

Measurements of Np-237 incineration in ADS setup QUINTA

E. Strugalska-Gola¹, S. Kilim¹, M. Bielewicz¹, M. Szuta¹, S. Tyutyunnikov², L. Zavorka², J. Adam², V. Stegailov², V. Chilap³,

1. National Centre for Nuclear Research, 05-400 Otwock-Świerk, Poland
2. Joint Institute for Nuclear Research, 141980 Dubna, Russia
3. CPTP "Atomenergomash", Moscow, Russia

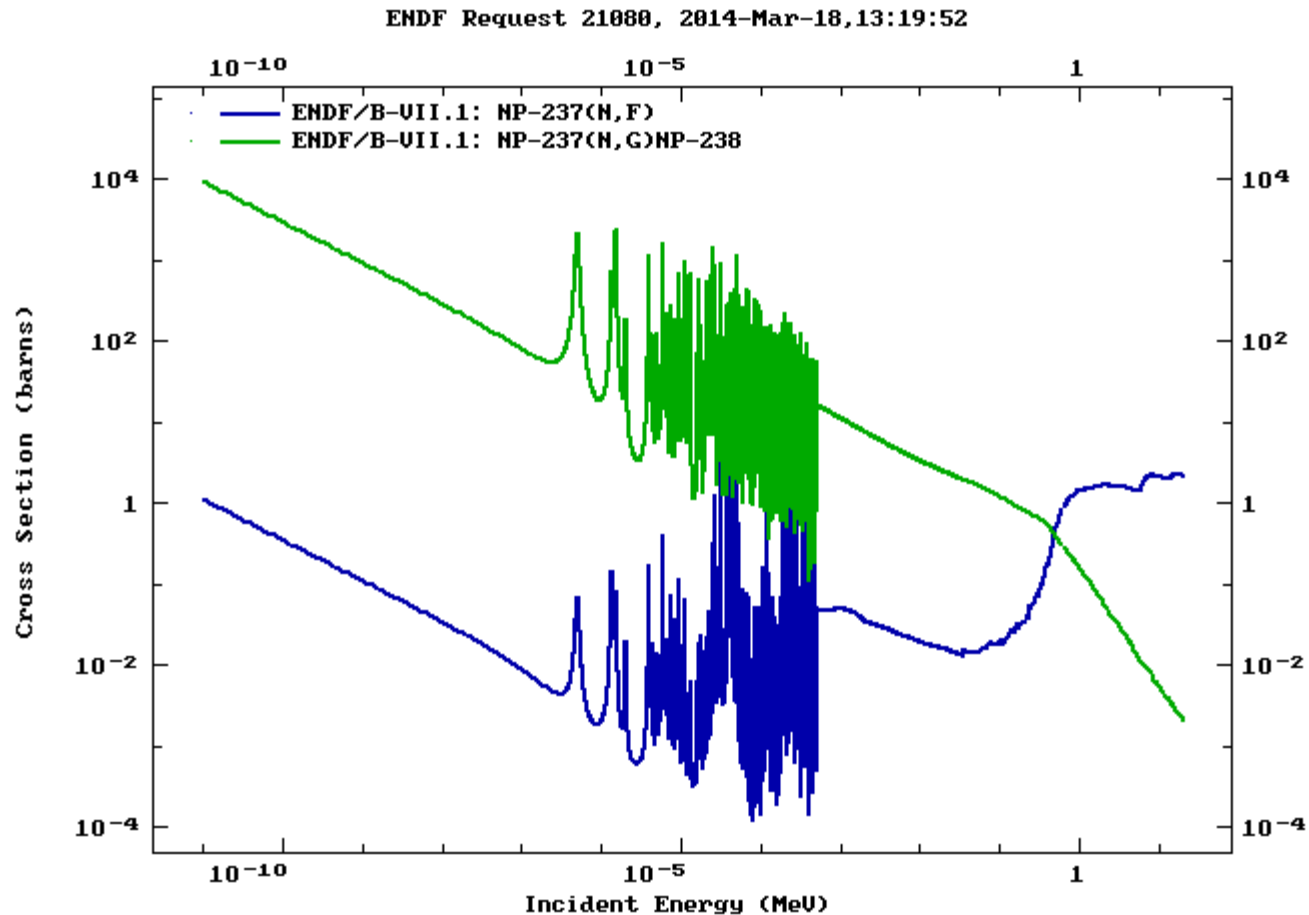
Some Np-237 introductory data

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.

