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## Neutron cross section measurements at n-TOF for ADS related studies

P F Mastinu<sup>18</sup>, U Abbondanno<sup>20</sup>, G Aerts<sup>7</sup>, H Álvarez<sup>35</sup>, F Alvarez-Velarde<sup>31</sup>, S Andriamonje<sup>7</sup>, J Andrzejewski<sup>26</sup>, P Assimakopoulos<sup>16</sup>, L Audouin<sup>12</sup>, G Badurek<sup>1</sup>, N Bustreo<sup>18</sup>, P aumann<sup>10</sup>, F Be vá<sup>6</sup>, E Berthoumieux<sup>7</sup>, F Calviño<sup>34</sup>, D Cano-Ott<sup>31</sup>, R Capote<sup>3,36</sup>, A Carrillo de Albornoz<sup>27</sup>, P Cennini<sup>37</sup>, V Chepel<sup>28</sup>, E Chiaveri<sup>37</sup>, N Colonna<sup>19</sup>, G Cortes<sup>33</sup>, A Couture<sup>41</sup>, J Cox<sup>41</sup>, M Dahlfors<sup>37</sup>, S David<sup>9</sup>, I Dillmann<sup>12</sup>, R Dolfini<sup>23</sup>, C Domingo-Pardo<sup>32</sup>, W Dridi<sup>7</sup>, I Duran<sup>35</sup>, C Eleftheriadis<sup>13</sup>, M Embid-Segura<sup>31</sup>, L Ferrant<sup>9</sup>, A Ferrari<sup>37</sup>, R Ferreira-Marques<sup>28</sup>, L itzpatrick<sup>37</sup>, H Fraiss-Kölbl<sup>4</sup>, K Fujii<sup>20</sup>, W Furman<sup>30</sup>, C Guerrero<sup>31</sup>, I Goncalves<sup>28</sup>, R Gallino<sup>22</sup>, E Gonzalez-Romero<sup>31</sup>, A Goverdovski<sup>29</sup>, F Gramegna<sup>18</sup>, E Griesmayer<sup>4</sup>, F Gunsing<sup>7</sup>, B Haas<sup>8</sup>, R Haight<sup>39</sup>, M Heil<sup>12</sup>, A Herrera-Martinez<sup>37</sup>, M Igashira<sup>25</sup>, S Isaev<sup>9</sup>, E Jericha<sup>1</sup>, Y Kadi<sup>37</sup>, F Käppeler<sup>12</sup>, D Karamanis<sup>16</sup>, D Karadimos<sup>16</sup>, M Kerveno<sup>10</sup>, V Ketlerov<sup>29,37</sup>, P Koehler<sup>40</sup>, V Konovalov<sup>30,37</sup>, E Kossionides<sup>15</sup>, M Krti ka<sup>6</sup>, C Lamboudis<sup>13</sup>, H Leeb<sup>1</sup>, A Lindote<sup>28</sup>, I Lopes<sup>28</sup>, M Lozano<sup>36</sup>, S Lukic<sup>10</sup>, J Marganiec<sup>26</sup>, L Marques<sup>27</sup>, S Marrone<sup>19</sup>, C Massimi<sup>21</sup>, A Mengoni<sup>3,37</sup>, P M Milazzo<sup>20</sup>, C Moreau<sup>20</sup>, M Mosconi<sup>12</sup>, F Neves<sup>28</sup>, H Oberhummer<sup>1</sup>, S O'Brien<sup>41</sup>, M Oshima<sup>24</sup>, J Pancin<sup>7</sup>, C Papachristodoulou<sup>16</sup>, C Papadopoulos<sup>14</sup>, C Paradela<sup>35</sup>, N Patronis<sup>16</sup>, A Pavlik<sup>2</sup>, P Pavlopoulos<sup>11</sup>, L Perrot<sup>7</sup>, R Plag<sup>12</sup>, A Plompen<sup>5</sup>, A Plukis<sup>7</sup>, A Poch<sup>33</sup>, C Pretel<sup>33</sup>, J Quesada<sup>36</sup>, T Rauscher<sup>38</sup>, R Reifarth<sup>39</sup>, M Rosetti<sup>17</sup>, C Rubbia<sup>23</sup>, G Rudolf<sup>10</sup>, P Rullhusen<sup>5</sup>, J Salgado<sup>27</sup>, L Sarchiapone<sup>37</sup>, I Savvidis<sup>13</sup>, C Stephan<sup>9</sup>, G Tagliente<sup>19</sup>, J L Tain<sup>32</sup>, L Tassan-Got<sup>9</sup>, L Tavora<sup>27</sup>, R Terlizzi<sup>19</sup>, G Vannini<sup>21</sup>, P Vaz<sup>27</sup>, A Ventura<sup>17</sup>, D Villamarin<sup>31</sup>, M C Vincente<sup>31</sup>, V Vlachoudis<sup>37</sup>, R Vlastou<sup>14</sup>, F Voss<sup>12</sup>, S Walter<sup>12</sup>, H Wendler<sup>37</sup>, M Wiescher<sup>41</sup>, and K Wisshak<sup>12</sup>

