

Np-237 transmutation efficiency dependence on beam particle, energy and sample position in QUINTA setup

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Abstract. Np-237 samples were irradiated with spallation neutrons produced at the ADS setup QUINTA. Six experiments were carried out at the JINR, in Dubna – one in carbon (C6+), three in deuteron, and two in proton beams. The energy in carbon was 24 GeV, in deuteron – 2, 4 and 8 GeV, respectively, and 660 MeV in the proton beam. In five cases the sample was located in a side window in a lead shield. In one case (660 MeV proton beam) two samples were located on the top of the QUINTA setup, one – on the top of section 2, and the second one – on the top of section 4. The transmutation study method was based on gamma-ray spectrometry. During the analysis of the spectra several fission products and one actinide were identified. Fission product activities yielded the number of fissions. The actinide (Np-238), a result of neutron capture by Np-237, yielded the number of captures.

The main goal of this work was to find out if and how the transmutation rate depended on the accelerator beam and sample location during the irradiation.

1 Introduction

Neptunium is one of 15 actinides produced artificially in power reactors as a by-product of energy production. Np-237 is its longest-lasting isotope with a half-life of $2.144 \cdot 10^6 y$ [1]. To eliminate its long-lasting activity, one has to split it somehow. There are two opportunities of neutron interactions with Np-237 – fission and radiative capture. Np-237 nuclear fission leads to the production of two new isotopes of masses statistically dispersed around two maxima. Some isotopes, also known as fission products (FP), are produced directly from Np-237 fission and the others – indirectly, as a result of the other mode of the fission product decay. The radiative neutron capture yields β -active Np-238 nuclei.

As shown in Fig. 1, the radiative neutron capture is a dominant way of interaction with neutrons in the energy range of 0–1 MeV. This is particularly important since the thermal region of neutron energy is the most available in present-day power reactors. Neutron capture induces the production of other actinides. The newly produced actinides affect the neutron balance so much that a fuel campaign is shortened. A neutron energy region exists at approximately 1 MeV and above where the fission of Np-237 dominates the radiative capture. To prevent/eliminate long-lasting activity/actinides, one needs a fast neutron source like a fast reactor or the accelerator-driven system (ADS).

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The Np-237 incineration results presented here are taken from experiments carried out at the ADS-type setup QUINTA in JINR, Dubna, Russia. Tab. 2 shows the data of each experiment. Some results presented in this work – the ones from deuteron beams – have already been published in [2]. Some other results, the ones from a 660 MeV proton and a C6+ beams have already been published in [3]. The results presented here are extended by another experiment with a 660 MeV proton beam where the QUINTA setup was configured without a lead shield.

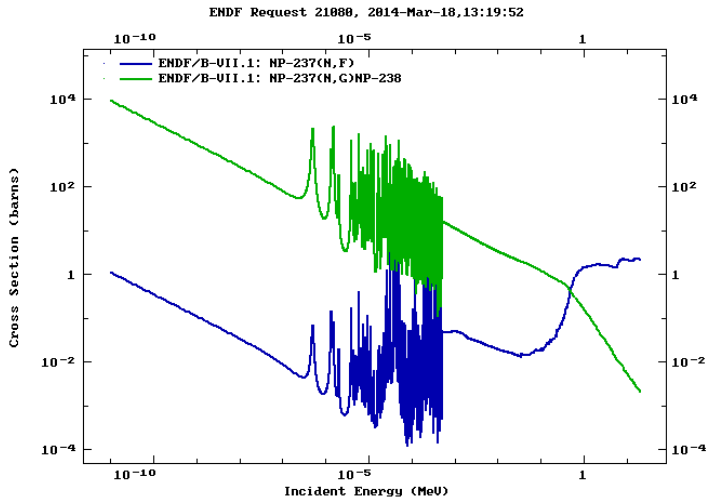


Figure 1. Np-237 neutron-caused fission (dark blue) and capture (green) cross section dependence on energy [5]

