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The fission programme at the CERN n_TOF facility

A. Tsinganis[a,b,*], M. Barbagallo[c], E. Berthoumieux[d], M. Calviani[b], E. Chiaveri[b], N. Colonna[c], M. Diakaki[a,d], I. Duran[e], C. Guerrero[b,f], F. Gunsing[b,d], E. Leal-Cidoncha[e], L.-S. Leong[g], C. Paradela[e], D. Tarrio[e], L. Tassan-Got[g], R. Vlastou[a], and the n_TOF Collaboration[h]

[a] *Dep. of Physics, National Technical University of Athens, Greece*
[b] *European Organisation for Nuclear Research (CERN), Geneva, Switzerland*
[c] *Istituto Nazionale di Fisica Nucleare (INFN) - Sezione di Bari, Bari, Italy*
[d] *Commissariat à l'Énergie Atomique (CEA) Saclay - Irfu, Gif-sur-Yvette, France*
[e] *Universidade de Santiago de Compostela, Spain*
[f] *Universidad de Sevilla, Spain*
[g] *Centre National de la Recherche Scientifique/IN2P3 - IPN, Orsay, France*
[h] *www.cern.ch/ntof*

Abstract

Since 2001, the scientific programme of the CERN n_TOF facility has focused mainly on the study of radiative neutron capture reactions, which are of great interest to nuclear astrophysics and on neutron-induced fission reactions, which are of relevance for nuclear technology, as well as essential for the development of theoretical models of fission. In particular, taking advantage of the high instantaneous neutron flux and high energy resolution of the facility, as well as of high-performance detection and acquisition systems, accurate new measurements on several long-lived major and minor actinides, from ^{232}Th to ^{245}Cm, have been performed so far. Data on these isotopes are needed in order to improve the safety and efficiency of conventional reactors, as well as to develop new systems for nuclear energy production and treatment of nuclear waste, such as Generation IV reactors, Accelerator Driven Systems and reactors based on innovative fuel cycles. A review of the most important results on fission cross-sections and fragment properties obtained at n_TOF for a variety of (radioactive) isotopes is presented along with the perspectives arising from the coming on line in the second half of 2014 of a new 19 m flight-path, which will allow n_TOF to expand its measurement capabilities to even more rare or short-lived isotopes, such as ^{230}Th, ^{232}U, ^{238,240}Pu and ^{244}Cm.

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* Corresponding author.
E-mail address: Andrea.Tsinganis@cern.ch

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1. Introduction

Although significant progress has been made since the discovery of fission over 70 years ago, the complexity of the phenomenon, with the interplay both of collective effects in the nucleus and single-particle interactions, is the main difficulty that needs to be overcome to formulate a comprehensive model of the fission that accurately describes all its aspects and has satisfactory predictive capabilities. To this day, however, theoretical calculations cannot be relied upon to accurately predict unknown fission cross-sections. New data – especially on neutron-induced fission – are therefore essential for the development of nuclear models and fundamental research in nuclear physics.

Beyond the basic physics interest in fission, the relevance of fission cross-section studies are obviously of great importance for nuclear energy applications, especially in the light of the renewed interest in this field. The continuous growth of the human population, coupled with the increase in per capita energy consumption, especially in developing countries as living standards improve, is leading to an ever more rapid increase in global energy demands. The serious environmental concerns posed by continuing reliance on fossil fuels are well known and have been highlighted once again in the recent IPCC (Intergovernmental Panel on Climate Change) assessment report (IPCC (2014)).

In the short and medium term, the growth of the global energy demand could be met with a more efficient use of existing energy sources by balancing a phased reduction of fossil fuel consumption with increased contributions from 'clean' or 'carbon-free' energy technologies. At present, over 80% of the total primary energy supply comes from fossil fuels (oil, coal and natural gas), which also account for roughly 70% of the electricity production. The rest of the electricity production is largely covered by nuclear (12%) and hydro-electric power (16%), while the contribution from other renewable energy sources, such as wind, solar, tide, geothermal energy etc. is still low (less than 5%). It is conceivable that with sustained investment in research on renewable sources of energy, these may eventually contribute a larger share. It seems unlikely, however, that they will be able to cover the entire energy demand in the foreseeable future, especially when it comes to providing power for increasingly large urban agglomerations with populations of several millions or even tens of millions (67% of the population is expected to reside in urban areas by 2050, against 52% in 2011). Nuclear energy will therefore likely remain an important component in the global energy mix and the nuclear output may even have to be increased to meet the challenges of the coming decades.

