Emission Probabilities of γ-rays from ^{238}Np and their use for determination of the Thermal Neutron Capture Cross Section of ^{237}Np

A. Letourneau[1], F. Marie
Irfu, CEA-Saclay, 91191 Gif-sur-Yvette, France
P. Mutti
Institut Laue-Langevin,38000 Grenoble, France
I. AlMahamid
Wadsworth Center, Laboratory of Inorganic and Nuclear Chemistry, Albany, NY, USA

Abstract

The relevant absolute γ-ray emission probabilities from the β-decay of ^{238}Np were measured by means of α and γ-spectroscopic techniques. We obtained values of (25.6±0.4) %, (8.9±0.2) % and (18.8±0.3) % for the 984.45, 1025.87 and 1028.54 keV γ-rays, respectively, in agreement with the previous measured ones. These intensities were used to deduce the thermal neutron capture cross section of ^{237}Np for which a value of (182.2±4.5) b is obtained higher by 11% than the recommended value.

Keywords: neptunium-237, γ-ray intensities, spectroscopy, thermal neutron capture, cross section, activation

1. Introduction

Neptunium-237 is a long lived ($T_{1/2}$=2.144 10^6 years) isotope which is present in spent-fuel of light water reactors and contributes significantly to the long-term radiotoxicity of nuclear wastes. The ^{237}Np capture cross section is an important parameter in the field of transmutation and also in the field of high burn-up fuel studies as it strongly affects the production of heavier elements. When bombarded with neutrons, ^{237}Np produces the intense α-emitter ^{238}Pu through the β-decay of ^{238}Np, generating additional radiotoxicity and causing a loss of neutrons in the fuel.
The most recent measurements of the ^{237}Np(n,γ) cross section as a function of the neutron energy in the range of 0.02 to 100 eV (Shcherbakov et al, 2005) have shown an overall good agreement with earlier measurements and evaluated data but have pointed out a problem related to the absolute normalization of the measurement. Generally, for time of flight measurements, the normalization is given by measurements based on activation techniques or sometimes calculated from resonance parameters as performed by Weston and Todd (1981). However, the thermal cross section values obtained by activation techniques and pile oscillation method present large discrepancies that affect the precision required for such cross section.
If we consider only the values measured by means of α-spectroscopic techniques from Brown and Hall (1956), Schuman and Berreth (1969), and Eberle et al. (1971), we obtain a mean cross section of 180 b with an error of 4 b that is compatible with the value of (181±6) b obtained by Weston and Todd (1981). A significant discrepancy is observed when comparing the above values to subsequent measurements done by γ-spectroscopic techniques where a mean value of 155.3 b with an error of 2.2 b is found in the original data of Jurova et al. (1984), Kobayashi et al. (1994) and Katoh et al. (2003). It was later shown by Harada et al. (2006) that part of the discrepancies between α and γ measurements comes from the γ-ray emission probabilities of the ^{238}Np decay. Indeed, all the experiments based on the γ-activation technique used the 984-keV γ-ray to determine the amount of ^{238}Np. Kobayashi et al. (1991), Katoh et al. (2003) and Jurova et al. (1984) used an intensity value of (27.8±0.8) % from Lederer (1981). In their experiment, Harada et al. (2006) used coupled γ- and α-spectroscopic techniques to measure the emission probability of this γ-ray. They found a value of (25.2±0.5) % that is lower than the evaluated one from Lederer (1981). They also measured the 312 keV γ-ray emission probability from the decay of ^{233}Pa. They used these two values to correct the three previous experiments. The corrected cross sections became compatible with the cross section measured by Harada et al. (2006), yielding a mean value of (170±3) b, while the value of Kobayashi et al. (1994) was increased by the correction to (196±4) b, increasing therefore the discrepancies between the data.
Kobayashi et al. (1994) used the thermal neutron facility of the Kyoto University Reactor (KUR) where the neutron energy distribution is in good agreement with a Maxwellian distribution at a temperature of 60 °C. On the other hand, most of the experiments were done using the Cd ratio technique, samples being irradiated into

[1]Corresponding Author: Tel. : +33 1 69 08 76 01, Fax : +33 1 69 08 75 84
E-mail address : aletourneau@cea.fr (A. Letourneau)

neutron fluxes with non-negligible epithermal component. For example, Katoh et al. (2003) and Harada et al. (2006) used the pneumatic tube facility of the (KUR). In this irradiation channel, the epithermal to thermal ratio is about 0.033 in the Westcott convention. As discussed in Katoh et al. (2003), the huge first resonance which is located near the Cd cut-off could have an impact on the final determination of the cross section.
In the present experiment we took advantage of the High Flux Reactor of the Laue-Langevin Institut (Grenoble – France) and the highly thermalized neutron flux of the H9 irradiation channel to extend the measurement on the absolute emission probabilities to the three most intense γ-rays from ^{238}Np and to explore the effect of the first resonance on the determination of the capture cross section. The ^{237}Np sample was irradiated in the H9 channel, which provides a neutron flux with an estimated epithermal to thermal ratio of 0.0021 in the Westcott convention. The γ-emission probabilities were determined from the γ-rate to the α-activity of ^{238}Pu. The cross section was deduced from the γ- and α-activities of the sample. This experiment is a part of a more complete program dedicated to the study of the transmutation chain of ^{237}Np when irradiated into thermal-neutron high-fluxes.

