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Comparison of neutron induced fission and capture in Np-237 and Pu-239 irradiated in QUINTA assembly with 660 MeV proton beam

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Abstract. Two Np-237 samples and one Pu-239 were irradiated in spallation neutrons produced in ADS setup QUINTA. The accelerated beam consisted of protons of energy 660 MeV. 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. In a similar manner the number of fissions in Pu-239 was determined. The Pu-240, a product of neutron capture by Pu-239, activity was impossible to measure.

1 Introduction

Np-237 and Pu-239 are produced artificially in a power reactor as a byproduct of energy production. They interact with reactor neutrons in two ways either their nuclei are split in two parts, fission products or they capture the neutrons resulting in production of other actinides. About 10-20% of fissionable isotopes are transmuted in reactor this way. As they are long lived and in a typical nuclear power reactor the parasitic neutron capture prevails, it is difficult to incinerate them. They accumulate. The only way to neutralise them is their fission.

Figure 1 shows Np-237 and Pu-239 fission product yield (the upper part) and fission and neutron capture cross section dependence on energy. It is seen that only for neutrons of energy above 1 MeV the fission prevails the capture. It means that either fast reactor or Accelerator Driven System (ADS) provide neutrons of the most appropriate energy to incinerate the actinides. In an international research project "Energy & Transmutation of Radio-Active Waste" (E&T RAW) 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.

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This work shows the results of Np-237 and Pu-239 incineration measurements. Proton beam accelerated in a PHASOTRON impinged on QUINTA core producing fast neutrons, spallation neutron field. Np-237 and Pu-239 samples were irradiated in it and resulting gamma activity was measured then.

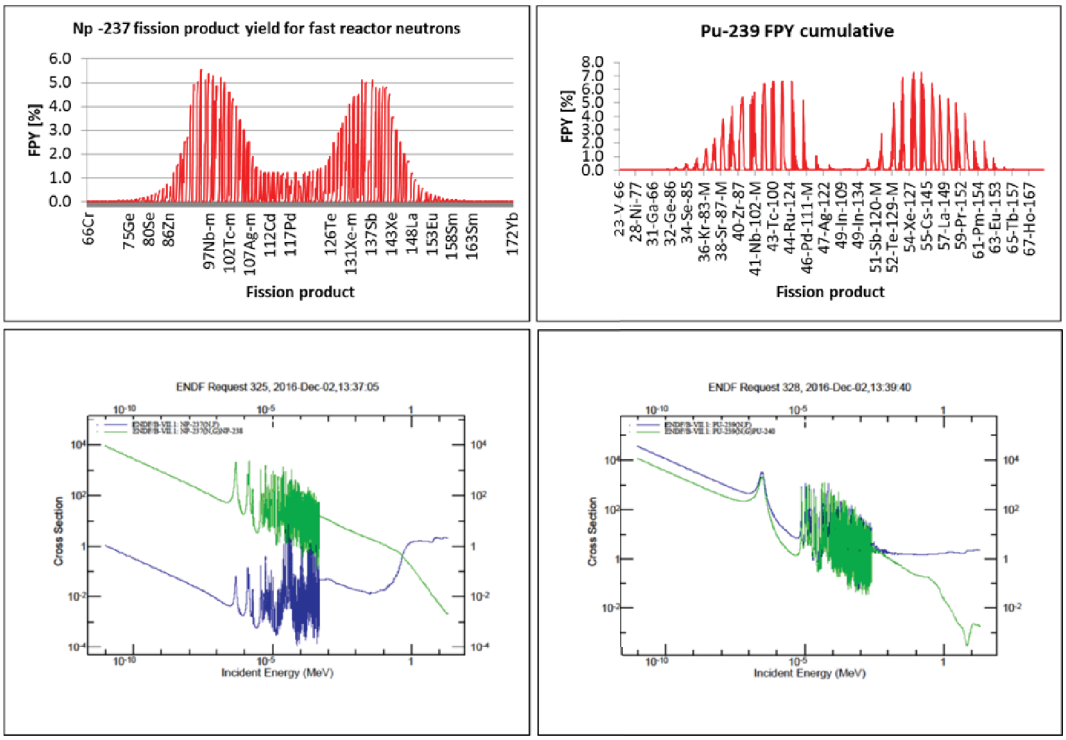

Figure 1. Np-237 (left) and Pu-239 (right) fission product yield (FPY) - upper view and (n,f) and (n,γ) cross section bottom view.

2 Experiment and work out description

The QUINTA setup is shown below - figure 2. Its core contains 500 kg of natural uranium elements cladded in aluminium. More details on QUINTA can be found in [1]. Actinide samples were located in the "window marked" on red, seen on the right drawing.

