

Neutron capture cross section measurements of ^{238}U , ^{241}Am and ^{243}Am at n_TOF

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

Spokespersons: Daniel Cano Ott, Frank Gunsing

Technical coordinator: Vasilis Vlachoudis

S. Andriamonje¹, J. Andrzejewski², L. Audouin³, V. Avrigeanu⁴, F. Becvar⁵, F. Belloni⁶, B. Berthier³, E. Berthoumieux⁷, M. Brugger¹, F. Calviño⁸, M. Calviani¹, D. Cano-Ott¹, C. Carrapiço^{7,12}, F. Cerutti¹, N. Colonna¹³, I. Dillmann¹⁴, C. Domingo-Pardo¹⁵, E. Dupont⁷, I. Duran¹⁶, A. Ferrari¹, W. Furman¹⁸, S. Ganesan¹⁹, B. Gomez⁸, I.F. Gonçalves¹², E. González¹¹, V. Gorlychev⁸, F. Gramegna⁹, C. Guerrero¹¹, F. Gunsing⁷, R. Haight²⁰, S. Harissopulos²¹, M. Heil¹⁵, M. Igashira²², K. Ioannides²³, E. Jericha²⁴, F. Käppeler²⁵, Y. Kadi¹, P. Koehler²⁶, F.G. Kondev²⁷, M. Krlická⁵, E. Leebos¹, C. Lederer²⁸, H. Leeb²⁴, R. Losito¹, J. Marganiec², S. Marrone¹³, T. Martínez¹¹, C. Massimi², P.F. Mastinu⁹, A. Mengoni^{30,31}, W. Mezentseva^{18,32}, M. Mirea⁴, P.M. Milazzo⁶, M. Mosconi³³, C. Paradela¹⁶, A. Pavlik³⁴, R. Plag¹⁵, J. Praena^{9,35}, J.M. Quesada³⁵, J. Perkowski², R. Reifarh^{15,36}, C. Rubbia¹, C. Sage⁷, R. Sarmiento¹², F. Sommerer¹, G. Tagliente¹³, J.L. Tain³⁷, L. Tassan-Got³, G. Vannini²⁹, V. Variale¹³, P. Vaz¹², A. Ventura³¹, D. Villamarin¹¹, V. Vlachoudis¹, R. Vlastou³⁸ and A. Wallner²⁸

(The n_TOF Collaboration, http://cern.ch/n_TOF/)

¹CERN, Geneva, Switzerland

²University of Lodz, Lodz, Poland

³Centre National de la Recherche Scientifique/IN2P3 - IPN, Orsay, France

⁴National Institute of Physics and Nuclear Engineering - IFIN, Bucharest, Romania

⁵Charles University, Prague, Czech Republic

⁶Istituto Nazionale di Fisica Nucleare, Trieste, Italy

⁷CEA Saclay, IRFU, F-91191 Gif-sur-Yvette, France

⁸Universidad Politécnica de Cataluña, Spain

⁹Istituto Nazionale di Fisica Nucleare,
Laboratori Nazionali di Legnaro, Italy

¹⁰Dipartimento di Fisica, Università di Padova, Italy

¹¹Centro de Investigaciones Energéticas Medioambientales y Tecnológicas – CIEMAT, Madrid, Spain

¹²Instituto Tecnológico e Nuclear - ITN, Lisbon, Portugal

¹³Istituto Nazionale di Fisica Nucleare, Bari, Italy

¹⁴Physik-Department E12, Technische Universität München,
Beschleunigerlaboratorium, Garching, Germany

¹⁵GSI, Darmstadt, Germany

¹⁶Universidade de Santiago de Compostela, Spain

¹⁸Joint Institute for Nuclear Research,

Frank Laboratory of Neutron Physics, Dubna, Russia

¹⁹BARC, Mumbai, India

²⁰Los Alamos National Laboratory, New Mexico, USA

²¹NCSR Demokritos, Athens, Greece

²²Tokyo Institute of Technology, Tokyo, Japan

²³University of Ioannina, Greece

²⁴Atominstytut der Österreichischen Universitäten,
Technische Universität Wien, Austria

²⁵Forschungszentrum Karlsruhe GmbH - FZK, Institut für Kernphysik, Germany

²⁶Oak Ridge National Laboratory, Physics Division, Oak Ridge, USA

²⁷Argonne National Laboratory, Nuclear Engineering Division, Argonne, Chicago, USA

²⁸VERA Laboratory - Isotopenforschung & Kernphysik,
Faculty of Physics, University of Vienna, Austria

²⁹Dipartimento di Fisica, Università di Bologna,
and Sezione INFN di Bologna, Italy

³⁰International Atomic Energy Agency - IAEA,
Nuclear Data Section, Vienna, Austria

³¹ENEA, Bologna, Italy

³²IRMM, Geel, Belgium

³³Physikalisch-Technische Bundesanstalt - PTB, Braunschweig, Germany

³⁴Institut für Isotopenforschung und Kernphysik, Universität Wien, Austria

³⁵Universidad de Sevilla, Spain

³⁶Universität Frankfurt am Main, Frankfurt, Germany

³⁷Instituto de Física Corpuscular, CSIC-Universidad de Valencia, Spain

³⁸National Technical University of Athens, Greece



Abstract

The increase of the world energy demand and the need of low carbon energy sources have triggered the renaissance and/or enhancement of nuclear energy in many countries. Fundamental nuclear physics can contribute in a practical way to the sustainability and safety of the nuclear energy production and the management of the nuclear waste. There exists a series of recent studies which address the most relevant isotopes, decay data, nuclear reaction channels and energy ranges which have to be investigated in more detail for improving the design of different advanced nuclear systems [1] and nuclear fuel cycles [2]. In this proposal, we aim at the measurement of the neutron capture cross sections of ^{238}U , ^{241}Am and ^{243}Am . All three isotopes are listed in the NEA High Priority Request List [37], are recommended for measurements [1] and play an important role in the nuclear energy production and fuel cycle scenarios. The measurements will provide as well valuable nuclear structure data necessary for the improvement of nuclear models and the statistical interpretation of the nuclear properties.