There are, nevertheless, three main concerns associated with the use of nuclear energy. These are: (i) the safety issues posed by current nuclear power plants, (ii) the long-term management of large volumes of radioactive waste and, finally, (iii) the proliferation of nuclear material and its potential use for military applications or terrorist activities. All three issues mentioned above, however, could be effectively addressed with the advanced nuclear reactor technologies presently under development.

Feasibility, design and sensitivity studies on new generation reactors require high-accuracy cross-section data for a variety of neutron-induced reactions from thermal energies to several tens of MeV. The NEA (Nuclear Energy Agency) 'Nuclear Data High Priority Request List' lists data requests for advanced reactor design, as well as other fields, while the OECD/NEA WPEC Subgroup 26 Final Report summarises the needs and target accuracies for nuclear data relevant for advanced nuclear systems. Capture and fission cross-sections of isotopes involved in the Th/U fuel cycle, long-lived Pu, Np, Am and Cm isotopes, long-lived fission fragments relevant for transmutation projects or isotopes considered as structural material for advanced reactors are included. Improved knowledge of these cross-sections is not only important for the design of advanced systems, but also for the operation of existing reactors, since safety margins can be more accurately defined, allowing for a more efficient use of available fuel resources.

The *n_TOF (**N**eutron **T**ime-**O**f-**F**light)* facility was constructed at CERN over 10 years ago to address these pressing needs of nuclear data for advanced nuclear technologies, as well as for nuclear astrophysics and for other fields of basic and applied nuclear physics.

2. The n_TOF facility

Between 1996 and 1997, an experimental campaign took place at CERN within the research programme on the Energy Amplifier (Rubbia (1995a,b)), a sub-critical fast neutron system driven by a proton accelerator. Among the main goals of the TARC (Transmutation by Adiabatic Resonance Crossing) experiment (Abanades (2002); Revol (1999)) was to demonstrate the feasibility of using Adiabatic Resonance Crossing (ARC) (Rubbia (1997)) as a means to destroy long-lived fission fragments in Accelerator Driven Systems (ADS), such as the Energy Amplifier. The experience accumulated, particularly with regard to the improved understanding of the physics and performance of a source of spallation neutrons based on a high-energy proton beam and a high-Z target, led to the proposal for the n_TOF facility at CERN by Rubbia (1998) with the aim of measuring neutron cross-sections over a wide energy range, from thermal to GeV, with a very high energy resolution and high instantaneous neutron flux. The facility, which includes an experimental area at the end of an approximately 185 m neutron flight-path, was commissioned and became operational in 2001.

Neutrons at n_TOF are spallation products created by a bunched 20 GeV/c proton beam delivered by CERN's PS (Proton-Synchrotron) accelerator onto a lead target. Since 2009 (Phase-II), the target is a lead cylinder 40 cm in length and 60 cm in diameter, while previously (Phase-I) it consisted of several lead blocks. A 1 cm-thick layer of water surrounds the target and is constantly circulated in order to cool it down. Along the direction of the beam, this layer is immediately followed by a second, 4 cm-thick layer which is filled either with demineralised water or borated water, with the addition of boric acid enriched in ^{10}B. The water present around the target, particularly in the beam direction, serves to moderate the neutrons produced in the spallation process and populate energies down to thermal. The two choices of moderator (water or borated water) strongly affect both the low energy neutron spectrum and the in-beam photon background. The beam-line consists of several sections of stainless steel tubes with progressively reduced diameters and two collimators that shape the beam. A dipole magnet is in place to deflect the charged particles travelling with the neutrons inside the vacuum tube. Since 2009, the experimental area meets the requirements to operate as a Type A Work Sector (Vlachoudis (2012)), meaning unsealed radioactive samples can be handled. More detailed presentations of the characteristics of the facility can be found in Abbondanno (2002), Guerrero (2013), Berthoumieux (2013) and Barbagallo (2013).

