Np-237 transmutation effectiveness dependence on beam particle and energy in QUINTA setup

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Some Np-237 introductory data

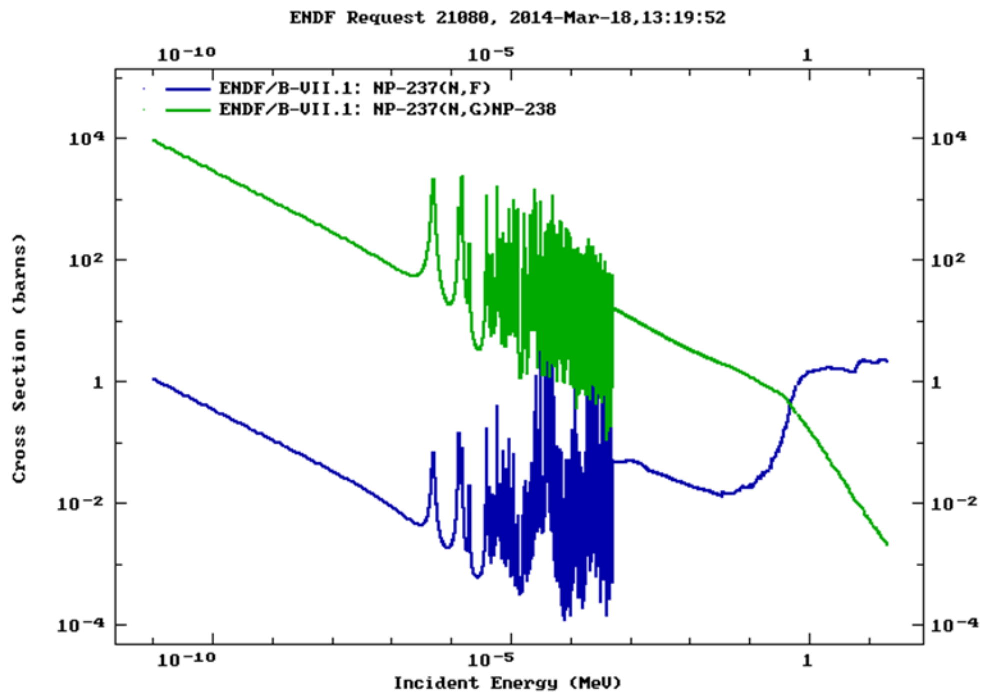
Radioactive, $T_{1/2} = 2.144 \times 10^6$ y

Produced in a reactor as a nuclear waste.

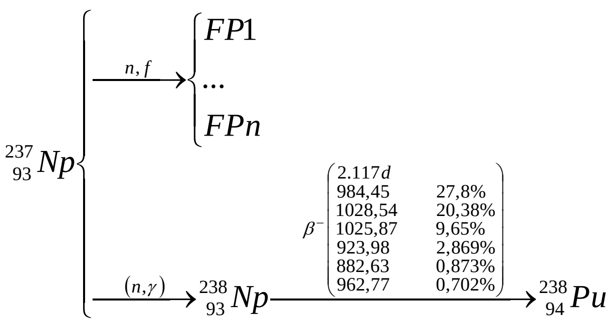
Difficult to burn in PWRs. It accumulates.

Basic Np-237 interaction modes with neutrons

Neutron capture produces another actinide. Np-237 fission is in fact the only way to get rid of its long lived activity. High energy neutrons needed to make fission prevail over capture.

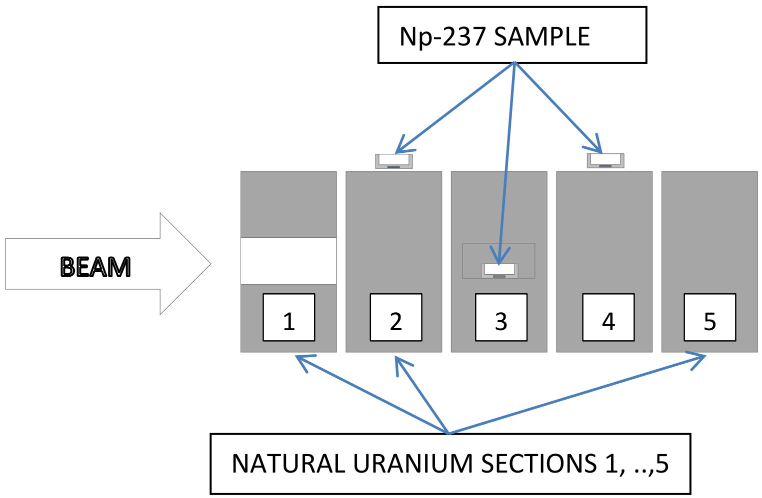


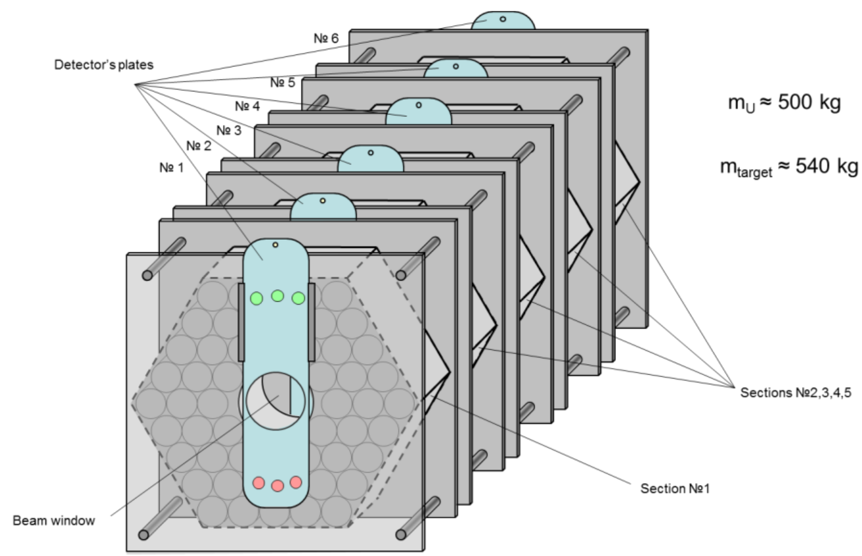
Np-237 fission and neutron capture CS dependence on energy



