

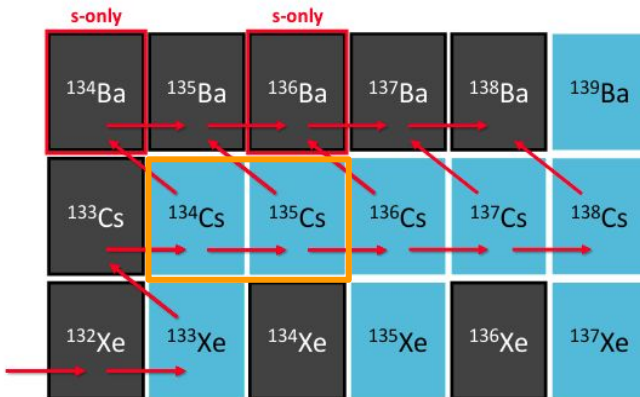
## EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

# Production of a $^{135}\text{Cs}$ sample at **ISOLDE** for (n, $\gamma$ ) activation measurements at **n\_TOF-NEAR**

**J. Leredegui-Marco, S. Carollo, C. Domingo-Pardo, F. Recchia, U. Köster,**  
V. Babiano, M. Bacak, J. Balibrea-Correa, A. Casanovas, F. Calviño, S. Cristallo, C. Guerrero, C. Lederer,  
C. Massimi, P. Milazzo, N. Patronis, S. Rothe, A. Tarifeño-Saldivia, E. Stamati, S. Stegemann, D. Vescovi

- **Motivation:  $^{135}\text{Cs}(n,\gamma)$  cross section for nucleosynthesis & transmutation**
- $^{135}\text{Cs}(n,\gamma)$  activation measurement at NEAR
- $^{135}\text{Cs}$  sample production at ISOLDE
- Summary and outlook



**S-process branchings at  $^{134}\text{Cs}$  and  $^{135}\text{Cs}$ : Fix the abundance ratio of the s-only  $^{134,136}\text{Ba}$ . Accurately measured from SiC from presolar grains of AGB origin**

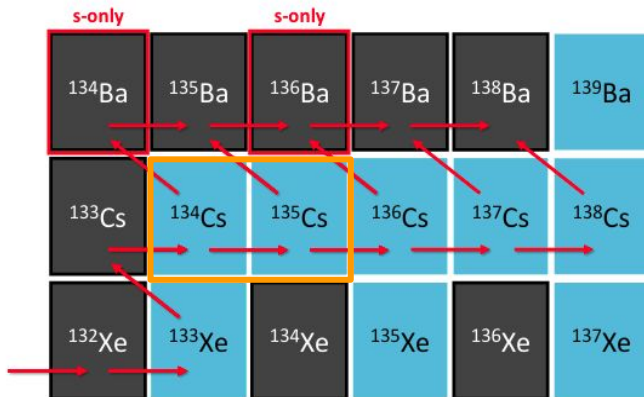
## The s process: Nuclear physics, stellar models, and observations

