## Neutron research at the N\_TOF facility (CERN): Results and perspectives

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## Neutron Research At The N\_TOF Facility (CERN): Results And Perspectives

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**Abstract.** In the first ten years of operation, the neutron time-of-flight facility n\_TOF at CERN has produced a large body of new and accurate data on neutron capture and fission cross sections, relevant to Nuclear Astrophysics and advanced nuclear technologies. In the next phase, with the construction of a second experimental area with a shorted flight path, the focus of the Collaboration will partially shift from high-resolution measurements of stable or long-lived isotopes, to high-flux measurements of isotopes of relatively short half-life, very low-cross sections, or available in very small quantities. The main results obtained so far at n TOF and the future perspectives are here presented.

**Keywords:** Neutron-induced reactions, neutron time-of-flight facility, spallation neutron source, stellar nucleosynthesis, advanced nuclear reactors.

**PACS:** 26.20.Kn; 28.20.Fc; 25.85.Ec; 28.65.+a; 29.25.Dz; 28.41.Vx

#### INTRODUCTION

Neutron-induced reactions play a crucial role in various fields of fundamental and applied Nuclear Physics. In Stars, neutron capture reactions are at the basis of the production of elements heavier than Fe. On Earth, neutron-induced fission of U and Pu isotopes are responsible for energy production in nuclear reactors. Furthermore, neutron-induced reactions are a useful tool for studying fundamental properties of nuclear matter, and can be of help in various applied fields, like Nuclear Medicine, non-destructive analysis of archeological and art objects, detection of fissile material, etc...

Neutron data are being collected at various facilities around the world. Since 2001, a new installation has joined the large family of neutron time-of-flight facilities: n\_TOF at CERN, Geneva. Based on a spallation neutron source, n\_TOF was built with the aim of providing fresh new data on neutron cross sections for a variety of isotopes relevant mainly to Nuclear Astrophysics and advanced nuclear technologies for energy production and nuclear waste incineration.

In the first ten years of operation, a large body of interesting results has been obtained at n TOF by an International Collaboration of more than one hundred researchers from thirty Institutes. An extensive experimental program on capture reactions has been carried out with the aim of improving current knowledge of Stellar Nucleosynthesis of heavy elements, and gaining a better insight on stellar evolution [1]. Furthermore, high accuracy and high resolution capture cross sections have been measured for some actinides, as requested for the design of advanced nuclear waste burners and/or Generation IV fast nuclear reactors. For the same purpose, neutroninduced fission reactions of U, Pu and Minor Actinides have been, or are being measured at n TOF [2]. For most isotopes, the n TOF results are characterized by

improved accuracy and/or higher resolution and/or a wider energy range than previous data.

To expand the range of measurements, in particular to radioactive isotopes of half-life as short as a few years, a second experimental area closer to the spallation target has been proposed and will be built in the next two years [3].

In this paper, after a brief description of the n\_TOF facility and its main features, an overview of the main results obtained so far is presented, with particular emphasis on the most recent achievements. The characteristics of the second experimental area and the program foreseen for the near future is finally discussed.

### THE NEUTRON TIME-OF-FLIGHT FACILITY AT CERN

The neutron beam at n TOF is produced by spallation of 20 GeV/c protons from the CERN Proton Synchrotron accelerator on a water-cooled Pb target [4]. Two different spallation targets have been used at n TOF. The first one, cooled by a 5 cm water layer acting also as moderator of the neutron spectrum, had to be replaced after four years of operation due to damage caused in some spots by inefficient cooling. The second target, installed in 2008, was equipped with a more efficient cooling system, and with a separate moderator circuit, to allow the use of different moderating materials. This target is made of a monolithic cylindrical block of lead, with length of 40 cm and diameter of 60 cm. On the exit face of the target, cooling is ensured by a water layer of 1 cm thickness, while moderation of the neutron spectrum is performed with a 4 cm thick layer of either normal, heavy or borated water. This last material is now mostly being used to optimize the spectral features of the neutron beam and, most importantly, to minimize the production of 2.2 MeV in-beam γ-rays, that constitute the main source of background in measurements of capture cross-sections in the keV neutron energy region.

