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Neutron cross-sections for next generation reactors: New data from n_TOF

N. Colonna [a,*], U. Abbondanno [b], G. Aerts [q], H. Álvarez [c], F. Álvarez-Velarde [d], S. Andriamonje [q], J. Andrzejewski [e], P. Assimakopoulos [f], L. Audouin [g], G. Badurek [h], P. Baumann [i], F. Becvar [j], E. Berthoumieux [q], M. Calviani [z], F. Calviño [k], D. Cano-Ott [d], R. Capote [l,m], A. Carrillo de Albornoz [n], P. Cennini [o], V. Chepel [p], E. Chiaveri [o], G. Cortes [r], A. Couture [s], J. Cox [s], M. Dahlfors [o], S. David [g], I. Dillman [t], R. Dolfini [u], C. Domingo-Pardo [v], W. Dridi [q], I. Duran [c], C. Eleftheriadis [w], L. Ferrant [g], A. Ferrari [o], R. Ferreira-Marques [p], H. Frais-Koelbl [l], K. Fujii [b], W. Furman [x], I. Goncalves [p], E. González-Romero [d], A. Goverdovski [y], F. Gramegna [z], E. Griesmayer [l], C. Guerrero [d], F. Gunsing [q], B. Haas [aa], R. Haight [ab], M. Heil [t], A. Herrera-Martinez [o], M. Igashira [ac], S. Isaev [g], E. Jericha [h], F. Käppeler [t], Y. Kadi [o], D. Karadimos [f], D. Karamanis [f], M. Kerveno [i], V. Ketlerov [y], P. Koehler [ad], V. Konovalov [x], E. Kossionides [ae], M. Krticka [j], C. Lampoudis [w,q], H. Leeb [h], A. Lindote [p], I. Lopes [p], M. Lozano [m], S. Lukic [i], J. Marganiec [e], L. Marques [n], S. Marrone [a], T. Martínez [d], C. Massimi [af], P. Mastinu [z], A. Mengoni [l,o], P.M. Milazzo [b], C. Moreau [b], M. Mosconi [t], F. Neves [p], H. Oberhummer [h], S. O'Brien [s], M. Oshima [ag], J. Pancin [q], C. Papachristodoulou [f], C. Papadopoulos [ah], C. Paradela [c], N. Patronis [f], A. Pavlik [ai], P. Pavlopoulos [aj], L. Perrot [q], M.T. Pigni [h], R. Plag [t], A. Plompen [ak], A. Plukis [q], A. Poch [r], C. Pretel [r], J. Quesada [m], T. Rauscher [al], R. Reifarth [ab], M. Rosetti [am], C. Rubbia [u], G. Rudolf [i], P. Rullhusen [ak], J. Salgado [n], L. Sarchiapone [o], I. Savvidis [w], C. Stephan [g], G. Tagliente [a], J.L. Tain [v], L. Tassan-Got [g], L. Tavora [n], R. Terlizzi [a], G. Vannini [af], P. Vaz [n], A. Ventura [am], D. Villamarin [d], M.C. Vicente [d], V. Vlachoudis [o], R. Vlastou [ah], F. Voss [t], S. Walter [t], H. Wendler [o], M. Wiescher [s], K. Wisshak [t], The n TOF Collaboration