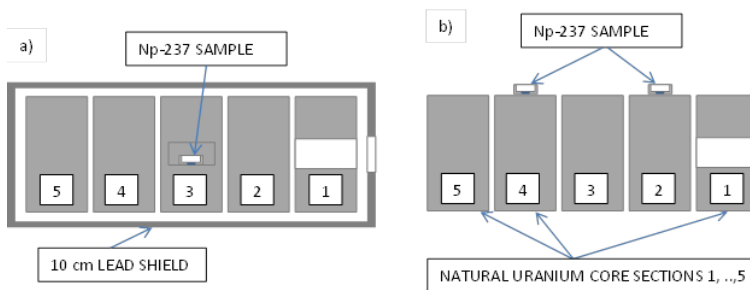


Figure 2. QUINTA setup vertical-axial cross section – configured with lead shield(a) and without it (b)

2 Experiment and data description

The core of the QUINTA setup (Fig. 2) consisted of aluminum-cladded elements of natural uranium arranged in five hexagonal sections. The total weight of the uranium was 500 kg. The uranium core was surrounded by a 10 cm-thick lead shield. For more details of the QUINTA setup construction see [4]. An accelerated beam of carbon ions (C6+), deuterons

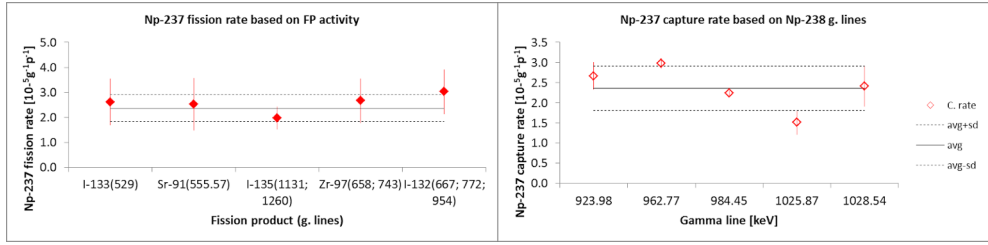


Figure 3. Np-237 fission and capture rate results (experiment 0.66 GeV/p/TS2)

or protons impinged on the uranium core causing nuclei spallation and the production of fast neutrons. The Np-237 sample was located in a side window about 20 cm from the axis of the beam. The spallation neutrons caused U-238 fission which generated a number of neutrons while converting into the Np sample. The Np-237 sample was formed of a disk 21 mm in diameter encapsulated in a U-shaped casing. The gamma spectrum of the sample was measured several times by a CANBERRA GR1819 spectrometer. The spectroscopy filters were made of Pb, Cd and Cu to reduce X-ray and low-energy gamma activity. Altogether it was composed of a 11.5 mm-thick Pb filter, in addition to 1 mm of Cd and 1 mm of Cu. Then, the following gamma spectra analysis aimed to identify gamma peaks as well as calculate their areas and parent isotopes. Co-60, Ba-133, Cs-137, Eu-152 and Th-228 samples were used as calibration ε_p sources for gamma spectrometer efficiency, ε_p . Generally some peaks stem from Np-237 fission products and the others from the neutron capture product, Np-238. The final measurements identified the number of Np-237 nuclei fissions and nuclei which absorbed neutrons. They were simply calculated from corresponding peak areas according to the formula:

$$R_{f\gamma} = \frac{S_\gamma}{m \cdot \phi \cdot \gamma_f \cdot \varepsilon_p \cdot I_\gamma} \cdot \frac{\lambda \cdot t_{irr}}{(1 - e^{-\lambda \cdot t_{irr}})} \cdot \frac{e^{\lambda \cdot t_+}}{(1 - e^{-\lambda \cdot t_{real}})} \cdot \frac{t_{real}}{t_{live}}, \quad (1)$$

$R_{f\gamma}$ – actinide fission rate, per beam particle and per gram,

γ – gamma line index,

f – reaction index ($f = \text{fission}$),

S_γ – gamma peak area,

γ_f – isotope production yield, % [5],

m – activation sample mass, g,

ε_p – gamma spectrometer efficiency,

I_γ – gamma line intensity, %,

ϕ – (deuteron/proton/C6+) integral number,

λ – isotope decay constant,

t_+ – cooling time,

t_{irr} – irradiation time,

t_{real} – real time of measurement,

t_{live} – live time of measurement.

In the simplest case the isotopes are produced directly from fission (like Sr-92, I-133 and I-135) or capture, and the beam intensity is constant. In the case of capture the formula is very similar to Eq. (1) except for the omitted γ_f coefficient.