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

Sample	Half-life (yr)	$Q$ value (MeV)	Comment
$^{63}\text{Ni}$	100.1	$\beta^-$ , 0.066	TOF work in progress (Couture, 2009), sample with low enrichment
$^{79}\text{Se}$	$2.95 \times 10^5$	$\beta^-$ , 0.159	Important branching, constrains s-process temperature in massive stars
$^{81}\text{Kr}$	$2.29 \times 10^5$	EC, 0.322	Part of $^{79}\text{Se}$ branching
$^{85}\text{Kr}$	10.73	$\beta^-$ , 0.687	Important branching, constrains neutron density in massive stars
$^{95}\text{Zr}$	64.02 d	$\beta^-$ , 1.125	Not feasible in near future, but important for neutron density low-mass AGB stars
$^{134}\text{Cs}$	2.0652	$\beta^-$ , 2.059	Important branching at $A = 134, 135$ , sensitive to s-process temperature in low-mass AGB stars, measurement not feasible in near future
$^{135}\text{Cs}$	$2.3 \times 10^6$	$\beta^-$ , 0.269	So far only activation measurement at $kT = 25$ keV by Patronis <i>et al.</i> (2004)
$^{147}\text{Nd}$	10.98 T d	$\beta^-$ , 0.896	Important branching at $A = 147/148$ , constrains neutron density in low-mass AGB stars
$^{147}\text{Pm}$	2.6234	$\beta^-$ , 0.225	Part of branching at $A = 147/148$
$^{148}\text{Pm}$	5.368 d	$\beta^-$ , 2.464	Not feasible in the near future
$^{151}\text{Sm}$	90	$\beta^-$ , 0.076	Existing TOF measurements, full set of MACS data available (Abbondanno <i>et al.</i> , 2004a; Wisshak <i>et al.</i> , 2006c)
$^{154}\text{Eu}$	8.593	$\beta^-$ , 1.978	Complex branching at $A = 154, 155$ , sensitive to temperature and neutron density
$^{155}\text{Eu}$	4.753	$\beta^-$ , 0.246	So far only activation measurement at $kT = 25$ keV by Jaag and Käppeler (1995)
$^{153}\text{Gd}$	0.658	EC, 0.244	Part of branching at $A = 154, 155$
$^{160}\text{Tb}$	0.198	$\beta^-$ , 1.833	Weak temperature-sensitive branching, very challenging experiment
$^{163}\text{Ho}$	4.570	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)
$^{170}\text{Tm}$	0.352	$\beta^-$ , 0.968	Important branching, constrains neutron density in low-mass AGB stars
$^{171}\text{Tm}$	1.921	$\beta^-$ , 0.098	Part of branching at $A = 170, 171$
$^{179}\text{Ta}$	1.82	EC, 0.115	Crucial for s-process contribution to $^{180}\text{Ta}$ , nature's rarest stable isotope
$^{185}\text{W}$	0.206	$\beta^-$ , 0.432	Important branching, sensitive to neutron density and s-process temperature in low-mass AGB stars
$^{204}\text{Tl}$	3.78	$\beta^-$ , 0.763	Determines $^{205}\text{Pb}/^{205}\text{Tl}$ clock for dating of early Solar System

**$^{134,135}\text{Cs}$**

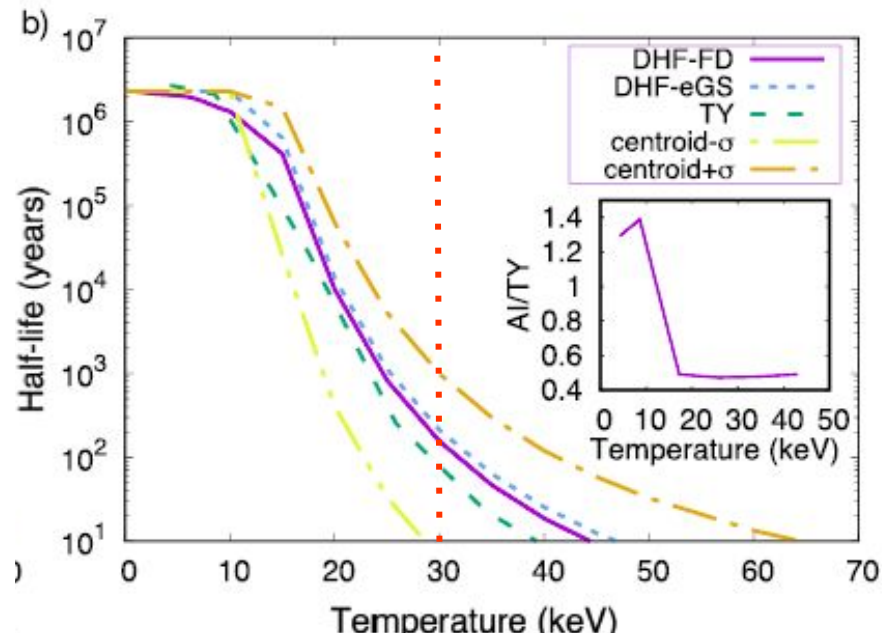
→ Among the **21 key s-nuclei** listed in Käppeler, *Rev. Mod. Phys.* **83**, 157 (2011)



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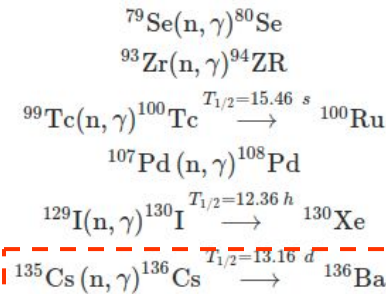
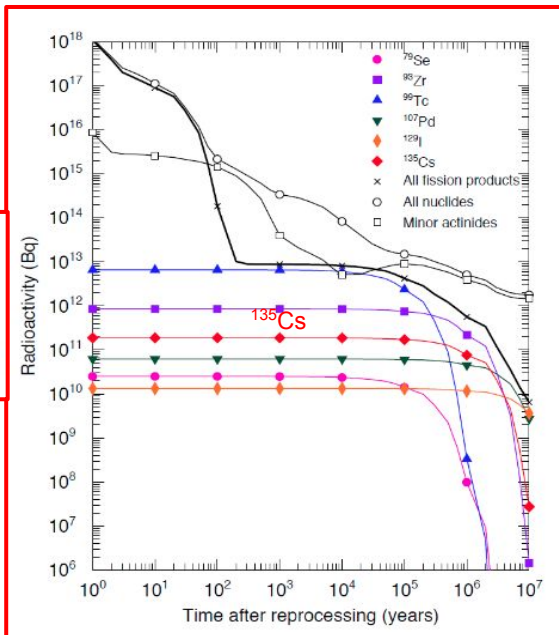
[Palmerini et al. \(2021\)](#): *Ba isotopic ratios sensitive to the  $(n,\gamma)$  CS of  $^{134}\text{Cs}$  and  $^{135}\text{Cs}$  + Temperature dependence of the  $\beta$ -decay rates*

