EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

First measurement of the ^{94}Nb(n, γ) cross-section[1]

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Abstract

Neutron capture on radioactive ^{94}Nb is of interest in both nuclear astrophysics and nuclear energy applications. It could play a critical role to constrain the ^{94}Mo production in AGB stars by the s-process, which cannot be reproduced by stellar models. There exist no previous ^{94}Nb(n,γ) experimental data for the resolved and unresolved resonance regions. We have recently produced a hyper-pure sample of ^{94}Nb for this (n,γ) experiment. We propose to perform the measurement at the high flux beam line EAR-2 in combination with low background C_6D_6 detectors, which allows us to overcome various experimental challenges. We have performed a careful feasibility study summarized in the text and detailed in the **appendices**[2].

Requested protons: $3{\cdot}10^{18}$ protons on target **Experimental Area**: EAR2

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[2] https://docs.google.com/document/d/1d4yw_zY5mIkZyUp8mz7Px_2J8oOO5zVRug89GzEWvI0/edit?usp=sharing

Motivation

Neutron capture cross-sections are key ingredients to understand the abundances of chemical elements heavier than iron due to the slow neutron capture process (s-process). The s-process is responsible for about half of the isotopic abundances. This process occurs in Asymptotic Giant Branch (AGB) stars of $M<4M_{sun}$ at temperatures of around $T=10^8$ K. The main neutron source is the $^{13}C(\alpha,n)^{16}O$ reaction during the recurrent He-shell flashes [1]. Our present knowledge of (n,γ) cross-sections is insufficient for the study of several radioactive isotopes, which act as branching points in the s-process path. In these cases, very important information on the physical stellar conditions, such as neutron densities and temperatures, can be gained because of the sensitivity of the local isotopic abundance pattern to the branching ratio, which is determined by the neutron capture rate, neutron density and stellar half-life. Several of the most challenging s-branching nuclei (^{93}Zr [2], ^{151}Sm [3] and ^{171}Tm [4]) have been recently measured at n_TOF.

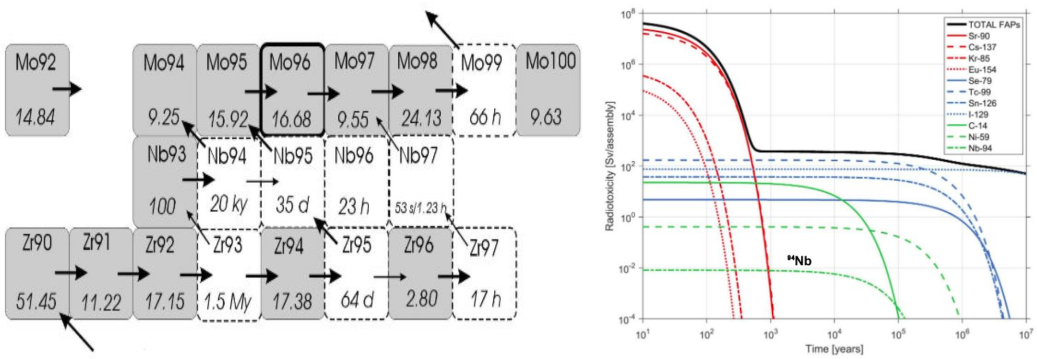

Fig.1 (Left) s-process path zoom on the Nb and Mo isotopes region. (Right) Main non-actinides contributors to low and intermediate nuclear waste radiotoxicity.

The solar-system abundance of ^{94}Mo represents yet a challenge for s-process models and an important topic of discussion. According to state-of-the-art stellar models [5,6,7], the predicted s-process abundance of ^{94}Mo is at most 1%[8,9,10]. On the other hand, accurate analysis of molybdenum isotopic compositions in single mainstream presolar SiC grains from the Murchison meteorite indicate an s-process abundance **five times larger[10,11]**. As indicated in [10,11], **more precise cross-section determinations are needed** to understand this **long-standing discrepancy**. One possible way to feed ^{94}Mo and compensate for such discrepancy is related to neutron capture on the initial ^{93}Nb. Neutron capture on ^{93}Nb leads to the formation of ^{94}Nb (t1/2 = 20ky). However, at the thermal conditions of the s-process (T=0.3GK) the effective half-life of ^{94}Nb drops to only 9 days[12], **strongly feeding ^{94}Mo**. Therefore, two yet unknown physics inputs are crucial to disentangle this contribution. One of them is the neutron capture cross-section on ^{94}Nb that we plan to measure in this project. As reported in the sensitivity study [13], $^{94}Nb(n,\gamma)$ cross-section is a key reaction for the production of ^{94}Mo in the thermal pulse of an AGB star, second only after $^{94}Mo(n,\gamma)$. The second ingredient is the effective stellar half-life of ^{94}Nb, which is only known theoretically [12] and will be experimentally tackled in the INFN-PANDORA project [12].

