High-Resolution Study of ^{237}Np Fission Cross Section from 5 eV to 1 MeV

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

The n_TOF Collaboration

[1]Atominstitut der Österreichischen Universitäten, Technische Universität Wien, Austria, [2]Institut für Isotopenforschung und Kernphysik, Universität Wien, Austria, [3]Fachhochschule Wiener Neustadt, Wiener Neustadt, Austria, [4]CERN, Geneva, Switzerland, [5]Centre National de la Recherche Scientifique/IN2P3 - IPN, Orsay, France, [6]Centre National de la Recherche Scientifique/IN2P3 - IReS, Strasbourg, France, [7]CEA/Saclay - DSM, Gif-sur-Yvette, France, [8]Forschungszentrum Karlsruhe GmbH (FZK), Institut für Kernphysik, Germany, [9]University of Ioannina, Greece, [10]Aristotle University of Thessaloniki, Greece, [11]ENEA, Bologna, Italy, [12]Laboratori Nazionali di Legnaro, Italy, [13]Istituto Nazionale di Fisica Nucleare, Bari, Italy, [14]Istituto Nazionale di Fisica Nucleare, Trieste, Italy, [15]Università degli Studi Pavia, Pavia, Italy, [16]CEC-JRC-IRMM, Geel, Belgium, [17]LIP - Coimbra & Departamento de Fisica da Universidade de Coimbra, Portugal, [18]Joint Institute for Nuclear Research, Frank Laboratory of Neutron Physics, Dubna, Russia, [19]Institute of Physics and Power Engineering, Kaluga region, Obninsk, Russia, [20]Centro de Investigaciones Energeticas Medioambientales y Technologicas, Madrid, Spain, [21]Consejo Superior de Investigaciones Cientificas - University of Valencia, Spain, [22]Universidad Politecnica de Madrid, Spain, [23]Universidad de Sevilla, Spain, [24]Universidade de Santiago de Compostela, Spain, [25]Universitat Politecnica de Catalunya, Barcelona, Spain, [26]Department of Physics and Astronomy - University of Basel, Basel, Switzerland, [27]Los Alamos National Laboratory, New Mexico, USA, [28]Oak Ridge National Laboratory, Physics Division, Oak Ridge, USA, [29]University of Notre Dame, Notre Dame, USA, [30]Instituto Tecnológico e Nuclear, Lisbon, Portugal, [31]Charles University, Prague, Czech Republic, [32]Centre National de la Recherche Scientifique/IN2P3 - CENBG, Bordeaux, France, [33]University of Lodz, Lodz, Poland, [34]Pôle Universitaire Léonard de Vinci, Paris La Défense, France, [35]Dipartimento di Fisica, Università di Bologna, and Sezione INFN di Bologna, Italy, [36]Dipartimento di Fisica Generale, Università di Torino and Sezione INFN di Torino, I-10125 Torino, Italy, [37]Tokyo Institute of Technology, Tokyo, Japan, [38]Japan Atomic Energy Research Institute, Tokai-mura, Japan, [39]NCSR, Athens, Greece, [40]National Technical University of Athens, Greece

Abstract. A series of measurements of ^{237}Np fission cross section have been performed at the CERN spallation neutron facility n_TOF which covers a wide energy range from 1 eV up to 250 MeV. A fast ionization chamber (FIC) was used as a fission fragment detector with registration efficiency of not less than 97 %. Particular attention was paid to correct the fission cross section with use of ^{235}U standard. Total experimental uncertainties are determined to be at the level of 3%. Analysis of the experimental data in the restricted neutron energy from 5 eV up to 1 MeV showed a systematic deviation from evaluated data (ENDF/B-VI). This discrepancy amounts to up to a factor 3 for resolved resonances in the neutron energy range of 5 eV - 2 KeV, and is in good agreement with some previous experiments. A similar disagreement at the level of 6-7% was found for higher energies around the threshold (E_n = 300 keV-1 MeV). This energy range is essential for the transmutation of neptunium in ADS or fast reactors. It is concluded that an updated evaluation of nuclear data for ^{237}Np is required.