**Accurate  $(n,g)$  cross section  $\rightarrow$  Stellar thermometer**



[Taioli et al. \(2022\)](#): **Half-life of  $^{135}\text{Cs}$  reduced to few hundred years under TPs conditions**

**$^{135}\text{Cs}$  ( $2.3 \times 10^6$  y): among LLFPs with largest contribution to long-term radiotoxicity**



Article | Open Access | Published: 24 October 2017

## Method to Reduce Long-lived Fission Products by Nuclear Transmutations with Fast Spectrum Reactors

Satoshi Chiba, Toshio Wakabayashi, Yoshiaki Tachi, Naoyuki Takaki, Atsunori Terashima, Shin Okumura & Tadashi Yoshida

## Handbook of advanced radioactive waste conditioning technologies

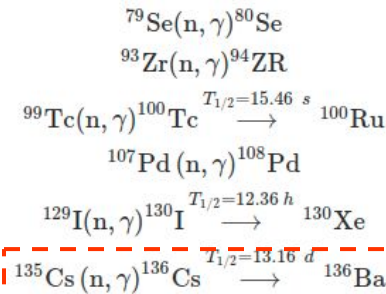
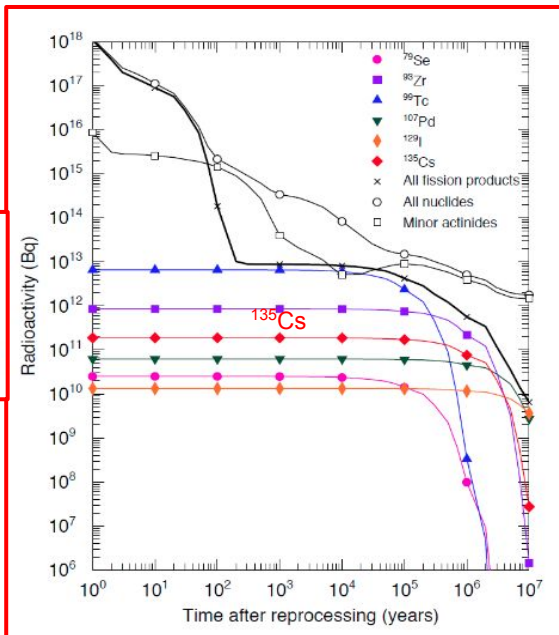
In terms of long-term radiotoxicity, however, long-lived fission products like Tc-99 and I-129, together with Se-79 and Cs-135, are the main contributors in addition to the above-mentioned actinides, and dominate the potential hazard in the case of HLW not containing actinides. Figure 13.2 compares

## Implications of Partitioning and Transmutation in Radioactive Waste Management

In the context of radiological risk reduction (concerning a deep geologic repository), the water soluble fission products  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{135}\text{Cs}$ ,  $^{79}\text{Se}$  and  $^{126}\text{Sn}$  are the most important radionuclides, due to a combination of toxicity, half-life and concentration.