The experimental programme at n_TOF has focused mainly on the study of radiative neutron capture reactions, which are of great interest to nuclear astrophysics and important for applications and neutron-induced fission reactions, which are of relevance to the development of new systems for nuclear energy production and the transmutation of existing nuclear waste. The characteristics of the facility, coupled with high-performance detection and acquisition systems, allow measurements of low-mass and/or radioactive samples of interest in these fields and others, such as basic nuclear physics and nuclear medicine.

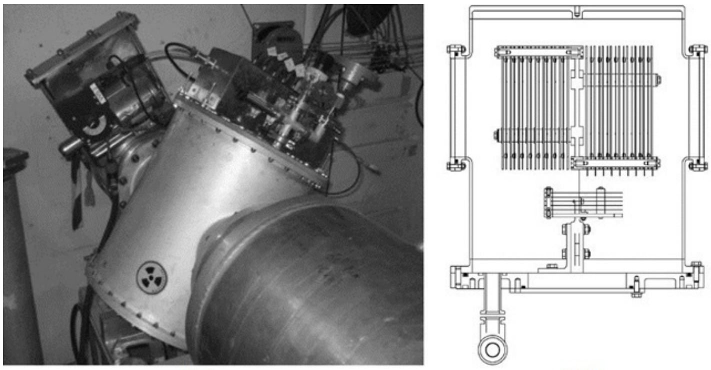
3. Experimental setups for fission

3.1. FIC

A variety of detectors are used for fission measurements at n_TOF. Most of them can be used to measure the cross-section of different isotopes simultaneously, thus optimising the use of beam-time. The Fission Ionisation Chamber (FIC) (Calviani (2008)) (Figure 1) consists of a stack of several parallel-plate chambers with 5 mm spacing between electrodes, operated with an Ar:CF_4 mixture (90:10) at a pressure of 720 mbar. The samples are deposited on both sides of a 100 μm thick aluminium foil, used as a cathode, while the anodes are made of a 15 μm thick foil. Samples used are typically of 8 cm in diameter and with up to 450 μg/cm^2 in thickness. Different variants of the FIC have been constructed for measurements at n_TOF.

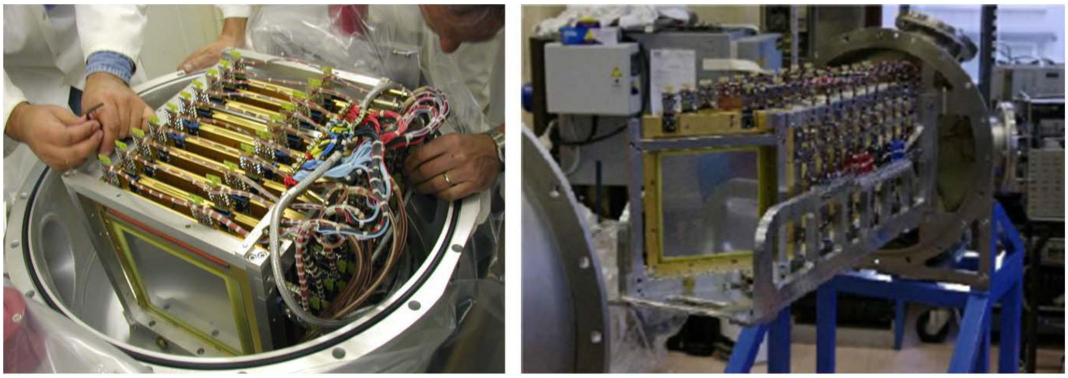
3.2. PPAC

The PPAC (Parallel Plate Avalanche Counter) assembly consists of a stack of parallel plate avalanche counters placed in a vessel in which low-pressure gas is circulated (Figure 2). This setup allows to detect both fission fragments emitted in a fission event in coincidence and it has the advantage of efficiently rejecting the α-particle background, as well as competing reactions. Originally, the detectors were placed perpendicularly with respect to the neutron beam. In the last few years, the detectors were tilted to achieve a reasonable efficiency at all emission angles of the fragments, avoiding the sharp drop in the efficiency at large angles typical of the previous setup and which led to larger uncertainties. The fission samples are deposited on 1.5 μm-thick mylar or 2 μm aluminum foils and are placed between two adjacent PPACs. The position sensitivity of this setup allows to measure the angular distribution of fission fragments in addition to the fission cross-section, as described in Tarrio (2014b).