<sup>1</sup>Atominstut der Österreichischen Universitäten, Technische Universität Wien, Austria, <sup>2</sup>Institut für Isotopenforschung und Kernphysik, Universität Wien, Austria, <sup>3</sup>International Atomic Energy Agency (IAEA), Nuclear Data Section, Vienna, Austria, <sup>4</sup>Fachhochschule Wiener Neustadt, Wiener Neustadt, Austria, <sup>5</sup>CEC-JRC-IRMM, Geel, Belgium, <sup>6</sup>Charles University, Prague, Czech Republic, <sup>7</sup>CEA/Saclay - DSM, if-sur-Yvette, France, <sup>8</sup>Centre National de la Recherche Scientifique/IN2P3 - CENBG, Bordeaux, France, <sup>9</sup>Centre National de la Recherche Scientifique/IN2P3 - IPN, Orsay, France, <sup>10</sup>Centre National de la Recherche Scientifique/IN2P3 - IReS, Strasbourg, France, <sup>11</sup>Pôle Universitaire Léonard de Vinci, Paris La Défense, France, <sup>12</sup>Forschungszentrum Karlsruhe GmbH (FZK), Institut für Kernphysik, Germany, <sup>13</sup>Aristotle University of Thessaloniki, Greece, <sup>14</sup>National Technical University of Athens, Greece, <sup>15</sup>NCSR, Athens, Greece, <sup>16</sup>University of Ioannina, Greece, <sup>17</sup>ENEA, Bologna, Italy, <sup>18</sup>Istituto Nazionale di Fisica Nucleare (INFN), Laboratori Nazionali di Legnaro, Italy, <sup>19</sup>Istituto Nazionale di Fisica Nucleare, Bari, Italy, <sup>20</sup>Istituto Nazionale di Fisica Nucleare, Trieste, Italy, <sup>21</sup>Dipartimento di Fisica, Università di Bologna, and

Sezione INFN di Bologna, Italy, <sup>22</sup>Dipartimento di Fisica, Università di Torino and Sezione INFN di Torino, Italy, <sup>23</sup>Università degli Studi Pavia, Pavia, Italy, <sup>24</sup>Japan Atomic Energy Research Institute, Tokai-mura, Japan, <sup>25</sup>Tokyo Institute of Technology, Tokyo, Japan, <sup>26</sup>University of Lodz, Lodz, Poland <sup>27</sup>Instituto Tecnológico e Nuclear(ITN), Lisbon, Portugal, <sup>28</sup>LIP - Coimbra & Departamento de Física da Universidade de Coimbra, Portugal, <sup>29</sup>Institute of Physics and Power Engineering, Kaluga region, Obninsk, Russia, <sup>30</sup>Joint Institute for Nuclear Research, Frank Laboratory of Neutron Physics, Dubna, Russia, <sup>31</sup>Centro de Investigaciones Energeticas Medioambientales y Tecnológicas, Madrid, Spain, <sup>32</sup>Istituto de Física Corpuscular, CSIC-Universidad de Valencia, Spain, <sup>33</sup>Universitat Politècnica de Catalunya, Barcelona, Spain, <sup>34</sup>Universidad Politécnica de Madrid, Spain, <sup>35</sup>Universidade de Santiago de Compostela, Spain, <sup>36</sup>Universidad de Sevilla, Spain, <sup>37</sup>CERN, Geneva, Switzerland, <sup>38</sup>Department of Physics and Astronomy - University of Basel, Basel, Switzerland, <sup>39</sup>Los Alamos National Laboratory, New Mexico, USA, <sup>40</sup>Oak Ridge National Laboratory, Physics Division, Oak Ridge, USA, <sup>41</sup>University of Notre Dame, Notre Dame, USA