In addition to the astrophysical motivation, ^{94}Nb ($T_{1/2}=2.0\cdot10^4$ y) is an isotope of interest in nuclear energy applications because of its presence in low and intermediate nuclear waste [14]. Since ^{94}Nb is produced in a multi-step process the contribution shown in Fig. 1 right would increase roughly with the square of the irradiation time, i.e. becomes more important for high burnup fuel.

Only a few experimental data-points at thermal neutron energies are available from activation measurements. The experimental thermal cross-section values are displayed in the left panel of Fig. 2. **There exist no previous ^{94}Nb(n,γ) experimental data for the resolved and unresolved resonance regions**. The absence of neutron capture data at higher energy is related to the limited mass that can be obtained for an experiment and the high activity of the sample. Due to the lack of experimental data the evaluated ^{94}Nb(n,γ) cross-section available in evaluated libraries is based only on statistical models. Furthermore, the lack of experimental data has introduced large variations in the Maxwellian Averaged Cross-Section (MACS) adopted over the years, as it is graphically shown in Fig.2-Right. The value of ^{94}Nb listed in the Karlsruhe Astrophysical database Kadonis[15] (482 ± 92 mb) covers only two of the five theoretical predictions available (see Fig.2-Right), which indicates that the uncertainty is probably larger than estimated.

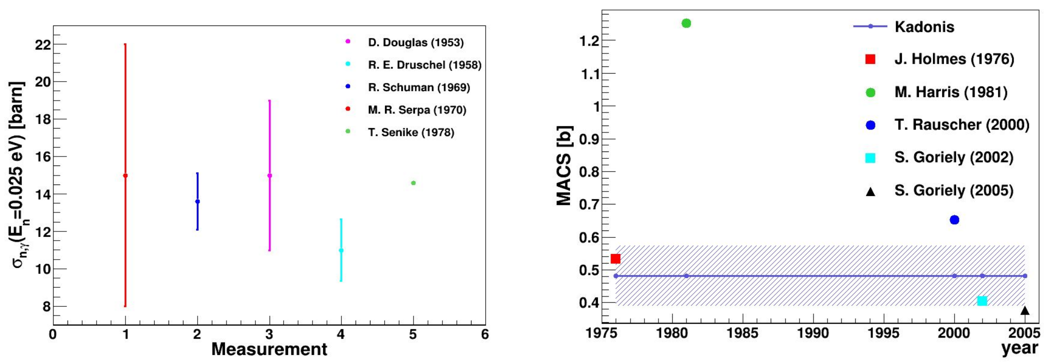

Fig.2.- (Left) Experimental thermal ^{94}Nb(n,γ) cross-section values.(Right) Theoretical estimations for MACS values over the years. The purple band corresponds to the recommended kadonis value..

The measurement of this particular neutron capture cross-section is quite challenging due to the aforementioned reasons of radioactivity and small sample mass. Presently, **such difficulties can be only overcome by means of a very high neutron luminosity as the one available at n_TOF EAR2, and a suitable sample with lowest possible contaminants**.

Sample production and characterization

The conventional methodology of producing ^{94}Nb from activation of ^{93}Nb materials was not applicable in this case. Although ^{93}Nb is naturally mono-isotopic, there is no supplier that can certificate less than about 100ppm of Ta. This quantity is far too much because it would take many years to sit out the ^{182}Ta produced in the neutron irradiation. Therefore, an alternative methodology was followed for this specific experiment. A **hyperpure sample of ^{93}Nb** originally produced at the Institute of Solid State and Materials Research of Dresden [16] was made available for this project thanks to a collaboration with ILL. The final ^{93}Nb material available for this experiment has a mass of 304 mg and **<1 ppm of Ta**. It was afterwards activated at ILL for 51 days and a power weighted fluence of 42 full-power days, thereby yielding **9.24×10^{18} atoms of ^{94}Nb**. This represents ~1% of the total number of atoms present in the bulk of the sample. A careful characterization of the activated sample was performed at PSI by means of HPGe gamma-ray spectroscopy [17,18]. The **activity of ^{94}Nb found in the sample was 10.1 MBq** with **no additional trace of contaminants**. This is therefore a rather **unique sample**, and its characterization became also of pivotal importance for a realistic assessment of the background level expected in the proposed experiment and to validate its feasibility.

Detection system and experimental area

The unique ^{94}Nb sample specifically produced for this experiment in combination with the characteristics of EAR2, with unparalleled instantaneous neutron flux [9.6·10^{5} n/cm2/pulse] and sufficiently good neutron energy resolution of 0.8% in the keV region, provide the first opportunity to measure the ^{94}Nb(n,γ) cross-section with high accuracy. Due to the high counting-rate and high sample activity C_6D_6 detectors are the best option to detect the prompt capture gamma cascade. They also present, up to now, the swiftest recovery from the γ-flash, thereby guaranteeing the quality of the data in the resolved neutron resonance region with negligible dead time corrections.

