## EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

## Neutron capture cross section measurements by the activation method at the n\_TOF NEAR Station

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#### Abstract

We propose to measure the spectral averaged neutron capture cross sections (SACS) of a series of isotopes by means of the activation technique in the newly commissioned NEAR station at n\_TOF. The main aim of the measurement is to verify that, with a suitable filtering of the neutron beam, the Maxwellian averaged cross section (MACS) at different temperatures can be determined with a reasonable accuracy as suggested by dedicated simulations and calculation.

To validate the use of the activation technique in the NEAR station for astrophysics-related measurements, we propose to irradiate five isotopes whose MACS at different temperatures have recently been determined with high accuracy at n\_TOF, from energy-differential cross sections measured in EAR1 and/or EAR2. In this respect the best candidates are:  $^{76}$ Ge,  $^{89}$ Y,  $^{94}$ Zr and  $^{140}$ Ce, as well as  $^{197}$ Au standard that will be used both as benchmark and reference target. A  $^{10}$ B-enriched B4C filtering system placed on the neutron beam in the NEAR station will be used to produce Mawellian-like spectra at different temperatures. As the neutron capture on  $^{89}$ Y produces a  $\beta$ -only emitter, we will test the possibility to perform activation measurements on such class of isotopes. The results of the proposed measurements could open the way to challenging measurements of MACS by the activation technique at n\_TOF, for rare and/or exotic isotopes of interest for Nuclear Astrophysics.

**Experimental Area**: NEAR **Requested protons**: none (7.0x10<sup>18</sup> protons used in parallel with EAR1 and EAR2 runs)

#### Introduction

All elements from iron up to uranium in the Universe are produced by sequences of neutron capture and beta-decay nuclear reactions in two main stellar scenarios, each being responsible for the production of about one half of the observed elemental abundances. Slow neutron capture process (or s-process in short), which mainly produces nuclei within the mass range of  $70 \le A \le 210$ , occurs during the advanced burning phases of stellar evolution [1]. Depending on the stellar mass, the s process operates in thermally pulsing low-mass Asymptotic Giant Branch (AGB) stars (main component) [2] or during core He and shell C burning in massive stars (weak component) [3]. The s-process is responsible for the production of one half of the heavy element abundance. The remaining half originates in explosive nucleosynthesis (occurring in supernovae events and/or neutron star mergers), where short-lived and very neutron-rich nuclei are produced via the rapid neutron capture called r-process [1], which later decay to the known stable isotopes. Recent works suggest that a likely source of r-process elements could be the catastrophic aftermath of mergers between neutron stars, which are the superdense cores of stars left behind after cataclysmic, explosive star deaths known as supernovas [4].

The main features of the chemical evolution of the Universe, and in particular heavy element production, are reasonably well described by stellar models incorporating the two neutron capture processes mentioned above. To this end, accurate stellar cross sections are a fundamental input in order to reproduce the observed abundances. At present, the knowledge of neutron cross section is still unsatisfactory for several isotopes, in particular bottleneck nuclei, so-called because of their very small cross section that represents an obstacle to heavier element production, and branching point isotopes, unstable nuclei with short half-life, whose decay effectively competes with further neutron capture. At n\_TOF, the energy-differential cross section of both classes of isotopes has been measured by the time-of-flight technique at the two experimental areas EAR1 and EAR2. However, the intrinsic limitations of the technique, in terms of signal-to-background ratio, has hindered up to now much-needed measurements for a number of important isotopes.

To overcome such limitations, a new experimental area at n\_TOF, the NEAR station, set at a

very short distance from the spallation source, can be considered. Combined with a dedicated laboratory for gamma and beta spectroscopy, based on HPGe detectors, the new high-flux irradiation station will allow to perform high-sensitivity activation measurements, thus contributing to a substantial improvement in experimental opportunities related to astrophysics, technological issues and basic nuclear physics.

Following the construction of the area near the target-moderator assembly at n\_TOF, a first commissioning of the neutron beam exiting the collimator in the new station has been performed [5], starting in July 2021. Preliminary results indicate that the neutron flux in the NEAR station is up to the expectations. To start exploiting the newly built measuring station, it is now crucial to study its performances. With the aim of verifying the potentialities of the activation technique at n\_TOF, we propose here a series of measurements of the Spectral Averaged Cross Section (SACS) by irradiating in the NEAR station isotopes whose Maxwellian Averaged Cross Section at different temperatures is well known, having been accurately previously determined on the basis of the energy-differential time-of-flight cross section measurements.