Mastinu@Inl.infn.it

**Abstract.** A neutron Time-of-Flight facility (n\_TOF) is available at CERN since 2001. The innovative features of the neutron beam, in particular the high instantaneous flux, the wide energy range, the high resolution and the low background, make this facility unique for measurements of neutron induced reactions relevant to the field of Emerging Nuclear Technologies, as well as to Nuclear Astrophysics and Fundamental Nuclear Physics. The scientific motivations that have led to the construction of this new facility are here presented. The main characteristics of the n\_TOF neutron beam are described, together with the features of the experimental apparatus used for cross-section measurements. The main results of the first measurement campaigns are presented. Preliminary results of capture cross-section measurements of minor actinides, important to ADS project for nuclear waste transmutation, are finally discussed.

## 1. Introduction

Recently, the problems related to energy provision are emerging as a global challenge, involving health, environmental, economical and social issues. At present nuclear power now accounts for 4.5% of total world's energy production/consumption, but already at this level, the accumulated spent fuel inventory, at the yearly production rate of 8,000 ton, will reach by 2020 about 200,000 ton. Assuming an increase of the contribution from conventional nuclear power plants to about 30% of the increased world's power consumption by 2050 (corresponding to an increase of 2.3%/yr), the yearly waste production would be as high as 100,000 ton/y. Such a large amount poses severe problems from the economic as well as from the environmental point of view. In particular the latter has generated public concern because the radiotoxicity of the produced waste is very high and the secondary produced actinides have a decay lifetime extending up to millions of years. A different solution has therefore to be found. It has been proposed that the amount and radiotoxicity of the nuclear waste can be reduced by using the Thorium fuel cycle, since this would strongly reduce the production of actinides like Am, Cm and Pu. Beside the Thorium fuel cycle process, an alternative/complementary solution would be the incineration of long-lived fission fragments and minor actinides by neutron capture and neutron-induced fission [1,2]. In this scenario, the long-lived radioisotopes would be transmuted in a new kind of reactor, the Acceleration Driven System (ADS), into short-lived ones, so reducing by orders of magnitude the necessary repository time. An ADS is a sub-critical reactor, in which the chain reaction

is sustained by an external neutron beam produced by means of the new high-current proton accelerator.

A reliable design and development of ADS requires high-accuracy cross-section data for neutron capture, neutron-induced fission and inelastic scattering on several isotopes, mainly radioactive ones. Capture and fission data are needed for fertile and fissile isotopes involved in the Th-cycle, such as  $^{232}\text{Th}$ ,  $^{231}\text{Pa}$ ,  $^{233}\text{U}$ ,  $^{234}\text{U}$  and  $^{236}\text{U}$ . Similarly, the design of ADS for nuclear waste incineration requires data on capture, fission and (n,xn) cross-sections for transuranic isotopes, in particular  $^{237}\text{Np}$ ,  $^{238,240,241}\text{Pu}$ ,  $^{241,243}\text{Am}$  and  $^{244,245}\text{Cm}$ , while the incineration scheme of long-lived fission products requires accurate data on capture reactions for  $^{79}\text{Se}$ ,  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{135}\text{Cs}$ ,  $^{151}\text{Sm}$ , etc. Also reaction, cross-sections for structural materials being considered as neutron-production target or as coolant, are still far from being accurately known, especially at high incident neutron energy.

Many of the aforementioned needs can be consistently addressed at the neutron time-of-flight, facility, n\_TOF, now operational at CERN, Geneva [3]. The innovative characteristics of the n\_TOF neutron beam, in particular the very high instantaneous neutron flux, make it unique in the world for measurements of cross-sections of radioactive or low-mass samples, relevant to many fields of applied and fundamental physics. Thanks to its unique features, the new facility is expected to produce also significant advances in the knowledge of capture processes involved in Stellar Nucleosynthesis, allowing to address a number of still open questions, mostly regarding radioactive species, such as the branching-point isotopes, or isotopes with very low cross-sections.

A large international collaboration, constituted by 150 researchers from 40 Institutes, mostly European, has constructed high performance experimental apparatus and an innovative data acquisition system, and is currently carrying out an intense experimental and theoretical program. The facility, experimental apparatus and first preliminary results at n\_TOF are here presented.