Basic Np-237 interaction modes with neutrons

QUINTA setup core vertical-axial cross section and 3D view





Experiment data

Beam energy/particle *	0.66 GeV/p	2 GeV/d	4 GeV/d	8 GeV/d	24 GeV/C6+	0.66 GeV/p	
Date	08 Nov 2014	04 Dec 2012	13 Dec 2012	22 Dec 2012	18 Dec 2013	22 Jun 2017	
Irradiation time (h)	5.72	6.27	9.35	16.17	22.8	5.25	
Total number of beam particles	8.64x10^{14}	3.052(9)x10^{13}	3.569(15) x10^{13}	1.390(8) x10^{13}	1.75 x10^{11}	7.78(9)x10^{14}	
Sample position	Left side window	Left side window	Left side window	Left side window	Left side window	Top of section 2	Top of section 4

*Particles: p – proton, d – deuteron, C6+ - carbon

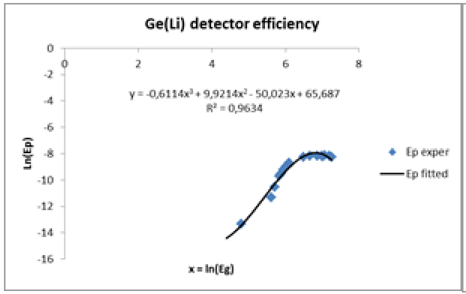
During the 22.06.2017 experiment the Np-237 samples were located on top of section 2 and section 4 of QUINTA while during the remaining ones in the left side window in lead shield.
During the 22.06.2017 experiment QUINTA was configured without lead shield.

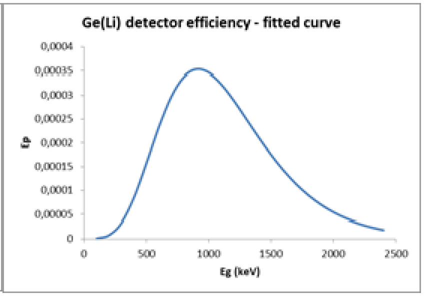
Experimental data work-out details

$$I_{f\gamma} = \frac{S_\gamma}{\gamma_f \cdot m \cdot \varepsilon_p \cdot I_\gamma \cdot \phi \cdot COI} \cdot \frac{\lambda_k \cdot t_{ir}}{\left(1 - e^{-\lambda \cdot t_{ir}}\right)} \cdot \frac{1}{\left(1 - e^{-\lambda t_{real}}\right)} \cdot \frac{t_{real}}{t_{live}} \cdot e^{\lambda t_+}$$

$I_{f\gamma}$ – 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 [%]
m – activation sample mass [g]
ε_p – gamma spectrometer efficiency
I_γ – gamma line intensity [%]

ϕ - deuteron beam integral
COI – correction for gamma quanta coincidence
λ_k – isotope decay constant
t_+ – cooling time
t_{ir} – irradiation time
t_{real} – real time of measurement
t_{live} – live time of measurement





$$\varepsilon_p = -0.6114x^3 + 9.921x^2 - 50.023x + 65.687$$
$$x = \ln(E)$$
$$R^2 = 0.9634$$

Experimental data work-out details

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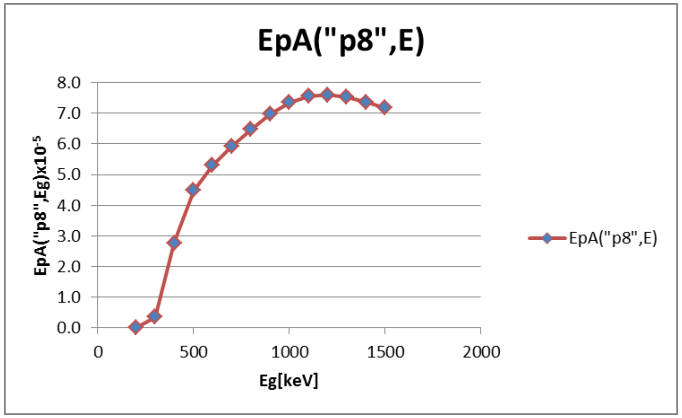


ε_p = 2.0027x5 + 1.0373x4 - 1.0403x3 - 1.0105x2 + 0.3977x - 9.5181
$x = \ln(E_g)$
$R^2 = 0.8745$

