EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

First measurement of the s-process branching ^{79}Se(n, γ)[1]

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Abstract

Selenium-79 is a key branching point in the slow neutron capture process (s-process) with relevant implications in nucleosynthesis. The products of the s-process nucleosynthesis after ^{79}Se are the s-only isotope ^{80}Kr and ^{82}Kr, whose abundance ratio is well characterized in presolar grains. This information, together with an accurate knowledge of ^{79}Se(n,γ) cross section will allow one to extract reliable constraints for the s-process site conditions, in particular the stellar temperature. This proposal aims at the first measurement of the neutron capture cross section of this key branching nucleus, that thus far could not be measured due to limitations in sample mass and detection sensitivity. Such a challenging measurement shall become now feasible, thanks to the high resolution and neutron luminosity of n_TOF EAR1 and EAR2 respectively, a special PbSe eutectic-alloy sample with 3 mg of ^{79}Se specifically produced for this experiment, and a new measuring technique and apparatus with enhanced sensitivity developed in the framework of an ERC Grant. Due to the relevance and challenge of this experiment, a careful study of its feasibility, including a detailed estimation of the expected results and astrophysical impact has been carried out.

Requested protons: 6×10^{18} protons on target
Experimental Area: EAR1 and EAR2

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1.- Motivation

The unstable ^{79}Se (terrestrial half-life $t_{1/2}$ = 3.27(8)×10^5 years [1]) represents one of the most relevant and debated s-branching nuclei [2] and the accurate knowledge of its capture cross section could provide a crucial test for the understanding of s-process nucleosynthesis both in massive stars (MSs) [3] and in asymptotic giant branch (AGB) stars [4,5]. In this respect, the ^{79}Se branching is particularly interesting because it is located in the transition region between weak (MSs) and the main (AGBs) s-process. The s-process path bifurcates at ^{79}Se due to the competing action of neutron capture and beta decay of this isotope [6]. Thus, a detailed knowledge of the ^{79}Se capture cross section is fundamental to fix the branching ratio to ^{82}Kr (neutron capture) or to ^{80}Kr (beta decay). In particular, the abundance of **the s-only ^{80}Kr, shielded from any contribution from the rapid neutron capture process (the r-process) by its stable isobar ^{80}Se, depends directly on such branching ratio**.

Recently, Prantzos et al. found an underproduction of ^{80}Kr in their Galactic Chemical Evolution model, compared to the abundances observed in the Sun [7]. The reason for such a discrepancy is probably related to the limited accuracy of the nuclear inputs. Therefore, a new measurement aiming at improving our knowledge of ^{80}Kr nucleosynthesis is highly desirable. Additionally, the Kr isotopic ratios have been measured in bulk SiC acid residues [4,5], providing details on AGB stars evolved prior to the formation of the Solar System. **Presolar grain measurements give the most precise data currently available on s-process nucleosynthesis (at least one order of magnitude better than spectroscopic observations)** and together with experimental cross sections can yield the most sensitive constraint for stellar models. The branching at ^{79}Se is particularly well suited for determining the thermal conditions of the stellar environment thanks to the strong thermal dependence of the beta decay rate of this isotope [8].

A recent Monte Carlo reaction rate variation study concluded that ^{79}Se(n,ɣ) is a key reaction in several investigated s-process nucleosynthesis models [9]. Moreover, calculations with the FUNs evolutionary code [10] to determine the sensitivity of the Kr isotopic ratios by variations of the ^{79}Se neutron capture, indicate that the stellar yields of ^{80}Kr and the ^{80}Kr/^{82}Kr abundance ratio vary in ±30% with the current uncertainty of a factor 2 in the cross section (see App. 4). The latter is representative of the dispersion in the theoretical MACS values obtained so far [6], although the true uncertainty could be significantly larger in view of recent estimates from indirect methods [11]. These sensitivity studies prove that, **once the ^{79}Se(n,γ) cross section has been measured, the temperature (and the effective half-life) could be accurately constrained by comparing the ^{80}Kr/^{82}Kr ratio calculated with stellar models and the experimental observations [4,5].**

Beyond the astrophysical motivation, the capture cross section of ^{79}Se is also of interest for nuclear transmutation studies because **^{79}Se is one of the main contributors among the fission products to the long-term radiotoxicity of spent fuel** due to its long half-live [12-14].