**$^{135}\text{Cs}$  ( $2.3 \times 10^6$  y): among LLFPs with largest contribution to long-term radiotoxicity**



**(n,γ) CS: fast energy range**

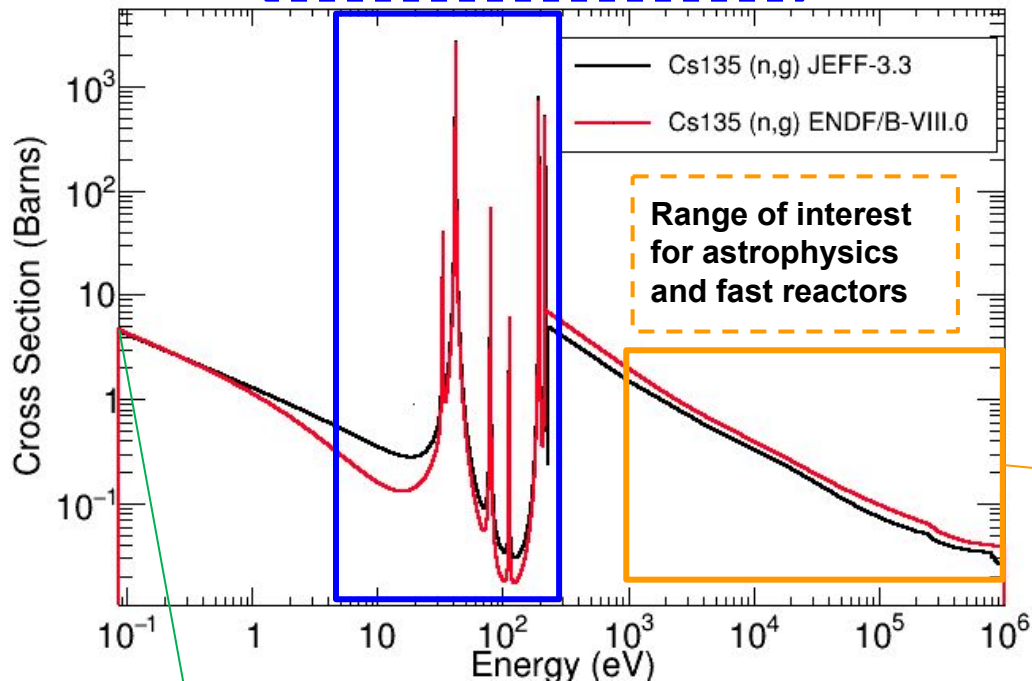
**Handbook of advanced radioactive waste conditioning technologies**

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RRR: 6 resonances from (n,tot)  
([V.A.Anufriev](#) 1987, not in EXFOR)



Range of interest  
for astrophysics  
and fast reactors

- (n,g) → MACS via activation**
- 2 Previous measurement at FZK
  - Only 2 energies: **@ 30 and 500keV:**
  - Corrections for the thermal contribution

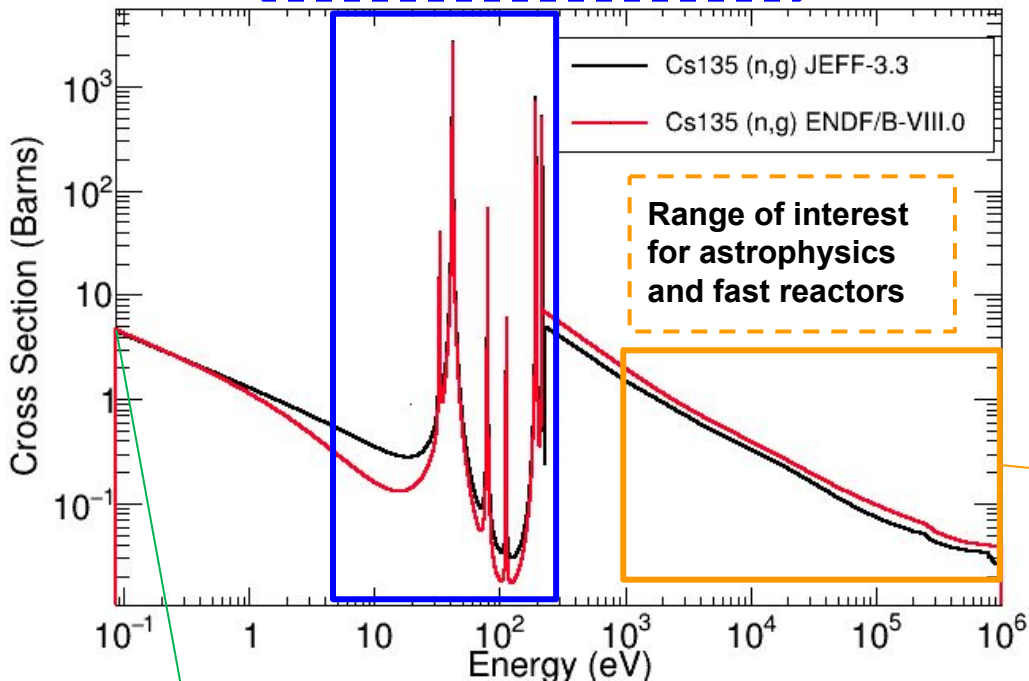
Sugarma (1949), Baerg (1958),  
Katoh (1997) (n,g) @ thermal

