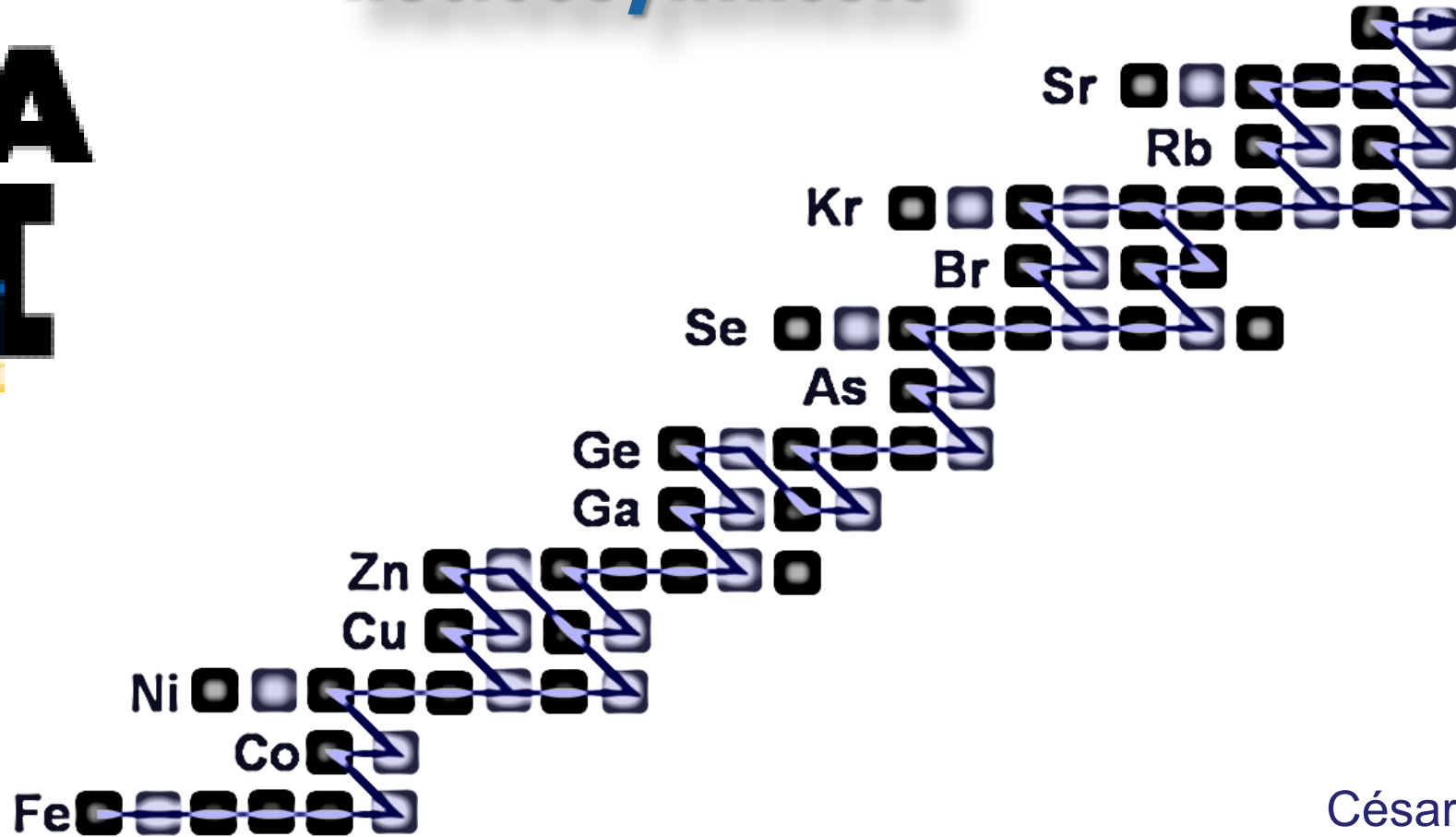


Neutron capture measurements for s-process nucleosynthesis



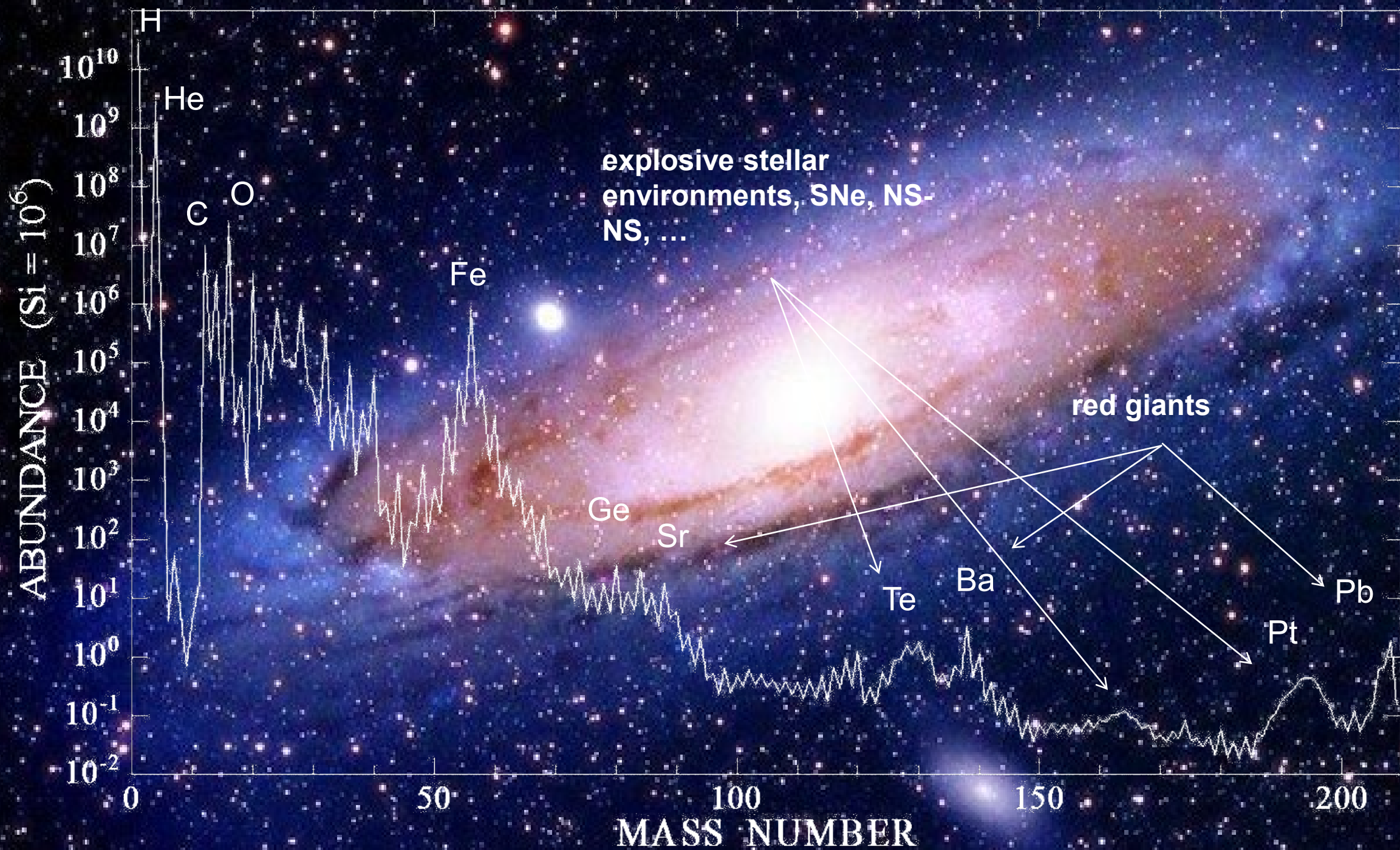
César Domingo Pardo

Nuclear Physics in Astrophysics XI, Dresden, 15-20 Sept. 2024



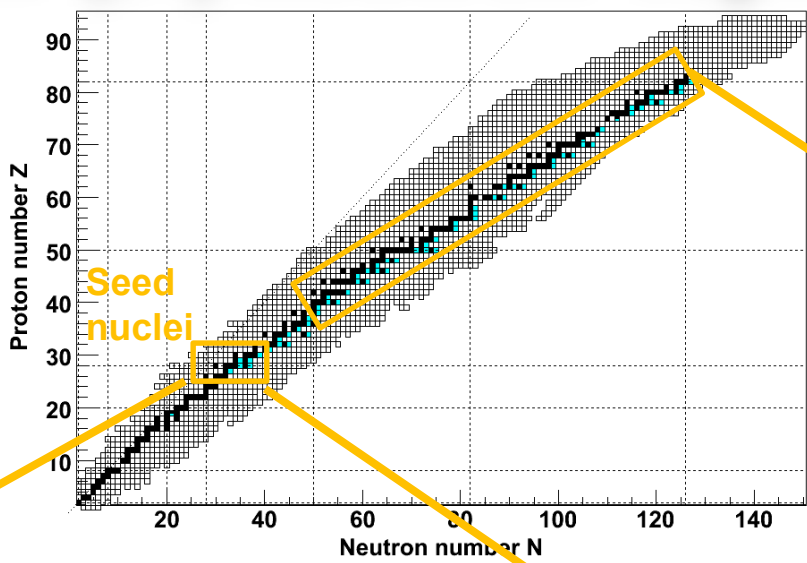
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The slow (s-) neutron-capture process

$$\tau_{n,\gamma} = \frac{1}{N_n \langle \sigma_{n,\gamma} v \rangle_{kT}}$$

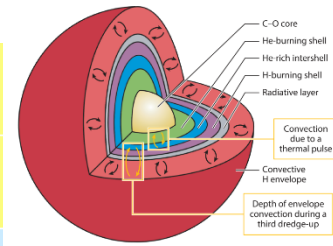


H-burning
 ~90 MK
 $kT=8 \text{ keV}$
 10^7 cm^{-3}
 $^{13}\text{C}(^4\text{He},n)^{16}\text{O}$

He-burning (TP)
 ~250 MK
 $kT=23 \text{ keV}$
 10^{11} cm^{-3}
 $^{22}\text{Ne}(^4\text{He},n)^{25}\text{Mg}$

$1 M_{\odot} < M < 3 M_{\odot}$
 Zr - Bi
 Main

^{205}Po 1.74 h	^{206}Po 8.80 d	^{207}Po 5.80 h	^{208}Po 2.90 a	^{209}Po 102.01 a	^{210}Po 138.43 d	^{211}Po 5.00 ms	^{212}Po 45.01 s
^{204}Bi 11.22 h	^{205}Bi 15.31 d	^{206}Bi 6.24 d	^{207}Bi 32.90 a	^{208}Bi 367.91 ka	^{209}Bi 100%	^{210}Bi 5.01 d	^{211}Bi 2.14 m
^{203}Pb 2.16 d	^{204}Pb 1.4%	^{205}Pb 1.80 Ma	^{206}Pb 24.1%	^{207}Pb 22.1%	^{208}Pb 52.4%	^{209}Pb 3.25 h	^{210}Pb 22.20 a
^{202}Tl 12.23 d	^{203}Tl 29.524%	^{204}Tl 3.78 a	^{205}Tl 7.476%	^{206}Tl 4.20 m	^{207}Tl 4.77 m	^{208}Tl 3.05 m	^{209}Tl 2.16 m
^{201}Hg 13.18%	^{202}Hg 29.86%	^{203}Hg 46.60 d	^{204}Hg 6.87%	^{205}Hg 5.14 m	^{206}Hg 8.15 m	^{207}Hg 2.90 m	^{208}Hg 41.00 m



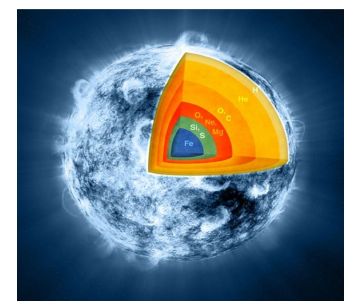
Lugaro M. et al.
 Annu. Rev. Nuc. Part. Sci. (2023)