## 2. The n\_TOF facility

The n\_TOF spallation neutron source is based on a 20 GeV/c proton beam, impinging on a  $40 \times 80 \times 80 \text{ cm}^3$  lead solid target surrounded by 5 cm water, which act as a coolant and moderator. The moderation effect strongly enhances the neutron flux at low energy and gives an almost flat isoenergetic distribution of neutrons in the range from 1 eV up to few MeV. Two collimators placed around 140 and 180 meters from the spallation target are used to shape the neutron beam to the desired dimension, while a sweeping magnet deflects charged particles away from the beam. Several concrete and iron shieldings ensure a very low background. The experimental area is located at 187.5 meters from the spallation target. At that distance, an energy resolution as good as  $dE/E=10^{-4}$  is reached for neutron energies up to 100 KeV. The neutron fluence in the experimental area is of the order of  $10^5 \text{ n/cm}^2/\text{pulse}$  [4].

The main features of the neutron beam are the extremely high intensity, white energy spectrum in the range 1 eV-250 MeV, low duty cycle, high resolution and low background. In particular, the high instantaneous flux allows the measurement of radioactive isotopes, nearly impossible to measure at other existing facilities.

## 3. n-TOF experimental apparatus

Several high-performance apparatus have been set up by the n\_TOF Collaboration for the neutron capture and neutron-induced fission cross-section measurements. Furthermore, to match the characteristics of the neutron beam and of the detection systems, an innovative data acquisition system (DAQ), purely based on fast Flash ADC, has been set up. A description of the different detectors and of their characteristics is given below.

### 3.1. The Silicon neutron flux monitor

Information about the neutron flux is extremely important in cross section measurements. The n\_TOF neutron beam intensity is monitored with a low-background system based on Silicon detectors

[5]. Four detectors,  $4 \times 6 \text{ cm}^2$  in area and  $300 \text{ }\mu\text{m}$  in thickness, are placed outside the beam around a thin Mylar foil with a deposit of  ${}^6\text{Li}$ , placed in the beam. The measurement of the neutron flux relies on the detection of secondary particles (tritons and alpha with energies of around  $2 \text{ MeV}$ ) produced after capture of low-energy neutrons. To minimize the background, the system is mounted in a carbon-fibre vacuum scattering chamber, at the entrance of the experimental area. As neutron converter, a pure  ${}^6\text{Li}$  sample of  $200 \text{ }\mu\text{g}/\text{cm}^2$  in thickness and  $6 \text{ cm}$  in diameter is used, deposited on a substrate of  $3 \text{ }\mu\text{m}$  mylar. To avoid  ${}^6\text{Li}$  from reacting with air, the deposit is sandwiched between two evaporated layers of C, less than  $10 \text{ }\mu\text{g}/\text{cm}^2$  in thickness each. The Silicon Monitor is typically used for relative normalization of the neutron fluence. During the commissioning of the facility, the system has also been used, together with other detectors (in particular a fission chamber from PTB), to determine the absolute neutron flux. In this case, it was necessary to introduce corrections for the geometric efficiency and, above  $1 \text{ keV}$ , for the anisotropy in the angular distribution of emitted tritons.