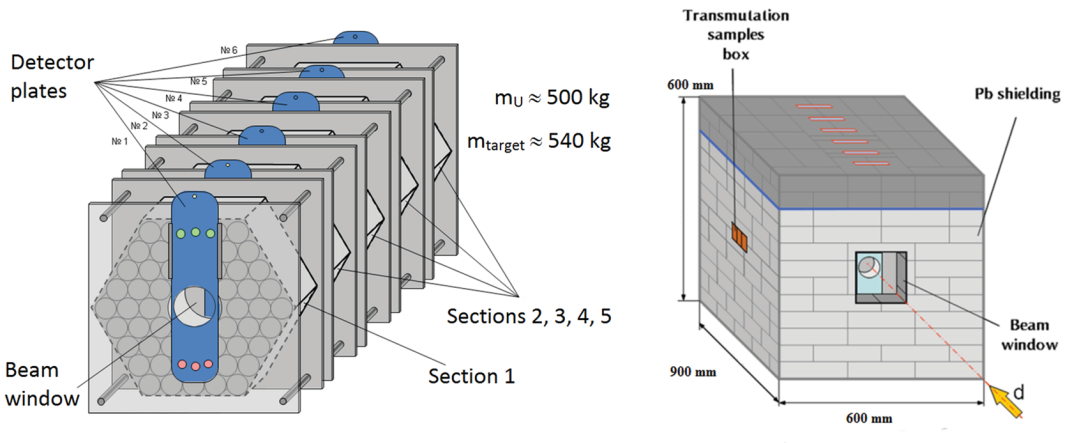

Figure 2. QUINTA setup view - internal core and front view.

Two experiments were carried out, see table 1. HPGE detector was used for gamma spectra registration then. To protect the detector against overload by X and low-energy gammas special filters were applied:

- 1mm Cu, 1 mm Cd and 12 mm Pb for Np-237 sample measurements.
- 1mm Cu, 1 mm Cd and 8 mm Pb for Pu-239 sample measurements.

The spectra were analyzed then with GammaMCA program to get a peak energy and a peak area. The peak energy lets us identify the parent isotope. There are two classes of the parent isotopes, fission products and absorption products.

The found peak areas were corrected for gamma line intensity, detector efficiency, fission product yield, cooling time and detector dead time formula 1. Some isotopes are chained, i.e. the decaying predecessor in chain gives additional rise to its quantity. More complicated formula 2 was used then. The final result was either fission rate, i.e. number off fissions per second, per sample gram and per one proton or capture rate. As concerns Pu-239 both fission and capture take place in neutron field. The main difference from Np-237 is in proportions. The fission prevails capture in Pu-239. In addition, neutron capture product Pu-240 undergoes α decay to U-236 with no gamma emission in 72.8% or with 45.242 keV X-radiation emission in 27.1%. Because of used Pb-filters and low activity caused by long (6563y) half life time it was impossible to determine number of captures in Pu-239.

Table 1. Experiments data

Experiment date	Irradiation duration	Beam	Energy	Fluence	Samples
2014-11-08	6h	Protons	660 MeV	$4.5x10^{15}$	Np-237-4
2015-12-04	5h 43m	Protons	660 MeV	$8.64x10^{14}$	Np-237-4 Pu-239-3

$$R_{f1g} = \frac{A_{1g}}{m\phi\gamma\epsilon_{pg}I_g} \frac{\lambda_1 t_{irr}}{(1 - e^{-\lambda_1 t_{irr}})} \frac{e^{\lambda_1 t_+}}{(1 - e^{-\lambda_1 t_{real}})} \frac{t_{real}}{t_{live}} \quad (1)$$

$$R_{f2g} = \frac{A_{2g}}{m\phi\epsilon_g I_g}\frac{t_{real}}{t_{live}}\frac{t_{irr}}{[c_1(1-e^{-\lambda_1 t_{real}})e^{-\lambda_1 t_+} + c_2(1-e^{-\lambda_2 t_{real}})e^{-\lambda_2 t_+}]}$$
$$c_1 = \frac{\gamma_1}{\lambda_1}(1-e^{-\lambda_1 t_{irr}})$$
$$c_2 = \frac{\gamma_2}{\lambda_2}(1-e^{-\lambda_2 t_{irr}}) \quad (2)$$

R_{fg} - actinide fission rate, per deuteron and per gram
g - gamma line index
f - reaction index (f = fission)
Ag - gamma peak area
γ - isotope production yield [%]
m - activation sample mass [g]
ϵ_{pg} - gamma spectrometer efficiency
I_g - 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 measurement

To get capture rate a very similar formula was used except the γ_f correction omitted. Isotope data are from table of Isotopes, 8E [2]. Fission product yield data come from ENDF/BVII.1 [3].

3 Results

The Np-237 results are both in graphical, see figure 3, and tabular forms below. Please, look at the γ_2[%] column in table 2. Case of γ_2 greater than zero means that the identified isotope has a decaying predecessor and formula 2 was used for reaction rate determination. Table 3 and table 4 present the summary results for each experiment.

The Pu-239 results are limited to graphical form only as their values are still preliminary - figure 4 shows identified fission products and determined preliminary fission rate - number of fissions per second, per sample gram and per one proton.