Despite of the high relevance of the ^{79}Se(n,ɣ) cross section, **no experimental data is available to date**. Evaluations and the Maxwellian-Averaged Cross Section (MACS) in KaDoNiS [6] are fully based on theoretical calculations. For the particular case of ^{79}Se(n,ɣ), direct measurements of the MACS via neutron activation are not feasible since the product nucleus ^{80}Se is stable. Hence, the time-of-flight technique is the only option available. Moreover, the reduced mass of this sample and its high activity make it a **unique case for CERN n_TOF, as demonstrated by the successful measurements of other high impact s-process branching isotopes** (^{63}Ni [15], ^{93}Zr[16,17], ^{151}Sm[18], ^{171}Tm[19], ^{204}Tl[20]). The feasibility of the proposed ^{79}Se(n,ɣ) measurement has been compared with the most challenging ones (see Ref. [21]).

In this context, a **Letter of Intent (LoI)** was already proposed to the Isolde and n_TOF Committee at CERN (INTC) [22]. After approval of this LoI, a high-quality sample has been prepared in collaboration with ILL, PSI and CERN. Also the novel detection system i-TED[23] has been developed [24-27] in the framework of the ERC-project HYMNS [28] to enhance the detection sensitivity to a level [29] that allows one to tackle for the first time a capture measurement of this isotope, which is the main objective of this proposal.

2.- Sample production and characterization

The ^{79}Se sample has been produced by means of irradiation of a ^{78}Se sample in the high-flux reactor at ILL. For the neutron irradiation a Pb-Se eutectic alloy was prepared at PSI to avoid the low melting point of pure Se (217 ºC). Among other possible alloys, lead was chosen due to the very small cross section of ^{208}Pb (0.36 mb at 30 keV). For the final sample, 3 g of metallic powder enriched to 99.34% in ^{78}Se were mixed with highly enriched lead (99% ^{208}Pb) to produce a pellet-alloy of 3.9028 g with a diameter of 14 mm and a thickness of 5 mm. The sample was encapsulated at CERN in a laser-welded casing of aluminum with a thickness of 0.5 mm (totaling 1.0240 g of 6N Al) before being irradiated at ILL with a power-weighted fluence of 42 full power days. The expected **amount of ^{79}Se is of about 3 mg**. This value is known with an accuracy of 10% from the reactor fluence data, and it will be accurately measured after the measurement by means of ICP-MS. The ^{79}Se sample was characterized at PSI in 2019, focusing on the **accurate determination of sample contaminants, which allowed for a realistic estimate of the background conditions in the actual capture experiment**. The details can be found in Ref. [30].

3.- Detection systems and experimental areas

Choosing the best combination of detection system and experimental area is key for the success of challenging capture measurements on unstable targets such as ^{79}Se(n,ɣ) which are available in very small quantities (~10^{19} atoms). In this section we discuss the reasons that lead to a combined proposal at EAR1 and EAR2.

First, given the small amount of ^{79}Se material (3 mg) and the activity of the sample, n_TOF-EAR2, featuring the largest instantaneous neutron flux world wide is the best solution to achieve good statistics and minimize the sample activity background. Moreover, a crucial aspect in this experiment will be the disentanglement of ^{78}Se and ^{79}Se levels, given that most of the sample is ^{78}Se. The most complete and accurate measurement of ^{78}Se was performed recently at n_TOF EAR1 [31]. Therefore, a reliable and systematic assessment of the ^{78}Se contribution to the capture yield, which is key to extract the ^{79}Se CS, requires a new measurement of the aforedescribed ^{78}Se+^{79}Se sample under similar experimental conditions. In this context, a measurement in EAR1, is also required to keep the systematic uncertainties under control in the Resolved Resonance Region.

