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Neutron measurements for advanced nuclear systems: The n_TOF project at CERN

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ABSTRACT

A few years ago, the neutron time-of-flight facility n_TOF was built at CERN to address some of the urgent needs of high-accuracy nuclear data for Accelerator Driven Systems and other advanced nuclear energy systems, as well as for nuclear astrophysics and fundamental nuclear physics. Thanks to the characteristics of the neutron beam, and to state-of-the-art detection and acquisition systems, high quality neutron cross-section data have been obtained for a variety of isotopes, many of which radioactive. Following an important upgrade of the spallation target and of the experimental area, a new measurement campaign has started last year. After a brief review of the most important results obtained so far at n_TOF, the new features of the facility are presented, together with the first results on the commissioning of the neutron beam. The plans for future measurements, in particular related to nuclear technology are finally discussed.

1. Introduction

The study of neutron-induced reactions has always been one of great importance for fundamental and applied nuclear physics. On the one hand, measurements of neutron cross-sections are a useful tool in nuclear structure, since they allow to study excited levels close to the neutron binding energy and to obtain information on nuclear properties such as level density, nuclear separation energy, etc. On the other hand, neutron data are essential in several interdisciplinary applications, such as nuclear astrophysics, nuclear medicine, dosimetry, radioprotection and, especially, nuclear energy.

In nuclear astrophysics, neutron data constitute a fundamental input in the modeling of stellar nucleosynthesis, because neutron capture reactions are responsible for the production of almost all nuclei heavier than Fe [1]. While the main features of the observed abundance distribution of elements in the Universe are now well understood and reproduced by theoretical models, a more detailed insight in the production mechanisms and in stellar evolution requires new data on a variety of isotopes, which could not be obtained in the past with the required accuracy.

In nuclear technology, neutron data are fundamental for a variety of applications. The field which relies the most on neutron data is by far nuclear energy. In recent years, new studies have been triggered by the urgent need to find safe, clean and possibly economic energy supplies to progressively replace fossil fuels, responsible for severe environmental problems, due to the large production of CO_2, which could be the cause of the greenhouse effect and related climatic changes. In this respect, it is now generally believed that an important role in the mix of energy sources of the future could be played by nuclear energy, in particular in developing countries.

If the advantages of nuclear energy by fission are evident, so are its disadvantages. The major drawbacks affecting current nuclear reactors are the low efficiency in the use of uranium resources, which could lead to availability problems in less than a century, and the abundant production of high-level nuclear waste, mostly made of long-lived fission fragments and trans-uranium (TRU) actinides.

Since a few years, new systems are being investigated, which could allow to overcome the mentioned limitations of current nuclear reactors. A promising solution to the waste problem is the use of subcritical Accelerator Driven Systems (ADS) [2], in which isotopes with long lifetime are transmuted in stable or short-lived nuclei by means of neutron-induced reactions (mainly capture and fission). In this case, a high-current, high-energy proton or deuteron accelerator would supply the neutron flux with sufficient energy to sustain the transmutation reactions, as schematically shown in Fig. 1. While an intense R&D is being conducted on several technological issues, in particular on high-intensity proton accelerators, progress in the knowledge of basic nuclear data related to the transmutation process is also needed.

Another possibility being investigated are the so-called Generation IV fast nuclear reactors [3]. In this case, a higher efficiency in the utilization of uranium resources is achieved by partial or full recycling of TRU actinides, with the further advantage of minimizing the final volume of high-level nuclear waste to be stored in geological repositories. As for ADS, new data on capture, fission and inelastic reactions for several isotopes, in particular for actinides, are needed for the development of Gen IV reactors [4]. Finally, studies are now being performed on alternative fuel cycles. An interesting possibility is the use of the Th/U cycle, eventually in conjunction with ADS and/or with Gen IV systems. The advantages of this fuel cycle are the availability of Th, which is 3–4 times more abundant than uranium in the Earth crust, and the low production of Pu and heavy actinides.