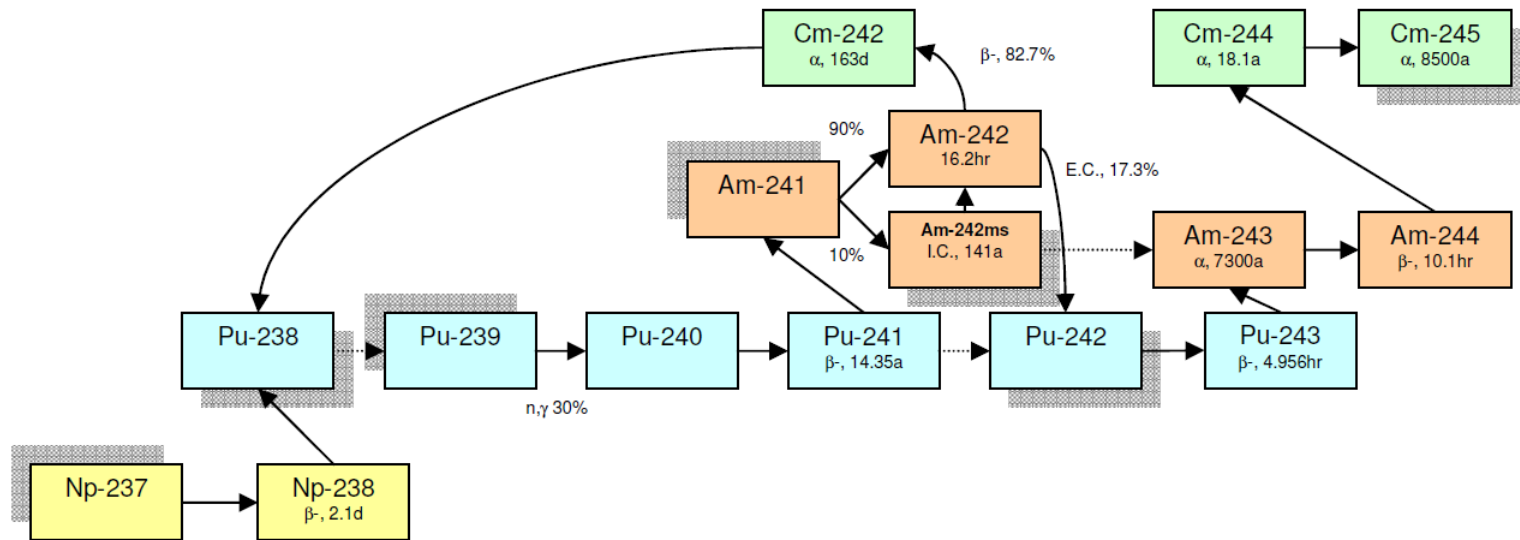
Np-237 fission and neutron capture CS dependence on energy



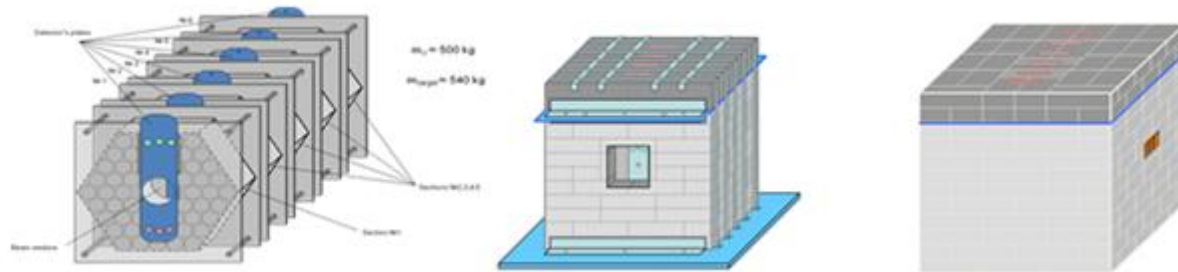
Neutron capture produces another actinide. Np-237 fission is in fact the only way to get rid of its long lived activity. High energy neutrons needed to make fission prevail over capture.