2. Description of the experiment

2.1 Target preparation

The ^{237}Np target was prepared at the Lawrence Berkeley National Laboratory. The ^{237}Np sample was electrodeposited by molecular plating technique (AlMahamid et al., 2005) on a 12 mm diameter Ni foil. The deposit area was a circle of 8 mm diameter. The thickness of the Ni plate was chosen to be 4 μm to allow the release of fission products from the target during irradiation in order to reduce the γ-ray background of the sample. The ^{237}Np target was mounted on a Ti holder together with a 6 mm diameter Al foil containing 1% (in mass) of ^{59}Co for measurement of the neutron flux. The distance between the ^{237}Np target and the ^{59}Co neutron flux monitor was 35 mm. Since the irradiation channel allows for a 4π irradiation geometry and the neutron density is quite constant in the irradiation area, the 35 mm distance does not introduce significant uncertainty on the neutron flux normalization.

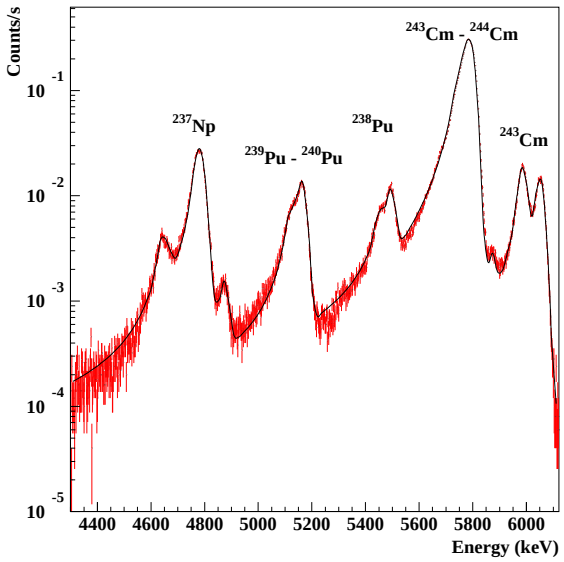

Fig. 1: Alpha energy spectrum prior to the irradiation, recorded at 5 cm from the target. Data were fitted with the α-energy function described in Marie et al. (2006). Shape parameters were determined once and kept identical for each α-ray.

The composition of the ^{237}Np sample and the quantity of ^{237}Np atoms were determined by α-spectroscopy (section 2.2) prior to the irradiation by using the Mini-INCA chamber (Marie et al., 2006). The α-energy spectrum is shown in Fig. 1, where we see that the α-activity is dominated by Cm isotopes. Using the unfolding procedure described in section 2.2, we extracted the activities for each isotopes and the isotopic composition of the ^{237}Np sample listed in Table 1. The ^{237}Np activity was determined in reference to a ^{244}Cm calibration source[2]

and amounts to (370.3±3.7) Bq. By using the nuclear parameters listed in Table 2, we deduced the number of ^{237}Np atoms present in the target to be (3.612±0.036) 10^{16} corresponding to a mass of (14.21±0.14) µg.
The initial isotopic composition of the target was taken into account to calculate the ^{238}Np and ^{238}Pu activities (see section 3).

Table 1: Percentage of atoms contained in the ^{237}Np target prior to irradiation.

^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu	^{243}Cm	^{244}Cm
99.7574 %	0.00135	0.1218	0.10635	0.0083	0.0048

2.2 Activity measurements

The ^{237}Np and ^{59}Co samples were irradiated for 2.733 hours in the so called diaphragm position of the Lohengrin spectrometer (H9 channel). After about half an hour, the samples were transferred into the Mini-INCA chamber to be analyzed by γ- and α-spectroscopy. The experimental setup and the data analysis are described in details in the reference Marie et al. (2006). Here, only a short description is given emphasizing specific aspects of the present measurement. The detection system consists of a High Purity Germanium (HPGe) detector with an intrinsic efficiency of 9.98%, an energy resolution of 1.7 keV at 1173 keV, and a Passive Implanted Planar Silicon (PIPS) detector with an energy resolution of 17 keV for 5.5 MeV α-rays. Signals from the HPGe detector are treated with a Digital Signal Processor (DSP) whereas those from the PIPS detector are treated with a conventional fast Amplifier and Analog to Digital Converter. The DSP calculates its live time with a precision better than 2% even when the electronic dead time is of 50%. Nevertheless, we only used runs for which the dead time was lower than 50%. The absolute efficiencies were established with calibrated γ and α sources[2] having geometrical characteristics similar to the samples. Measurements were made 2-3 days after the end of the irradiation to reduce the total activity due to short-lived contaminants.

