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

Neutron capture at the *s*-process branching points ^{171}Tm and ^{204}Tl

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Abstract

Branching points in the s-process are very special isotopes for which there is a competition between the neutron capture and the subsequent b-decay chain producing the heavy elements beyond Fe. Typically, the knowledge on the associated capture cross sections is very poor due to the difficulty in obtaining enough material of these radioactive isotopes and to measure the cross section of a sample with an intrinsic activity; indeed only 2 out o the 21 *s*-process branching points have ever been measured by using the time-of-flight method.
In this experiment we aim at measuring for the first time the capture cross sections of ^{171}Tm and ^{204}Tl, both of crucial importance for understanding the nucleosynthesis of heavy elements in AGB stars. The combination of both (n,γ) measurements on ^{171}Tm and ^{204}Tl will allow one to accurately constrain neutron density and the strength of the ^{13}C(α,n) source in low mass AGB stars. Additionally, the cross section of ^{204}Tl is also of cosmo-chronolgical relevance and, in combination with the ^{205}Pb/^{205}Tl abundance ratio, it will provide valuable information on the time passed since the last nucleosynthesis contributing to the chemical composition of our solar system.

Requested protons: $7.5\text{x}10^{18}$ protons on target
Experimental Area: EAR-1

1. Introduction, motivation and objectives

The s- and r-processes are the responsible for the formation in the stars of practically all the chemical elements heavier than iron. The phenomenological picture of the classical s process was formulated about 50 years ago in the seminal papers of Burbidge et al. [1] and of Cameron [2] in 1957, where the entire s-process panorama was already sketched in its essential parts. They explain how, in this process, the elements heavier than iron are produced by a continuous chain of neutron capture reactions and beta-decays that give rise to the heavy elements. The phenomenology of the s-process implies that the solar abundance distribution is composed of two parts, a main component, which is responsible for the mass region from Y to Bi, and a weak component, which contributes to the region from Fe to Sr. The main and weak components can be assigned to low mass stars (between 1 and 3 solar masses) and to massive stars (more than 8 solar masses), respectively. Accordingly, the Galactic enrichment with s-process material starts with the lighter s elements, because massive stars evolve much quicker. For a recent and comprehensive review see Ref. [3].

The s-process requires a neutron source that sustains the reaction chain. These neutrons are mainly produced by the $^{22}Ne(\alpha,n)^{25}Mg$ and $^{13}C(\alpha,n)$ reactions, but at rather different temperatures and neutron densities. During core He burning, neutrons are produced near core He exhaustion at temperatures of $T\sim3x10^8$ K for about 10^4 years with neutron densities below 10^6 cm^{-3}, whereas the higher temperatures of $T\sim10^9$K during the subsequent carbon shell burning phase give rise to peak values of about 10^{12} cm^{-3}. More details can be found in Raiteri et al. [4-5] and Limongi et al. [6].

A quantitative description of the abundances arising from the s-process requires both, the neutron capture rates and the b-decay probabilities of all the isotopes involved. Along the s-process path, unstable nuclei with relatively long (y) and with very long (Gy) half-lives become of utmost interest, as they can be used to constrain the conditions of the environment (density, temperature) and the age of the nucleosynthesis process, respectively. Despite of their pivotal role, as of today, only the capture cross section of 2 out of the 21 s-process branching points isotopes (see [3]) have been measured by neutron time-of-flight (the most accurate method for neutron capture studies). The focus of the present proposal is to determine, for the first time, the capture cross section of two additional branching point isotopes, with relevance for the understanding of the environment conditions and for a cosmochronology study s-process contribution to the solar system composition.

As mentioned above, the neutron capture cross section of s-process branching points has been measured by time-of-flight only for two cases: ^{63}Ni (Lederer et al. [7] at n_TOF/CERN and Weigand [8] at DANCE/LANL) and ^{151}Sm (Abbondanno et al. [9] at n_TOF/CERN).

Apart from the very large instantaneous neutron flux required for this kind of studies, the availability of (unstable) sample material in sufficient amount and with enough purity represents an additional challenge for accessing this valuable information.

For this reason, only indirect methods such as surrogate reactions or studies based on the inverse reaction (γ,n) in combination with the detailed balance (see e.g. Raut et al. [10]), have provided some constraint or indication about the (n,γ) cross sections of several s-process branching point nuclei.

Within the EC *NeutAndalus* project [11] there is the opportunity of making a big step forward in the field, by making three samples of the corresponding *s*-process branching points ^{147}Pm, ^{171}Tm and ^{204}Tl available for measurements. The project includes the aim of measuring their capture cross sections at the CERN n_TOF facility [12].

While ^{147}Pm must be measured in the future n_TOF EAR-2 neutron beam (see related LoI within this INTC session), the available mass of ^{171}Tm and ^{204}Tl is such that the measurements are possible at n_TOF EAR-1, representing the latter two measurements the core of this proposal.