Actinides accumulation ways when starting from Np-237

Samuel E. Bays; Reactor Physics Characterization of Transmutation Targeting Options in a Sodium Fast Reactor; INL/CON-07-12439



Experiment description



QUINTA setup 3D view – inner core, front and rear view



Deuteron beam run – Ed = 2, 4 and 8 GeV respectively

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 (10¹³)	3.052(9)	3.569(15)	1.390(8)

Experimental data work-out details

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

$I_{f\gamma}$ – actinide fission rate, per deuteron and per gram

γ – gamma line index

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

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

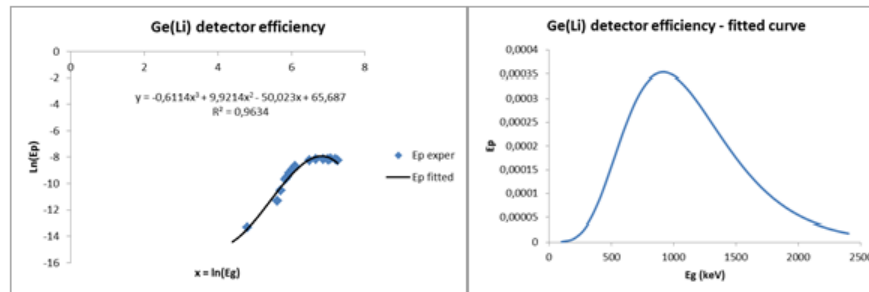
λ_k – isotope decay constant

t_+ – cooling time

t_{ir} – irradiation time

t_{real} – real time of measurement

t_{live} – live time of measurement



$$\varepsilon_p = -0.6114x^3 + 9.921x^2 - 50.023x + 65.687$$

$$x = \ln(E)$$

$$R^2 = 0.9634$$

Basic gamma lines identified

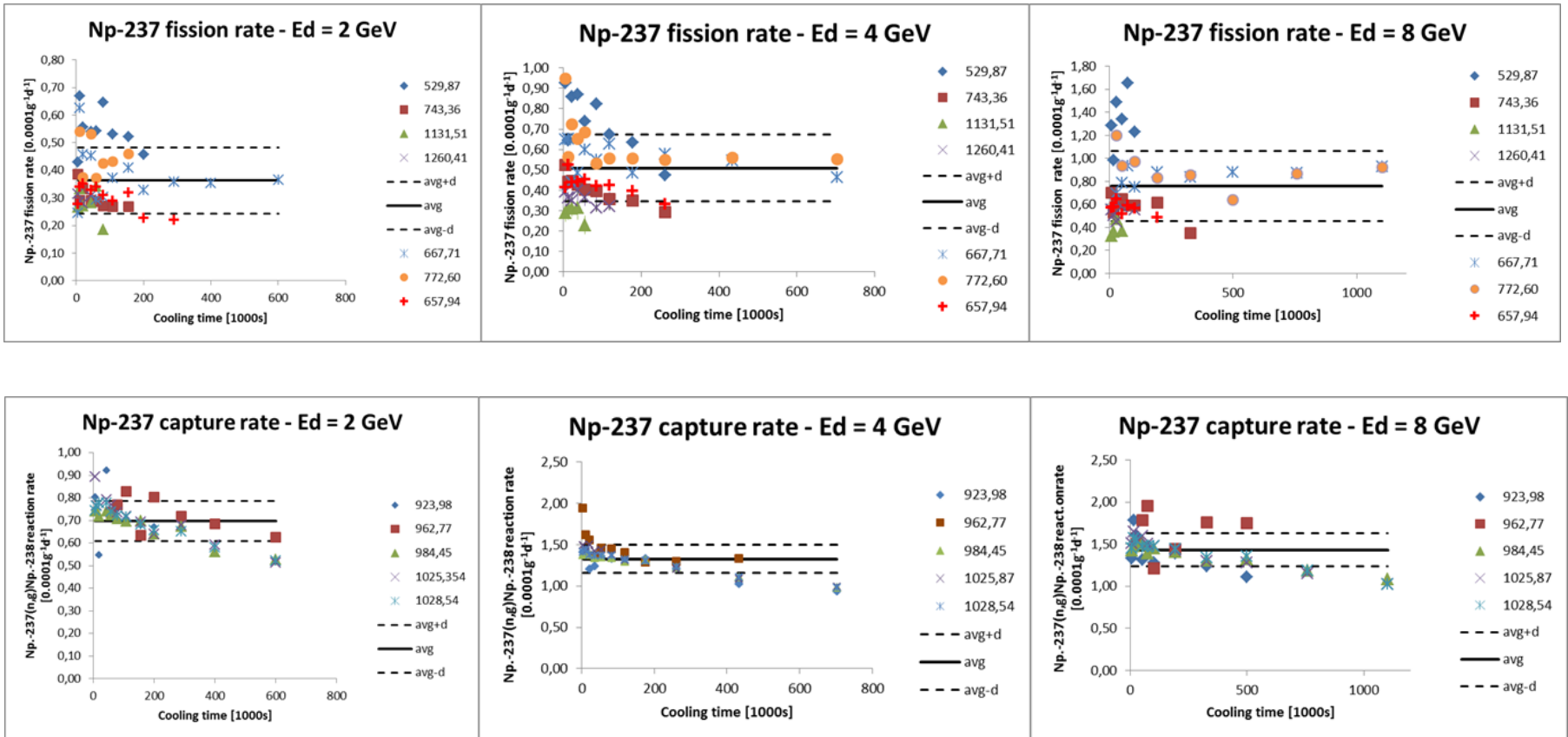
E-gamma	Isotope	Source	T1/2	Fission yield [%] [4]	I-gamma [%] [3]
529.87	133I	FP	20.87h	4,45	87
657.94	Zr-97->97Nb*	FP	16.744h	5,38	98,23
667.71	Te-132->I-132**	FP	3.26d	4,39	98,7
743.36	Zr-97	FP	16.744h	5,35	93,6
772.6	Te-132->I-132**	FP	3.26d	4,39	75,6
1131.51	I-135	FP	6.57h	4,16	22,6
1260.41	I-135	FP	6.57h	4,16	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.