I. Introduction

New concepts for nuclear systems are being explored to improve the sustainability of nuclear energy that appears in the EU SET plan [3] as an unavoidable component in the mix for the energy generation. These new concepts cover from evolutionary light water reactors, that most probably will remain the main component of the nuclear park along this century, to fast reactors foreseen in generation IV, as the only really long term sustainable systems by optimizing the natural resources, and include the subcritical fast systems, ADS, especially suited for the intensive waste minimization.

Despite the many previous measurements and recent efforts, the optimization and future industrial deployment of these new concepts still challenge the present level of basic nuclear data knowledge. The availability of all the required data, their quality of and the description of their uncertainties in the most recent databases and simulation tools need to be improved.

The main challenges come from:

1. Isotopes which have become relevant due to their increased presence in the fuels for the new generation of reactors. This applies in particular to the systems aiming at the Pu and Minor Actinides (MA) recycling: fast reactors and ADS, but also Gen III(+) reactors with a high Pu load.
2. The proposal of fast systems, which has enhanced the relevance of the region from 1eV to several MeV for all isotopes and, very especially, for high mass actinides.
3. New fuel cycles with multi-recycling of actinides that could drive to new levels of accumulation of minor isotopes, thus generating additional risks and/or costs at several stages of the fuel cycle. One of the most important aspects is the propagation of the uncertainties as the number of irradiation cycles increases.
4. New requirements on the level of precision. In the nuclear industry as everywhere else, the computer simulations should help minimizing (but not replacing) the need for small scale and costly experimental demonstrators. This new role of the

simulation tools will require a higher level of precision that can only be achieved if the precision of initial basic nuclear data is previously improved.

5. Better assessment of the uncertainties on the data and on the derived magnitudes. This requires a more realistic evaluation of the cross section uncertainties and their correlations between different energies and different channels. Only in this way the estimations from the simulations will profit from the enhanced basic data precision and from the constraints set by the integral experiments.

The n_TOF facility at CERN was built in 2000 with the goal of providing to the international nuclear data community a unique place where to measure neutron induced cross sections of highly radioactive isotopes available only in small amount. Since then, it has done essential contributions to the field of the nuclear data for the science and the technology and, in particular, has produced excellent data sets on the neutron capture cross section of ^{237}Np and ^{240}Pu [4].

The n_TOF collaboration proposes to continue this successful program with the cross sections of Actinides and Minor Actinides with the three measurements in this proposal: ^{238}U , ^{241}Am and ^{243}Am . All three isotopes are priority and appear in the NEA High Priority List [5], the NEA WPEC Subgroup 26 Report on the accuracy of nuclear data for advanced reactor design [1] and the EU 6th Framework Programme IP-EUROTRANS/NUDATRA reports [2].

In addition, such measurements are also of relevance to fundamental science in many aspects. Improved resonance parameters of p-waves in ^{238}U can lead to a better determination of the parity violation effects observed in ^{239}U resonances [6]. The excellent characteristics of the n_TOF facility have been demonstrated in a previous experiment on the $^{232}\text{Th}(n,\gamma)$ cross section [7]. The use of the n_TOF total absorption calorimeter as a γ -ray spectrometer will provide unique data for the Photon Strength Functions models and nuclear level densities in heavy nuclei [8].

II. The ^{241}Am (n, γ) cross section

Two different TOF capture data sets are available in the resolved resonance region: the (n,abs) data from Weston [9] and the (n, γ) data from Vanpraet [10]. Both sets correspond to massive and highly radioactive ^{241}Am samples, consisting of 2 g of AmO_2 and 1.7 g of metallic Am respectively.

Large differences up to 30% are found between the two data sets since Vanpraet's values are not corrected for self-screening effects. Additional experimental resonance parameters have been obtained from absorption [9] and indirect transmission/fission [11] measurements.

A more recent measurement has been performed at Los Alamos National Laboratory LANSCE facility with the DANCE total absorption calorimeter [12]. However, the short flight path of 18 m used at LANSCE makes the resonance analysis at energies above a few tens of eV nearly impossible.

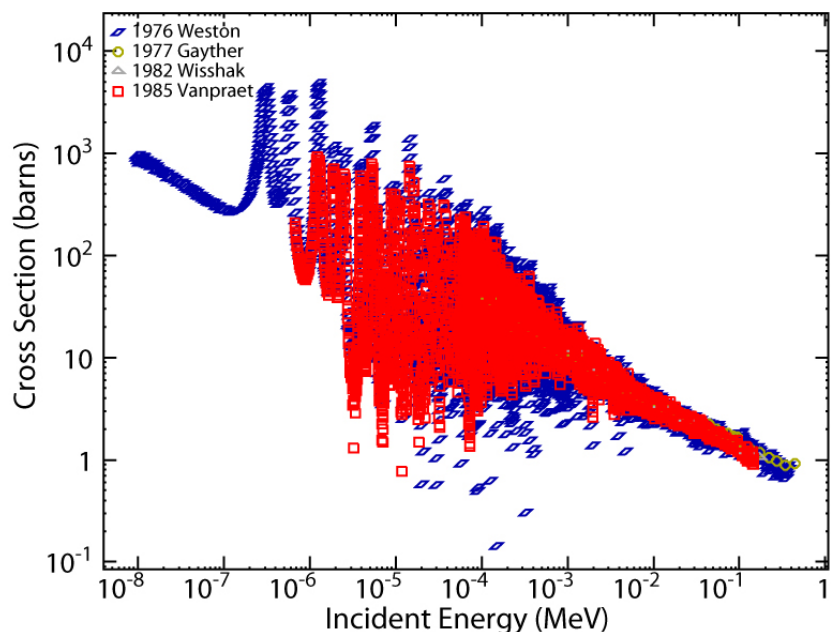


Figure 1. ^{241}Am (n, γ) cross section data available at the EXFOR [15] database by April 2009.

