at PSI) is preparing the chemical purification.

Step 4: shaping of the samples for use at n_TOF

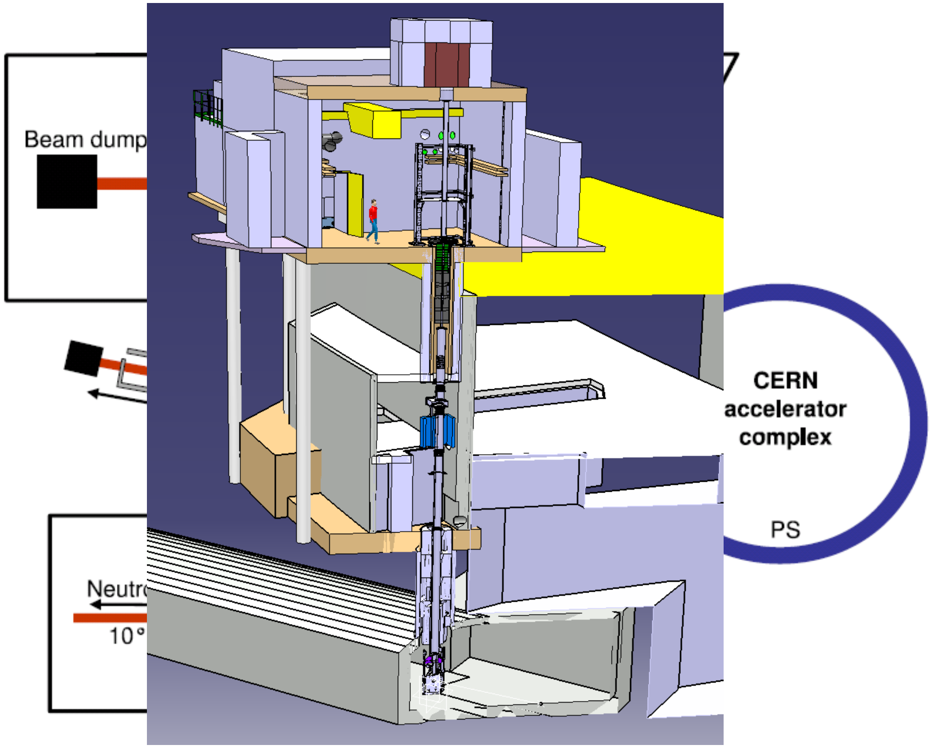
Better/possible option under discussion:

droplets of Pm/Tm$(NO_3)_3$

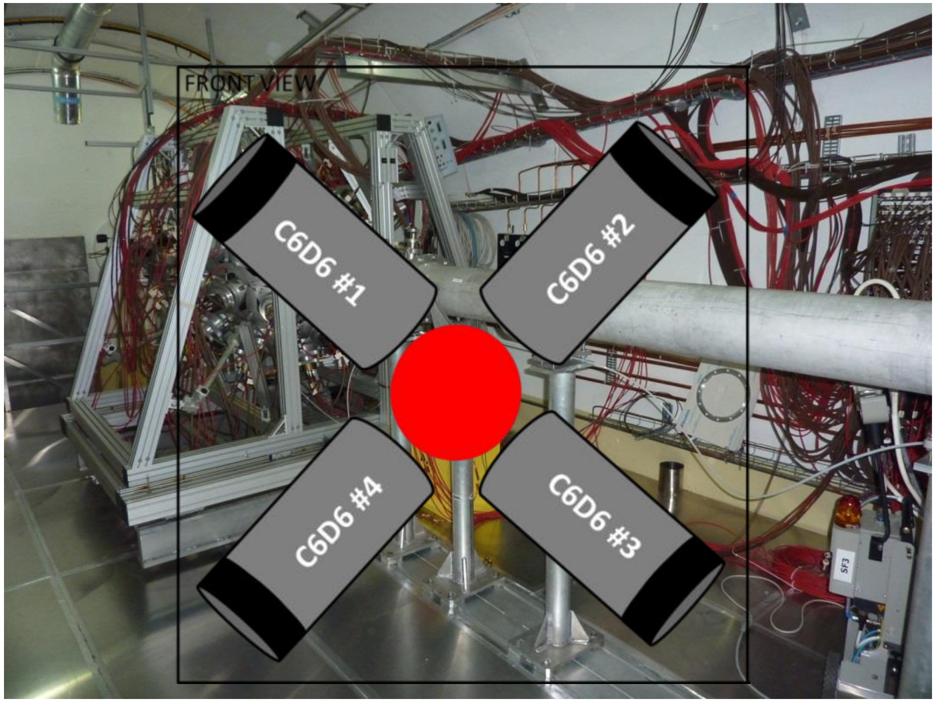
solution into BeO oxide matrix,

…

Experimental set-up



Mastinu et al., CERN-n_TOF-PUB-2013-002, *“New*



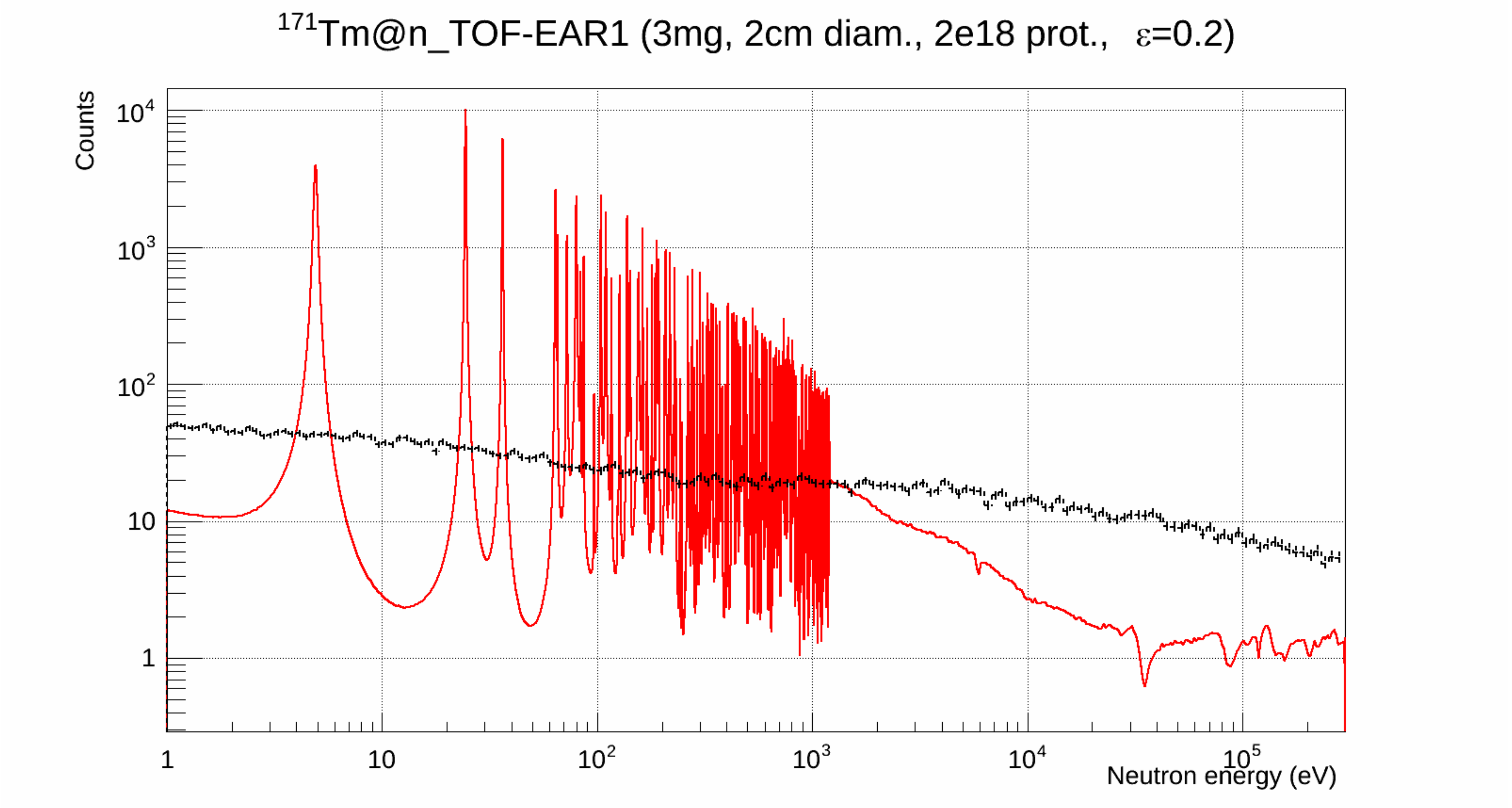
C6D6 vs. BaF$_2$:

- BaF2: more affected by g-flash and neutron sensitivity (high CRate won’t be a problem)
- C6D6: lower efficiency and less background discrimination

The choice will depend on the success of the gated PMT and the background reduction in EAR-1

Beam Time request: ^{171}Tm(n,γ)

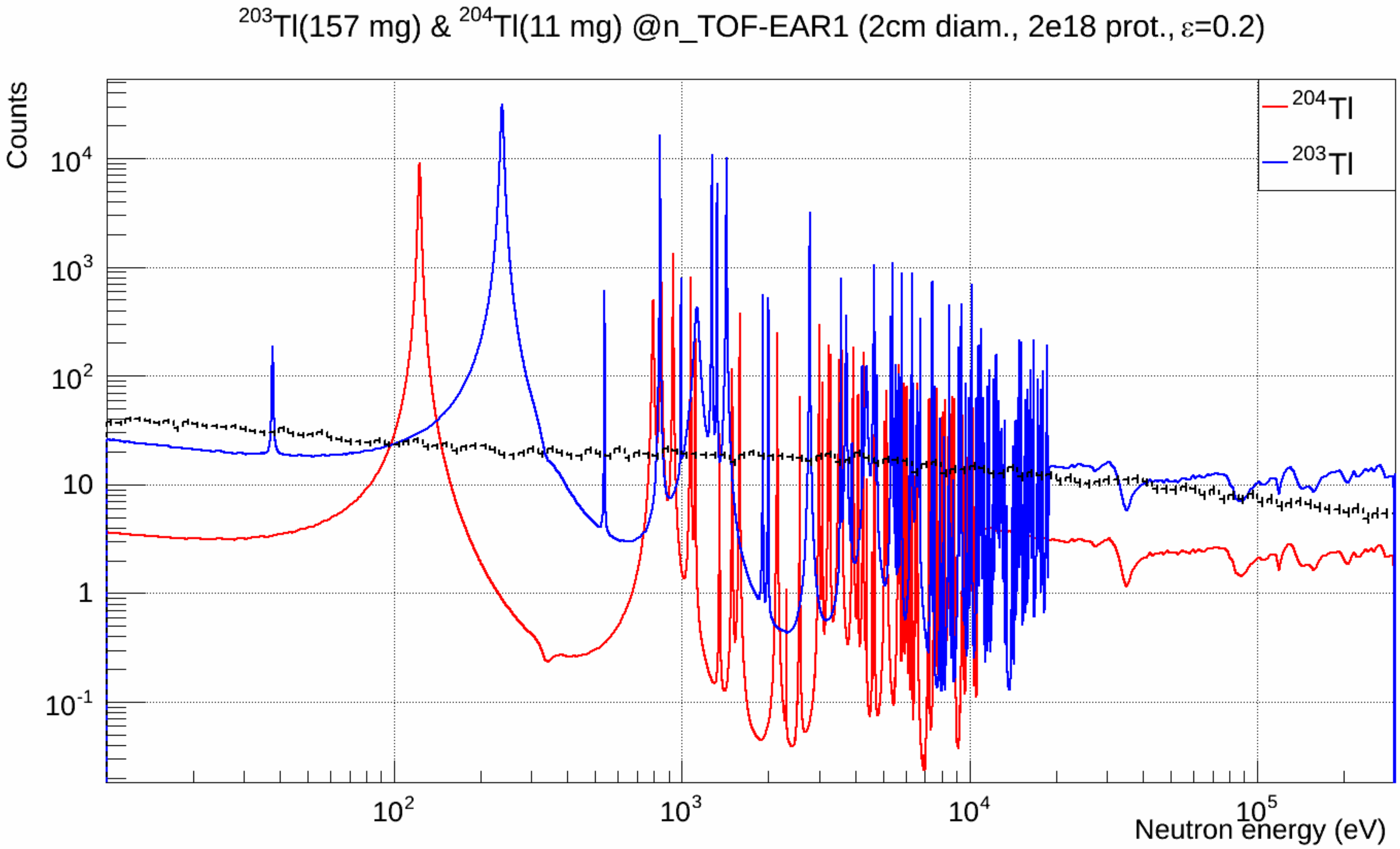
Beam time request based on the only evaluation available: TENDL-2012 (TALYS)