(a) (b)

Fig. 1. One of the FIC detector variants placed along the n_TOF beam line (a) and a drawing of the interior (b), where the stack of samples and electrodes is clearly visible.



(a) (b)

Fig. 2. Internal view of the PPAC assembly used at n_TOF. (a) The original configuration with detectors placed perpendicularly with respect to the neutron beam and (b) the tilted setup.

3.3. Micromegas

The most notable design variation of the Micromegas compared to a more traditional parallel plate avalanche chamber, which features a single gas volume between the anode and cathode, is the separation of the gas volume into two regions: a *drift region* whose width can vary from a few hundreds of μm to a few cm and an *amplification region*, typically of a few tens of μm. The two regions are separated by a *micromesh*, a thin (~5 μm) conductive layer with 35 μm diameter holes on its surface at a distance of 50 μm from each other. For neutron measurements, it is of particular importance to minimise the amount of material present in the beam in order to avoid generating background neutrons that lead to the production of additional secondary particles, such as γ-rays, which are an undesirable background especially for capture measurements. With the minimisation of these effects in mind, the *microbulk* design (Andriamonje (2010, 2011)) was developed (Figure 3). This variant was utilised both for measurements and for monitoring of the neutron fluence. Detectors with an active area of 10 cm in diameter are typically used, but variants with diameters down to a few centimetres have also been produced. Among the advantages of Micromegas detectors are their robustness and radiation hardness, as well as a high signal-to-noise ratio, even for small deposited energies.

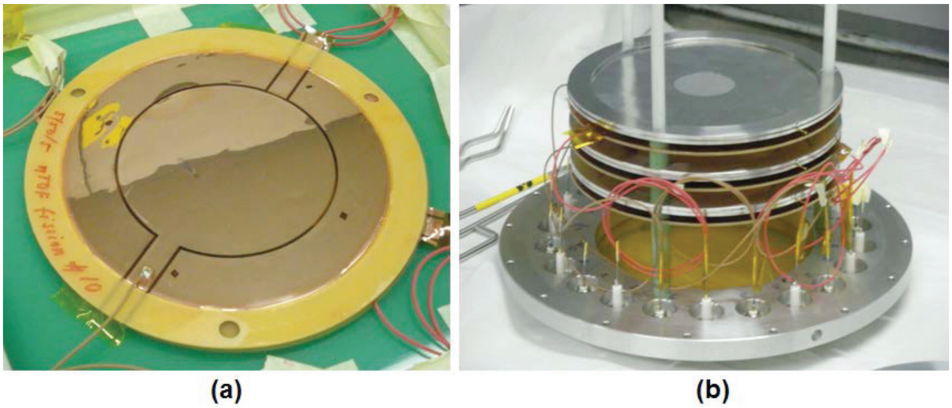


Fig. 3. A Micromegas detector of the 'microbulk' variant (a) and (b) a partial setup of Micromegas detectors and Pu samples as they are mounted inside the specially constructed chamber.

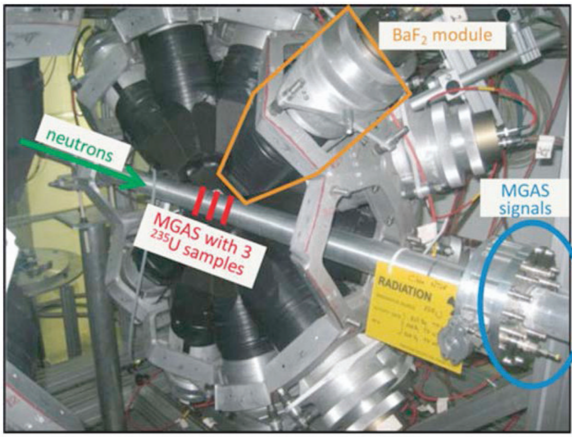


Fig. 4. A view of the 'fission tagging' setup at n_TOF. In this picture, one hemisphere of the BaF_2 array is visible, along with the chamber that holds the Micromegas detectors in the centre of the array.