### 3.2. $\text{C}_6\text{D}_6$ liquid scintillator detectors for capture studies

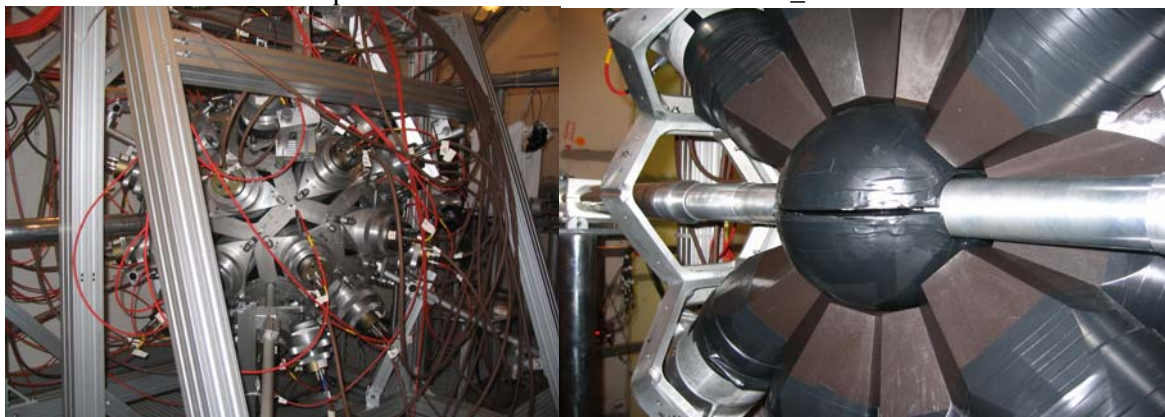
In the first two years of operation of the n\_TOF facility, neutron capture experiments were performed using  $\text{C}_6\text{D}_6$   $\gamma$ -ray detectors, in combination with the pulse height weighting technique, a method that makes the detection efficiency independent of the  $\gamma$ -ray cascade of the  $(n,\gamma)$  reaction. The  $\text{C}_6\text{D}_6$  detectors have a low  $\gamma$ -ray efficiency over the range of interest, between  $0.1\text{-}10 \text{ MeV}$ , but have very low sensitivity to scattered neutrons as compared to other gamma-ray detectors. The neutron sensitivity of the detectors was further reduced by using a very thin carbon fiber container glued directly onto the photomultiplier tube [6]. In this way, all parts which are not absolutely essential were removed, including the window of the cell and the complete photomultiplier housing. The neutron sensitivity of the optimized detectors is of the order of  $10^{-6}$ . Two  $\text{C}_6\text{D}_6$  detectors have been used in the measurement campaigns of 2002 and 2003. The detectors were mounted in direct contact with the beam line, in close geometry with respect to the sample. A carbon-fibre sample changer, specifically made for n\_TOF in order to minimize the background related to neutrons elastically scattered from the samples, was used in the measurements.

The use of  $\text{C}_6\text{D}_6$  detectors relies on the PHWT, which consist in a software modification of the detector  $\gamma$ -ray response in order to obtain an efficiency for the detection of a capture event independent of the details of the cascade (in particular, the multiplicity). In the PHWT, the response distribution  $R_j$  is modified through the introduction of a set of weighting factors  $W_i$  determined from the condition that the efficiency for a single  $\gamma$ -ray is proportional to its energy  $E_\gamma$ , so that the detection efficiency of the capture cascade is independent of the actual cascade path, provided that the probability of simultaneous detection of more than one cascade  $\gamma$ -ray is negligible. The appropriate weighting function for each specific  $(n,\gamma)$  set-up can only be obtained by Monte-Carlo simulations of the detector response, performed with GEANT (3.21 and 4) and MCNP-X. The accuracy of the method and of the simulations was verified in the first measurement campaign with standard cross-sections. An uncertainty of less than  $2 \%$  is typically associated with the PHWT [7].

### 3.3. The $4\pi$ Total Absorption Calorimeter (TAC)

Although the  $\text{C}_6\text{D}_6$  capture setup is characterized by a very low neutron sensitivity, it is not possible with such detectors to distinguish whether the detected  $\gamma$  rays originate from the  $(n,\gamma)$  reaction, the radioactive background or from competing reaction channels like fission or inelastic scattering. The best signature for the identification of neutron capture events in  $(n,\gamma)$  cross section measurements via the TOF technique is the total energy of the  $\gamma$ -cascade by which the product nucleus de-excites to its ground state. Hence, accurate measurements of  $(n,\gamma)$  cross sections can be made by using a detector that operates as a calorimeter with good energy resolution. In the  $\gamma$ -spectrum of such a detector, all capture events would fall in a line at the neutron binding energy (typically between  $5$  and  $10 \text{ MeV}$ ), well separated from the  $\gamma$ -ray backgrounds that are inevitable in neutron experiments, and independent of the multiplicity of the  $\gamma$ -ray cascade.

A new Total Absorption Calorimeter (TAC) has been constructed and used for neutron capture cross section measurements at n\_TOF. The device is particularly suited for the measurement of minor actinides which are subject of nuclear waste transmutation. The TAC consists of 12 pentagonal and 28 hexagonal shaped  $\text{BaF}_2$  crystals. Both types of crystals are 15 cm long and are placed at 11 cm from the geometric centre of the TAC. Such a configuration covers 95.2% of the total solid angle. Figure 1 shows an external and internal picture of the calorimeter mounted at n\_TOF.