PHYSICAL REVIEW C **69**, 025803 (2004)

### Neutron capture studies on unstable $^{135}\text{Cs}$ for nucleosynthesis and transmutation

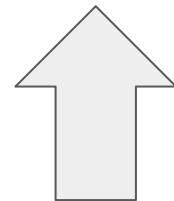
N. Patronis,<sup>1</sup> S. Dababneh,<sup>2,\*</sup> P. A. Assimakopoulos,<sup>1</sup> R. Gallino,<sup>3</sup> M. Heil,<sup>2</sup> F. Käppeler,<sup>2,†</sup> D. Karamanis,<sup>1</sup> P. E. Koehler,<sup>4</sup> A. Mengoni,<sup>5</sup> and R. Plag<sup>2</sup>

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**(n,g) via activation at NEAR:**

- Capability to tune the energy spectrum
- MACS in a wider range (1 keV-1MeV) of interest for astrophysics & fast reactors



**(n,g) → MACS via activation**

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- Only 2 energies: **@ 30 and 500keV:**
- Corrections for the thermal contribution

Sugarma (1949), Baerg (1958),  
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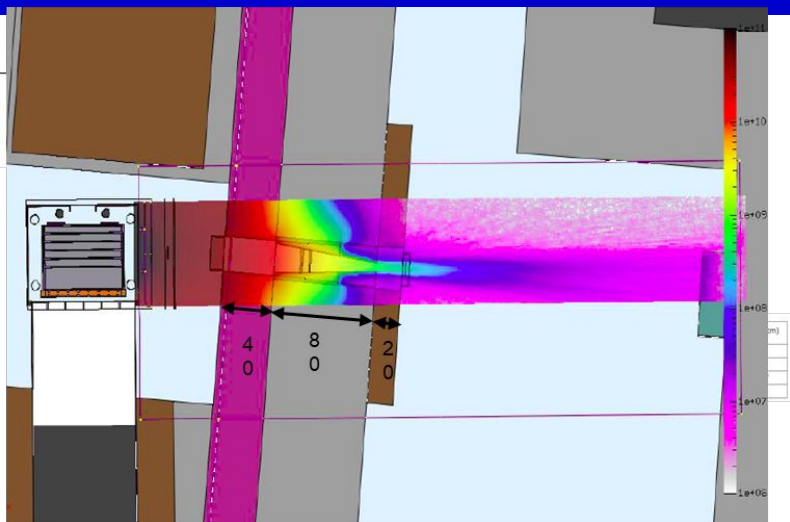
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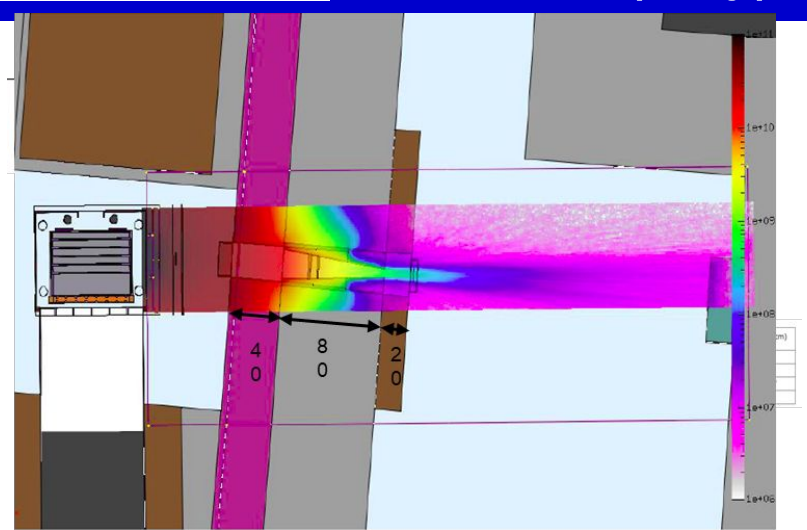


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## Physics at the NEAR:

- High flux ( $\times \sim 100$  EAR2 outside the collimator)
  - Activation measurements
  - Small mass
  - Unstable isotopes
- e.g. s-process branchings not accessible via TOF