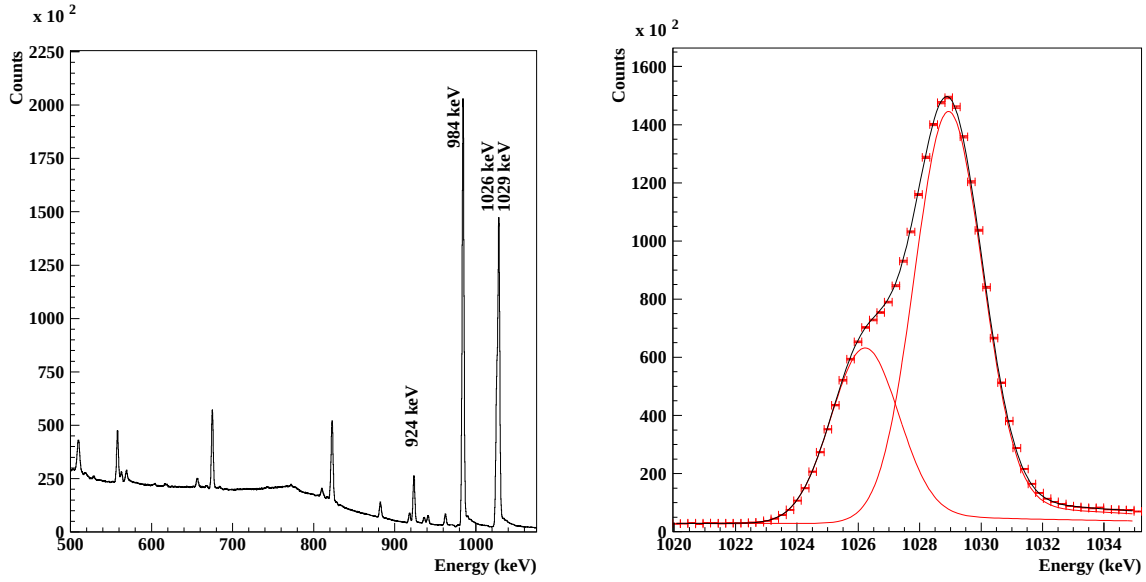

Fig. 2: Energy spectrum of emitted γ-rays by ^{238}Np at 79 cm from the target. The main γ-rays used for the analysis are energy-labeled. Right: zoom-in on the 1026 and 1029-keV doublet, with the results from the multi-peak minimization (lines).

An example of γ-ray energy spectrum is shown in Fig. 2, where the three main γ rays from ^{238}Np (see Table 2) are clearly separated from the background. We also note the presence of contaminants coming from the activation of the target holder. A significant part of the input counting rate is not visible on the figure as it corresponds to γ-rays coming from ^{198}Au (the most intense γ-ray is at 411-keV) and X-rays from the Pb shielding. These γ-rays were responsible for about 80% of the input counting rate. They were also responsible for a depopulating of the ^{238}Np γ-rays by fortuitous coincidences that can not be rejected by the Pile-up rejection

[2] From the Radioactivity Standards Laboratory (LEA) of CERCA. The activities were certified to be known with 1.25% of uncertainty (1σ) for ^{60}Co and ^{152}Eu γ-sources and 2% and 0.7% for the ^{241}Am and ^{244}Cm α-sources, respectively.

system of the electronic. We evaluated the amount of counts lost $dA(E_i)$ in each ^{238}Np photopeak activity $A(E_i)$ from the activities measured in the sum peak areas $A(E_i+E_j)$:

$$dA(E_i) = \sum_j A(E_i + E_j)\frac{(B+S)}{S} \quad (1)$$

where the photopeak signal (S) over the total interacting signal (S+B), (B) being the Compton background, was estimated with an accurate MCNP simulation of the HPGe detector for γ-energies ranging between 100 and 1200 keV. The correction factor that have been applied for the three main γ-rays of ^{238}Np amounts to (5±1)%. It was deduced from the most active γ-rays of the spectra. The photopeak activities were determined from a multi-peak chi-square minimization procedure, each peak being modeled by a Gaussian function added to an exponential background. We added an exponential tail convoluted with the Gaussian function to describe the tail of the photopeak at high energy due to the pile-up with low-energy photons. As can be seen on Fig. 2, the statistics in each photopeak is very high, so that the minimization procedure is very sensitive to the modeling of the peak. To take care of the imperfections in the modeling description we increased the errors obtained by the fit to reduce the chi-square by degrees of freedom close to unity. The final errors of the γ-activities contained the errors of the fit and the errors on the photopeak efficiency (2%).

Table 2: Evaluated nuclear data used in this experiment.

