# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

# Neutron capture on <sup>205</sup>TI: depicting the abundance pattern of lead isotopes in s-process nucleosynthesis

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#### Abstract

We propose to use the TOF technique to measure the neutron capture cross section of  $^{205}Tl(n,\gamma)$ over the full energy range of stellar interest. An accurate measurement of this cross section is needed for a complete and consistent understanding of the s-process nucleosynthesis of the heaviest nuclei which are produced in low-mass and low metallicity AGB-stars.

The only previous TOF measurement has yield only a partial information, insufficient for a reliable analysis of the complex branching pattern around <sup>205</sup>Pb and <sup>205</sup>Tl. Furthermore, there is also a discrepancy of 40% between the two previous activation measurements made at kT=24 keV.

The cross section of  ${}^{205}Tl(n,\gamma)$  is particularly relevant because it affects the equilibrium that is established in some stellar conditions between the  ${}^{205}\text{Tl} \rightarrow {}^{205}\text{Pb}$  bound-state  $\beta$ -decay and the  $^{205}$ Pb  $\rightarrow$   $^{205}$ Tl E.C. decay. This effect induces a complex interplay which influences the final sprocess abundance of both nuclei. We propose to measure accurately and with high resolution the  $^{205}$ Tl(n, $\gamma$ ) cross section by using a set of four C<sub>6</sub>D<sub>6</sub> detectors in combination with the pulsed neutron-source of CERN n TOF.

**Requested protons**: 3x10<sup>18</sup> protons on target **Experimental Area**: EAR1

## 1. Introduction and motivations

The nucleosynthesis of elements heavier than iron in the Universe is mainly produced by a series of neutron capture reactions and beta-decays in the so-called *slow* (s) and *rapid* (r) processes. The stellar sites and basic mechanisms of the s-process are well identified [KAP11]. The s-process mechanism operates during core He-burning and shell C-burning in massive stars (also called *weak* s-process), as well as in H-burning and He-burning layers of Thermally-Pulsing low-mass stars of the Asymptotic Giant Branch (TP-AGBs), in what is known as the *main* s-process.

The main s-process is the dominant source for elements of A>90. During the quiescent phase between Thermal Pulses (TP) in low mass AGB stars, H-burning fusion takes place in a thin layer at the base of the convective envelope of the star. In the ashes of the H-burning shell the conditions are such that a <sup>13</sup>C pocket is formed, and neutrons are produced through the reaction <sup>13</sup>C( $\alpha$ ,n)<sup>16</sup>O. This reaction starts to become relevant at a temperature of about 0.9·10<sup>8</sup> K, and produces neutron densities of 10<sup>6</sup>-10<sup>8</sup> cm<sup>-3</sup>. 95% of the total s-process neutron irradiation is reached during <sup>13</sup>C pocket nucleosynthesis.

A second stage of the s-process takes place during the convective TP, which is caused by the rapid burning of the He accumulated during the H-burning phase. During the TP, the temperature can exceed  $2.5 \cdot 10^8$  K, which enables the release of neutrons through the reaction  $^{22}$ Ne( $\alpha$ ,n) $^{25}$ Mg. This neutron irradiation is short, lasting for a few hundred years, but the neutron density achieved is considerably higher, up to  $10^{10}$  neutrons/cm<sup>3</sup>. This intense neutron burst strongly activates the so-called *branching points*, unstable nuclei with long enough half-lives that the capture process can compete with the beta decay. Thus, even if the contribution to the overall neutron exposure is low (~5%), the TP s-process has a strong influence to the final abundances of nuclei around the *branching points*.

An area of special interest is the termination of the s-process reaction flow, i.e. the nuclei in the mass range A=203-210 (figure 1). Some nuclei of interest are the double magic nucleus <sup>208</sup>Pb, whose local overabundance constitutes the third elemental abundance peak, and the s-only isotopes <sup>204</sup>Pb and <sup>205</sup>Pb. The importance of <sup>205</sup>Pb is that on top of being an s-process only, it is radioactive, decaying by E.C. to <sup>205</sup>Tl with a t<sub>1/2</sub>=15 million years. This would enable the use of the abundance ratio <sup>205</sup>Pb/<sup>204</sup>Pb as a chronometer of the last s-process events that contributed to the solar system abundances [YOK85]. But this, of course, is only possible if the final <sup>205</sup>Pb abundance at the end of the AGB phase is well known. Any accurate model of the s-process requires the knowledge of the capture and decay rates of the isotopes involved. Here it is worth mentioning that the authors have



Figure 1. Left: Termination of the s-process chain. Right: Solar elemental abundances, with the third s-process peak highlighted

successfully measured in the past the cross section of the reaction  $^{204}$ Pb(n, $\gamma$ ) [DOM07], and more recently, that of the branching nucleus  $^{204}$ Tl [CAS18], which had not been measured previously (these results are to be published in 2018). These measurements have contributed (or will contribute) to reduce significantly the uncertainty in the evaluation of the  $^{205}$ Pb abundance, which is the purpose of this proposal as well.