^{60}Zn 2.38 m	^{61}Zn 1.48 m	^{62}Zn 9.19 h	^{63}Zn 38.47 m	^{64}Zn 48.63%	^{65}Zn 243.63 d	^{66}Zn 27.9%	^{67}Zn 4.1%	^{68}Zn 18.75%
^{59}Cu 1.36 m	^{60}Cu 23.70 m	^{61}Cu 3.33 h	^{62}Cu 9.67 m	^{63}Cu 69.17%	^{64}Cu 12.70 h	^{65}Cu 30.83%	^{66}Cu 5.12 m	^{67}Cu 2.58 d
^{58}Ni 68.077%	^{59}Ni 75.99 ka	^{60}Ni 26.223%	^{61}Ni 1.14%	^{62}Ni 3.634%	^{63}Ni 100.11 a	^{64}Ni 0.926%	^{65}Ni 2.52 h	^{66}Ni 2.27 d
^{57}Co 271.76 d	^{58}Co 70.86 d	^{59}Co 100%	^{60}Co 5.27 a	^{61}Co 1.65 h	^{62}Co 1.50 m	^{63}Co 27.40 s	^{64}Co 300.00 ms	^{65}Co 1.20 s
^{56}Fe 91.754%	^{57}Fe 2.119%	^{58}Fe 0.282%	^{59}Fe 44.50 d	^{60}Fe 1.50 Ma	^{61}Fe 5.98 m	^{62}Fe 1.13 m	^{63}Fe 6.01 s	^{64}Fe 2.00 s

$M > 8 M_{\odot}$ Weak Fe - Zr

core He-burning
 300 MK
 $kT=26 \text{ keV}$
 10^6 cm^{-3}
 $^{22}\text{Ne}(^4\text{He},n)^{25}\text{Mg}$

shell C-burning
 ~ GK
 $kT=91 \text{ keV}$
 10^{11} cm^{-3}
 $^{12}\text{C}(^{12}\text{C},n)^{23}\text{Mg}$



The very first s-process neutron-capture experiments

REVIEWS OF

MODERN PHYSICS

VOLUME 29, NUMBER 4

OCTOBER, 1957

Synthesis of the Elements in Stars*

E. MARGARET BURBRIDGE, G. R. BURBRIDGE, WILLIAM A. FOWLER, and F. HOYLE

Kellogg Radiation Laboratory, California Institute of Technology, and Mount Wilson and Palomar Observatories, Carnegie Institution of Washington, California Institute of Technology, Pasadena, California

"It is the stars, The stars above us, govern our conditions";
(King Lear, Act IV, Scene 3)

but perhaps

"The fault, dear Brutus, is not in our stars, But in ourselves,"
(Julius Caesar, Act I, Scene 2)

If a sufficient number of neutrons is available, the last of the isotopes in the s chain will achieve the full value of $n(A)$ given by this equation and we shall have

$$n(A)\sigma(A) = \text{constant}$$

1272

NATURE

June 30, 1962 No. 4835

Neutron Capture in Tin Isotopes at Stellar Temperatures

THEORIES of stellar nucleosynthesis have emphasized the importance of neutron capture (s- and r-processes occurring in giant stars and supernovae respectively) in producing the heavy elements^{1,2}. For the s-process, cosmic isotopic abundances are predicted to show an inverse proportionality to the neutron capture cross-sections. While some nuclei can be produced by only one process or the other, the abundances can be most accurately measured for isotopes of a single element. Thus the many isotopes of tin provide the best opportunity to test the theories and to determine such parameters as the relative contribution of supernovae to the material of the solar

paring the ratio N_r/N_s (Table 1, col. 7) with the latest published estimates², which are shown in col. 8.

R. L. MACKLIN
T. INADA*
J. H. GIBBONS

Oak Ridge National Laboratory,
Oak Ridge, Tennessee.

* Visiting scientist from National Institute of Radiological Sciences, Chiba, Japan.

¹ Burbridge, E. M., Burbridge, G. R., Fowler, W. A., and Hoyle, F., *Rev. Mod. Phys.*, **28**, 547 (1957).

² Clayton, D. D., and Fowler, W. A., *Ann. Phys.*, **18**, 51 (1961).

³ Gibbons, J. H., Macklin, R. L., Miller, P. D., and Neiler, J. H., *Phys. Rev.*, **122**, 182 (1961).

⁴ Moxon, M. C., and Rae, E. R., in *Neutron Time-of-Flight Methods*, edit. by Spaepen, J. (Euratom, Brussels, 1961).

(1)	(2)	(3)	(5)
Nucleus	σ_s (mb.)	Isotopic abundance N^*	$N_s\sigma_s = (N - N_r)\sigma_s$
Sn 116	$92 \pm 19\%$	0.1424	13.1
117	$390 \pm 82\%$	0.0757	13.9
118	$59 \pm 12\%$	0.2401	11.5
119	$243 \pm 51\%$	0.0858	11.1
120	$35 \pm 7\%$	0.3297	10.0
122	23 ± 5	0.0471	—
124	23 ± 4	0.0598	—

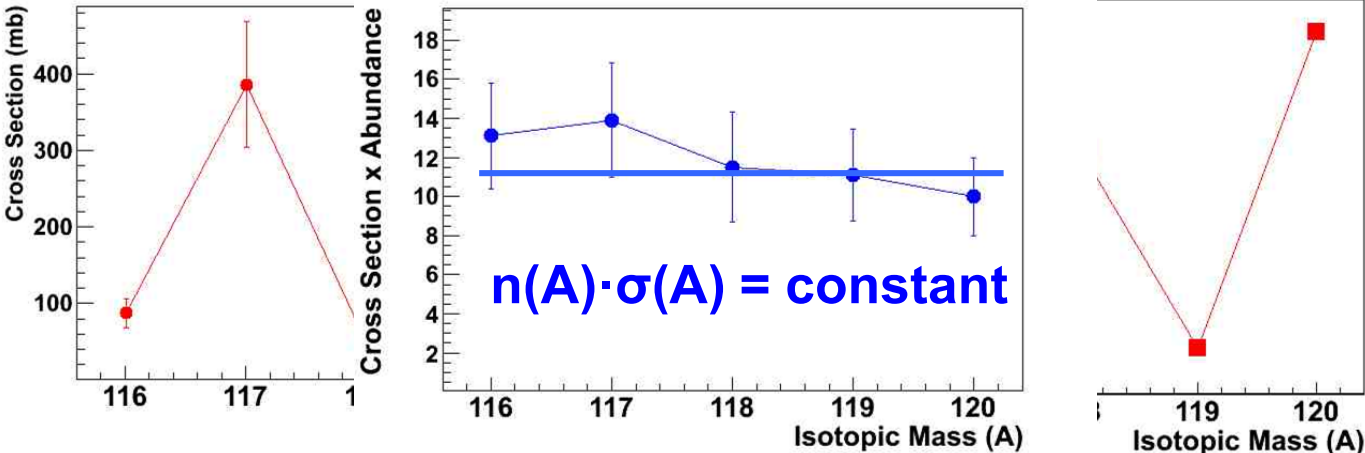
→ First quantitative test of s-process theory!

Scintillator Tank

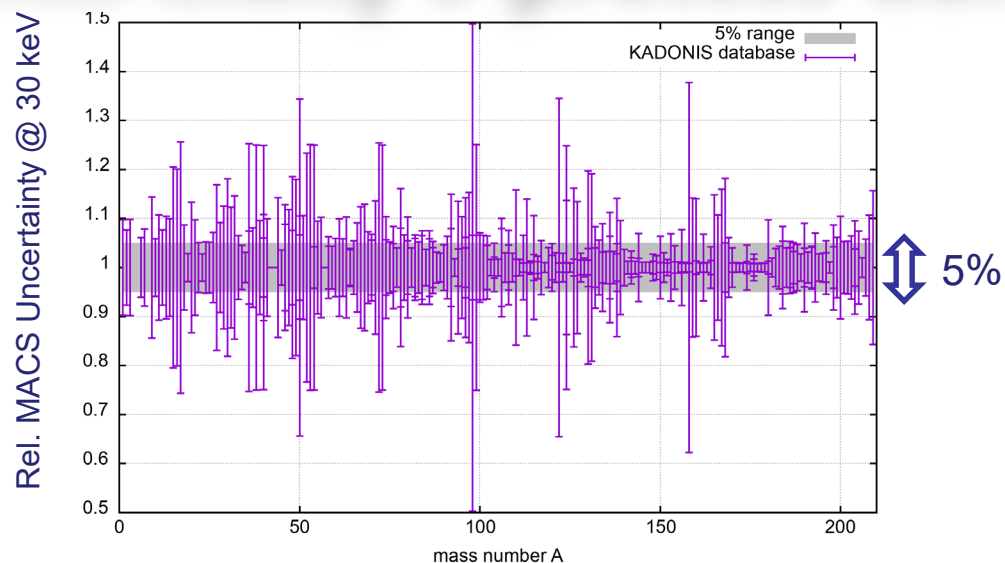
1 OoM!

Moxon-Rae Detector

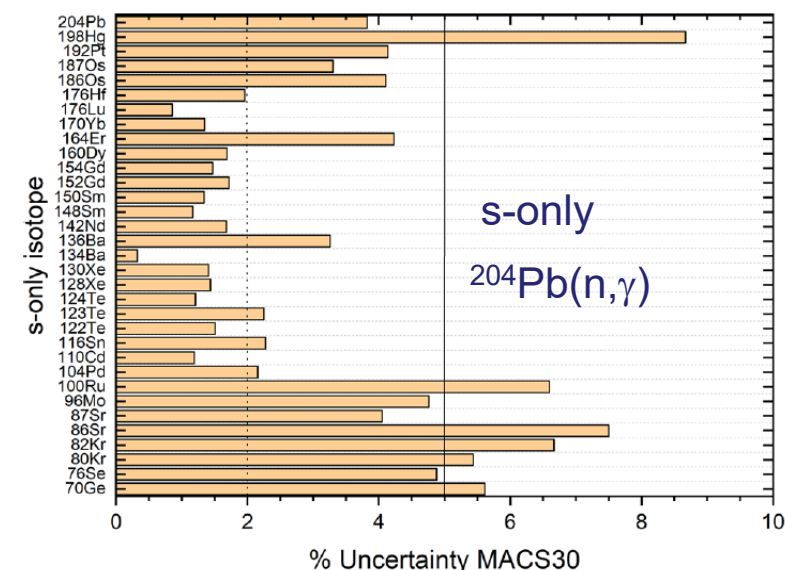
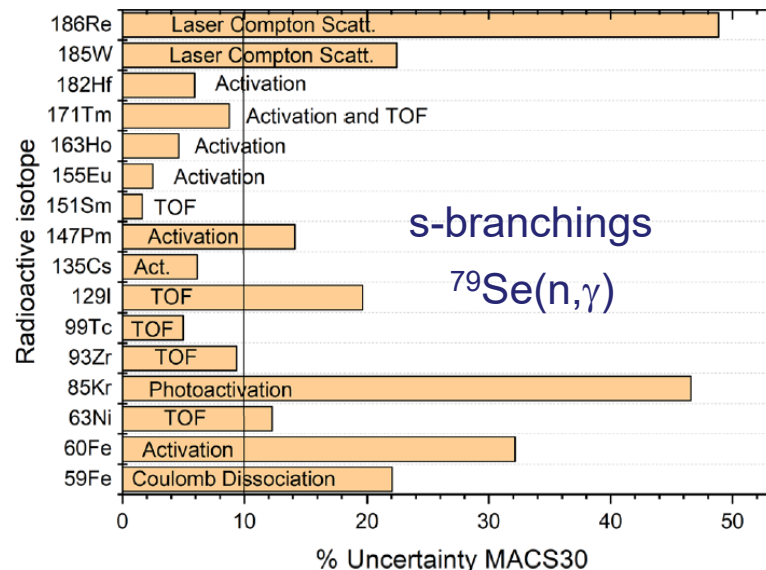
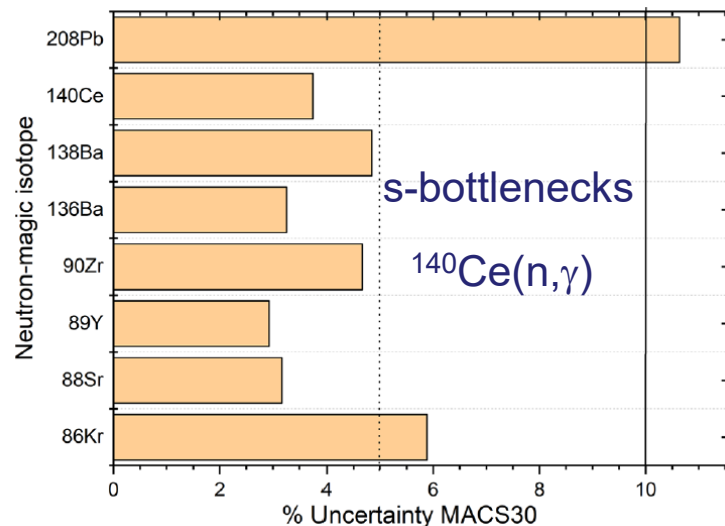
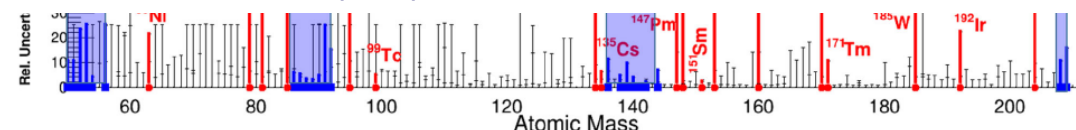
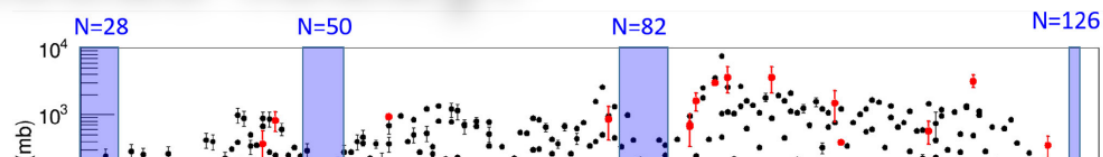
$$\epsilon_\gamma = k \epsilon_y$$



