

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of neutron capture cross section on ^{134}Cs through surrogate reaction $(d,p\gamma)$ at ISS

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Abstract: The neutron capture (n,γ) cross sections of ^{134}Cs and ^{135}Cs are relevant to fix the branching ratio between the two s-only ^{136}Ba and ^{134}Ba , well characterized from SiC in presolar grains. The $^{135}\text{Cs}(n,\gamma)$ reaction is also of interest for the potential transmutation of this long-lived ($T=2\cdot 10^6$ y) fission product. A new project has been launched to measure both $^{134,135}\text{Cs}(n,\gamma)$ by direct and surrogate methods. In this proposal, we present the method for the indirect measurement through the surrogate reaction $(d,p\gamma)$, and propose to measure the first of the two reactions, $^{134}\text{Cs}(d,p\gamma)$, using the Isolde Solenoidal Spectrometer and the SpecMAT scintillation array.

Summary of requested shifts: 25 shifts



1 Introduction

About 50% of the elements heavier than iron are produced by the so-called s-process, where the typical time scale of a neutron capture is longer than the half-life of β -decays of the unstable nuclei involved [1]. Asymptotic Giant Branch stars (AGB), and in particular those with mass lower than $3 M_{\odot}$ are responsible for the production of the main component of the s-process (i.e. nuclei from Sr to Bi).

In thermally-pulsing asymptotic-giant branch (TP-AGB) stars the main neutron source results from the reaction $^{13}\text{C}(\alpha, n)^{16}\text{O}$, whereas a weaker and recurrent contribution arises from the $^{22}\text{Ne}(\alpha, n)^{25}\text{Mg}$ reaction during the recurrent He-shell flashes. In the interpulse period between He shell flashes, a neutron density of less than 10^7 cm^{-3} is maintained in the ^{13}C pocket, a thin layer in the He shell, where the ^{13}C source operates under radiative conditions and at the relatively low temperatures of $k_B T = 8 \text{ keV}$. When a sufficiently high temperature is reached at the bottom of the He-burning zone, the ^{22}Ne source is activated during the following He shell flash. Though the total neutron exposure is smaller than contributed by the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction because the ^{22}Ne source is limited to a few years, much higher neutron densities of the order of 10^{10} cm^{-3} are reached at higher temperatures of $k_B T = 23 \text{ keV}$ during this highly convective instability.

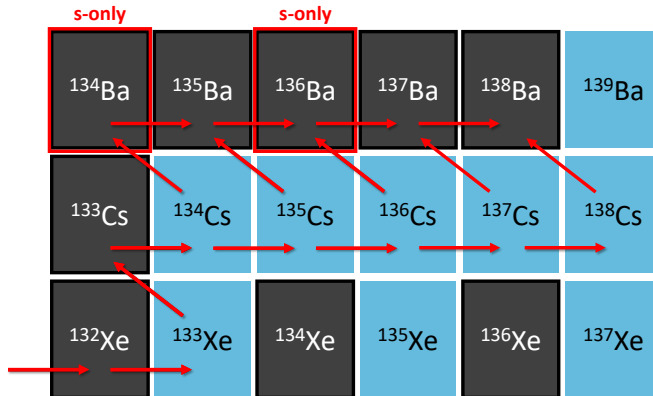


Figure 1: The s-process neutron capture flow through the Xe-Cs-Ba region. The stable isotopes are the ones in black.

In the s-process path, unstable nuclei that are sufficiently long lived such that neutron capture can compete with β decay act as branching points and yield a local isotopic pattern which is very sensitive to the physical conditions of the stellar environment [1]. ^{134}Cs and ^{135}Cs are both considered branching points of the s-process [1]. Moreover, as pointed out by Palmerini et al. [3], both the ^{134}Cs and ^{135}Cs branching points may have a temperature dependence, differently from what was assumed previously (Takahashi & Yokoi [4]): this makes them potential s-process thermometers to constrain the thermal conditions during the evolution of thermally-pulsing asymptotic-giant branch (TP-AGB) stars [1, 2]. The temperature dependence of ^{134}Cs half-life for β decay has been recently calculated by Taioli et al. [5] and is reported in Fig. 2.