In the unresolved resonance region up to 1 MeV there exist only two TOF data sets by Wisshak [13] and Weston [14], ranging from 0.3 keV to 100 keV and 5 keV to 100 keV respectively. No data are available above 100 keV. As it is shown in Figure 7, there is a systematic bias between the two data sets of about 10%. One more experimental point for the neutron capture cross section is available above 1 MeV from an activation measurement [16].

The measurement at n_TOF will provide high resolution and high accuracy data up to neutron energies of a few tens of keV. The resonance analysis, combined with a recent transmission measurement performed at IRMM will lead to an improved and high quality set of resonance parameters.

III. The ^{243}Am (n, γ) cross section

The amount of available data for ^{243}Am (n, γ) is scarce. In the resolved resonance region, experimental radiative widths have been obtained indirectly from transmission and fission TOF measurements [17][18][19]. No direct TOF cross section data are available in the EXFOR database.

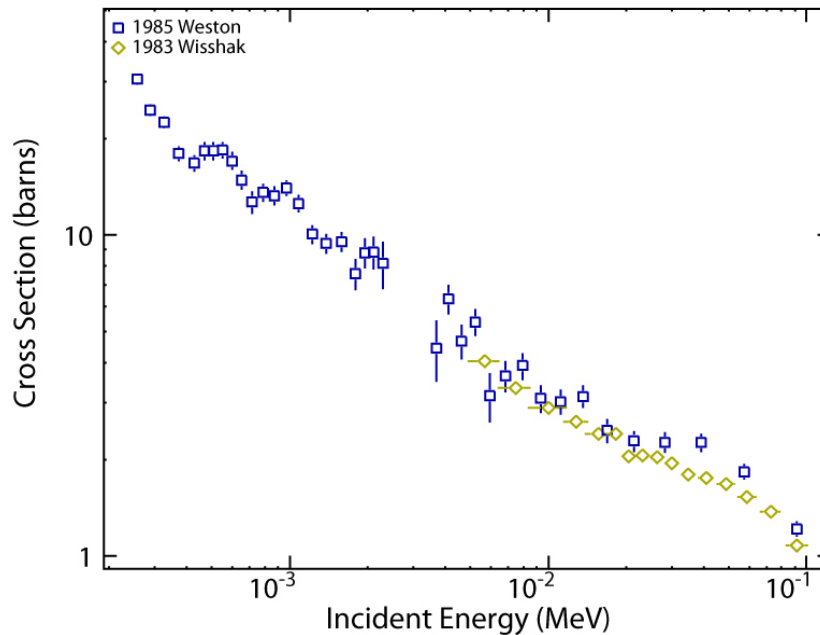


Figure 2. ^{243}Am (n,γ) cross section data available at the EXFOR [15] database by April 2009.

In the unresolved resonance region two additional sets of TOF data from Gayther [20] and Wisshak [21] are available. Gayther's data show a good agreement with those from Vanpraet and Weston up to 2 keV, where differences start to be sizeable. According to the authors, the uncertainties of the measurements are of the order of 10% or higher. As it can be seen in Figure 6, above 2 keV the data from Gayther start to deviate systematically by 10% or more. Such a discrepancy could be attributed to uncertainties in the normalization, as suggested in a recent evaluation work from Ignatyuk [22]. The data by Wisshak [21] range from 10 keV to 250 keV and are in good agreement with Gayther's measurement within the uncertainties of 5% to 20%. No other experimental points are available above 400 keV except the data from Flerov, standing for the $^{241\text{m}}\text{Am}$ isomeric state capture cross section, and the values from several activation measurements [23][24][16].

The evaluated uncertainty of the ^{243}Am neutron capture cross section is, on average, 7-10% in the range between thermal energies up to 100 keV. One should notice that such a value stands from one single capture data set in the fast region and should be taken under caution. On the other hand, the target accuracy request for an ADS is 2-3%. Thus, a new accurate measurement is mandatory and has been proposed as part of the EU 6th Framework Programm project IP-EUROTRANS under the domain NUDATRA [25].

IV. The ^{238}U (n,γ) cross section

In contrast to the Am isotopes, the number of measurements on the $^{238}\text{U}(n,\gamma)$ is much larger, over 25 datasets in subregions of the resolved resonance region, and a few less in the unresolved energy region are available in the EXFOR database. Nevertheless, inconsistencies are still present for the capture cross section up to 25 keV. The goal of proposing a new measurement is the reduction of the actual uncertainty in the cross

section to a value below 2% in the range of a few eV up to hundred keV. Such a challenge is only achievable through an ongoing European effort, which consists in a series of measurements combining different facilities, experimental techniques and analysis methodologies (See Section V for details). The data obtained at n_TOF with the Total Absorption Calorimeter [26] and the carbon fibre C₆D₆ [27] detectors will be combined with two new transmission and capture measurements to be performed at the IRMM GELINA facility [28]. The entire data set will be analyzed in common with two different resonance analysis codes, SAMMY [30] and REFIT [31], in order to produce the best ²³⁸U(n,γ) cross section data in the world and, eventually, lead to a new standard.

V. The experimental setups

The quantity determined in a neutron capture experiment is the capture yield, i.e. the fraction of neutrons incident on a sample (with thickness n atoms per barn) and undergoing the (n,γ) interaction. The capture yield $Y(E)$, with $0 < Y(E) < 1$, for the first interaction can be written as:

$$Y(E) = (1 - e^{-n\sigma_T(E)}) \frac{\sigma_\gamma(E)}{\sigma_T(E)} \approx \begin{cases} n\sigma_\gamma & n\sigma_T \ll 1 \\ \sigma_\gamma / \sigma_T & n\sigma_T \gg 1 \end{cases}$$

with σ_T the total and σ_γ the neutron capture cross section. The two limiting cases are approximations for thin respectively thick samples.