INTRODUCTION

Neptunium-237 is the most important actinide in the problem of spent fuel transmutation. While the development of power facilities of a new generation like fast reactors or accelerator driven systems (ADS) need accurate nuclear data, large discrepancies in the fission cross section data available up to now in the resolved resonances region have been detected. This situation motivated experimental activities devoted to repeat fission cross section measurements in a wide neutron energy range, aiming at obtaining high accuracy cross section data. This experimental programme has been setup at the CERN neutron time-of-flight facility n_TOF[1]. This is a spallation neutron source where neutrons are generated by the interaction of the 20 GeV proton beam from the CERN PS accelerator with a massive water-cooled lead target. The spallation neutrons have an energy spectrum that covers the range 1 eV to at least 250 MeV with intensity sufficient for the irradiation of thin neptunium samples installed in an ionization chamber. The time of flight (TOF) technique gives high-energy resolution (up to 10^{-4}) with a width of the proton bunch of less than 7 ns and a flight path of 186m. These experimental conditions allowed us to perform detailed studies in the resolved resonance region and simultaneously in the fast region. The results of the analysis reported here cover the neutron energy range from 20 eV to approximately 700 KeV, chosen to point out the most important discrepancies between experimental and evaluated nuclear data for this isotope.

EXPERIMENTAL DETAILS

The ^{237}Np and ^{235}U targets were installed back-to-back in a multiple ionization chamber. All targets were fabricated from high purity material using the painting technique and analyzed with alpha-counting with semiconductor detector in good geometry. The total masses were 12.82±0.08 mg for neptunium and 31.8±0.2 mg for uranium.

A fast parallel plate ionization chamber (FIC0) was used as a fragment detector. The detector chamber is a cylindrical stainless steel assembly 30 cm long and 40 cm in diameter with 2 mm Al windows. The operating gas was a mixture 90%Ar + 10% CF_4 at a pressure of 600 mbar. The distance between electrodes was chosen to be 5 mm to get fast signals with 300 V high voltage supply. Our past experience in the use of analogous chambers showed a time resolution of 2-3 ns with minimal overlapping of fission fragments and α-particles well suited for the n_TOF facility. This is sufficient for n_TOF timing and neutron flux. The registration efficiency of the detector was near 98%.

The ionization detector was equipped with fast electronics including time-digital converters (TDC) and wave-form digitizers (WFD, or FADC). TDC are used for long times covering the neutron energy range from 0.1 eV to 1 MeV. FADC have been used for measurements above 20 KeV.

The energy calibration was performed using the resolved resonances of ^{235}U(n,f) and an example for a limited energy range is presented in Fig. 1. The results show very good agreement with known experimental data. This allowed us to normalize the experimental fission yield for ^{237}Np(n,f) reaction into absolute fission cross section as a function of neutron energy.

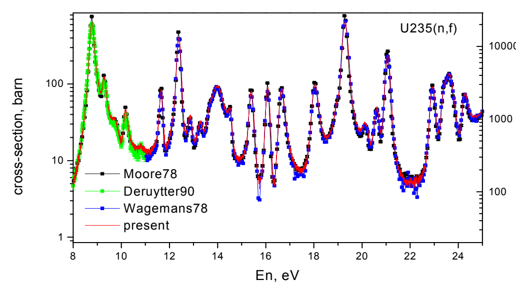

FIGURE 1. Comparison of ^{235}U fission cross section with the existing experimental data (from EXFOR).

RESULTS AND DISCUSSION

The results of the fission cross-section measurements around well-known resonance cluster at the neutron energy E_n = 39.9 eV are presented in Fig. 2 together with other available data [2,3,4]. Disagreement between different data sets looks too large to satisfy practical needs. The list of calculated fission cross section integrals $A_\lambda = \int \sigma_f \, dE_n$ for neutron energies between 38 and 42 eV is presented in Table 1. A factor of 3 higher value is found in a comparison of the present data and the data of Plattard *et al.* [3].