Still many s-process data needs today!



- Today: Situation is far from satisfactory!
- Re-measuring and improving (!) neutron-capture CSs on many stable isotopes is still very important!
- See e.g. M. Spelta (UT-INFN), Poster#248 on $^{30}\text{Si}(n,\gamma)$, Poster#249 on $^{64}\text{Ni}(n,\gamma)$, Talk#79 R. Mucciola



I. Dillmann et al. Eur. Phys. Jour. A (2023)
 F. Käppeler et al. Rev. Mod. Phys. (2011)
 C. Domingo-Pardo et al. Eur. Phys. Jour. A (2023)

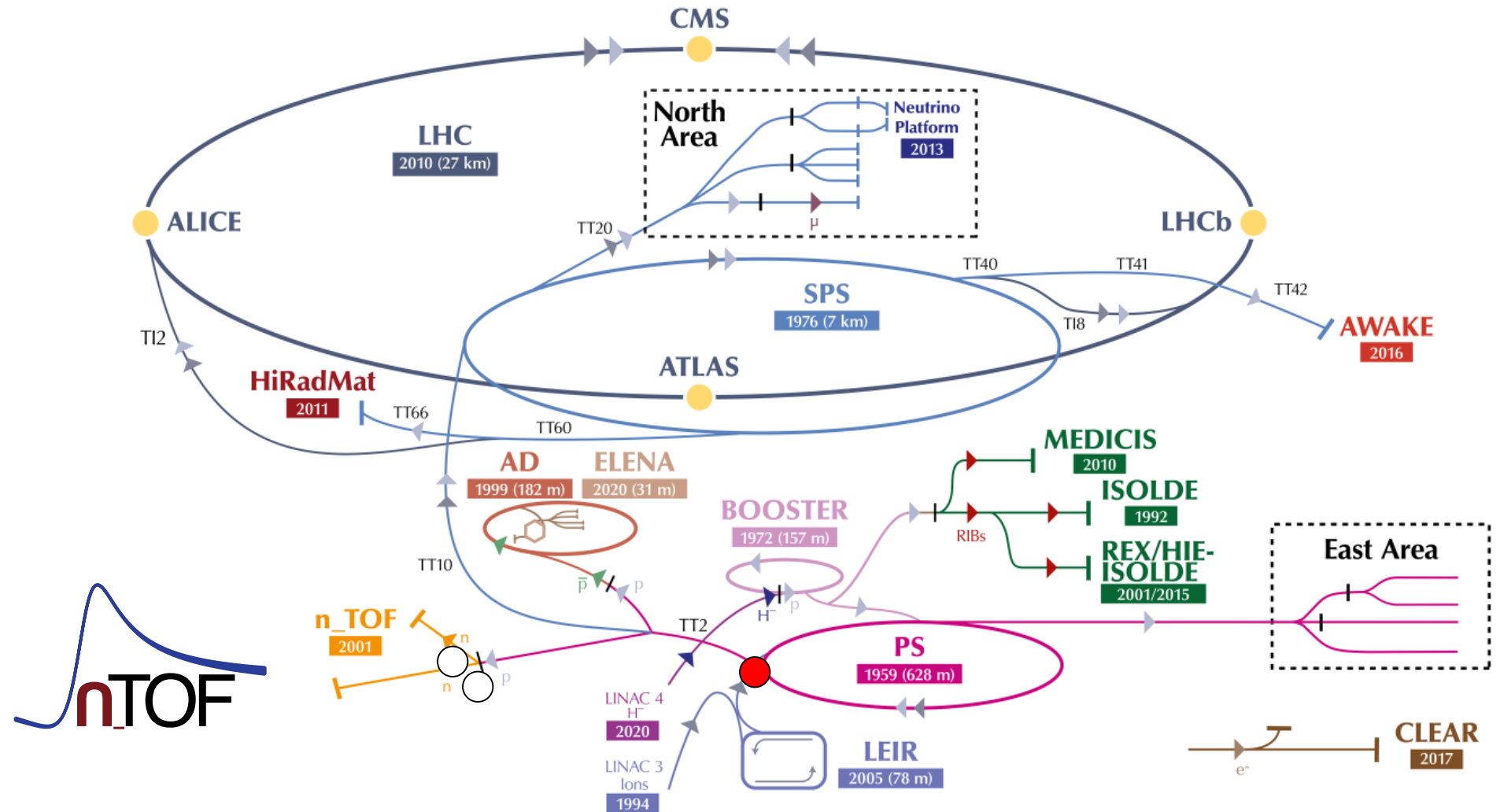
G. Cescutti et al. MNRAS (2018)
 N. Nishimura et al. MNRAS (2016)

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n_TOF @ CERN

The CERN accelerator complex *Complexe des accélérateurs du CERN*

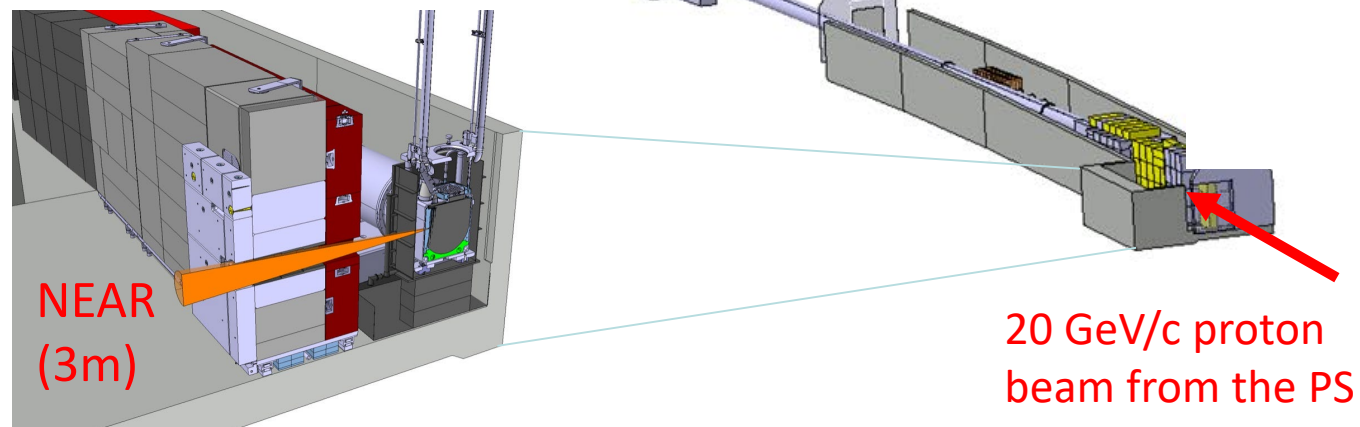
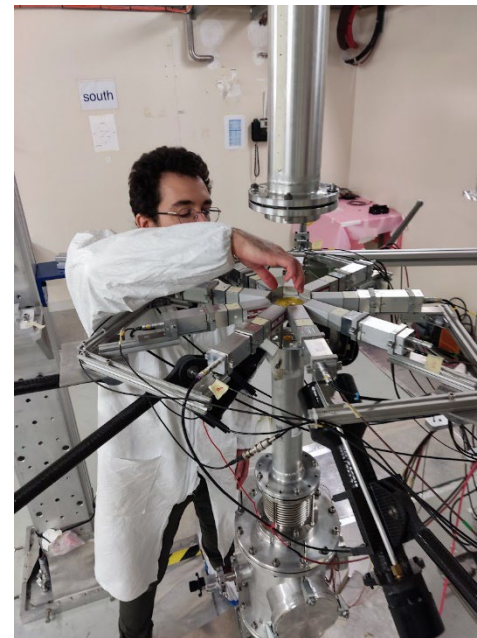
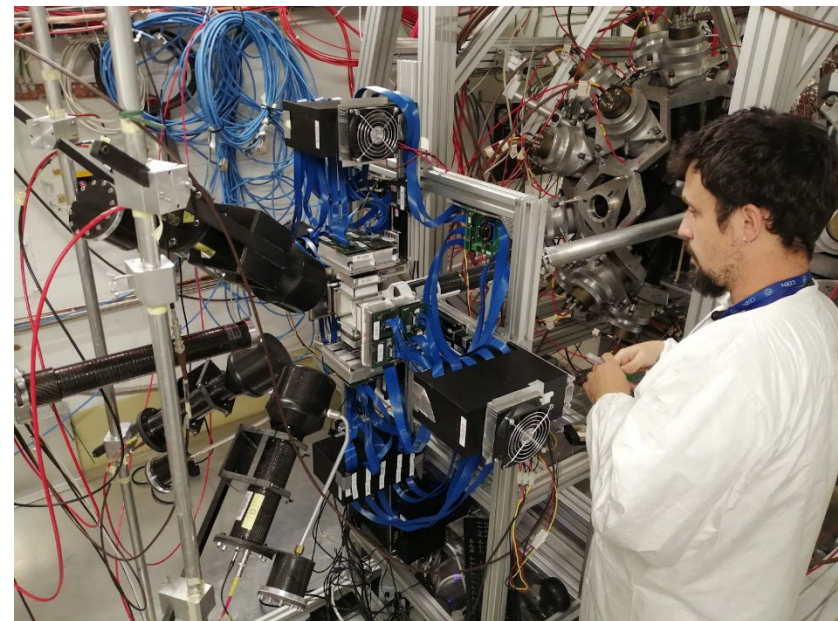