[a] *Istituto Nazionale di Fisica Nucleare, V. Orabona 4, 70126 Bari, Italy*
[b] *Istituto Nazionale di Fisica Nucleare, Trieste, Italy*
[c] *Universidade de Santiago de Compostela, Spain*
[d] *Centro de Investigaciones Energeticas Medioambientales y Technologicas CIEMAT, Madrid, Spain*
[e] *University of Lodz, Lodz, Poland*
[f] *University of Ioannina, Greece*
[g] *Centre National de la Recherche Scientifique/IN2P3-IPN, Orsay, France*
[h] *Atominstitut der Österreichischen Universitäten, Technische Universität Wien, Austria*
[i] *Centre National de la Recherche Scientifique/IN2P3-IReS, Strasbourg, France*
[j] *Charles University, Prague, Czech Republic*
[k] *Universidad Politecnica de Madrid, Spain*
[l] *International Atomic Energy Agency (IAEA), Nuclear Data Section, Vienna, Austria*
[m] *Universidad de Sevilla, Spain*
[n] *Instituto Tecnologico e Nuclear (ITN), Lisbon, Portugal*
[o] *CERN, Geneva, Switzerland*
[p] *LIP-Coimbra & Departamento de Fisica da Universidade de Coimbra, Portugal*
[q] *CEA/Saclay-DSM/DAPNIA, Gif-sur-Yvette, France*
[r] *Universitat Politecnica de Catalunya, Barcelona, Spain*
[s] *University of Notre Dame, Notre Dame, USA*
[t] *Forschungszentrum Karlsruhe GmbH (FZK), Institut für Kernphysik, Germany*
[u] *Università degli Studi Pavia, Pavia, Italy*
[v] *Instituto de Fisica Corpuscular, CSIC-Universidad de Valencia, Spain*
[w] *Aristotle University of Thessaloniki, Greece*
[x] *Joint Institute for Nuclear Research, Frank Laboratory of Neutron Physics, Dubna, Russia*
[y] *Institute of Physics and Power Engineering, Kaluga region, Obninsk, Russia*
[z] *Istituto Nazionale di Fisica Nucleare (INFN), Laboratori Nazionali di Legnaro, Italy*
[aa] *Centre National de la Recherche Scientifique/IN2P3-CENBG, Bordeaux, France*
[ab] *Los Alamos National Laboratory, New Mexico, USA*
[ac] *Tokyo Institute of Technology, Tokyo, Japan*
[ad] *Oak Ridge National Laboratory, Physics Division, Oak Ridge, USA*
[ae] *NCSR, Athens, Greece*
[af] *Dipartimento di Fisica, Università di Bologna, and Sezione INFN di Bologna, Italy*
[ag] *Japan Atomic Energy Research Institute, Tokai-mura, Japan*
[ah] *National Technical University of Athens, Greece*

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[ai] Institut für Isotopenforschung und Kernphysik, Universität Wien, Austria
[aj] Póle Universitaire Léonard de Vinci, Paris La Défense, France
[ak] CEC-JRC-IRMM, Geel, Belgium
[al] Department of Physics and Astronomy-University of Basel, Basel, Switzerland
[am] ENEA, Bologna, Italy

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ABSTRACT

In 2002, an innovative neutron time-of-flight facility started operation at CERN: n_TOF. The main characteristics that make the new facility unique are the high instantaneous neutron flux, high resolution and wide energy range. Combined with state-of-the-art detectors and data acquisition system, these features have allowed to collect high accuracy neutron cross-section data on a variety of isotopes, many of which radioactive, of interest for Nuclear Astrophysics and for applications to advanced reactor technologies. A review of the most important results on capture and fission reactions obtained so far at n_TOF is presented, together with plans for new measurements related to nuclear industry.

1. Introduction

Accurate neutron cross-section data are of primary importance for basic and applied Nuclear Physics. In particular neutron data are essential in Nuclear Astrophysics for understanding the production of heavy elements in the Universe, which occurs mainly through slow and rapid neutron capture processes, during the various phases of stellar evolution (Käppeler, 1999). In nuclear technology, a renewed interest in nuclear energy production has triggered new studies aimed at developing future generation systems that would address major safety, proliferation and waste concerns. In particular a possible solution to the waste problem currently being considered is the transmutation of the highly radiotoxic nuclear waste in Accelerator Driven Systems or in Generation-IV fast nuclear reactors (NEA, 2002). Since the incineration process relies on neutron capture or neutron-induced fission (for transuranic elements), it is of crucial importance for the design and safe operation of the new systems to collect accurate neutron cross-section data on the involved isotopes, in particular plutonium, minor actinides, long-lived fission fragments, and structural materials. To perform such measurements, an innovative neutron time facility has been constructed at CERN, Geneva.

