Neutron Capture Cross Section Measurements at n_TOF of ^{237}Np, ^{240}Pu and ^{243}Am for the Transmutation of Nuclear Waste

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Abstract. Accurate and reliable neutron capture cross section data for actinides are necessary for the proper design, safety regulation and precise performance assessment of transmutation devices such as Fast Critical Reactors or Accelerator Driven Systems. In particular, the neutron capture cross sections of ^{237}Np, ^{240}Pu and ^{243}Am play a key role in the design and optimization of strategies for the Transmutation of Nuclear Waste. The listed cross sections have been measured in 2004 at n_TOF [1] with a high accuracy due to a combination of features unique in the world: high instantaneous neutron fluence and excellent energy resolution of the n_TOF facility, innovative Data Acquisition System based on flash ADCs and the use of a high performance BaF$_2$ Total Absorption Calorimeter as a detection device.

Keywords: ^{237}Np,^{240}Pu, ^{243}Am, neutron capture cross section, nuclear waste, transmutation, total absorption calorimeter
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INTRODUCTION

Nuclear waste transmutation has been proposed as a way to reduce substantially (in a factor of 1/100 or more) the radiotoxicity inventory of the long lived component of the nuclear waste, mainly the trans-uranium actinides. Actinide transmutation is proposed to take place by fission in nuclear systems like critical reactors or subcritical Accelerator Driven Systems (ADS). The detailed engineering designs, safety evaluations and the detailed performance assessment of dedicated transmutation ADS and critical reactors (i.e. with fuels highly enriched in transuranic isotopes) require more precise and complete basic nuclear data [2].

EXPERIMENT

Isotopically enriched targets of minor actinides are available only in small amounts, typically of the order of 1 - 100 mg. Even then, the activities involved present major difficulties related both to the experimental techniques and the radioprotection aspects. The targets of ^{237}Np (43.3 mg, 1.29 MBq), ^{240}Pu (51.2 mg, 458 MBq) and ^{243}Am (10 mg, 75 MBq) measured at n_TOF [1] were all assembled in the same way: the radioactive material was sandwiched between two thin Al layers (total mass < 75 mg) and canned inside a 0.35 mm thick Ti canning with ISO 2919 certification (requested by the safety regulations at CERN). Their isotopic purity was determined by γ-ray spectrometry and is >98% in all cases.

In capture measurements at n_TOF [3] with a 4 cm diameter neutron beam spot, the instantaneous fluence amounts to 10^5 neutrons/cm^2/pulse for neutron energies between 0.1 eV and 20 GeV. This is one of the key features for measuring the (n,γ) cross section of low mass and highly radioactive isotopes with a good signal to noise ratio. During the measurements, the repetition rate of the PS was on average 3 - 4 proton pulses for a PS supercycle of 16.4 s. In addition, the n_TOF facility provides an excellent energy

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resolution of 10^{-3} to 10^{-4} at a 185 m flight path between the Pb spallation target and the counting station.

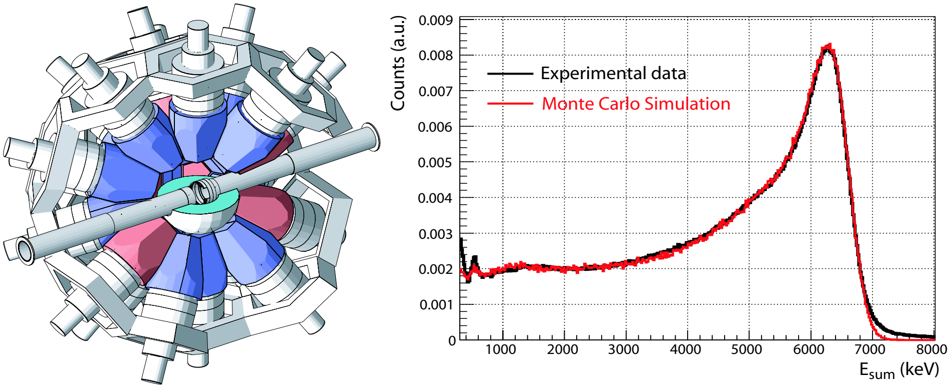

FIGURE 1. Left: view of the Total Absorption Calorimeter as it is implemented in the code GEANT4[9] used for the MC simulations. Right: experimental (black) and Monte Carlo simulated (red) energy deposition spectra in the TAC for capture events in the ^{197}Au(n,γ) reaction at 4.9 eV.

The capture cross section measurements at n_TOF have been done relative to the standard capture cross section ^{197}Au(n,γ). For this reason, several independent monitors were used permanently for a proper normalization between the ^{197}Au(n,γ) and main measurements.

The Data Acquisition System (DAQ)[4] used in the measurements consists in 54 channels of high performance flash ADCs [5]. Each channel has 8 Mbytes memory and was operated at a sampling rate of 500 Msamples/s, thus allowing to record the full detector history for neutron energy ranges between 0.3 eV and 20 GeV. After zero suppression and data formatting, the raw data are sent to CERN's massive storage facility CASTOR [6] via several Gigabit links. In parallel, especially designed pulse shape analysis routines run on a PC farm and extract from the digitized detector signals the necessary information for the data analysis. The n_TOF DAQ offers unique features such as an extremely low dead time (< 10 ns), good signal analysis and pileup discrimination among others, resulting in an excellent mechanism for controlling all kind of systematic uncertainties associated to the detector's behavior.