The design of advanced nuclear energy systems requires accurate neutron data for a large number of isotopes. In particular, high-accuracy, high-resolution cross-section data are needed on neutron induced reactions (capture, fission and inelastic reaction) for several actinides, long-lived fission fragments and structural materials.

To address some of the pressing needs of accurate new data for nuclear astrophysics and for advanced nuclear technologies, an innovative neutron time-of-flight facility, n_TOF, was constructed ten years ago at CERN, Geneva. The convenient features of the neutron beam, combined with high-performance detection and data acquisition systems, have allowed to collect important data in a first measurement campaign, that lasted between 2002 and 2004. In the past two years, some important modifications of the facility and of the experimental area have been implemented, with the aim of improving the features of the neutron beam and make possible to perform a number of difficult measurements, in particular on minor actinides. After a brief introduction of the n_TOF facility and of the experimental setups, the main characteristics of the n_TOF neutron beam are described. The most important results obtained in the first experimental campaign are then briefly reviewed, with particular emphasis on reactions of interest for ADS and Gen IV projects. The modifications recently implemented on the spallation target, on the cooling and moderation systems, and on the experimental area are then presented, together with the first results of the commissioning of the new neutron beam. The plans for future measurements related to nuclear astrophysics and nuclear technologies are discussed at the end.

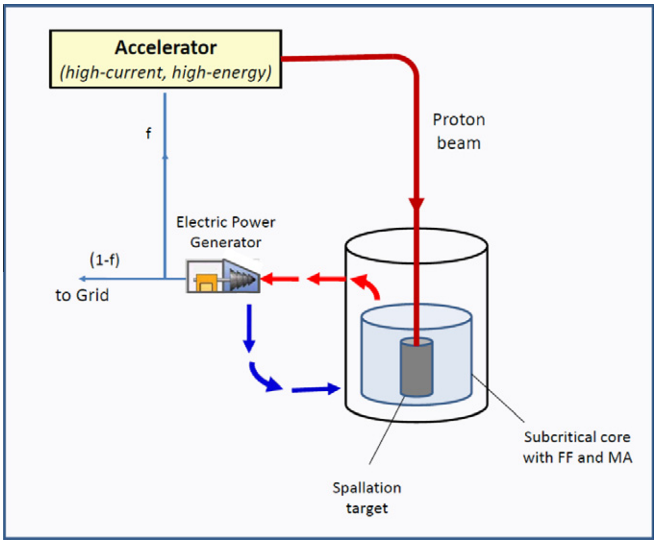

Fig. 1. Schematic drawing of the principles of Accelerator Driven Systems for the incineration of nuclear waste, in particular fission fragments (FF) and minor actinides.

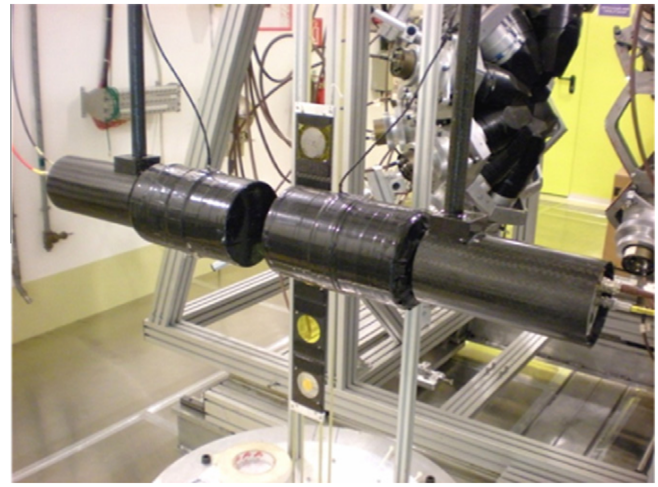

Fig. 2. Photo of the setup used for measurements of capture reactions, based on the C_6D_6 liquid scintillator detectors. To minimize the neutron sensitivity of the apparatus, the housing and support of the detectors, as well as the sample holder, are made of carbon fiber.

