

# Hunting the *s*-process branching points <sup>147</sup>Pm, <sup>171</sup>Tm and <sup>204</sup>TI at CERN

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Branching points along the s-process path in stellar nucleosynthesis are key isotopes in which there is a competition between two processes: neutron capture  $(n,\gamma)$  and  $\beta^{-}$  decay. This particularity makes the local isotopic abundance pattern around these isotopes very sensitive to their half-life and  $(n,\gamma)$  cross section as well as to the neutron density and the temperature at the stellar scenario being considered. Therefore important information about the latter quantities can be inferred when one knows the corresponding  $(n,\gamma)$  cross sections. This is however difficult because such radioactive isotopes do not exist in nature and thus the material to perform accurate measurements is scarce. Indeed, the  $(n,\gamma)$  cross section of only two, <sup>63</sup>Ni and <sup>151</sup>Sm, of the 21 most important branching points have been measured by time-of-flight. In order to improve significantly the situation we have launched a program to produce new samples of branching point isotopes and measure their capture cross section at the CERN n\_TOF facility. A significant amount of material (~mg) of  ${}^{147}$ Pm [t<sub>1/2</sub>=2.6y],  ${}^{171}$ Tm [t<sub>1/2</sub>=1.9y] and  ${}^{204}$ Tl [t<sub>1/2</sub>=3.8y]  $(^{79}$ Se is also foreseen in the near future) has been produced by irradiations of highly enriched precursors in the ILL high-flux reactor and this material serves for preparing high purity samples at PSI. The experiment proposals have been already approved and the capture cross section measurements are scheduled in 2014 and 2015.

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## 1. Introduction and Motivation

The s- and r-processes are responsible for the formation 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 (see Figure 1). For a recent and comprehensive review see Ref. [3].

A quantitative description of the abundances arising from the s-process requires both, the neutron capture rates and the  $\beta$ -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 means of the time-of-flight technique, which represents the most accurate method for energy-dependent neutron capture studies in the full stellar energy-range of relevance. One of the main objectives of the forthcoming experimental campaign at CERN n\_TOF is the measurement, for the first time, of the capture cross section of three additional branching point isotopes, with relevance for the understanding of the environment conditions and for a cosmo-chronology study of the *s*-process contribution to the solar system composition.



*Figure 1. Example of the s-process path and branching around A=204-205.* 

As mentioned above, the neutron capture cross section of s-process branching points has been measured by time-of-flight only in two cases: <sup>63</sup>Ni [4,5] and <sup>151</sup>Sm [6]. For all other cases, the cross section values used for calculating abundances in stellar models are purely based on theoretical predictions (see KADoNiS [7], where typically, discrepancies between a factor of 2 to 3 are found between theoretical predictions). Within the EC NeutAndalus project there is the opportunity of making a big step forward in the field, by making three samples of the corresponding s-process branching points <sup>147</sup>Pm, <sup>171</sup>Tm and <sup>204</sup>Tl (see [3] and [8] for details on the importance of these particular isotopes) and measure their capture cross section at the CERN n\_TOF facility [9]. These will be the first ever time-of-flight measurement for these isotopes.

### 2. Production of isotopes at ILL and sample preparation at PSI

The production of the isotopes of interest is achieved in this case by neutron capture reactions on stable isotopes and, in some case, the subsequent beta decay. The irradiation has taken place during eight weeks in the high-flux reactor of ILL (France) [10], which features the highest thermal neutron flux in Europe. We have irradiated pressed powder pellets of 5 mm in diameter made of <sup>146</sup>Nd, <sup>170</sup>Er and <sup>203</sup>Tl in order to produce <sup>147</sup>Pm, <sup>171</sup>Tm and <sup>204</sup>Tl through the following reactions and reaction chains:

The irradiated pellets have already been shipped from ILL to PSI (Switzerland) where they are now being transformed into suitable samples for neutron capture experiments. In particular, <sup>147</sup>Pm and <sup>171</sup>Tm will be chemically separated from the corresponding matrix of Nd and Er target material, and the resulting material in non-carrier-added quality will be electrodeposited on two thin (7  $\mu$ m) aluminum backings that will then be sandwiched in order to have "encapsulated" samples with negligible risk of contamination during handling. In the case of <sup>204</sup>Tl, chemical separation is not possible and thus the pellet, with a  $\beta^-$  (E<sub>max</sub>=763 keV) activity of ~200 GBq, will be encapsulated in a 1 mm thick aluminum capsule that will reduce the corresponding beta dose.

The isotopic composition of all the samples will be determined by means of Inductively Coupled Plasma Mass Spectrometry (ICP-MS) at PSI. For the cases of <sup>147</sup>Pm and <sup>171</sup>Tm, a comparison with standard solutions will provide also the precise mass of the isotopes of interest.

A summary of the expected sample's composition, masses and geometries is given in Table 1.

Isotope of	Sample	Mass (atoms) of the	Geometry
interest	composition	isotope of interest	
<sup>147</sup> Pm	$^{147}Pm_2O_3$	$0.3 \text{ mg} (1.2 \cdot 10^{18})$	20 mm diameter deposited on $2x7 \ \mu m$
			aluminum
<sup>171</sup> Tm	$^{171}\text{Tm}_2\text{O}_3$	$3.6 \text{ mg} (1.3 \cdot 10^{19})$	20 mm diameter deposited on 2x7 μm
			aluminum
<sup>204</sup> Tl	$Tl_2O_3$	$11 \text{ mg} (3.2 \cdot 10^{19})$	5 mm diameter thick pressed pellet
	(5% of <sup>204</sup> Tl )		encapsulated in 1 mm thick aluminum

Table 1. Summary sample's composition, masses and geometries (expected values as of Summer 2014).

### 3. Planned experiments at the CERN n\_TOF facility

The neutron time-of-flight facility [9] at CERN features two neutron beam lines with 185 (EAR1) and 19 meters (EAR2) long flight paths, having EAR2 a neutron flux ~30 times more intense than that at EAR1.

The measurements of the neutron capture cross sections of <sup>171</sup>Tm and <sup>204</sup>Tl have been proposed in EAR1 and the one of <sup>147</sup>Pm, where less than 1 mg is available, will require the higher neutron flux of EAR2. The measurement proposals have been approved by the CERN Scientific Committee and will take place between 2014 and 2015. The measurements will be carried out using low neutron sensitivity  $C_6D_6$  scintillators (made of carbon fiber, see Figure 2) in combination with the Pulse Height Weighting Technique [11]. The combination of good neutron resolution of the n\_TOF-EAR1 and the low background in the  $C_6D_6$  detectors should give access to the full resolved resonance region of the associated capture cross sections, while the investigation of the unresolved resonance region (above 1-30 keV, depending on the isotope) will be challenging due to the peak-to-background conditions in that energy range. The outcome from the experiment with <sup>147</sup>Pm at n\_TOF-EAR2 is still uncertain, as the beam line is still under commissioning. However we expect at least to determine the average level spacing and resonance strength so that these values can serve as input for Hauser-Feshbach calculations.



Figure 2. Picture of the new low neutron sensitivity  $C_6D_6$  detectors at n\_TOF.

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