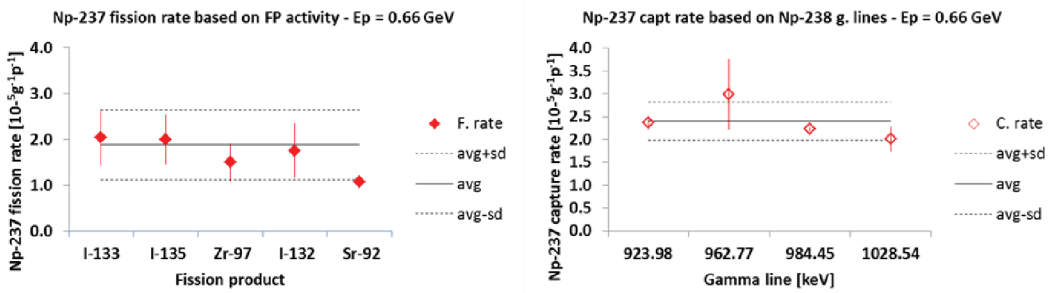

Figure 3. Np-237 fission (left) and capture (right) rates based on experiment 2014-12.

Table 2. Gamma peaks identified in Np-237 spectra.

$E_g[keV]$	$Isotope$	$T_{1/2}$	$I_g[\%]$	$\gamma_1[\%]$	$\gamma_2[\%]$
529.87	133I	20.87h	87	4.45	
555.57	91Sr	9.63h	95	2.67	
630.19	132I	2.295h	13.3	3.98	0.0407
641.28	142La	91.1 min	47.4	4.5	
657.94	97Nb	72.1m	98.23	5.35	0.031
661.66	137Cs	30.05y	85.1	5.11	
667.71	132I	2.295h	98.7	3.98	0.0407
743.36	97Zr	16.744h	93.6	5.35	
749.80	91Sr	9.63h	23.61	2.67	
756.72	95Zr	64.032d	54.38	5.54	
772.60	132I	2.295h	75.6	3.98	0.0407
954.55	132I	2.295h	17.6	3.98	0.0407
1038.76	135I	6.57h	8.01	4.16	
1131.51	135I	6.57h	22.6	4.16	
1260.41	135I	6.57h	28.7	4.16	
1383.93	92Sr	2.66h	90	4.01	
1457.56	135I	6.57h	8.73	4.16	
1678.03	135I	6.57h	9.62	4.16	
1791.20	135I	6.57h	7.77	4.16	
923.98	238Np	2.117d	2.869	100	
962.77	238Np	2.117d	0.702	100	
984.45	238Np	2.117d	27.8	100	
1028.54	238Np	2.117d	20.38	100	

Table 3. Experiment 2014-12-08 results summary for Np-237 sample.

	Avg	**Std. dev.**	**Std. dev. [%]**
Fission rate $[g^{-1}p^{-1}]$	1.67E-05	3.53E-06	21.10
Capture rate $[g^{-1}p^{-1}]$	2.40E-05	4.17E-06	17.40
fission/capture	0.70	0.19	27.35

Table 4. Experiment 2015-12-04 results summary for Np-237 sample - preliminary.

	Avg	**Std. dev.**	**Std. dev. [%]**
Fission rate $[g^{-1}p^{-1}]$	8.62E-06	2.17E-06	25.24
Capture rate $[g^{-1}p^{-1}]$	1.49E-05	1.65E-06	11.07
fission/capture	0.58	0.16	27.55

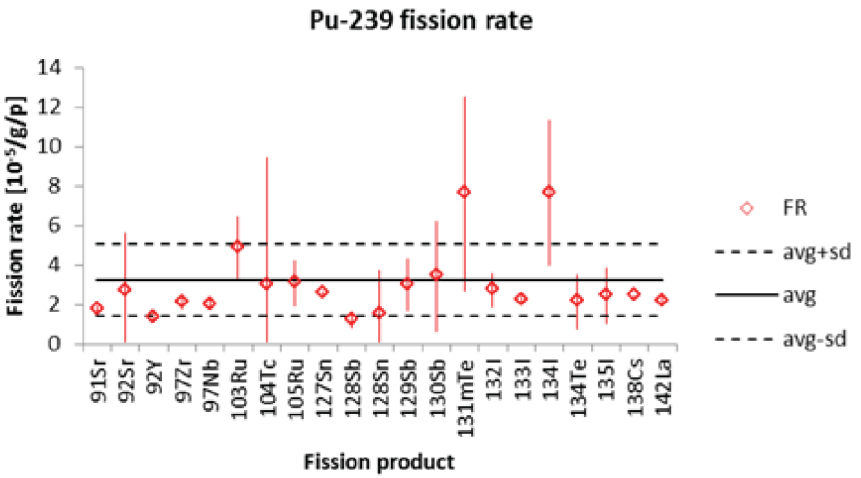

Figure 4. Pu-239 fission rate based on fission product activities - preliminary.

4 Conclusions

Np-237 and Pu-239 samples were irradiated in spallation neutron field to search for incineration feasibility. Both Np-237 fission and neutron capture products were identified. The fission to capture ratio was found to be below 1 which means that neutron capture still prevails in QUINTA neutron field.

Pu-239 fission products were identified as well but the fission rate value is still preliminary. The applied method did not allow to determine capture rate.

Acknowledgments

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