NEUTRON CROSS-SECTION FOR P&T AND ADS AT THE N_TOF FACILITY AT CERN

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Abstract

An innovative neutron time of flight facility (n_TOF) has recently become operative at CERN. Neutrons in a wide energy range (1 eV<E_n<250 MeV) are generated by spallation of 20 GeV/c protons on a lead target. The instantaneously very intense neutron flux, low duty cycle, high resolution and low background make this facility unique for cross-section measurements relevant to Nuclear Astrophysics, fundamental Nuclear Physics, and particularly for Nuclear Technology, where most of the isotopes of interest are highly radioactive and available only in small samples. The n_TOF collaboration has proposed a vast experimental programme on capture, fission and (*n*,*xn*) reaction. [1] The main objective is the study of isotopes and reactions relevant to Accelerator-driven Systems (ADS) for nuclear waste transmutation and nuclear energy production. Studies of capture reactions relevant to Nuclear Astrophysics will also benefit from the innovative characteristics of the n_TOF neutron beam. This paper presents the present status and the first raw results.

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Introduction

From the middle of the nineties, when the new interest and tendencies for advance nuclear fuel cycle based on partitioning and Transmutation, P&T, started to be consolidated, it was clear that actinide transmutation will be based on their fission in nuclear systems (critical reactors or sub-critical ADS), in most cases of fast neutron energy spectra and using specific fuel compositions, much richer on high mass trans-uranium actinides. As a consequence, new isotopes, new energy regions and new reactions will play a major role on the behaviour of these transmutation nuclear systems and the corresponding cross-section data will be needed with better quality than available. In addition, the transmutation of Long Lived Fission Fragments has also been proposed using neutron absorption (mainly by radioactive capture) normally in the thermal and epithermal neutron energy spectra. Indeed the topic was discussed in several forums including the 5th NEA/OECD International Exchange Meeting, 5IEM, [1] and notably at CERN, resulting in the proposal [2] of construction of an specific installation, named n_TOF, to perform a large measurements campaign of the nuclear data required for advanced nuclear fuel cycles and the transmutation devices involved in those cycles. This proposal was accepted by CERN and financed by the European Commission within a shared cost action of the 5th Framework Programme, contract n_TOF_ADS FIKW-CT-2000-00107. The initial design [3] and expected characteristics of the n_TOF installation at CERN were indeed presented in the 6IEM of the NEA/OECD. [4] The facility initiated its commissioning in 1999 providing its first characteristics in 2000 [5] and after some shielding improvements during 2001 the present configuration and performance were reached in 2001. [6] The exploitation of the facility started in April 2002, and the actually measured n_TOF characteristics and the first raw results will be presented in this 7IEM.

n_TOF Programme motivations

The n_TOF_ADS has as objectives the measurements of cross-sections for two generic topics, the Thorium nuclear fuel cycle and the ADS for P&T. The main reactions in the Thorium cycle fuels are the neutron capture by ^{232}Th that, after fast radioactive decays breeds ^{233}U, and the fission of the ^{233}U that finally provides the neutron multiplication. ^{232}Th has a smaller nuclear mass than ^{238}U, so its neutron irradiation produce mainly uranium isotopes and much less trans-uranium actinides, although protactinium is also produced. This allows to generate a better nuclear waste in nuclear fuels with ^{232}Th matrix than in ^{238}U matrix fuels. For this reason, nuclear fuels with ^{232}Th matrix had been proposed both: for the transmutation devices for the present nuclear wastes, and for a new generation of “cleaner” nuclear energy production systems with a much smaller generation of trans-uranium actinides in the irradiated fuel. In both cases, fast and thermal neutron spectra had been proposed.

The list of new isotopes that play an important role in the thorium fuel cycle includes: ^{230,232}Th, ^{232,233,234,236}U and ^{231,232,233}Pa, as can be appreciated in the following reactions of this cycle:

^{232}Th + n → $\underline{^{233}\text{Th}}$ → $\underline{^{233}\text{Pa}}$→ $\underline{^{233}\text{U}}$, ^{233}U + n→ $\underline{^{234}\text{U}}$ … (7 neutrons before Pu)
^{233}U + n → Thermal and Fast fission, ^{232}Th + n → Fast fission
^{232}Th + n → 2n + ^{231}Th→ $\underline{^{231}\text{Pa}}$; ^{231}Pa + n → $\underline{^{232}\text{Pa}}$→ $\underline{^{232}\text{U}}$; ^{233}U + n → 2n + $\underline{^{232}\text{U}}$

The second generic field of nuclear data needs in n_TOF_ADS is related to the use of ADS for nuclear waste transmutation. Effective nuclear waste transmutation requires the use of special fuels with reduced intrinsic safety features (lower β_{eff}, lower Doppler effect, …) making more difficult to operate in critical condition. In addition new chemical forms of the proposed fuels and the need for a much extended fuel burn-ups require larger operational flexibility than what is available in critical reactors. The sub-criticality of the ADS and its external intense neutron source provides the required flexibility without loss of safety margins. However, to operate a sub-critical device requires a neutron spallation source that brings even faster neutrons (E_n>5MeV) into the system (although only few per cent) and consequently new reactions become more important, in particular (*n,xn*). In addition, the fact that lead and Pb/Bi alloys have often an important role in ADS, both as the target of the spallation source and as coolant of the nuclear core, make specially relevant the cross-sections of these elements.