Counting rate estimates, feasibility study and expected results

The estimation of the counting rate statistics has been calculated using the 2014-2015 evaluated flux of n_TOF EAR2. The background used corresponds to the one measured during the experiments of the same campaign. The counting rate estimate is made by the convolution of the neutron flux, taking into account the experimental resolution of the setup, and using JEFF3.3 and TENDL-2019 evaluated libraries for ^{93}Nb and ^{94}Nb isotopes, respectively. The (n,γ) efficiency and the counting rate associated with the sample activity have been determined by means of accurate Monte Carlo (MC) simulations of the C_6D_6 detectors situated at a distance of 10 cm from the sample. The estimated intercepted fraction of the beam by the sample and the Resolution Function were taken from MC simulations of the current spallation target [19,20]. During the actual experiment we expect that the neutron energy resolution will be significantly better than the assumed resolution, due to installation of a new and optimised spallation target during LS2 Therefore, the results shown in this text are somewhat conservative.

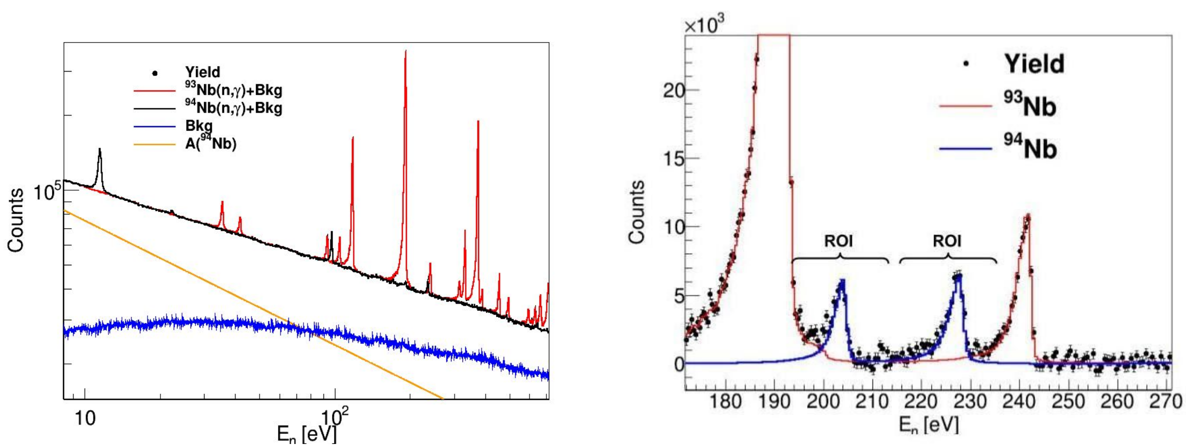

Fig 4. (Left) Estimated Total yield from one of the Monte Carlo experiments; ^{93}Nb*,* ^{94}Nb *counting rate and expected background as a function of the neutron energy in the experiment are displayed by the different colors . (Right) Monte Carlo reaction yield after the subtraction of the empty+activity background to the total yield.*

Due to the large activity of the sample, as in similar previous situations, it was determined that the best compromise between efficiency optimization and background suppression is to use a high detection energy threshold in the C_6D_6 detectors of 500 keV, that may be compensated afterward by the Pulse-Height Weighting Technique [21]. Fig. 4 shows the expected counting spectra including realistic backgrounds, as determined in MC simulations for the proposed experimental setup. These results were obtained as follows. Given the limited signal-to-background ratio expected for many of the ^{94}Nb resonances (see Fig.4) a methodology was developed in order to realistically **assess the feasibility of the experiment and to quantify the expected results**. For such study, 300 TALYS[22] MC simulations of the ^{94}Nb(n,γ) cross-section were used. For further details the reader is referred to **Appendix A**. From the TALYS MC ^{94}Nb(n,γ) cross-sections, the expected counting rate was calculated, and then resampled proton pulse by proton pulse using a Poisson distribution whose mean value corresponds to the value of the different components. The number of protons used for the calculation are summarized in the table 1.

According to this MC study, and since the ^{93}Nb(n,γ) cross-section was also recently measured at n_TOF EAR2 [23] (See **Appendix D**), the most suitable strategy is to analyze the experimental yield of ^{93}Nb(n,γ) and ^{94}Nb(n,γ) altogether from the capture data in the same sample. Statistically it becomes more accurate to not subtract the dominant contribution of ^{93}Nb, but rather identify the corresponding neutron resonances of ^{93}Nb(n,γ) and carry out a simultaneous R-matrix analysis of both isotopes.