The n_TOF facility

EAR2 @ 20m

n_TOF

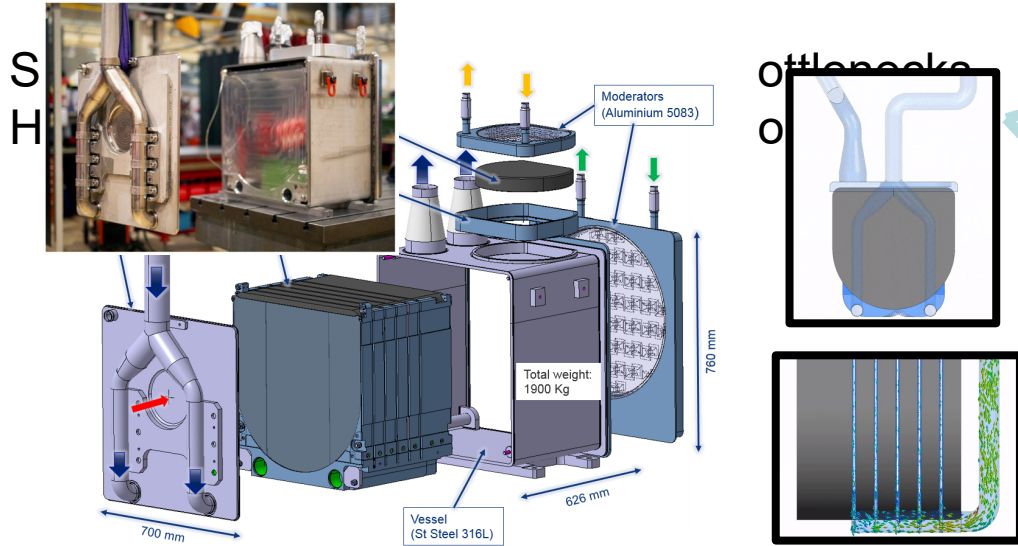
EAR1@186 m



Uniqueness of n_TOF

- instantaneous intensity and energy distribution
- repetition rate of the proton driver
- time/neutron-energy resolution (long flight-path)
- background conditions

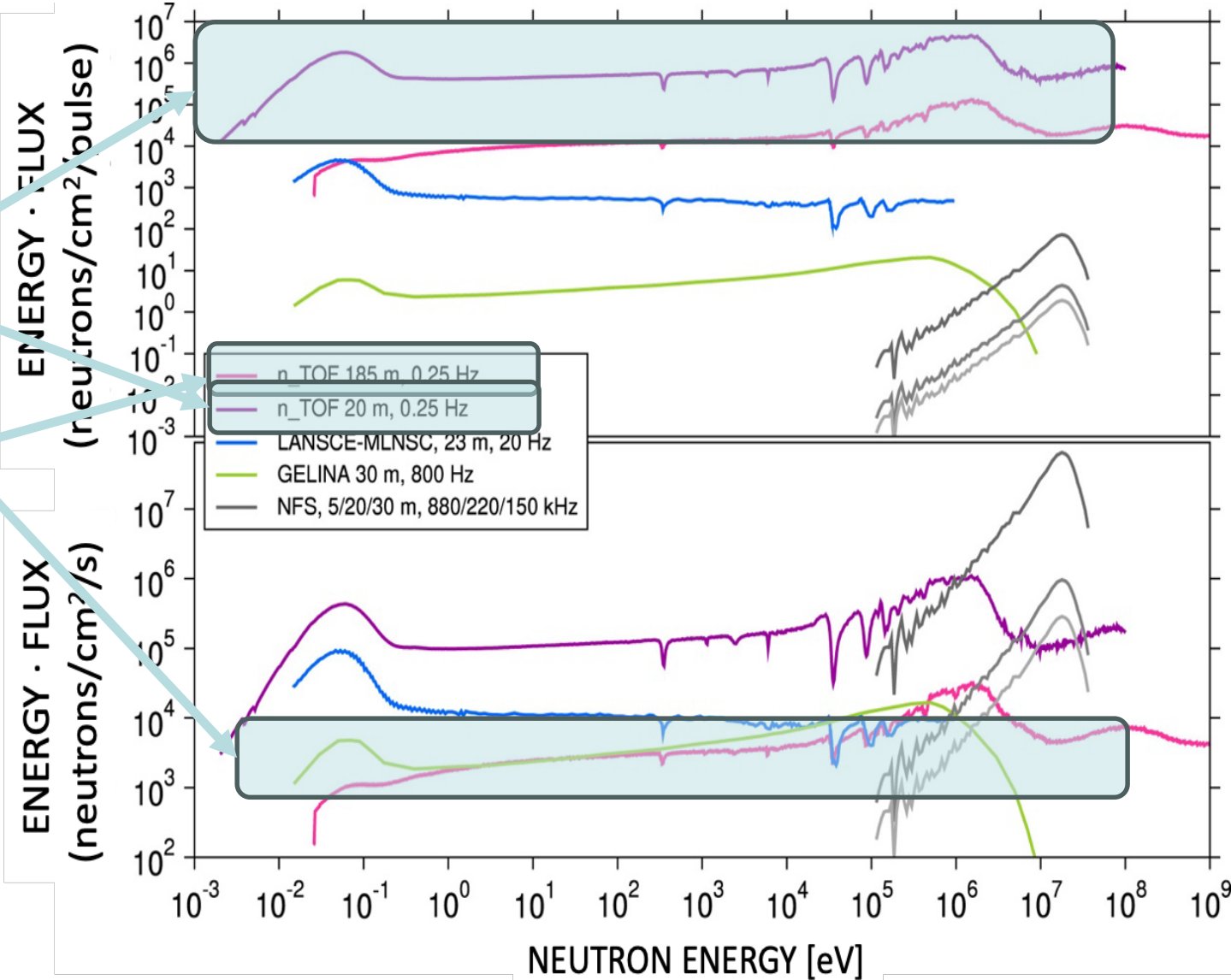
Radioactive samples → s-branchings
(sample-activity dominated background)



Neutron Physics with particle accelerators

N. Colonna, F. Gunsing, and F. Käppeler

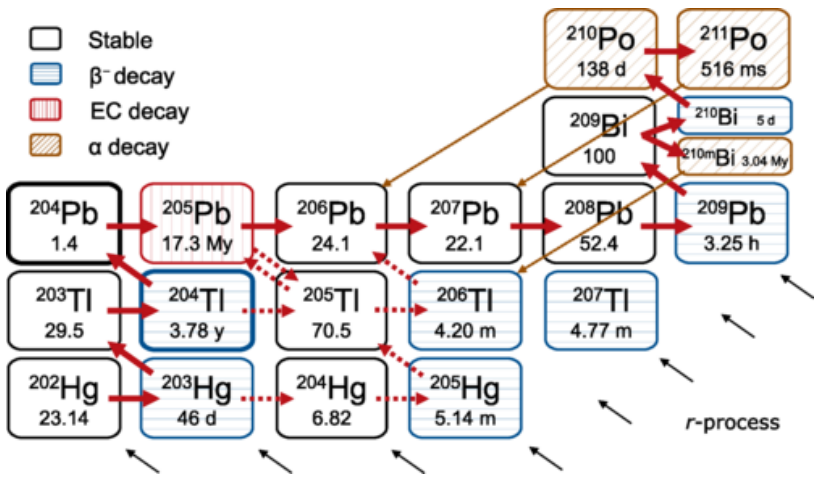
[Progress in Particle and Nuclear Physics, 101, 177 \(2018\)](#)



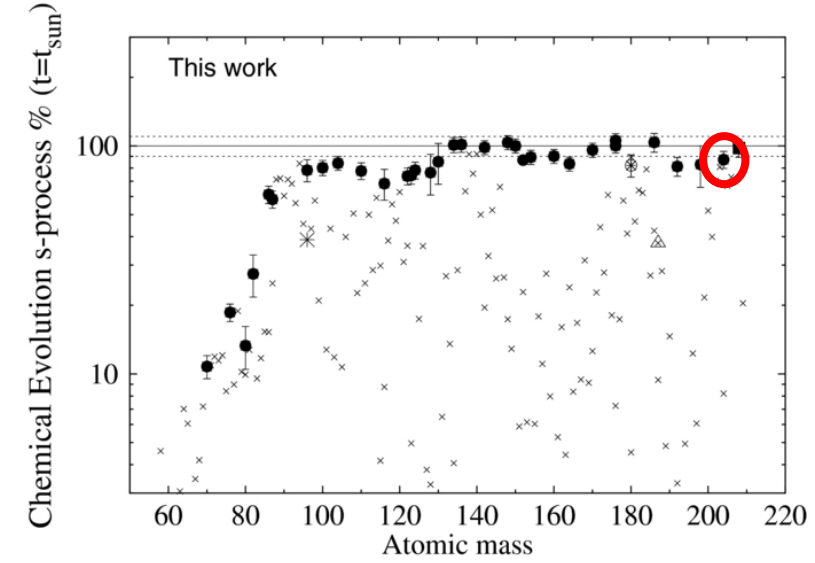
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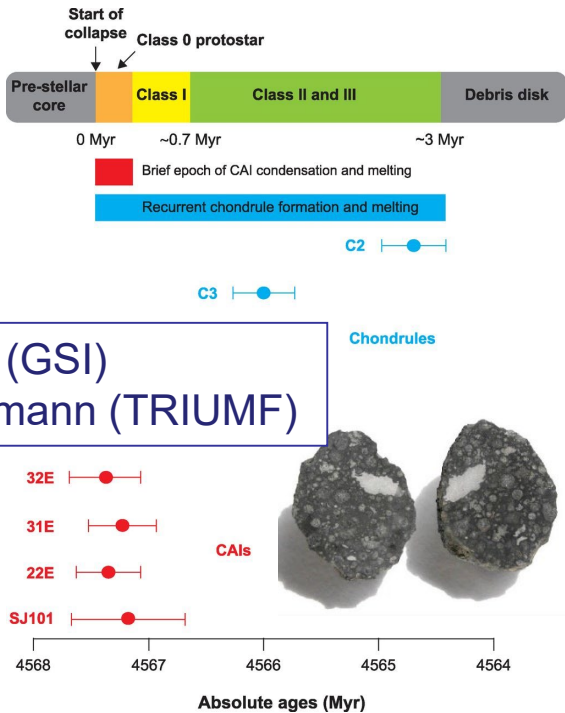
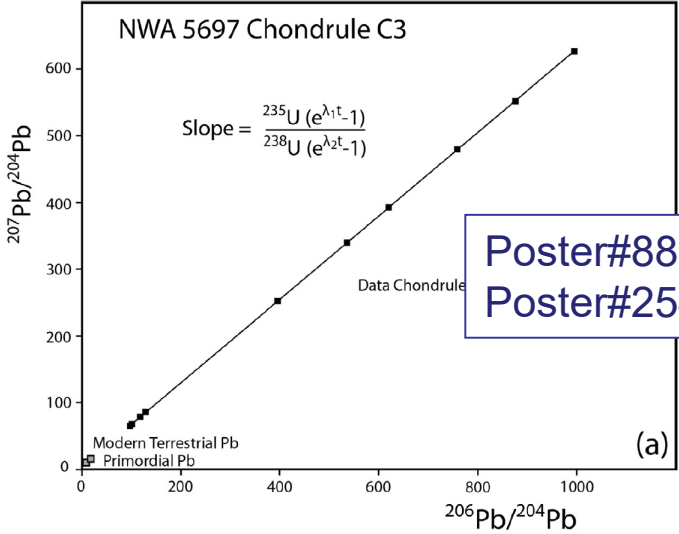
Origin of the heaviest s-only isotope ^{204}Pb : Neutron capture on ^{204}Tl (3.78y)



- Pure s-process origin
 - No r-process contribution (shielded)
 - No radiogenic contribution from U/Th
- Benchmark for AGB- and GCE-models (Travaglio 2004, Bisterzo 2014)
- Primordial abundance preserved: the reference for Pb-Pb clock (Connelly 2012)
- Its nucleosynthesis is impacted (dominated) by the branching at ^{204}Tl
- Thus far only theoretical estimates existed for the latter (x2 variations)!
- γ -process contribution (Pignatari 2016) and/or fractionation in ESS (González 2014)?