2. The n_TOF facility

The neutron time-of-flight facility n_TOF is based on a spallation neutron source [5]. Neutrons are produced by 20 GeV/c protons from the CERN Proton Synchrotron accelerator, impinging onto a lead block, surrounded by a water layer acting as coolant and moderator of the neutron spectrum. The target/moderator material and geometrical configuration were chosen taking into account the large body of data on neutron production by high-energy protons [6,7] (for more recent work on the subject see also [8] and references therein), as well as on the basis of extensive simulations of the neutron production and transport, performed with the most reliable Monte Carlo codes, such as FLUKA and MCNP. Although it was not the first time a Pb spallation targets was used in a neutron facility (see Ref. [9] for example), the high energy and peak current of the proton beam posed new challenges on the design of the n_TOF target and cooling system.

The innovative features of the n_TOF neutron beam are derived directly from the three main characteristics of the PS accelerator: high energy, low duty cycle and extremely high peak current. The high energy and high current of the PS accelerator complex lead to a neutron beam characterized by a wide energy range and very high instantaneous flux (i.e. the flux per bunch). The wide energy spectrum, spanning over nine orders of magnitude from thermal energy to approximately 1 GeV, is a particularly convenient feature for fission measurements, since it allows to extend the knowledge of cross-sections well above current limits, in the region of hundreds of MeV neutron energy, which is of interest for Accelerator Driven Systems, as well as for fundamental nuclear physics. Similarly, the very high instantaneous source intensity, of the order of 2×10^{15} neutrons/pulse, makes n_TOF particularly suited for cross-section measurements on radioactive isotopes, since it allows to minimize the signal-to-background ratio. Finally, the low duty cycle of the PS (<0.5 Hz), results in a practically absent wrap-around problem, which occurs when two consecutive neutron bunches overlap. Another important aspect of n_TOF is the possibility to perform high-resolution measurements. Thanks to the very high source intensity, it is possible to increase considerably the flight path, still keeping an average neutron flux comparable to, and an instantaneous flux up to three order of magnitude higher than, other facilities. At present, the measuring station is located at 187.5 m from the spallation target, allowing to reach an energy resolution of 10^{-4} up to a few keV neutron energy. Finally, a series of beam shaping collimators, thick iron and concrete shielding walls, and a sweeping magnet ensure a very low ambient background in the experimental area.

3. The experimental setups

The convenient features of the neutron beam are complemented with state-of-the-art detection and data acquisition systems. The neutron flux is monitored with a low-mass device made of a thin foil with ^{6}Li deposit surrounded by an array of Silicon detectors outside the beam. For capture measurements, two different detectors have been set up: an array of deuterated benzene (C_6D_6) liquid scintillator detectors, and a 4π BaF_2 total absorption calorimeter (TAC). The first apparatus is characterized by a low neutron sensitivity, further minimized at n_TOF by reducing the amount of material surrounding the detectors, and by using carbon fiber for detector housing and support (because carbon is characterized by a very low neutron capture cross-section). A photo of the apparatus, as currently used at n_TOF, is shown in Fig. 2. Corrections for the detection efficiency of this apparatus rely on detailed Monte Carlo simulations of the experimental setup [10]. For highly radioactive and fissile isotopes, in particular for minor actinides of interest for advanced nuclear energy systems, capture measurements are performed by means of the so-called calorimetric method, where the entire γ-ray cascade is detected in a large volume 4π detector. At n_TOF, a total absorption calorimeter (TAC) made of 40 BaF_2 crystals was built. The neutron sensitivity of the apparatus was minimized by inserting an inner sphere of moderating and absorbing material (based on ^{6}Li), and by encapsulating the crystals in ^{10}B-loaded carbon fiber.