In fact, the main relevance for s-process nucleosynthesis of  $^{205}$ Tl, the most abundant thallium isotope (70%) in Nature, is that it becomes unstable at stellar temperatures, decaying to  $^{205}$ Pb by bound-state beta decay. This decay rate increases dramatically as temperature rises, and becomes equivalent to the  $^{205}$ Pb decay rate at a temperature of approx. T~1.6  $\cdot 10^8$  K, which is quickly reached and surpassed during the recurrent TP.

Therefore, the <sup>205</sup>Tl stellar decay could contribute decisively to the <sup>205</sup>Pb produced; actually, as pointed by Yokoi et al., it may be crucial to its survival. But if <sup>205</sup>Tl is an important source of <sup>205</sup>Pb through its decay, it can be deduced that changes in the abundance of <sup>205</sup>Tl during s-process nucleosynthesis must affect the abundance of <sup>205</sup>Pb.

It is in this context that the  ${}^{205}\text{Tl}(n,\gamma)$  reaction rate becomes relevant, since it is the reaction that affects more sensibly the abundance of  ${}^{205}\text{Tl}$  during s-process nucleosynthesis. A decrease (increase) in this rate will yield an over (under)-production of  ${}^{205}\text{Tl}$ , which in turn will lead to an increase (decrease) in the amount of  ${}^{205}\text{Pb}$  produced by the decay of  ${}^{205}\text{Tl}$ .

In order to illustrate the astrophysical relevance of the proposed measurement, a NuGrid [NUGRID] post-processing nucleosynthesis network calculation was carried out to simulate the  $^{205}Pb/^{204}Pb$  production ratio in a 3 solar mass star, for two different metallicities: half solar metallicity (Z=0.006) and very low metallicity (Z=1e-4)[RITT17]. The calculation has been performed for a whole  $^{13}C$  pocket (H-burning shell) and the follow-up TP (He-burning intershell) episode. The whole MACS for  $^{205}Tl(n,\gamma)$  has been varied  $\pm 33\%$ , which is a conservative estimate of the present uncertainties in this cross section (see section 2). The  $^{205}Pb/^{204}Pb$  abundance ratio for the Z=0.006 model is shown in Fig. 2. The impact of the MACS variation on the abundance of the stellar envelope is not negligible. Changes of roughly -6% (MACS +33%) and +11% (MACS - 33%) to the  $^{205}Pb/^{204}Pb$  ratio are obtained from the calculation for both stellar models.

Finally, it is relevant to say that <sup>205</sup>Tl has no excited levels at low energy, which could be populated under stellar conditions [RAU00] and, therefore, the laboratory measurement will directly provide the relevant MACS for a straight-forward astrophysical



Pb-205/Pb-204 abundance ratio in the stellar envelope, M=3, Z=0.006

Figure 2. Variation of the <sup>205</sup>Pb/<sup>204</sup>Pb abundance ratio in the stellar envelope as a function of time (cycles), for a  $3M_{\odot}$ , Z=6e-3 star, for three different rates of the <sup>205</sup>Tl(n, $\gamma$ ) reaction: the Kadonis reference rate (in blue), the reference rate multiplied by a 1.33 factor (in orange), and multiplied by a 0.66 factor (in green).

interpretation, this is to say, without any need of so-called Stellar Enhancement Corrections [RAU00]. In addition, the <sup>205</sup>Tl capture cross section has not been measured yet in any modern facility. The only experimental data available dates from the seventies and show discrepancies between the different measurements (see section 2).