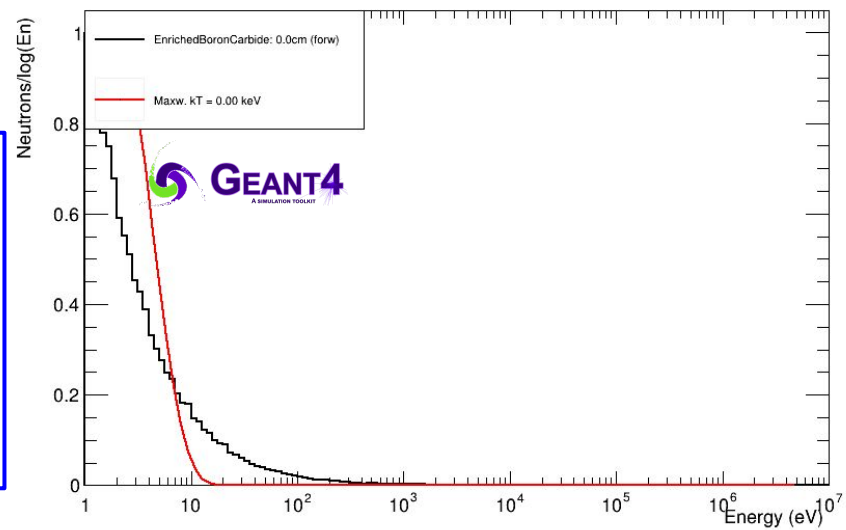
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**Neutron spectra + filter ( $\text{B}_4\text{C}$ , ...) after the collimator exit:**

- Measure SACS @ various stellar temperatures from 0.1 to several hundreds of keV.
- E. Stamati et al., [CERN-INTC-2022-008: INTC-P-623 \(2022\)](#): benchmark with long-lived ( $n,\gamma$ ) products



**Flux:** FLUKA Simulations up to marble wall + Geant4 (95% Enriched  $\text{B}_4\text{C}$ )

**Irradiation time:**  
 $3 \times T_{1/2}^{136}\text{Cs}$  (13 d)

**GEAR:** HPGe decay station

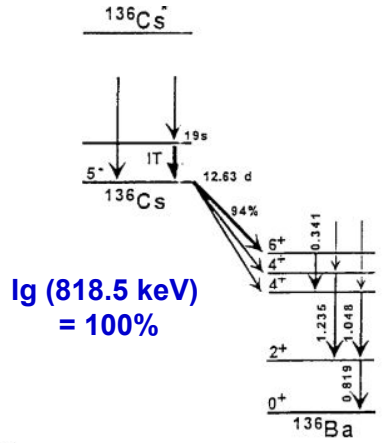
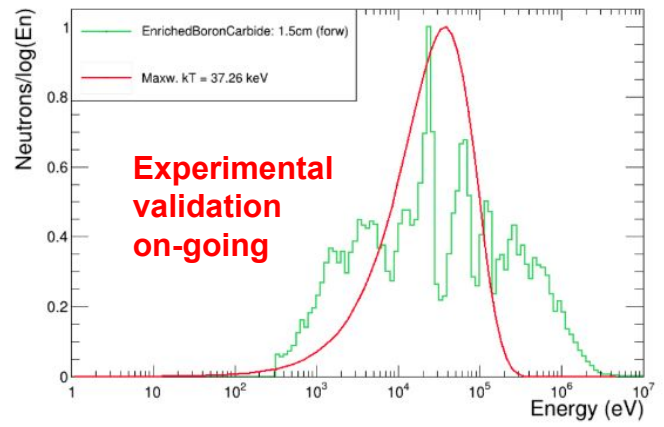


Figure 3: Simulated neutron flux with enriched  $\text{B}_4\text{C}$  filter 1.5 cm thick (normalized).

Figure 4: Decay scheme of  $^{136}\text{Cs}$ .

Cross section (mb)	n_TOF irradiation time	Activation measurement time	$\beta - \gamma$ coincidences at 818.5 keV	Neutron flux (neutrons/s/cm <sup>2</sup> )	$^{135}\text{Cs}$ atoms needed
$160 \pm 10$	40 days	39 days	200	$5.4 \cdot 10^7$	$2.4 \cdot 10^{15}$