Fission cross-section measurements have been carried out with different detector systems. Fission fragments are detected in a Fission Ionization Chamber (FIC), constituted by a stack of several parallel-plate chambers with 5 mm spacing between electrodes, operated with argontetrafluormethane (90% Ar + 10% CF_4) at 720 mbar. The samples are deposited on both sides of one of the electrodes. More recently, the FICs have been replaced with high-performance MicroMegas detectors, a gas detector originally developed for tracking in high-rate high-energy experiments [11]. The detector is composed of a gas volume separated into two regions by a thin micromesh. In the first section, the ionization and drift of the electrons occur, while in the second one, typically 50–160 μm thick, a multiplication process takes place, thanks to a high electric field (40–70 kV/cm) between the micromesh and the anode. The anode can be segmented into strips or pads for position sensitivity. The detector has several advantages, such as robustness and high radiation hardness, as well as a high signal-to-noise ratio even for small deposited energies. These qualities have been exploited at n_TOF, where MicroMegas detectors have already been extensively used for measuring and monitoring the neutron flux, as well as for determining the neutron beam profile. In all these cases, neutron detection is achieved by using a deposit of a suitable converter material (in particular, ^{10}B and ^{235}U). A new version of the MicroMegas has recently been developed, based on the micro-bulk principle, in which a grid of kapton pillars of small thickness (25 or 50 μm), obtained by means of a chemical process, ensures a perfect uniformity of the very thin multiplication gap. The detector is operated with a mixture of Ar (2%) and isobutene, at the pressure of 1 bar. When employed as fission detector, MicroMegas are characterized by a good discrimination of α-particles from fission fragments. For this reason, the detector will be used in future measurements of fission cross-sections on actinides.

A second method used at n_TOF for measuring neutron-induced fission cross-sections relies on the detection of both fission fragments in coincidence, by means of a stack of position-sensitive Parallel Plate Avalanche Counters (PPACs). The main advantage of the coincidence technique is the very efficient rejection of α-particles. The samples are deposited on a very thin mylar or aluminum foil (a few μm), mounted between two adjacent PPACs. The position sensitivity of the device allows to measure, together with the cross-section, the angular distribution of fission fragments. New measurements on this subject are now ongoing for several actinides. The stack of PPACs used in such measurements are shown in Fig. 3. All detectors are placed inside a vessel in which gas at low pressure is circulated.

All setups mentioned above allow to measure simultaneously the fission cross-sections of several isotopes, thus optimizing the use of the beam time. In all cases, ^{235}U and ^{238}U samples are also measured as reference, because their fission cross-sections constitute well established standards.

The n_TOF data acquisition systems is based on a set of Flash Analog to Digital Converters (FADC), with sampling rates ranging up to 2 GS/s [12]. Off-line reconstruction of the digitized signal allows to extract relevant information on the neutron time-of-flight (and thus on the neutron energy), as well as on other relevant information such as energy deposited in the detectors and particle identification.

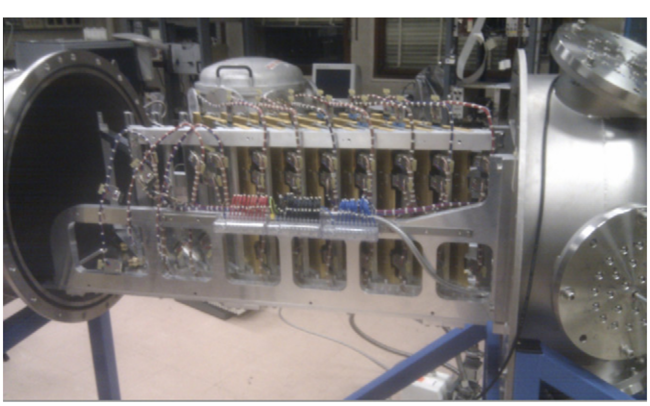

Fig. 3. Photo of the detection system based on position-sensitive Parallel Plate Avalanche Counters, now in use for the measurement of the angular anisotropy of fission fragments in neutron-induced fission reactions of various actinides. The detectors are tilted by 45 degrees relative to the direction of the neutron beam, in order to extend the angular range investigated.