Example detector efficiency function

Basic Np-237 FP identified decay chains – part 1/2

$$Fission \xrightarrow{\gamma_1=3.93\%} {}^{91}_{38}Sr \xrightarrow{\beta^-\begin{pmatrix} & 9.63h & \\ 1024.3\ keV & & 33\% \\ 749.8\ keV & & 23.61\% \end{pmatrix}} {}^{91}_{39}Y \xrightarrow{IT\begin{pmatrix} & 49.71m & \\ 555.58\ keV & & 100\% \end{pmatrix}} {}^{91}_{39}Y$$

$$Fission \xrightarrow{\gamma_1=4.37\%} {}^{92}_{38}Sr \xrightarrow{\beta^-\begin{pmatrix} & 2.71h & \\ 1383.93\ keV & & 90\% \\ 953.31\ keV & & 3.52\% \end{pmatrix}} {}^{92}_{39}Y \xrightarrow{\beta^-\begin{pmatrix} & 3.54h & \\ 934.46\ keV & & 13.9\% \\ 1405.28\ keV & & 4.8\% \end{pmatrix}} {}^{92}_{40}Zr$$

$$Fission \xrightarrow{\gamma_1=5.67\%} {}^{95}_{40}Zr \xrightarrow{\beta^-\begin{pmatrix} & 64.02d & \\ 756.729\ keV & & 54\% \\ 724.199\ keV & & 44.1\% \end{pmatrix}} {}^{95}_{41}Nb \xrightarrow{\beta^-\begin{pmatrix} & 34.975d & \\ 765.794\ keV & & 100\% \end{pmatrix}} {}^{95}_{42}Mo$$

$$\left.\begin{matrix} Fission \xrightarrow{\gamma_1=6.11\%} {}^{97}_{40}Zr \xrightarrow{\beta^-\begin{pmatrix} & 16.97h & \\ 743.36\ keV & & 93\% \\ 507.64keV & & 5.03\% \end{pmatrix}} \\ Fission \xrightarrow{\gamma_2=6.14\%} \end{matrix}\right\} {}^{97}_{41}Nb \xrightarrow{\beta^-\begin{pmatrix} & 72.1m & \\ 658.08\ keV & & 98\% \end{pmatrix}} {}^{97}_{42}Mo$$

$$Fission \xrightarrow{\gamma_1=0.683\%} {}^{131}_{52}Te \xrightarrow{\beta^-\begin{pmatrix} & 30h & \\ 149.716\ keV & & 69\% \\ 452.323\ keV & & 18.18\% \\ 492.66\ keV & & 4.826\% \\ 602.039\ keV & & 4.187 \end{pmatrix}} {}^{131}_{53}I \xrightarrow{\beta^-\begin{pmatrix} & 8.9207d & \\ 364.489\ keV & & 81.7\% \\ 636.989\ keV & & 7.17\% \\ 284.305\ keV & & 6.14\% \\ 722.911\ keV & & 1.773\% \end{pmatrix}} {}^{132}_{54}Xe$$

$$\left.\begin{matrix} Fission \xrightarrow{\gamma_1=4.75\%} {}^{132}_{52}Te \xrightarrow{\beta^-\begin{pmatrix} & 3.204d & \\ 228.16\ keV & & 88\% \\ 49.72keV & & 15\% \end{pmatrix}} \\ Fission \xrightarrow{\gamma_2=4.85\%} \end{matrix}\right\} {}^{132}_{53}I \xrightarrow{\beta^-\begin{pmatrix} & 2.295h & \\ 667.718\ keV & & 99\% \\ 772.6\ keV & & 75.6\% \\ 954.55\ keV & & 17.6\% \\ 630.19\ keV & & 13.3\% \end{pmatrix}} {}^{132}_{54}Xe$$

$$Fission \xrightarrow{\gamma_1=6.46\%} {}^{133}_{53}I \xrightarrow{\beta^-\begin{pmatrix} & 20.8h & \\ 529.872\ keV & & 87\% \end{pmatrix}} {}^{133}_{54}Xe$$

Basic Np-237 FP identified decay chains – part 2/2

$$\left.\begin{matrix} Fission \xrightarrow{\gamma_1=4.41\%} {}^{134}_{52}Te \xrightarrow{\beta^-\begin{pmatrix} & 41.8m & \\ 767.2\ keV & & 29.5\% \\ 210.465\ keV & & 22.7\% \\ 277.951\ keV & & 21.2\% \\ 79.445\ keV & & 20.9\% \\ 435.06\ keV & & 18.9\% \\ 565.992\ keV & & 18.6\% \\ 180.891\ keV & & 18.3\% \\ 742.58\ keV & & 15.3\% \end{pmatrix}} \\ Fission \xrightarrow{\gamma_2=6.95\%} \end{matrix}\right\} {}^{134}_{53}I \xrightarrow{\beta^-\begin{pmatrix} & 52.8m & \\ 847.025\ keV & & 95.4\% \\ 884.09\ keV & & 64.9 \\ 1072.55\ keV & & 14.9\% \\ 595.362\ keV & & 11.1\% \\ 621.79\ keV & & 10.6\% \\ 1136.16\ keV & & 9.1\% \\ 677.34\ keV & & 7.9\% \\ 540.825\ keV & & 7.63\% \\ 405.451\ keV & & 7.35\% \\ 857.29\ keV & & 6.68\% \end{pmatrix}} {}^{134}_{54}Xe$$

$$\left.\begin{matrix} Fission \xrightarrow{\gamma_1=6.71\%} {}^{135}_{53}I \xrightarrow{\beta^-\begin{pmatrix} & 6.57h & \\ 1260.409\ keV & & 28.9\% \\ 1131.511\ keV & & 22.74\% \end{pmatrix}} \\ Fission \xrightarrow{\gamma_2=1.37\%} \end{matrix}\right\} {}^{135}_{54}Xe \xrightarrow{\beta^-\begin{pmatrix} & 9.1h & \\ 249.77\ keV & & 90\% \\ 608.151\ keV & & 2.90\% \end{pmatrix}} {}^{135}_{55}Cs$$