2. The n_TOF facility

The CERN n_TOF facility is a time-of-flight installation based on a spallation neutron source (Abbondanno et al., 2002). Neutrons are produced by 20 GeV/c protons from the CERN Proton Synchrotron accelerator, impinging onto a $80\times80\times60$ cm^3 lead block, surrounded by a water layer acting as coolant and moderator of the neutron spectrum. The resulting energy spectrum spans over nine orders of magnitude, from thermal energy to approximately 1 GeV. Together with the wide energy range, other important features of the n_TOF facility are the very high instantaneous flux in the experimental area ($\sim10^6$ n/pulse), which makes it particularly suited for cross-section measurements on radioactive isotopes, and the low duty cycle (0.5 Hz). The measuring station is located at 187.5 m from the spallation target, allowing to reach a very high resolution in neutron energy. Finally, a series of beam shaping collimators and thick iron and concrete shieldings result in a very low ambient background. The neutron flux available in the experimental area for capture and fission measurements is shown in Fig. 1.

3. The experimental setups

The innovative features of the neutron beam have been 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 in the beam, surrounded by an array of Silicon detectors. For capture measurements, two different apparatus are typically used: a set of deuterated benzene (C_6D_6) liquid scintillator detectors, and a 4π BaF_2 calorimeter. The first apparatus is characterized by a low neutron sensitivity, further optimized at n_TOF by minimizing the amount of material surrounding the detectors, and by building the scintillator container and the sample changer in carbon fiber, since carbon is characterized by a low neutron capture cross-sections. To correct for the dependence of the efficiency of the C_6D_6 detectors on the multiplicity of the cascade, the pulse height weighting technique was applied in the analysis of capture data, with the weights determined from accurate Monte Carlo simulations of the experimental setup (Abbondanno et al., 2004). For highly radioactive and fissile isotopes, such as for minor actinides,

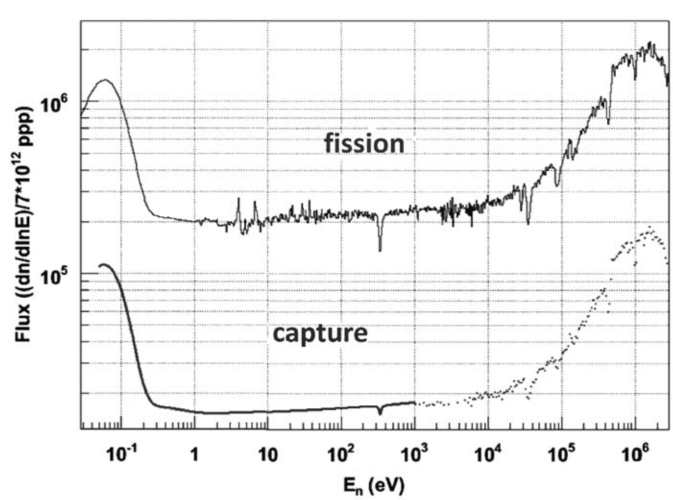

Fig. 1. The energy distribution of the n_TOF neutron fluence in the experimental area, for two different collimators, 1.9 and 6 cm in diameter, which are used for capture and fission measurements, respectively.

* Corresponding author. Tel.: +39 080 5442351; fax: +39 080 5442470.
E-mail address: nicola.colonna@ba.infn.it (N. Colonna).

the calorimetric method was used, where the whole deexcitation cascade is detected in a device with large detection efficiency and complete solid angle coverage. The n_TOF total absorption calorimeter (TAC) is made of 40 BaF_2 crystals. In order to minimize the neutron sensitivity of the apparatus, an inner sphere of a material containing ^6Li ($C_{12}H_{20}O_4(^6Li)_2$) is used, for moderating and absorbing neutrons scattered from the sample. Furthermore, each of the crystals is enclosed in a ^{10}B-loaded carbon fiber capsule, acting as a shield for thermal neutrons. A complete list of capture cross-section measurements performed with the two detection systems is given in Table 1.

The fission cross-section measurements have been carried out with two independent detector systems. The fission ionization chamber (FIC), consists of 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 a 100 μm thick aluminum foil, used as cathode, while the anode are made of a 15 μm thick foil. The samples are 8 cm in diameter and 4–450 μg/cm^2 in thickness. Different FIC's have been built and used in the n_TOF measurements. One of them, containing highly radioactive species like ^{233}U, $^{241,243}Am$ and ^{245}Cm, was qualified as a "sealed source" compliant with the ISO 2919 norm (Calviani et al., 2008a).