4. The results of n_TOF Phase-I

An international collaboration of over 120 researchers from 40 universities and research institutes, mostly European, is responsible for proposing and carrying on a large experimental program at n_TOF. High-quality data on a large number of reactions have been collected in the first measurement campaign. For most isotopes, the n_TOF results are characterized by improved accuracy, higher resolution and a wider energy range than previous data. In some cases, the n_TOF data have also helped to solve existing discrepancies between previous experimental and/or evaluated cross-sections. A large fraction of the measurements performed so far at n_TOF have already been published, and the results have been made available in EXFOR, the experimental database, for use by the nuclear astrophysics and/or the nuclear reactor communities. A list of capture and fission cross-section measurements performed at n_TOF, together with a comprehensive list of publications, can be found on the n_TOF web site. Among the most important results obtained so far, we recall the high accuracy capture data of ^{237}Np and ^{240}Pu [13], which are some of the minor actinides most abundantly produced in current reactors. Similarly, high quality results have been obtained on fission cross-sections of ^{233}U [14], of fundamental importance for the development of the alternative Th/U fuel cycle, or on some minor actinides (^{241}Am, ^{243}Am and ^{245}Cm), for which currently available cross-sections are affected by uncertainties too large for the desired applications. The n_TOF facility offers a unique opportunity in this respect, since it combines the very high instantaneous neutron flux, ideal for measuring the highly radioactive transuranic elements, with the high resolution in neutron energy, which allows to extend the resolved resonance region to higher energies, making calculations of self-shielding effects in reactor fuel elements more reliable. Finally, the wide energy range of the n_TOF neutron beam allows to measure fission cross-sections up to several tens of MeV, a feature important for applications to transmutation projects, as well as for improving model calculations of fission reactions.

5. n_TOF Phase-II

Following the end of the first measurement campaign, the facility was shut-down for three years in order to substitute the spallation target, upgrade the cooling systems and the ventilation of the target area and, finally, to upgrade the experimental area so to allow handling of highly radioactive samples without certified sealing. The substitution of the spallation target was necessary after a visual inspection revealed the presence of surface oxidation in some areas, as well as of pitting corrosion at the proton impact location. The degradation of the target was mostly attributed to inefficient cooling in some particular positions. For this reason, it was decided to modify the design of the target, to avoid mechanical instabilities and deformations related to the modular geometry. The new spallation target, schematically shown in Fig. 4, is now made of a monolithic cylindrical block of lead, 40 cm length and 60 cm diameter. Besides the shape of the lead block, important modifications were implemented also on the cooling system with the aim of ensuring a more uniform and efficient cooling, as well as to allow the use of an independent moderation circuit. On the exit face of the target, cooling is ensured by a water layer of 1 cm, while moderation of the neutron spectrum is performed with a 4 cm thick layer of either normal, heavy or borated water. The use of different moderators allow to change the spectral features of the neutron beam and, most importantly, to minimize the production

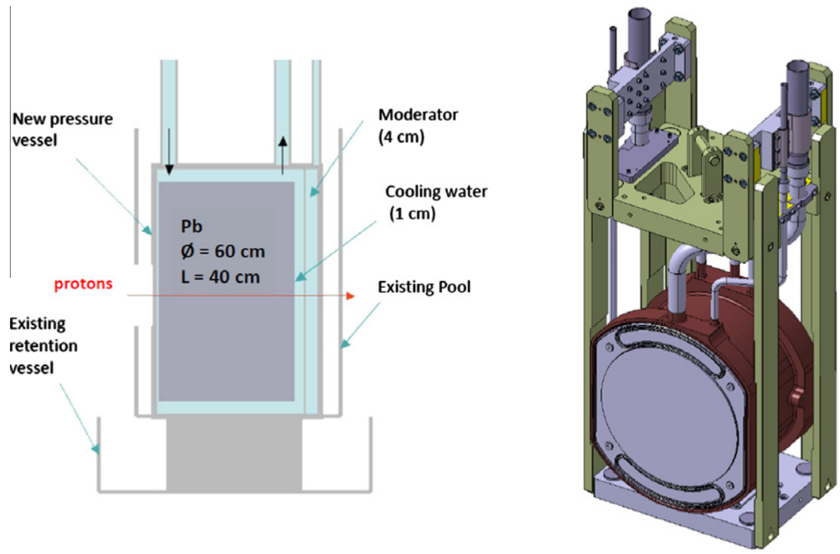

Fig. 4. Schematic drawings of the new spallation target at n_TOF. The figure on the left shows the various elements of the new target (Pb block, cooling and moderation circuit, etc.).

of 2.2 MeV in-beam γ-rays, which are abundantly produced in the cooling system by the radiative capture on hydrogen. Scattering of these γ-rays by the sample constitutes the main source of background in measurements of capture cross-sections in the keV neutron energy region. The use of a borated water considerably reduces such contamination, since thermal neutrons are absorbed by means of $^{10}B(n,\alpha)$ reactions.

