

## Time-of-flight and activation experiments on $^{147}\text{Pm}$ and $^{171}\text{Tm}$ for astrophysics

C. Guerrero<sup>1,a</sup>, J. Lerendegui-Marco<sup>1</sup>, C. Domingo-Pardo<sup>2</sup>, A. Casanovas<sup>3</sup>, R. Dressler<sup>4</sup>, S. Halfon<sup>5</sup>, S. Heinitz<sup>4</sup>, N. Kivel<sup>4</sup>, U. Köster<sup>6</sup>, M. Paul<sup>7</sup>, J.M. Quesada-Molina<sup>1</sup>, D. Schumann<sup>4</sup>, A. Tarifeño-Saldivia<sup>3</sup>, M. Tessler<sup>7</sup>, L. Weissman<sup>5</sup>, O. Aberle<sup>8</sup>, J. Andrzejewski<sup>9</sup>, L. Audouin<sup>10</sup>, M. Bacak<sup>8,12</sup>, J. Balibrea<sup>13</sup>, M. Barbagallo<sup>14</sup>, F. Becvar<sup>15</sup>, E. Berthoumieux<sup>12</sup>, J. Billowes<sup>16</sup>, D. Bosnar<sup>17</sup>, A. Brown<sup>18</sup>, M. Caamaño<sup>19</sup>, F. Calviño<sup>3</sup>, M. Calviani<sup>8</sup>, D. Cano-Ott<sup>13</sup>, R. Cardella<sup>8</sup>, F. Cerutti<sup>8</sup>, Y.H. Chen<sup>10</sup>, E. Chiaveri<sup>1,8</sup>, N. Colonna<sup>14</sup>, G. Cortés<sup>3</sup>, M.A. Cortés-Giraldo<sup>1</sup>, L. Cosentino<sup>20</sup>, L.A. Damone<sup>14,21</sup>, M. Diakaki<sup>12</sup>, E. Dupont<sup>12</sup>, I. Durán<sup>19</sup>, B. Fernández-Domínguez<sup>19</sup>, A. Ferrari<sup>8</sup>, P. Ferreira<sup>22</sup>, P. Finocchiaro<sup>20</sup>, K. Göbel<sup>23</sup>, A.R. García<sup>13</sup>, A. Gawlik<sup>9</sup>, S. Gilardoni<sup>8</sup>, T. Glodariu<sup>24</sup>, I.F. Gonçalves<sup>22</sup>, E. González<sup>13</sup>, E. Griesmayer<sup>11</sup>, F. Gunsing<sup>8,12</sup>, H. Harada<sup>25</sup>, J. Heyse<sup>26</sup>, D.G. Jenkins<sup>18</sup>, E. Jericha<sup>11</sup>, F. Käppeler<sup>27</sup>, Y. Kadi<sup>8</sup>, A. Kalamara<sup>28</sup>, P. Kavragin<sup>11</sup>, A. Kimura<sup>25</sup>, N. Kivel<sup>18</sup>, M. Kokkoris<sup>28</sup>, M. Krťicka<sup>15</sup>, D. Kurtulgil<sup>20</sup>, E. Leal-Cidoncha<sup>19</sup>, C. Lederer<sup>29</sup>, H. Leeb<sup>11</sup>, S. Lo Meo<sup>30,31</sup>, S.J. Lonsdale<sup>26</sup>, D. Macina<sup>8</sup>, J. Marganiec<sup>2,29</sup>, T. Martínez<sup>13</sup>, A. Masi<sup>1</sup>, C. Massimi<sup>30,31</sup>, P. Mastinu<sup>34</sup>, M. Mastromarco<sup>7</sup>, E.A. Mauger<sup>4</sup>, A. Mazzone<sup>14,35</sup>, E. Mendoza<sup>13</sup>, A. Mengoni<sup>30</sup>, P.M. Milazzo<sup>36</sup>, F. Mingrone<sup>8</sup>, A. Musumarra<sup>20,37</sup>, A. Negret<sup>24</sup>, R. Nolte<sup>32</sup>, A. Oprea<sup>24</sup>, N. Patronis<sup>38</sup>, A. Pavlik<sup>39</sup>, J. Perkowski<sup>2</sup>, I. Porras<sup>40</sup>, J. Praena<sup>40</sup>, D. Radeck<sup>32</sup>, T. Rauscher<sup>41,42</sup>, R. Reifarh<sup>23</sup>, P.C. Rout<sup>43</sup>, C. Rubbia<sup>8</sup>, J.A. Ryan<sup>16</sup>, M. Sabaté-Gilarte<sup>1,8</sup>, A. Saxena<sup>43</sup>, P. Schillebeeckx<sup>26</sup>, A.G. Smith<sup>16</sup>, N.V. Sosnin<sup>16</sup>, A. Stamatopoulos<sup>28</sup>, G. Tagliente<sup>14</sup>, J.L. Tain<sup>2</sup>, L. Tassan-Got<sup>10</sup>, A. Tsinganis<sup>28</sup>, S. Valenta<sup>15</sup>, G. Vannini<sup>30,33</sup>, V. Variale<sup>14</sup>, P. Vaz<sup>22</sup>, A. Ventura<sup>31</sup>, V. Vlachoudis<sup>8</sup>, R. Vlastou<sup>28</sup>, A. Wallner<sup>44</sup>, S. Warren<sup>16</sup>, C. Weiss<sup>8</sup>, P.J. Woods<sup>29</sup>, T. Wright<sup>16</sup>, P. Žugec<sup>8,17</sup>, and the n\_TOF Collaboration

