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Measurements of neutron capture cross-sections at n_TOF

G. Tagliente[1,a], U.Abbondanno[2], G.Aerts[3], F.Alvarez-Velarde[4], H.Álvarez-Pol[5], S.Andriamonje[3], J.Andrzejewski[6], G.Badurek[7], P.Baumann[8], F.Bečvář[9], J.Benlliure[5], E.Berthoumieux[3], F.Calviño[10], D.Cano-Ott[4], R.Capote[11], P.Cennini[12], V.Chepel[13], E.Chiaveri[12], N.Colonna[1], G.Cortes[10], D.Cortina[5], A.Couture[14], J.Cox[14], S.Dababneh[15], M.Dahlfors[12], S.David[16], R.Dolfini[17], C.Domingo-Pardo[18], I.Duran[5], M.Embid-Segura[4], L.Ferrant[16], A.Ferrari[12], R.Ferreira-Marques[13], H.Frais-Köelbl[19], W.Furman[20], I.Goncalves[21], R.Gallino[22], E.Gonzalez-Romero[4], A.Goverdovski[23], F.Gramegna[24], E.Griesmayer[19], F.Gunsing[3], B.Haas[25], R.Haight[26], M.Heil[15], A.Herrera-Martinez[12], S.Isaev[16], E.Jericha[7], F.Käppeler[15], Y.Kadi[12], D.Karadimos[27], M.Kerveno,[8], V.Ketlerov[23], P.Koehler[28], V.Konovalov[20], M.Krtička[9], C.Lamboudis[29], H.Leeb[7], A.Lindote[13], I.Lopes[13], G.Lorusso[1], M.Lozano[14], S.Lukic[8], J.Marganiec[6], S.Marrone[1], J.Martinez-Val[30], C.Massimi[36], P.Mastinu[24], A.Mengoni[12] , P.M.Milazzo[2], A.Molina-Coballes[18], C.Moreau[2], M.Mosconi[15], F.Neves[13], H.Oberhummer[7], S.O'Brien[14], J.Pancin[3], T.Papaevangelou[12], C.Paradela[5], A.Pavlik[31], P.Pavlopoulos[32], J.M.Perlado[30], L.Perrot[3], M.Pignatari[22], R.Plag[15], A.Plompen[33], A.Plukis[3], A.Poch[10], A.Policarpo[13], C.Pretel[10], J.Quesada[11], W.Rapp[15], T.Rauscher[34], R.Reifarth[26], C.Rubbia[17], G.Rudolf[8], P.Rullhusen[33], J.Salgado[21], J.C.Soares[21], C.Stephan[16], J.L.Tain[18], L.Tassan-Got[16], L.Tavora[21], R.Terlizzi[1], G.Vannini[36], P.Vaz[21], A.Ventura[35], D.Villamarin[4], M.C.Vincente[4], V.Vlachoudis[12], F.Voss[15], H.Wendler[12], M.Wiescher[14], K.Wisshak[15]

[7]*Atominstitut TUWien, Austria,* [31]*Inst. für Isotopenforschung und Kernphysik, Univ. Wien, Austria,* [19]*Fachhochschule Wiener Neustadt, Austria,* [12]*CERN, Geneva, Switzerland,* [16]*CNRS/IN2P3 - IPN, Orsay, France,* [8]*CNRS/IN2P3 - IReS, Strasbourg, France,* [3]*CEA/Saclay - DSM, Gif-sur-Yvette, France,* [15]*FZK, Karlsruhe, Germany,* [27]*Univ.Ioannina, Greece,* [29]*Aristotle Univ. Thessaloniki, Greece,* [35]*ENEA, Bologna, Italy,* [24]*INFN, LNL, Italy,* [1]*INFN, Bari, Italy,* [2]*INFN, Trieste, Italy,* [17]*Univ. Pavia, Italy,* [33]*CEC-JRC-IRMM, Geel, Belgium,* [13]*LIP - Coimbra & Dep. Fisica Univ. Coimbra, Portugal,* [20]*JINR, Dubna, Russia,* [23]*Institute of Physics and Power Engineering, Obninsk, Russia,* [4]*CIEMAT, Madrid, Spain,* [18]*CSIC, Valencia, Spain,* [30]*Univ. Politecnica Madrid, Spain,* [11]*Univ. Sevilla, Spain,* [5]*Univ. Santiago de Compostela, Spain,* [10]*Universitat Politecnica de Catalunya, Barcelona, Spain,* [34]*Dep. Physics and Astronomy - Univ. Basel, Switzerland,* [26]*LANL, New Mexico, USA,* [28]*ORNL, Oak Ridge, USA,* [14]*Univ. Notre Dame, USA,* [21]*ITN, Lisbon, Portugal,* [9]*Charles Univ., Prague, Czech Republic,* [25]*CNRS/IN2P3 - CENBG, Bordeaux, France,* [6]*Univ. Lodz, Poland,* [32]*Pôle Universitaire Léonard de Vinci, Paris, France,* [36]*Dip. Fisica, Univ. Bologna and INFN Bologna, Italy,* [22]*Dip. Fisica Generale, Univ. Torino and INFN Torino, Italy*

