PROCEEDINGS OF SCIENCE

Measurements of Np-237 incineration in ADS setup QUINTA

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Np-237 samples were irradiated in spallation neutrons produced in ADS setup QUINTA. Three experiments were carried out. The accelerated beam consisted of deuteron ions of energy 2, 4 and 8 GeV respectively. The method was based on gamma-ray spectrometry measurement. During analysis of the spectra several fission products and one actinide were identified. Fission product activities gave the number of fissions. The actinide (Np-238), a result of neutron capture by Np-237 gave the number of captures.

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Introduction

The actinides is a name of a series of 15 chemical elements of atomic number from 89 (actinium) to 103 (lawrencium). The two of them – uranium and plutonium – called mayor actinides are basic components of the nuclear reactor fuel. Of the remaining the three – neptunium, americium and curium (called minor actinides) – are byproducts of energy production in reactor resulting in effect of parasitic neutron capture. About 10-20% of fissionable isotopes are transmuted in reactor into minor actinides. As they are long lived and in typical nuclear power reactor the parasitic neutron capture prevails, it is difficult to incinerate them. They accumulate. At present, approximately 2500 tons of spent fuel containing about 25 tons of plutonium and 3.5 tons of the "minor actinides" (MAs) neptunium, americium, and curium, as well as 3 tons of long-lived FPs (out of a total of about 100 tons of FPs) are produced annually in the European Union [1]. Sooner or later comes the time for mankind to face the problem of active actinides. Np-237 is a typical minor actinide. Fig. 1 shows its fission and neutron capture dependence on energy. It is seen that only for neutrons of energy above 1 MeV the fission prevails the capture. QUINTA setup is a kind of ADS-type spallation neutron source made of natural uranium surrounded by lead shield. It is located in Dubna, Russia in Joint Institute for Nuclear Research. Np-237 samples were irradiated in it and resulting gamma activity was measured then. The main result of this work is fission to capture ratio for Np-237.

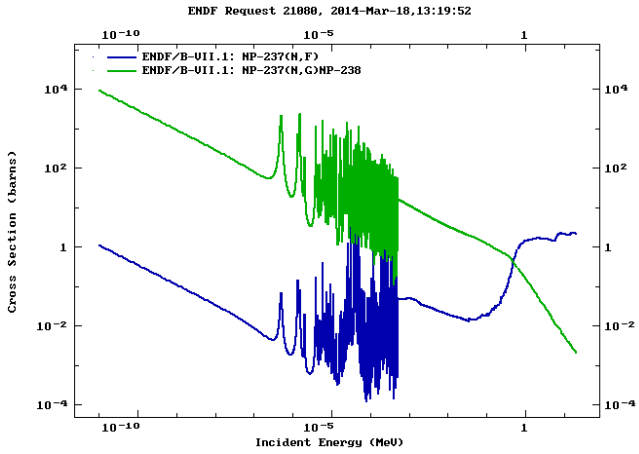

Figure 1. Np-237 neutron caused fission (dark blue) and capture (green) cross section dependence on energy [2].

1. Experiment description

Beams of deuterons provided by the NUCLOTRON Accelerator impinged on uranium assembly QUINTA. Spallation of uranium nuclei was initiated. The QUINTA setup is shown below – Fig. 2. More details on QUINTA can be found in [3]. Actinide samples were located in the "window" marked on red, seen on the right drawing. Beam run is shown on Fig. 3 and some more data in Table 1.