<sup>1</sup> Universidad de Sevilla, Sevilla, Spain

<sup>2</sup> Instituto de Fisica Corpuscular IFIC, Valencia, Spain

<sup>3</sup> Universitat Politècnica de Catalunya UPC, Barcelona, Spain

<sup>4</sup> Paul Scherrer Institute PSI, Villigen, Switzerland

<sup>5</sup> Soreq Nuclear Research Center, Yavne, Israel

<sup>6</sup> Institut Laue-Langevin ILL, Grenoble, France

<sup>7</sup> Hebrew University, Jerusalem

<sup>8</sup> European Organization for Nuclear Research (CERN), Switzerland

<sup>9</sup> University of Lodz, Poland

<sup>10</sup> Institut de Physique Nucléaire, CNRS-IN2P3, Univ. Paris-Sud, Université Paris-Saclay, 91406 Orsay Cedex, France

<sup>11</sup> Technische Universität Wien, Austria

<sup>12</sup> CEA Saclay, Irfu, Gif-sur-Yvette, France

<sup>13</sup> Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT), Spain

<sup>14</sup> Istituto Nazionale di Fisica Nucleare, Sezione di Bari, Italy

<sup>15</sup> Charles University, Prague, Czech Republic

<sup>16</sup> University of Manchester, UK

<sup>17</sup> University of Zagreb, Croatia

<sup>18</sup> University of York, UK

<sup>19</sup> University of Santiago de Compostela, Spain

<sup>20</sup> INFN Laboratori Nazionali del Sud, Catania, Italy

<sup>21</sup> Dipartimento di Fisica, Università degli Studi di Bari, Italy

<sup>22</sup> Instituto Superior Técnico, Lisbon, Portugal

<sup>23</sup> Goethe University Frankfurt, Germany

<sup>24</sup> Horia Hulubei National Institute of Physics and Nuclear Engineering, Romania

<sup>25</sup> Japan Atomic Energy Agency (JAEA), Tokai-mura, Japan

<sup>26</sup> European Commission, Joint Research Centre, Geel, Retieseweg 111, 2440 Geel, Belgium

<sup>27</sup> Karlsruhe Institute of Technology, Campus North, IKP, 76021 Karlsruhe, Germany

<sup>28</sup> National Technical University of Athens, Greece

<sup>29</sup> School of Physics and Astronomy, University of Edinburgh, UK

<sup>30</sup> Agenzia nazionale per le nuove tecnologie (ENEA), Bologna, Italy

<sup>31</sup> Istituto Nazionale di Fisica Nucleare, Sezione di Bologna, Italy

<sup>a</sup>e-mail: [cguerrero4@us.es](mailto:cguerrero4@us.es)