Nuclide	Half-live	Radiation	Energy (keV)	Intensity (%)
^{237}Np [a]	2.144 10^6 y (7)	α	4771.4 (8)	23.15 (29)
		α	4788.0 (9)	47.64 (6)
^{238}Np [b]	2.117 d (2)	γ	984.45 (2)	
		γ	1025.87 (2)	
		γ	1028.54 (2)	
^{238}Pu [b]	87.7 y (3)	α	5456.3 (3)	28.98 (10)
		α	5499.03 (20)	70.91 (10)
^{60}Co [c]	1925.1 d (5)	γ	1173.237	99.9736 (7)
		γ	1332.501	99.9856 (4)

[a] Singh and Tuli (2005)
[b] Chukreev et al. (2002)
[c] Tuli (2003)

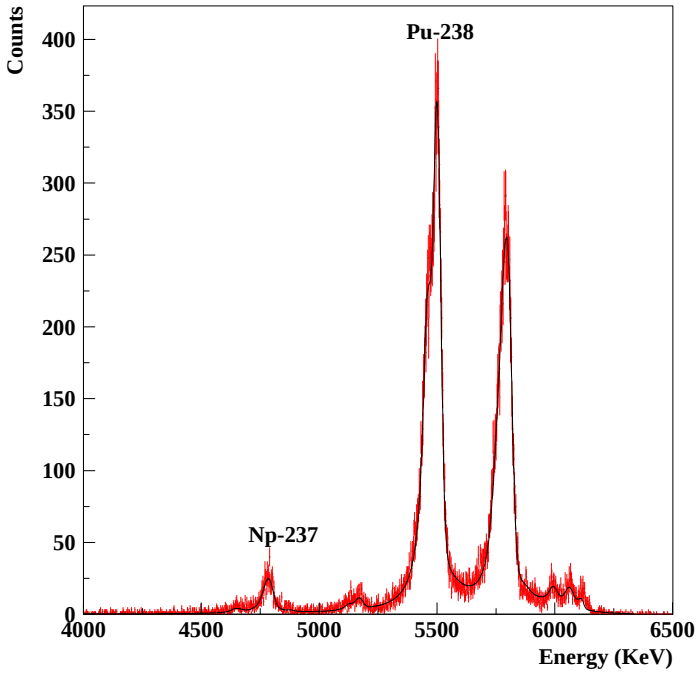

Fig. 3: Alpha-energy spectrum recorded after 3 days of cooling at a distance of 29 cm from the target. Data were fitted with the function described in Marie et al. (2006). Shape parameters were determined once and kept identical for each α-peak.

An example of α-energy spectrum is shown in Fig. 3 where we see the α-rays from ^{238}Pu (see Table 2). For these measurements the electronic dead time was controlled to be less than 12%. We tested that for dead time lower than this value the estimated live time get a precision better than 1%. The dead time was mainly driven by low energy events produced by β and γ rays coming from the target and interacting with the detector. These events, when piled-up with α-rays, are also responsible for the small tail that can be seen at high energy.

The resulting measured activities are shown in Table 3 for ^{237}Np, ^{238}Pu and the 984.45 keV, 1028.54 keV and 1025.87 keV γ-rays, which originates from the (2+) → (2+) and (2+) → (0+) transitions of the 1028.545 keV level in ^{238}Pu, and from the (3+) → (2+) transition of the 1069.943 keV level, respectively. We see that for each measurement, the ^{237}Np activities are compatible within error bars with the activity measured before irradiation. It confirms that the burn-up of the target could be considered as negligible and that there was no loss of material during irradiation.

Table 3: Measured activities for ^{237}Np, the three most intense γ-rays of ^{238}Np and ^{238}Pu.

Cooling time (days)	^{237}Np (Bq)	^{238}Np (MBq)			^{238}Pu (Bq)
		984.45 keV	1025.87 keV	1028.54 keV	
2.4037	371.2±3.6	15.31±0.36	5.32±0.21	11.25±0.34	4890±83
2.5709	367.5±3.4	14.44±0.34	5.05±0.20	10.62±0.32	5184±85
2.7381	367.4±3.4	13.67±0.32	4.72±0.19	9.99±0.31	5263±85
2.9049	387±5	13.06±0.36	4.54±0.26	9.61±0.39	5387±129

3. Absolute γ-ray emission probabilities from ^{238}Np

The absolute γ-ray emission probabilities from ^{238}Np were determined in reference to the measured α activities of ^{238}Pu. As ^{238}Np nuclides decay to ^{238}Pu, their activity as a function of time is linked. The time dependent relation can be established by solving the Bateman equations:

$$A_3(t) = A_3(T_{irr})e^{-\lambda_3 t} + \frac{\lambda_3}{\lambda_3 - \lambda_2} A_2(T_{irr})\left[e^{-\lambda_2 t} - e^{-\lambda_3 t}\right] \quad (2)$$

where $A_2(T_{irr})$ is the ^{238}Np activity by the end of irradiation, $A_3(t)$ is the ^{238}Pu activity after a cooling time t and λ_i are the β or α decay rates. The activities by the end of irradiation can be written as:

$$A_2(T_{irr}) = \lambda_2 \hat{\sigma}_1^c \phi N_1(0)\left(\frac{e^{-\hat{\lambda}_1 T_{irr}} - e^{-\hat{\lambda}_2 T_{irr}}}{\hat{\lambda}_2 - \hat{\lambda}_1}\right) \quad (3)$$