3.4. The 'fission tagging' setup

Recently, a system combining the 4π Total Absorption Calorimeter (TAC), consisting of 40 BaF_2 crystals, and specially constructed Micromegas detectors was tested at n_TOF for the simultaneous measurement of the fission and capture cross-sections of fissile isotopes, as described in Guerrero (2012). Fission events are 'tagged' with the Micromegas detector and the γ-rays detected in coincidence are assigned to a fission event, rather than capture. A picture of the setup can be seen in Figure 4.

3.5. The data acquisition system

The analogue detector signals are input into the n_TOF Data Acquisition System, which is based on 8-bit *Acqiris* (now *Agilent*) flash-ADCs and is described in detail in Abbondanno (2005). The use of fast electronics is essential for measurements such as those performed at n_TOF, where fast timing is a critical element in order to resolve short times-of-flight and high counting rates in detectors. By using fADCs, the analogue waveform of the detector signals can be sampled at a chosen sampling rate between 100 MS/s and 1 GS/s. Due to the 8 MB memory limit of the buffer, a maximum acquisition window of 80 ms can be obtained, which corresponds to neutron energies of ~30 meV for the 185 m flight-path. In order to minimise the volume of data to be transferred and recorded, a zero-suppression algorithm is employed to avoid recording long sequences of noise where no useful signals are present. The data are finally stored in CASTOR (CERN Advanced STORage manager), the CERN central data storage system. The recorded data are analysed off-line by means of routines that determine the signal baseline, search for signals and determine the corresponding neutron time-of-flight (and thus the incident neutron energy) and the pulse height, among other quantities. The DAQ system is presently being upgraded with the use of new 10-bit digitisers.

4. Results from n_TOF

The n_TOF Collaboration, which consists of more than 130 scientists from over 30 institutes, has produced a significant collection of high-quality data on a large number of reactions, including fission cross-sections of many isotopes. In most cases, the n_TOF results are characterised 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.

The many fission measurements performed at n_TOF so far include several isotopes of uranium, that is $^{233-236,238}$U, ^{232}Th and ^{237}Np (including fission fragment angular distributions), as well as ^{nat}Pb, ^{209}Bi, ^{241,243}Am, ^{242}Pu and ^{245}Cm. A list of measurements and related references are given in Table 1.

Table 1. Neutron-induced fission cross-section measurements performed at n_TOF, with information on the setup used in each case and with the corresponding references. Several isotopes have been measured with different setups and fission fragment angular distributions have also been measured in some cases.

Isotope	Setup – Method	References
^{nat}Pb, ^{209}Bi	PPAC – coincidence	Tarrio (2011)
^{232}Th$^{(*)}$	PPAC – coincidence	Tarrio (2014a)
^{233}U	FIC – single fragment detection	Calviani (2009); Belloni (2011b)
^{234}U	FIC – single fragment detection	Karadimos (2014)
^{234}U,^{237}Np	PPAC – coincidence	Paradela (2010); Leal-Cidoncha (2014)
^{235}U(n,f/γ)	TAC and Micromegas – 'fission tagging'	Guerrero (2012)
^{235}U(n,f/γ)	TAC – Calorimetric Shape Decomposition (CSD)	Carrapico (2013)
^{236}U	FIC – single fragment detection	Sarmento (2011)
^{237}Np	FIC – single fragment detection	Diakaki (under preparation)
^{235,238}U$^{(*)}$, ^{237}Np$^{(*)}$	PPAC – coincidence	Leong (2013)
^{238}U	FIC – single fragment detection and	Paradela (2014)
	PPAC – coincidence	*same as above*
^{241,243}Am	FIC – single fragment detection	Belloni (2013, 2011a); Mastromarco (under preparation)
^{242}Pu	Micromegas – single fragment detection	Tsinganis (2014c,b)
^{245}Cm	FIC – single fragment detection	Calviani (2012)

$^{(*)}$ Fission fragment angular distributions (FFAD).

