EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Production of a ^{135}Cs sample at ISOLDE for (n,γ) activation measurements 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=$2\cdot10^6$ y) fission product. A new project has been launched to measure both ^{134,135}Cs(n,γ) by direct and surrogate methods. The direct measurement of ^{135}Cs(n,γ) is foreseen 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. A crucial factor for the success of these measurements, is the production of a high quality target. In this context, this proposal aims at the production of a high purity ^{135}Cs sample of about $2{\cdot}10^{15}$ atoms by means of implantation using the ISOLDE beams and the general purpose separator (GPS).

Requested shifts: Parasitic (different options are proposed)

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 of thermal neutrons are quite low, for this reason the use of fast neutron flux has been

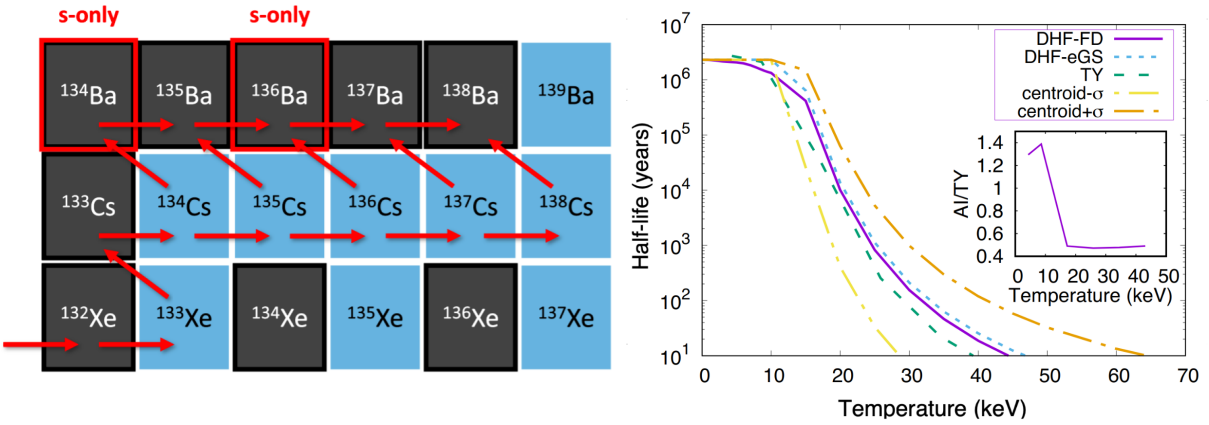

Figure 1: Left: The *s*-process neutron capture flow through the Xe-Cs-Ba region. The stable isotopes are the ones in black. Right: Half-life of ^{135}Cs vs. temperature, calculated in [5], using a variety of models. They all predict a sharp reduction of lifetime as soon as the stellar temperature reaches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

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 (^{134,135}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. 2). 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, 25 keV) and for transmutation in fast reactors. Extending the energy range in the direct measurement of the ^{135}Cs(n,γ) cross section is also of interest for the validation of the surrogate method, which typically provides a coarse resolution of 80-100 keV. The indirect method is the only one currently available to experimentally access the ^{134}Cs(n,γ) cross section.

In neutron induced measurements on radioactive nuclei, the quality of the target (mass, purity, homogeneity) is crucial for the success of the experiment. In this proposal we

propose the production of a high-purity sample of ^{135}Cs using the beams of ISOLDE. To justify the proposal, Sec. 2 presents the feasibility study for the experiment at NEAR and quantifies the minimum number of ^{135}Cs atoms required. The details of the proposed sample production and implantation at ISOLDE are described in Sec. 3.

2 ^{135}Cs(n,γ) at n_TOF NEAR and required mass

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) [14]. With a suitable choice of filter, the energy distribution of out-coming neutrons can be shaped to a Maxwellian-like neutron spectrum at stellar temperatures (from a few keV to several hundreds of keV) [14]. The first benchmark of the feasibility to measure Maxwellian averaged cross section (MACS) at stellar temperatures in this facility is currently ongoing [15].
The n_TOF collaboration has already expressed the interest for this scientific proposal and is willing to provide the resources and beam time needed. However, the accurate beam time estimates require first from the experimental characterization of the filtered neutron fluxes at a-NEAR and the validation of the simulations [15]. Moreover, additional work in the GEAR decay station is required to optimize its efficiency. For these reasons, the n_TOF proposal to measure ^{135}Cs(n,γ) at NEAR will be submitted in a future INTC call, once the facility and the decay setup are fully characterized experimentally, and the sample has been successfully produced.
This section describes the main characteristics of the future activation experiment at NEAR on the basis of the first MC simulations and discusses the minimum number of ^{135}Cs atoms that should contain the sample. For the calculation presented herein the ^{135}Cs(n,γ) cross section considered is the MACS at 30 keV from KADoNiS $\sigma = 160 \pm 10$ mb [16]. The neutron capture cross section will be measured using the activation method. After the irradiation at the new NEAR facility, the target will be moved to the GEAR decay station [14].
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 registered over a scorer with a radius of 20 cm. However, taking into account the smaller dimension of the samples, only the central part of the scorer with a radius of 5 cm has been considered. Different filter thicknesses were simulated to model the expected flux at different average neutron energies. For this calculation, we took the 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. 2. From the output of the simulations and assuming an average power of the proton beam of $1.3 \cdot 10^{12}$ protons/s, the average flux is found to be $\Phi = 5.4 \cdot 10^7$ neutrons/cm^2/s. The irradiation time considered is 40 days, which corresponds to about three times the half-life of ^{136}Cs, since reaching saturation ($\sim 5\tau$) will request a too long time and will not improve significantly the