The innovative features of the n TOF neutron beam derive from the high energy of the proton beam (20 GeV/c), low duty cycle (< 0.5 Hz) and extremely high peak current of the primary proton beam (7x10<sup>12</sup> p/bunch). These features are at the origin of very high source intensity, of the order of 2×10<sup>15</sup> neutrons/pulse, which allows to obtain both a high instantaneous flux and high resolution in the experimental area, located at ~190 m from the spallation target. Two collimators, various iron and concrete shielding walls, and a sweeping magnet complete the installation. At the beginning of the second experimental campaign, the experimental area was upgraded to a "Work Sector Type A", with a series of safety and monitoring systems, so to allow measurements of high-activity samples without certified sealing. This crucial modification was essential for exploiting the full strength of the facility.

The convenient features of the neutron beam are complemented by state-of-the-art detection and data acquisition systems. The neutron flux in the experimental area is continuously monitored with a low-mass device, the SiMon detector, made of a thin foil with <sup>6</sup>Li deposit surrounded by an array of Silicon detectors outside the beam. Measurements of the energy-dependent neutron flux were performed in the commissioning phase of the new target with MicroMegas detectors equipped with <sup>10</sup>B and <sup>235</sup>U layers, as well as with a calibrated fission chamber. These measurements are periodically repeated so to be able to detect even the slightest modification with time of the spectral features of the neutron beam. A combination of the four different flux measurements has led to an accuracy on the neutron flux between 2 and 5% in a very wide neutron energy range, from thermal energy to tens of MeV.

#### THE EXPERIMENTAL PROGRAM

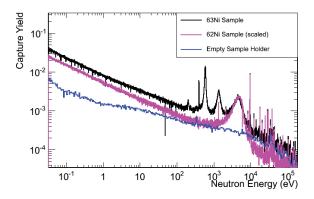
Measurements have mainly been performed at n\_TOF in two campaigns. The first one (Phase-I) was carried on between 2001 and 2004, while the current Phase-II started in 2008. The list of measured isotopes, together with a comprehensive list of publications, can be found on the n\_TOF web site [5]. Mostly, the measurements have focused on capture and fission reactions, while more recently a program on (n,charged particle) reactions has started. In the following, the main results obtained so far are discussed.

#### **Capture Reactions**

For capture measurements, two different detection systems have been set up: an array of deuterated liquid scintillator detectors ( $C_6D_6$ ), and a  $4\pi$  BaF<sub>2</sub> Total Absorption Calorimeter (TAC). The first apparatus is characterized by a low neutron sensitivity, further minimized at n TOF by the use of carbon fiber for detector housing and support. For highly radioactive and fissile isotopes, in particular for minor actinides, capture measurements are performed with the TAC, which allows the identification and rejection of background and competing reactions by reconstructing the total energy of the γ-ray cascade. The relatively large neutron sensitivity of the apparatus is reduced by inserting an inner sphere of moderating and absorbing material (based on <sup>6</sup>Li), and by encapsulating the crystals in <sup>10</sup>B-loaded carbon fiber.

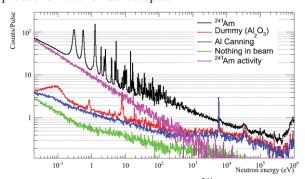
An intense program on Nuclear Astrophysics has been carried out at n TOF both in Phase I and II. In particular data have been collected with the C<sub>6</sub>D<sub>6</sub> detectors on isotopes with very small cross-sections. To this class belong <sup>139</sup>La and different Zr isotopes, that represent a bottle-neck in Stellar Nucleosynthesis, as well as some Mg and Pb isotopes. In all these cases, capture cross sections have been determined with better accuracy than in the past, thanks to the characteristics of the neutron beam and optimized neutron sensitivity of the apparatus. Recently, measurements of the capture cross-sections have also been completed for different isotopes of Fe and Ni, two elements at the beginning of the path of the s process nucleosynthesis. It should be mentioned that many of the above isotopes are also relevant to reactor technology, as part of structural material.