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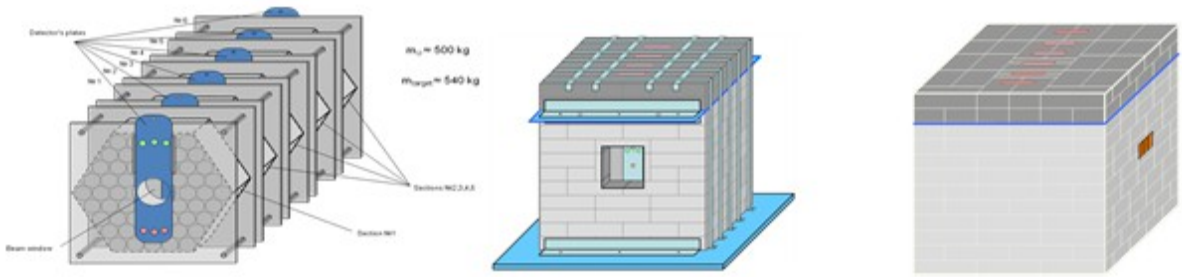

Figure 2. QUINTA setup view - internal core, front view and rear view.

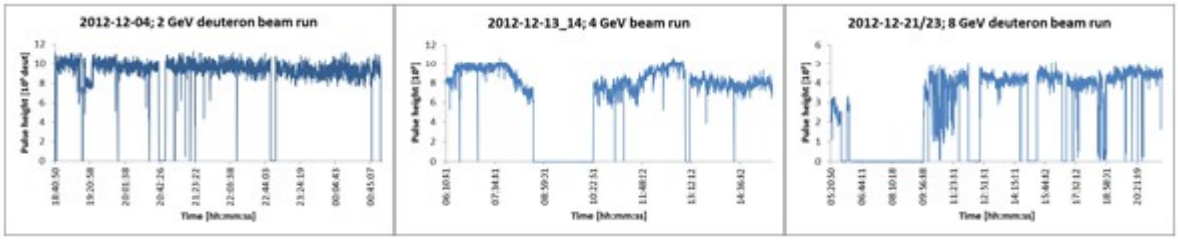

Figure 3. Deuteron beam run during 2, 4 and 8 GeV energy experiments

Each Np-237 sample had cylinder form of 21 mm diameter encapsulated in U-shaped casing. The sample activity was measured tenfold at least at the CANBERRA GR1819 spectrometer. The spectroscopy filters made of Pb, Cd and Cu were used to reduce X and low gamma activities.

Table 1. December 2012 experiments data [4]

Beam energy	**2 GeV**	**4 GeV**	**8 GeV**
Date	04 Dec 2012	13 Dec 2012	22 Dec 2012
Irradiation time (h)	6.27	9.35	16.17
Total number of deuterons (10^{13})	3.052(9)	3.569(15)	1.390(8)

2. Experimental data work-out details

Gamma spectra were analyzed with DEIMOS program [5]. Identified peak area was corrected then for cooling time, irradiation time, detector efficiency and fission product yield to get *Np-237* fission rate $I_{f\gamma}$.

$$I_{f\gamma} = \frac{W_1}{W_2} \cdot C_{IH} \tag{1}$$

$$W_1 = \frac{100 \cdot S_\gamma}{\varepsilon_p \cdot I_\gamma} \cdot \frac{1}{\left(1 - e^{-\lambda t_{real}}\right)} \cdot \frac{t_{real}}{t_{live}}$$

$$W_2 = \frac{m \cdot \phi \cdot \gamma_f \cdot \left(1 - e^{-\lambda \cdot t_{ir}}\right)}{100 \cdot \lambda \cdot t_{ir}} \cdot e^{-\lambda t_+}$$

$I_{f\gamma}$ – actinide fission rate, per deuteron and per gram
γ – gamma line index
f – reaction index (f = fission)
S_γ – gamma peak area
γ_f – isotope production yield [%]

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m – activation sample mass [g]
γ_p – gamma spectrometer efficiency
I_γ – gamma line intensity [%]
ϕ - deuteron integral number
λ – isotope decay constant
t_+ – cooling time
t_{ir} – irradiation time
t_{real} – real time of measurement
t_{live} – live time of measuremet
C_{IH} – correction for irradiation history

Formula for neutron capture rate was very similar except the γ_f correction omitted.