The improvements required include:

- Measurements of the basics cross-sections (elastic, capture, fission, total and inelastic) on many high mass transuranic isotopes The most important energy range covers from thermal to about 1 MeV for capture an thermal to several (20) MeV for fission.
- Measurements of the basics cross-sections of medium and long-lived fission and spallation products.
- Measurements of fast neutron reactions, in the energy range from few keV to 20 MeV (capture, fission, elastic, inelastic, (*n,xn*), (n,α), etc), with usual actinides, fission products, coolant and structural materials.
- Evaluation of available experimental data to compute cross-sections and dissemination of the evaluated cross-sections results through the international agencies co-ordinating the distribution of these data.

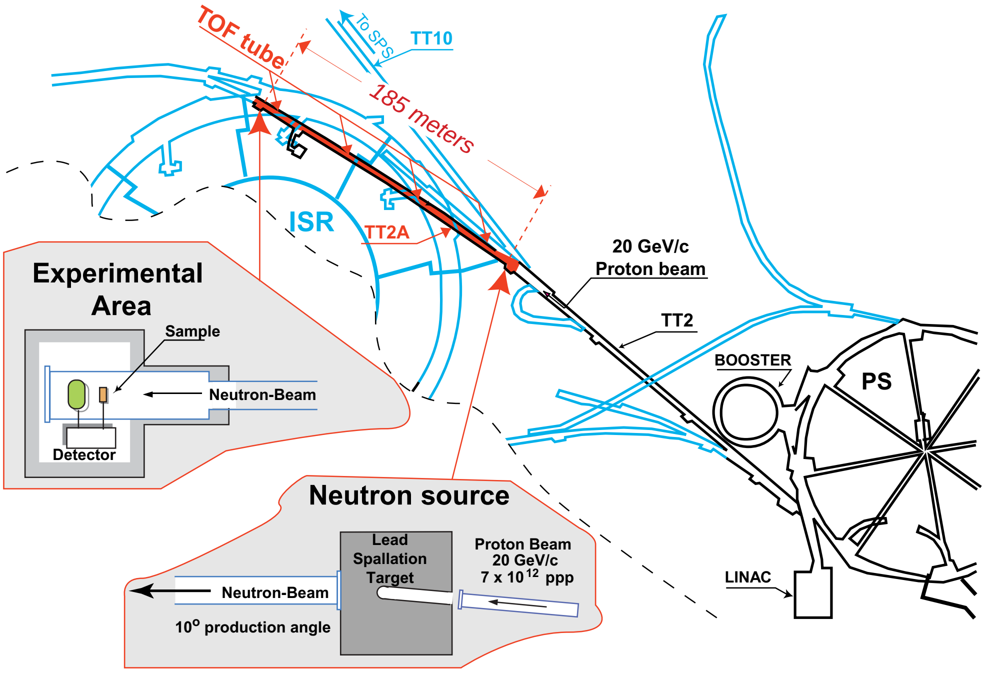
The resulting wish list of isotopes and reactions for nuclear waste transmutation should include, as actinides: ^{230}Th, ^{232}Th, ^{231}Pa, ^{232}Pa, ^{233}Pa, ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{244}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{242,243,244,245,246,247,248}Cm; as long lived fission fragments: ^{99}Tc, ^{129}I, ^{151}Sm, ^{79}Se, ^{126}Sn, ^{135}Cs, ^{93}Zr, ^{107}Pd; and as new coolants and structural materials: ^{206,207,208}Pb, ^{209}Bi.

On the other hand the main reactions are: *Capture*, that equilibrates fission in the neutron multiplication by parasite absorption of neutrons and generates the actinide nuclear wastes but at the same time breeds fissile isotopes and is the intermediate step to the transmutation fission for some actinides and the main transmutation reaction for LLFF; *Fission*, that produce the neutron multiplication and the reactor energy and is the final actinide transmutation reaction; *Inelastic scattering*, that shapes the neutron spectra in fast reactor cores (coolant, fuel and structural materials); and *(n,xn)*, that contribute to the shaping of the neutron spectrum and to the neutron multiplication near the spallation target but also generates the most dangerous nuclear waste in the Thorium cycle.

The n_TOF CERN facility

Most of the n_TOF-ADS measurements are and will be made in a new facility that has been set-up at CERN. The n_TOF facility at CERN, Figure 1**,** uses the existing CERN Proton Synchrotron, PS, to send 20 GeV/c protons on a lead target, surrounded by a 5 cm thick water layer. A fraction of the neutrons produced by spallation and moderated in the water is conducted by a 185 m long, wide vacuum pipe to an experimental area. In this area, the samples to be measured are exposed to those neutrons and the secondary particles, produced by the neutron interactions with the sample, are detected. An optimised set of neutron collimators and shielding elements had been introduced in the neutron path from the spallation target to the samples, to conform the neutron beam shape and position and to minimise the neutron and photon background in the experimental area. [5]