- <sup>32</sup> Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany  
<sup>33</sup> Dipartimento di Fisica e Astronomia, Università di Bologna, Italy  
<sup>34</sup> Istituto Nazionale di Fisica Nucleare, Sezione di Legnaro, Italy  
<sup>35</sup> Consiglio Nazionale delle Ricerche, Bari, Italy  
<sup>36</sup> Istituto Nazionale di Fisica Nucleare, Sezione di Trieste, Italy  
<sup>37</sup> Dipartimento di Fisica e Astronomia, Università di Catania, Italy  
<sup>38</sup> University of Ioannina, Greece  
<sup>39</sup> University of Vienna, Faculty of Physics, Vienna, Austria  
<sup>40</sup> University of Granada, Spain  
<sup>41</sup> Department of Physics, University of Basel, Switzerland  
<sup>42</sup> Centre for Astrophysics Research, University of Hertfordshire, UK  
<sup>43</sup> Bhabha Atomic Research Centre (BARC), India  
<sup>44</sup> Australian National University, Canberra, Australia

**Abstract.** The neutron capture cross section of several key unstable isotopes acting as branching points in the *s*-process are crucial for stellar nucleosynthesis studies, but they are very challenging to measure due to the difficult production of sufficient sample material, the high activity of the resulting samples, and the actual  $(n,\gamma)$  measurement, for which high neutron fluxes and effective background rejection capabilities are required. As part of a new program to measure some of these important branching points, radioactive targets of  $^{147}\text{Pm}$  and  $^{171}\text{Tm}$  have been produced by irradiation of stable isotopes at the ILL high flux reactor. Neutron capture on  $^{146}\text{Nd}$  and  $^{170}\text{Er}$  at the reactor was followed by beta decay and the resulting matrix was purified via radiochemical separation at PSI. The radioactive targets have been used for time-of-flight measurements at the CERN n\_TOF facility using the 19 and 185 m beam lines during 2014 and 2015. The capture cascades were detected using a set of four  $\text{C}_6\text{D}_6$  scintillators, allowing to observe the associated neutron capture resonances. The results presented in this work are the first ever determination of the resonance capture cross section of  $^{147}\text{Pm}$  and  $^{171}\text{Tm}$ . Activation experiments on the same  $^{147}\text{Pm}$  and  $^{171}\text{Tm}$  targets with a high-intensity 30 keV quasi-Maxwellian flux of neutrons will be performed using the SARAF accelerator and the Liquid-Lithium Target (LiLiT) in order to extract the corresponding Maxwellian Average Cross Section (MACS). The status of these experiments and preliminary results will be presented and discussed as well.

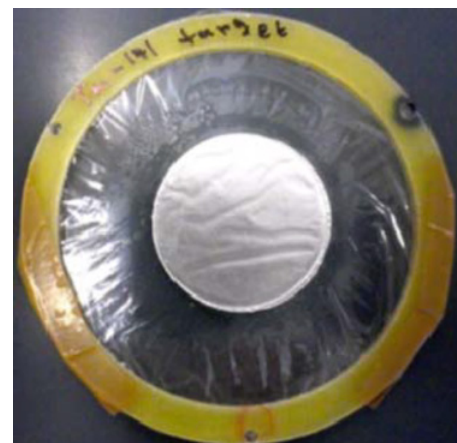
## 1. Introduction

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].

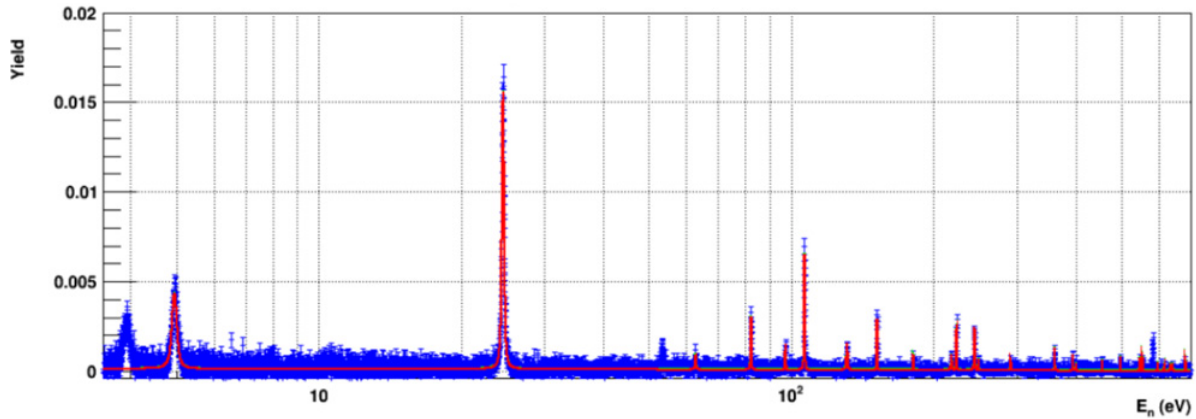
A quantitative description of the abundances arising from the *s*-process requires both, the neutron capture rates and the  $\beta$ -decay probabilities of all the isotopes involved. Along the *s*-process path, unstable nuclei with relatively long ( $\gamma$ ) and with very long (Gy) half-lives, known as branching point isotopes, become of utmost interest: their destruction via either beta decay or neutron capture depends on the conditions of the environment (density, temperature). Hence the importance of knowing the corresponding capture cross sections. Despite of their pivotal role, as of today, only the capture cross section of