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Some decaying isotopes are produced in two ways – directly from Np-237 fission and indirectly, as a result of other the fission products decay chain. Te-132 → I-132 decay chain is a very illustrative example of this case [6].

$$\left.\begin{array}{l} Fission \xrightarrow{\gamma_1} {}^{132}_{52}Te \xrightarrow{\beta^-\begin{pmatrix}3.204d\\228.16keV\,88\%\\49.72keV\,15\%\end{pmatrix}} {}^{132}_{53}I \xrightarrow{\beta^-\begin{pmatrix}2.295h\\667.718keV\,99\%\\772.6keV\,75.6\%\\954.55keV\,17.6\%\end{pmatrix}} \\ \qquad\qquad\qquad\qquad Fission \xrightarrow{\gamma_2} {}^{132}_{53}I \xrightarrow{\beta^-\begin{pmatrix}2.295h\\667.718keV\,99\%\\772.6keV\,75.6\%\\954.55keV\,17.6\%\end{pmatrix}} \end{array}\right\} {}^{132}_{54}Xe \quad (2)$$

In fact Te-132 isotope can have several predecessors but their life time is so short that their contribution can be included into γ_1 making it a cumulative fission yield while γ_2 is an individual one. Based on [7] fission yield data one has γ_1=3.98% and γ_2=0.407%.

The above decay chain specificity is that I-132 half life time (2.29h) is much shorter than the Te-132 one (3.204d). It causes the I-132 lines (667.71, 772.6 and 954.55 keV) to decay not according to 2.295h but 3.204d half life time. Therefore W_2 expression in (1) gets for I-132 lines changed to:

$$W_2 = m \cdot \phi \cdot \left(\frac{\gamma_1 \cdot \left(1 - e^{-\lambda_1 \cdot t_{ir}}\right)}{100 \cdot \lambda_1 \cdot t_{ir}} \cdot e^{-\lambda_1 t_+} + \frac{\gamma_2 \cdot \left(1 - e^{-\lambda_2 \cdot t_{ir}}\right)}{100 \cdot \lambda_2 \cdot t_{ir}} \cdot e^{-\lambda_2 t_+} \right) \quad (3)$$

The spectrometer efficiency curve was based on Co-60, Ba-133, Cs-137, Eu-152 and Th-228 calibration sources activity measurements. The detector efficiency (ε_p) curve is shown on Fig. 4.

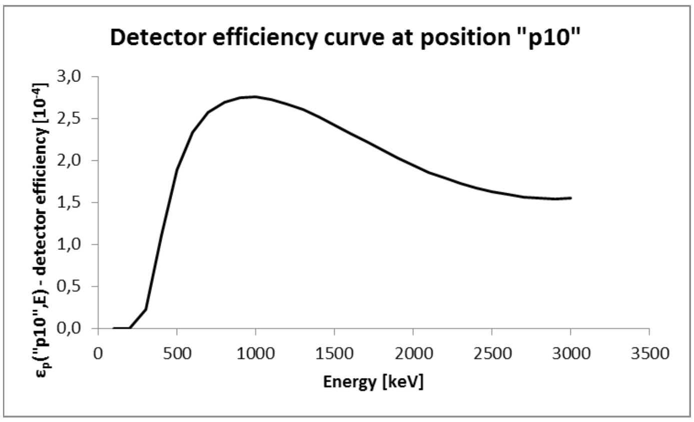

Figure 4. HPGe detector efficiency dependence on energy

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Looking at Fig. 3 one can see beam run or irradiation history. During experiment 2 (4 GeV) and 3 (8 GeV) there was quite a big time interval with no beam. The deviation of real beam run from the "flat" one was corrected by formula:

$$C_{IH} = \left(\sum_{i=1}^{ir} \varphi_i e^{-\lambda(t_{ir}-t_i)} \right) \frac{\lambda t_{ir}}{\phi\left(1-e^{-\lambda t_{ir}}\right)} \tag{4}$$

Here φ_i means number of deuterons in *i-th* beam shot (pulse), ϕ total number of deuterons, i numbers the beam shots. The correction equals 1 for experiment 1 (2 GeV), mostly 1 but 1.02 for I-135 and 1.07 for Sr-92 for experiment 2 (4 GeV) and much bigger for experiment 3 (8 GeV) where it reaches 1.21 and 1.43 for I-135 and Sr-93 respectively.