The neutron capture detection system consists in a segmented Total Absorption Calorimeter made of 40 BaF_2 crystals with ^{10}B loaded carbon fibre capsules and placed at 185 m flight path from the spallation source. The TAC has a nearly 100% detection efficiency for electromagnetic cascades (i.e. capture events) and a good energy resolution (14% at 662 keV and 6% at 6.1 MeV). The radioactive targets are placed at the geometric center of the TAC and surrounded by a $C_{12}H_{20}0_4(^6Li)_2$ neutron absorber placed inside the inner hole of the TAC. The neutron absorber and the ^{10}B loaded carbon fibre capsules reduce the sensitivity of the detector to the scattered neutrons, do not reduce the capture detection efficiency (even though they lower the total absorption efficiency) and also help in attenuating the low energy component (10 - 100 keV) of the sample

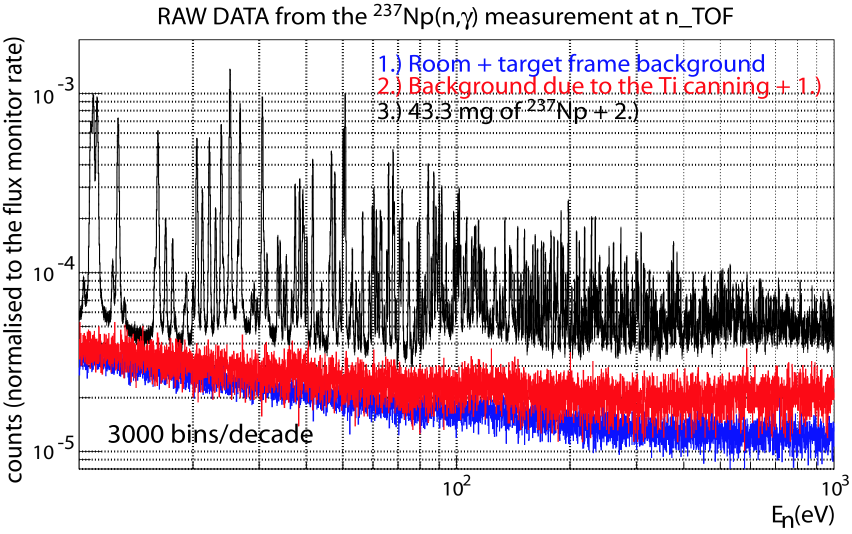

FIGURE 2. Raw data from the ^{237}Np (n,γ) measurement in the resolved resonance region between 10 eV and 1 keV. Below, the background due to the Ti canning and the empty TAC (without any sample in the beam).

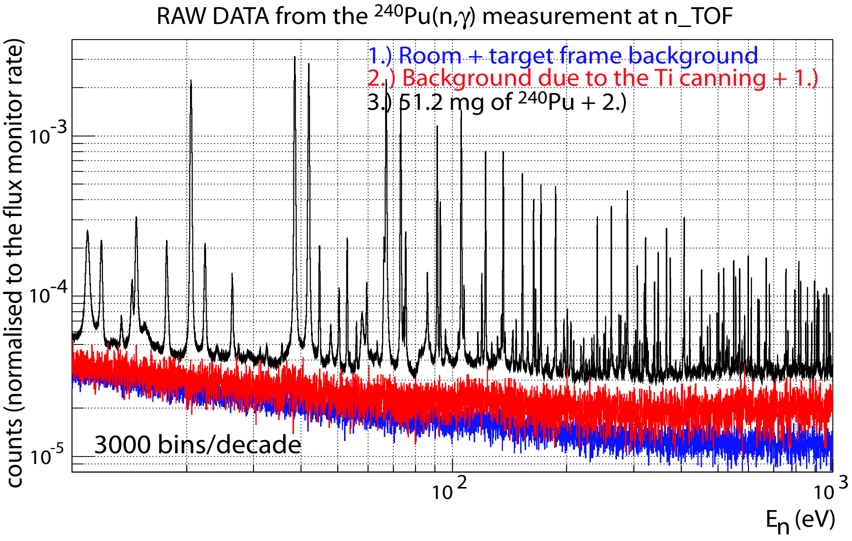

FIGURE 3. Raw data from the ^{240}Pu (n,γ) measurement in the resolved resonance region between 10 eV and 1 keV. Below, the background due to the Ti canning and the empty TAC (without any sample in the beam).

radioactivity. For the measurement of ^{243}Am, a cylindrical Pb shielding of 1 mm thickness around the target and outside the beam was necessary for suppressing the strong γ-activity with energies about 100 - 200 keV.

Fig. 1 shows a view of the experimental setup, as it is implemented in the code GEANT4[9] used for the detailed MC simulations of the TAC. The performance of the TAC has been investigated both experimentally (with standard calibration sources and the reference ^{197}Au(n,γ) cross section) and by Monte Carlo simulations [7] [8]. Furthermore, all sources of background have been measured and are being simulated for performing the background corrections necessary for an accurate capture cross section analysis.

RESULTS

The capture data for ^{237}Np, ^{240}Pu and ^{243}Am have been taken in summer 2004. A large number of resonances have been observed in the resolved resonance region with good statistics in both the raw data for ^{237}Np and ^{240}Pu, as it is shown in fig. 2 and 3, respectively. Furthermore, the level of the background there is fairly low, thus allowing to conclude already that the extraction of accurate resonance parameters is possible. The cross section analysis in the unresolved resonance region will require a more detailed study and subtraction of the background, dominated mainly by the effect of the Ti canning of the samples. However, clear capture signals are visible in the TAC energy deposition spectra up to a few tens of keV.

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