One of the main challenges of this measurement is the high beam-induced background related to the dominant abundance of lead in the sample. The usage of the innovative i-TED detection system [23,28], which exploits the Compton Imaging technique to enhance the detection sensitivity in a factor 5-10 compared to the setup of four C_6D_6 detectors [30] is the best solution. This improvement should enable the systematically accurate measurement of the ^{79}Se(n,γ) cross section at EAR1. An additional strength of i-TED is its good energy resolution (4.5% FWHM at 662keV / $LaCl_3$) which will enable the extraction of spectroscopic information from the (n,ɣ) cascade, which is completely unknown for this isotope. At variance with C_6D_6 detectors, the energy resolution of i-TED will also allow selections in deposited energy, hence controlling better the different sources of background and contributing to the **best control of systematic uncertainties at EAR1**. The good performance and background rejection capabilities of i-TED at EAR1 have been validated during the commissioning of a first prototype in 2018 [29,32]. However, its maximum DACQ acquisition rate is of 500 kEvents/s which represents, at this time, a limitation for its use at EAR2. In the latter area, the common C_6D_6 detectors are better suited.

For all the above-mentioned reasons, we propose a combined measurement using i-TED at EAR1 and four C_6D_6 at EAR2. The first part of the proposal aims at measuring the **first resonances with high energy resolution and small systematic uncertainties** by means of the enhanced detection sensitivity and the well-known performance of the detectors in **EAR1**. The measurement at **EAR2,** that will be validated with the one at EAR1, will profit on the 40 times larger flux to **significantly improve the statistical uncertainty, minimize the contribution of the MBq sample activity and allow the measurement of the thermal cross section**. Previous capture experiments have also combined both EARs to improve the quality of the final data [19,33].

4.- Counting rate estimates, feasibility and expected results

The counting rate estimates have been calculated using the evaluated flux of n_TOF EAR1 and EAR2. The capture cross sections of ^{79}Se, ^{78}Se and ^{27}Al (casing) were obtained from the JEFF-3.3 evaluation. We have to note that the ^{79}Se cross section is based on a theoretical calculation (TENDL-2015), which is affected by an uncertainty of a factor of two, or more. The (n,ɣ) efficiency and the sample activity background have been determined by means of accurate MC simulations of i-TED and C_6D_6 set-ups. The beam-related backgrounds, including the contribution of the lead in the PbSe sample, have been taken from experimental data measured with C_6D_6 and scaled in the case of i-TED assuming a factor 5 gain in capture to background with respect to the former [29]. Last, the fraction of the beam intercepted by the sample and the Resolution Function have been taken from the MC simulations of the current target [34,35]. A detailed description of the expected ^{79}Se(n,ɣ) counting rates compared to the individual background contributions can be found in App. 3. **The present proposal focuses on the measurement of the RRR in the 1eV-10keV energy range, which will be sufficient to provide a stringent constraint of the MACS in the relevant stellar energy range.**

SAMPLE	EAR1: i-TED	EAR2: C_6D_6
Se-79+Se-78 (PbSe sample)	$2{,}5.10^{18}$ p	$1{,}5.10^{18}$ p
Dummy (Pb + Al)	5.10^{17} p	5.10^{17} p
Au, C, Pb, Filters	~5.10^{17} p	~5.10^{17} p
TOTAL	**$3{,}5.10^{18}$ p**	**$2{,}5.10^{18}$ p**

Table 1.- Summary of the requested number of protons for the two measurements in this proposal.

Because of the large impact -and challenge- of this measurement, we have included in this proposal a study -more detailed than usual- to evaluate its feasibility and the expected results. In the RRR, the number of observable resonances will depend on the statistics, the uncertainty associated with the background subtraction and the energy and strength of the resonances. To realistically simulate the statistical uncertainties we have implemented a MC resampling method and assigned a given number of protons to the sample (^{79}Se + ^{78}Se + ^{208}Pb + ^{27}Al) and the dummy (^{208}Pb + ^{27}Al) measurements. The sample activity, which is the dominant source of background at EAR1 (see App. 3), has been fitted to 1/v and subtracted without adding any additional uncertainty. The number of protons considered for this MC experiment for each sample and each EAR are summarized in Table 1.

The resulting ^{78}Se + ^{79}Se counting rate in different energy ranges is presented in Fig. 1 for the two measurements in this proposal. This result resembles the experimental capture yield, where we will carry out a combined R-Matrix analysis of the ^{78}Se and ^{79}Se capture cross sections using the SAMMY code [36]. An accurate knowledge of the resonance parameters of ^{78}Se is expected from the recent measurement of ^{78}Se(n,ɣ) at n_TOF, currently under analysis [31]. Fig. 1 shows the main ^{79}Se(n,ɣ) resonances that will be observed with high resolution on top of the ^{78}Se contribution in EAR1, thereby providing an accurate normalization and contaminant (^{78}Se) contribution assessment for the high-statistics measurement at EAR2. According to this MC experiment, at EAR2 we will be able to analyze resonances beyond 1 keV. The results at EAR2 are somehow conservative since in this "MC experiment" we have used the current RF, which is expected to improve significantly with the new spallation target [37].

