EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Activation measurements of the ^{135}Cs(n,γ) cross-section at n_TOF-NEAR

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Abstract:

The neutron capture (n,γ) cross sections of ^{134}Cs and ^{135}Cs are relevant for the *s*-process since they fix the branching ratio between the two s-only ^{136}Ba and ^{134}Ba, well characterized from SiC in presolar grains. The ^{135}Cs(n,γ) reaction is also of interest for the potential transmutation of this long-lived (T=2·10^{6} y) fission product. A new project has been launched to measure both ^{134,135}Cs(n,γ) by direct and surrogate methods. This proposal aims at the direct measurement of ^{135}Cs(n,γ) at the recently built n_TOF-NEAR facility, which thanks to its very high neutron flux, allows to experimentally access the (n,γ) cross section of unstable nuclei with very low mass samples.

Requested protons: 6.2×10^{18}.
Experimental Area: NEAR

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 about 3 $M_{\odot}$ are responsible for the production of the main component of the *s*-process (i.e. nuclei from Sr to Bi).
Along 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]. These two branchings fix the abundance ratio of the *s*-only ^{134,136}Ba isotopes, as shown in the left panel of Fig. 1. Moreover, as pointed out by Palmerini et al. [2], both the ^{134}Cs and ^{135}Cs branching points may have a temperature dependence, differently from what was assumed previously (Takahashi & Yokoi [3]): 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, 4]. The temperature dependence of ^{135}Cs half-life for β decay has been recently calculated by Taioli et al. [5] and is reported in the right panel of Fig. 1.
Analyses of presolar SiC grains of AGB origin found in pristine meteorites provide crucial information on ^{134}Ba and ^{136}Ba relative abundances produced during AGB nucleosynthesis [2]. Thus, presolar grain data for Ba isotope ratios 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.
Moreover, ^{135}Cs is among the long-lived products of Uranium fission, and so is present in radioactive waste coming from nuclear energy production. Due to its toxicity, half-life and concentration, it is one of the most important radionuclides in the context of radiological risk reduction [6]. One of the possible solutions for the problem of radioactive waste disposal is the partitioning and transmutation (P&T) of these long-lived fission isotopes into short-lived or stable nuclides. Transmutation could take place in nuclear reactors or in accelerator driven transmutation systems (ADS) [7]; however, cross sections for capture

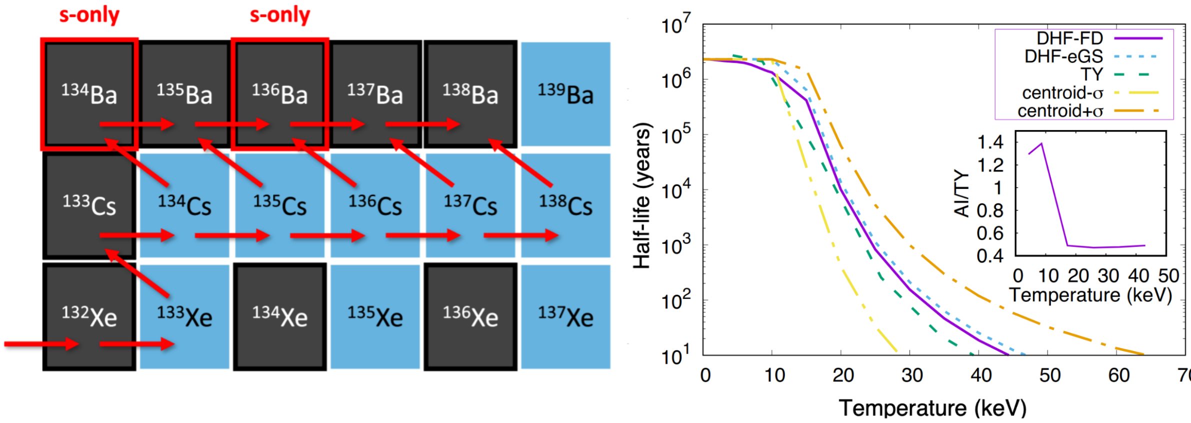

Figure 1: Left: The s-process neutron capture flow through the Xe-Cs-Ba region. The stable isotopes are the ones in black. Right: Half-life of ^{135}Cs vs. temperature, calculated in [5], using a variety of models. They all predict a sharp reduction of lifetime as soon as the stellar temperature reaches k_BT=20 keV.