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Abstract. Capture cross-sections on isotopes relevant to accelerator driven systems for energy production and nuclear waste transmutation, and to stellar nucleosynthesis have been studied at the innovative neutron time of flight facility n_TOF at CERN. The extremely high instantaneous neutron flux and the low background conditions in the experimental area make this facility unique for accurate measurements on low-mass or radioactive samples. The n_TOF facility is described, together with the features of the experimental apparata used. First results of the experimental campaign 2002-2004 are presented.

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1. Introduction

The experimental determination of neutron cross-sections is of great importance in several fields of fundamental and applied Nuclear Physics. High resolution neutron capture cross-section data have gained much interest in recent years due to the development of new ideas related to nuclear energy production, such as transmutation of nuclear waste or the thorium fuel cycle [1].
In currently operating nuclear fission reactors the spent fuel elements account for the largest part of the nuclear waste in terms of radiation hazard. Apart from uranium, they consist of fission product and isotopes of plutonium and of the minor actinides neptunium, americium and curium. The high activity and long lifetimes of several isotopes requires a long term storage under safe conditions on a time scale of thousands of years. One solution to this problem is to store the nuclear waste in some deep underground repository as the Yucca Mountain in the US that has a capacity of 70,000 ton with a cost of 15 G$. This solution could result largely insufficent if in the future, as projected, there will be an increase of nuclear power with conventional critical reactors. Another solution is the transmutation of the nuclear waste by means of neutron capture or fission, which could considerably reduce the radiotoxicity inventory.
Another approach is to reduce the amount of nuclear waste, the higher actinides, by using a fuel cycle based on ^{232}Th. The isotope ^{232}Th is not fissile but after neutron capture followed by β-decay, the fissile isotope ^{233}U is formed. The build-up of the minor actinides, especially americium and curium, is strongly suppressed due to the lower atomic and mass number of thorium.
A related development concerns subcritical reactors, driven by charged particle accelerators, the so-called accelerator driven system (ADS), proposed in configuration for waste trasmutation but also for energy production in combination with the thorium fuel cycle. The new developments in the field of emerging nuclear technology require new and accurate neutron cross-section data. An improved accuracy in the neutron capture cross section is also a pre-requisite for improvements in the field of Nuclear Astrophysics. In fact the reliability of models of the stellar s-process nucleosynthesis can only be tested by new data in neutron capture cross-sections, with uncertainties of a few percent. Moreover, recent developments have emphasized the importance of neutron capture nucleosynthesis for probing the deep interior of Red Giant stars and for following the enrichment of heavy elements during the galactic evolution. Data required in the energy range from 0.3 to 300 keV are largely missing, particularly in the mass region A $\leq$ 100, where cross-sections are small and dominated by single resonances[2].
The n_TOF performances are optimal for accurate capture cross-section measurements on low-mass and radioactive samples.

2. The n_TOF facility

The n_TOF facility, based on an idea by Rubbia et al.[3], became fully operational in May 2002, when the scientific program started [4]. Neutrons are produced by spallation reactions induced by a pulsed, 6 ns wide, 20 GeV/c proton beam with up to 7 x 10^{12} protons per pulse, impinging on a 80x80x60 cm^3 lead target. A 5 cm water slab surrounds the lead target acting as a coolant and as a moderator of the initial fast neutron spectrum. An isolethargic neutron flux distribution is produced over a wide energy range (1 eV - 250 MeV). Neutrons emerging from the target propagate in a vacuum pipe inside the 200 m long time-of-flight tunnel. Two collimators are mounted along the flight path, one 13.5 cm in diameter placed at 135 m from the lead target and one at 180 m with a diameter of 2 cm for the capture measurements. This collimation results in a Gaussian-shaped beam profile [5]. A 1.5 T sweeping magnet placed at 40 m upstream the experimental area is used to deflect charged particles traveling along the vacuum pipe. For an efficient background suppression, several concrete and iron walls are placed along the time-of-flight tunnel. The measuring station is located at 187.5 m from the spallation target. The neutron beam is monitored up to 1 MeV by a low-mass system, based on a thin Mylar foil with a ^{6}Li deposit placed in the beam, surrounded by an array of silicon detectors placed outside the beam. The detection of the tritons and α's produced in the ^{6}Li(n,α)^{3}H reaction gives a direct measure of the neutron flux. The data acquisition system is based on flash ADCs with a sampling rate up to 1 GHz for recording the detector signals during nearly 20 ms for off-line analysis [7].