After completing the installation of the new spallation target and of the cooling and moderation systems, a set of measurements for the characterization of the new neutron beam were performed. The neutron flux has been determined by means of four different measurements. A calibrated fission chamber from PTB [15], containing ^{235}U samples, MicroMegas (MGAS) detectors with ^{10}B and ^{235}U deposit, and a Silicon Monitor (SiMon) consisting of 4 silicon detectors viewing a ^{6}Li target. The highest accuracy in the absolute neutron flux determination is provided by the PTB fission chamber, which is therefore used as reference for all other measurements. However, the use of the other detectors with different neutron-converting reactions is useful in order to determine the energy dependence of the flux with an uncertainty around 2%. The results for normal water as moderator are shown in Fig. 5 (all data are normalized to the PTB results in the region 0.1–1 eV). The observed neutron flux distribution is very similar to the one produced with the first spallation target, with a peak in the thermal energy region, a nearly isolethargic distribution between 1 eV and several hundreds of keV, and an evaporation peak in the MeV region. The dips observed in the keV region are due to absorption in the aluminum windows at the entrance of the evacuated flight path. The results are in good agreement with simulations, performed with FLUKA and MCNP-X.

The use of borated water as moderator changes the neutron flux distribution, mainly at low energies. Fig. 6 shows the preliminary results of the analysis of three different measurements of the neutron flux, based on the standard cross-section of ^{6}Li, ^{10}B and ^{235}U. The data were taken with a concentration of boric acid of 1.28%, with 96% ^{10}B enrichment. Compared with the neutron flux for normal water, the thermal peak is strongly suppressed, due to the neutron capture by ^{10}B. A reduction in the flux is observed up to 100 eV, while at higher energy the use of borated water as moderator has practically no effect.

As mentioned above, the main advantage in the use of borated water is the strong reduction of the in-beam γ-rays, which contaminate the neutron beam in the keV energy region. Fig. 7 shows the capture yield measured with a sample of ^{nat}Fe, with normal (red) and borated water (black) as moderators. It can be clearly observed that the background in the keV region is reduced by at least an order of magnitude with the borated water moderator. This is an important improvement for the n_TOF neutron beam, since it allows to reach higher accuracy, with respect to n_TOF Phase-I, for the capture cross-section in the keV energy region. As evident in Fig. 7, the much smaller contamination of in-beam γ-rays in the neutron beam now allows one to study even small resonances, or resonance structures, which were previously masked by the background. This improvement is particularly important for the measurements of capture cross-section of actinides in the unresolved resonance region.

Together with the flux, dedicated measurements were performed after the refurbishing of the facility, to characterize the neutron beam in terms of spatial profile and resolution functions. The precise determination of the beam profile is of primary importance since the samples used in most measurements of neutron capture reactions are smaller than the neutron beam itself (in this

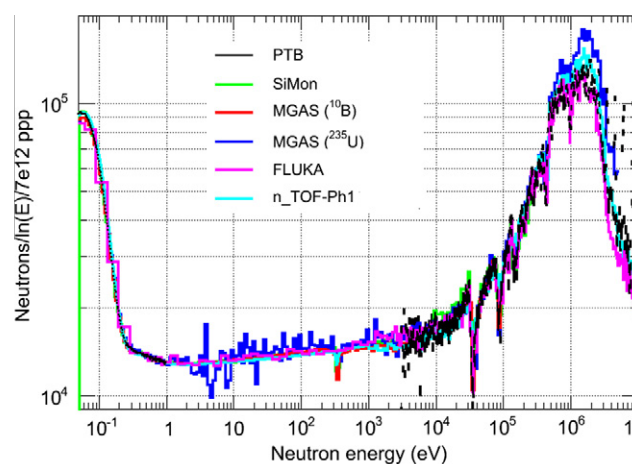

Fig. 5. The neutron flux measured with different detectors during the commissioning of the new spallation target, with 5 cm water layer. Apart for small details, the neutron flux in n_TOF Phase-II is similar to the one available in the first measurement campaign.