The main idea behind the proposed measurements is that, by means of a suitable filtering setup, the energy distribution of the neutron beam in the NEAR station can be shaped so to resemble a Maxwellian-like distribution. Analytical calculations, confirmed by detailed Monte Carlo simulations performed with the GEANT4 toolkit (described below), show that indeed this can be achieved by using <sup>10</sup>B-based filters of adequate thickness, that remove the low-energy component of the original spectrum of the neutron beam entering the NEAR station. In principle, the use of a moderator to suppress (or reduce) the high-energy component ( $E_n > 1$  MeV) as well would also be necessary. An extended simulation campaign is planned, which will allow to select a proper geometry and material for a moderator, to be successively installed close to the n\_TOF target assembly. However, considering that the capture cross section typically decreases rapidly above 1 MeV, reasonable results can be obtained even without a moderator. An interesting feature at the NEAR station is that Maxwellian-like neutron spectra corresponding to different thermal energies *kT* can be obtained simply by varying the thickness of the <sup>10</sup>B-based filter.

Preliminary calculations indicate that Maxwellian Averaged Cross Sections at different temperatures can be determined by the activation technique in the NEAR station, up to 100 keV, with reasonable accuracy for most of the isotopes. We propose to verify these calculations by measuring the SACS at three different temperatures (8, 25 and 30 keV), for isotopes whose energy-differential cross sections have recently been measured in EAR1 and EAR2 at n\_TOF [6], thus allowing the accurate determination of their SACS at all temperatures. Together with <sup>197</sup>Au, that will be used both as a benchmark and reference for the other isotopes, we propose to measure by activation the spectral averaged cross section of <sup>76</sup>Ge, <sup>89</sup>Y, <sup>94</sup>Zr and <sup>140</sup>Ce. The filtering systems, and related simulations are described in detail in the next session, together with the setup for gamma and beta spectroscopy.

Among the proposed isotopes,<sup>89</sup>Y has been chosen to verify the ability of a dedicated setup to measure the activation of  $\beta$ - only emitters. In fact, the neutron capture product <sup>90</sup>Y undergoes an almost pure  $\beta$ - decay to <sup>90</sup>Zr with a half-life of 64.1 h and a decay energy of 2.28 MeV, corresponding to an average beta energy of 0.934 MeV. The induced <sup>90</sup>Y activity will be

measured by placing the sample in close geometry between a HPGe detector (with a carbon epoxy window) and a plastic scintillator detector. Through this configuration, the covered solid angle for both detectors will be maximized. Accordingly, the excellent energy resolution of the HPGe detector will be combined with the high detection efficiency of betas in the plastic detector. In this way, the ability to perform direct activation measurements of MACS, even in those cases where the product nucleus is a beta-only emitter, will be confirmed.

#### **Monte Carlo simulations**

The neutron energy distribution at the irradiation position was estimated by Monte Carlo GEANT4 simulations. The geometry of the irradiated object consists of one or more samples contained in the center of a cylinder made of boron carbide (B4C) highly enriched in <sup>10</sup>B (commercially available from different providers, see for instance <u>https://www.3m.com/</u>). A study of the neutron energy distribution as a function of the cylinder's thickness, in the range from 0.5 to 2 cm for a fixed radius of 5 cm was performed. In this configuration the cylinder can house several samples of 1 cm radius (Figure 1). The neutron energy distribution inside the boron carbide changes with position, along its radius and thickness. To reduce this effect, the samples are located in the same plane (i.e. experiencing the same filter thickness) with respect to the beam axis and symmetrically from the axis. The used Monte Carlo simulation included the concrete walls and the objects in and around the NEAR station, so as to take into account the effect of the environmental shielding materials.



Figure 1: Schematic view of the setup with the filter material and sample. On the left figure, the exit of the collimator into the NEAR Station area is shown with the support for the sample+filter. On the right panel the sample+filter assembly, consisting of two B4C cylinders are shown, arranged to sandwich the sample in the middle.