The expected number of measurable resonances in the ^{94}Nb(n,γ) yield depends on the overlap with resonances of ^{93}Nb(n,γ), their relative strengths and the overall counting statistics. It is worth to remark that **for the statistical and astrophysical interpretations a minimum number of 10 resonances becomes necessary**. This number, in turn, determines the total beam-time request of the present proposal. In order to estimate, in a realistic way, how many ^{93}Nb+n levels will be observed, a Region of Interest (ROI) is defined at the position of the individual ^{94}Nb resonances in the ^{93}Nb+^{94}Nb netto yield (see Fig.5). Assuming Gaussian statistics, a significance D, can be defined as:

$$D = (I_{yield}(ROI) - I_{93\,Nb}(ROI))/(ROI)$$

Where $I_{yield}(ROI)$ and $I_{93\,Nb}(ROI)$ are the total counts (black dots in Fig. 4) and the contribution of ^{93}Nb (red line) and (ROI) the quadratic sum of the statistical uncertainty from the counting MC yield and a fair 10% systematic uncertainty for the ^{93}Nb reaction yield. D will determine the detection of the resonance by setting a threshold in the significance. This process, after the repetition over all resonances in all 300 MC yields, can be used to determine an educated guess for the expected number of detected resonances. Attending to a conservative significance of D>3, the expected number of detected resonances during the experiment is 13(5) in the neutron energy range up to 1.2 keV for the number of protons requested as in Table 1. Further details of the calculation can be found in **Appendix B**. This result, when propagated into the average resonance parameters would lead to statistical uncertainties of about 10%, 18% and 40% in Γ_γ, D_0 and S_0, respectively. These values are **sufficient to constrain the cross-section in the unresolved resonance region and the values of the MACS in the relevant stellar energy range**. This statement is demonstrated in **Appendix C**.

The requested amount of protons for the proposal which corresponds to this calculation are summarized in table 1.

Configuration	Protons **10^{18}**
Nb-94 sample in place	2.0
Background estimation	0.5
Au, C, Filters	0.5

Total	**3.0**

Table 1: *Summary of requested protons for the measurement. The number of protons devoted to ancillary measurements could be reduced if the measurements are shared with other proposals.*

Summary and Conclusions

The ^{94}Nb(n,γ) reaction is highly interesting both in nuclear astrophysics and nuclear energy applications. So far, **no measurement has been made either in the resolved and unresolved neutron resonance region** due to its radioactivity, resulting in availability of only low mass samples, and high sample activity.
A hyperpure Nb sample with 9.24×10^{18} atoms of ^{94}Nb has been produced for the present experiment in a collaboration between ILL, PSI and CSIC. Based on the present knowledge, such **a unique ^{94}Nb sample**, in combination with the high neutron luminosity of n_TOF EAR2 would make the first measurement ever on this isotope feasible.
Because of the conspicuous challenges in such an experiment Monte Carlo "experiments" based on an extensive set (300 histories) of statistical level parameters have been performed in order to assess the feasibility of this experiment. **About 13(5) resonances in the neutron energy range up to 1.2 keV should be accessible for the first time.** From this reduced set of resonances the average resonance parameters can be obtained (as e.g. in the recent ^{171}Tm experiment[4]) with sufficient statistical uncertainty to extract valuable astrophysical information. As a consequence, the cross section at higher energies and the MACS at kT=8keV can be experimentally constrained (see Fig.5 and **Appendix C**).

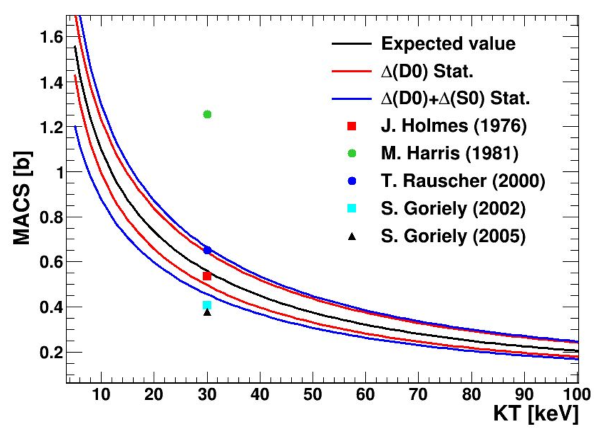

Fig 5. Expected MACS values as a function of the thermal energy. The red and blue lines are the statistical uncertainty estimated from D_0 and S_0. In the same plot is included the values taken from kadonis.

It is important to remark that the calculations were made using the n_TOF-Phase3 Resolution Function, which was one of the limitations in some of the previous (n,γ) cross-section measurements at EAR2. In the forthcoming campaign we expect a significantly improved RF, which will lead to a higher peak-to-background ratio. In this respect, the estimates presented here can be considered conservative.

Summary of requested protons: 3×10^{18}

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