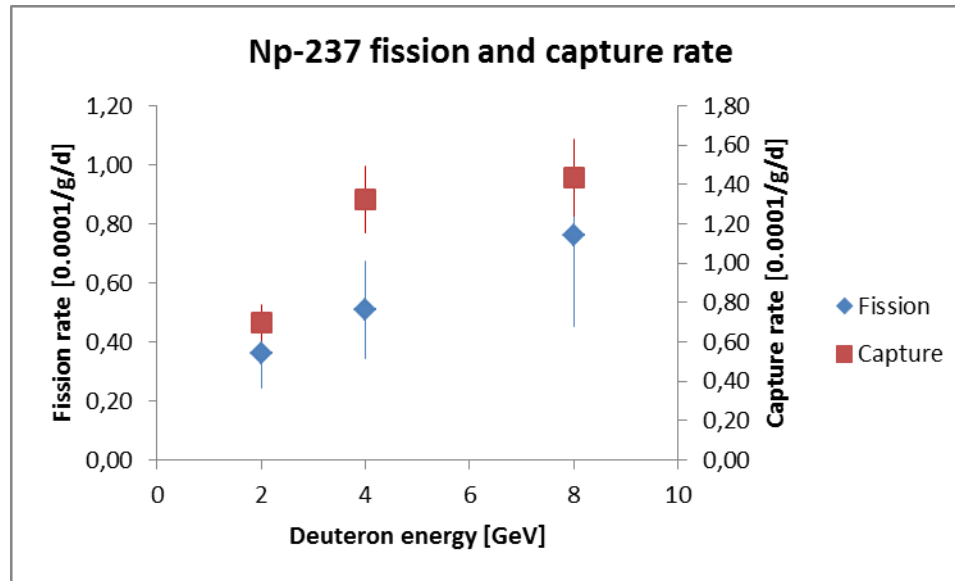
*Line 657.94 keV stems in fact from Nb-97 beta decay ($T_{1/2} = 72.1$ min), but its quantity is modified by Zr-97 decay rate ($T_{1/2} = 16.744$ h) [3,4]. Therefore Zr-97 decay constant (16.744h) approximates the line 657.94 activity decreasing.