The observable quantity in these measurements is the counting rate C as a function of the energy, which is related to the reaction yield by

$$Y(E) = \frac{C(E) - B(E)}{\varepsilon \Phi(E)}$$

where B accounts for the background rate, ε is the efficiency of the detector for counting an (n,γ) reaction and $\Phi(E)$ is the incident neutron flux. The systematic uncertainties in these type of measurements are given by the accuracy in the determination of the detection efficiency, the characterization of the background and the neutron fluence.

Two techniques are used at n_TOF for measuring (n,γ) cross sections:

- The total absorption technique, which consists in the use of a nearly 100% efficiency calorimeter which absorbs all the gamma rays emitted in the radiative capture process and therefore counts the number of (n,γ) directly.
- The total energy detector technique, which consists in the use of so-called total energy detectors [38] that have a γ-ray detection efficiency which is proportional to the γ-ray energy. In this way, if the detector efficiency is low, one can count (n,γ) reactions with an efficiency proportional to the total energy of the electromagnetic cascade associated to the radioactive capture process.

Both are complementary and have their own advantages and disadvantages. The total absorption, based on large inorganic scintillators, allows using the information on the multiplicity (due to segmentation) and the energy (due to the good energy resolution) for the suppression of background events during the analysis. In this way, the

background due to the sample and environmental radioactivity, typically of low multiplicity, can be separated very efficiently from the (n,γ) electromagnetic cascades. The technique is ideal for measuring highly radioactive samples of isotopes available in small amount and has been applied very successfully at n_TOF in the past [4].

On the other hand, the total energy detectors are usually organic scintillators with a low energy resolution and nearly null photopeak efficiency. Therefore, the background separation becomes more involved and requires a careful characterization and subtraction. The advantage here becomes significant when measuring isotopes with a large elastic to capture cross section ratio. One of the most harmful sources of background that appear in neutron capture measurements is due to the detection of neutrons scattered by the sample (or any other material) along the beam line. Such a scattering can follow the resonant structure of the cross section and lead to an inaccurate determination of the resonance parameters. Such effects have been noticed in the past [31,40,45] and for this reason, the n_TOF collaboration has built new total energy detectors based on the C_6D_6 scintillator and encapsulated in a carbon fibre housing [27]. They have an extremely low neutron sensitivity and will be necessary mainly for the ^{238}U measurement.

The measurements proposed here will combine the two techniques separately for reducing as much as possible the systematic uncertainties associated to the different methodologies involved.

V.a. The Total Absorption Calorimeter (TAC)

The n TOF Total Absorption Calorimeter (TAC) is an improved version of the BaF_2 calorimeter at FZK [32] and has been designed in order to fulfill all the requirements of an ideal total absorption detector: large solid angle coverage, high γ -ray total absorption efficiency, good energy resolution, high segmentation, low neutron sensitivity, and fast time response.

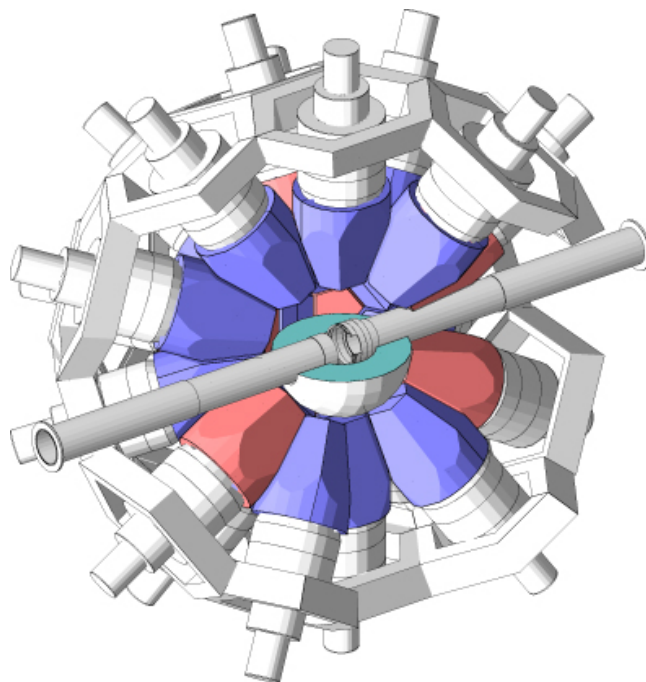


Figure 3. The n_TOF Total Absorption Calorimeter.

The TAC consists of 40 BaF₂ crystals, 12 pentagonal and 28 hexagonal, that form a sphere with an inner radius of 10.5 cm covering a large solid angle (95% of 4π) with respect to the sample position at the centre of the TAC. Two holes in opposite positions are left in order to allow the entrance and exit of the beam line through the detector. Both types of crystals, pentagonal and hexagonal, are shaped from BaF₂ cylinders 14 cm in diameter and 15 cm in thickness, with raw and final weights of 12 kg and 7.5 kg, respectively.