There are some lines – like 658, 667, 772.6 and 954 keV – which behave in a more complicated way described in [3]. Tab. 1 shows all identified gamma lines and their data (isotope

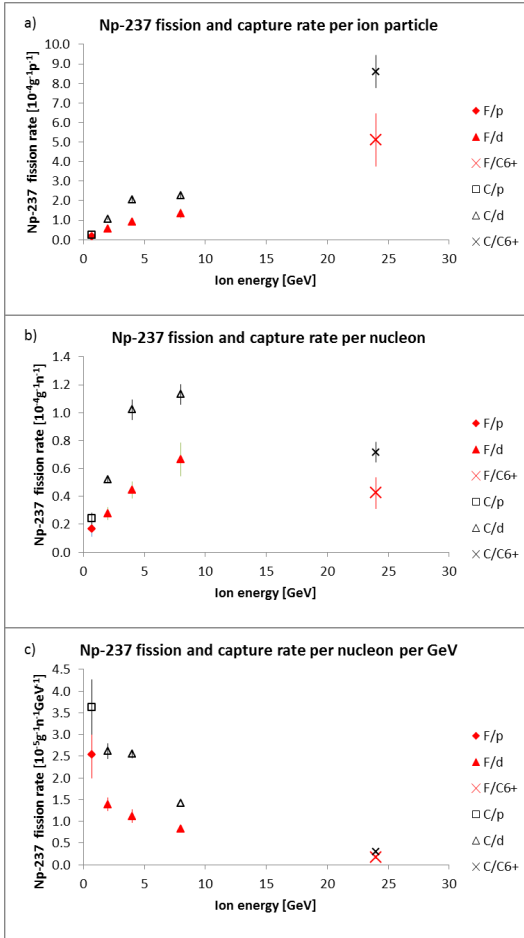


Figure 4. Np-237 fission and capture rate results and their successive recalculation for samples located in left side window (LSW) – a) – “bare” results (per sample gram and one beam particle), b) – per sample gram, one beam particle and one (ion) nucleon, c) – per sample gram, one beam particle, one (ion) nucleon and ion energy unit. Legend – reaction/particle – for example F/p – fission/proton

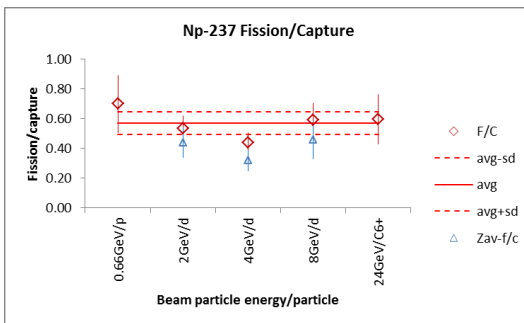


Figure 5. Np-237 fission-to-capture ratio for each experiment for samples located in the left side window (LSW)

source, half-life, intensity and corresponding FP yield). Data averaging was performed over two steps – averaging over measurements for each isotope and then over isotopes or gamma lines in the capture case (Fig. 3).

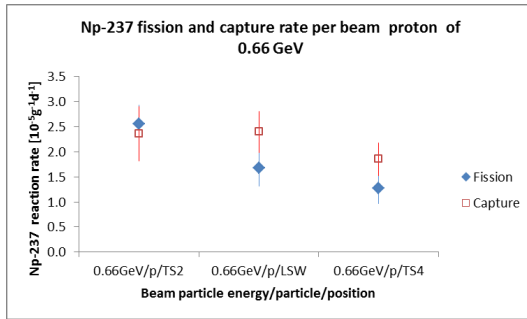


Figure 6. Np-237 fission and capture rate results for 0.66 GeV proton beam dependence on sample location - for samples located on top of QUINTA's core (TS2, TS4) and in the left side window (LSW)

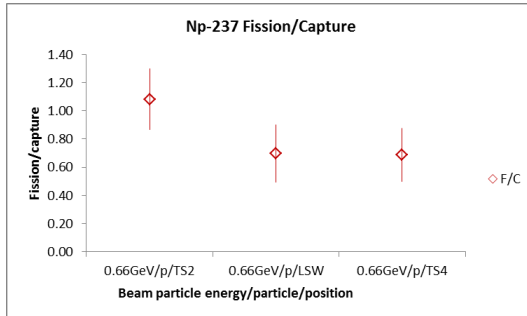


Figure 7. Np-237 fission-to-capture ratio for 0.66 GeV proton beam dependence on sample location - for samples located on top of QUINTA's core (TS2, TS4) and in the left side window (LSW)