The ratio between Ba isotopes is known from SiC crystals from presolar grains of AGB origin that were captured in pristine meteorites [3], giving us information on the isotopic

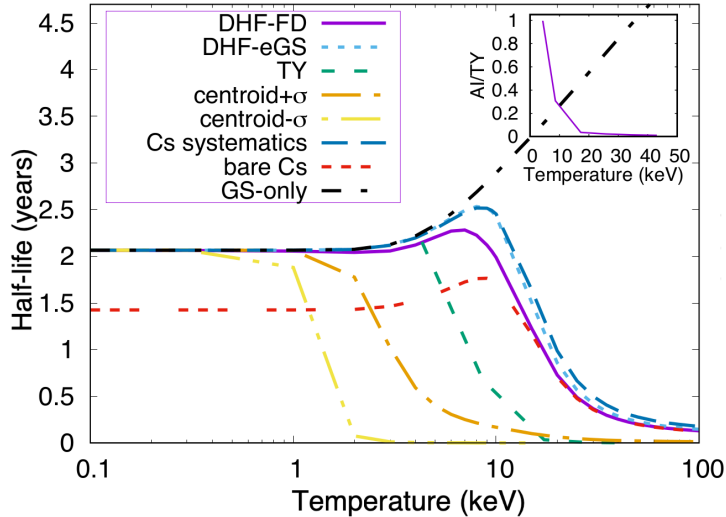


Figure 2: Half-life of ^{134}Cs vs. temperature, calculated in [5], using a variety of models.

composition during their formation. Thus, the data obtainable from these crystals together with the measured neutron capture cross sections of both ^{134}Cs and ^{135}Cs can constrain the thermal conditions in state-of-the-art AGB models.

Following the aforementioned scientific motivations, a collaborative project has been established to measure both $^{134,135}\text{Cs}(n,\gamma)$ cross sections combining direct measurements ($^{135}\text{Cs}(n,\gamma)$) at n_TOF-NEAR and surrogate reactions in inverse kinematics ($^{134}\text{Cs}(n,\gamma)$). Measuring the $^{134}\text{Cs}(n,\gamma)$ cross section using the surrogate reaction method would establish the possibility of performing this kind of experiment at ISOLDE. Furthermore, as a consequence of the short half-life of the nucleus and of the presence of γ radiation in its β decay, this cross section cannot be studied using presently available methods: on one side measurements at a n_TOF type facility are hindered by the γ activity of the sample, on the other side activation methods cannot be used because the activity of ^{134}Cs will dominate by many order of magnitude over the long lived ^{135}Cs produced in the activation process. The indirect measurement proposed here is thus the only possibility at present to obtain an experimental estimation of the neutron capture cross section.

2 Surrogate reaction method and reaction proposed

Since it is challenging to determine many astrophysical reaction rates due to the radioactivity of the isotopes involved, indirect approaches are becoming increasingly common. The $(d,p\gamma)$ reaction can be used as surrogate for (n,γ) to obtain the neutron capture cross section: the method for the indirect measurement and the theoretical framework are explained and tested in [11]. This method has also been tested by Ratkiewicz *et al.* in [12], where they were able to correctly reproduce the neutron capture cross section on ^{95}Mo , a stable isotope, in direct kinematics. We recently performed an experiment at Argonne National Laboratory to measure the $^{85}\text{Kr}(d,p\gamma)$ reaction. In this experiment we used a ^{85}Kr beam and the HELIOS magnetic spectrometer, while the γ -rays were detected

using the Apollo scintillator array. In this experiment we were able to detect proton- γ coincidences around the neutron separation energy of ^{86}Kr , and observe the γ -rays from the $2_1^+ - 0_1^+$ transition. The analysis is still on-going, but from the preliminary results it is possible to observe the γ transition in the matrix γ -ray energy vs excitation energy (Fig. 3).