7% uncertainty

Table 1: Summary of the data used for the calculation

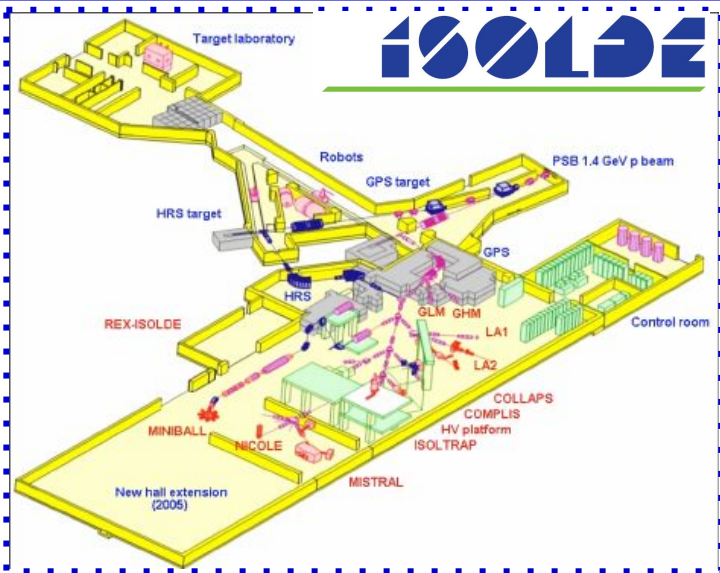
**HPGe Efficiency:**  
 @ 3 cm: 1.6% @818keV  
 x 20% Beta-gamma coincidence  
 (precise characterization ongoing + potential efficiency upgrade)

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## Why at ISOLDE?

- Yields  $1 \times 10^9 - 1 \times 10^{10}$  at/uC  $\rightarrow 10^{15}$  at  $^{135}\text{Cs}$  in several days to weeks
- Separation: GPS  $\rightarrow$  neighbouring masses  $<0.1\%$  +  $^{136}\text{Cs}$  decays in few months (Ba isotopes stable)
- **Foster ISOLDE-n\_TOF synergy: radioactive samples production + measurement at NEAR**



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ISOLDE Yield Database



ISOLDE Yield database: FLUKA

Molten La metal:  
8.5e9 at/ $\mu\text{C}$

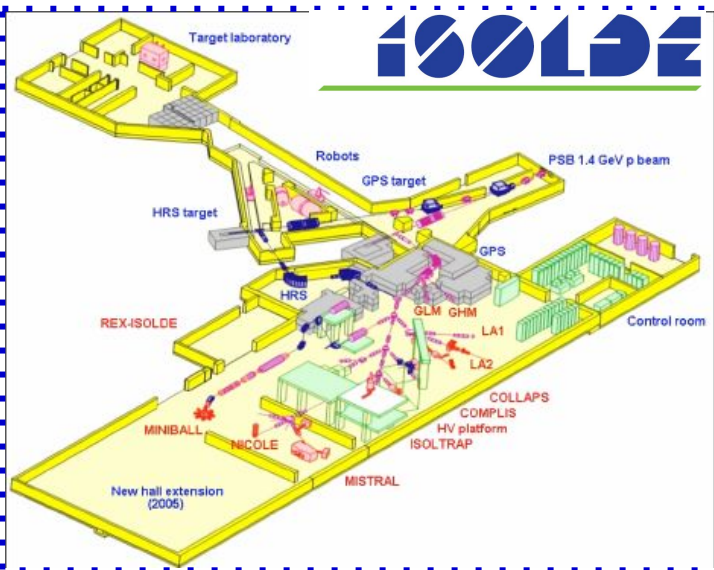
U carbide:  
2.4e9 at/ $\mu\text{C}$

La carbide:  
1.3e9 at/ $\mu\text{C}$

ISOLDE Yield Database



ISOLDE Yield Database



**ISOLDE Yields:  
FLUKA**

**1.5 uA (avg)  
protons**

**Extract eff.  
>=50%**

**To implant  
2.4e15 at**

Target	In-target (at/ $\mu$ C)	In-target (at/s)	Implanted (at/s)	Implanted (at/day)	Required Days <sup>a</sup>	Beam mode
Molten La	$8.50 \cdot 10^9$	$1.28 \cdot 10^{10}$	$6.38 \cdot 10^9$	$5.51 \cdot 10^{14}$	4.4	Exclusive
U carbide	$2.40 \cdot 10^9$	$3.60 \cdot 10^9$	$1.80 \cdot 10^9$	$1.56 \cdot 10^{14}$	15.4	Parasitic
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## Proposed production alternatives:

- 1) **U Carbide target "parasitic mode"** : in parallel to other running experiments one could collect in the SSP collection chamber (@ GLM or GHM beamlines). **15 days required for  $2.4 \times 10^{15}$  atoms.**
- 2) **U carbide target "parasitic mode" + offline GPS**: Old U Carbide target @ ISIS irradiation point at GPS until sufficient  $^{135}\text{Cs}$   $\rightarrow$  GPS (when available) & collection @ GLM or GHM. **Advantages: irradiation and collection separated in time**  $\rightarrow$  scheduling could be facilitated + ion beam would be optimized for the collection.