Among the interesting results achieved at n\_TOF, the measurement of the capture cross section of three rare Os isotopes - <sup>186,187,188</sup>Os - has allowed to reduce the uncertainty in the nuclear input of the so-called Os/Re cosmo-chronometer. More details on these results, and related publications, can be found at [6].



**FIGURE 1.** The <sup>63</sup>Ni capture yield, measured at n\_TOF with an enriched and freshly purified sample [7]. Once corrected for the contribution of the <sup>62</sup>Ni contamination and for other background components, the <sup>63</sup>Ni(n, $\gamma$ ) cross section allows the determination of Maxwellian Averaged Cross Sections at various temperatures, thus shedding some light on this first s-process branching point.

The convenient features of the n TOF facility are fully exploited in the measurement of capture cross sections of radioactive isotopes. In Nuclear Astrophysics, unstable isotopes are in some case responsible for a branching of the s-process path. Apart for affecting the production of heavier elements, branching point isotopes can provide information on the stellar environment and evolution. In this respect, interesting data have recently been obtained, for the first time with high resolution, on the capture cross section of <sup>63</sup>Ni, the first branching points in the sprocess chain [7]. The measurement was possible thanks to an enriched sample, with very low Cu contamination, purified at the Paul Sherrer Institute (PSI), Switzerland. Figure 1 shows the preliminary yield measured at n TOF. Although the sample <sup>63</sup>Ni enrichment was only 10%, the capture yield of <sup>63</sup>Ni is, except in a small region, well above the background, including the one originating from the 62Ni contamination in the sample. These data have allowed the experimental determination for the first time of the Maxwellian Average Cross Sections at various temperatures, with preliminary values up to a factor of two higher than previously thought, mostly on the basis of theoretical calculations. Such a result bears important consequences on the prediction of the production rate for Cu isotopes.



**FIGURE 2.** Preliminary results on the  $^{241}$ Am(n, $\gamma$ ) reaction measured at n\_TOF with C6D6 detectors [9]. Thanks to the high instantaneous neutron flux, the large background related to the natural radioactivity of the sample does not impede to determine the cross section for this relatively short-lived minor actinide, of interest for Accelerator Driven Systems and Generation IV fast reactors.

Together with Nuclear Astrophysics, capture cross sections play an important role in applications. In this respect, data for various U and Pu isotopes, as well as

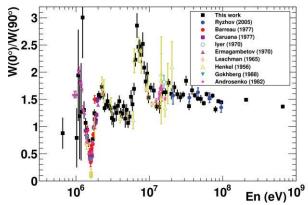
for several minor actinides, are needed for the design of advanced nuclear systems, or for the development of new fuel cycles. In the two experimental campaigns at n TOF, capture cross-sections have been measured for  $^{233,234,238}$ U,  $^{237}$ Np,  $^{240}$ Pu and  $^{241,243}$ Am. To minimize the background related to the competing fission reactions, the TAC has been used, allowing to reach uncertainties as low as 5%. A further reduction of the uncertainty, down to a 1-2% level, has recently been attempted, by combining measurements performed with the two different detection systems, the C<sub>6</sub>D<sub>6</sub> array and the TAC, a method that was proven successful in minimizing systematic uncertainties in the analysis of capture cross section of <sup>197</sup>Au in the Resolved Resonance Region [8]. Recently this method was applied to the capture cross section of <sup>241</sup>Am, a difficult measurement for current facilities, due to the relatively short half-life of this minor actinide. Figure 2 shows the preliminary yield measured with the C<sub>6</sub>D<sub>6</sub> array [9]. The high instantaneous neutron flux in this case is extremely useful in reducing to a manageable level background related to the natural radioactivity of the sample, thus allowing to obtain high quality results from thermal to several hundred keV.

#### **Fission Reactions**

As for capture, fission cross section measurements have been carried out with different detection systems. A multi-stack Fission Ionization Chamber (FIC), using in the first campaign, has been replaced in Phase II by high-performance Micromegas detectors, characterized by a better signal-to-noise ratio, thanks to some electron multiplication. A second method used at n TOF relies on the detection of both fission fragments in coincidence. To this purpose, a stack of position-sensitive Parallel Plate Avalanche Counters (PPACs) is used, which also the determination of the angular distribution of fission fragments. In all fission measurements, the ratio method is used, which consists in determining the cross section of a given isotope relative to that of 235U, a well established standard. To this purpose, reference samples of <sup>235</sup>U (and <sup>238</sup>U) are always mounted in the same detector and measured simultaneously.