Figure 1. **General layout of the n_TOF facility**



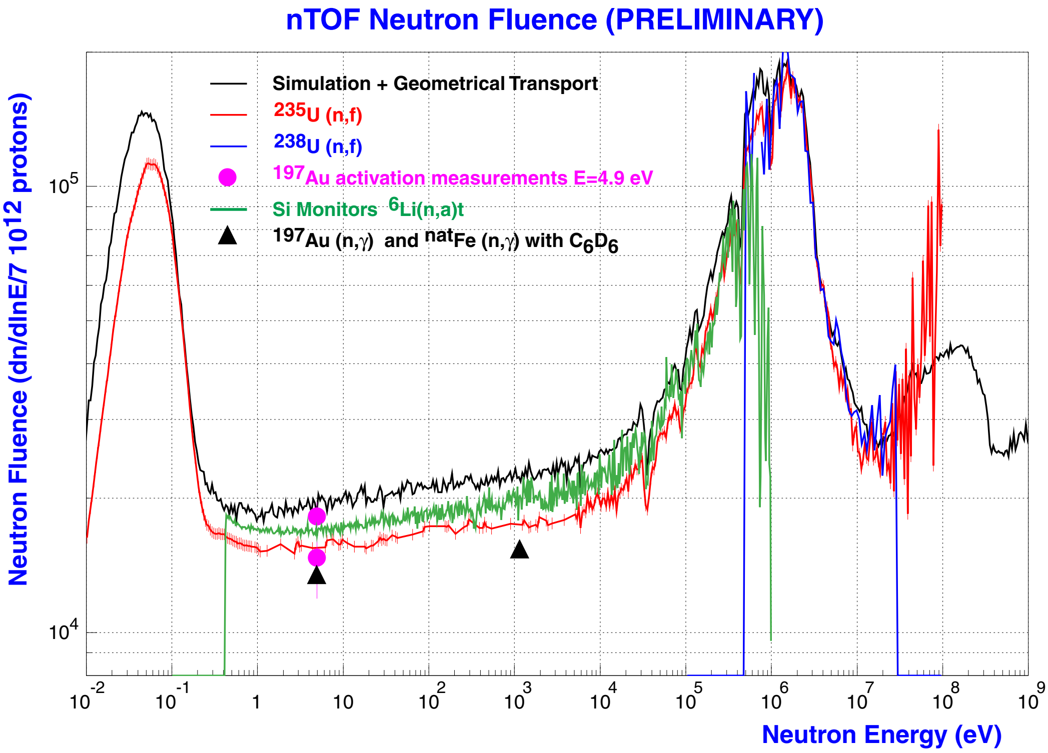
The time of flight, TOF, of the neutrons along the 185 m, that separate their production in the spallation target and the sample position, is measured with up to 1 ns precision. This should allow to determine the neutron energy with resolutions better than 10^{-3} for neutrons of energies up to several MeV. The identification of the reaction taking place in the sample is based on the measurement of the secondary products. The counting rate of different types of reactions allow to compute their cross-sections, using the previously determined neutron flux, taking both as functions of the neutron energy.

The PS is able to provide 7×10^{12} protons per pulse with an FWHM pulse duration of 5 ns, this together with the high proton energy provides the most instantaneously intense neutron source available for cross-section measurements. The maximum repetition rate is one shot every 1.2 s, although n_TOF will not take normally every pulse. These characteristics make n_TOF a world-wide unique facility for neutron cross-section in the intermediate energy region, from 1 keV to many MeV, and for radioactive, expensive or rare material samples. In addition the facility has the possibility of reaching up to several hundred MeV, although with decreasing intensity.

The neutron beam characteristics

The beam characterisation relies on 4 detectors: The Si detectors and the PPACs for the neutron energy spectrum determination and the micromegas and the CR-39-TED for the beam geometry. In addition a set of three BF_3 long counters located at the end of the neutron escape line provide fast integral neutron beam intensity and position monitoring.

Figure 2. **Measured neutron flux at the sample position**



The low energy part of the neutron energy spectrum is measured by four Silicon detectors viewing a ^{6}LiF sample placed at the entrance of the experimental area. Both the α and the tritons produced in the ^{6}Li(n,α)^{3}H reaction are detected by the silicons. The high energy part (and also the low energy part) of the neutron energy spectrum is measured by fission detectors observing reference samples of ^{235}U, ^{238}U and one sample of ^{209}Bi. ^{197}Au activation foils measurements and observed reaction rates of the ^{197}Au capture on its main resonances provide additional reference points. Figure 2 shows the comparisons of the different measurements of the neutron flux in the 2002 campaign and the Monte Carlo predictions, for the energy range covering from 10^{-2} to 10^{8} eV.