of thermal neutrons are quite low, for this reason the use of fast neutron flux has been proposed. Nevertheless, the capture cross-sections for most of these isotopes are still not known, and their accurate measurement in the fast energy region (1 keV-1 MeV) is required to study the feasibility of transmutation and optimize the irradiation and cooling cycles [7].

Following the aforementioned scientific motivations, a collaborative project has been established to measure both ^{134,135}Cs(n,γ) cross sections combining direct measurements (^{135}Cs(n,γ)) at n_TOF-NEAR and surrogate reactions in inverse kinematics (^{134}Cs(n,γ)). Details on the indirect method can be found in Ref. [8].

Focusing in the measurement of ^{135}Cs(n,γ), the current knowledge of this cross section is still scarce and only a small number of experimental data exist because of the difficulty in obtaining a pure ^{135}Cs sample [9]. In the keV energy range, a first measurement of the MACS at k_BT= 25 keV was performed by activation at FZK [10], finding a result of 198 ± 17 mb. The same sample, that was produced by implantation and is no longer available, was used for another set of activation measurements of the MACS at k_BT= 30 keV (164 ± 10 mb) and k_BT= 500 keV (34.8 ± 3.0 mb), again in FZK [4].

A new set of activation measurements at the n_TOF NEAR would first confirm the previous results and, in addition, extend the measurement of the cross section to additional energies where presently there is no experimental information available [11, 12], thanks to the tunable energy of the neutron spectra at NEAR (see Sec. 4). This would allow to cover the whole range from 1 keV to 1 MeV of interest both for the main s-process nucleosynthesis (k_BT = 8 keV, 30 keV) and for transmutation in fast reactors.

In this proposal, we plan to measure the neutron capture cross section on ^{135}Cs at the new NEAR facility at n_TOF, using the activation method. In order to compare to previous measurements, we will perform an activation with an average energy of 30 keV for one of the runs. Furthermore, we propose to extend the range of previous measurements by performing another run at lower energy (8 keV).

2 The n_TOF NEAR station

The new n_TOF NEAR station, located very close ($\sim$ 3 m) to the spallation target, features an extremely high neutron fluence which makes it particularly well suited for the measurement of very small mass samples of radioactive nuclei [13].

Detailed **Monte Carlo simulations** have been performed to determine the neutron flux at the irradiation station for activation measurements at NEAR (a-NEAR) [15]. The simulations indicate that with a suitable choice of filter, the energy distribution of outgoing neutrons can be shaped to a Maxwellian-like neutron spectrum at stellar temperatures (from a few keV to several hundreds of keV) [15]. This is validated by the **commissioning runs** performed at the NEAR facility that has made significant progresses. The preliminary experimental results of the neutron flux in the activation position carried out by means of the multi-foil activation and the neutron slowing-down spectrometry techniques agree with the simulations with a deviation smaller than 20% [16]. The simulations have been further validated with the measurement of the off-beam neutron background with CR39 dosimeters [17].

A **first experiment** [18] using the same experimental setup was performed at the NEAR facility to measure Maxwellian averaged neutron capture cross section (MACS) at stellar temperatures using the activation method. After the irradiation the target has been moved to the GEAR decay station [15], which was already fully characterized and employed for activation measurements in the previous benchmark campaign [18]. The measurement has been successfully completed and is currently under analysis.

3 ^{135}Cs sample preparation

The target for this proposal will be produced in the first months of 2024 by implantation at ISOLDE, as proposed in [14], and it is expected to have around $2.4\cdot10^{15}$ atoms.