**Figure 1:** Photograph of the external and internal view of the  $\text{BaF}_2$  calorimeter. The inner ball of neutron absorbing material is visible in the right panel, as well as the Carbon-fibre capsules around the crystals

To minimize the TAC neutron sensitivity, a neutron absorber was placed inside the calorimeter, and each crystal was encapsulated in a  $^{10}\text{B}$ -loaded carbon fibre capsule. The neutron absorber consisted of a spherical shell of 10 cm external radius, made of  $(^6\text{LiOOC}(\text{CH}_2)_{10}\text{COO}^6\text{Li})$ , placed inside the TAC and surrounding the target. Its composition was suitably chosen in order to moderate (through scattering on H) and capture (on  $^6\text{Li}$ ) neutrons scattered by the targets that otherwise could be captured in the detector and surrounding materials, leading to background signals that can be misidentified with real capture events. For some highly radioactive samples, (such as  $^{243}\text{Am}$ ), a 1mm thick, 9 cm long and 3,2 cm radius cylindrical lead shielding had to be placed around the target holder, in order to absorb the low energy gamma rays.

#### 3.4. Parallel Plate Avalanche Counters (PPACs)

The measurement of neutron-induced fission cross-sections are performed at n\_TOF with two different fission fragment detectors. Position sensitive Parallel Plate Avalanche Counters (PPAC) have been used for isotopes related to the Thorium-Uranium fuel cycle (energy release) and for minor actinides relevant to nuclear waste incineration. A PPAC consists of two thin parallel stretched foils with a very low gas pressure in between. A high electric field between electrodes ensures a small time spread. Time determination is obtained on a central anode made of 1.5 micron thick Mylar foil aluminized on both sides. Cathodes placed on each side of the anode are made from 1.5 micron thick Mylar foils, with deposited aluminum strips 2 mm wide, for two-dimensional position information.

A series of  $20 \times 20 \text{ cm}^2$  area PPACs are mounted in stacks, perpendicularly to the beam, with different samples placed between two of them. The distance between the sample and each PPAC is 15 mm. A sample, typically made of material spread as a layer of  $300 \mu\text{g}/\text{cm}^2$  and with few cm in diameter. It is placed perpendicular to the neutron beam. The target is deposited on a thin substrate to allow the detection of the two fission fragments in coincidence. Materials with standard fission cross sections, like  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{209}\text{Bi}$ , are permanently used both for normalization of the relative fission yields as well as for flux monitoring. A total of 10 samples, including those used as flux monitors and standards, can be measured simultaneously.

### 3.5. Fission Ionization Chamber (FIC)

The measurement of fission reactions with a counter based on the absorption of the fission fragments in gas has also been foreseen at the n\_TOF Facility at CERN. After the good results achieved during the second n\_TOF Commissioning [4] by using a Parallel Plate Induction Fission Chamber from PTB, it was decided to build a dedicated counter based on the same principle but optimized for the facility. This kind of detector is simple and easy to operate. Apart from the windows, made with Kapton foils 125  $\mu\text{m}$  in thickness, all metallic parts are made of aluminum or aluminum alloy. The detector is filled with gas (90% Ar, 10%  $\text{CF}_4$ ) at 0.72 bar. No gas circulation is needed. The targets are stacked so that total of 16 targets and 18 electrodes could be mounted together leaving a distance of 5 mm between layers. There is also another stack of electrodes (3 targets and 4 electrodes) mounted perpendicularly to the beam axis, that is equipped with reference isotopes to evaluate the background produced by scattered neutrons. In both stacks one place is reserved for a “dummy” target (support without deposit). Samples of few mg total mass are typically used in the measurements. Samples of  $^{238}\text{U}$  and  $^{235}\text{U}$  are mounted at all times, to serve as a reference and as a monitor of the neutron flux.