$$Fission \xrightarrow{\gamma_1=6.25\%} {}^{137}_{55}Cs \xrightarrow{\beta^-\begin{pmatrix} & 30.7y & \\ 661.657\ keV & & 85.1\% \end{pmatrix}} {}^{137}_{56}Ba$$

$$\left.\begin{matrix} Fission \xrightarrow{\gamma_1=5.25\%} {}^{138}_{54}Xe \xrightarrow{\beta^-\begin{pmatrix} & 14.08m & \\ 258.411\ keV & & 31.5 \\ 434.562\ keV & & 20.3 \\ 1768.26\ keV & & 16.7\% \\ 2015.82 & & 12.25 \end{pmatrix}} \\ Fission \xrightarrow{\gamma_2=0.466\%} \end{matrix}\right\} {}^{138}_{55}Cs \xrightarrow{\beta^-\begin{pmatrix} & 33.41m & \\ 462.798\ keV & & 18.6\% \\ 1435.795\ keV & & 19\% \end{pmatrix}} {}^{138}_{56}Ba$$

$$Fission \xrightarrow{\gamma_1=4.97} {}^{142}_{57}La \xrightarrow{\beta^-\begin{pmatrix} & 91.1m & \\ 641.285\ keV & & 47\% \\ 2397.8\ keV & & 13.3\% \\ 2542.7\ keV & & 10\% \\ 894.9\ keV & & 8.34\% \\ 1901.3\ keV & & 7.16\% \end{pmatrix}} {}^{142}_{58}Ce$$

Basic gamma lines identified

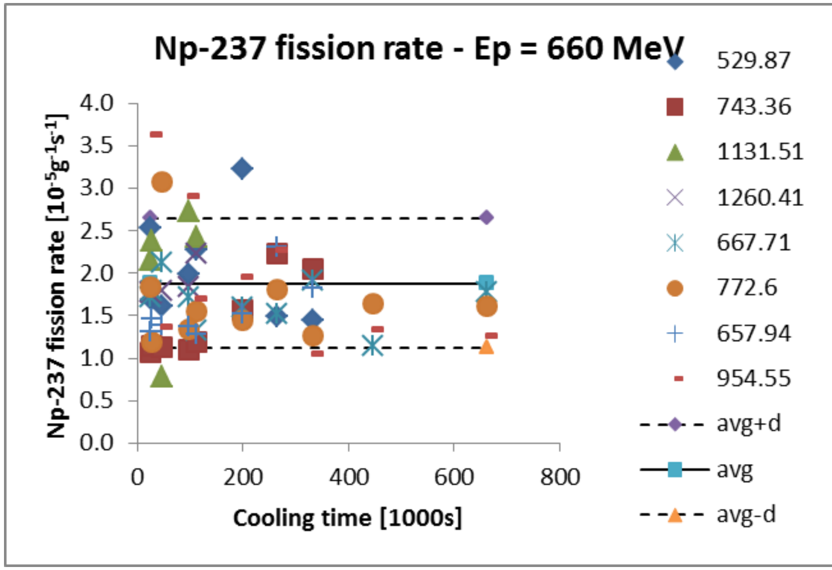
E-gamma, keV	Isotope	Source	T1/2	Fission yield [%] [5]	I-gamma [%] [4]
529.87	I-133	FP	20.87h	6.46	87
658.08	Zr-97->97Nb*	FP	16.744h	6.11	98.23
667.71	Te-132->I-132**	FP	3.26d	4.85	98.7
743.36	Zr-97	FP	16.744h	6.11	93.6
772.6	Te-132->I-132**	FP	3.26d	4.85	75.6
1131.51	I-135	FP	6.57h	6.71	22.6
1260.41	I-135	FP	6.57h	6.71	28.7
923.98	Np-238	CP	2.117d	N/A	2.869
962.77	Np-238	CP	2.117d	N/A	0.702
984.45	Np-238	CP	2.117d	N/A	27.8
1025.87	Np-238	CP	2.117d	N/A	9.65
1028.54	Np-238	CP	2.117d	N/A	20.38

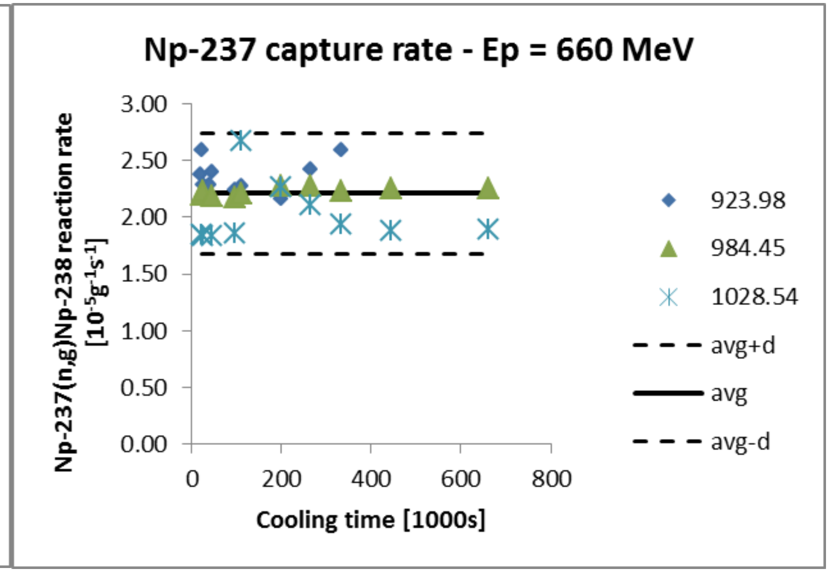
FP – fission product. CP – neutron capture product.