These two isotopes are important s-process branching points [3]. The unstable isotope ^{171}Tm (half-life of 1.92 years) represents a branching in the s-process path that is independent of stellar temperature [3] and therefore suited to constrain explicitly the *s*-process neutron density in low mass AGB stars. The branching analysis will benefit from the fact that Tm is a rare earth element and thus the relative abundances of the existing isotopes are known with high accuracy. The case of ^{204}Tl (half-life of 3.8 years) is particularly interesting; its decay product is ^{204}Pb, which produces ^{205}Pb when undergoing neutron capture. The fact that both ^{204}Pb and ^{205}Pb are screened from the *r*-process by the stable ^{204}Hg and ^{205}Tl combined with the previous measurements of the capture cross section of ^{204}Pb at n_TOF [13] will provide highly interesting chronometric information about the time span between the last s-process nucleosynthetic events that were able to modify the composition of the proto-solar nebula and the formation of solar system solid bodies. See Yokoi et al. [14] for a detailed discussion on the principles behind the ^{205}Pb/^{205}Tl *s*-process chronometry.

At present the values of these two cross sections used for calculating abundances in stellar models are purely based on theoretical predictions. For example, Figure 1 shows the values of the calculated Maxwellian Averaged Cross Section (MACS) along the years, which vary within a wide range of values up to factor of three from minimum to maximum values.

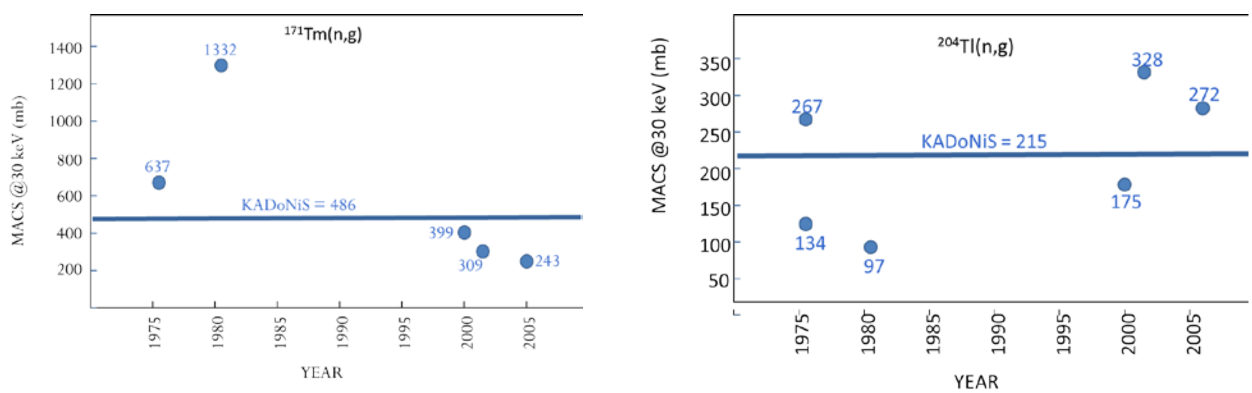

Figure 1. Historical series of MACS predicted values at 30 keV for ^{171}Tm and ^{204}Tl since 1975. All values are theoretical predictions.

Presently, such a large uncertainty on the cross section of these isotopes is hindering a reliable interpretation of the astrophysical aspects discussed above. For example, a variation of the (n,γ) cross section of ^{204}Tl by a factor of two up and down produces a corresponding decrease (increase) in the abundance of ^{205}Tl (^{204}Pb) by more than 30%. Such behaviour may impact by a factor of 2, or more, the ^{205}Pb/^{205}Tl ratio of interest for the cosmo-chronological study described above. This is illustrated in the sensitivity study shown in Figure 2, which is based on a classical s-process analysis (density $1\text{x}10^3$ g/cm^3, 10^8 n/cm^3 and $T_9 = 0.2$ K).

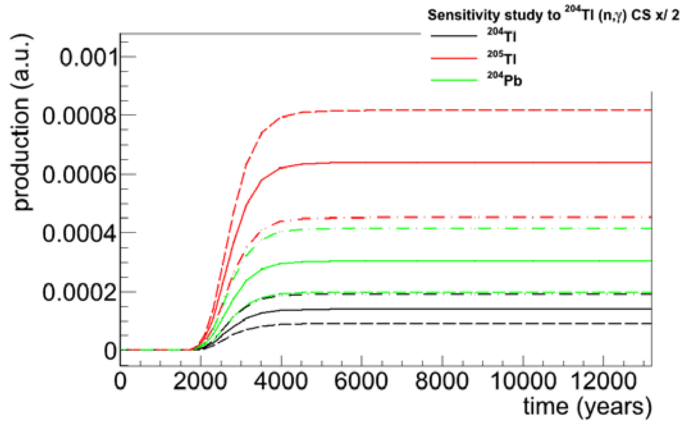

Figure 2 Sensitivity study based on a variation of the cross section of 204Tl by a factor of two up (dot-dashed) and down (dashed) lines.