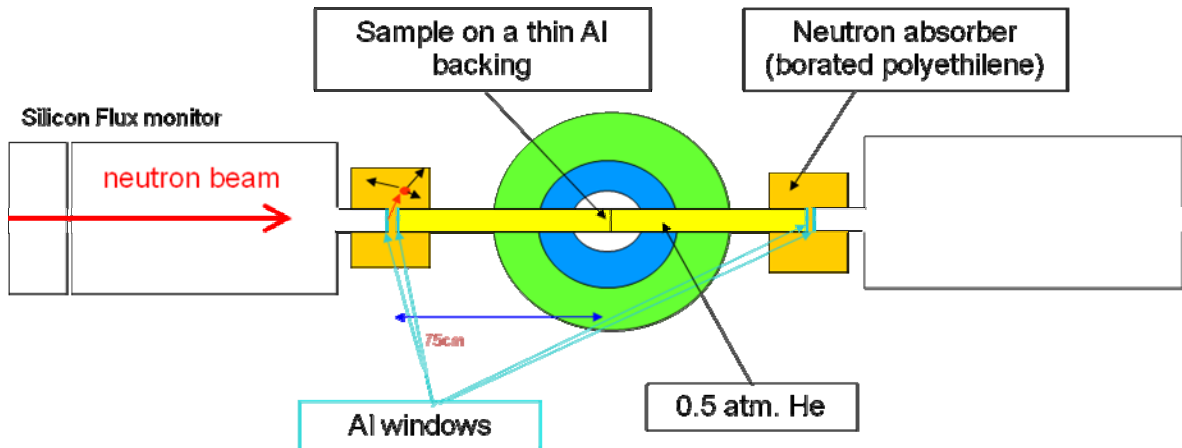


Figure 4. Scheme of the TAC used in combination with long sample containers.

The neutron sensitivity of the TAC set-up is minimized by the combination of a neutron moderator/absorber surrounding the sample and the use of crystal capsules with a high content in ¹⁰B. The idea behind this design is that neutrons scattered at the sample position are moderated before reaching the crystals and then absorbed either inside the actual moderator or in the capsules of the crystals by means of ¹⁰B(n,γ)⁷Li reactions. Furthermore, the scattering in dead material like vacuum windows has been minimized as well by using a long sample container: the samples will be deposited on a thin aluminum backing inside a sealed container whose windows are outside the TAC. In this way, it will be possible to shield the windows with thick borated polyethylene neutron absorbers and reduce the neutron sensitivity in a factor of 10 with respect to measurements performed in 2004.

The neutron sensitivity will be determined experimentally as a function of the neutron energy by measuring the response of the TAC to the neutrons scattered by a sample with low capture cross section (typically graphite).

V.b. The low sensitivity C₆D₆ detectors

The neutron capture experiments of ²³⁸U will be performed as well using C₆D₆ γ-ray detectors together with a pulse height weighting technique [33-35]. The pulse height weighting is the method of making the detection efficiency independent of the gamma-ray cascade of the (n,γ) reaction and is based on low-efficiency detectors in order to detect not more than a single gamma ray per cascade.

We plan to use C₆D₆-based γ-ray detectors in a standard 90 degrees geometry. The overall detection efficiency for a capture event, taking into account the γ-ray

multiplicity, is about 20% in this configuration. A schematic view of the setup is shown in figure 5.

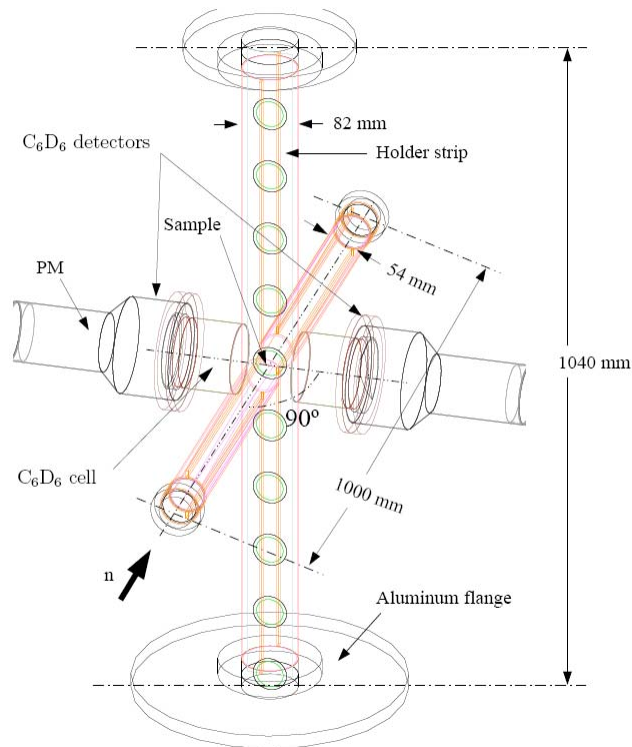


Figure 5. Scheme of the setup with two C_6D_6 detectors and a carbon fibre sample changer.

The liquid scintillator detectors based on deuterated benzene C_6D_6 have an overall efficiency (including solid angle) of a few percent for γ -rays in the range of interest between 0.1-10 MeV. Taking into account the gamma-ray multiplicity, with this setup the efficiency for a capture event is about 20%. They have also the advantage of being the least sensitive to scattered neutrons as compared to other γ -ray detectors. However, it is not possible with C_6D_6 -based detectors in the standard setup to distinguish whether the detected γ -rays originate from the (n,γ) reaction, the radioactive background or from competing reaction channels like fission or inelastic scattering. Therefore, the separate background components will have to be measured independently (when possible) or estimated by Monte Carlo simulations.

VI. The samples

All the radioactive samples should have an isotopic enrichment $> 99\%$ and the masses are listed in Table 1. The ^{241}Am and ^{243}Am samples will be encapsulated inside aluminum containers fulfilling the ISO-2919 norm. The containers will be about 1.5 m long and have a diameter of 5 cm. The samples will be placed inside the container, centered in both radial and longitudinal coordinates and deposited on a very thin Al backing. The atmosphere inside the container will be He at low pressure. The ^{238}U sample will not be encapsulated and placed either in air or in vacuum.

In addition to the actinides, gold samples of various thicknesses will be used as a reference at 4.9 eV (first resonance) and in the high energy region above 100 keV, where the (n, γ) cross section is a standard.