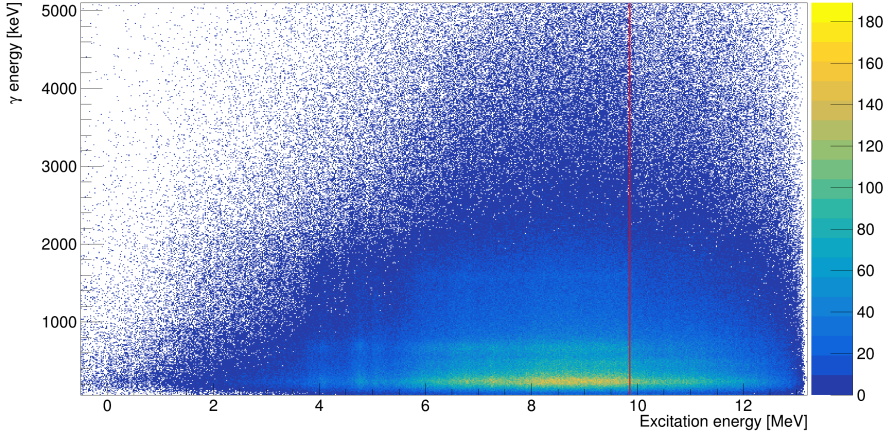


Figure 3: Preliminary result from the $^{85}\text{Kr}(d,p\gamma)$ experiment. In the upper panel: γ -ray energy vs excitation energy, the red line indicates the neutron separation energy in ^{86}Kr .

Therefore, we propose to perform a $(d,p\gamma)$ reactions using Isolde Solenoidal Spectrometer (ISS), with a ^{134}Cs beam with 7 MeV/u energy and a CD_2 target. The protons emitted will be detected with the Si array in ISS, while for γ -rays it is possible to use the CeBr_3 scintillator array from SpecMAT. The expected beam intensity at the ISS of ^{134}Cs is 10^7 pps, the same used for the $^{85}\text{Kr}(d,p\gamma)$ experiment. The neutron separation energy of ^{135}Cs is 8.76 MeV and the Q-value of the reaction is 6.54 MeV; therefore, it is possible to populate states around S_n with the silicon array placed backward, at a distance of 50 mm from the target, with a magnetic field of 2 T, as shown from the kinematics simulation in Fig. 4.

To estimate the statistics and the beam time needed, we calculated the expected cross section using the Distorted Wave Born Approximation (DWBA) with the finite-range code PTOLEMY [13]. The cross sections were integrated on the angular range accepted by the silicon array for the specific reaction: in the case of $^{134}\text{Cs}(d,p\gamma)$, the angular coverage is $0^\circ - 50^\circ$ in the center of mass frame; however, since the protons emitted between 0° and 10° are the one with energy lower than 1.5 MeV and thus are not used in the analysis because they mainly contribute to noise, the cross section has been integrated in the range $10^\circ - 50^\circ$. The ground state of ^{134}Cs has $J^\pi = 4^+$, so transfers with $\ell = 0$ or $\ell = 2$ can both populate a $9/2^+$ state. The cross section for populating a fictional $9/2^+$ state at 8.5 MeV excitation energy with a 7 MeV/u beam was found to be 2.23 mb and 10.97 mb, for $\ell = 0$ and $\ell = 2$ transfer, respectively. In total, the cross section is 13.2 mb. This result were compared to the one found for the case of the ^{85}Kr , in which $\ell = 0$ and $\ell = 2$ transfers to a fictional 4^+ state at 9.5 MeV were considered, finding 0.62 mb and 3.40 mb, for a total of 4.02 mb, in an angular range of $10^\circ - 45^\circ$ (found in the same way that in the