$$A_3(T_{irr}) = A_3(0)e^{-\hat{\lambda}_3 T_{irr}} + \left\{\frac{\lambda_3 \lambda_2 \hat{\sigma}_1^c \phi N_1(0)}{\hat{\lambda}_2 - \hat{\lambda}_1}\left[\left(\frac{e^{-\hat{\lambda}_1 T_{irr}} - e^{-\hat{\lambda}_3 T_{irr}}}{\hat{\lambda}_3 - \hat{\lambda}_1}\right) - \left(\frac{e^{-\hat{\lambda}_2 T_{irr}} - e^{-\hat{\lambda}_3 T_{irr}}}{\hat{\lambda}_3 - \hat{\lambda}_2}\right)\right]\right\} \quad (4)$$

where the quantities $\hat{\lambda}_i = \lambda_i + (\hat{\sigma}_i^c + \hat{\sigma}_i^f)\phi$ are the total disappearance probabilities, $\hat{\sigma}_i^c$ and $\hat{\sigma}_i^f$ being the capture and fission cross sections, Φ is the neutron flux, and the index $i=1$ stands for ^{237}Np. As the burn-up of the target is negligible ($\hat{\lambda}_1 \approx 0$) and by inserting Eq. (3) into Eq. (4), we get the relation between the ^{238}Pu and ^{238}Np activities by the end of irradiation:

$$A_3(T_{irr}) \approx A_3(0)e^{-\hat{\lambda}_3 T_{irr}} + \frac{\lambda_3 A_2(T_{irr})}{(\hat{\lambda}_3 - \hat{\lambda}_2)}\left[1 - \frac{\hat{\lambda}_2}{\hat{\lambda}_3}\left(\frac{1 - e^{-\hat{\lambda}_3 T_{irr}}}{1 - e^{-\hat{\lambda}_2 T_{irr}}}\right)\right] \quad (5)$$

If the quantity $A_3(0)$ is sufficiently small to be neglected and if $\hat{\lambda}_2 \approx \lambda_2$, $\hat{\lambda}_3 \approx \lambda_3$, as it is the case in our experimental conditions, we see that the ratio $A_3(T_{irr})/A_2(T_{irr})$ does not depend on any cross section values but only on the decay rates.

Finally, the measured activities for the γ-rays from ^{238}Np and α-rays from ^{238}Pu can be expressed as a function of the isotope activities taking into account the integration time of the measurement (*RT = 4 hours*):

$$Y_2(t) = I_\gamma A_2(T_{irr})e^{-\lambda_2 t}\frac{(1 - e^{-\lambda_2 RT})}{\lambda_2 RT} \quad (6)$$

$$Y_3(t) = I_\alpha \left\{ A_3(T_{irr}) e^{-\lambda_3 t} \frac{(1-e^{-\lambda_3 RT})}{\lambda_3 RT} + \frac{\lambda_3 A_2(T_{irr})}{\lambda_3 - \lambda_2} [e^{-\lambda_2 t} \frac{(1-e^{-\lambda_2 RT})}{\lambda_2 RT} - e^{-\lambda_3 t} \frac{(1-e^{-\lambda_3 RT})}{\lambda_3 RT}] \right\} \quad (7)$$

where I_γ and I_α are the absolute emission probabilities.

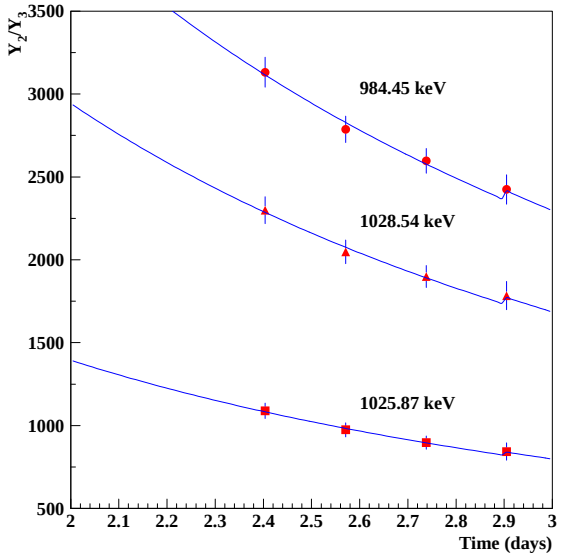

Fig. 4: Ratio of the measured ^{238}Np over ^{238}Pu activities for the three main γ-rays of ^{238}Np. Lines are the fit results with Eq. (6), Eq. (7) and Eq. (5). The step for the later point is due to a shorter time measurement.