2 out of a list of 21 important *s*-process branching points isotopes (see [3]) have been measured by neutron time-of-flight.

In this work, we add three more items to the list of measured isotopes. We have produced sizable quantities of  $^{147}\text{Pm}$ ,  $^{171}\text{Tm}$  and  $^{204}\text{Tl}$  inside the ILL high flux reactor, purified the material, made suitable targets out it [4], and measured the corresponding capture cross section by time-of-flight at the CERN n\_TOF facility [5,6] and by activation at LiLiT [7]. Since the data analysis is ongoing, this paper does not include final results but a description of the experiments and an outlook of the analysis and expected results.



**Figure 1.** Picture of the  $^{171}\text{Tm}$  target.



**Figure 2.** Experimental capture yield of the  $^{171}\text{Tm}(n,\gamma)$  measurement at CERN n\_TOF.

## 2. Experiments

### 2.1. Production of the radioactive targets

The isotopes  $^{147}\text{Pm}$ ,  $^{171}\text{Tm}$  and  $^{204}\text{Tl}$  have been produced by neutron irradiation at the high flux reactor at Institute Laue-Langevin (ILL), Grenoble of 98.2 mg of  $^{146}\text{Nd}_2\text{O}_3$  enriched to 98.8%, 238 mg of  $^{170}\text{Er}_2\text{O}_3$  enriched to 98.1%, and 263 mg of  $^{203}\text{Tl}_2\text{O}_3$  enriched to 99.5%. The powder of each isotope was pressed into pellets, each of which was then encapsulated into a high purity quartz ampule sealed by a flame torch. These ampules were irradiated at ILL for a period of 55 days with an average neutron flux of  $8.2 \times 10^{14}$  n/cm<sup>2</sup>/s and, after a cooling period of approximately 1.5 years, the samples were shipped to PSI where they underwent chemical processing.

While the  $^{204}\text{Tl}$  target was left inside the quartz ampule for the subsequent measurements due to its prohibitive dose rate, the irradiated Nd (150 GBq) and Er (3 GBq) pellets were chemically purified prior to making suitable targets. The material was electroplated onto 5 μm thick aluminum backings resulting in two high quality targets of 22 mm diameter with a total of 3.8 mg of  $^{171}\text{Tm}$  and 85 μg of  $^{147}\text{Pm}$ . A picture of one of the targets is shown in Fig. 1.

### 2.2. Time-of-flight experiments at n\_TOF

The CERN time-of-flight facility features two neutron beam lines: a shorter one with increased neutron flux at only 19 m [5] and a longer one with better energy resolution at 185 m [6]. Both beam lines look at the neutrons produced by spallation induced by a pulsed 20 GeV/c proton beam impinging on a cylindrical lead block every (at best) 1.2 seconds.

After traveling through the chosen beam line, a fraction of the neutrons incident on the target under study ( $^{147}\text{Pm}$ ,  $^{171}\text{Tm}$  and  $^{204}\text{Tl}$  in this case) undergoes neutron capture reactions, and the subsequent  $\gamma$ -rays are detected by an array of four C<sub>6</sub>D<sub>6</sub> liquid scintillators [8]. These are detectors with very low neutron sensitivity that allow one eliminating the background due to neutrons scattered in the target. After the corresponding background subtraction, the measured distributions of capture counts as a function of time-of-flight, i.e., neutron energy, are transformed into the capture yield applying the so-called Pulse Height Weighting Technique (PHWT) [9] and using the saturated resonance of  $^{197}\text{Au}$  for an absolute normalization [10].