*Line 658.08 keV stems in fact from Nb-97 beta decay ($T_{1/2}$ = 72.1 min), but its quantity is modified by Zr-97 decay rate ($T_{1/2}$ = 16.744h) [4]. Therefore Zr-97 decay constant (16.744h) approximates the line 658.08 activity decreasing.

**Lines 667.71 and 772.6 keV stem from I-132 ($T_{1/2}$ = 2.295h) but their activities are modified by Te-132 decay rate ($T_{1/2}$ = 3.26d) [4]. Therefore Te-132 decay constant (3.26d) approximates the lines activity decreasing.

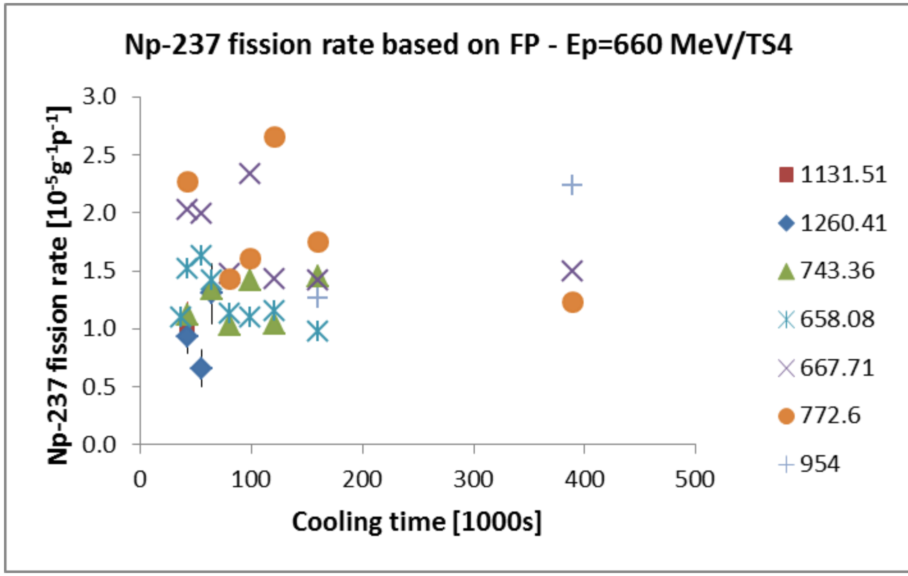
Np-237 fission and capture rate example partial results put together – experiment with 0.66 GeV proton beam

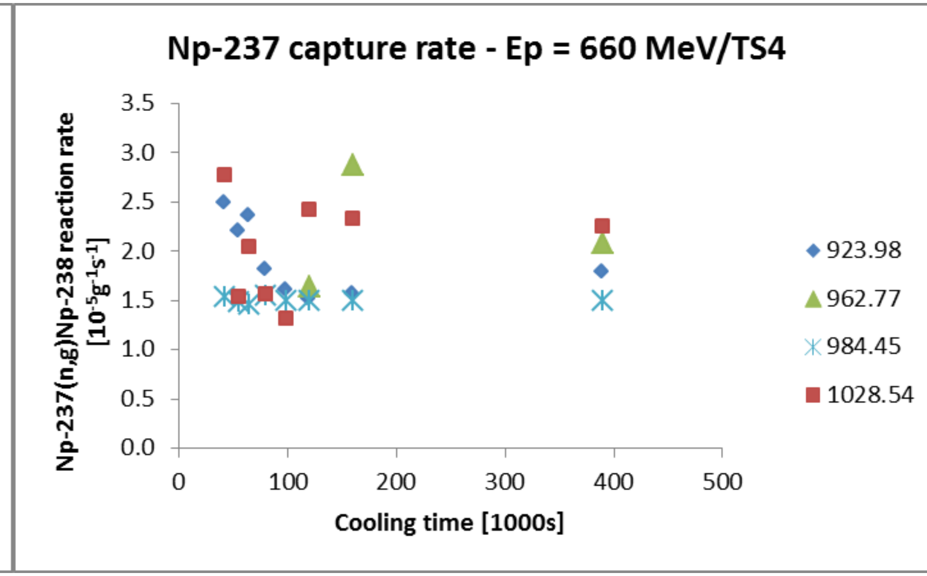




Np-237 sample located in QUINTA's left side window in lead shield

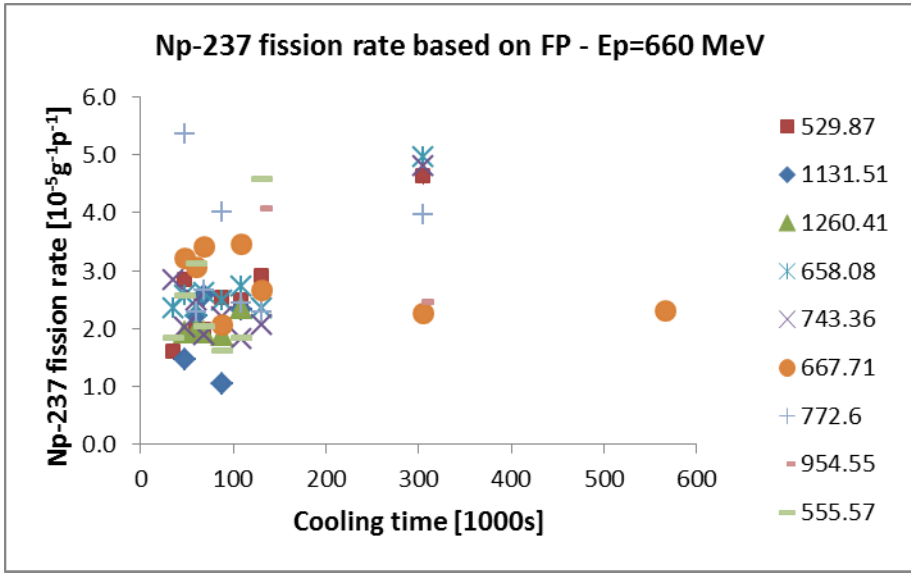
Np-237 fission and capture rate example partial results put together – experiment with 0.66 GeV proton beam