Background scaled by 0.33 (gain after PHWF)

Beam Time request: ^{204}Tl(n,γ)

Beam time request based on the only evaluation available: TENDL-2012 (TALYS)


^{203}Tl(157 mg) & ^{204}Tl(11 mg) @n_TOF-EAR1 (2cm diam., 2e18 prot., ε=0.2)

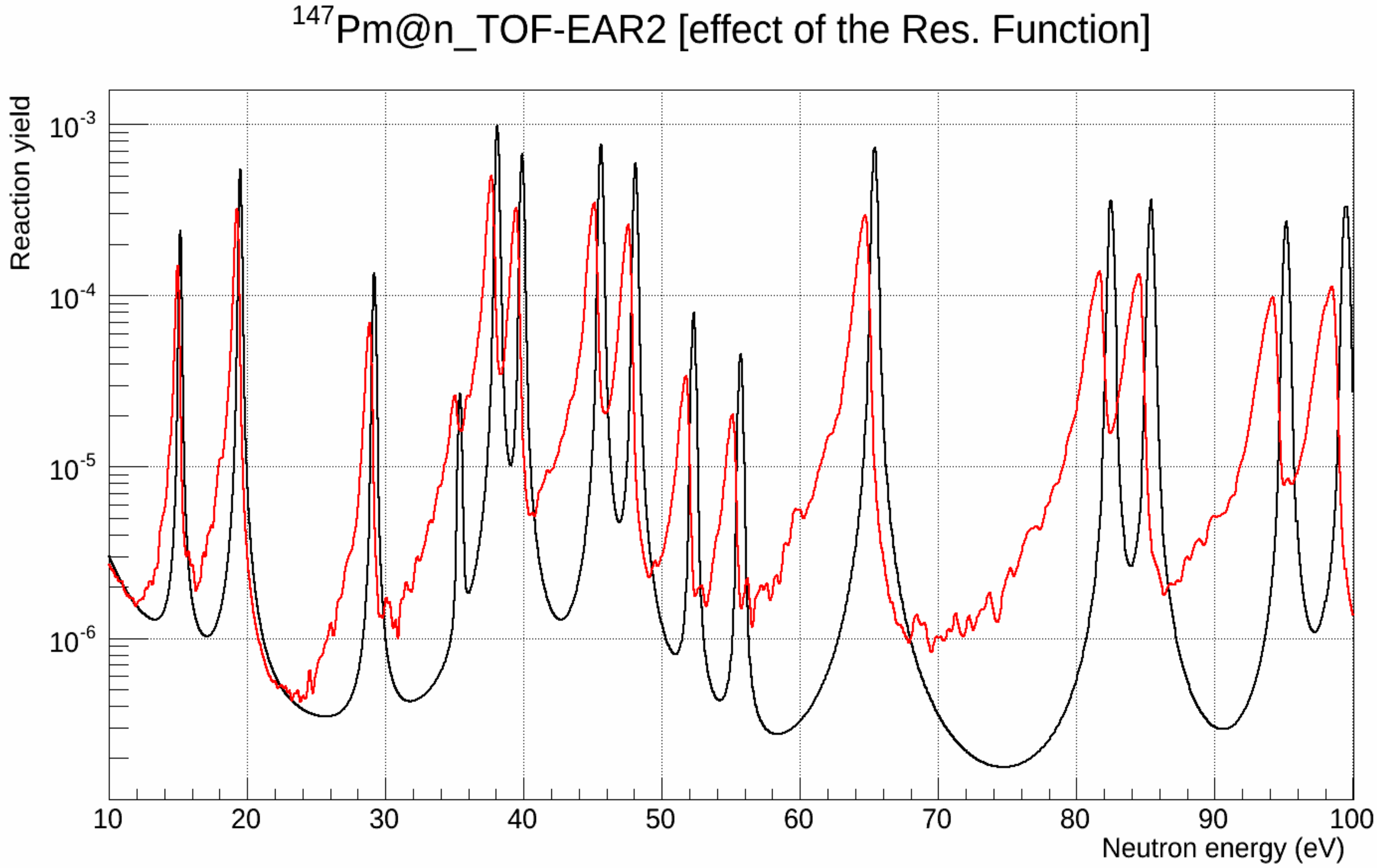