Table 1. Sample Np-237-identified gamma lines and their data. FP – fission product. CP – neutron capture product. (*) Nb-97 quantity and decay rate are modified by its parent isotope Zr-97. (**) I-132 quantity and decay rate are modified by its parent isotope Te-132

E-gamma	Isotope	Source	$T_{1/2}$	Fission yield, % [4]	I-gamma, % [5]
529.87	^{133}I	FP	20.87h	4.45	87
658.08	$^{97}\text{Nb}^*$	FP	16.744h	5.38	98.23
667.71	$^{132}\text{I}^{**}$	FP	3.26d	4.39	98.7
743.36	^{97}Zr	FP	16.744h	5.35	93.6
772.60	$^{132}\text{I}^{**}$	FP	3.26d	4.39	75.6
954.55	$^{132}\text{I}^{**}$	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
1383.93	^{92}Sr	FP	2.66h	4.01	90
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

Table 2. Experimental data

Beam energy/particle*	0.66 GeV/p	0.66 GeV/p	2 GeV/d	4 GeV/d	8 GeV/d	24 GeV/C6+
Date	22/06/17	08/11/14	04/12/12	13/12/12	22/12/12	18/12/13
Irradiation time (h)	5.25	5.72	6.27	9.35	16.17	22.8
Total number of beam particles	$7.78(9) \times 10^{14}$	8.64×10^{14}	$3.052(9) \times 10^{13}$	$3.569(15) \times 10^{13}$	$1.390(8) \times 10^{13}$	1.75×10^{11}
Sample position	Top of section 2	Left side window	Left side window	Left side window	Left side window	Left side window
	Top of section 4	Left side window	Left side window	Left side window	Left side window	Left side window

Table 3. Basic Np-237 incineration parameters for each experiment. (*) Particles: p – proton, d – deuteron, C6+ – carbon. (**) Sample positions: TS2 – top of section 2, TS4 – top of section 4, LSW – left side window in lead shield

Avg. fission and capture	Particle energy/particle*/sample position**						
	0.66 GeV/p/TS2	0.66 GeV/p/LSW	0.66 GeV/p/TS4	2 GeV/d/LSW	4 GeV/d/LSW	8 GeV/d/LSW	24 GeV/C6+/LSW
fission ($10^{-5} \text{ g}^{-1} \text{ p}^{-1}$)	2.56(38)	1.67(35)	1.28(30)	5.56(87)	8.95(12)	13.3(24)	51.1(135)
capture ($10^{-5} \text{ g}^{-1} \text{ p}^{-1}$)	2.36(55)	2.40(42)	1.85(33)	10.10(43)	20.40(14)	22.6(14)	86.1(84)
fission ($10^{-5} \text{ g}^{-1} \text{ p}^{-1} \text{ nucleon}^{-1}$)	2.56(38)	1.67(35)	1.28(30)	2.78(43)	4.47(60)	6.65(12)	4.26(11)
capture ($10^{-5} \text{ g}^{-1} \text{ p}^{-1} \text{ nucleon}^{-1}$)	2.36(55)	2.40(42)	1.85(33)	5.22(22)	10.21(71)	11.31(7)	7.17(7)
fission ($10^{-5} \text{ g}^{-1} \text{ p}^{-1} \text{ nucleon}^{-1} \text{ GeV}^{-1}$)	3.88(58)	2.53(53)	1.93(46)	1.39(22)	1.12(15)	0.83(15)	0.18(5)
capture ($10^{-5} \text{ g}^{-1} \text{ p}^{-1} \text{ nucleon}^{-1} \text{ GeV}^{-1}$)	3.58(83)	3.63(63)	2.81(49)	2.61(11)	2.55(18)	1.41(9)	0.30(3)
F/C	1.08(30)	0.70(19)	0.69(20)	0.53(9)	0.44(7)	0.59(11)	0.59(17)
F/A	0.52(14)	0.41(11)	0.41(12)	0.35(6)	0.30(5)	0.37(7)	0.37(11)