We used the ratio of the measured γ activities of ^{238}Np over the α activity of ^{238}Pu (Y_2/Y_3) to deduce the γ-ray intensities I_γ (Fig. 4). Data were fitted using Eq. (6), Eq. (7) and Eq. (5). The errors on the decay rates and α-intensities (Table 2) were propagated by iterating the fit procedure over 5000 iterations. For each iteration the decay rates and intensities were randomly generated within their Gaussian error. The error summary can be written as follow:

(i) The error on the decay rates	< 1%
(ii) The error on the α-ray efficiency and modeling of the α-peak	1%
(iii) The statistical error on the α-ray measurement of ^{238}Pu	<1%
(iv) The error on the emission probability of α rays from ^{238}Pu	<0.3%
(v) The error on the γ-ray photopeak efficiency	2%
(vi) The error on the γ-ray measurement of ^{238}Np (including the statistical errors)	1-3%
(vii) The error on the combustion cross section of ^{238}Np (negligible in our case)	16%

Results of the fits are given in Table 4. The obtained values are in good agreement with the recent evaluation of Chukreev et al. (2002) and with previous measurements. This good agreement with the results of experiments that used different techniques provides a good confidence in our measurement. It should be noted that the absolute value from Lederer (1981) was deduced from an intensity balance of the ground state of ^{238}Pu.

Table 4: Resulting γ-ray intensities, per 100 decays, measured in this experiment and compared with earlier results, in addition to the recommended values of Chukreev et al. (2002). The Winter's values were normalized to our 984.45 keV intensity.

Authors (year)	984.45 keV	1025.87 keV	1028.54 keV	Methods
Present work	25.6±0.4	8.9±0.2	18.8±0.3	αγ
Chukreev et al. (2002)	25.19±0.21	8.72±0.15	18.29±0.23	Eval
Harada et al. (2006)	25.2±0.5			αγ
Rengan et al. (2006)	25.17±0.13	8.766±0.45	18.23±0.93	βγ
Chang et al. (1990)	25.19±0.21	8.71±0.15	18.29±0.23	4πβγ
Lederer (1981)	27.8	9.7±0.6	20.3±0.8	γ
Winter et al. (1972)	25.4	8.76±0.43	18.4±0.9	γ

4. Capture cross section of ^{237}Np

The obtained I_γ values (Table 4) were used to deduce the effective ^{237}Np capture cross section from the fit of the γ and α activities using Eq. (6) and (7) and Eq. (3) and (4) therein (see Fig. 5). Over 5000 fit iterations were performed to propagate properly the errors associated with the fixed parameters, their value being randomly generated within their Gaussian error for each iteration. The neutron flux was measured by means of the standard ^{59}Co(n,γ)^{60}Co reaction. The γ activities at energies of 1173.2 keV and 1332.5 keV were measured after irradiation giving an activity of (6.00±0.06)10^5 Bq for ^{60}Co. The effective capture cross section of ^{59}Co was evaluated by means of a precise simulation of the HFR reactor with the MCNP code (Marie et al., 2006). Considering a value of (37.18±0.06) b (Mughabghab, 2006) for the 0.0253 eV capture cross section of ^{59}Co, we obtained an effective cross section of (31.1±0.9) b. The deduced neutron flux was (6.57±0.15)10^{14} n/cm^2/s. The main uncertainty is attributed to the uncertainty of the initial quantity of ^{59}Co, which is known within an error of 2%.

The effective ^{237}Np capture cross sections deduced from the ^{238}Np activity measurement amounts to (149.9±3.7) b and from the ^{238}Pu activity measurement to (150.3±3.7)b, resulting in an effective cross section of $\hat{\sigma}$= (150.1±3.7) b. The final error includes the previous listed errors added to the following ones:

(i) the error on the neutron flux	2.2%
(ii) the error on the initial quantity of ^{237}Np atoms	1.1%

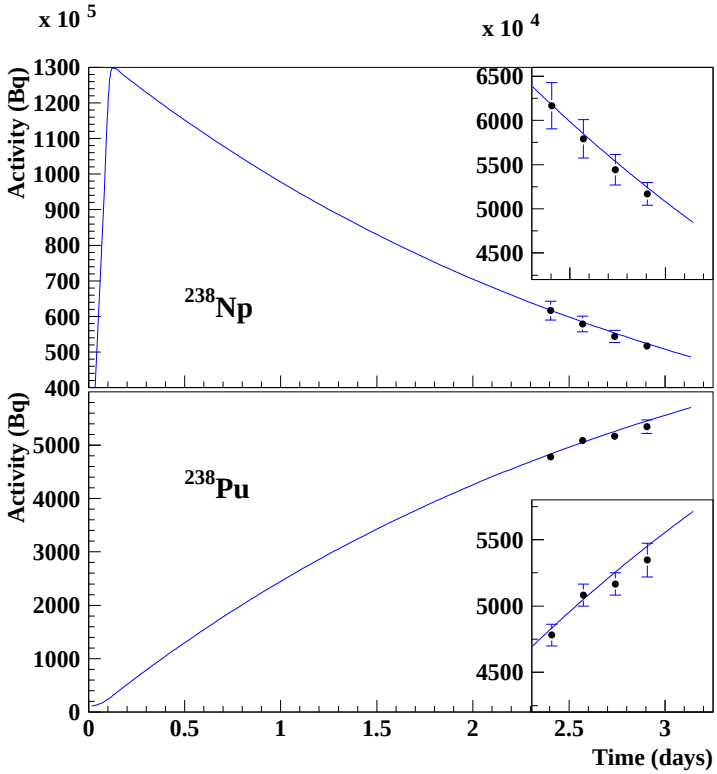

Fig. 5: Measured ^{238}Np and ^{238}Pu activities (symbols) fitted with the Bateman solutions (Eq 6 and 7).