Among the many interesting results of the n_TOF fission programme, several highlights are here presented. The ^{233}U fission cross-section was measured from thermal to 2 MeV in a single measurement with the FIC detector and with an accuracy of 5%. The n_TOF data indicated that significant corrections to evaluations were in order in the region between 10 eV and 100 keV, as shown in Figure 5a. The ^{245}Cm(n,f) measurement was also performed with the FIC detector and was particularly challenging due the very high activity of the samples (87 MBq per sample). Below 30 eV, two very old previous measurements existed with large discrepancies, while, above that energy, a single measurement with neutrons from a nuclear explosion had been performed. As can be seen is Figure 5b, the n_TOF data confirmed the previous data above 30 eV and showed that significant corrections were necessary in evaluations at lower energies.

Another interesting case is the fission cross-section of ^{236}U, where n_TOF data showed that JEFF-3.2 and ENDF/B-VII.1 were overestimating the cross-section at thermal energies by a factor of more than 100. In the case of ^{237}Np, the data reported by Paradela (2010) above 1 MeV (Figure 6a) are about 7% higher than previous measurements. These results, however, are not confirmed by more recent n_TOF results. The most recent fission measurement in Phase-II is the cross-section of ^{242}Pu that was measured with a setup based on Micromegas detectors and was the first fission

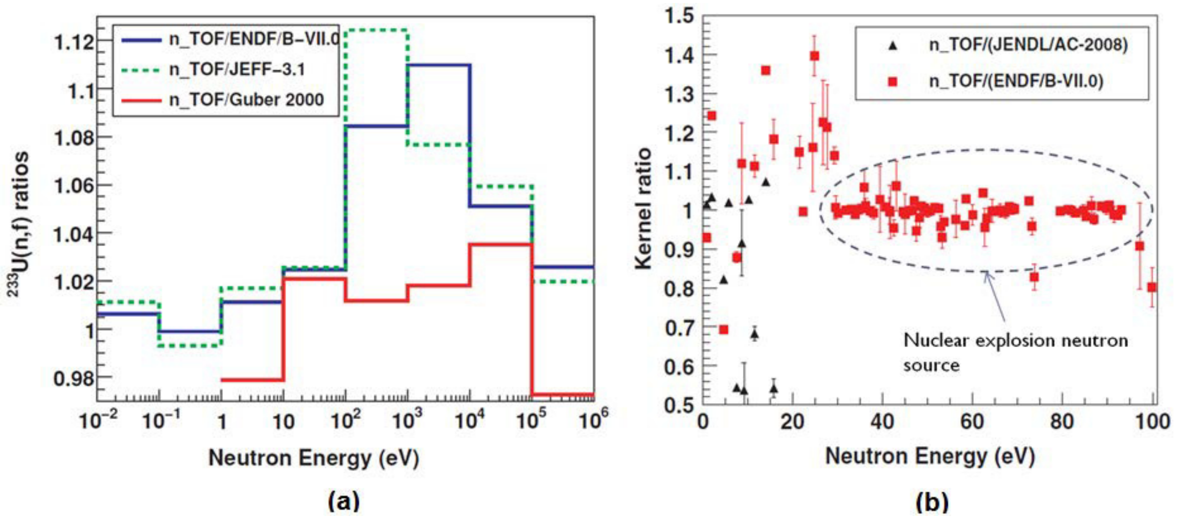


Fig. 5. Ratio of n_TOF data on ^{233}U (a) and ^{245}Cm(n,f) (b) with evaluations. Both these results indicated the need for significant corrections to evaluations.