The second fission setup consists of a stack of parallel plate avalanche counters (PPACs) to detect both fission fragments in coincidence. The main advantage of the coincidence technique is the very efficient rejection of α-particles. The fission samples were deposited on 1.5 μm thin mylar or 2 μm aluminum foils and are placed between two PPACs.

Both setups allowed to measure simultaneously the fission cross-sections of several isotopes, thus optimizing the use of the beam time. A list of fission cross-section measurements performed at n_TOF in the first measurement campaign is reported in Table 2. In both setups, ^{235}U and ^{238}U samples were used as reference, because their fission cross-sections are considered as fission standards.

The n_TOF data acquisition system is based on a set of 8 bit Acqiris Flash analog to digital converters (FADC) modules, with sampling rates ranging from 100 MS/s for the relatively slow fission detectors to 1 Gs/s for the fast C_6D_6 scintillators used in capture measurements (Abbondanno et al., 2005). The buffer memory of the FADC allows to record data for up to 80 ms, corresponding to neutrons in the thermal energy region. The signals generated in the detectors during a neutron bunch are acquired and stored, after a zero-suppresion procedure, in the CERN central data storage facility. An off-line reconstruction of the digitized signal allows one to extract relevant information on the neutron time-of-flight (and thus on the neutron energy), as well as on the energy deposited in the detectors and on particle identification.

Table 1
Capture cross-sections measurements performed at n_TOF in the first experimental campaign with the two different techniques.

Isotopes	Experimental setup
^{197}Au, ^{151}*Sm*, ^{204,206,207,208}Pb, ^{209}Bi, ^{139}La, ^{232}*Th*, ^{24,25,26}Mg, ^{90,91,92,93,94,96}Zr, ^{186,187,188}Os	C_6D_6—pulse height weighting technique
^{197}Au, ^{233,234}*U*, ^{237}*Np*, ^{240}*Pu*, ^{243}*Am*	BaF_2—total absorption calorimeter

Radioactive isotopes are indicated in italic.

Table 2
Measurements of neutron-induced fission cross-sections performed at n_TOF in the first experimental campaign with the two different experimental setups.

Isotopes	Experimental setup
^{nat}Pb, ^{209}Bi, ^{232}*Th*, ^{237}*Np*, ^{233,234,235,238}*U*	PPAC—coincidence technique
^{232}*Th*, ^{237}*Np*, ^{233,234,235,236,238}*U*, ^{241,243}*Am*, ^{245}*Cm*	FIC—single fragment detection

Radioactive isotopes are indicated in italic.

4. The results

The data collected at n_TOF have allowed to improve the accuracy of neutron cross-sections for a great number of isotopes, helping to solve existing discrepancies between previous experimental data or among cross-section databases. In some cases, new evaluations accounting for the n_TOF results have been performed, making available to the nuclear industry long-needed reliable data for the design of advanced systems for energy production. Among the various results, the measurement of the $^{232}Th(n,\gamma)$ reaction is one of the most relevant. Thanks to the features of the neutron beam, and to the performance of the experimental setup, the cross-section measured at n_TOF for this isotope is characterized by high accuracy (~5%) and high resolution. In particular, in the energy region between 10 keV and 1 MeV, the n_TOF data, shown in Fig. 2, have allowed to finally solve a 40% discrepancy between previous measurements, as shown in Aerts et al. (2006), and provided the basis for a new, more reliable evaluation than currently available (Sirakov et al., 2008). Important results on capture reactions have also been obtained with the TAC on ^{247}Np and ^{240}Pu, produced in current nuclear power reactors. These data are needed in transmutation projects or for the design of Gen IV fast nuclear reactors (Guerrero et al., 2008). The n_TOF results on these reactions, when completely analyzed, will be among the most accurate available.