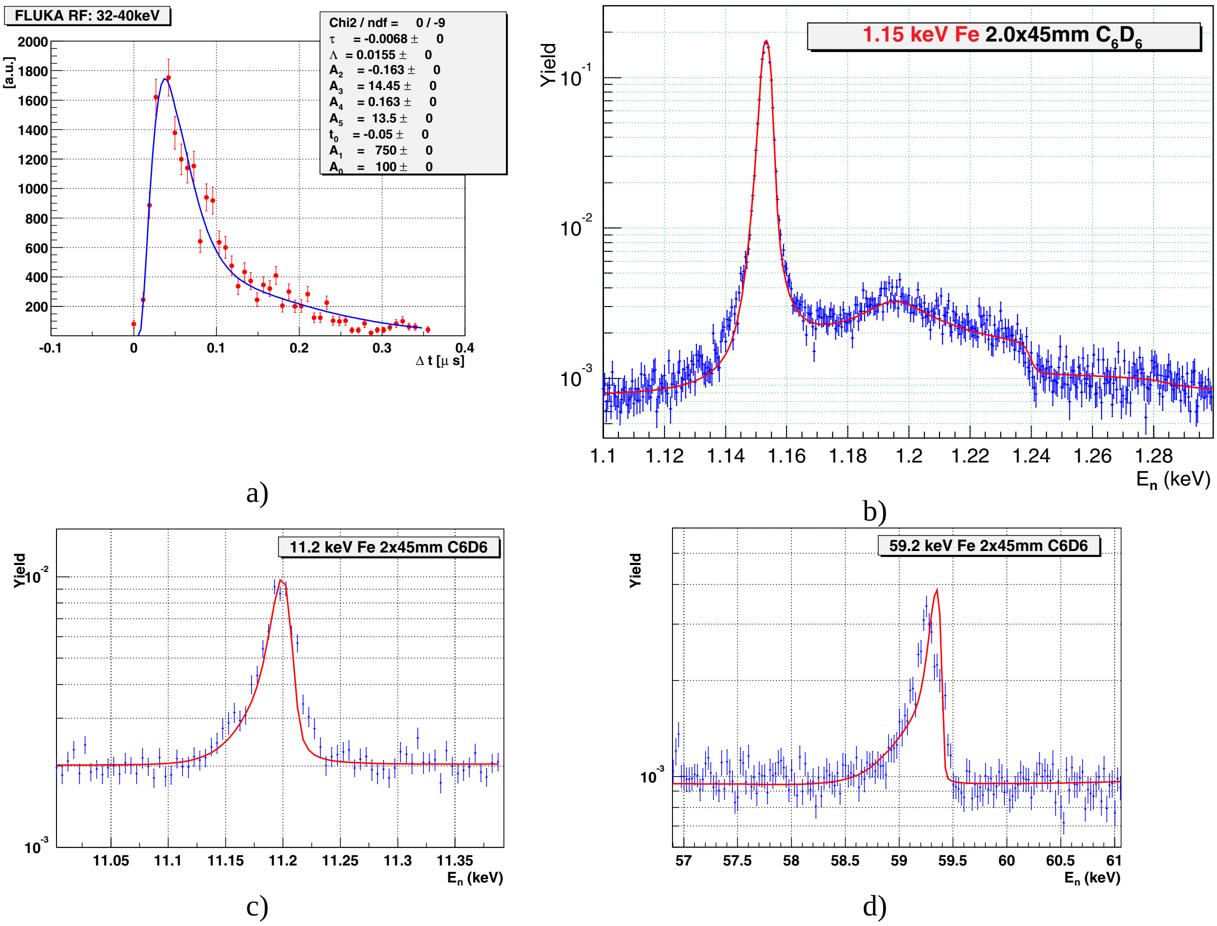
The beam geometry, position and profile, is determined for a very wide energy range by the micromegas detector. At low energies this detector uses a ^{6}Li converter using the ^{6}Li(n,α)^{3}H reaction to produce charged particles from the neutrons. The fine segmentation of the detector allows to determine the projection of the beam profile on its charge collecting strips. The recoils of nuclei included in the detector gas, particularly H, extend the operation of the detector to much higher neutron energies. In addition, the neutron beam profile, at energies in the range of few eV, is measured by CR-39 detectors observing the products of the ^{10}B(n,α)^{7}Li reaction from a thin boron foil covered by a 1mm thick Cd foil. Also the PPACs fission detector observing the reference samples provide beam profile information.

Capture measurements in the 2002 campaign

Most of the 2002 measurement campaign has been devoted to the characterisation of the installation properties and the cross-section measurements using capture detectors. For the 2002 campaign the detectors used to detect the secondary particles produced after the interaction of the neutron beam with the samples are low neutron sensitivity C_6D_6 detectors. Two versions of these detectors had been used. First, four commercial BICRON detectors modified to reduce the neutron sensitivity and improve the response time. Second two C_6D_6 detectors specifically designed for n_TOF, with containers made of carbon fibre in order to further reduce the neutron sensitivity. Both sets of detectors had been commissioned during the years 2001 and 2002, and the corresponding weighting functions for the different configurations of detectors and samples had been obtained by the Monte Carlo simulation validated by the 2001 commissioning measurements.

Reference samples of ^{197}Au and ^{56}Fe in combination with these C_6D_6 detectors have been used to explore the energy resolution and the characteristic time response function of the n_TOF installation. Figure 3 shows on one hand the predicted time response function (a) and on the other hand the measured capture reaction rate on three well isolated ^{56}Fe resonances (b-d), and the corresponding Monte Carlo predictions. The figure shows a reasonable understanding of the time response function and a resolution better that 10^{-3} up to 1MeV.

Figure 3. **Monte Carlo simulation of the time response function and experimental capture reaction rate on well isolated ^{56}Fe resonances**



These measurements in combination with measurements of Pb and C samples and in absence of sample, as well as the measurements with different upstream “black” filters have allowed to estimate the beam induced backgrounds. The results show a substantial reduction in the beam related background after the completion of the tunnel and lead target shielding. The ambient background induced by the beam is so low that for all the measurements of the 2002 campaign, the beam induced background is generated by the beam interaction with the samples. In addition geometrical rearrangement of the detector set-ups had allowed to further reduce the photon background for the capture measurements.