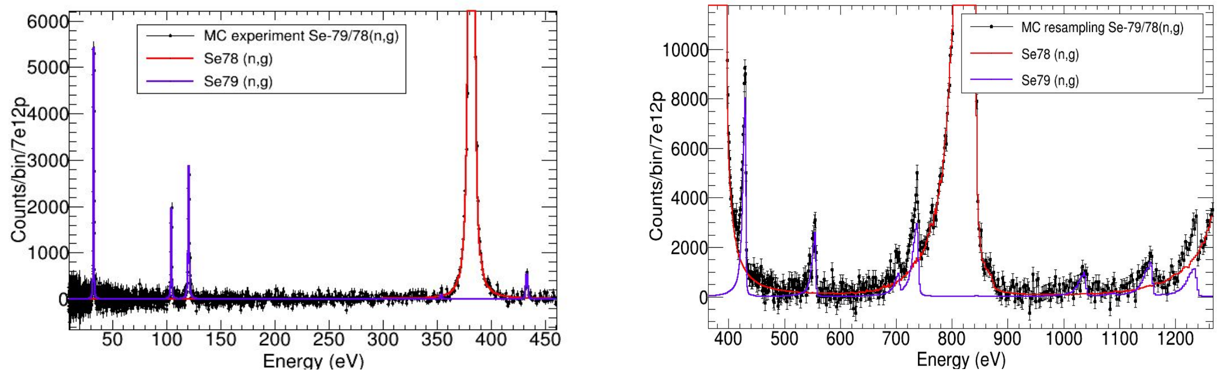

Fig. 1.- Total ^{79}Se + ^{78}Se counts as a function of the neutron energy obtained from the "MC experiment" together with the contribution of each isotope: i-TED at EAR1 in the energy range below 500 eV (left) and C_6D_6 at EAR2 from 400 to 1250 eV (right).

The final number of observed ^{79}Se(n,ɣ) levels determines the accuracy of the average resonance parameters used to calculate the MACS (see next section). This value depends on the unknown and possible overlap with the dominant resonances of ^{78}Se (see Fig. 1). To analyze the possible impact related to the unknown resonance energies and strength in the ^{79}Se+n, the same MC experiment has been repeated for **300 different ^{79}Se(n,ɣ) sets of resonances compatible with the default average resonance parameters in TALYS [38]** (see Table A1.1). The number of observed resonances in the 300 possible cross sections, has been evaluated in terms of an statistical detection limit D, as it is detailed in App. 1. The results of the detection limit study indicate that between 12(3) and 16(3) resonances will be observed up to 1.5 or 2 keV (see App. 1). **A minimum number of 15 resonances is required to extract average resonance parameters with a reasonable uncertainty (40% in S_0) to calculate the cross section in the URR and constrain the MACS (see next Section). This clearly justifies the requested number of protons in Table 1.**

Beyond the RRR, the discrepancies in the ^{79}Se(n,ɣ) cross section also affect the thermal point, where JEFF-3.3 (10.97 b) and ENDF/B-VIII.0 (50 b) deviate in almost a factor 5. According to our estimates, the measurement at EAR2 will allow us to measure the thermal cross section with a negligible statistical uncertainty and a systematic uncertainty dominated by that of the dummy background. If the actual thermal cross section is significantly smaller than predicted by the evaluations we will be able to at least determine a lower limit (see App. 2).

5.- Expected astrophysical impact: MACS

In capture experiments of stable isotopes with large sample masses the MACS can be directly determined from the pointwise TOF data (10-100 keV). However, in the measurement of small-mass radioactive isotopes, only the RRR is experimentally accessible. In such cases, the R-Matrix analysis of the observed s-wave resonances using the SAMMY [36] code, followed by a statistical analysis of the individual parameters leads to a set of average parameters D_0, S_0 and $\langle \Gamma_\gamma \rangle_0$. These s-wave values together with the p-wave parameters (from systematics, see Table

A1.1) can be plugged into the FITACS code [39] (implemented in SAMMY) to calculate a semi-empirical cross section up to 300 keV, from which the MACS at different k_BT can be then determined (see, for instance, Ref. [19]).