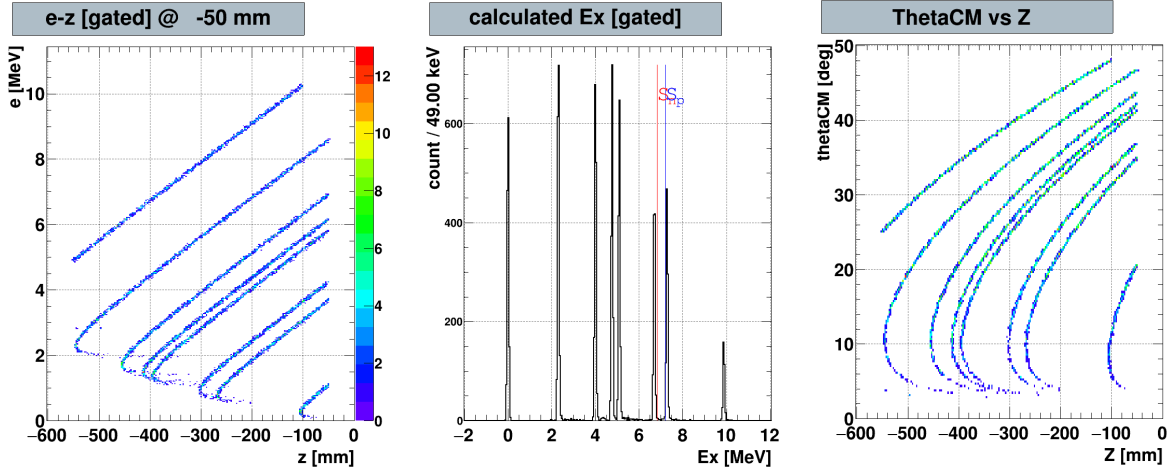


Figure 4: Kinematic simulation of the $^{134}\text{Cs}(d,p\gamma)$ reaction with the silicon array at 50 mm distance from the target, backward. On the left, proton energy vs z position. In the center, excitation energy of the product, ^{135}Cs . On the right: angle of emission of the proton in the center of mass vs z position.

^{135}Cs case). In that case, the experimental counts of proton- γ coincidences in a 500 keV range around 9.5 MeV excitation energy were $1.4 \cdot 10^4$ (Fig. 5), for 6 days of beam time. For estimating the expected experimental counts for the $^{134}\text{Cs}(d,p\gamma)$ reaction, the ratio of the cross sections and the ratio of the efficiencies of the arrays for γ detection has to be taken into account, while the efficiencies of the silicon arrays are considered to be similar. The γ -detection efficiency of Apollo is around 20%, while the efficiency of the SpecMAT array obtained from simulations [14] varies from 3% to 1.5% in an energy range between 0.5 MeV and 1.5 MeV; thus we considered a reduction of a factor of 10 in the expected statistics. The comparison between the geometry of Apollo and the one of SpecMAT is shown in Fig. 6. The expected counts in 6 days of beam time in a range of excitation energy of [8.25 MeV, 8.75 MeV] are $4.6 \cdot 10^3$.

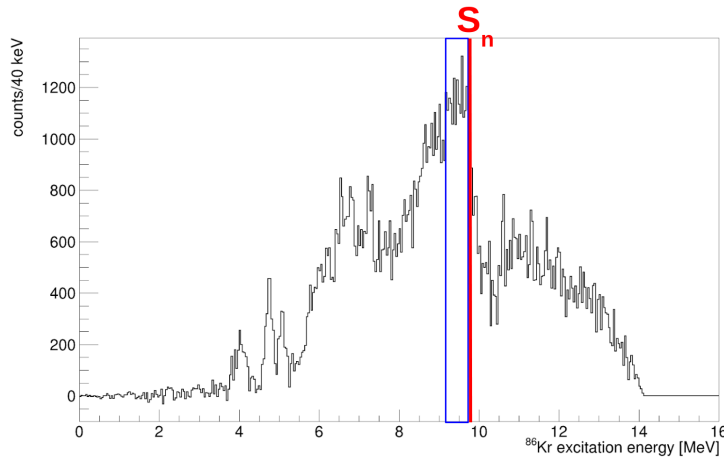


Figure 5: Excitation energy spectrum of ^{86}Kr after subtraction of the carbon background.