A total of 13 shifts with the molten La target should in principle allow to collect the required number of atoms. Cs is efficiently released and surface ionized from the target. After mass separation using the General Purpose Separator (GPS), leading to contributions of neighboring masses $<$0.1%, it will possible to collect ^{135}Cs in the GLM or GHM beam lines. We note that ^{135}Cs beams may be accompanied by stable ^{135}Ba isobars, but this is not disturbing for the planned activation experiment since neutron capture on ^{135}Ba does not form long-lived radionuclides.

The sample will be implanted in a backing of tenths of μm thick of aluminium or graphite, as in previous n_TOF targets implanted in ISOLDE [19]. The backing will be glued to an Al frame, 30 mm in diameter compatible with the B_4C filters available at NEAR. To limit self-sputtering of the sample, we will spread the beam to make an area of about 2x2 cm^2, still smaller than the spot of maximum flux in the activation position at NEAR.

According to our estimates, the potential contaminants of the implanted sample include some ^{134}Cs tailing into the collection with an expected activity below the kBq. This ^{134}Cs activity should not affect the decay measurement at GEAR since the ^{134}Cs decay lines are at lower energies than those of ^{136}Cs. Moreover, applying $\beta - \gamma$ coincidences, as discussed in Sec. 4, would also help to clean this background considerably.

4 Experimental setup and required beam time

The activation experiments of this proposal require an irradiation to be performed using the neutron beam of NEAR, followed by a decay measurement where the activated ^{136}Cs nuclei will be quantified.
As for the irradiation, the neutron beam will be shaped using the same neutron filtering assembly developed for previous campaigns and shown in Figure 2. The sample will be encased within two disks made of boron carbide (B_4C), ≥95% enriched in ^{10}B. Each cylinder is 60 mm in diameter. The B_4C disks currently available at NEAR can be assembled so that the total thickness on each side of the target (T in Fig. 2) varies from 0.5 mm to 10 mm. As it is shown by the results of the previous campaign [20] and confirmed using Monte Carlo calculations, thicker filters would be required to obtain neutron spectra with an average temperatures above 10 keV.

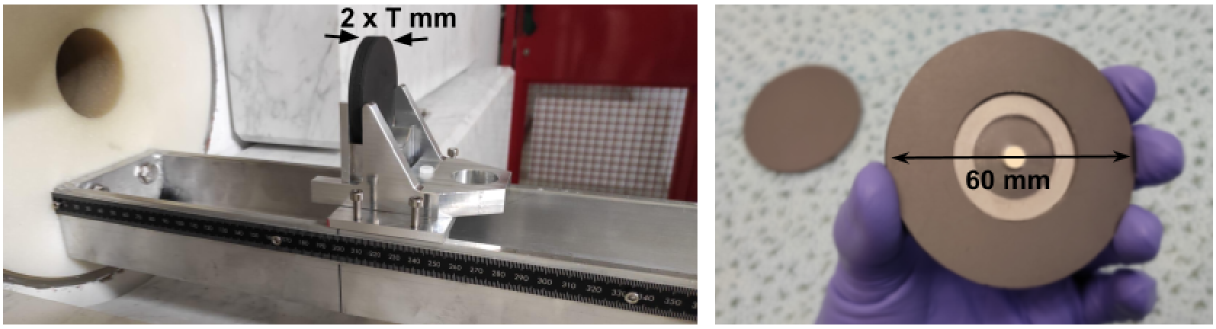

Figure 2: Left: Activation setup at NEAR with the neutron beam filtering assembly. Right: Opened B_4C filter showing the irradiated sample inside.

