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Measurement of the radiative capture cross section of the *s*-process *branching points* ^{204}Tl and ^{171}Tm at the n_TOF facility (CERN)

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Abstract. The neutron capture cross section of some unstable nuclei is especially relevant for s-process nucleosynthesis studies. This magnitude is crucial to determine the local abundance pattern, which can yield valuable information of the *s*-process stellar environment. In this work we describe the neutron capture (n,γ) measurement on two of these nuclei of interest, ^{204}Tl and ^{171}Tm, from target production to the final measurement, performed successfully at the n_TOF facility at CERN in 2014 and 2015. Preliminary results on the ongoing experimental data analysis will also be shown. These results include the first ever experimental observation of capture resonances for these two nuclei.

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 main features and basic characteristics of both processes were already well sketched in the seminal papers by Burbidge et al. [1] and Cameron [2]. An up to date review of the *s*-process, including the main stellar sites where it takes place, can be found in Käppeler et al. [3].The *s*-process mechanism operates during core He-burning and shell C-burning in massive stars of $M>8M_{sun}$ (also called *weak s*-process), as well as in H-burning and He-burning layers of Thermally-Pulsing low-mass stars (1-3 M_{sun}) of the Asymptotic Giant Branch (TP-AGBs), in what is known as the *main s*-process. While the weak *s*-process is the main contributor to the abundances of the elements from Fe to Sr, the main *s*-process is the dominant source for elements of A>90.

Any reliable model of the *s*-process nucleosynthesis aims to reproduce faithfully the solar elemental abundances, and for this requires a precise knowledge of the neutron capture cross sections and, in some cases also the beta decay rates, of ideally all nuclei involved. Among all these nuclei, of particular importance are some nuclides which are radioactive, with half-lives from years to Gy. This means that during the *s*-process its decay process competes with the neutron capture; since they effectively split the *s*-process flow, these nuclides are known as *branching points*. The determination of the neutron capture cross section of these nuclei is crucial to determine the elemental abundances around the branching point, and also enables us to give some constraints on several variables of the *s*-process stellar environment, such as temperature, neutron density or pressure. Their radioactive nature, which is what makes them relevant, is also what makes the measurement of the capture cross section especially challenging. In this work we describe the first ever capture measurement on two of these branching points, ^{171}Tm and ^{204}Tl, including the target preparation, the experimental setup, as well as preliminary results for both nuclei. Preliminary results for ^{203}Tl(n,γ), necessary for the ^{204}Tl data analysis procedure, are also shown.

2. Target preparation

The ^{204}Tl and ^{171}Tm samples were produced by neutron irradiation, at the high neutron flux of the nuclear reactor at ILL. Samples of 263 mg of $^{203}Tl_2O_3$, enriched to 99.5%, and 238 mg of $^{170}Er_2O_3$, enriched to 98.1% were used as primary materials. These samples had been pressed into cylindrical pellets at PSI, and enclosed in quartz ampoules to make them suitable for irradiation at ILL. While the Er irradiated seed could be chemically purified and then electroplated to produce a 22 mm diameter target containing 3.47 mg of ^{171}Tm, the ^{204}Tl had to be left untouched inside the ampoule due to the very high beta activity of the sample, 200 GBq. By the time of the experiment the sample contained 9 mg of ^{204}Tl (4% relative Tl concentration), plus 4 mg (2%) of its daughter ^{204}Pb. An impurity of 370kBq of ^{60}Co was also present in the ^{204}Tl sample. Additionally, since the precise shape and spatial distribution of the $^{203\text{-}204}$Tl sample inside the ampoule was uncertain after irradiation, a gamma scanning procedure of the sample was performed prior to the capture measurement [4].

3. Capture measurement

Both ^{171}Tm and ^{204}Tl capture measurements were carried out in the Experimental Area 1 (EAR1) of the n_TOF facility at CERN [5], in the fall of 2014 and summer of 2015, respectively. At n_TOF a pulsed neutron beam is produced by spallation of a 20 GeV/c pulsed proton beam from the PS impinging on a massive lead target. The energy of the neutrons is determined with a high resolution via the time-of-flight technique. A set of 4 C_6D_6 liquid scintillation detectors were used to detect the prompt capture γ-rays. These detectors are optimized for a very low sensitivity to the neutrons scattered by the sample. In the case of the ^{204}Tl, the high beta activity, together with the high decay energy of these electrons (763 keV), produced an additional and very intense gamma-ray background due to bremsstrahlung in the quartz ampoule. This made it necessary to shield the detectors with a 2 mm thick lead foil, which also helped to overcome a few other experimental effects, like possible shifts in detector gain observed during the ^{171}Tm campaign (up to 15%) or in the ^{241}Am(n,γ) measurement [6].