3 Results

The results are presented in a selective way to illustrate some work out stages. Fig. 3 shows the rates of Np-237 fission (left side) and radiative capture (right side). Punctual marks represent the average values for one isotope (FP) or gamma line (capture) and the error value corresponds to their standard deviation, σ . The solid line represents the average value, averaged over all isotopes - the left-hand side case, or gamma lines - the right-hand side one. Dashed lines represent the $\pm\sigma$ (standard deviation) range. Fig. 4 shows the number of Np-237 fission and capture rate results and their successive recalculation for samples located in left side window (LSW) - a) -“bare” results (per sample gram and one beam particle), b) - per sample gram, one beam particle and one (ion) nucleon, c) - per sample gram, one beam particle, one (ion) nucleon and ion energy unit. Legend - reaction/particle - for example F/p - fission/proton. X-axis shows the ion energy in GeV. Fig. 5 illustrates the fission-to-capture ratio (F/C) for each experiment for samples located in the left side window (LSW). Punctual marks represent the value of F/C for one experiment. Blue triangles (Zav-f/c) represent the data derived from [7]. Experiment mnemonics is shown as x-axis symbols. A summary of the results is shown in Tab. 3. The fission-to-capture ratio seems to be the most appropriate parameter for analysis because division eliminates some possible common errors.

Fig. 6 shows Np-237 fission and capture rate results for 0.66 GeV proton beams dependence on sample location - for samples located on the top of QUINTA's core (TS2, TS4) and in the left side window (LSW). The specified positions are considered to be equivalent.

Fig. 7 shows Np-237 fission-to-capture ratio for 0.66 GeV proton beams dependence on the sample location - for samples located on the top of QUINTA's core (TS2, TS4) and in the left side window (LSW).

The results of the five experiments identified which ion and which energy level are the most appropriate for Np-237 incineration, and the general incineration of actinides. Fig. 4 is helpful in this regard. First, the reaction rate must be related to the mass of the sample and to one beam ion particle - Fig. 4a. Then, it must be related to one beam ion particle and to one beam ion nucleon - Fig. 4b. And, finally, it must be related to one ion nucleon and one unit of energy - GeV - Fig. 4c. According to this analysis the proton is the most efficient in Np-237 incineration. Nevertheless, the results from deuteron are disturbing. The efficiency of the proton is approximately twice as big as that of deuteron. It looks like the energy brought into the target by the neutron of deuteron is lost, i.e. the neutron does not produce spallation neutrons or at least produces them much less than the ion particle proton. Intuitively this is acceptable because a proton-target nucleus interaction, the electrical (+ nuclear) one, covers a much larger range than the neutron-nucleus one, the nuclear one. The C6+ ions are much less efficient than the proton and deuteron. The efficiency of deuteron decreases with energy as expected, because the greater the energy is the shorter is the time of flight through the target nucleus. Generally, the incineration potential peaks when the energy of the deuteron beam is equal to 2 GeV.

It is not clear why the capture rate behaves a little differently to the fission one. As shown in Fig. 5, the fission-to-capture ratio deviates from its average value of 0.57(8) by more than statistics allows. According to statistics 68.27% of results must fall within $\pm\sigma$ (2 standard deviations) but only 60% have fallen within this range in this experiment.

As for the 0.66 GeV/p results (Figs. 6 and 7), the dependence on the sample position the results from the top of section 2 (TS2) has shown some increase in comparison to the TS4 and LSW results.

4 Conclusions

Five experiments have been carried out using the QUINTA-ADS-type setup to investigate the incineration potential of Np-237 in the left side window position. The ion beam consisted of protons (0.66 GeV), deuterons (2, 4 and 8 GeV) and carbon C6+ (24 GeV). But, the sixth experiment was carried out to investigate the mentioned potential dependence on the sample position, i.e. the axial position.

Beam ion particle neutrons are considered not to contribute or contribute much less than protons to the incineration of Np-237. The most appropriate parameters of comparison were the number of fissions per gram of sample material, per beam ion particle, per beam ion particle nucleon, and per beam energy unit. The dependence of neutron capture on ion energy is different to that of fission. The fission-to-capture (F/C) and fission-to-absorption ratios are the other parameters which characterize the efficiency of incineration.

Generally the fission-to-capture ratio of Np-237 was less than 1, i.e. actinides still cumulate in QUINTA, with one exception – 0.66GeV/p/TS2 experiment where the fission-to-capture ratio was 1.08.

The fission-to-capture ratio deviates from its expected average value by more than statistics allow. 60% of results fall within $\pm\sigma$ (2 standard deviations) instead of 68.27%.

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