After beam characterisation and the commissioning of the detectors and experimental area, the measurements campaign had finally started on 2002. The proton beam allocation for year 2002 has allowed to measure the capture cross-section of ^{151}Sm, ^{209}Bi, ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb and ^{232}Th. Figures 4 to 7 show the draft reaction rates profiles as functions of the neutron energy. It should be noted that the ^{151}Sm measurement is the first time-of-flight capture measurement for this isotope. The Bi and Pb isotopes cover all stable isotopes of the proposed elements for most ADS spallation target and of the coolant of several ADS and critical fast reactor designs. Finally the ^{232}Th measurement show the interest of the high intensity and low repetition cycle of n_TOF, reducing the intrinsic sample radioactive background to very small levels even in the valleys between the first Th resonances. Its should also be noted that the resonances structures of ^{232}Th can be resolved up to nearly 10 KeV. All these measurements are on the process of analysis to extract the resonance parameters and the average cross-section in the unresolved resonance region. The energy range of the evaluation of this cross-section is expected to extend up to 1 MeV. Finally it should also be noted that the resonance structures of the 204 and 207 lead isotopes do not exactly match the information on the standard nuclear databases indicating the need of a detailed revision of the content of these databases.

Figure 4. **^{151}Sm preliminary observed capture reaction rate**

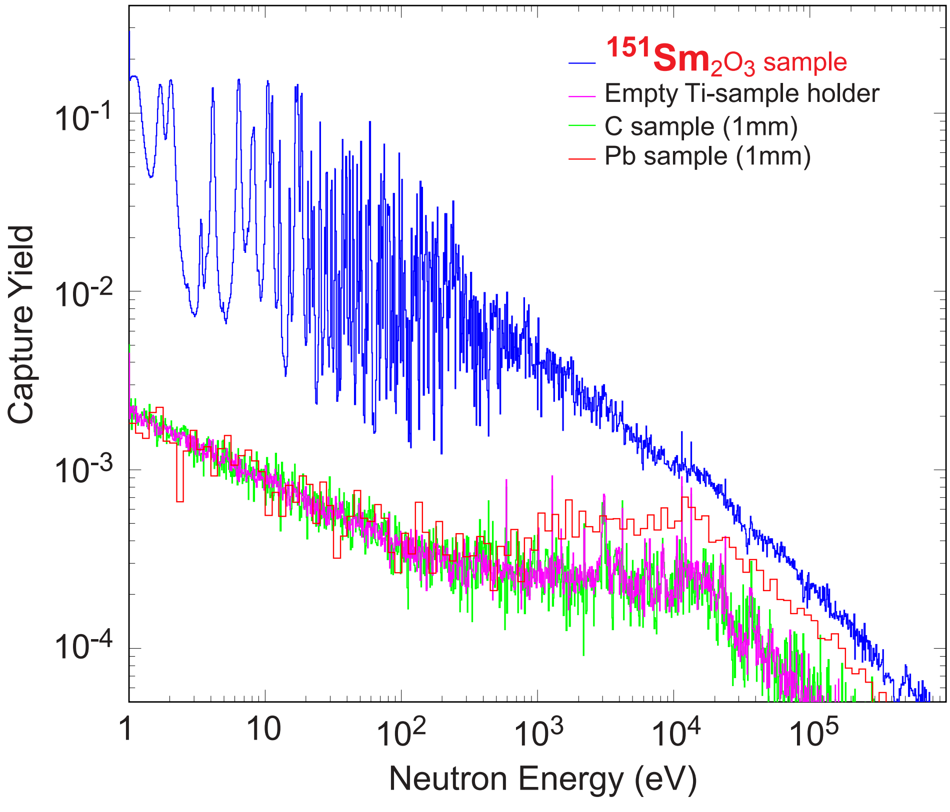


Figure 5. **^{232}Th preliminary observed capture reaction rate**

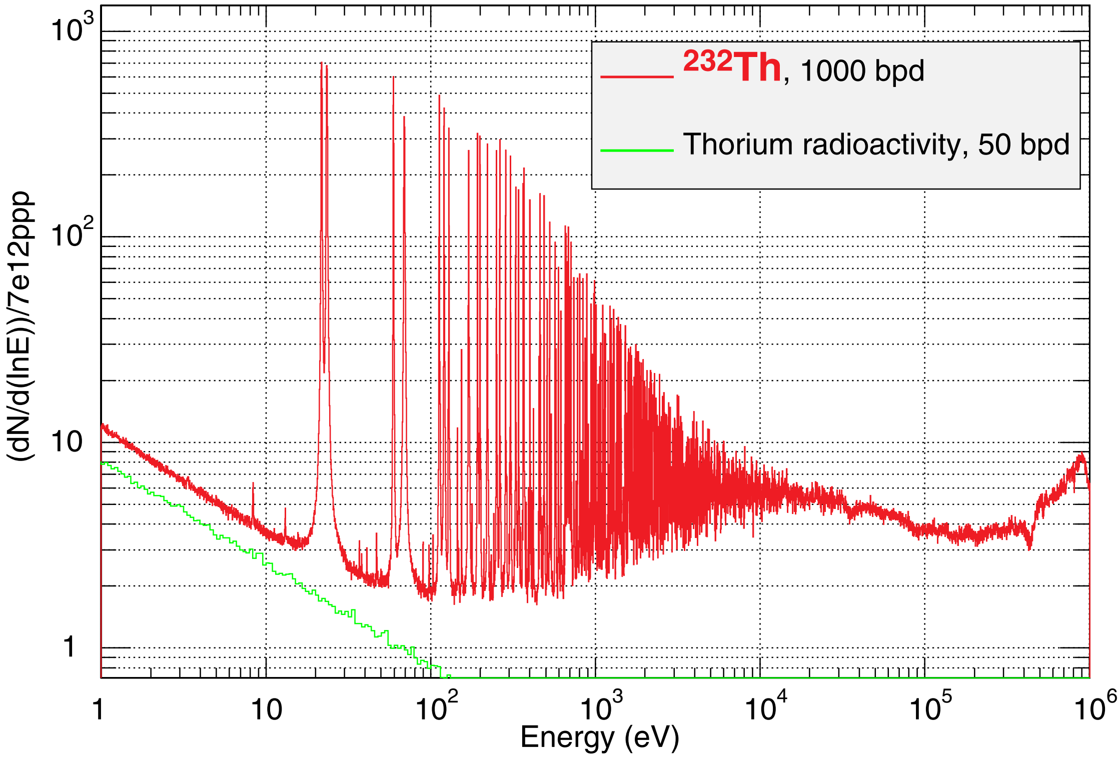


Figure 6. **^{209}Bi preliminary observed capture reaction rate**

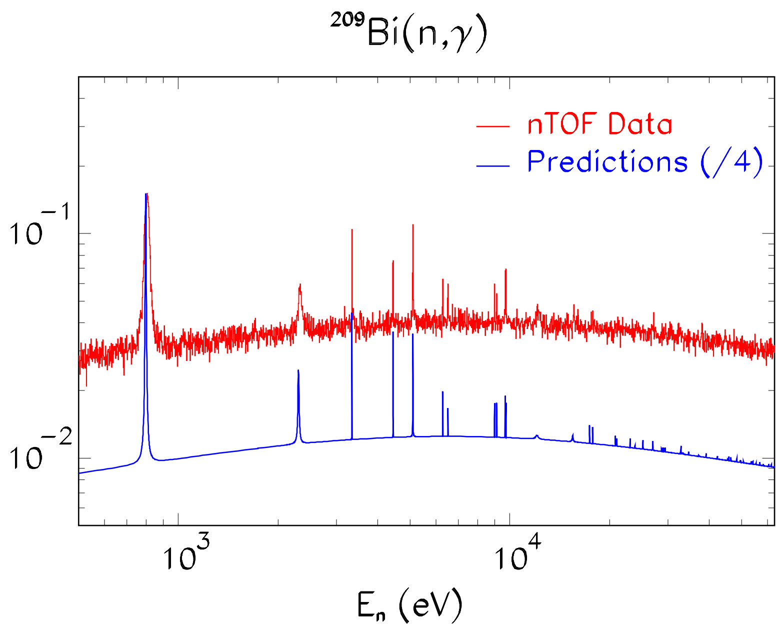
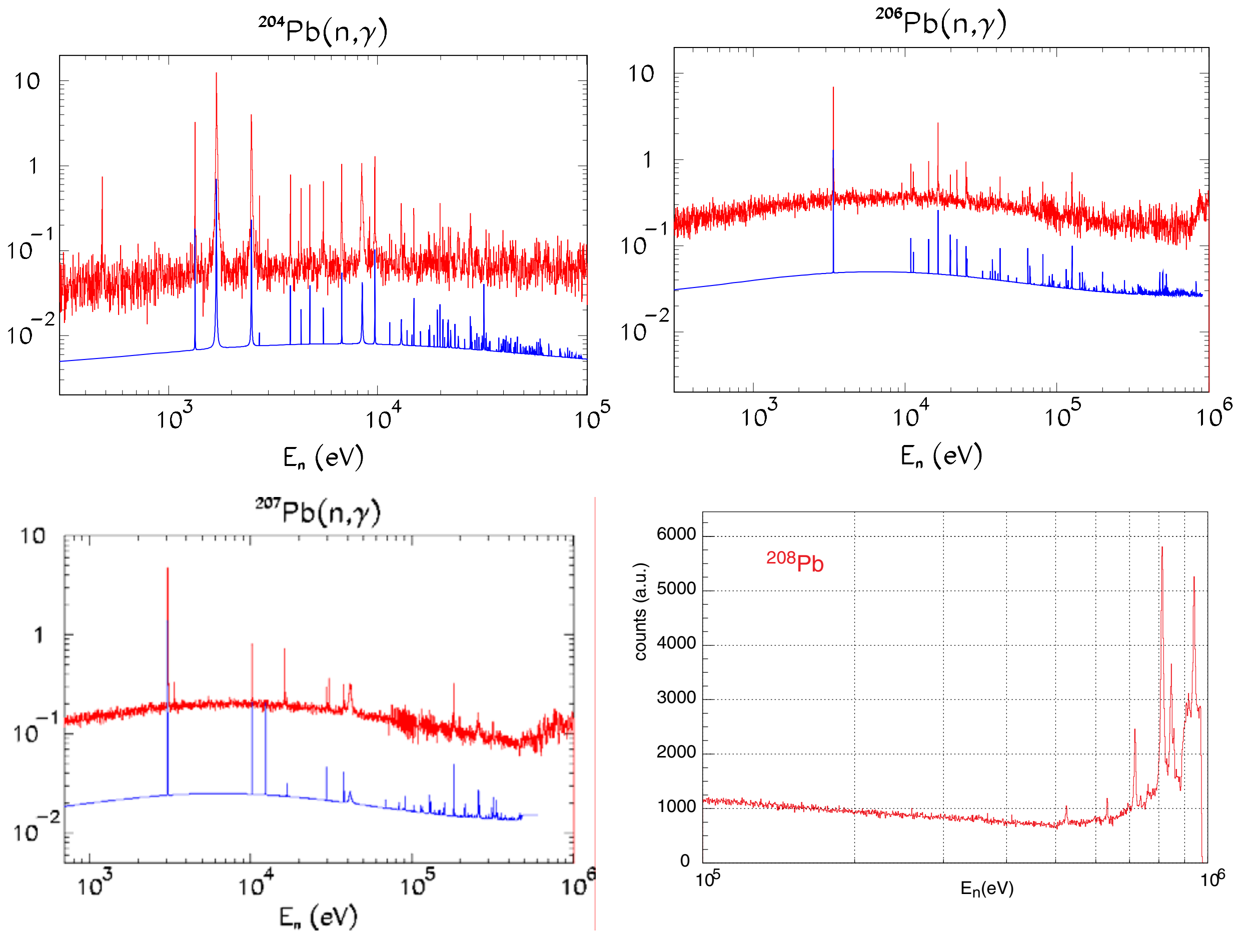