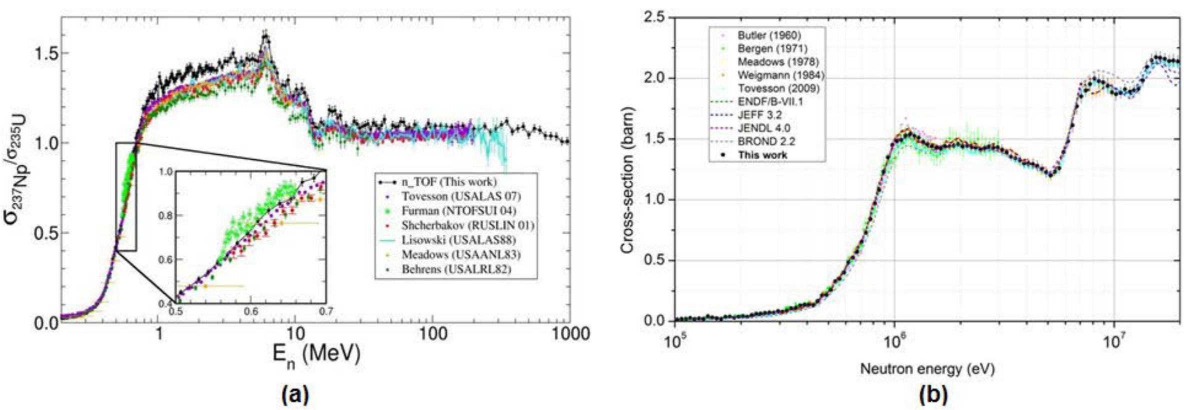


Fig. 6. Data on the ^{237}Np(n,f) (a) and ^{242}Pu(n,f) (b) cross-sections obtained at n_TOF. The data on ^{237}Np(n,f) from the PPAC detector are systematically higher than previous measurements by about 7% above 1 MeV, but are not confirmed by more recent n_TOF results. The ^{242}Pu(n,f) results are in good agreement with recent results obtained at LANL by Tovesson (2009).

measurement performed with this setup. The results above the fission threshold are shown in Figure 6b, while several resonances were also visible above the background from contaminants and from spontaneous fission events.

During the second n_TOF experimental campaign, a concerted effort took place to obtain an evaluated neutron flux. This involved the use of experimental data from different detectors using different reference reactions and comparing results with detailed Monte-Carlo simulations. This work is detailed in Barbagallo (2013). An unexpected outcome of this study was the observation of a possible overestimation of the ^{235}U fission cross-section in the 10-30 keV region by about 6-8% in the evaluations. As can be seen in Figure 7, the flux determined based on the ^{235}U fission cross-section is systematically lower than the evaluated one determined from all setups, as well as from the simulations. Furthermore, the evaluated cross-section also seems to be systematically higher than experimental data in this region. Additional indications from the ^{235}U(n,γ) measurement at DANSCE by Jandel (2012), where the capture cross-section was measured relative to the fission cross-section, also add value to this conclusions and a dedicated measurement is planned to further investigate this issue (Barbagallo (2014)).

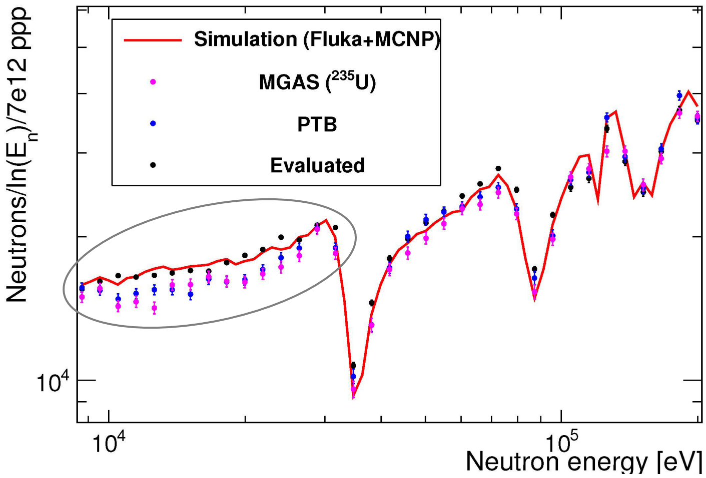


Fig. 7. The comparison of results based on the ^{235}U(n,f) cross-section with the weighted average of all measurements and with simulations, indicates a systematic overestimation of the cross-section in the 10-30 keV range.