**Lines 667.71 and 772.6 keV stem from I-132 ($T_{1/2} = 2.295$ h) but their activities are modified by Te-132 decay rate ($T_{1/2} = 3.26$ d) [3,4]. Therefore Te-132 decay constant (3.26d) approximates the lines activity decreasing.

Np-237 fission rate and capture rate results

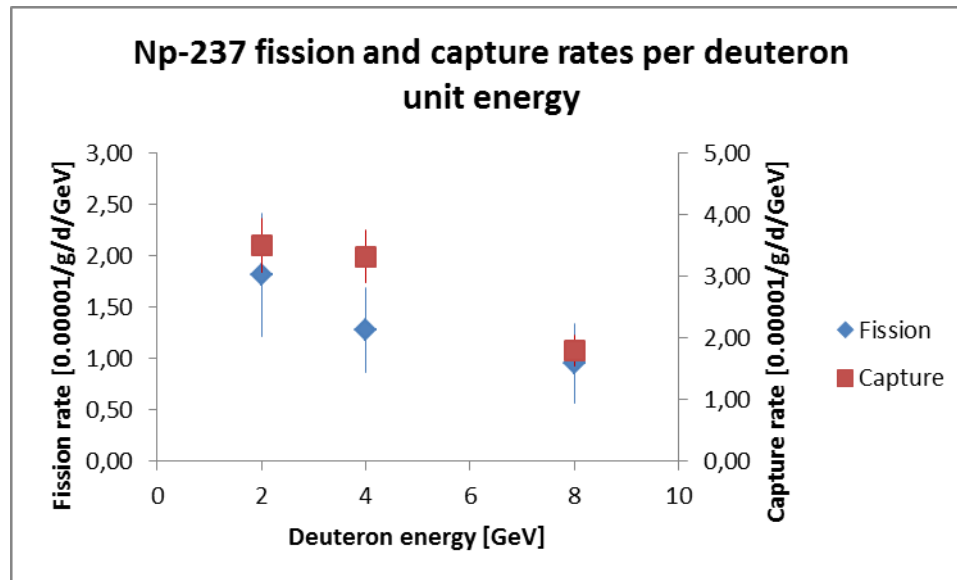


Np-237 fission and capture dependence on deuteron energy

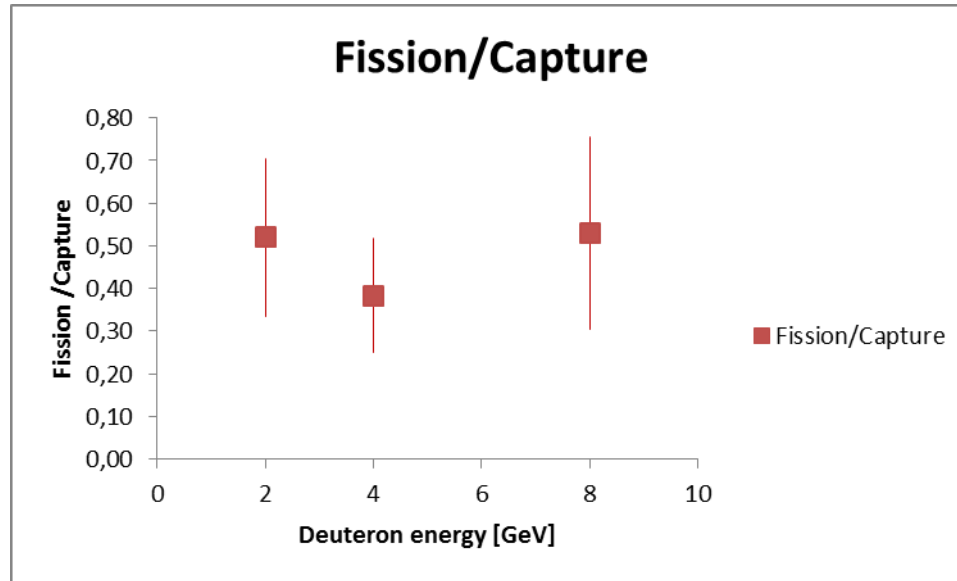


Beam deuteron energy [GeV]	Fission rate [$10^{-4} \text{g}^{-1} \text{d}^{-1}$]	Standard deviation [$10^{-4} \text{g}^{-1} \text{d}^{-1}$]	Standard deviation [%]	Capture rate [$10^{-4} \text{g}^{-1} \text{d}^{-1}$]	Standard deviation [$10^{-4} \text{g}^{-1} \text{d}^{-1}$]	Standard deviation [%]
2	0.363	0.12	33.04	0.699	0.0887	12.69
4	0.509	0.164	32.26	1.33	0.171	12.88
8	0.76	0.306	40.3	1.43	0.196	13.67