### 3.6. The Data Acquisition System

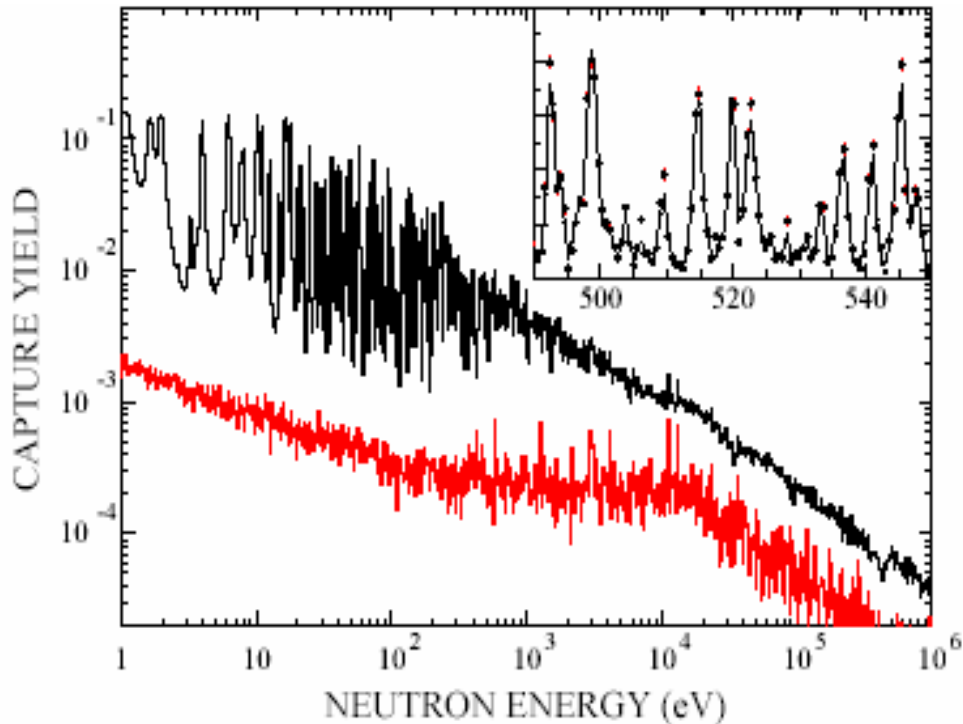
An innovative, general purpose Data Acquisition System (DAQ) has been developed, to match the specific characteristics of the n\_TOF neutron beam and the performances of the experimental apparatus [8]. A new system was necessary in particular to resolve the expected large number of pile-up events, a direct consequence of the high instantaneous neutron flux. The n\_TOF DAQ was designed to accommodate the various detector requirements without any loss of efficiency or dead time, in a user-friendly software environment. The system is based on fast Flash ADC, which sample and store the full analogue waveform of the detector signals for each channel and beam burst, with on-line zero-suppression. Additional data reduction is achieved by recording the data in compressed “tar” file format. The digitizer modules used are the DC270 and DC240 8 bit resolution models manufactured by Acqiris, with sampling rates up to 1 and 2 Gsamples per second (Gs/s) respectively, bandwidths of 250 and 500 MHz and 8 Mbytes on-board memory. The detector signal characteristics define the sampling rate and the dynamic range to be used. Particularly demanding was the configuration of the n\_TOF DAQ for the measurements of capture cross-sections with the calorimeter, with as many as 49 flash ADC channels employed simultaneously. The DAQ is typically triggered by the signal produced by the proton beam impinging on the lead target. The data are transferred via cPCI/PCI adapters into the readout PCs, which compress the data and transfer them to the CERN Central Data Recording facility for storage.

Once the raw data were written on tape, it can be read by the Data Processing Software. The relevant signal parameters such as time, energy and other pertinent information (like particle identification) are extracted and written on Data Summary Files for final analysis. The amount of raw data is very large, if the Total Absorption Calorimeter is used: i.e. total of 138.4 TBytes have been accumulated as raw data and 2.7 TBytes as DSTs (average reduction factor of 50), which represents a data rate of 2.5 Tbytes/day over a period of 2 months.

## 4. Experimental campaign

Starting in 2002, a vast experimental program has been undertaken by the n\_TOF Collaboration on capture and fission cross-sections. Table 1 reports a complete list of the measured isotopes, many of which are radioactive. Some of the data were collected with the aim of improving the comprehension of Stellar Nucleosynthesis, while other were required for the design of an ADS for energy release and nuclear waste incineration. An example of the quality of the n\_TOF data is represented by the results of  $^{151}\text{Sm}(n,\gamma)$  reaction, the first measurements performed at the n\_TOF facility. This reaction is of interest for Nuclear Astrophysics since the radioactive  $^{151}\text{Sm}$  represents a branching point in the s-process path, with the branching ratio (capture versus  $\beta$ -decay) sensitive to the temperature at which the s-process nucleosynthesis takes place. An accurate determination of the neutron capture cross-

section is therefore fundamental to obtain information on thermodynamics of AGB stars. The natural radioactivity of the sample, which has hindered this measurement at other neutron facilities, did not constitute a problem at n\_TOF, thanks to the high instantaneous neutron flux, as clearly seen in the figure. The results allowed to extract for the first time an experimental value of the Maxwellian Averaged Cross-Section.