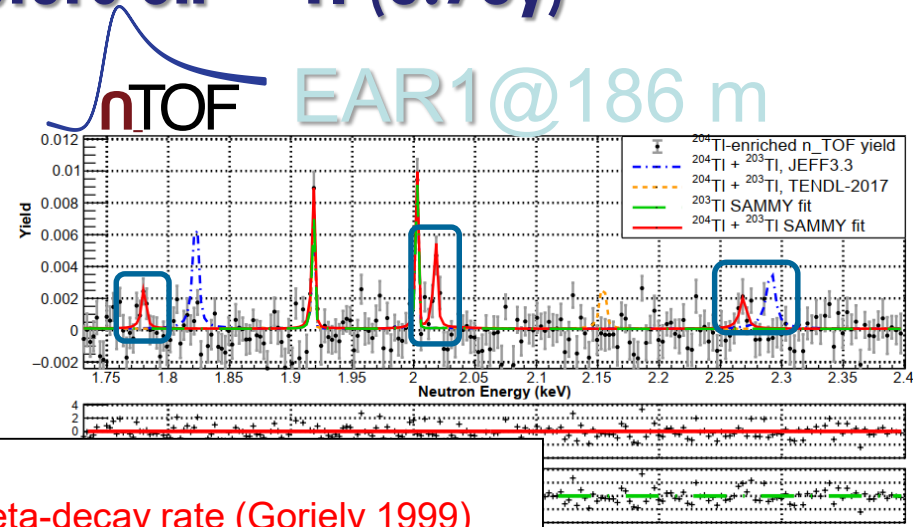
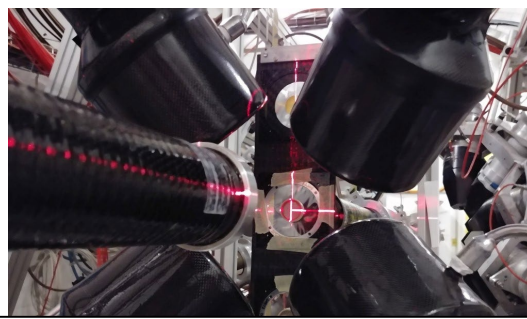
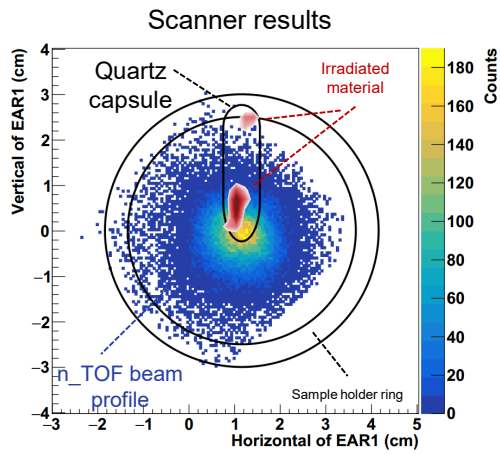
$$\left(\frac{{}^{20x}\text{Pb}}{{}^{204}\text{Pb}}\right)_{\text{today}} = \left(\frac{{}^{20x}\text{Pb}}{{}^{204}\text{Pb}}\right)_{t_0} + \left(\frac{{}^{23x}\text{U}, {}^{232}\text{Th}}{{}^{204}\text{Pb}}\right)_{\text{today}} (e^{\lambda t} - 1)$$



A Casanovas-Hoste *et al.* (The n_TOF Collaboration)
Physical Review Letters **133**, 052702 (2024)
DOI: [10.1103/PhysRevLett.133.052702](https://doi.org/10.1103/PhysRevLett.133.052702)

J.N. Connelly et al. Science **338** 2012
DOI: [10.1126/science.1226919](https://doi.org/10.1126/science.1226919)

Origin of the heaviest s-only isotope ^{204}Pb : Neutron capture on ^{204}Tl (3.78y)



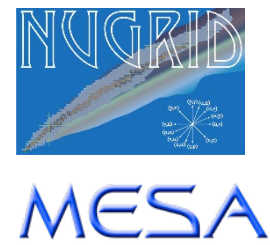
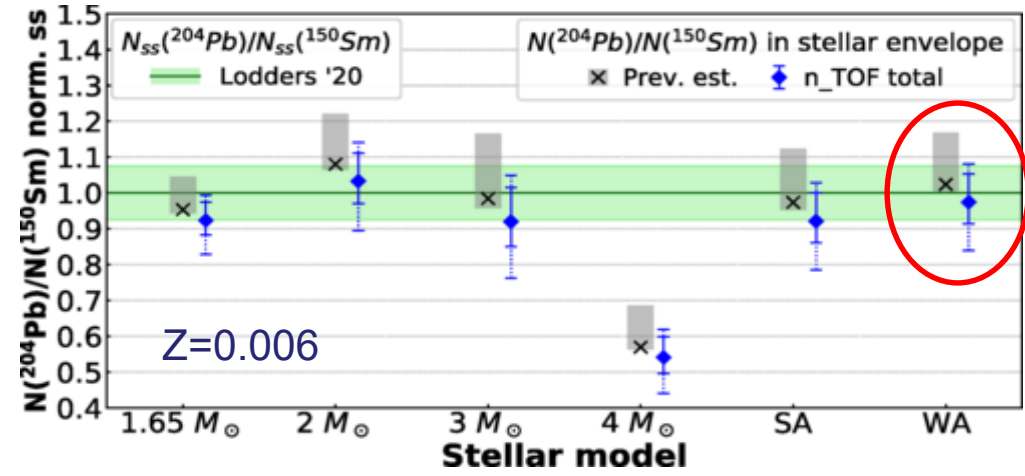
Pellet containing 225 mg of ^{203}Tl , 99.5% isotopic purity, produced at **PSI** by machine pressing and inserted into quartz container

Irradiated at **ILL reactor** with thermal neutrons for 55 days: **9 mg of ^{204}Tl** produced

180 GBq of β activity plus radioactive impurities and bremsstrahlung



→ Remaining open questions:
- Thermal dependency of the beta-decay rate (Goriely 1999)
- Strength of the $^{22}\text{Ne}(\alpha, n)$ source

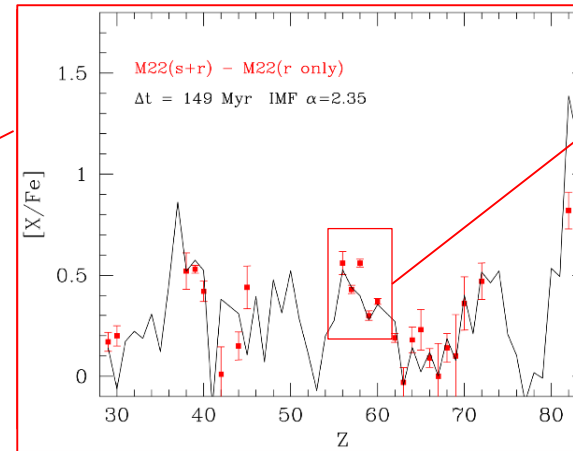


The uncertainty arising from the $^{204}\text{Tl}(n, \gamma)$ cross section on the s-process abundance of ^{204}Pb has been reduced from $\sim 30\%$ down to $+8\%/-6\%$, and the s-process calculations are in agreement with K. Lodders in 2021.

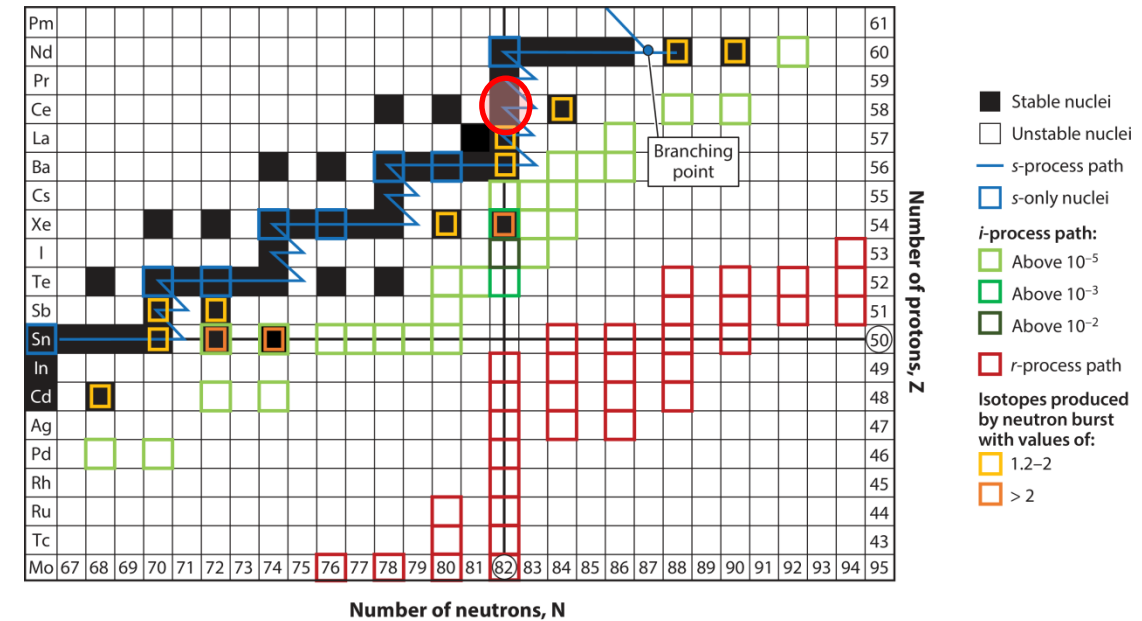
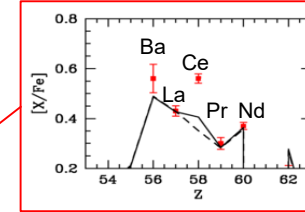
The ^{140}Ce s-process bottleneck puzzle



2nd gen. M22 stars (N – Nr)



O. Straniero et al., ApJ (2014)

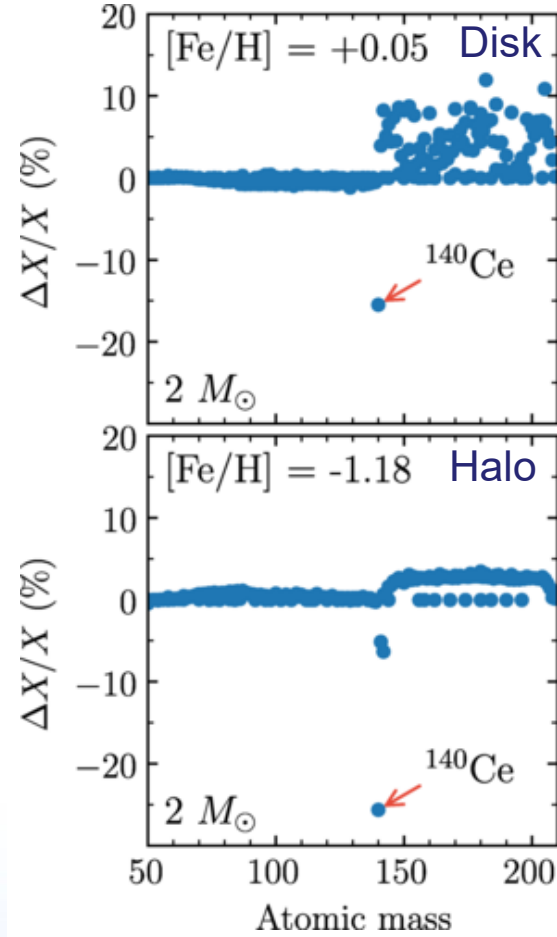
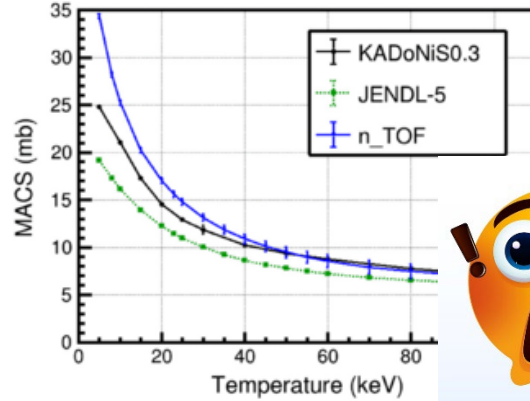
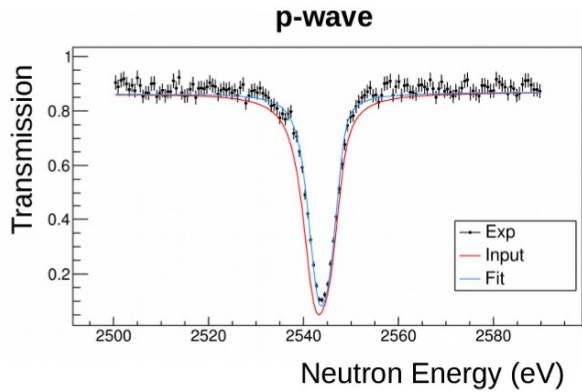
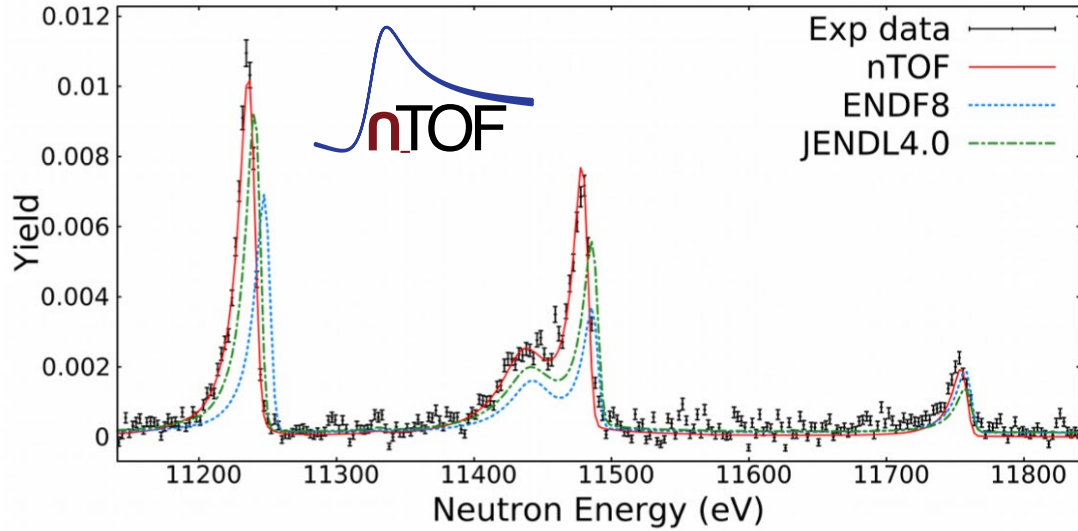