Np-237 fission and capture rates per deuteron unit energy

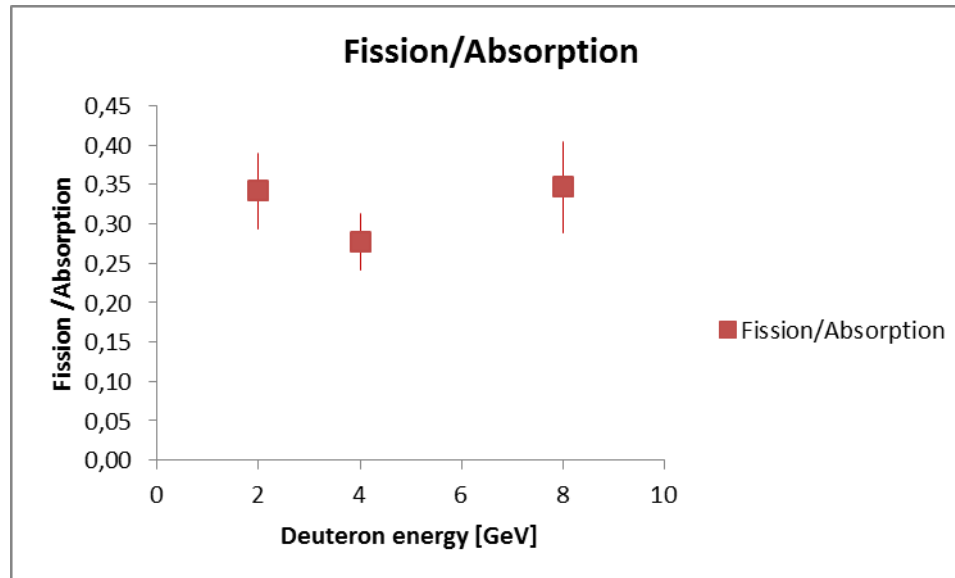


Np-237 fission to capture ratio dependence on deuteron energy



Beam deuteron energy [GeV]	Fission/Capture ratio	Standard deviation	Standard deviation [%]
2	0.52	0.18	35.40
4	0.38	0.13	34.74
8	0.53	0.23	42.56