The statistical uncertainty in the experimental level spacing D_0, and the neutron strength function S_0 depends on the number of analyzed resonances. According to the results of the previous section, the MACS has been calculated with the parameters of Table A1.1 assuming two scenarios where 9 (worst case) and 19 (best case) resonances are observed in ^{79}Se+n. The calculation with FITACS leads to an interval of confidence for the cross section in the URR which propagates to a range in the MACS, as it is shown in Fig. 2. Additionally, the stellar enhancement factor at kT = 8, 30 keV is 1.0 [9] and, therefore, no additional theoretical correction due to capture on excited states will be needed. The expected MACS at 30 keV is 187 mb, of which 45% comes from the s-wave contribution which can be constrained with the measurement of the RRR (see Fig. 2). The expected uncertainty in the MACS associated with the envisaged number of observed s-wave resonances ranges from 20 to 26%. **The strong uncertainty reduction in the MACS from the current factor of >2 would represent the most stringent empirical constraint for the thermal conditions of TP-AGB and MSs.**

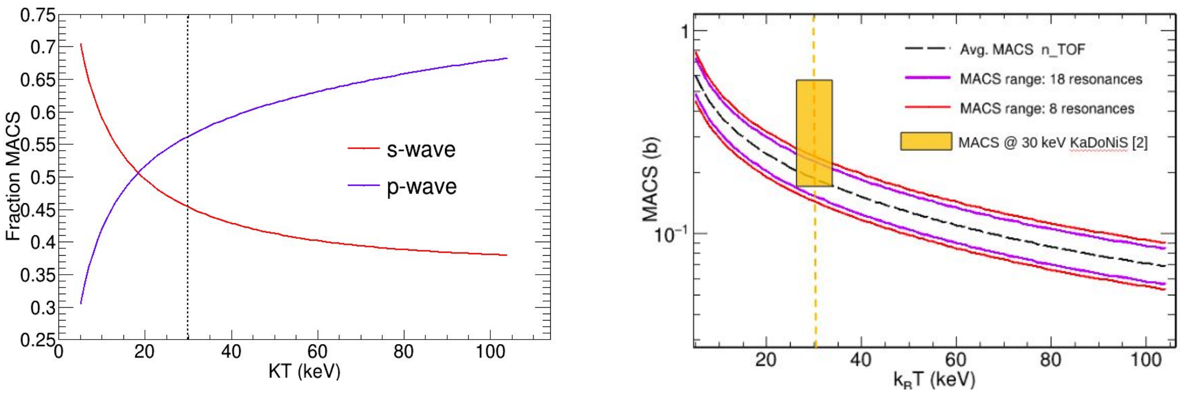

Fig. 2.- Expected contribution of the s- and p-wave resonances to the MACS as a function of the temperature (left). Expected MACS at different stellar temperatures (black dashed) and uncertainty range related to the limited number of observed resonances compared to the range of theoretical calculations compiled in KaDoNiS (yelow band) [6] (right).

6.- Summary & outlook

The measurement described in this proposal will become the **first ever experimental capture data on ^{79}Se(n,ɣ)**, a **key astrophysical s-process branching point** that has been extensively debated and discussed in the literature [2-8].

This proposal presents a reliable approach to tackle this **challenging and high-impact experiment**. The limited mass and high activity of the sample makes this measurement an **n_TOF unique case**. The relevance of this measurement has motivated a **realistic and conservative risk-assessment study**, which shows the feasibility of the proposed experiment and the adequacy of the proposed methodology and beam-time request. The expected data outcome has been used to assess its expected astrophysical impact.

The required detection sensitivity will be achieved with i-TED, a **novel detection system which has been specifically developed (HYMNSERC-CoG Project) to accomplish this measurement at CERN n_TOF.** The last step of the **ERC project (due date May 2022),** after a very successful technological development [24-27] and validation [29] phase, **will be this high-impact scientific output.**

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Appendices: https://drive.google.com/file/d/14OMR7LS_LZ5Uj3rldXznq2jYq6YgwK38/view?usp=sharing