Lugaro M, et al. 2023
 Annu. Rev. Nucl. Part. Sci. 73:315–40

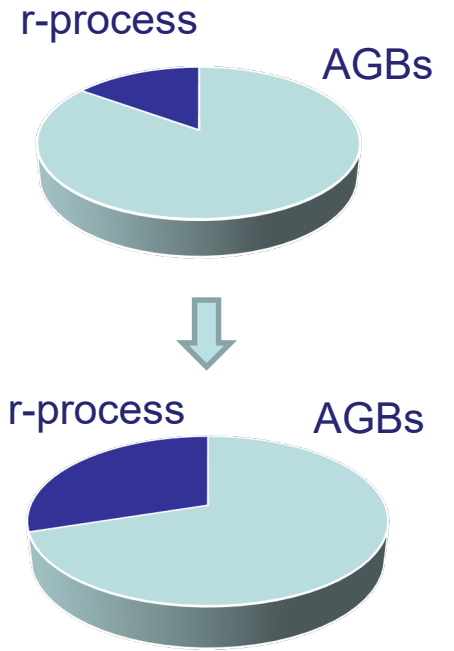
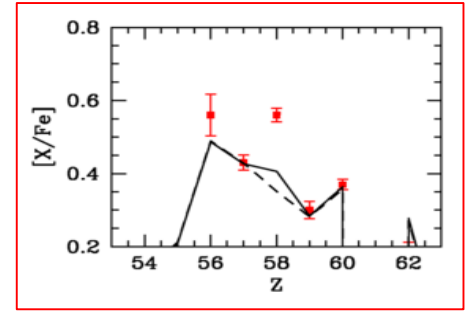
- Abundances in s-process enhanced stars in M22 show a strong overproduction of Ce wrt AGB-models (Strainero, 2014).
- Ce abundance should be dominated by ^{140}Ce , 80% s-process (Prantzos et al. 2020)
- Neutron-magic (verly low CS) implies that previous CS determinations may be affected by neutron-sensitivity effects
- Is the $^{140}\text{Ce}(n,g)$ cross section actually smaller (?)

The ^{140}Ce s-process bottleneck puzzle

EAR1@186 m



FUNS, Straniero 2006, Cristallo 2011



Poster#261 R.N. Sahoo (INFN-Bologna)
Tue. 17th, 17:55 M. Friedman #171

S. Amaducci *et al.* (The n_TOF Collaboration)
Physical Review Letters **132**, 122701 (2024)
DOI: [10.1103/PhysRevLett.132.122701](https://doi.org/10.1103/PhysRevLett.132.122701)

- Possibly increase r-process by a factor x2 $\rightarrow$ GCE model calculations
- Possible i-process contribution? $\rightarrow$ Larger ^{140}Ba production ($\downarrow$ CS)
- 35% higher than SARAF-activation measurement R.N. Sahoo *et al.*, Phys. Rev. C, Volume **109** Issue 2 025808 (2024) $\rightarrow$ Has to be understood (!)

N. Prantzos *et al.*, Mon. Not. R. Astron. Soc. **491**, 1932 (2020)

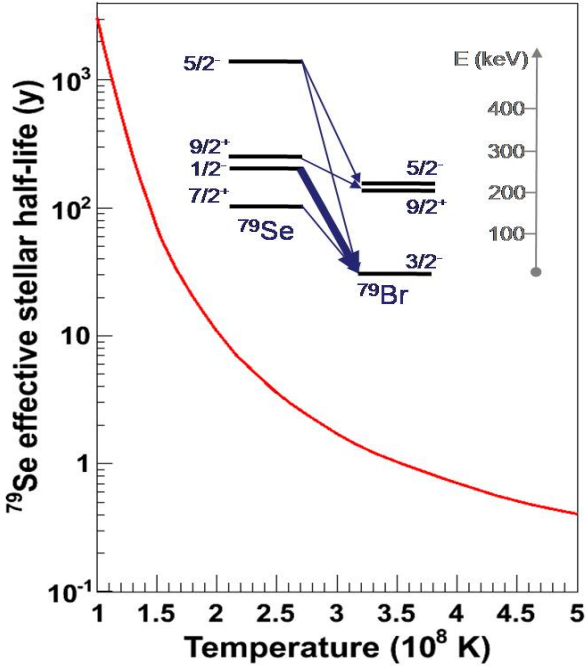
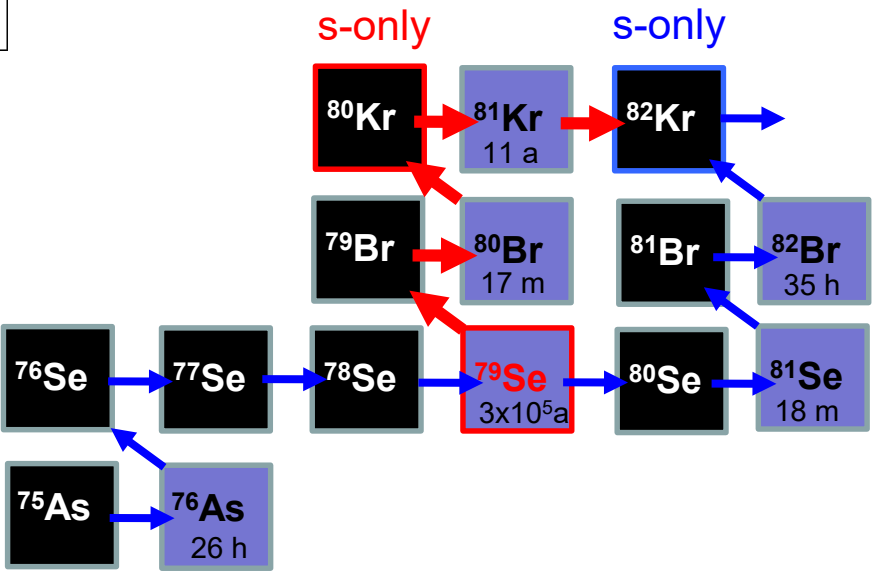
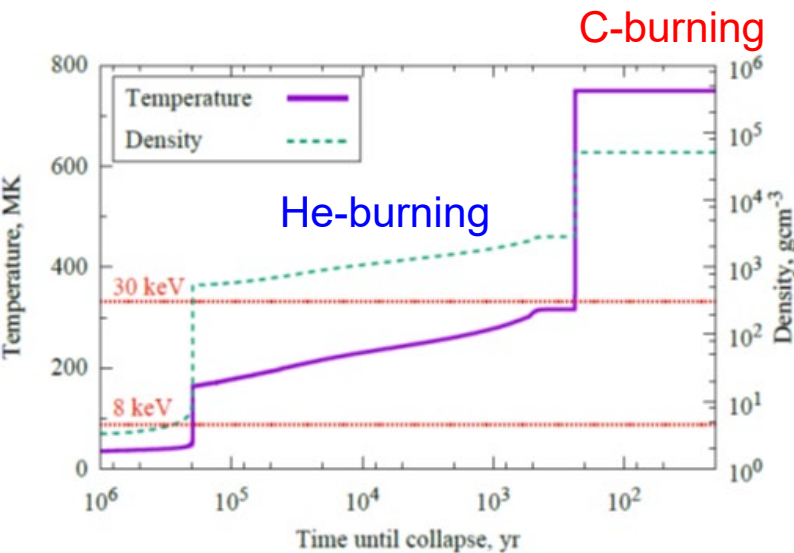
Weak s-process temperature via the ^{79}Se branching

letters to nature

Nature **332**, 700 - 702 (21 April 1988); doi:10.1038/332700a0

S-process krypton of variable isotopic composition in the Murchison meteorite

ULRICH OTT*, FRIEDRICH BEGEMANN*, JONGMANN YANG†‡ & SAMUEL EPSTEIN†



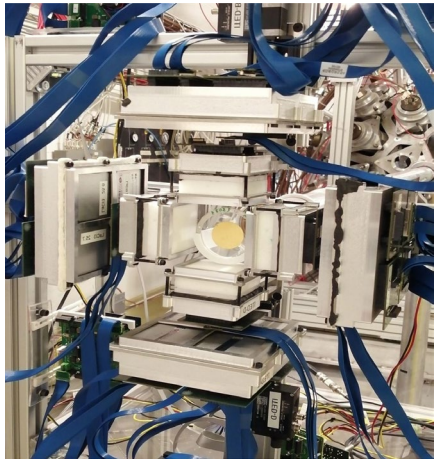
N. Nishimura et al., MNRAS469, 1752–1767 (2017)

Weak s-process temperature via the ^{79}Se branching

ILL: ~ 3 mg of ^{79}Se via ^{78}Se n-activation



6MBq γ -ray emitters
3 mg of ^{79}Se
1.6 MBq of ^{60}Co
5 MBq of ^{75}Se



PSI: ^{208}Pb ^{78}Se alloy

2.8 g of ^{208}Pb
1.0 g of ^{78}Se



Nuclear Inst. and Methods in Physics Research, A 1029 (2022) 166-143

Contents lists available at ScienceDirect

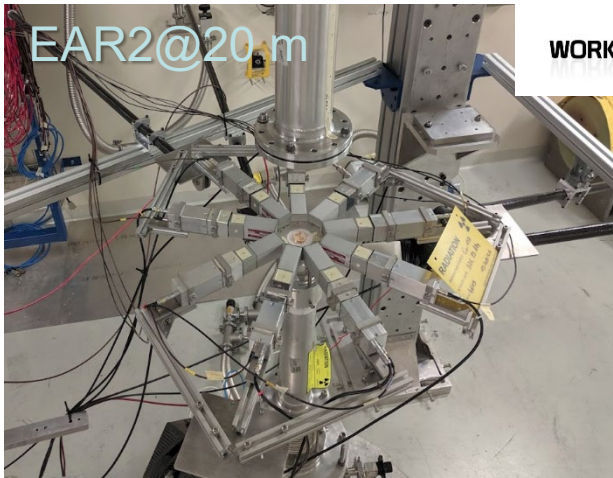
Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