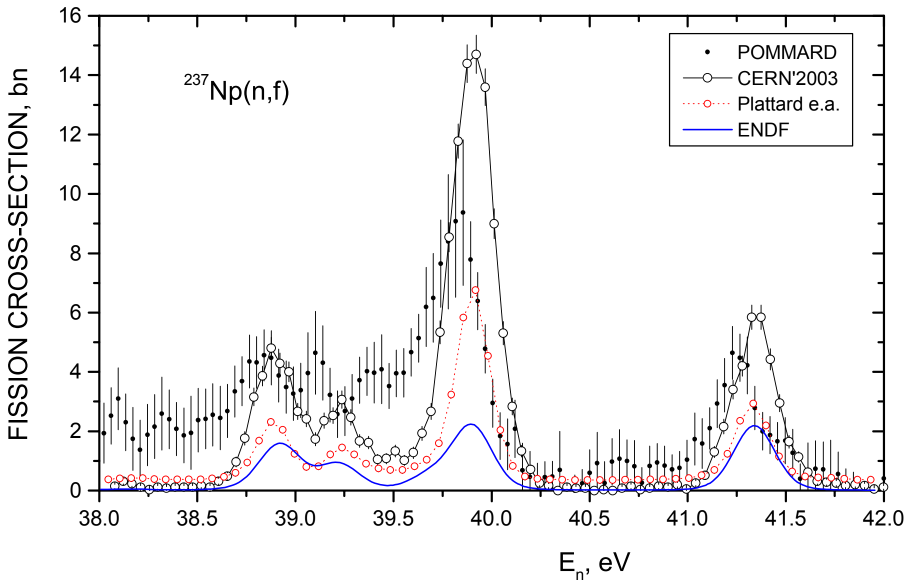

FIGURE 2. ^{237}Np(n,f) cross-section between 38 and 42 eV.

TABLE 1. A comparison of fission cross-section integrals from different experiments. The ENDF/B-VI value is lower than the value of Plattard *et al.* (see Fig. 2).

Author	A_λ, b*eV	Comment
n_TOF 2003	8.34 ± 0.10	Spallation source
Brown *et al.* [2]	10.2 ± 2.5	POMMARD
Plattard *et al.* [3]	2.88 ± 0.03	Linac, Saclay
Dermenjiev *et al.* [4]	9.14 ± 1.37	IBR-30, Dubna

The extremely good n_TOF energy resolution allowed us to get more accurate and detailed data above 600 eV. This is shown in Fig. 3 where the present data are compared with the data of Plattard *et al.* [3]. Resonance doublets around E_n = 1150, 1270, 1320 eV are clearly resolved. The determination of more accurate resonance parameters is possible. This work is in progress with SAMMY code.

The experimental data for fast neutrons are shown in Fig. 4. The ratio of the fission cross sections of ^{237}Np to that of ^{235}U is used as experimental observable. Agreement between the different data sets is good excluding the region above 450 KeV. To clarify the situation fission cross-sections measured with only TOF technique are presented in Fig. 5. Obviously, all TOF experiments show higher fission cross sections than contained in present in evaluated data libraries. The discrepancy is of the order of 6-7%.

REFERENCES

1. CERN n_TOF Facility: Performance Report- CERN-SL-2002-053 ECT.
2. W.K. Brown *et al.* – Nucl. Phys. **A156**, 609 (1970).
3. S. Plattard – Ph.D. thesis, Saclay, 1973.
4. E. Dermenjiev *et al.* – Phys. At. Nucl. **57**, 1362 (1994).