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- 3) **Molten La target**: Highest yield but less/not compatible to other experiments  $\rightarrow$  **exclusive shifts.**



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**x~2 lower yield**  
**→Unfavoured**

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## TECHNICAL DETAILS:

*Information: Ulli Köster*

### Beam-lines

- Low mass (GLM) or high mass (GHM) beam lines parasitic to HIE- ISOLDE experiments with In, Sn, Sb, Te, Cs or Ba beams in the central beamline

### Implantation matrix

- Should be conductive (e.g. Be, Al, C or metallized mylar, etc.)

### Sample size

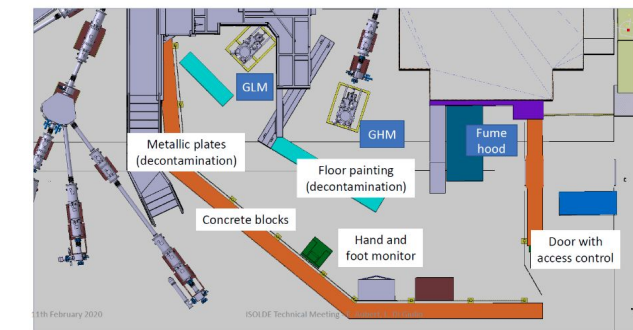
- Small diameter (about 3 mm without much losses) possible.
- Limit self-sputtering of the sample → spread the beam over an area of (cm<sup>2</sup>)+ adapt to n\_TOF optimum size

### Isotopic purity

- GPS mass separator: <0.1% of neighbouring masses
- <sup>136</sup>Cs: Sample cooled for > 10 x T<sub>1/2</sub> (13 d) → remove <sup>136</sup>Cs
- <sup>134</sup>Cs: kBq activities but strongest gamma lines of <sup>134</sup>Cs decay are below those of <sup>136</sup>Cs
- Cleaner spectrum with β-γ coincidences: <sup>136</sup>Cs decay g-rays emitted delayed (<sup>136m</sup>Ba isomer with T<sub>1/2</sub> = 0.3s)

### <sup>135</sup>Cs mass characterization

- Preliminary estimation: Beam current ( upper limit for the <sup>135</sup>Cs content due to the presence of isobaric <sup>135</sup>Ba)
- Final determination: After the n\_TOF experiment by dissolving the target + ICP-MS.



- Motivation:  $^{135}\text{Cs}(n,\gamma)$  cross section for nucleosynthesis and transmutation
- Project: Combining direct & surrogate reactions to measure  $^{134,135}\text{Cs}(n,\gamma)$
- $^{135}\text{Cs}(n,\gamma)$  activation measurement at NEAR
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## Motivation

- $^{134}\text{Cs}$  &  $^{135}\text{Cs}(n,\gamma)$ : relevant s-process branching  $\rightarrow$  s-only  $^{134,136}\text{Ba}$  (SiC presolar grains)
- $^{135}\text{Cs}(n,\gamma)$  ( $T_{1/2} = 2\text{e}6 \text{ y}$ )  $\rightarrow$  also relevant for transmutation of LLFP

## $^{135}\text{Cs}(n,\gamma)$ : NEAR + GEAR

- Previous MACS @ 30 and 500 keV (N. Patronis, FZK, 2004).
- Aim of the experiment at NEAR: Covering a wider energy range thank to the spectrum-shaping capability.
- Preliminary study: neutron spectra from MC simulations & experimental efficiency of the GEAR setup.
- Estimated minimum required mass of  $^{135}\text{Cs}$ :  $2.4 \times 10^{15}$  atoms

## Aim of the Proposal: $^{135}\text{Cs}$ sample production at ISOLDE

- Strengthen collaboration ISOLDE/MEDICIS for the production of future samples of interest for (n,g) at NEAR.
  - Technical feasibility and details have been discussed with ISOLDE experts.
  - **Options for production:**
1. Parasitic production/implantation (I): U carbide target while other experiments running  $\rightarrow$  15 days for the aimed number of atoms
  2. Alternative Parasitic approach (II):  $^{135}\text{Cs}$  in Old U carbide + offline extraction when GPS available
  3. Alternative exclusive shifts (III): 4 days with Molten metallic La target

## Outlook towards the $^{135}\text{Cs}(n,\gamma)$ at NEAR + GEAR

- Experimental characterization / validation of the NEAR “quasi-estelar” beams + upgrade of the GEAR setup
- Final beam-time estimation for the  $^{135}\text{Cs}(n,\gamma)$  via activation at NEAR + GEAR  $\rightarrow$  **INTC Proposal**

THANK YOU FOR  
YOUR ATTENTION!