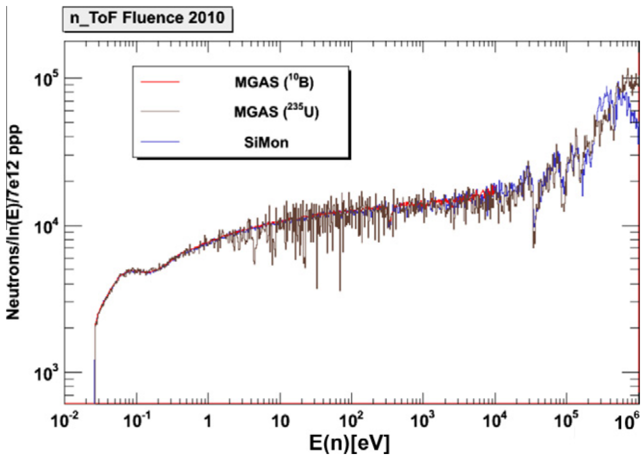

Fig. 6. Preliminary results of the measured neutron flux energy distribution produced with a 4 cm thick borated water moderator. The measurements were performed with a MicroMegas chamber equipped with ^{10}B and ^{235}U deposits, as well as with the Silicon Monitor described in the text. As expected, the main effect of the new moderator is the strong reduction of the thermal peak. Above 1 keV, no appreciable differences are observed compared to the use of normal water as moderator.

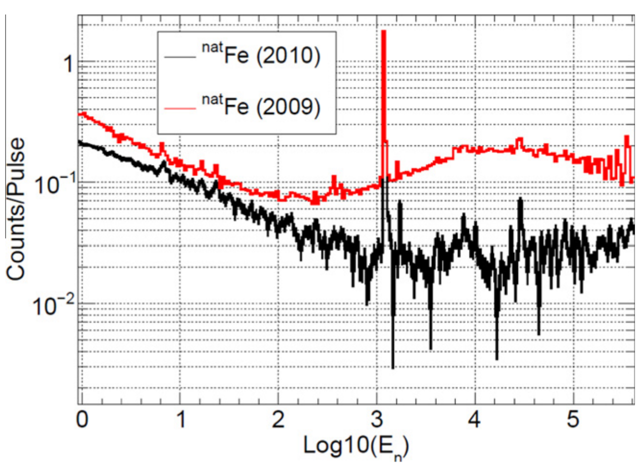

Fig. 7. Comparison between the capture yield measured for ^{nat}Fe with two different moderators: normal water (red histogram) and borated-water (black). As evident in the figure, the strong reduction of the background in the keV region allows to collect more accurate capture cross-section data than in the past, in the 1 keV–1 MeV energy range, in particular for actinides. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

way uncertainties related to inhomogeneities of the sample are minimized, while the reaction yield is maximized). However, corrections for the fraction of beam intercepted by the sample, which depends on neutron energy, have to be performed. A specific position-sensitive detector, the *X–Y MicroMegas* (XYMG), was developed for the determination of the n_TOF beam profile, with a spatial resolution of a few hundred microns. The measured beam profile has a nearly Gaussian shape with a width of ~7 mm changing slightly with neutron energy, in agreement with simulations. Finally, a measurement was performed to determine the energy resolution of the n_TOF neutron beam, which is related to the time resolution of the proton beam and, especially, to the time spread introduced by the moderation process. The resolution measured with the new spallation target was found to be practically the same as the one observed with the previous target.