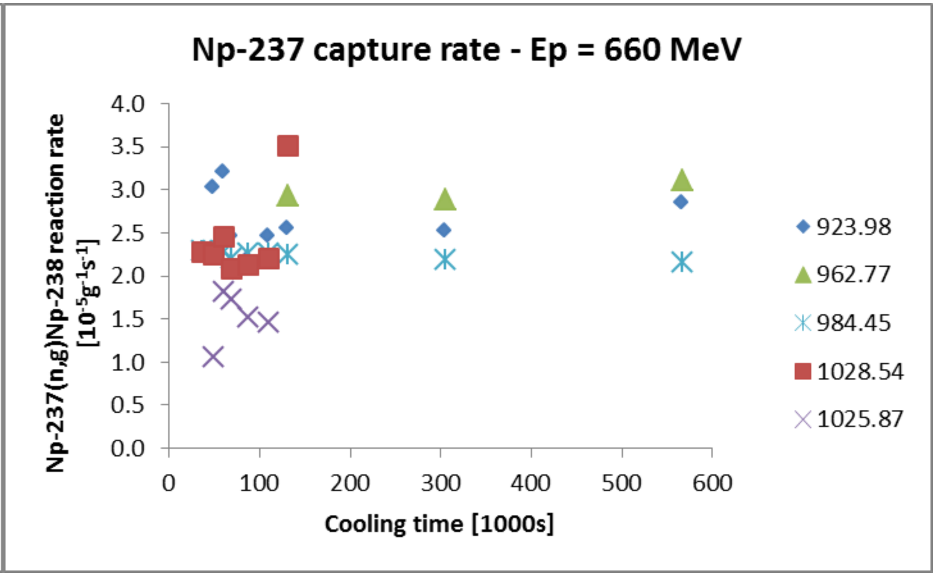




Np-237 sample located on top of section 4.

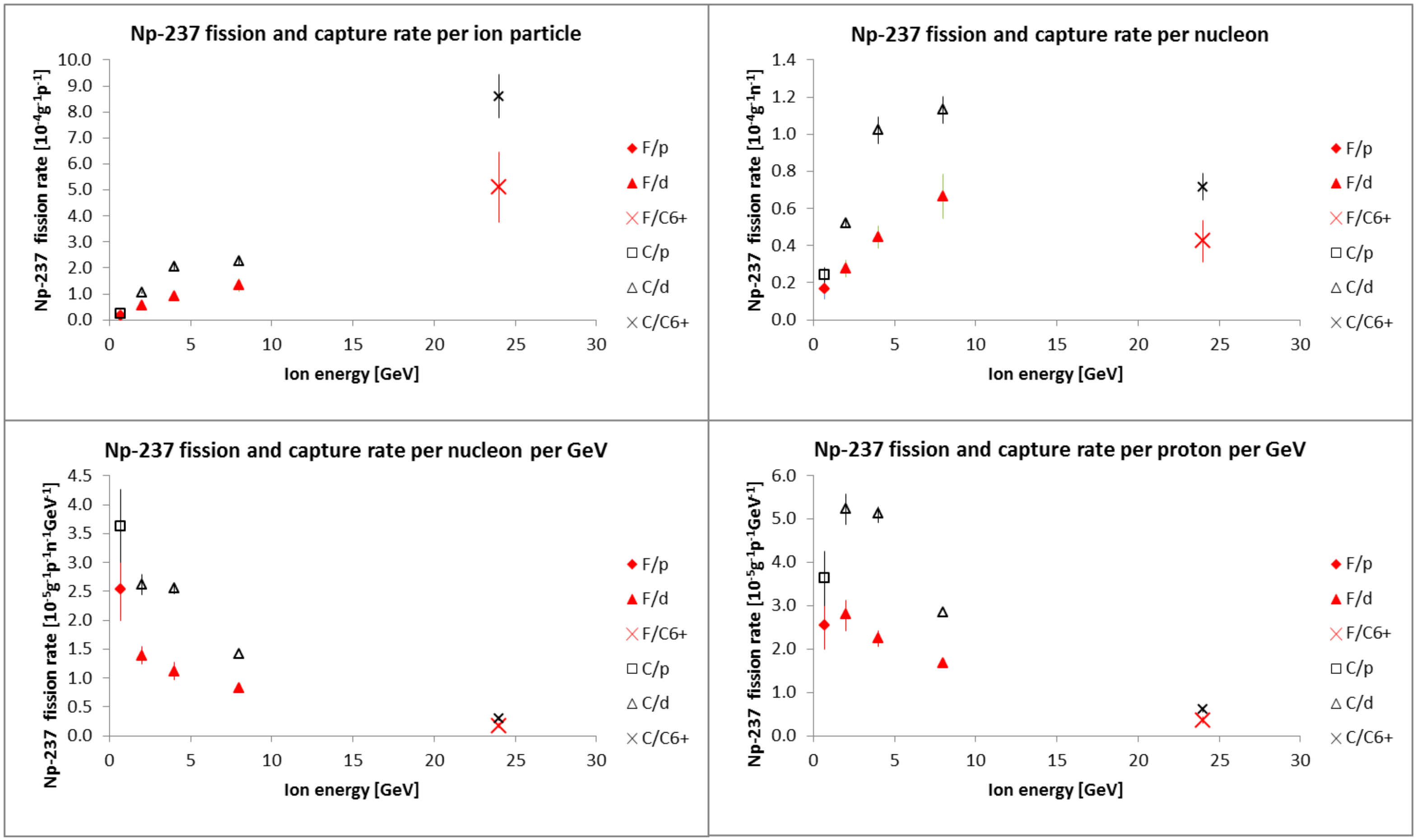
Np-237 fission and capture rate example partial results put together – experiment with 0.66 GeV proton beam