The present value was compared to the effective cross section calculated from the three most used nuclear data libraries by the MCNP simulation of the reactor (see Table 5). While the JEFF 3.1 and ENDF-BVI values are in a good agreement with our value, the latest release of ENDF and JENDL values are lower by about 11 %. Nevertheless, we see that the ratio between the effective cross section and the 0.0253 eV value does not vary significantly. So that we used a mean ratio value of r=0.824±0.003 to convert the effective cross section into the E_0=0.0253 eV value. We found σ_0=(182.2±4.5) b that confirm the value of Weston and Todd (1981) and JEFF 3.1/ENDF-BVI. Our value was also confirm by the recent integral experiment OSMOSE (Noguere et al., 2007) at the Minerve reactor (CEA-Cadarache - France). In this experiment, they showed that our ^{237}Np cross section value drives the best agreement between the calculated and the measured reactivity.

Table 5: Effective ($\hat{\sigma}$) and 0.0253 eV (σ_0) neutron capture cross sections of ^{237}Np.

	σ_0 (b)	$\hat{\sigma}$ (b)	$r = \hat{\sigma}/\sigma_0$
Present work	182.2±4.5	150.1±3.7	
JEFF 3.1 / ENDF-BVI	181	149	0.827
ENDF-BVII / JENDL 3.3	162	133	0.821

5. Discussion

In Table 6, the existing ^{237}Np(n,γ)^{238}Np cross section values from the literature are compared with the value obtained in the present work. We see the good agreement with α-activation based measurements and with the transmission measurement of Smith et al. (1957) if we consider the large uncertainty of this measurement.

Table 6: Present and existing ^{237}Np capture cross section values.

Authors (year)	Methods	σ_0 (b)	σ_0 [a] (b)	σ_0 [b] (b)
Present work	γ –α activation	182.2±4.5 b		
Brown and Hall (1956)	α activation	172±7 b		
Smith et al. (1957)	Transmission	170±22 b		
Tattersall et al. (1960)	Pile oscillator	169±3 b		
Schuman and Berreth (1969)	α activation	185±12 b		
Eberle et al.(1971)	α activation	184±6 b		
Weston and Todd (1981)	Time of flight	181±6 b		
Jurova et al. (1984)	γ activation	158±4 b	172±4 b	174±5
Kobayashi et al. (1994)	γ activation	158±3 b	196±4 b	181±4
Katoh et al. (2003)	γ activation	141.7±5.4 b	168±6 b	156±6
Harada et al. (2004)	γ –α activation	169±6 b		157±7

[a] Corrected values from Harada et al. (2006).

[b] Corrected values when using the recommended values of (38.5±0.4)% (Singh and Tuli, 2005) for the 312-keV γ-ray and (25.19±0.21)% for the 984-keV γ-ray (Chukreev et al., 2002).

However, there is discrepancy between our value and those obtained using γ-activation technique. This discrepancy can be reduced for some of them by using the recent evaluated γ-ray emission probabilities. Indeed, Kobayashi et al. used the 312-keV γ-ray emitted from ^{233}Pa to determine the amounts of ^{237}Np in the sample with an intensity value of 37.0 %. Katoh et al. did the same but with a value of (38.6±0.4)% that is close to the recommended value of (38.5±0.4) % proposed by Singh and Tuli (2005). Jurova et al. did not used this γ-ray but the 86 keV one. Harada et al. (2006) have used a value of (41.6±0.9) % to determine the amount of ^{237}Np. This value, which is much higher than the recent measured ones, was determined is that experiment and used to correct the three other γ-measurements (see Table 6). Here is a brief summary of the existing values of the emission probability of the 312-keV γ-ray. In the framework of the EUROMET project (No. 416) this emission probability was recently measured. Three independent results were reported: (38.7±0.4) % by Woods et al. (2000), (38.5±0.4) % by Schötzig et al. (2000), and (37.8±0.6) % by Luca et al. (2000). More recently Shchukin et al. (2004) have reported a value of (37.5±0.24) %, and older measurements give values of (38.6±0.5) % for Gehrke et al. (1979) and (38±4) % for Berdikov et al. (1964). All these values are in agreement with the one used in the Katoh's experiment and we believe that the value obtained by Harada et al. is overestimated.

If we correct the cross sections obtained in the γ-activation experiences, as Harada et al. did, by using the recommended values of (38.5±0.4) % from Singh and Tuli (2005) for the 312-keV γ-ray and (25.19±0.21) % from Chukreev et al. (2002) for the 984-keV γ-ray, we found the values indicated in Table 6. A very good agreement is observed between our value and the Kobayashi's one and a compatibility with the Jurova's one whereas the discrepancy with Katoh and Harada still exist. We do not have any interpretation at this time for this discrepancy but it probably shows the importance to take care of the first resonance when extracting the cross section.