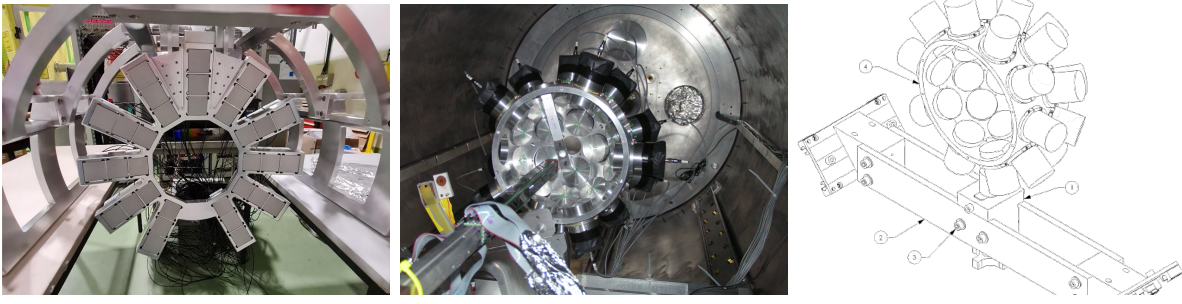


Figure 6: Comparison between the geometry of the SpecMAT array (on the left, from [14]) and Apollo (center and right).

Similarly to the measurement of the $^{85}\text{Kr}(d,p\gamma)$, the heavy recoils cannot be separated in angle from the unreacted beam and thus it is not possible to use a recoil detector. For this reason, the only way to remove the contribution from the carbon in the CD_2 target is to take some runs with a carbon target and later subtract it. This will require 2 additional days of beamtime.

Because ^{134}Cs is long-lived it is possible to perform the experiment during the winter physics campaign. Moreover, considering the existence in ^{134}Cs of a 8^- isomer with a lifetime of 4.2 hours, the running in the winter campaign will simplify the measurement as the isomer will be fully decayed to the ground state.

In summary, we ask for a total of 8 days of beam time, of which 6 days will be used for measuring on the CD_2 target and 2 days for the carbon target. One additional shift is considered for the preparation, for a total of **25 shifts**.

References

- [1] F. Käppeler et al., Review of Modern Physics, vol. 83, Issue 1, pp. 157-194 (2011).
- [2] N. Patronis et al., Phys. Rev. C 69, 025803 (2004).
- [3] S. Palmerini et al., Astrophys. J. 2021, 921, 7.
- [4] K. Takahashi et al., Atomic Data and Nuclear Data Tables, Vol. 36, p.375 (1987).
- [5] S. Taioli et al., Astrophys. J. 2022, 933, 158.
- [6] INTERNATIONAL ATOMIC ENERGY AGENCY, Implications of Partitioning and Transmutation in Radioactive Waste Management, Technical Reports Series No. 435, IAEA, Vienna (2004).
- [7] S. Chiba, T. Wakabayashi, Y. Tachi et al., Sci. Rep. 7, 13961 (2017).
- [8] Y. Shibahara, S. Nakamura, A. Uehara et al., J Radioanal Nucl Chem 325, 155–165 (2020).
- [9] S. Jaag, F. Käppeler, and P. Koehler, Nucl. Phys. A621, 247c (1997).
- [10] <https://isoyields2.web.cern.ch/InTargetProductionChart.aspx>
- [11] J. E. Escher et al., Phys. Rev. Lett. 121, 052501 (2018).
- [12] A. Ratkiewicz et al., Phys. Rev. Lett. 122, 052502 (2019).
- [13] M. H. Macfarlane and S. C. Pieper, ANL-76-11 Rev. 1, ANL Report (1978) and <http://www.phy.anl.gov/theory/ptolemy/>.
- [14] O. Poleshchuk, private communication.