The starting point of the simulations were the neutrons scored at the marble surface towards the irradiation point. The neutrons were sampled from a given energy and momentum distribution

provided by the CERN FLUKA team. In our simulation, the center of the irradiation point is positioned 5 cm from the marble surface, with the axis of the cylinder coincident with the beam axis. In Figure 2 the starting energy distribution at the marble position and at the sample position inside the boron carbide cylinder are shown.



Figure 2: Neutron energy distribution at NEAR station (in blue) and at the sample position after filtering inside a 1-cm thickness B4C cylinder.

The role of the B4C on the thermal and epithermal region is evident. In fact, neutrons are moderated and eventually absorbed by the filtering system. As a result, the mean energy of the distribution is shifted at higher energies and a large fraction of the lower energy neutrons is lost. The advantage is that the neutron energy distribution presents a dominant peak, roughly shaped to resemble a Maxwell-Boltzmann distribution, as showed in Figure 3. Although not completely satisfying, mainly because of the tail at high energies, this configuration can already provide strong indication on the effectiveness of the proposed setup. Future upgrades, in particular the installation of a moderator inside the spallation-target hall will improve the agreement between shaped and expected Maxwell-Boltzmann distributions.



Figure 3: Neutron energy distribution obtained at the irradiation point, using a 5 mm thickness B4C cylinder and comparison with a Maxwell-Boltzmann distribution with most probable thermal energy kT =

#### 0.15 keV.

It is important to note here that different B4C cylinder's thickness results in different mean of the energy distribution, thus enabling SACS measurements corresponding to different thermal energies. Our Monte Carlo simulations show that with B4C thickness of up to 20 mm, all ranges of thermal energies can be obtained, up to kT = 28 keV (see Figure 4).



Figure 4: Neutron energy distribution obtained at the irradiation point, using a 20 mm thickness B4C cylinder and comparison with a Maxwell-Boltzmann distribution with most probable thermal energy kT = 27.7 keV.

We have also estimated the variation of the neutron energy distribution as a function of the distance of the irradiated sample from the center of the B4C cylinder. In particular, while the total number of neutrons reduces by approximately 30% moving from the center to the position at 3 cm from the center, the shape of the energy distribution does not change appreciably, as reported in Figure 5.



### **Activity Measurements**

Following the irradiation of the samples, their induced activity will be measured in the newly developed Gamma-ray Spectroscopy Experimental Area (GEAR), located in building 802 (TT2A). In any gamma-ray measurement, the most important criteria to be met are the high detection efficiency, high energy resolution and the lowest possible background energy deposition in the detector. In order to satisfy these conditions, an electrically cooled HPGe will be utilized. With an energy resolution of the order of 2 keV at 1.33 MeV, this type of detector allows for a precise identification of the observed photo-peaks. The newest of the two HPGe detectors at n\_TOF has a relative efficiency as high as 55% and a carbon epoxy window, enabling the measurement of gamma-rays in a very wide energy spectrum ranging from 3 keV up to 10 MeV. Furthermore, to optimize the background conditions as much as possible, the detector will be installed and operated inside a specially designed lead shielding (CANBERRA 747).

For the counting rate estimation, an irradiation time of 14 days was considered  $(1.4 \times 10^{18} \text{ protons})$  in parallel with EAR1 and EAR2 runs), along with a cooling time of 4 hours between irradiation and measurement. The adopted masses of each isotope, as given in Table *1*, was determined in order to keep statistical uncertainties below 3%, in all cases. The sample-detector distance for all cases except the  ${}^{89}Y(n,\gamma)$  will be 6 cm.

Reaction	Product half-life	Mass (g)
<sup>76</sup> Ge(n,γ)	11.2 h	0.09
$^{94}$ Zr(n, $\gamma$ )	64 d	0.2
$^{140}Ce(n,\gamma)$	32.5 d	0.6
<sup>197</sup> Au(n,γ)	2.69 d	0.015
<sup>89</sup> Y(n,y)	64 h	0.14

 

 Table 1: Mass of the isotopes of interest needed to maintain the statistical uncertainty of the measurement below 3%.

In summary, the proposed measurements represent a fundamental benchmark for the measurements at the n\_TOF NEAR Station. The proposed measurements/irradiation will be performed during the standard operation runs at n\_TOF EAR1 and EAR2. Therefore, **no additional proton request** is implied by the present proposal.

The results of the proposed measurements will pave the way to perform challenging measurements of MACS of interest for Nuclear Astrophysics by the activation technique at  $n_{TOF}$ .

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