Figure 7. **^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb preliminary observed capture reaction rate**



Programme of neutron cross-section measurements at n_TOF

A large number of measurements will be made at the n_TOF facility. The list of measurements programmed within the n_TOF_ADS EU contract includes:

Capture cross-sections *of Th-cycle isotopes*: Including measurements of ^{232}Th, ^{231}Pa, ^{233}U, ^{234}U and ^{236}U in the range from 1eV to ~1 MeV. Some of these isotopes will be measured first with C_6D_6 during 2002 and 2003, although some measurements will be repeated with the 4π calorimeter at 2004.

Capture cross-sections *of transuranic isotopes*: ^{240}Pu, ^{242}Pu, ^{241}Am, ^{243}Am and ^{245}Cm will be measured at n_TOF mainly on 2004 with the 4π calorimeter. ^{237}Np has been measured by the n_TOF collaboration at IRMM at Geel. The aimed energy range of measurements will be from 1eV to approx. 1 MeV.

Capture cross-sections *of Long Lived Fission Fragments*: ^{151}Sm has already been measured by n_TOF at CERN with C_6D_6 detectors during 2002. ^{151}Sm is a medium lived fission fragment (90y) with additional interest for astrophysics. On the other hand, ^{99}Tc (2.1×10^5y) and ^{129}I (1.6×10^7y) are the LLFF with highest impact on the long term radiotoxicity inventory and the two with highest possibilities of reaching technical feasibility of transmutation. They are/will be measured by n_TOF at IRMM at Geel. ^{79}Se (6.5×10^4y) could be measured at n_TOF depending on sample and proton availability.

Capture cross-section*s of Coolants and Structural materials*: As already mention, lead and Pb/Bi alloys have an important role in ADS, both as the target of the spallation source and as coolant of the nuclear core. Po produced by Pb and Bi activation is a source of concern from the point of view of radioactive wastes and radiation protection in ADS operation. Furthermore, Pb and Bi isotopes will contribute to the neutron balance and spectrum definition by capture, elastic and inelastic scattering and by the (*n,xn*) reactions. As a consequence, ^{204,206,207,208}Pb and ^{209}Bi had already been measured by n_TOF during 2002.

Fission cross-section*s of Th-cycle and transuranic isotopes*: The isotopes considered are ^{237}Np, ^{239}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, ^{245}Cm, ^{232}Th, ^{231}Pa, ^{233}U, ^{234}U and ^{236}U (plus ^{235}U and ^{238}U – reference standard isotopes). The main objective will be to cover the energy range from 1eV to 20 MeV, but the high energy limit will be extended as much as allowed by statistics. Fission measurements are very time consuming and so 2 types of complementary detectors will be used, and both foresee the possibility to use simultaneously multiple samples. These measurements are foreseen for the year 2003.

Total cross-section*s*: Performed by transmission, most probably in the IRMM facilities. The presently proposed isotopes are ^{237}Np, ^{129}I, ^{239}Pu and ^{240}Pu.