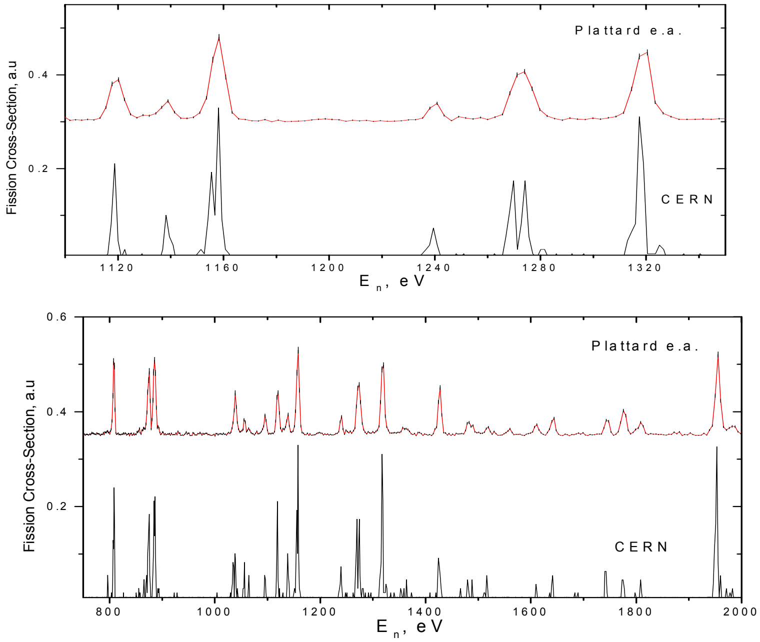

FIGURE 3. Resolution effects in ^{237}Np fission cross section (upper part) and general view between 700 and 2000 eV.

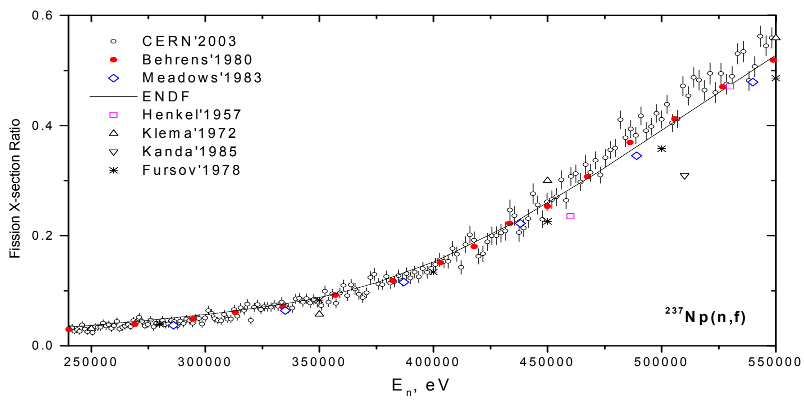

FIGURE 4. Fission cross-section of ^{237}Np(n,f) relative Uranium-235.

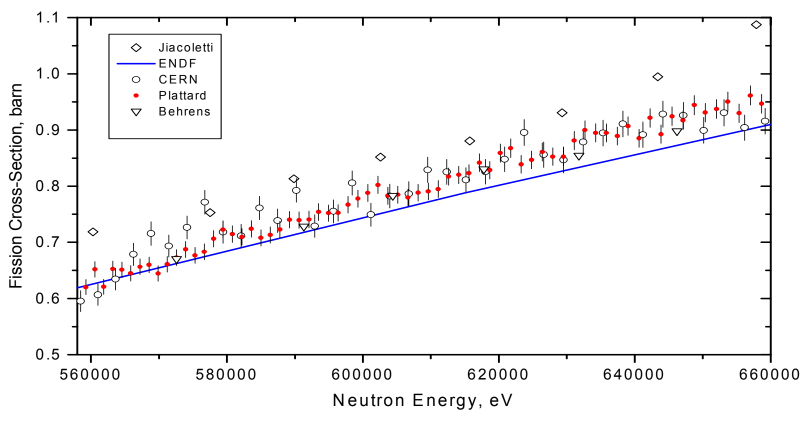

FIGURE 5. Fission cross-section of ^{237}Np(n,f) relative ^{235}U in the energy range up to 660 KeV. Only experimental data from TOF measurements are shown for comparison.