statistics.

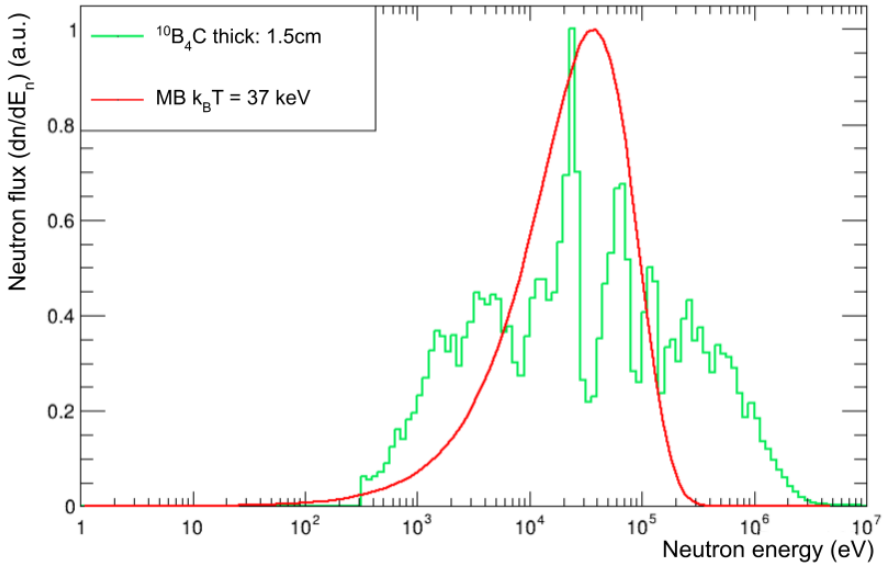

Figure 2: Simulated neutron flux filtered with an enriched B_4C filter 1.5 cm thick.

Concerning the measurement of the activated sample, ^{136}Cs β decays with 100% probability to the excited states of ^{136}Ba. To quantify the number of neutron captures to ^{136}Cs we will detect the γ-rays from the de-excitation of ^{136}Ba produced in the β-decay of ^{136}Cs, in particular the γ from the first 2^+ state to the ground state at 818.5 keV. In order to have good statistics, we required a minimum of 1000 counts in the photo-peak.
The efficiency of the HPGe detector installed in the GEAR station has been characterized at various distances of the sample [14]. For the estimate presented in the proposal, the closest distance of 3 cm has been considered, where $\epsilon = 1.6\%$ 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. Operating the HPGe in coincidence with a β detector can be used to reduce the background to negligible level. To take into account the efficiency in $\beta - \gamma$ coincidence we assumed a factor 5 of reduction of the counts observed in the main peak.The half-life of ^{136}Cs is 13 days, so we propose to measure its decay for 39 days, that corresponds to 3 half-lives. The expected coincidences in the peak at 818.5 keV are 200 without significant background and this will lead to an statistical uncertainty of 7% according to Poisson statistics.

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

Table 1: Summary of the data used for the estimation of the minimum ^{135}Cs mass.

Considering all the factors listed above, the number of atoms of ^{135}Cs needed in the target is $2.4 \cdot 10^{15}$. All the data reported are summarized in Table 1.

3 Production of a ^{135}Cs sample at ISOLDE

Different production mechanisms for the ^{135}Cs sample have been discussed, being the production and implantation at ISOLDE the preferred one for various reasons. First, at ISOLDE we will be able to implant the aimed mass in a reasonable time. Moreover, thanks to the mass separation, a high isotopic purity can be achieved that will reduce the background in the measurement of the ^{136}Cs decay at GEAR. Last, it will help also to foster the ISOLDE-n_TOF synergy in terms of producing radioactive samples, and subsequently measuring them at the neighbouring and recently built NEAR facility.
In order to estimate the number of required shifts to implant $2.4 \cdot 10^{15}$ atoms, we have taken into account the following factors. ^{135}Cs can be produced with reasonably high yields at ISOLDE using various targets. For the aim of this proposal, the calculated in-target production rates can provide a good guidance [17]. According to the FLUKA calculations, the best production rate of 8.5×10^9 at/μC would be obtained with the molten La metal target. By using the more common U Carbide (UCx) target, 2.4×10^9 at/μC would be produced. Then, assuming a release and ionization efficiency of at least 50% for long-lived Cs isotopes and a proton current of 1.5 μA on target averaged over time, one would need 4 and 15 days to produce the aimed number of atoms with the molten La target and UCx target, respectively. Recent yield measurements of long-lived Cs isotopes (132,134,136) from UCx and LaCx targets showed overall efficiencies of 60% to 100% with respect to the FLUKA estimates, i.e. the assumed 50% is conservative. The estimated yields, implantation rates and required days are summarized in Table 2.