3 Details for the Technical Advisory Committee

3.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

- Permanent ISOLDE setup: *ISS*
 - To be used without any modification
 - To be modified: The SpecMAT scintillator array is needed
- Travelling setup
 - Existing setup, used previously at ISOLDE
 - Existing setup, not yet used at ISOLDE
 - New setup

3.2 Beam production

- Requested beams:

Isotope	Production yield in focal point of the separator ($/\mu\text{C}$)	Minimum required rate at experiment (pps)	$t_{1/2}$
^{134}Cs	$7.66 \cdot 10^9$	10^7 pps	2.0648 y

- Full reference of yield information: yield database and, for the rate at the experiment, private communication by Alberto Rodriguez Rodriguez (15th November 2023)
- Target - ion source combination: Surface ion source with Th carbide target or La Molten target. Also surface ion source with U carbide target was used for other Cs isotopes with similar mass.
- RILIS? No
 - Special requirements:
- Additional features?
 - Neutron converter:
 - Other:
- Expected contaminants: Expected beam purity of 99% (private communication by Alberto Rodriguez Rodriguez)
- Acceptable level of contaminants: less than 5%
- Can the experiment accept molecular beams? No
- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? Yes, with IS721.

3.3 HIE-ISOLDE

- HIE ISOLDE Energy: 7 MeV/u;
 - Precise energy determination required
 - Requires stable beam from REX-EBIS for calibration/setup?
- REX-EBIS timing
 - Slow extraction
 - Other timing requests

3.4 Shift breakdown

Summary of requested shifts:

With protons	Requested shifts
Optimization of experimental setup using isotope ^{134}Cs	1
Data taking, isotope ^{134}Cs	24

3.5 Health, Safety and Environmental aspects

3.5.1 Radiation Protection

- If radioactive sources are required:
 - Purpose: calibration
 - Isotopic composition:
 - * ^{148}Gd , ^{239}Pu , ^{241}Am , ^{244}Cm (unsealed)
 - * ^{60}Co , ^{137}Cs , ^{22}Na (sealed)
 - Activity: from 1 kBq to 10 kBq
 - Sealed/unsealed: both
- Describe the hazards generated by the experiment:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	<input type="checkbox"/>	[pressure] [bar], [volume][l]
	Vacuum	<input type="checkbox"/>	
	Machine tools	<input type="checkbox"/>	
	Mechanical energy (moving parts)	<input type="checkbox"/>	
	Hot/Cold surfaces	<input type="checkbox"/>	
Cryogenic Safety	Cryogenic fluid	<input type="checkbox"/>	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	<input type="checkbox"/>	[voltage] [V], [current] [A]
	High Voltage equipment	<input type="checkbox"/>	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	<input type="checkbox"/>	[fluid], [quantity]
	Toxic/Irritant	<input type="checkbox"/>	[fluid], [quantity]
	Corrosive	<input type="checkbox"/>	[fluid], [quantity]
	Oxidizing	<input type="checkbox"/>	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	<input type="checkbox"/>	[fluid], [quantity]
	Dangerous for the environment	<input type="checkbox"/>	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	<input type="checkbox"/>	[laser], [class]
	UV light	<input type="checkbox"/>	
	Magnetic field	<input checked="" type="checkbox"/>	2 T
Workplace	Excessive noise	<input type="checkbox"/>	
	Working outside normal working hours	<input type="checkbox"/>	
	Working at height (climbing platforms, etc.)	<input type="checkbox"/>	
	Outdoor activities	<input type="checkbox"/>	
Fire Safety	Ignition sources	<input type="checkbox"/>	
	Combustible Materials	<input type="checkbox"/>	
	Hot Work (e.g. welding, grinding)	<input type="checkbox"/>	
Other hazards			