6. Conclusion

We used a combination of α- and γ– spectroscopic techniques to analyze an irradiated ^{237}Np sample in a quasi-pure Maxwellian neutron flux. From this measurement we deduce the absolute emission probabilities of the three most intense γ-rays from ^{238}Np. The obtained intensities agree within 1% with the existing data and with the recommended values of Chukreev et al. (2002). Then we used these new intensities, to determine the capture cross section of the ^{237}Np, one of the main contributors to the long-term radiotoxicity of nuclear waste. We found a value which is in good agreement with previous measurements based on α-spectroscopy technique and with

some of the recent γ-spectroscopy techniques after correction of the γ-intensities from ^{233}Pa and ^{238}Np with the recommended values. There is still a discrepancy with the two most recent measurements done in the Kyoto University Reactor by using the pneumatic tube facility. Finally, our value is in agreement with JEFF3.1 and about 11% higher than the value present in the latest releases of ENDF and JENDL nuclear data libraries. Since these two libraries seem to have adopted the same evaluation, a new evaluation should be envisaged.

Acknowledgements

The authors would like to thank the personnel of ILL for their technical support. Dr. AlMahamid is grateful to Dr. David Shuh for his kind help and the Lawrence Berkeley National Laboratory for providing her with a Visiting Scientist status.

References

AlMahamid, I., Shaughnessy, D., Sudowe, R., Proceedings of Nuclear fission and Fission-product spectroscopy, Cadarache, 11-14 May 2005, AIP Conference Proceeding, 798, Melville, New York, 157.
Berdikov, V.V., Silantev, A.N., 1964, Izv Acad. Nauk. SSSR, Ser. Fiz., 28, 394.
Brown, F., Hall, G.R., 1956, J. Inorg. Nucl. Chem. 2, 205.
Chang, Y., Zhu, B., Yan, C., Shi, G., 1990, Chin.J.Nucl.Phys. 12, No 1, 65.
Chukreev, F. E., Makarenko V. E., Martin M. J., 2002, Nuclear Data Sheets 97, 129.
Eberle, S.H., Bleyl H.J., Gantner E., Reinhardt J., Krueckeberg, C., 1971, Wirkungsquerschnitte, KFK 1456, 51.
Gehrke, R.J., Helmer, R.G., Reich, C.W., 1979, Nucl. Sci. Eng., 70, 298.
Harada, H., Nakamura S., Ohta M., Fujii T., Yamana H., 2006, J. Nucl. Sci. Technology, 43, 1289.
Jurova, L.N. Poljakov V.P., Rukhlo V.P. et al., 1984, Vop. At.Nauki i Tekhn.,Ser.Yadernye Konstanty, 1/55, 3.
Katoh, T., Nakamura K., Furutaka K., et al., 2003, J. Nucl. Sci. Technology 40, 559.
Kobayashi, K., Yamanaka, A., Kimura, I., 1994, J. Nucl. Sci. Technology 31, 1239.
Lederer, C.M., 1981, Phys. Rev. C24, 1175.
Luca, A., Etcheverry, M., Morel, J., 2000, Appl. Radiat. Isot., 52, 481.
Mughabghab, S., 2006, Atlas of Neutron resonance, 5th Ed, Elsevier.
Marie, F., Letourneau, A. et al., 2006, Nucl. Instr. and Method A556, 547.
Noguere, G., Bernard, D., De Saint Jean, C., Iooss, B., Gunsing, F., Kobayashi, K., Mughabghab S.F., Siegler, P., 2007, Nucl. Science and Engineering 160, 108-122.
Rengan, K., Devries, D., Griffin, H., 2006, Nucl. Instr. And Method A565, 612.
Schötzig, U., Schönfeld, E., Janszen, H., 2000, Appl. Radiat. Isot., 52, 883.
Schuman, R.P. and Berreth, J.R., 1969, Prog. Report of Idaho Nuclear Corp, IN-1296.
Shcherbakov, O., Furutaka K., Nakamura S., et al., 2005, J. Nucl. Sci. Technology 42, 135.
Shchukin, G., Iakovlev, K., Morel, J., 2004, Appl. Radiat. Isot., 60, 239.
Singh, B., Tuli, J.K., 2005, Nuclear Data Sheets 105, 109.
Smith, M.S., Smith, R.R., Joki, E.G., Evans, J.E., 1957, Phys. Rev. 107, 525.
Tattersall, R.B., Rose, H., Patenden, S.K., Jowitt, D., 1960, J. Nucl. Energy Part A: Reactor Sci., 12, 33.
Tuli, J.K., 2003, Nuclear Data Sheets 100, 347-481.
Weston, L.W. and Todd , J.H., 1981, Nuclear Science and Engineering 79, 184.
Winter, W.J.B., Wapstra, A.H., Goudsmit, P.F.A., Konijn, J., 1972, Nucl. Phys. A197, 417.
Woods, S.A., Woods, D.H., de Lavison P., Jerome, S.M., Makepeace, J.L., Woods, M.J., Husband, L.J., Lineham, S., 2000, Appl. Radiat. Isot., 52, 475.