Target	In-target (at/μC)	In-target (at/s)	Implanted (at/s)	Implanted (at/day)	Required Days[a]	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
La carbide	$1.30{\cdot}10^9$	$1.95{\cdot}10^9$	$9.75{\cdot}10^8$	$8.42{\cdot}10^{13}$	28.5	Parasitic

Table 2: Possible options for the ^{135}Cs sample production at ISOLDE. The preferred options, that will allow the implantation in parasitic mode to other running experiments, use the U carbide target (more details in the text). a) To implant $2.4{\cdot}10^{15}$ atoms.

While a run with a molten La target should in principle allow to collect the required activity in about 13 shifts (see Table 2), this would require a dedicated target for this experiment (molten La targets are not frequently used at ISOLDE) and it carries the risk that this target could fail before reaching the required beam dose. Other molten metal targets are operated in a special mode (STAGISO) with lower proton beam intensity to prevent obstructing the chimney to the ion source by "splashes" and such a careful approach might be required for the molten La target too.

Operation of UCx and LaCx targets is more predictable and such targets are more frequently scheduled at ISOLDE. The use of UCx is favoured over LaCx due to its twice higher ^{135}Cs yield (see Table 2). Cs is efficiently released and surface ionized in any Nb/Ta/W/Re cavity, i.e. present as beam whenever no specific counter-measures (quartz transfer line or LIST) are applied. Hence, whenever another ISOLDE experiment employs beams of In, Sn, Sb, Te, Cs or Ba with RILIS or surface ionization and a mass A=115-132 or A=139-154 is sent to the central beam line, it is possible to collect ^{135}Cs in parallel in the GLM or GHM beam line respectively. Such a "parasitic mode" is particularly efficient when the central mass is kept constant for several days, typically during HIE-ISOLDE experiments. Multiple collections might be necessary until the desired sample mass of 2.4×10^{15} atoms is reached. 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.
An alternative approach would consist on irradiating an old UCx target at the ISIS irradiation point at GPS until the required number of ^{135}Cs atoms has built up in the target. Then, whenever GPS is not in use, the sample could be collected at GLM or GHM. In this way, irradiation and collection could be separated in time and scheduling could be facilitated and the ion beam would be optimized for the collection rather than delivery to HIE ISOLDE experiments.
As for the implantation, the matrix will be made of a conductive material such as Be, Al, C or metallized mylar. To limit self-sputtering of the sample, it would be preferential to spread the beam over an area of few cm^2. This requirement matches well with the sample size required for the future activation measurement at NEAR. The final geometry of the sample to be irradiated at a-NEAR is still to be decided. After the sample is produced, the ^{135}Cs mass will be characterized. The current will provide the first upper limit on the number of implanted ^{135}Cs atoms but it is affected by the presence of the isobaric ^{135}Ba background. The precise determination of the ^{135}Cs content will be performed after the n_TOF experiment by dissolving the target and carrying out and ICP-MS measurement. A sufficiently high isotopic purity will be achieved with the General Purpose Separator (GPS), expecting contributions of neighboring masses $<0.1\%$. To further reduce the impurity of ^{136}Cs ($T_{1/2}$ = 13 d), we will wait sufficiently long time before the experiment to ensure that its concentration has decayed to a negligible level. Last, there might also be 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. 2, would also help to clean this background considerably.

In conclusion, to reach the minimum number of $2.4{\cdot}10^{15}$ ^{135}Cs atoms needed for the activation measurement at n_TOF-NEAR, we ask for a total of 15 days of ^{135}Cs beam using the U carbide target and running in parasitic mode to other experiments. Other alternatives have been also mentioned to facilitate the scheduling at ISOLDE and the compatibility to other experiments.

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., EPJ-A (submitted), arXiv e-prints (2022): arXiv:2208.02163.

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

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

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

[17] https://isoyields2.web.cern.ch/InTargetProductionChart.aspx

DESCRIPTION OF THE PROPOSED EXPERIMENT

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

Part of the experiment	Design and manufacturing
Implantation to be carried out at the in the SSP collection chamber at GLM/GHM beamlines, fixed ISOLDE installation.	☒ To be used without any modification ☐ To be modified
No flexible or transported equipment will be brought to the CERN site.	☐ 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: No 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			