The neutron-induced fission cross section of several isotopes, essentially all long-lived actinides, has been measured so far at n\_TOF, in some cases up to 1 GeV. High accuracy results are now available for <sup>232</sup>Th, <sup>233,234,236</sup>U, <sup>237</sup>Np, <sup>241,243</sup>Am and <sup>245</sup>Cm, while a measurement of <sup>240,242</sup>Pu(n,f) cross section is currently under way. The use of the ratio method allows the determination of the energy dependence of the cross section with accuracy close to 3%. In most cases, cross sections are also determined in absolute value with 3-

5% accuracy. As for capture reactions, n TOF fission data are typically characterized by a higher accuracy and/or better resolution than past measurements. The high quality of the data is also a consequence of the wide energy range, from thermal to hundreds of MeV, covered simultaneously in a single measurement, a feature that allows minimizing systematic uncertainties associated with the absolute normalization of the data. As an example, in the very difficult measurement of the <sup>245</sup>Cm fission cross section [10], normalization to recently determined thermal values allowed the determination of the absolute cross section with a 5% accuracy over more than seven decades in neutron energy. At low energy, where only two, largely discrepant data sets existed before, the high accuracy; high resolution n TOF data will serve as basis for a more precise re-evaluation.



**FIGURE 3.** Preliminary results on the anisotropy of fission fragment from <sup>232</sup>Th(n,f) reaction [13]. Once refined, the n\_TOF data will represent the most accurate and complete data set up to date, covering the whole energy range from fission threshold all the way up to 1 GeV.

Among the most interesting results, high quality results in a wide energy range have been obtained on fission cross-sections of <sup>233</sup>U [11,12] of fundamental importance for the development of the alternative Th/U fuel cycle, while a striking and somewhat unexpected result, confirmed by a recent re-analysis of integral measurements, regards <sup>237</sup>Np, whose fission cross section above threshold was found to be 6-8% higher than current evaluations [13]. Furthermore, the angular anisotropy of fission fragments from <sup>232</sup>Th and <sup>234</sup>U has been recently measured with the PPAC system, and the results will soon be available. A preliminary result on the angular anisotropy of <sup>232</sup>Th is shown in Figure 3 [14].

#### **Other Measurements**

Recently, the n\_TOF Collaboration has undertaken new lines of experimental activity, such as the simultaneous measurement of capture and fission cross sections, and the measurement of (n,charged particle) reactions. In the first case, the possibility to measure capture reactions with fission veto is very useful for some actinides characterized by a fission cross-section much higher than that for capture. To this end, a fission tagging MicroMegas detector has been used in combination with the TAC [15], allowing the determination of the capture-to-fission ratio for several resonances of <sup>235</sup>U. The method will be used in the future for some difficult isotopes, such as <sup>233</sup>U, whose capture cross section is more than one order of magnitude smaller than the fission one.

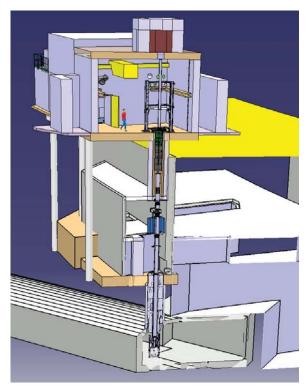
Developments of new detectors, such as MicroMegas chambers and diamond detectors, have recently opened the way to  $(n,\alpha)$  measurements, of interest for Nuclear Astrophysics, reactor technology and medical applications. A measurement was recently performed for the  $^{33}S(n,\alpha)$  cross-section with a Micromegas detector, while the important  $^{59}Ni(n,\alpha)$  reaction will soon be measured with a dedicated system specifically developed at n\_TOF, based on single crystal, chemical vapor deposition diamond detectors.