3. Results

There are two sources of gamma quanta – fission product (FP) activity and neutron capture product (CP) activity. The identified products and their data are shown in Table 2.

Table 2. Sample Np-237 identified gamma lines and their data

E-gamma	Isotope	Source	T1/2	Fission yield [%] [7]	I-gamma [%] [6]
529.87	^{133}I	FP	20.87h	4,45	87
657.94	^{97}Zr->^{97}Nb*	FP	16.744 h	5,38	98,23
667.71	^{132}Te->^{132}I**	FP	3.26d	4,39	98,7
743.36	^{97}Zr	FP	16.744 h	5,35	93,6
772.6	^{132}Te->^{132}I**	FP	3.26d	4,39	75,6
954.55	^{132}Te->^{132}I**	FP	3.26d	4,39	17,6
1131.51	^{135}I	FP	6.57h	4,16	22,6
1260.41	^{135}I	FP	6.57h	4,16	28,7
923.98	^{238}Np	CP	2.117d	N/A	2,869
962.77	^{238}Np	CP	2.117d	N/A	0,702
984.45	^{238}Np	CP	2.117d	N/A	27,8
1025.87	^{238}Np	CP	2.117d	N/A	9,65
1028.54	^{238}Np	CP	2.117d	N/A	20,38

FP – fission product. CP – neutron capture product.

*Line 657.94 keV stems in fact from Nb-97 beta decay, but its quantity is modified by Zr-97 decay rate [6,7], (16.8h).

Isotope	T1/2	Y-individual [%]	Y-cum [%]
^{97}Y	3.76s	3.22	4.25
^{97}Zr	16.8h	1.09	5.35
^{97}Nb-m	58.1s	3.83E-03	5.03
^{97}Nb	1.23h	3.10E-02	5.38

**Lines 667.71, 772.6 and 954.55 keV stem from I-132 but their activities are modified by Te-132 decay rate [6,7], (3.26d).

Isotope	T1/2	Y-individual [%]	Y-cum [%]
^{132}Sb	4.2m	5.59E-01	6.00E-01
^{132}Te	3.26d	2.47	3.98
^{132}I	2.28h	4.07E-01	4.39
^{132}Xe	stable	7.10E-03	4.40

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The results of measurements are shown on Fig. 5 grouped by experiment (rows) and fission, capture (columns). No data were rejected. They were treated as equivalent members of the same population and their average value is an arithmetical mean while the error, σ is a standard deviation of the population. The average values are shown on Fig. 6 and Table 3, while fission to capture ratio on Fig. 7 and Table 4.