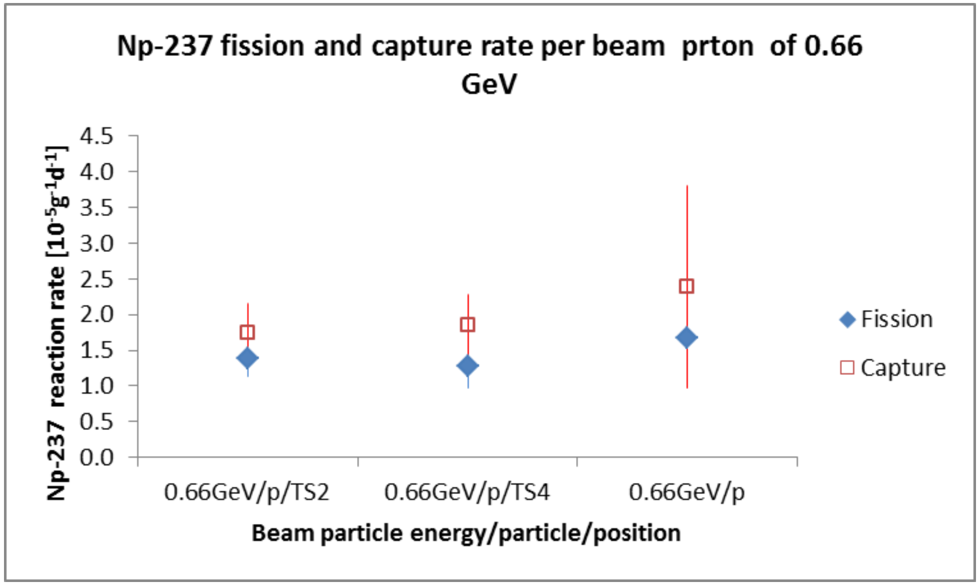
Np-237 sample located on top of section 2.

Np-237 fission and capture rate results dependence on beam particle, beam ion energy at sample location – QUINTA's left side window

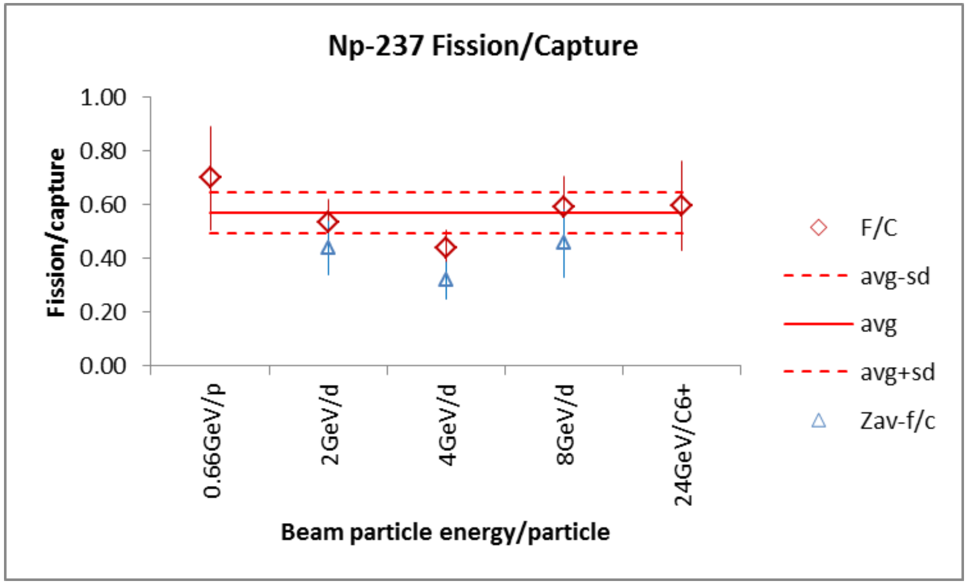


Legend: reaction/particle – for example F/p – fission/proton.
Reactions: F – fission, C – neutron capture.
Particles: p – proton, d – deuteron, C6+ - carbon 6-times ionized

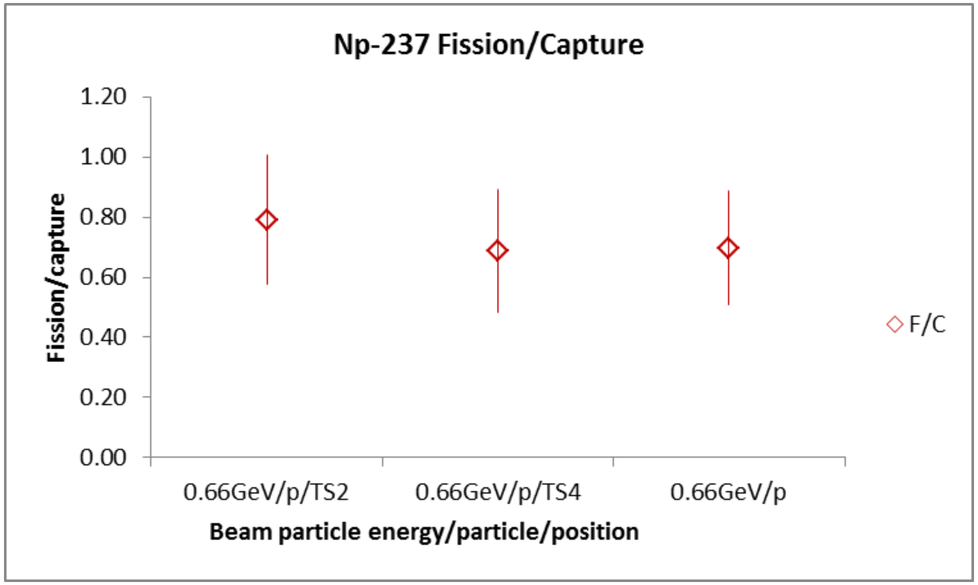
Np-237 fission and capture rate dependence on sample location for proton beam