3. The experimental set-up and analysis technique

In the first phase of the n_TOF project, neutron capture measurements were carried out with an array of C_6D_6 (deuterated benzene) liquid scintillator cells. This detector has the advantage of being the least sensitive to scattered neutrons. Cylindrical cells 12.7 cm diameter by 7.8 cm thickness have been specifically designed and constructed for n_TOF [8],with a carbon fibre container, and a minimization of the support material, which has led to a very low neutron sensitivity, allowing to perform measurements of isotopes with a large scattering to capture ratio.
Due to the small solid angle coverage and the low intrinsic efficiency of the C_6D_6 detectors, only one γ-ray per event is detected from the de-excitation cascade following neutron capture. For an accurate cross-section determination, the efficiency of the set-up has to be made independent on the details of the de-excitation cascade, in particular on the γ-ray multiplicity. To this aim the pulse height weighting technique (PHWT) has been used. It consists in modifying by software, in suitable way, the detector response so that the efficiency ε_γ is proportional to the photon energy E_γ (ε_γ=kE_γ). Under these conditions the efficiency for detecting a cascade becomes proportional to the known cascade energy E_c and independent on the actual cascade path ($\varepsilon_c \approx \Sigma_j \varepsilon_{\gamma j} = kE_c$). The proportionality of the efficiency with γ-ray energy is achieved by modifying the detector energy response distribution

R(E) with a pulse height weighting factor W(E) applied to the recorded spectrum. The weighting functions are determined on the basis of simulations of the detector response performed with the code GEANT. The weighting function technique has been validated by measuring isotopes with well-known capture cross-sections, such as ^{nat}Fe, ^{nat}Ag and ^{197}Au. An overall accuracy of 2% has been found [6][9].
In the second phase of the n_TOF project the neutron capture measurements have been performed with a Total Absorption Calorimeter (TAC). The design of the n_TOF TAC is based on 40 BaF_2 crystals, 15 cm in thickness, in the form of truncated pyramids. Each of the pentagonal and hexagonal crystals subtends the same solid angle with respect to the sample in the centre.
On average the crystals exhibit an energy resolution of 14% at 662 keV and an excellent time resolution of about 500 ps. To minimize the background induced by neutron scattered by the sample and captured in the detector, a central neutron absorber consisting of a ^{6}Li-loaded moderator and ^{10}B-loaded carbon fibre capsules covering the crystal have been implemented. Further features of the TAC are photomultiplier tubes with special voltage dividers, and a data recording system based on flash-ADCs [7]. These features are essential in order to reduce the sensitivity to scattered neutrons as well as for achieving the required capacity and quality for data acquisition and storage. Thanks to these features, the measurements can take full advantage of the general TAC characteristics, as the unique signature of capture events, which can be identified via the recorded information on the total energy and on multiplicity of the event. This allows to efficiently discriminate against ambient background and competing reaction channels such as inelastic scattering and fission. Figure 1 shows a view of the TAC, as implemented in the code GEANT4 for detailed MC simulations. The performance of the TAC has been investigated

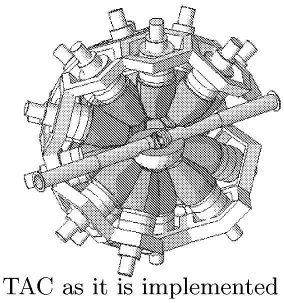

Fig. 1. View of the TAC as it is implemented in the code GEANT4.

both experimentally (with standard calibration sources and the reference ^{197}Au(n,γ) cross reaction) and by MC simulation [10]. Furthermore, all sources of background have been measured and compared to GEANT4 simulations for performing the corrections necessary for an accurate capture cross section analysis.