The energy distribution and magnitude of the neutron flux at the sample position has been obtained from GEANT4 simulations that included on both sides of the sample a sandwich of Boron carbide filters (B_4C) 95% enriched in ^{10}B. The neutron flux has been extracted from neutrons impinging the target in the most central part with a radius of 5 cm. Different filter thicknesses were simulated to resemble the actual setup in Fig. 2 and the expected flux at different average neutron energies was calculated.
As we introduced before, two irradiations have been considered in this proposal corresponding to two different average energies, 30 and 8 keV. For the first one, we assumed the simulated neutron flux obtained with a B_4C thickness of 1.5 cm, which provided the closest average neutron energy to 30 keV. The resulting logarithmic neutron flux is shown is Fig. 3. Assuming an average intensity of the proton beam of $1.3 \cdot 10^{12}$ protons/s, the average flux is expected to be $\Phi = 5.4 \cdot 10^7$ neutrons/cm^2/s. As for the ^{135}Cs(n,γ) cross section, we assumed the MACS at 30 keV from KADoNiS $\sigma = 160 \pm 10$ mb [21]. The same calculation has been repeated for the irradiation with an average energy of 8 keV. In this case, the MACS reported in KADoNiS is 382 mb [21]. The filter thickness that provides the closest average temperature is 12 mm, for which the expected average flux is $\Phi = 6.0 \cdot 10^7$ neutrons/cm^2/s.
During the decay measurement we will identify the 818.5 keV gamma-ray from the de-excitation of the first 2^+ state in ^{136}Ba that is populated with a branching of 99.7% in the

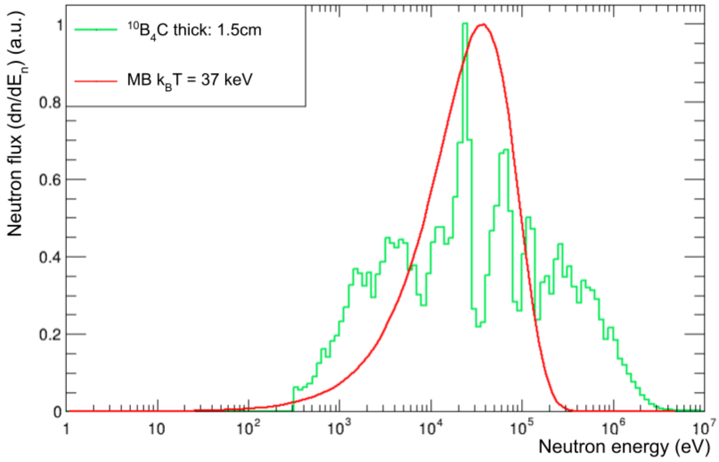

Figure 3: Simulated neutron flux filtered with an enriched B_4C filter 1.5 cm thick.

decay of ^{136}Cs. For the beam-time estimates we have assumed that the HPGe detector currently present in the GEAR station will be used. The efficiency of this detector has been characterized at different distances from the sample to the detector [15]. For the estimate presented in the proposal, the closest distance of 3 cm has been considered, where $\varepsilon = 1.6\%$ at 818.5 keV. This estimate may be conservative since the sample could be placed even closer to the detector and the setup could be upgraded in the future.
Considering the target described in Sec. 3, containing $2.4 \cdot 10^{15}$ atoms of ^{135}Cs, the fluxes obtained from the simulation and the efficiency of the GEAR setup, we calculated the irradiation time needed to obtain 1000 counts in the photo-peak after a measurement of the activated samples of three times the half-life of ^{136}Cs. The latter provides a good trade off between statistics and signal-to-background ratio. The irradiation time needed at NEAR to achieve these counting statistics is, assuming 10^{17} protons per day, 40 days for the average energy of 30 keV. In the case of the irradiation at 8 keV, due to the larger flux and MACS compared to KT = 30 keV, 20 days of irradiation would be needed.
To reduce the background to negligible level the HPGe will be operated in coincidence with the available plastic cylindrical BC408 detector of 4 cm in diameter and 10 mm thick for the detection of beta particles. A special sample holder setup is foreseen so that the sample will be "sandwiched" between the HPGe and the plastic detectors at close geometries [15]. Taking into account a reasonable 20% efficiency in $\beta - \gamma$ coincidence the expected coincidences in the peak at 818.5 keV are 200 without significant background and this will lead to a 7% contribution to the uncertainty due to Poisson statistics.
Considering all the factors listed above, summarized in Table 1, the number of protons requested for the two irradiations of the ^{135}Cs sample at NEAR are $4.0 \cdot 10^{18}$ and $2.0 \cdot 10^{18}$, respectively for the spectra centered at about 30 and 8 keV. In this proposal, the MACS of ^{135}Cs will be from the experimental SACS in reference to that of ^{197}Au, since both the stellar neutron capture cross section of ^{197}Au [22] and the parameters of the ^{198}Au decay are accurately known. For this purpose, a Au sample of the same dimensions of the implanted ^{135}Cs target will be irradiated exactly in the same position. This requires $1.0 \cdot 10^{17}$ protons for each spectra. All the irradiations will run in parasitic mode to the