The neutron scattering background and in-beam γ -ray background (see [36] for details) will be characterized by measuring the response of the detection systems to ^{nat}C and ^{nat}Pb samples. Such measurements are necessary for the background subtraction and should be performed with sufficient statistics.

VII. Beam time request

The beam time request is based on the experience acquired in previous measurements and on the reaction rates (Figures 5 to 7) with the most recent simulations of the neutron beam fluence. It is summarized in Table 1.

Isotope	Mass	Requested Number of protons			
		TAC		C_6D_6	
		days (at 0.2 Hz)	protons	days (at 0.2 Hz)	protons
^{238}U	70 mg	20	2.42E+18	30	3.63E+18
^{241}Am	2 mg	25	3.02E+18	3	3.63E+17
^{243}Am	10 mg	20	2.42E+18	3	3.63E+17
^{197}Au	100 mg	2	2.42E+17	3	3.63E+17
^{nat}C	100mg	2	2.42E+17	3	3.63E+17
^{nat}Pb	100 mg	1	1.21E+17	1.5	1.81E+17
Empty container	-	1	1.21E+17	1.5	1.81E+17
Total		71	8.59E+18	45	5.44E+18
Grand total					1.40E+19

Table 1. Beam time request for the different measurements. For the calculation of the beam time period (express in days) an average PS frequency of 0.2 Hz was assumed.

The criteria adopted for the TAC measurements are:

- ^{238}U . To reach an average statistical uncertainty below 2% in the pseudo-resonance integrals at 1 keV.
- ^{241}Am and ^{243}Am . To reach an average statistical uncertainty below 3% in the pseudo-resonance integrals at 1 keV in each measurement.

The criteria adopted for the measurements with the C_6D_6 are:

- ^{241}Am and ^{243}Am . Three days of beam time are requested at a PS pulse rate of 0.2 Hz in order to determine the resonance parameters of the lowest lying (and strongest) resonances in the eV region and for investigating the feasibility of a measurement with the C_6D_6 at higher neutron energies.
- ^{238}U . The first goal is to reach a statistical uncertainty $< 2\%$ in the energy range between 10 keV and 100 keV for a binning of 100 bins/decade. Such a condition will lead to sufficient statistics in the resolved resonance region. As a second goal we would like to reduce the statistical error to a negligible level so that only

systematic uncertainties will limit the accuracy of the measurement. Since it seems unrealistic to increase the beam time, we will study the possibilities to be able to use ^{238}U with a mass of the order of grams.

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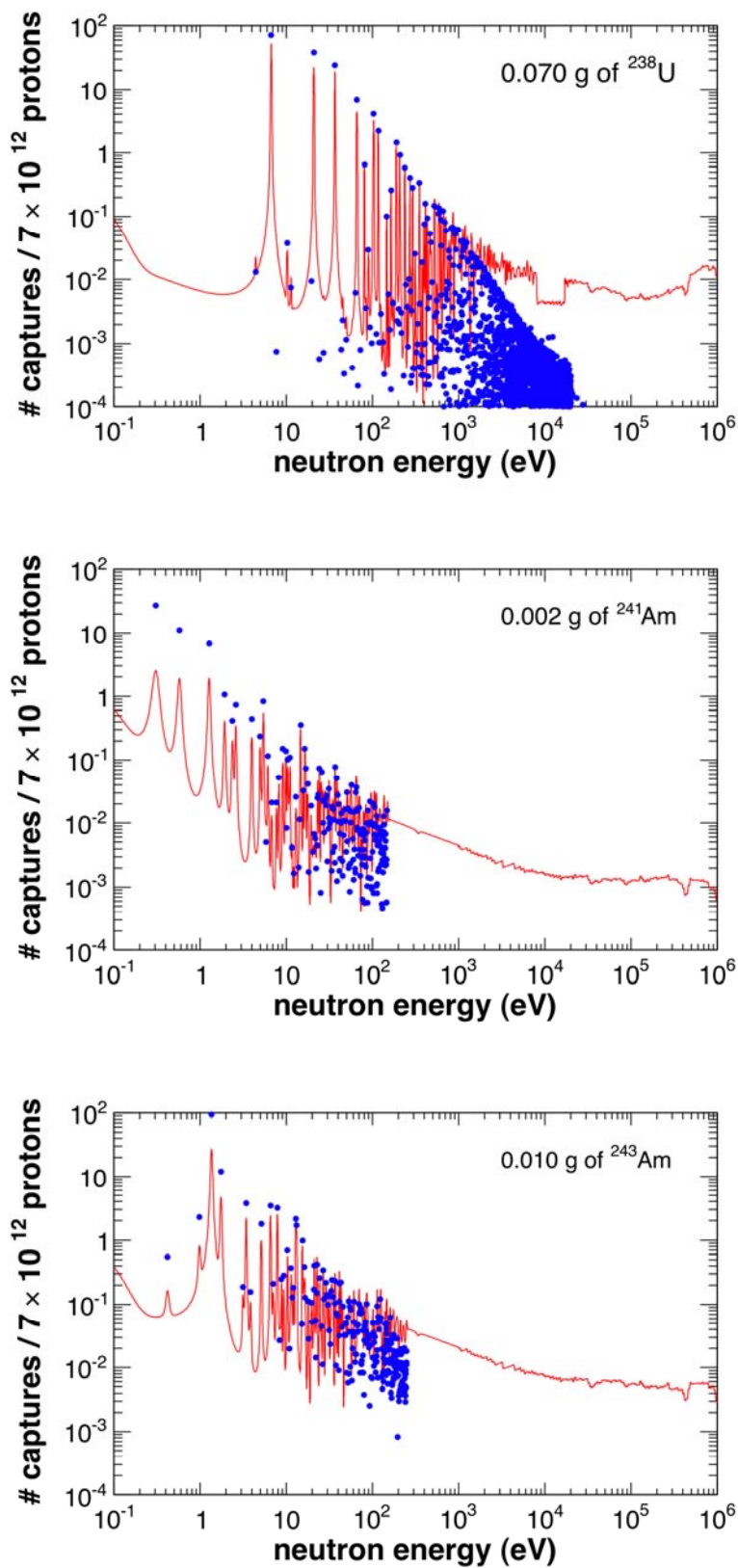


Figure 6. The reaction rates of $^{238}\text{U}(n,\gamma)$, $^{241}\text{Am}(n,\gamma)$ and $^{243}\text{Am}(n,\gamma)$ for a single normalized proton bunch. The red curve shows the number of capture reactions per bin using 100 bins per energy decade. The blue points indicate the number of capture reactions integrated over the resonance.