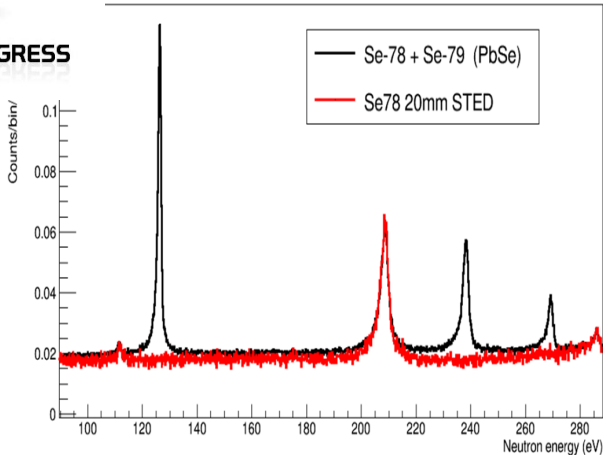
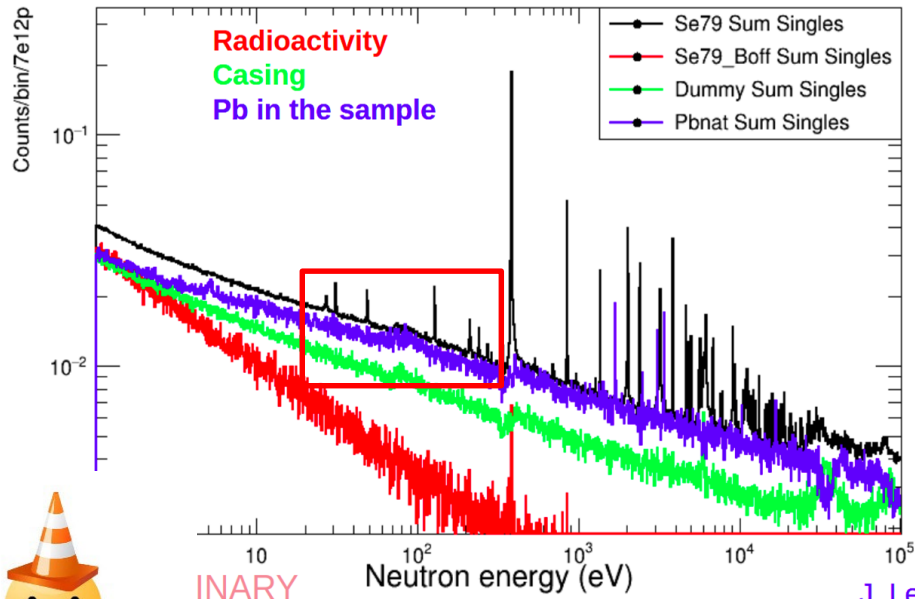
Preparation of PbSe targets for ^{79}Se neutron capture cross section studies

Nadine M. Chiera ^{a,*}, Emilio Andrea Maugeri ^a, Ivan Danilov ^a, Javier Balibrea-Correa ^b, Cesar Domingo-Pardo ^b, Ulli Köster ^c, Jorge Lerendegui-Marco ^b, Mario Veicht ^{a,d}, Ivan Zivadinovic ^{a,e}, Dorothea Schumann ^a, the n_TOF collaboration

^a Paul Scherrer Institut, Switzerland
^b Institut de Física Corpuscular - Consejo Superior de Investigaciones Científicas/Universidad de Valencia, Spain
^c Institut Laser Langevin, France
^d Ecole Polytechnique Fédérale de Lausanne, Switzerland
^e Eidgenössische Technische Hochschule Zürich, Switzerland



WORK IN PROGRESS



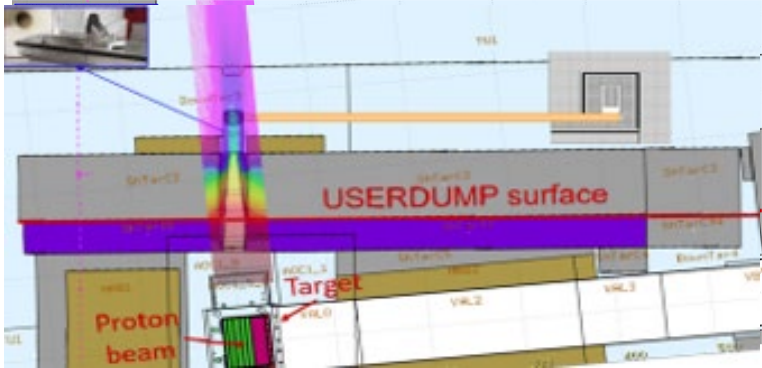
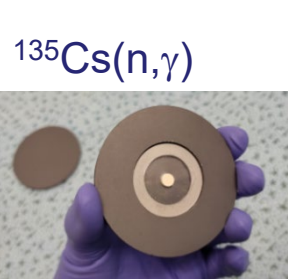
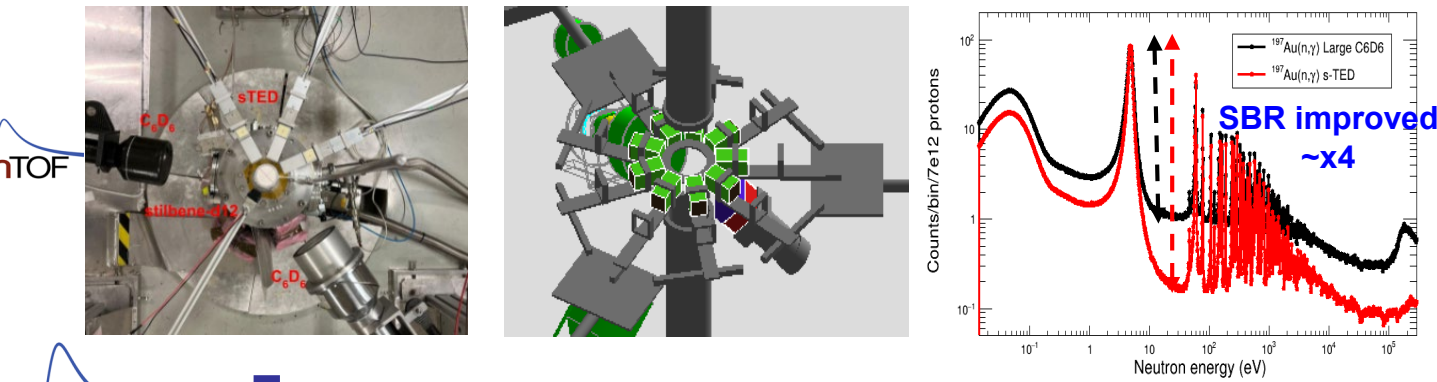
J. Lerendegui-Marco et al. (analysis in progress)

Outline

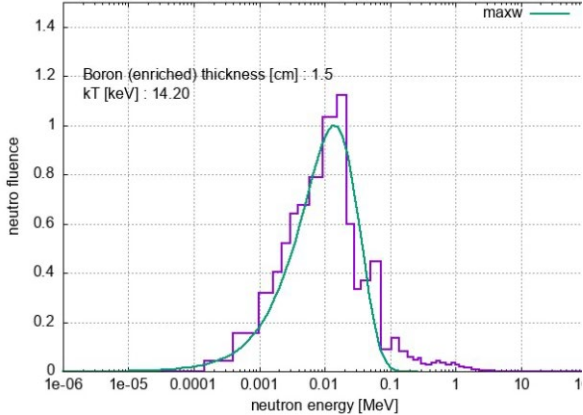
- Introduction: s-process nucleosynthesis and why more (n, γ) measurements
- The CERN n_TOF facility: a growing family of stations!
- Recent s-process experiments (selected examples):
 - Origin of the heaviest s-only ^{204}Pb : Neutron capture on $^{204}\text{Tl}(3.78\text{y})$
 - The ^{140}Ce s-process bottleneck “puzzle”
 - Weak s-process temperature via the ^{79}Se branching
- Future (experimental) prospects
- Summary & Outlook

What comes next?

- Novel detector developments (d12-stilbene array) STAR



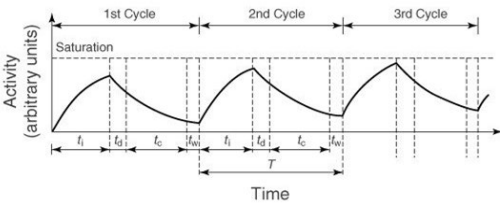
We are here



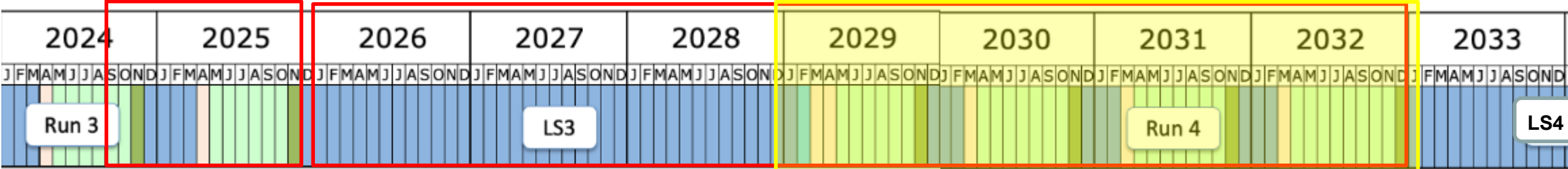
Sample	Half-life (yr)	Comment
⁶³ Ni	100.1	TOF work in progress (Couture, 2009), sample with low enrichment
⁷⁹ Se	2.95 × 10 ⁵	Important branching, constrains s-process temperature in massive stars
⁸¹ Kr	2.29 × 10 ⁵	Part of ⁷⁹ Se branching
⁸⁵ Kr	10.73	Important branching, constrains neutron density in massive stars
⁹⁵ Zr	64.02 d	Not feasible in near future, but important for neutron density low-mass AGB stars
¹³⁴ Cs	2.0652	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	So far only activation measurement at kT = 25 keV by Patronis <i>et al.</i> (2004)
¹⁴⁷ Nd	10.981 d	Important branching at A = 147/148, constrains neutron density in low-mass AGB stars
¹⁴⁷ Pm	2.6234	Part of branching at A = 147/148
¹⁴⁸ Pm	5.368 d	Not feasible in the near future
¹⁵¹ Sm	90	Existing TOF measurements, full set of MACS data available (Abbondanno <i>et al.</i> 2004a; Wiesshak <i>et al.</i> 2006c)
¹⁵⁴ Eu	8.593	Complex branching at A = 154, 155, sensitive to temperature and neutron density
¹⁵⁵ Eu	4.753	So far only activation measurement at kT = 25 keV by Jaag and Käppeler (1995)
¹⁵³ Gd	0.658	Part of branching at A = 154, 155
¹⁶⁰ Tb	0.198	Weak temperature-sensitive branching, very challenging experiment
¹⁶³ Ho	4570	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	Important branching, constrains neutron density in low-mass AGB stars
¹⁷¹ Tm	1.921	Part of branching at A = 170, 171
¹⁷⁹ Ta	1.82	Crucial for s-process contribution to ¹⁸⁰ Ta, nature's rarest stable isotope
¹⁸⁵ W	0.206	Important branching, sensitive to neutron density and s-process temperature in low-mass AGB stars
²⁰⁴ Tl	3.78	Determines ²⁰⁵ Pb/ ²⁰⁵ Tl clock for dating of early Solar System