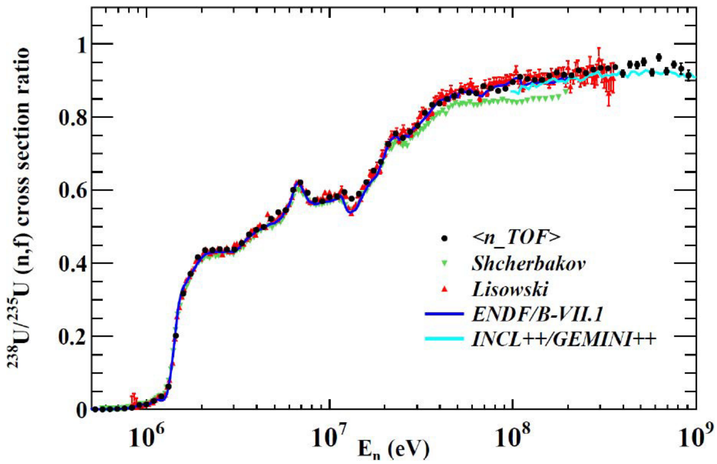


Fig. 8. The n_TOF ^{238}U(n,f)/^{235}U(n,f) ratio, obtained as the weighted average of four datasets obtained with the FIC and PPAC detectors (Paradela (2014)), compared to previous results and model calculations.

Finally, four different datasets obtained with the FIC and PPAC setups on the ^{238}U/^{235}U fission cross-section ratio have been combined by Paradela (2014) to produce an n_TOF ratio, which is shown in Figure 8, compared to previous data, evaluations and model calculations. The n_TOF results may help solve a long-standing discrepancy between the two most important experimental dataset available above 20 MeV, while extending the neutron energy range up to 1 GeV for the first time.

5. Experimental Area II and future scientific programme

A new experimental area (Experimental Area II or EAR-2) (Chiaveri (2012)) is being commissioned in the second half of 2014. EAR-2 is located at the end of a 19 m neutron beam-line placed vertically above the spallation target. The proximity to the target has yielded a gain in flux of more than 20 times compared to the existing experimental area

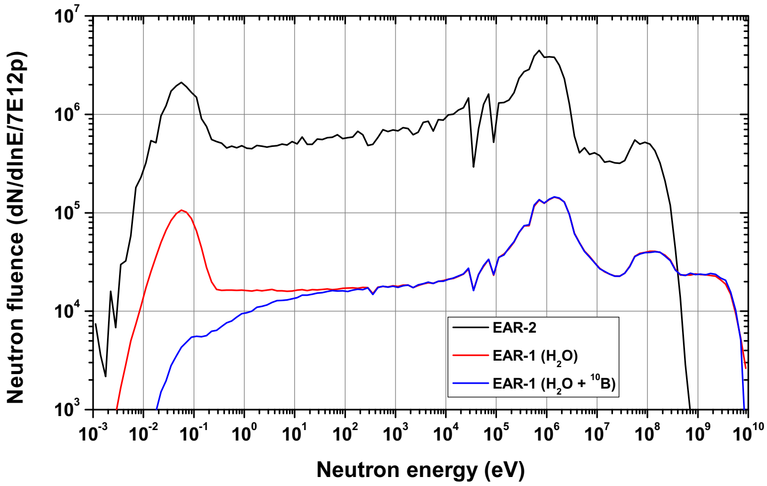


Fig. 9. Comparison of the neutron fluence of Experimental Area II with the fluence in Experimental Area I for the two choices of moderator (water and borated water with ^{10}B enrichment). For most of the energy range, a gain of at least a factor of 20 is obtained.

(EAR-1), as can be seen in Figure 9. The new area will allow n_TOF to expand its measurement capabilities to even more short-lived and rare isotopes, such as ^{230}Th, ^{232}U, ^{238,240}Pu and ^{244}Cm. The scientific programme of EAR-2 is scheduled to begin with the measurement of the fission cross-section of ^{240}Pu (Tsinganis (2014a)) with Micromegas detectors.

6. Conclusions

Since 2001, n_TOF has carried out an extensive research programme that has produced new results on capture and fission cross-sections of relevance to the fields of nuclear astrophysics and advanced nuclear technologies. A variety of detection systems are employed for these measurements and more are being tested for future use. In recent years, a setup for the simultaneous study of capture and fission cross-sections of fissile isotopes has been successfully tested. The commissioning in the second half of 2014 of a new 19 m flight-path (Experimental Area II), with a neutron flux over 20 times higher compared to the existing 185 m beam-line, opens new perspectives for fission measurements at n_TOF with low-mass samples and short-lived isotopes, such as ^{230}Th, ^{232}U, ^{238,240}Pu and ^{244}Cm, while high resolution measurements can still be performed in Experimental Area I during the next measurement period (Phase-III).

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