$S_n(^{203+1}\mathrm{Tl})$=6.7 MeV

$S_n(^{204+1}\mathrm{Tl})$=7.5 MeV

Background scaled by 0.33 (gain after PHWF)

Beam Time request: ^{147}Pm(n,γ)

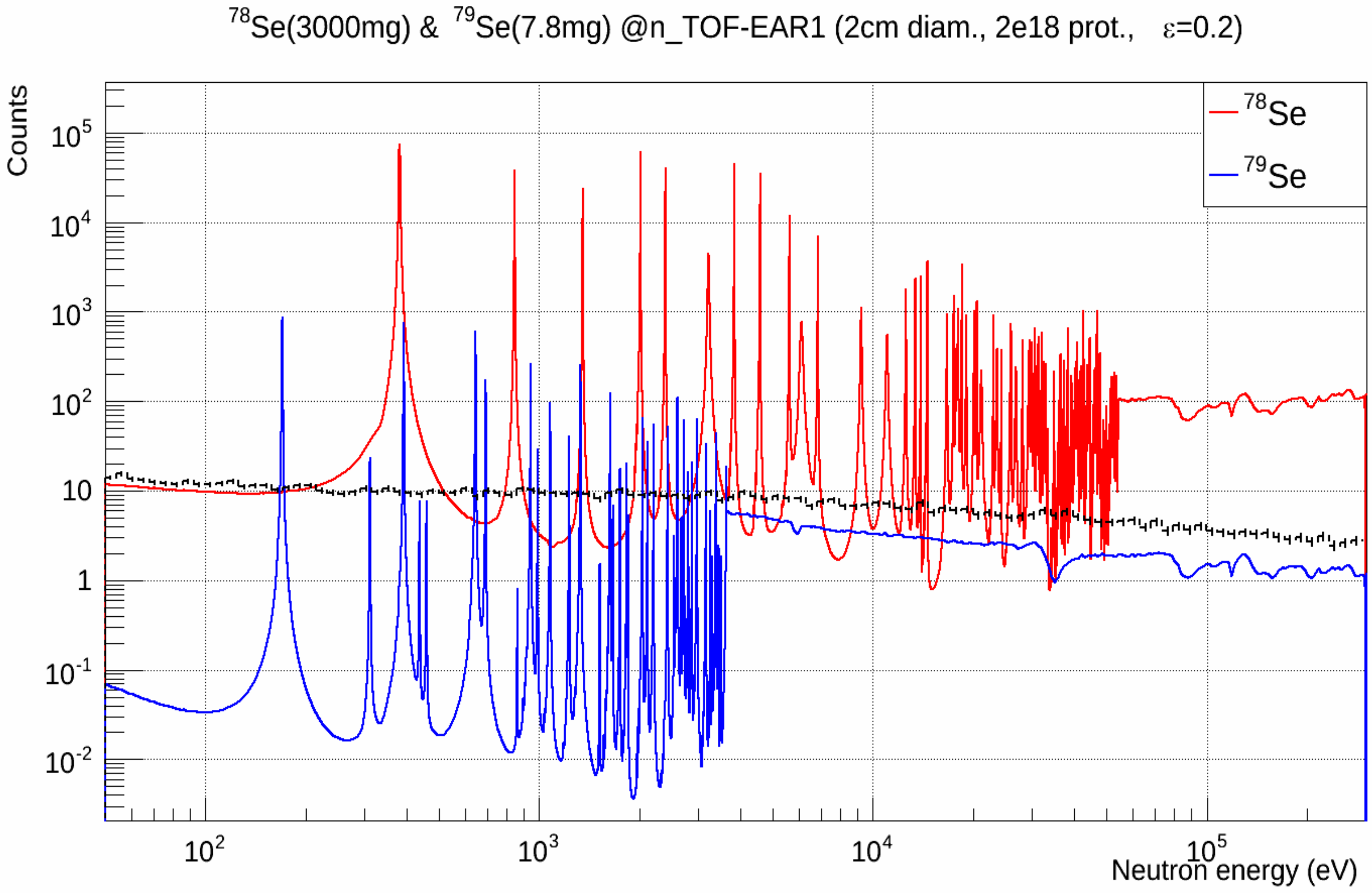
Beam time request based on the only evaluation available: TENDL-2012 (TALYS)