Np-237 fission to capture ratio dependence on beam particle and energy



Np-237 fission to capture ratio dependence on sample location for proton beam



Basic Np-237 incineration parameters for each experiment

Particle energy/particle \ Avg. fission and capture	0.66GeV/p	2GeV/d	4GeV/d	8GeV/d	24GeV/C6+	0.66GeV/p/TS2	0.66GeV/p/TS4
fission (10^{-5} $g^{-1}p^{-1}$)	1.67(35)	5.56(87)	8.95(12)	13.3(24)	51.08(135)	1.38(25)	1.28(30)
capture (10^{-5} $g^{-1}p^{-1}$)	2.40(42)	10.10(43)	20.40(14)	22.6(14)	86.06(84)	1.75(36)	1.85(33)
fission (10^{-5} $g^{-1}p^{-1}nucleon^{-1}$)	1.67(35)	2.78(43)	4.47(60)	6.65(12)	4.26(11)	1.38(25)	1.28(30)
capture (10^{-5} $g^{-1}p^{-1}nucleon^{-1}$)	2.40(42)	5.22(22)	10.21(71)	11.31(7)	7.17(7)	1.75(36)	1.85(33)
fission (10^{-5} $g^{-1}p^{-1}nucleon^{-1}GeV^{-1}$)	2.53(53)	1.39(22)	1.12(15)	0.83(15)	0.18(5)	2.09(38)	1.93(46)
capture ($g^{-1}p^{-1}nucleon^{-1}GeV^{-1}$)	3.63(63)	2.61(11)	2.55(18)	1.41(9)	0.30(3)	2.64(54)	2.81(49)
fission (10^{-5} $g^{-1}p^{-1}proton^{-1}GeV^{-1}$)	2.53(53)	2.78(43)	2.24(30)	1.66(30)	0.35(9)	2.09(38)	1.93(46)
capture (10^{-5} $g^{-1}p^{-1}proton^{-1}GeV^{-1}$)	3.63(63)	5.22(22)	5.11(35)	2.83(18)	0.60(6)	2.64(54)	2.81(49)
F/C	0.70(19)	0.53(9)	0.44(7)	0.59(11)	0.59(17)	0.79(22)	0.69(20)
F/A	0.41(11)	0.35(6)	0.30(5)	0.37(7)	0.37(11)	0.44(12)	0.41(12)

Notes:

1. Experiments with samples at TS2 and TS4 position – QUINTA with no lead shield. The remaining – QUINTA in lead shield.
2. Experiments with 0.66 GeV proton beam (0.66 GeV/p, 0.66 GeV/p/TS2, 0.66 GeV/p/TS4) - both fission and capture rate and fission to capture ratio are very close to each other.

TS2 – top of section 2. TS4 – top of section 4 of QUINTA.
Remainding – sample in side window in lead shield

Conclusions

1. Fission rate per beam energy unit suggests proton beam to be the best one for Np-237 incineration. It suggests that neutron contained in beam particle nucleus gives much less contribution than proton to target nuclei spallation.
2. The presented data show transmutation rate dependence on beam energy, but it is impossible to state where the maximum is.
3. For fixed beam particle (proton) and beam energy the incineration rate shows no dependence on Np-237 position. Fission to capture ratio shows no dependence on sample position what suggests the neutron spectrum to be uniform throughout entire QUINTA body.

References

1. S. Kilim et al.; Np-237 incineration study in various beams in ADS setup QUINTA; Nukleonika 63, 17–22, 2018, https://doi.org/10.1515/nuka-2018-0003
2. W. Furman et al.; Recent results of the study of ADS with 500 kg natural uranium target assembly QUINTA irradiated by deuterons with energies from 1 to 8 GeV at JINR NUCLOTRON*; PoS(Baldin ISHEPP XXI)086.*
3. S. Kilim et al.; Measurements of Np-237 incineration in ADS setup QUINTA; *PoS(Baldin ISHEP XXII)056*
4. Evaluated Nuclear Data File (ENDF). Interpreted ENDF file. Np-237(FY_cum) Cumulative Fission-Product Yields and (n, ind_Fy) Independent Fission-Product Yields.
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6. L. Zavorka et al. (2015); Neutron-induced transmutation reactions in 237Np, 238Pu, and 239Pu at the massive natural uranium spallation target; *Nuclear Instruments and Methods in Physics Research B 349 (2015) 31–38;* Retrieved September 14, 2015 from http://dx.doi.org/10.1016/j.nimb.2014.12.084; 0168-583X/_ 2015 Elsevier B.V.
7. S. R. Hashemi-Nezhad et al. “Optimal ion beam, target type and size for accelerator driven systems: implications to the associated accelerator power,” *Ann. Nucl. Energy 38, 1144–1155 (2011).*