F. Käppeler et al. Rev. Mod. Phys. (2011)

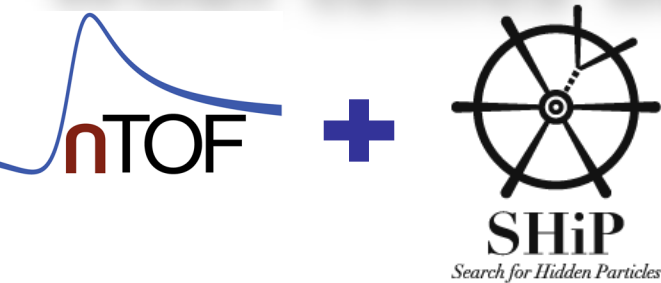
- Examples: 59Fe, 94Nb, 125Sb, 134Cs, **135Cs**, 144Ce, 148Pm, 154Eu, 155Eu, 160Tb, 170Tm, 171Tm, and 181Hf (s-process), Cs-137, 66Ni, 72Zn (i-process)



H. Beer, et al. (FZK), Nucl. Inst. Meths. 337, 2–3 (1994)

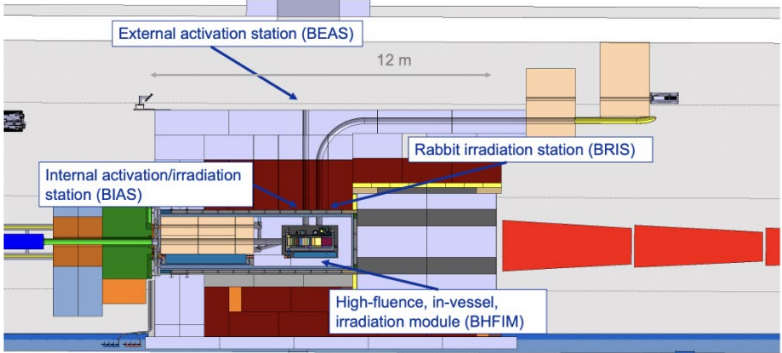
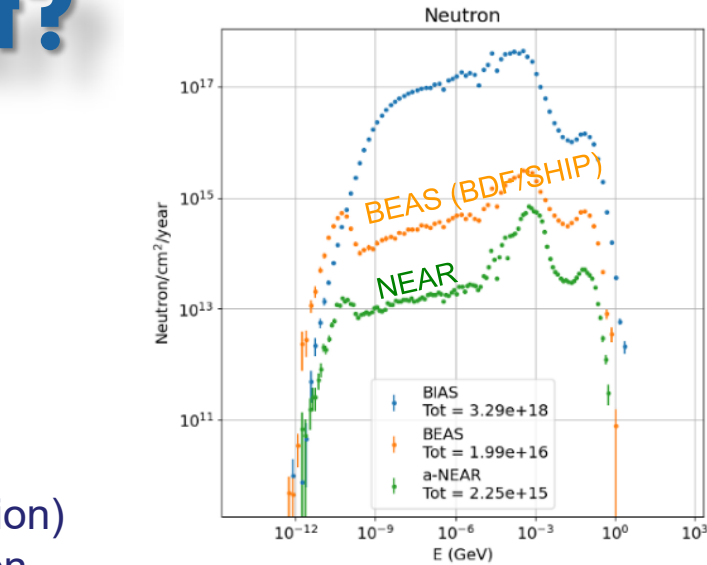
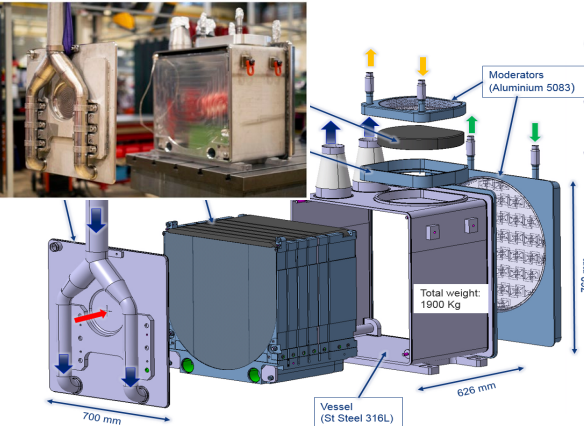


What comes next?



@LS4:
New (IV
Generation)
Spallation
Source

$\phi_n \times 4$

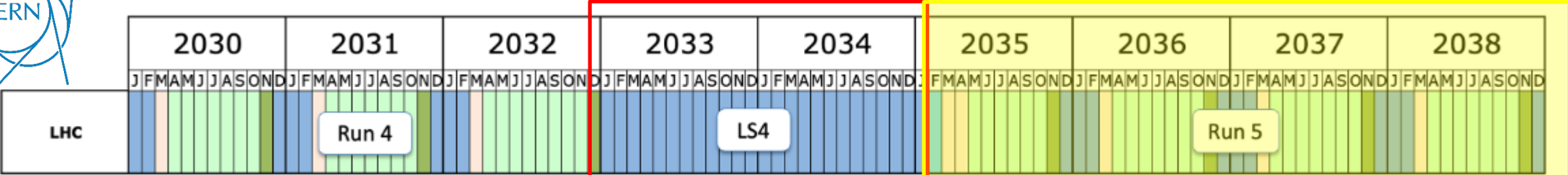


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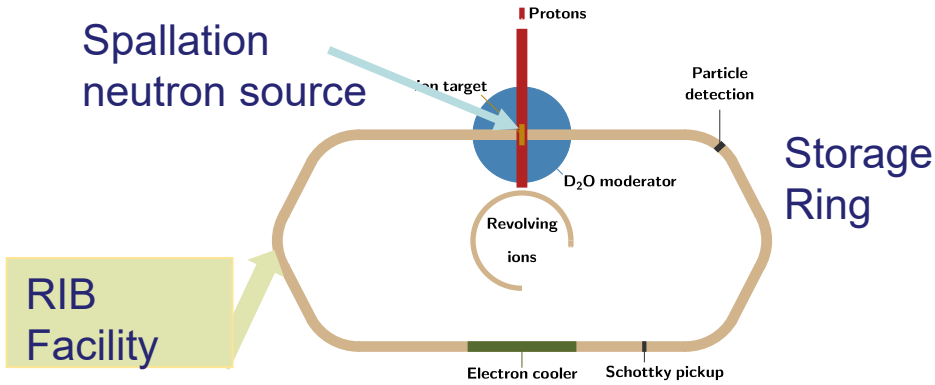
Figure 3: Top view of the BDF target station, modified in this sketch to provide a view of the potential location of the additional irradiation and activation stations.

Life-time >10y.



What comes next?

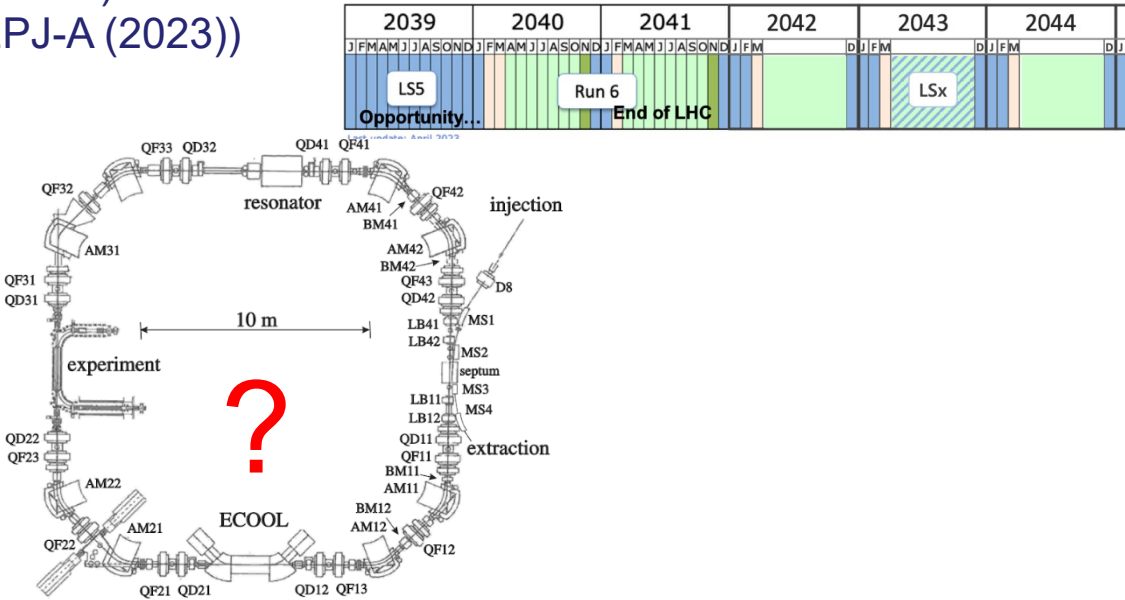
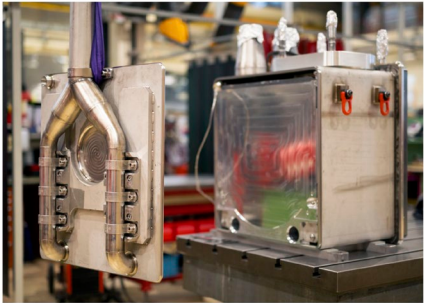
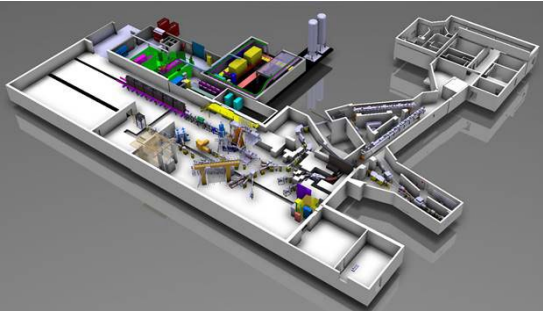
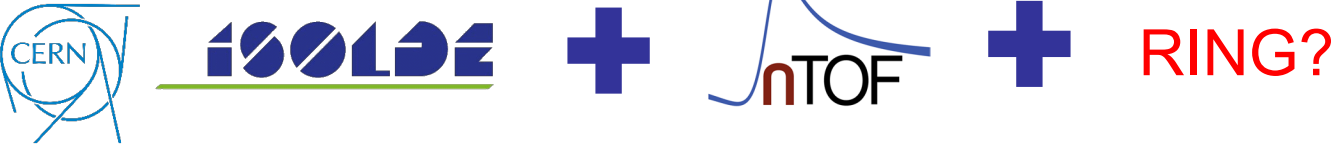
- Exploring novel approaches: neutron-sources & rings (LANL, TRIUMF)



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- Proof-of-concept project ongoing @ Los Alamos (R.Reifarth, A. Couture et al.)
- LE-customized Storage Ring + NGs @ TRIUMF (see I. Dillmann et al. EPJ-A (2023))
- Why not at CERN n_TOF&ISOLDE? Only one ring is missing...



Summary & Outlook

- Over the last years, concomitant efforts in neutron-beam **facilities** (Gen-III Spal Source, n_TOF EAR2, NEAR, etc), **detection systems** (sTED, i-TED, STAR, etc) and **sample-production techniques** (ILL, PSI, ISOLDE) have enabled a significant step forward in the measurement of some of the most challenging neutron-capture rates for stellar nucleosynthesis, including $^{94}\text{Nb}(n,\gamma)$, $^{79}\text{Se}(n,\gamma)$, $^{140}\text{Ce}(n,\gamma)$, $^{204}\text{Tl}(n,\gamma)$, etc.
- There are still a lot of neutron-capture cross-sections whose accuracy needs to be improved to the 5% level, or less, which should be doable for **stable isotopes** with state-of-the-art instrumentation and facilities.
- Improving CSs of unstable nuclei, both in accuracy and covered energy range, will require **further developments** in terms of detection systems (STAR) and facilities (4th Gen. Target, NEAR-CYCLING, BDF/SHIP, etc)
- Novel concepts, such as **inverse-kinematic measurements** utilizing storage rings combined with very-large intensity neutron sources, shall enable one to tackle for the first time neutron-capture rates of the most exotic s-process branching nuclei (^{85}Kr , ^{95}Zr , ^{135}Cs , ^{147}Nd , $^{170,171}\text{Tm}$, etc), as well as a significant number of i-process n-rich isotopes.