experiments being carried out in EAR1 and EAR2.

Average kT	Cross section (mb)	^{135}Cs atoms	Activation measurement time	$\beta-\gamma$ coincidences at 818.5 keV	Neutron flux (n/s/cm^2)	n_TOF beam time
30 keV	160 ± 10	$2.4 \cdot 10^{15}$	39 days	200	$5.4 \cdot 10^7$	40 days
8 keV	382 ± 10	$2.4 \cdot 10^{15}$	39 days	200	$6.0 \cdot 10^7$	20 days

Table 1: Summary of the data used for the estimation of the beam time request.

References

[1] F. Käppeler et al., Rev. Mod. Phys. **83(1)**, 157-194 (2011).

[2] S. Palmerini et al., Astrophys. J., **921**, 7 (2021).

[3] K. Takahashi et al., Atomic Data and Nuclear Data Tables **36**, 375 (1987).

[4] N. Patronis et al., Phys. Rev. C **69**, 025803 (2004).

[5] S. Taioli et al., Astrophys. J., **933**, 158 (2022).

[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] A. Ratkiewicz et al, Phys. Rev. Lett. **122**, 052502 (2019).

[9] Y. Shibahara, S. Nakamura, A. Uehara et al., J. Radioanal. Nucl. Chem. **325**, 155–165 (2020).

[10] S. Jaag, F. Käppeler, and P. Koehler, Nucl. Phys. A **621**, 247 (1997).

[11] C. Domingo-Pardo et al., Eur. Phys. J. A **59**, 8 (2023).

[12] V.V. Zerkin, B. Pritychenko, Nucl. Inst. and Methods A **888**, 31-43 (2018).

[13] A. Mengoni et al.,CERN-INTC-2020-073 (2020).

[14] J. Lerendegui-Marco, S. Carollo, CERN-INTC-2022-040 (2022).

[15] N. Patronis et al., EPJ-C (submitted), arXiv e-prints (2022): arXiv:2209.04443

[16] P. Torres, M. Mastromarco, P. Perez-Maroto et al., n_TOF Collaboration & Analysis Meeting, 25-26 May 2023. https://indico.cern.ch/event/1279454/contributions/

[17] J. Lerendegui-Marco, M. Bacak et al., n_TOF Collaboration & Analysis Meeting, 25-26 May 2023. https://indico.cern.ch/event/1279454/contributions/5405357/

[18] E. Stamati, A. Manna et al., CERN-INTC-2022-008 (2022).

[19] E.A. Maugeri et al., Nuclear Instruments and Methods A **889**, 138-144 (2018).

[20] E. Stamati et al., n_TOF Collaboration General Meeting, 22-24 November 2023. https://indico.cern.ch/event/1309447/timetable/

[21] https://www.kadonis.org/

[22] C. Lederer et al., Phys. Rev. C **83**, 034608 (2011).

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
If relevant, write here the name of the fixed installation you will be using: irradiation setup at NEAR, HPGe and beta plastic detector at the GEAR decay station present at the n_TOF installation	☒ To be used without any modification ☐ To be modified
If relevant, describe here the name of the flexible/transported equipment you will bring to CERN from your Institute: None	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]

Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			