**Figure 2:** Data from the  $^{151}\text{Sm}(n,\gamma)$  reaction. The black histogram show the capture data, while the red one represents the overall background. In the upper-right insert, some of the resonances are shown (symbols) together with the results of a resonance analysis.

Another important measurement performed at n\_TOF was the capture reaction of  $^{232}\text{Th}$ . This isotope is crucial for the investigation of the alternative nuclear fuel cycle based on Th/U. The earlier measurements of the  $^{232}\text{Th}(n,\gamma)$  cross-section presented severe discrepancies, of up to 30%, an uncertainty much too large for one of the key elements of the fuel. At n\_TOF, the measurement was performed with the same setup used for the  $^{151}\text{Sm}(n,\gamma)$  reaction, based on  $\text{C}_6\text{D}_6$  liquid scintillators. The much more accurate results of n\_TOF, characterized by an uncertainty of around 5%, will allow to generate more reliable nuclear data library for  $^{232}\text{Th}$ , as requested for the implementation of the thorium fuel cycle in existing or in innovative nuclear power devices.



Capture	Fission
<sup>151</sup> Sm, <sup>204,206,207,208</sup> Pb, <sup>209</sup> Bi, <sup>232</sup> Th	<sup>233,234,236</sup> U
<sup>139</sup> La, <sup>24,25,26</sup> Mg	<sup>232</sup> Th
<sup>90,91,92,93,94,96</sup> Zr	<sup>237</sup> Np
<sup>186,187,188</sup> Os	<sup>241,243</sup> Am
<sup>233,234</sup> U, <sup>237</sup> Np, <sup>240</sup> Pu, <sup>243</sup> Am	<sup>245</sup> Cm
<sup>197</sup> Au, <sup>nat</sup> C, <sup>nat</sup> Pb	<sup>235,238</sup> U

**Table 1:** List of isotopes measured at n\_TOF. The bottom line shows the isotopes used for normalization or background rejection

Another important measurement performed at n\_TOF was the capture reaction of <sup>232</sup>Th. This isotope is crucial for the investigation of the alternative nuclear fuel cycle based on Th/U. The earlier measurements of the <sup>232</sup>Th(n,γ) cross-section presented severe discrepancies, of up to 30 %, an uncertainty much too large for one of the key elements of the fuel. At n\_TOF, the measurement was performed with the same setup used for the <sup>151</sup>Sm(n,γ) reaction, based on C<sub>6</sub>D<sub>6</sub> liquid scintillators. The much more accurate results of n\_TOF, characterized by an uncertainty of around 5 %, will allow to generate more reliable nuclear data library for <sup>232</sup>Th, as requested for the implementation of the thorium fuel cycle in existing or in innovative nuclear power devices.

Since the 4π calorimeter has become operational, high quality data have been collected on the capture cross-section of a series of minor actinides. Thanks to the high instantaneous neutron flux, the radioactivity of the samples, which has hindered the measurements at other facilities, affected only marginally the n\_TOF measurements. For this reason, the data collected at n\_TOF are much more accurate than those from previous measurements, and in some cases (as for example for the <sup>243</sup>Am), represent the first data ever collected on capture cross-sections for highly radioactive samples.

Finally, new and accurate results were also obtained at n\_TOF on the fission cross-sections for <sup>232</sup>Th, Uranium isotopes and a variety of minor actinides. In most cases, the superior resolution of the n\_TOF neutron beam has resulted in higher precision data, with new resonances observed, or with the clear identification of multiplets where single bumps had been previously observed. As for capture, the innovative features of the n\_TOF facility, coupled with the high performance of the experimental apparatus and of the acquisition system, have provided nuclear data with the high accuracy required for the development of new nuclear technologies.

In conclusion, the innovative features of the n\_TOF facility, coupled to the high performance experimental apparatus and acquisition systems, allowed to collect long needed high quality data on capture and fission cross-sections. The n\_TOF data are expected to improve current models of Stellar Nucleosynthesis, as well as the databases used for the development of advanced nuclear technologies.

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