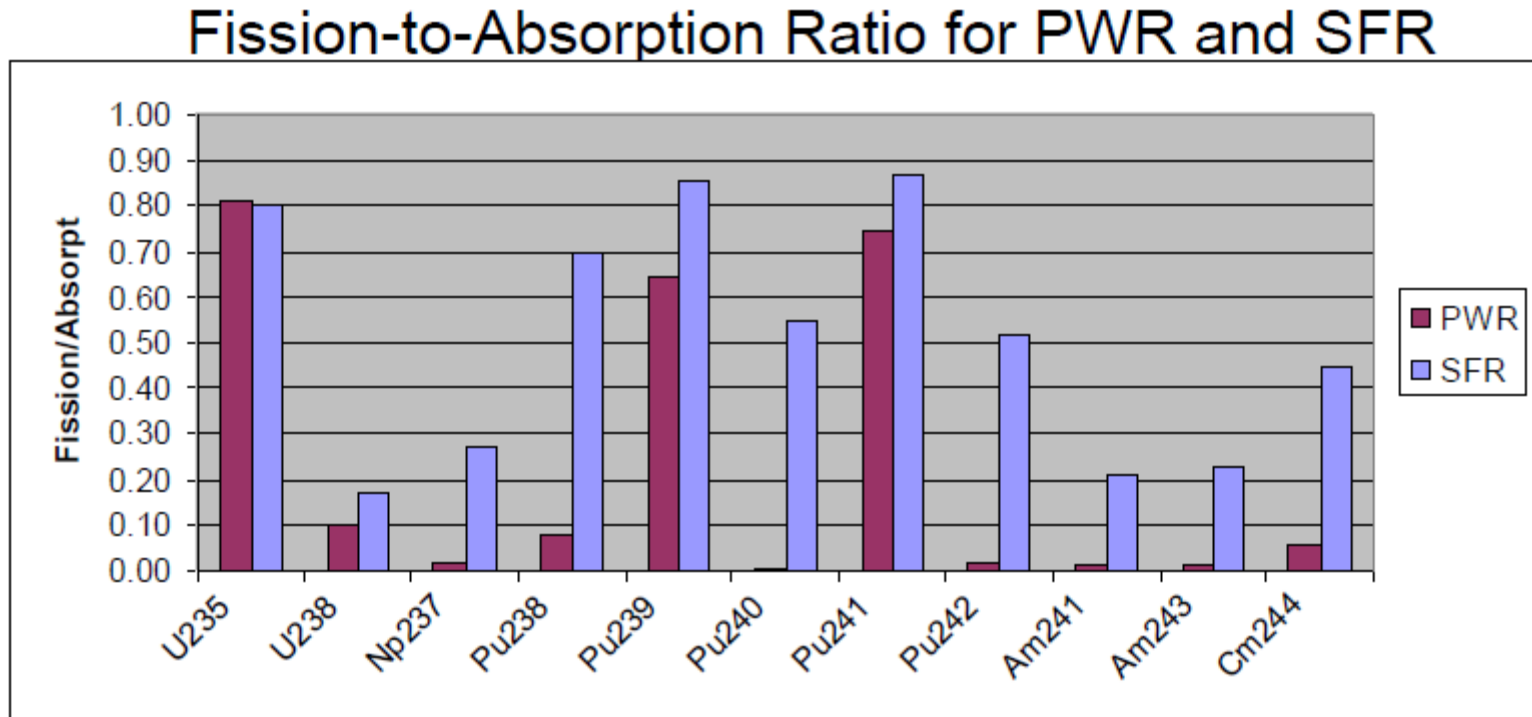
Np-237 fission/absorption ratio dependence on deuteron energy



Beam deuteron energy [GeV]	Fission to absorption ratio $I_f/(I_f+I_c)$	Error	%Error
2	0.34	0.05	14.05%
4	0.28	0.04	12.91%
8	0.35	0.06	16.58%

Literature example of actinide fission/absorption ratio

M. Salvatores, G. Palmiotti; Radioactive waste partitioning and transmutation within advanced fuel cycles: Achievements and challenges; Progress in Particle and Nuclear Physics 66 (2011) 144–166.



Conclusions

- Two ways of Np-237 interaction with neutrons – fission and capture.
- Fast neutrons needed to destroy Np-237 – fission/capture ratio grows with neutron energy growth.
- Np-237 fission to capture ratio was measured on QUINTA setup for three deuteron energies 2, 4 and 8 GeV.
- The fission to capture ratio seems to be constant for the specified above energies.
- The fission/capture ratio is of order 0.5 for the three deuteron energies.
- The fission/absorption ratio is of order 0.3 for the three deuteron energies.

References

1. Samuel E. Bays; Reactor Physics Characterization of Transmutation Targeting Options in a Sodium Fast Reactor; INL/CON-07-12439
2. Frána J.: Program DEIMOS32 for Gamma-Ray Spectra Evaluation, J. Rad. Nucl. Chem., V. 257, No. 3 P. (2003) 583-587.
3. TABLE OF ISOTOPES, 8E
4. Fission Product Yields per 100 Fissions for ^{237}Np High-Energy Neutron Fission Decay, T.R. England and B.F. Rider, LA-UR-94-3106, ENDF-349 ; <http://ie.lbl.gov/fission/237nph.txt>
5. M. Salvatores, G. Palmiotti; Radioactive waste partitioning and transmutation within advanced fuel cycles: Achievements and challenges; Progress in Particle and Nuclear Physics 66 (2011) 144–166.

Thank you for attention

Back up slides

Some Np-237 fission product data – chains, production yield, decay

Fission Product Yields per 100 Fissions for ²³⁷Np High-Energy Neutron Fission Decay, T.R. England and B.F. Rider, LA-UR-94-3106, ENDF-349 ; <http://ie.lbl.gov/fission/237nph.txt>

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

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