### 2.3. Activation experiments at LiLiT

The Liquid Lithium Target (LiLiT) [7] installed at the SARAF facility (Israel) represents the most intense quasi-Maxwellian neutron beam worldwide. The SARAF accelerator provides a proton beam of 1–2 mA with an energy of  $\sim 1.93$  MeV (just above the threshold of the  $^7\text{Li}(p,n)$  reaction) that is driven into a thin (1.5 mm) liquid lithium layer, hence providing the quasi-Maxwellian neutron energy distribution (see [11] for details). At LiLiT, Maxwellian Averaged Cross Sections (MACS) are measured via the activation technique. The targets are exposed to the neutron beam and the number of capture reactions is determined from the number of  $^{A+1}\text{Z}$  nuclei produced. Assuming that the  $^{A+1}\text{Z}$  isotope is radioactive, the number of unstable nuclei is quantified using a Ge detector looking at the associated emission of  $\gamma$ -rays. In this case, the MACS of  $^{197}\text{Au}$  serves as a reference.

## 3. Preliminary and expected results

The time-of-flight measurements of both  $^{171}\text{Tm}$  and  $^{204}\text{Tl}$  were carried out at the n\_TOF long neutron beam line, EAR1. In both cases, despite of the severe background conditions arising for the high activity of the targets, we have obtained a nice data set that allows resolving capture resonances for the first time.

In the case of  $^{171}\text{Tm}$  the preliminary capture yield is displayed in Fig. 2, showing resonances up to 700 eV and illustrating the good resolution of n\_TOF-EAR1. This data set will provide a complete set of resonance parameters and the unresolved resonance region will be derived from these with the help of the Hauser-Feshbach statistical model. In the case of  $^{204}\text{Tl}$  the observed resonance are actually in the keV region of interest in astrophysics. In the case of  $^{147}\text{Pm}$ , the mass of the target is so small (85 μg) that it is at the detection limit of the n\_TOF short beam line, EAR2. In this case only a few resonances have been observed and therefore the data will not allow extracting a cross section value in the keV energy region of interest.

In the case of the activation measurements, both  $^{147}\text{Pm}(n,\gamma)$  and  $^{171}\text{Tm}(n,\gamma)$  have been successfully measured at LiLiT. Indeed, due to the high neutron beam intensity at LiLiT we have significantly increased the

statistics achieved in the previous experiment and will therefore be able to use more  $\gamma$ -ray lines, improving the accuracy and increasing the reliability of our results. The results will be the measured MACS at 30 keV with an expected accuracy of 10%.

The authors acknowledge financial support by Spanish FPA2013-45083-P and FPA2014-53290-C2-2-P projects and the EC FP7 projects NeutAndalus (Grant No. 334315) and CHANDA (Grant No. 605203).

## References

- [1] E. Burbidge, G. Burbidge, W. Fowler, F. Hoyle, *Rev. Mod. Phys.* **29** (1957)
- [2] A. Cameron, A.E.C.L. Chalk River, Canada, Technical Report No. CRL-41 (1957)
- [3] F. Käppeler, R. Gallino, S. Bisterzo, Wako Aoki, *Rev. Mod. Phys.* **83** (2011)
- [4] S. Heinritz, et al., in preparation
- [5] C. Weiss, et al., *Nucl. Instrum. Meth. A* **799** (2015)
- [6] C. Guerrero, et al., *Eur. Phys. J. A* **49**, 27 (2013)
- [7] S. Halfon, et al., *Rev. Sci. Instrum.* **85** (2014)
- [8] P.F. Mastinu, et al., CERN-n.TOF-PUB-2013-002
- [9] U. Abbondanno, et al., *Nucl. Instrum. Meth. A* **521** (2004)
- [10] R. Macklin, J. Halperin, R. Winters, *Nucl. Instrum. Meth. A* **164** (1979)
- [11] W. Ratynski, F. Käppeler, *Phys. Rev. C* **37** (1988)