The design of advanced nuclear reactors for energy production, in particular of Gen IV fast reactors, requires new data on fission reactions for several minor actinides, since currently available cross-sections are too uncertain 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 to 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

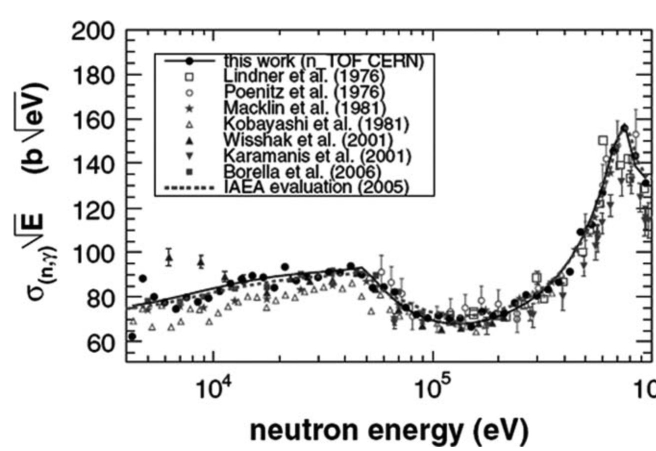

Fig. 2. The $^{232}Th(n,\gamma)$ cross-sections measured at n_TOF compared with evaluations and previous results, in the unresolved resonance region, between 1 keV and 1 MeV neutron energy. A recent evaluation including the new n_TOF results is also shown in the figure by the dashed curve.

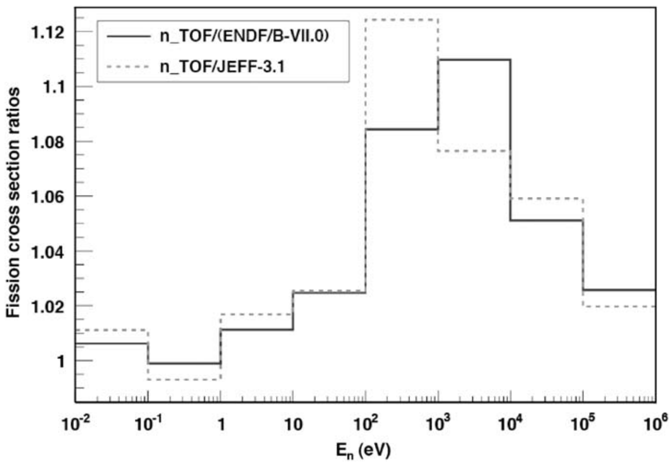

Fig. 3. The ratio between the $^{233}U(n,f)$ cross-section measured at n_TOF and two recent evaluations. The shortcoming of the cross-section databases is evident in the region between 10 eV and 100 keV, where the n_TOF results show a cross-section 5%–10% higher than currently evaluated.

to measure fission cross-sections up to several tens of MeV, a feature important for applications to fast reactors and to extract fundamental information on fission barriers. A typical example of the high quality results on fission cross-section is presented in Fig. 3, where the results for the $^{233}U(n,f)$ reaction are shown, relative to the current evaluations in the ENDF/B-VII.0 and JEFF-3.1 data libraries (Calviani et al., 2008b, 2009). We would like to remark that a great advantage offered by the n_TOF facility is the possibility to measure simultaneously the fission cross-sections in an energy range extending over several orders of magnitude, from thermal to several hundreds of MeV, allowing to minimize uncertainties related to the absolute normalization of the cross-sections.

The high quality data collected at n_TOF will allow to improve the current accuracy of cross-section databases, a fundamental prerequisite for the design of new reactors based on the Th/U fuel cycle.

5. Conclusions and perspectives

The first experimental campaign at the n_TOF facility was successfully completed with a wealth of new results on capture and fission cross-sections of interest for emerging nuclear technologies, as well as for other important applications, such as Nuclear Astrophysics and basic Nuclear Physics. Of particular importance are the measurements of highly radioactive samples, for which accurate new data have been obtained, thanks to the high instantaneous neutron flux of the facility. After a refurbishment of the spallation target, a second measurement campaign, n_TOF_Ph2, will take place with a series of new capture and fission measurements of interest for applications to the nuclear industry, in particular to Gen IV fast reactors. Among others, measurements on high-mass Pu isotopes and minor actinides are foreseen, in some cases with experimental setups modified to detect capture and fission reactions simultaneously. Together with the application 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 neutron flux 100 times larger than the present one, making possible measurements currently not feasible.

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