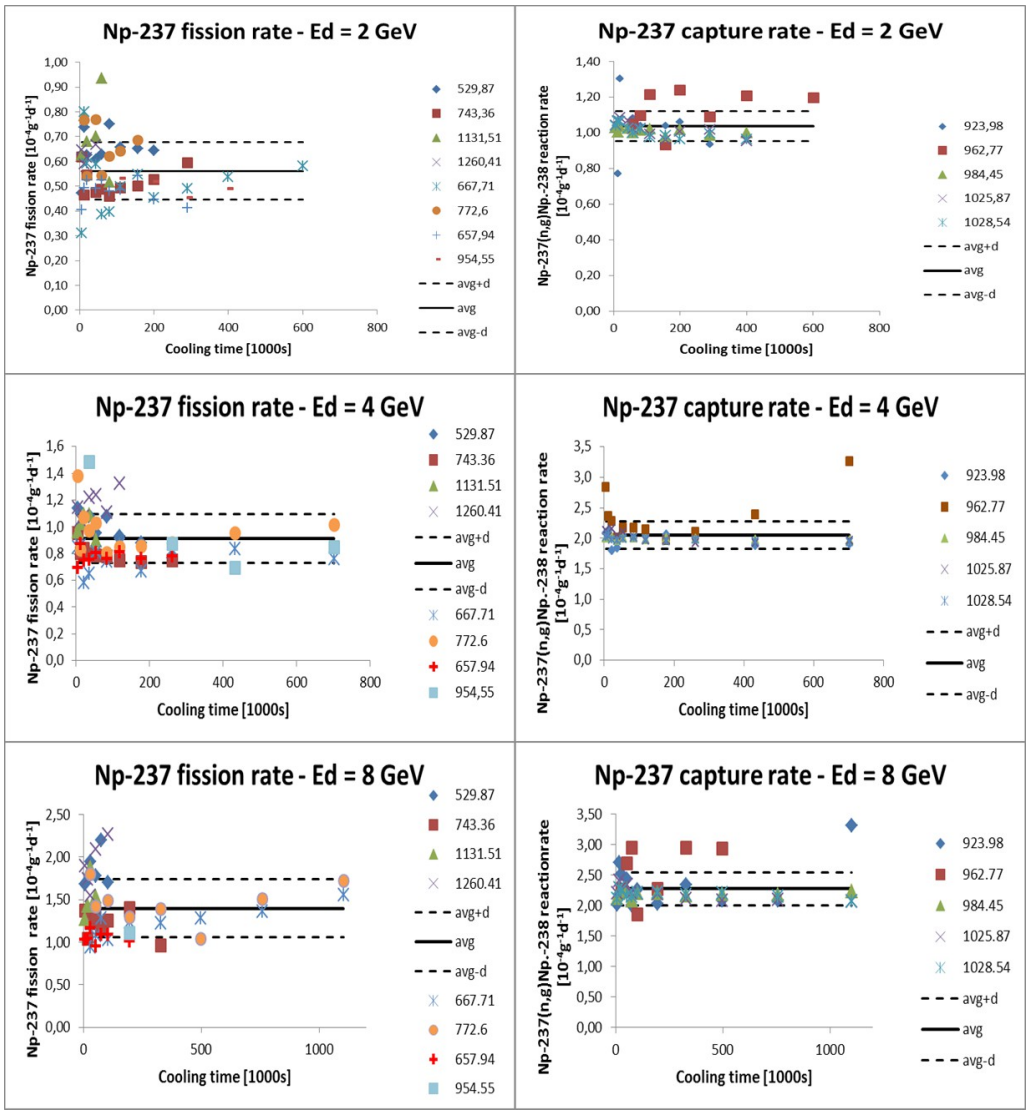

Figure 5. Np-237 fission and capture rate based on identified gamma lines for beam deuteron energies 2, 4 and 8 GeV. Solid and dashed lines show ±σ (standard deviation of population) range.

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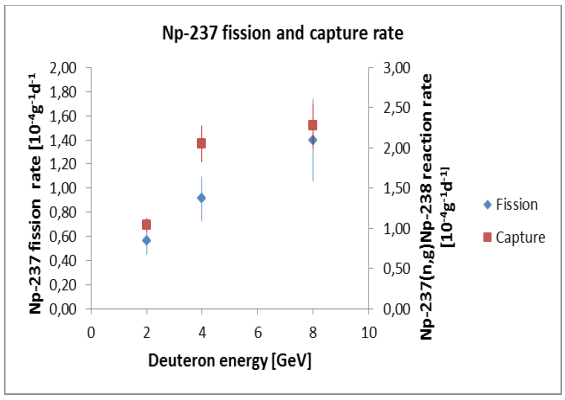

Figure 6. Np-237 average fission and capture rate per gram and per 1 deuteron dependence on energy