2. Sample preparation

As in most challenging capture cross section experiments, the material availability and samples preparation are crucial and not trivial. In the case of ^{171}Tm and ^{204}Tl, this is the reason why these experiments have not been carried out before. Within the *NeutAndalus* project we have established a collaboration between CERN, the Institute Laue Langevin ILL (Grenoble, France) and the Paul Scherrer Institute PSI (Villigen Switzerland) in order to produce the isotopes of interest and prepare the corresponding samples. Along the Summer of 2013, two 5 mm diameter pellets of 210 mg of ^{170}Er (isotopic purity 98.1%) and 225 mg of ^{203}Tl (isotopic purity 99.5%) have been irradiated with thermal neutrons for a total of 56.7 days at the ILL experimental nuclear reactor. The pellets are shown in Figure 3.

Figure 3. Pellets of ^{146}Nd, ^{170}Er and ^{203}Tl before their irradiation at ILL.

This irradiation has produced 3.6 mg and 11 mg of ^{171}Tm and ^{204}Tl, respectively. After the irradiation, the ^{204}Tl sample can be directly used for the (n,γ) measurement at CERN n_TOF because there is no possibility to separate isotopically the ^{204}Tl from the initial ^{203}Tl. On the other hand, ^{171}Tm is produced after b-decay of the ^{171}Er (7.5 h) resulting from neutron capture in ^{170}Er. Thus, advanced chemical purification techniques will be used to separate Tm from the Er in the sample. This separation will be carried out at the RadWaste Analytic unit at PSI using the technique of ion exchange chromatography.

3. Experimental set-up

There are two different systems for measuring neutron capture cross sections at n_TOF, both based on the detection of the prompt gamma-rays which follow each capture event, the low neutron sensitivity C_6D_6 detectors [15,16] and the 4π BaF_2 Total Absorption Calorimeter (TAC) [17]. In the case of the C_6D_6 detectors, a new set-up with four new detectors with even more reduced neutron sensitivity will be available for the experiment (see Ref. [18] for details). The two detection systems are illustrated in Figure 4.

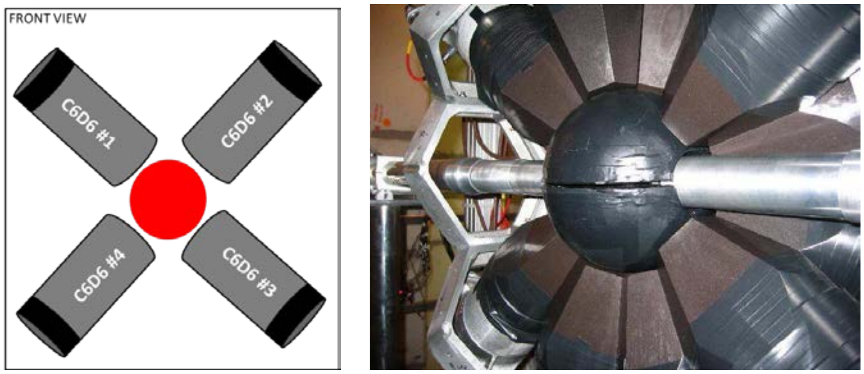

Figure 4. The C_6D_6 (left) and TAC (right) detection systems employed in neutron capture cross sections measurements at n_TOF, both to be used in this proposal.

In our case, taking into consideration that the ^{171}Tm sample will be highly pure and thus the C_6D_6 detectors are a better choice, mainly because of their smaller neutron scattering background conditions. In the case of the ^{204}Tl sample where ^{203}Tl atoms account for 93% of the sample, only the use of the TAC will allow distinguishing from capture cascades coming from one (^{203}Tl) or another (^{204}Tl) isotope in the sample. This will be observed in the TAC at different deposition energies, since the neutron separation energy values are pretty different: $S_n(^{203+1}$Tl$)$=6.7 MeV and $S_n(^{204+1}$Tl$)$=7.5 MeV.