The counting rate TOF (or neutron-energy equivalent) spectra were transformed, after background subtraction, into capture yield by applying the so-called Pulse Height Weighting Technique (PHWT) [7]. The gold saturated resonance method [8] was employed for absolute yield normalization. For the analysis of the ^{204}Tl measurement the special sample details, such as the quartz encapsulation and the uncertain geometry of the activated material involved also the additional measurement of a $^{203}Tl_2O_3$ pure sample, nearly identical to the one irradiated.

4. Preliminary results

The capture yield analysis in the Resolved Resonance Region (RRR) is analysed and parameterized using the Bayesian R-matrix analysis code SAMMY [9]. In the case of ^{171}Tm, it has been possible to analyse the RRR up to 700 eV, which allowed to extract individual resonance parameters for 28 *s*-wave resonances for the first time ever [10].

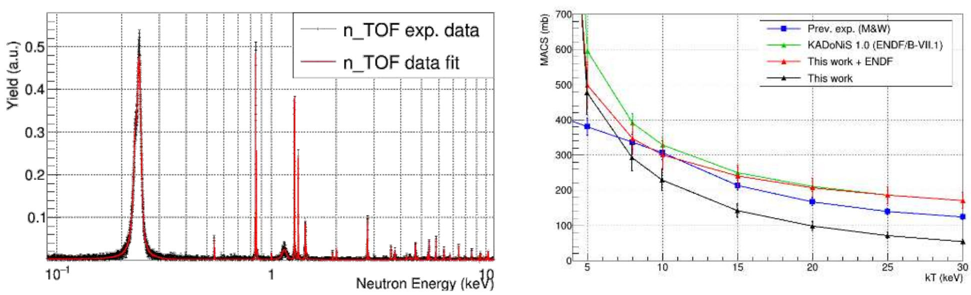

Figure 1. *Left:* Capture Yield analysis for the ^{203}Tl(n,γ) reaction. *Right*: In black, the MACS obtained employing the new resonance data from 30 eV up to 26 keV. This is compared to the MACS calculated with the ENDF evaluation data (green), and the MACS obtained in the experimental measurement by Macklin and Winters (blue).

For the analysis of the $^{203\text{-}204}$Tl sample (n,γ) yield, the first step was to analyze the ^{203}Tl pure sample yield with the highest possible accuracy, in order to reliably account for it in the later analysis of the ^{204}Tl(n,γ). In ^{203}Tl(n,γ) more than 70 resonances, from 37 eV up to 26 keV, have been identified and analysed with SAMMY. This included the first experimental measurement for resonances at a neutron energy lower than 3 keV (fig. 1). These new

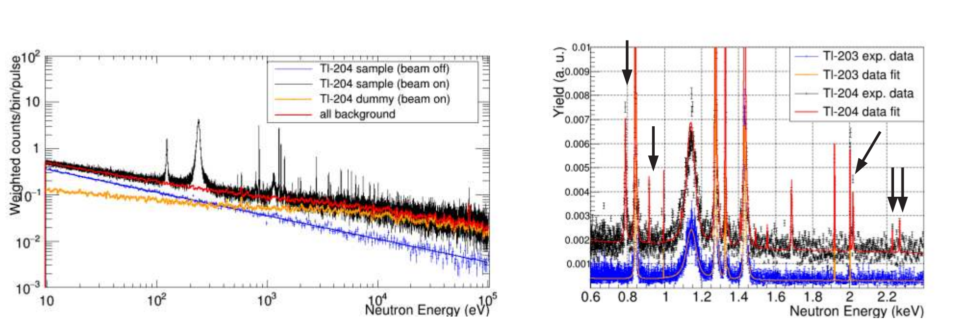

Figure 2. *Left:* Counting rate vs neutron energy spectra for the $^{203\text{-}204}$Tl(n,γ) measurement, showing the different background contributions. *Right*: Comparison of the ^{203}Tl pure sample and $^{203\text{-}204}$Tl sample capture yields, with arrows indicating the ^{204}Tl(n,γ) resonances identified up to now.

resonances have a significant impact in the Maxwellian Averaged Cross Section (MACS) at 5-8 keV when compared to the commonly accepted MACS in KADoNiS v0.3 [11], which is based on the previous measurement [12]. Finally, the analysis of the ^{204}Tl sample is ongoing; at the present time, six resonances of ^{204}Tl in the energy range from 122 eV up to 2.2 keV have been identified for the first time (fig. 2, right). A few additional candidates are under evaluation.

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