## 2. Status of the data

The examination of the literature and databases shows that the cross section for the  $^{205}$ Tl(n, $\gamma$ ) $^{206}$ Tl reaction in the regions of astrophysical interest remains still uncertain. TOF measurements of natural thallium isotopes have been carried out by Macklin et al at ORNL in 1976 [MAC76] and by Couture et al at LANSCE in 2007 [COU07]. For the particular case of  ${}^{205}$ Tl(n, $\gamma$ ) reaction, just partial or preliminary results were reported, but the final results of both experiments were never published. Indeed, for the ORNL experiment, the accessible data is an EXFOR entry (13734.003) with only resonance energies and resonance kernels. Resonance parameters are not provided and the uncertainties are not quoted or evaluated in EXFOR. The available experimental information makes difficult the proper assessment of the astrophysical aspects concerning the  ${}^{205}$ Tl(n, $\gamma$ ) ${}^{206}$ Tl reaction. The examination of the ENDF/B-VII.1 evaluated data for this reaction suggest that the evaluation in the region of astrophysical interest ( $\sim 1 - 100 \text{ keV}$ ) is based on the ORNL data from 1976 [MAC76]. It is worth mentioning that the CS measurements carried out at ORNL in the seventies were later corrected for a common systematic error (~5-10%) found in the data processing routines. An effective correction factor (0.9507) was only provided for the  ${}^{203}$ Tl(n, $\gamma$ ) measurement [MAC81], and thus the case of  ${}^{205}$ Tl(n, $\gamma$ ) CS remains unclear. The inspection of the JEFF3.2 evaluation shows notable differences with respect to ENDF/B-VII.1 (see Fig. 3). In fact, for neutron energies up to 30 keV, there are more than ten resonances in ENDF/B-VII.1 which are not present in the JEFF3.2 evaluation (which is based on the TENDL evaluation), thus affecting the calculation of the relevant input for nucleosynthesis calculations (MACS) in this region.



Figure 3. Comparison between ENDF/B-VII.1 and JEFF 3.2 evaluations for the  ${}^{205}Tl(n,\gamma)$  cross section, which illustrates the present mismatch between evaluations owing to the scarcity of experimental data.

In the KADONIS database (v1.0) the recommended total MACS value at 30 keV is  $52.6\pm3.9$  mb. This value is based on the ENDF/B-VII.1 evaluation and assuming a 7.4% uncertainty as in [MAC76]. Two additional MACS values at 24 keV from activation measurements are also listed in KADONIS. Both measurements present notable differences:  $30\pm8$  mb by Hasan et al [HAS68] and  $50\pm4$  mb by Kononov et al [KST58]. Finally, the recommended uncertainties for the MACS values increase as the temperature departs from 25 keV, being as much as 30% at 5 keV. The lack of experimental data for reliable MACS calculation below 10 keV is important since the <sup>13</sup>C pocket mechanism of s-process nucleosynthesis starts operating at approximately T~10<sup>8</sup> K (kT=8.6 keV).

#### 3. Experimental set-up and target

For this measurement we plan to use the current capture setup at the EAR1, based on 4  $C_6D_6$  detectors, which are made of carbon fiber for achieving a very low neutron sensibility [PM13]. EAR1 offers the best neutron energy resolution, and a high neutron flux. Concerning the sample, our aim it is to produce a cylindrical sample of 4 g of <sup>205</sup>Tl (99% pure) oxide, with a diameter of 30 mm. Such dimensions would maximize the sample mass while keeping the thickness in reasonable terms. At the same time it will ensure that the fraction of the neutron beam intercepting the sample is the largest, thus making the best possible use of the allocated beam time.

#### 4. General remarks and beam time request

It is expected that the new measurement of the  ${}^{205}\text{Tl}(n,\gamma)$  cross section proposed here, which relies on the well-known value of the  ${}^{197}\text{Au}(n,\gamma)$  cross section, will solve the present ambiguity of the  ${}^{205}\text{Tl}(n,\gamma)$  MACS from 5 to 30 keV. This will be achieved measuring accurately the cross section in the region of astrophysical interest from 1 to 100 keV, which, in the case of  ${}^{205}\text{Tl}$ , coincides with the range of the Resolved Resonance Region (RRR).

The expected counting rate for the whole experiment, with 3000 bins per decade, is shown in figure 3 for the whole energy range and for the 20 to 80 keV region in detail. The background corresponds to a recent August 2017 measurement ("empty frame"). The total efficiency of the 4 detectors is estimated to be 15%.



Figure 4. Expected counts in the 4  $C_6D_6$  detectors for the <sup>205</sup> $Tl(n,\gamma)$  measurement with, with zooms in the 20 to 80 keV range and in the 29-30 keV range, respectively

# Summary of requested protons

The overall beam time request is summarized in Table 1. The usual gold, for normalization purposes, and background measurements are all included.

Sample	Purpose	Protons
<sup>205</sup> TI	<sup>205</sup> Tl(n,γ) with C6D6	2·10 <sup>18</sup>
Dummy	<sup>205</sup> TI sample background	5·10 <sup>17</sup>
Au, Pb, C	Normalization, beam induced background estimation	5·10 <sup>17</sup>
TOTAL		3.10 <sup>18</sup>

Table 1. Summary of beam time request

The  $2 \cdot 10^{18}$  protons allocated to the <sup>205</sup>Tl sample should allow to reduce the statistical uncertainty in the 30 keV resonances to around 2.5% (figure 4, detail), which we assume will be already below the sum of systematic uncertainties.

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