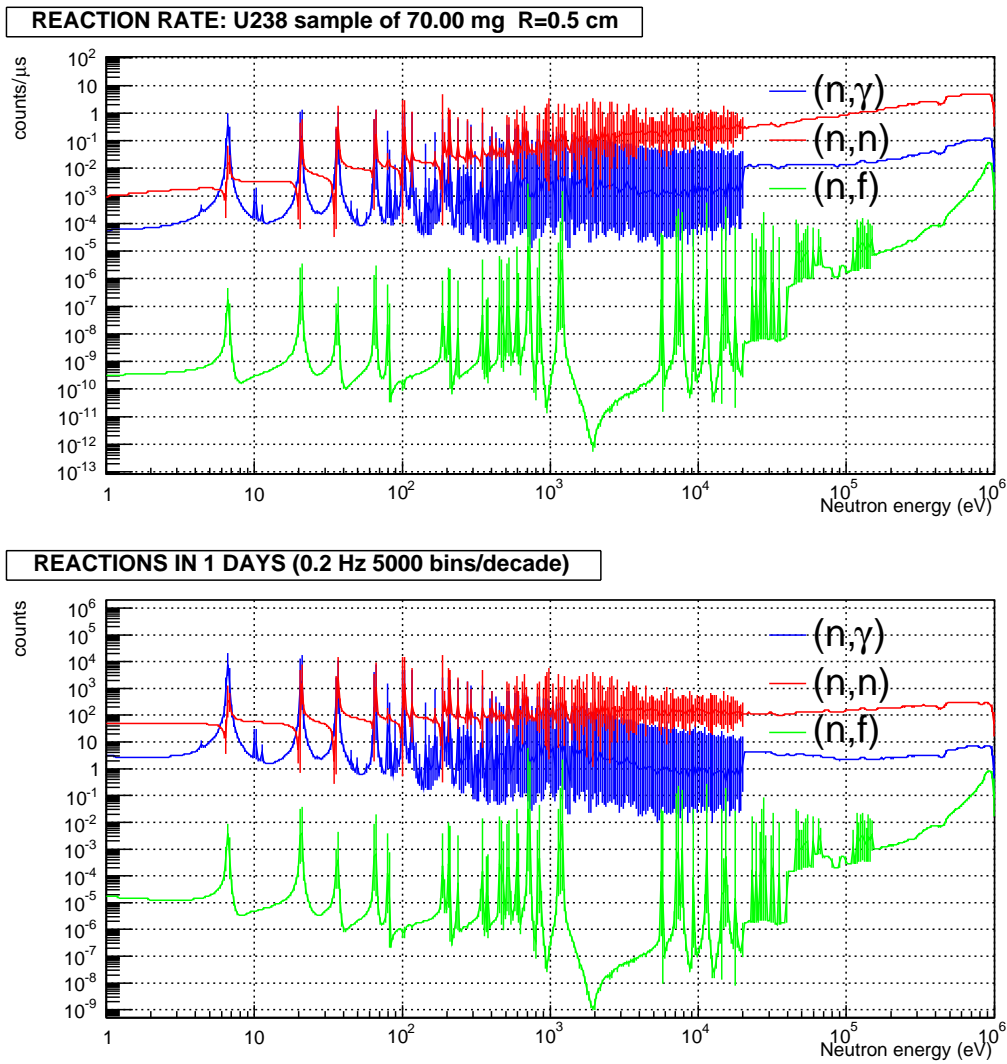
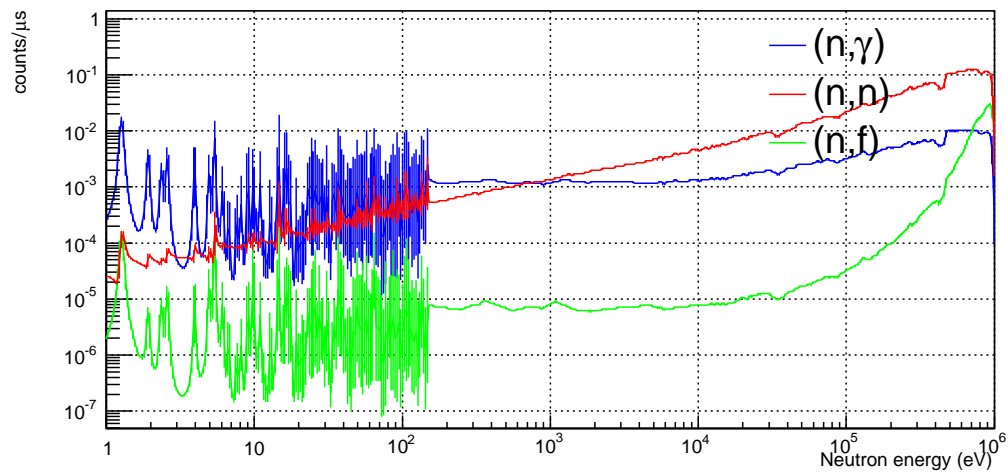


Figure 7. Top: $^{238}\text{U}(n,\gamma)$ reaction rates (in red) in counts/ μs compared to the elastic (blue) and neutron induced fission (green) rates for one PS pulse of $7 \cdot 10^{12}$ protons. Bottom: Number of reactions in one day of beam time. An 0.2 Hz PS average repetition rate was assumed.