4. Experimental Campaign

The n_TOF experimental campaigns 2002-2004 have covered capture (listed in Table 1) and fission cross-section measurements for a large number of samples. The motivations and the physics case of the various measurements can be found in the proposals submitted to the CERN INTC Committee [11].

Table 1. Capture cross-sections measured at n_TOF

Reaction	detectors set-up	year
^{24,25,26}Mg(n,γ)	C_6D_6	2003
^{90,91,92,93,94,96}Zr(n,γ)	C_6D_6	2003
^{139}La(n,γ)	C_6D_6	2003
^{151}Sm(n,γ)	C_6D_6	2002
^{186,187,188}Os(n,γ)	C_6D_6	2003
^{204,206,207,208}Pb(n,γ)	C_6D_6	2002
^{209}Bi(n,γ)	C_6D_6	2002
^{232}Th(n,γ)	C_6D_6	2002
^{237}Np(n,γ)	TAC	2004
^{240}Pu(n,γ)	TAC	2004
^{243}Am(n,γ)	TAC	2004

5. Results

5.1. ^{151}Sm(n,γ) cross section

This measurement is important in Nuclear Astrophysics because the unstable isotope ^{151}Sm ($t_{1/2}$=93 yr) is a branching point in the s-process path. In particular, this branching is sensitive to the temperature at which the s-process is taking place. The accurate determination of the neutron capture cross-section of this isotope can thus provide crucial information on thermodynamical conditions in AGB stars. The ^{151}Sm(n,γ) cross section has also important implications in nuclear technologies, since it is a fission fragment accumulating during nuclear reactor operation. For this reason a specific request is included in the NEA "High Priority Nuclear Data Request List". The measurement has been performed with an enriched 200 mg $^{151}SmO_2$ sample, encapsulated in a 0.1 mm thick Ti-can. The result obtained at n_TOF yield a value for the Maxwellian averaged cross-section $< \sigma(n,\gamma) > = 3100$ $\pm$ 160 mb, a value larger than that estimated by model calculations (see Fig 2)[12]. The stellar neutron capture rate, for the first time based on experimental values, allowed to reach important conclusions with respect to the nucleosynthesis in this mass region: i) the classical model, based on the phenomenological study of the s-process fails to describe the branching at ^{151}Sm consistently; ii) the p-process contributes 30% of the solar system observed abundance of ^{152}Gd.

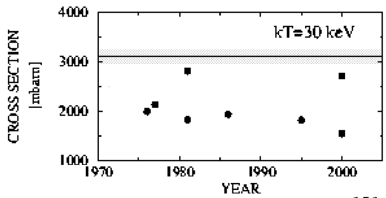

Fig. 2. Maxwellian averaged (n, γ) cross section of ^{151}Sm (shaded band) and previous calculations (symbols).

5.2. ^{232}Th(n,γ) cross section measurements

The ^{232}Th(n,γ) cross-section is crucial for the investigation of the nuclear fuel cycle based on Th/U, an alternative to the present standard cycle based on Pu/U. The cross-section has been measured previously, but severe discrepancies resulted from these early measurements. In particular, in the neutron energy region 5 keV $\leq E_n \leq$ 15 keV, the discrepancy reaches 30%. This is, of course, far too large for the capture cross-section of a key element of the fuel. The measurement was performed at n_TOF in the 2002 with the same detector set-up used in the ^{151}Sm measurement. The much improved accuracy of the result obtained at n_TOF will allow the generation of new ^{232}Th data which can be used for the implementation of the thorium fuel cycle in the existing or innovative nuclear power devices.

6. Conclusion

The innovative features of the n_TOF facility and the high performance of the experimental apparata and acquisition system have allowed to collect accurate data on capture cross-section measurements. The results on the ^{151}Sm(n,γ) and ^{232}Th(n,γ) reactions demonstrate the high quality of the data collected at n_TOF. Even for these radioactive samples, the very high instantaneous neutron flux has allowed to extract cross-sections with high accuracy, while the high resolution of the neutron beam has led to improvements in the resolved resonance region. The vast experimental program undertaken at n_TOF will provide new data needed for improving current stellar models, as well as for making reliable databases for the development of new ideas in the field of emerging nuclear technologies.

Notes

a. Permanent address: INFN Bari, Via Orabona 4, I-70126 Bari, Italia;
E-mail: giuseppe.tagliente@ba.infn.it

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