(n,xn) cross-section*s*: Performed in two ways, by detection of prompt photons in TOF installations at CERN and IRMM and by activation methods in several facilities at Europe providing monoenergetic neutrons. Adding together both types of installations, measurements are proposed for ^{237}Np, ^{232}Th, ^{231}Pa, ^{239}Pu, ^{241}Pu, ^{241}Am, ^{243}Am, ^{233}U, and ^{207}Pb. The (*n,xn*) measurements based on the prompt γ identification are described in another paper presented to this 7IEM. [7]

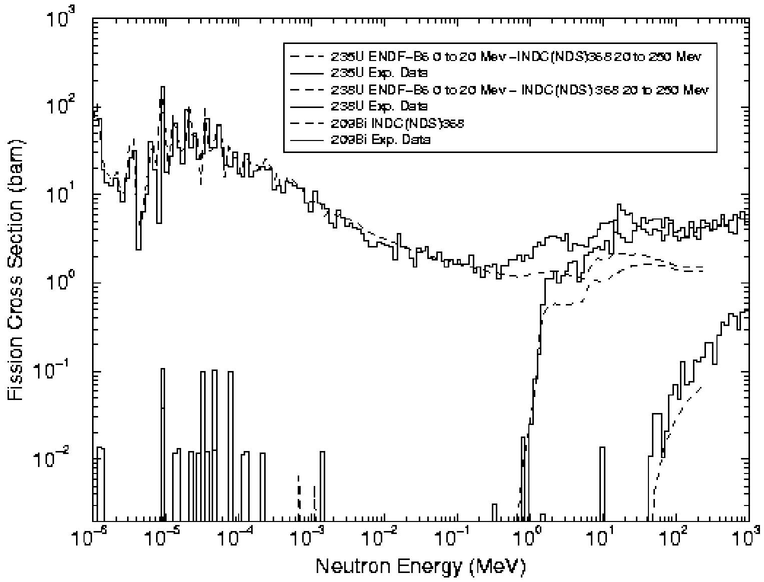
The n_TOF Facility will stop its operation (as the rest of the CERN accelerators) during 2005, but it is scheduled to continue operation during 2006 and afterwards, extending the cross-section measurements campaign.

n_TOF detectors for future measurements

During the first two years of the n_TOF experiment, capture measurements will be made with the previously described C_6D_6 detectors. At 2004, capture cross-sections will be performed with a high performance, 4π total absorption γ calorimeter, inspired on the design of existing BaF_2 FZK calorimeter presently under construction. This detector will be mainly devoted to capture measurements of fissionable isotopes (actinides) and highly radioactive samples. The main advantages with respect to C6D6 detectors will be: the possibility to separate capture from fission events; the discrimination between capture events and other sources of γ's (radioactive decay, background, etc..) by measuring the total cascade energy, a better control of systematics (no need of weighting function or similar corrections and in addition the tails of the total energy absorption allow to estimate the efficiency and contamination); and a higher absolute event efficiency where we expect to improve by a factor 5 with respect to the present C_6D_6 detectors.

Two types of Fission detectors hade been built for the experiments at n_TOF. On one hand, a set of up to 11 Parallel Plates Avalanche Chambers, PPACs, and on the other an ionisation chamber (Fast Induction Detector) with capability of 16 double sided samples. Both detectors are designed to operate in the energy range from thermal energies up to 250 MeV. The performance of the fission detectors and the capabilities of the n_TOF facility for measuring fission cross-sections been tested during 2001 and 2002 tested with samples of standard cross-sections, namely ^{235}U, ^{238}U and ^{209}Bi. The counting rate was analysed, contrary to what will be done normally, to extract the cross-section from a flux (a rough flux estimation from Monte Carlo simulations). Figure 8 shows the comparison of the obtained apparent cross-sections from the PPACS with the databases. This figure shows that with the same setup it is possible to measure the fission cross-section for energies ranging from 1eV to several hundred MeV (nearly 9 orders of magnitude). It also shows that the data reproduce correctly the fission thresholds and the characteristic energies for opening of the (n,nf) and (n,2nf) reactions in the U isotopes. In general the absolute value of the cross-section (actually of the effective fluence) was correctly reproduced even by the rough Monte Carlo, within a factor 2. A more detailed simulation and analysis in progress, should substantially improve this agreement. The last month of the 2002 campaign has indeed been devoted to test measurements of the ^{232}Th and ^{234}U fission cross-sections.

Figure 8. **^{235}U, ^{238}U and ^{209}Bi observed fission reaction rate**



Two additional key elements of the n_TOF experiments the DAQ and the Simulation. The Flash-ADC based DAQ, has allowed to operate with a largely reduced dead-time, allowing to use piled-up signals, and to extend the energy measurements well within the prompt particles flash of the capture detectors. This system however generates very large data volumes that require specific data transmition, storage and processing developed by n_TOF. The simulation that has allowed to design the collimation and shielding systems and to understand the sources of background up to more than 7 orders of magnitude in flux respect to the beam, has been able to describe the neutron and photon interactions from thermal up to nearly 20 GeV of kinetic energy and very high energetic charged particle, inside a very long and rather complex geometry.

Summary

Following the new needs of nuclear data for Partitioning and Transmutation advanced fuel cycles, a new installation for time of flight cross-section measurements has been set-up at CERN. After an one year delay, and an exhaustive beam, shielding and detector commissioning, the facility was ready to initiate its long cross-section measurement campaign at the spring of 2002. The measured beam characteristics, including the beam intensity, energy profile, shape, resolution and time response functions have reached their design values and the beam induced background has been reduced to the beam-sample interaction. The resulting beam instantaneous intensity, energy range and background levels are the best available world-wide.

The extraordinary performances of the n_TOF facility had been already exploited during 2002 by measuring the capture cross-sections of ^{151}Sm, ^{209}Bi, ^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb and ^{232}Th that at present are being analysed and will be later on evaluated. Several of these measurements will improve the existing time-of-flight direct data available up to now.

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