REACTION RATE: Am241 sample of 2.00 mg R=0.5 cm



REACTIONS IN 1 DAYS (0.2 Hz 5000 bins/decade)

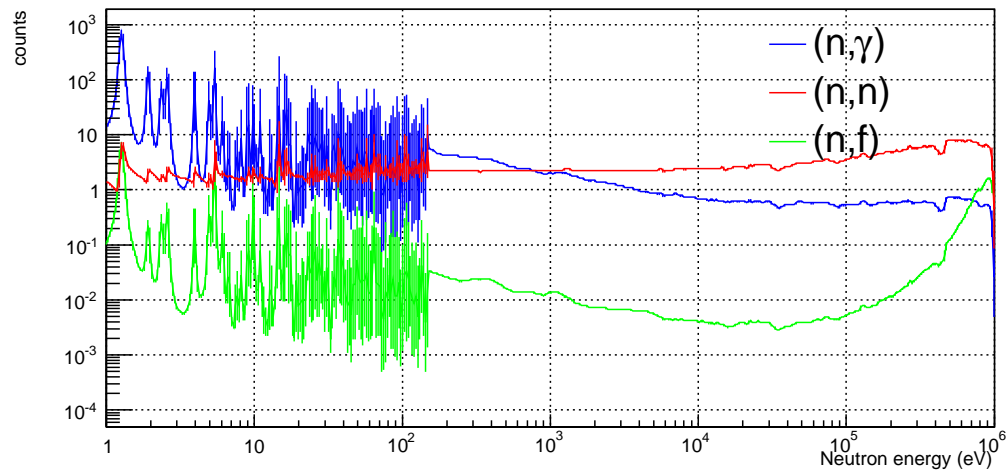
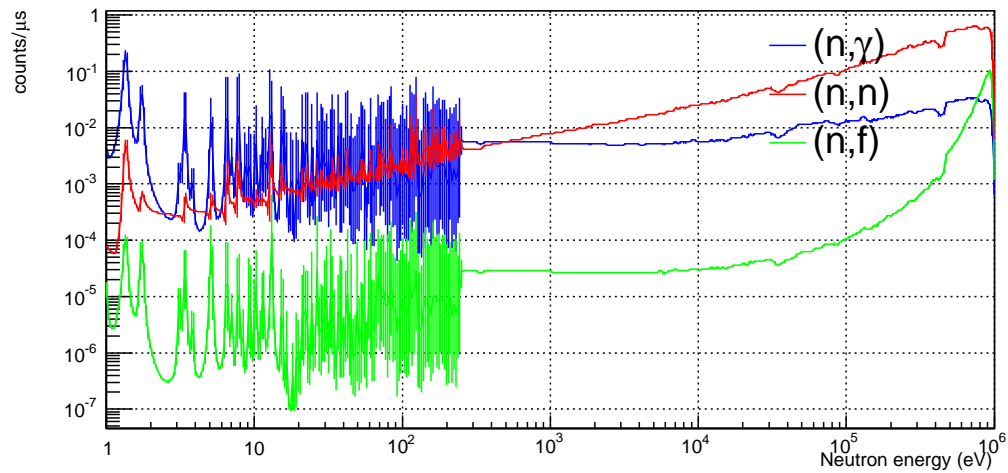


Figure 8. Top: $^{241}\text{Am}(n,\gamma)$ reaction rate (in red) in counts/μs compared to the elastic (blue) and neutron induced fission (green) rates for one PS pulse of $7 \cdot 10^{12}$ protons. Bottom: Number of reactions in one day of beam time. An 0.2 Hz PS average repetition rate was assumed.

REACTION RATE: Am243 sample of 10.00 mg R=0.5 cm



REACTIONS IN 1 DAYS (0.2 Hz 5000 bins/decade)

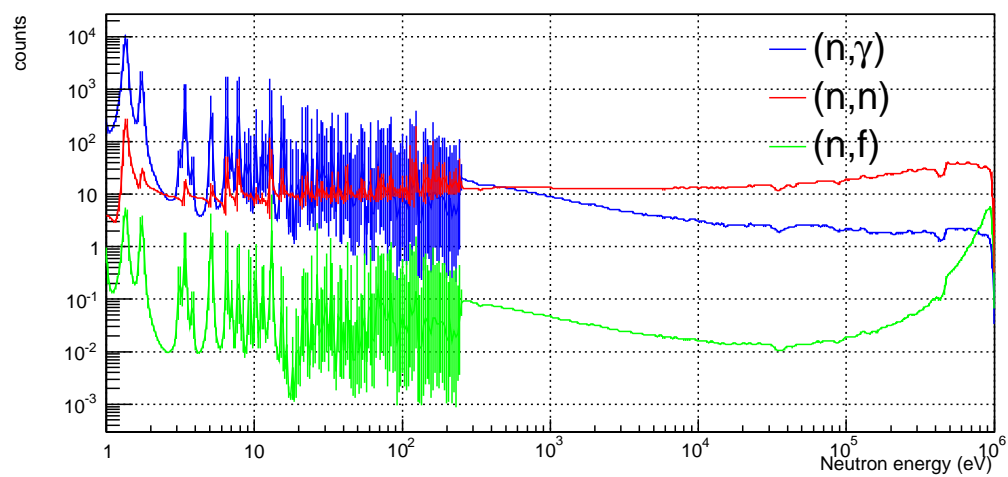


Figure 9. Top $^{243}\text{Am}(n,\gamma)$ reaction rate (in red) in counts/ μs compared to the elastic (blue) and neutron induced fission (green) rates for one PS pulse of $7 \cdot 10^{12}$ protons. Bottom: Number of reactions in one day of beam time. An 0.2 Hz PS average repetition rate was assumed.