Table 3. Np-237 fission and capture rate per sample gram and per 1 deuteron

Beam deuteron energy [GeV]	Fission rate [$10^{-4}g^{-1}d^{-1}$]	Standard deviation [$10^{-4}g^{-1}d^{-1}$]	Standard deviation [%]	Capture rate [$10^{-4}g^{-1}d^{-1}$]	Standard deviation [$10^{-4}g^{-1}d^{-1}$]	Standard deviation [%]
2	0.562	0.115	20.41	1.04	0.084	8.1
4	0.913	0.181	19.84	2.05	0.226	11.04
8	1.40	0.339	24.28	2.27	0.270	11.89

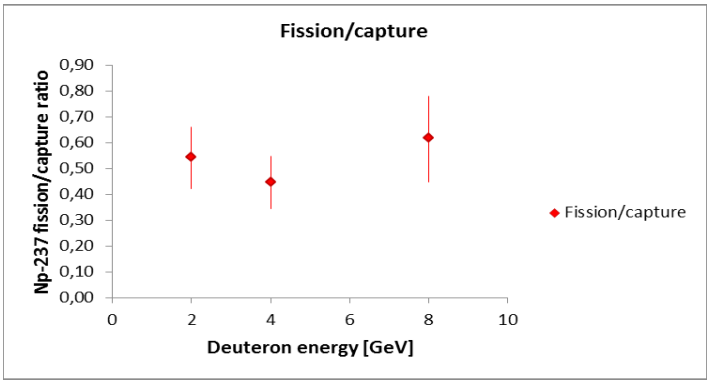

Figure 7. Np-237 fission to capture ratio dependence on beam deuteron energy

Table 4. Np-237 fission/Capture ratio dependence on beam deuteron energy

Beam deuteron energy [GeV]	Fission/Capture ratio	Standard deviation	Standard deviation [%]	Fission/absorption ratio	Standard deviation	Standard deviation [%]
2	0.541	0.119	21.96	0.35	0.08	22.26
4	0.445	0.101	22.70	0.31	0.07	22.12
8	0.615	0.166	27.03	0.38	0.10	27.00

Based on these data fission/capture seems not to depend on energy, what suggests the neutron

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spectrum not to depend on energy as well. To compare these data with literature [1] – Fig. 8 - fission to absorption ratio is given in Table 4 as well. The comparison says the QUINTA setup to be about 1.4 times more effective in Np-237 incineration than Sodium-cooled Fast Reactor (SFR). Nevertheless the neutron capture is still about two times higher than the fission (incineration) what means the trans-Np-237 actinides cumulate.

4. Conclusions

The fission/capture ratio seems to be constant in the explored deuteron energy range (2 – 8 GeV), of order 0.53 while fission to absorption ratio is of order 0.35.

Comparison of the determined fission to absorption ratio with literature data says the QUINTA setup to be about 1.4 times more effective in Np-237 incineration than Sodium-cooled Fast Reactor (SFR). Nevertheless the incineration does not prevent the trans-Np-237 actinides accumulation.

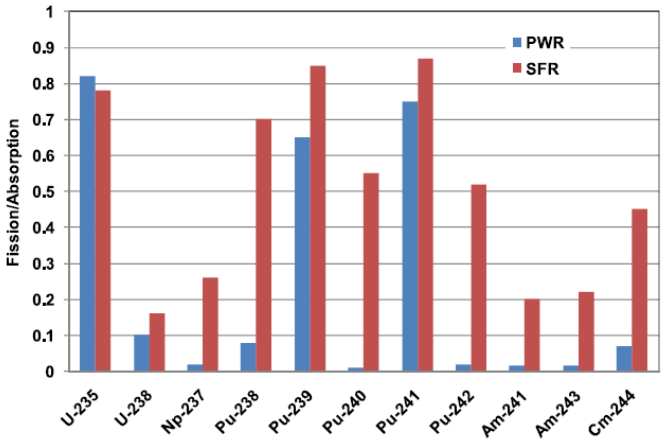

Figure 8. Actinides fission to absorption ratio for PWR and SFR [1]

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