#### THE SECOND EXPERIMENTAL AREA

The n TOF Collaboration has recently proposed the construction of a second experimental area (EAR-2) at the vertical of the spallation target, at a much shorter distance than the present measuring station (20 vs 200 m). The extremely high neutron flux that would be available in EAR-2, approximately a factor of 25 higher than in EAR-1, and the still reasonable energy resolution, will open the way to a complete set of new measurements, currently not feasible at n TOF (or at any other facility around the world). In particular, the second experimental area will allow the measurement of isotopes with very low cross section or available in very small quantity (<1 mg). Most importantly, the combination of the much higher flux and shorter timeof-flight will make possible the measurement of neutron cross sections of unstable isotopes with very high specific activity, i.e. with half-life has short as a few years, thanks to a drastic improvement of the signal-to-background ratio, being the background related to the natural radioactivity of the sample. The range of possible measurements, of interest for Nuclear Astrophysics and nuclear technology, is rather vast, with the limitation related mostly to the availability of samples. In this regard, samples of short lived isotopes of interest for Nuclear Astrophysics could be produced by implantation of radioactive beams at the RIB facility ISOLDE at CERN.

The very high flux of EAR-2 could also allow the use of very thin samples, a great advantage in the measurements of (n, charged particle) reactions, in particular if low-energy  $\alpha$ -particles are emitted. This is the case, for example, of the Astrophysics-relevant  $^{25}Mg(n,\alpha)$  reaction, in which  $\alpha$ -particle of energy less than 500 keV are produced.

Another possible application of the intense neutron beam in EAR-2 will be irradiation of various material and electronic devices for dosimetry studies, detector development and other applications. For some specific cases, requiring extremely high doses or a high radiation environment, the Collaboration is planning to set-up an additional irradiation point at 1.5 m from the spallation target, along the vertical flight path, which could enhance the range of possible uses of n\_TOF.



**FIGURE 4.** A schematic drawing of the 20 m vertical flight path and of the second experimental area that will soon be constructed at n\_TOF. A permanent magnet (in blue), a collimator just below the experimental area, and a beam dump on top of the building will be used to shape the beam and minimize the background in the measuring station.

The preliminary design study of the second experimental area has been recently completed and construction will start in a few months. A schematic drawing of the vertical neutron beam line at n\_TOF is shown in Figure 4. Along the flight path, a permanent magnet and a collimator will be installed, to minimize the background in the experimental area. The first

neutron beam is expected to be delivered in EAR-2 in the second half of 2014. After the commissioning phase, and for the first few years of operation, the Collaboration is considering measurements of the capture cross section of  $^{79}\mathrm{Se}$  and other short lived isotopes of interest for Nuclear Astrophysics, as well as measurements of (n,p) and (n,  $\alpha$ ) reactions on  $^{7}\mathrm{Be},$   $^{25}\mathrm{Mg},$  and  $^{26}\mathrm{Al},$  of interest for stellar and primordial Nucleosyntesis. On the nuclear technology side, simultaneous measurement of capture and fission cross sections of  $^{238}\mathrm{Pu}$  and  $^{244}\mathrm{Cm}$  are being planned, together with capture cross section of  $^{245}\mathrm{Cm}$  and fission cross section and angular anisotropy of  $^{230}\mathrm{Th}$  and  $^{232}\mathrm{U}$  [3].

#### **CONCLUSIONS**

In more than ten years of operation, the n TOF facility at CERN as allowed the succesfull measurements of a large number of neutron cross sections, of interest for Nuclear Astrophysics and for advanced nuclear technologies. The features of the facility and high performance of experimental devices have generally led to high quality data, serving as the basis for advances in the understanding of stellar nucleosynthesis and for improving the accuracy of nuclear data libraries used in the field of nuclear energy. The construction of a second flight path at the shorter distance, foreseen for the near future, will open new measurement opportunities, in particular on shortlived isotopes, currently out of reach at n TOF and other neutron facilities around the world. As in the past, the goal of the n\_TOF Collaboration for the next years is to collect excellent quality neutron data of interest for basic and applied Nuclear Physics.

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