Another important upgrade regarded the experimental area, which was modified in order to allow measurements of high-activity samples. In the first experimental campaign, radioprotection regulations of CERN required that all samples with activity exceeding certain limits had to be sealed according to ISO2919 prescriptions. A large background was however produced by the sealing capsules. To minimize this background, the experimental area was upgraded to a so-called "Work Sector Type A", i.e. an area complying with radioprotection regulations for manipulation of unsealed radioactive sources. To this aim, the area was equipped with a series of safety systems, such as controlled ventilation, anti-fire doors, continuous radiation monitoring, etc., and strict access procedures were introduced for all personnel working at n_TOF. This crucial modification was essential for exploiting the full strength of the facility.

After the commissioning phase, a series of measurements of interest for nuclear astrophysics and emerging nuclear technologies have now started. Measurements of the capture cross-sections of various Fe and Ni isotopes have been completed. These measurements are important for improving the comprehension of stellar nucleosynthesis, being these two elements at the beginning of the chain of neutron capture processes, as well as in nuclear technology, since they enter in the composition of many reactor components. Another very important measurement recently completed is the capture measurement of ^{241}Am, one of the most important minor actinides involved in transmutation projects with ADS and in the design of Generation IV fast reactors. To improve the present accuracy of the ^{241}Am(n,γ) cross-section, data have been collected at n_TOF with the two different detection systems, the C_6D_6 and the TAC, described in a previous section. The analysis of these data is in the very early stage, but the improved features of the n_TOF neutron beam is expected to lead to very high quality results on this important isotope.

The n_TOF Phase-II program foresees several new important measurements to be performed in the next years. The capture cross-sections of ^{238}U and ^{236}U will be measured in the near future, while fission cross-sections measurements are planned for ^{240}Pu and ^{242}Pu. Finally, the angular anisotropy of various actinides, such as ^{232}Th, ^{234}U and ^{237}Np, are in the process of being measured.

The n_TOF Collaboration is also planning to perform capture measurements with fission veto for some actinides characterized by a fission cross-section much higher than that for capture. In this case, capture events are difficult to discriminate from fission, resulting in uncertain capture cross-sections. As a possible solution, a fission tagging detector has been developed at n_TOF to be used in combination with the total absorption calorimeter. This method could allow in the near future the very difficult measurement of the ^{233}U(n,γ) cross-section, which is an order of magnitude lower than for the fission reaction. Finally, developments of new detectors, such as diamond detectors, may open the way to (n,α) measurements at n_TOF. A pilot experiment on ^{33}S(n,α) cross-section with a MicroMegas detector is already planned for the next months.

6. Conclusions and perspectives

The first experimental campaign at the n_TOF facility was successfully completed with new results on capture and fission cross-sections of interest for nuclear astrophysics and for emerging nuclear technologies. The performances of the facility have been recently improved by refurbishing the spallation target, the cooling and moderator circuits, and the experimental area. In particular, while the flux and resolution of the neutron beam have not changed significantly relative to n_TOF Phase-I, the background produced by the in-beam contamination of γ-rays has been drastically reduced, thanks to the use of a borated-water moderator. Furthermore, the upgrade of the experimental area to a ''Work Sector Type A'' allows nowadays to perform at n_TOF measurements on high-activity samples, in particular actinides, for which accurate new data are needed for advances in basic and applied nuclear science.

Following these modifications, a second measurement campaign has started and is now in progress, with a series of new capture and fission measurements of fundamental importance for the development of advanced nuclear energy systems, such as Accelerator Driven Systems and Gen IV reactors. Among other key reactions, the neutron capture on ^{241}Am has been just measured, while fission measurements on the heavier Pu isotopes are foreseen next. Finally, an upgrade of the experimental setups has been completed, which will allow the simultaneous study of capture and fission reactions. Together with the applicative aspects, a very interesting program on nuclear astrophysics will also be pursued in Phase-II. Finally, an upgrade of the facility with a second flight path at the shorter distance of 20 m has been proposed. Such a short flight path will result in a 100 times larger neutron flux, which will open new vistas to measurements, which are currently not feasible.

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