Background scaled by 0.33 (gain after PHWF)

Beam Time request: ^{79}Se(n,γ)

Beam time request based on the only evaluation available: TENDL-2012 (TALYS)



$S_n(^{78+1}Se)$=7.0 MeV

$S_n(^{79+1}Se)$=9.9 MeV

Background scaled by 0.33 (gain after PHWF)

Objectives of the measurements

Measure the corresponding capture cross sections by ToF for the first time ever:

At least:

Observe the resonances in the eV region

Determine an accurate set of average resonance parameters: D_0, S_0 and $<\Gamma_\gamma>$

At best (only if background in the keV significantly reduced)**:**

Measure the URR (1-300 keV) with 20 bins/decade with σ_{stat}<10%

<u>**As a result we shall provide the first MACS ever based on measurements for all nuclei.**</u>

Summary of choices

Measure the corresponding capture cross sections by ToF for the first time ever:

At least:

- Observe the resonances in the eV region
- Determine an accurate set of average resonance parameters: D_0, S_0 and $<\Gamma_\gamma>$
- MACS from average resonance parameters

At best (only if background in the keV reduced)**:**

- Measure the URR (1-300 keV) with 20 bins/decade with σ_{stat}<10%

	Detection Set-Up		Experimental Area		Choice
	L6D6	TAC	EAR-1	EAR-2	
^{171}Tm (3 mg)	X	X	X	X	L6D6@EAR-1
^{204}Tl (11mg)	-	X	X	X	TAC@EAR-1
^{147}Pm (0.3 mg)	X	X	-	X	L6D6@EAR-2
^{79}Se (7.8 mg)	-	X	-	X	TAC@EAR-?

Summary of beam request

Sample	Set-Up	Goal	Protons
^{171}Tm (3mg)	L6D6@EAR-1	RRR up to 1keV, URR only if background reduced	2x10^{18}
^{204}Tl (11mg)	TAC@EAR-1	RRR up to 5 keV (too much ^{203}Tl for URR)	3x10^{18}
^{203}Tl(157mg)	TAC@EAR-1	Background for ^{204}Tl measurement	1x10^{18}
^{147}Pm (0.3mg)	C6D6@EAR-2	RRR & URR (depends on background at EAR-2)	1x10^{18}
Dummy		Background	6x10^{17}
^{nat}Pb		Background	3x10^{17}
^{197}Au		Validation	6x10^{17}

^{171}Tm and ^{204}Tl @ EAR-1 → 7.5x10^{18} protons
^{147}Pm @ EAR-2 → 2.5x10^{18} protons (nice to test EAR-2!)

They shall be performed in 2014 because of short half-lives