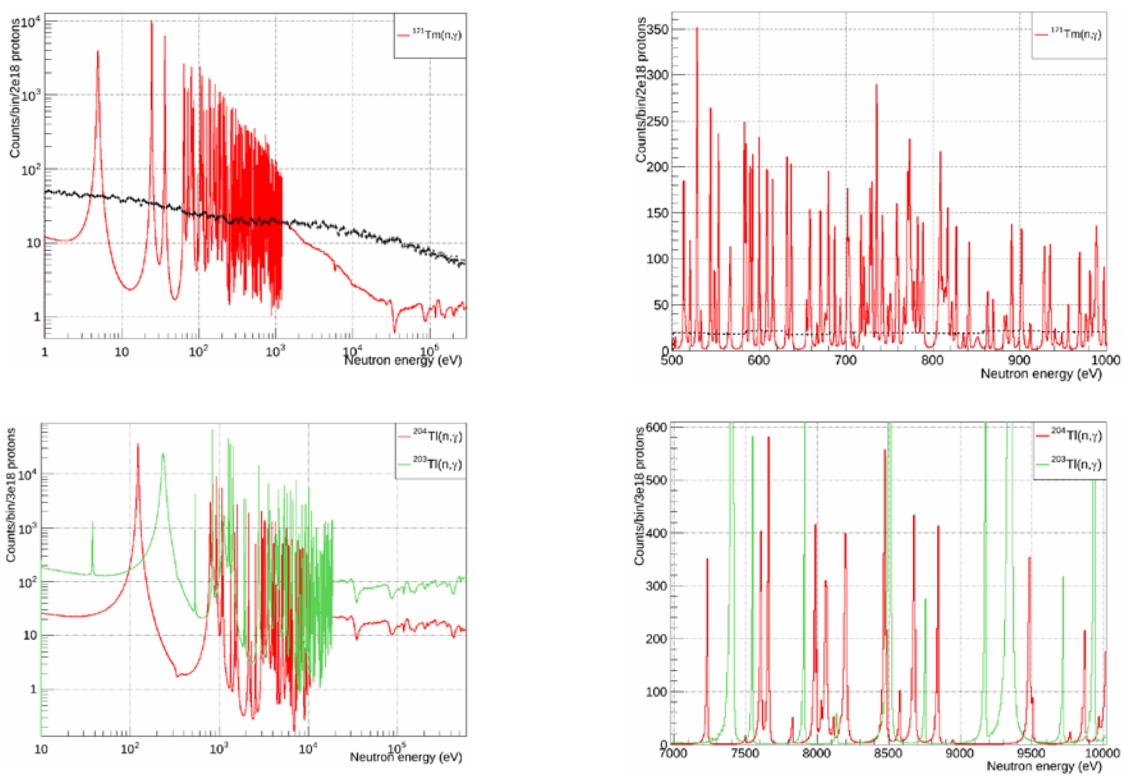

Figure 5. Expected counts for the ^{171}Tm (C_6D_6) and ^{204}Tl (TAC) measurements, with zooms in the high energy.

4. Objectives and beam time request

The astrophysical neutron energy range of interest spans between 1 keV and 300 keV. Because ^{171}Tm and ^{204}Tl are heavy isotopes, the cross section resonances appear at low energies and thus the Unresolved Resonance Region (URR) starts already at neutron energies of a few keV. The measurement of the cross section in the URR becomes more challenging owing to its smooth (decreasing) behaviour and the lower signal-to-background conditions. Considering the background level measured in the previous campaign at CERN n_TOF, we expect to determine the MACS in the URR up to a few hundreds of keV, both point-wise and by means of Hauser-Feshbach theory using the resonance parameters obtained at low energy. The former is governing the beam request, since we aim at having a minimum of 100 counts/bin with 100 bins per decade in the URR in both cases.

The expected distributions of counts for the ^{171}Tm and ^{204}Tl measurements are shown, with 3000 bins/decade in Figure 5 (only 100 bins/decade in the URR). The efficiency used for ^{171}Tm, using four C_6D_6 detectors, is 20%. For the ^{204}Tl case a typical TAC efficiency of 60% has been employed but this will be reduced when one sets the energy conditions for distinguishing between ^{203}Tl and ^{204}Tl cascades. The actual values cannot be stated now but it will have to be found during the measurement; and the background level will change substantially with these energy conditions, which is why no background level is shown in the ^{204}Tl case.

Summary of requested protons

In addition to the beam time allocated to the ^{171}Tm and ^{204}Tl measurements, background and normalization measurements will be carried out. The overall beam time request is summarized in Table 1.

Table 1. Summary of the beam time request.

Sample / Set-Up	Objective	Protons
^{171}Tm/C_6D_6	Capture Cross section of ^{171}Tm with C_6D_6	$2x10^{18}$
^{204}Tl / TAC	Capture Cross section of ^{204}Tl with TAC	$2.5x10^{18}$
^{203}Tl / TAC	Background from ^{203}Tl on the ^{204}Tl sample with TAC	$1x10^{18}$
Empty / C_6D_6	Overall beam-on background	$0.6x10^{18}$
Dummy / C_6D_6&TAC	Sample backing related background	$0.8x10^{18}$
^{197}Au / C_6D_6&TAC	Normalization and validation	$0.6x10^{18}$
Total		**$7.5x10^{18}$**

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