Seminarium UZ3

# Application of minor actinides as neutron fluency and average neutron energy detectors in the place of their location.

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# Application of minor actinides as neutron fluency and average neutron energy detectors in the place of their location.

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# 1. Proposed method of actinide sample application as neutron fluency detector.

- The fast neutron fluence measurement method consists in utilizing neutron irradiated actinide samples for estimating neutron fluence and average neutron energy inside the volume of samples. The idea is to search the neutron energy ($E_d$) for the ratio ($\alpha(E_d)$) of fission cross section ($\sigma_f(E_d)$) to capture cross section ($\sigma_c(E_d)$) of the selected actinide isotope from the nuclear data base that is equal to the measured ratio ($\alpha_m$) of fissioned ($N_{yf}$) and captured ($N_{yc}$) actinide isotopes (spectral indexes):

$$\alpha(E_d) = \frac{\sigma_f(E_d)}{\sigma_c(E_d)} = \alpha_m = \frac{N_{yf}}{N_{yc}} = \frac{\overline{\sigma}_f}{\overline{\sigma}_c} \qquad (1)$$

# 1. Proposed method of actinide sample application as neutron fluency detector.

Since the measured spectral indexes ($\alpha_m$) is defined as the ratio of average fission ($\bar{\sigma}_f$ ) and capture ($\bar{\sigma}_c$ ) cross sections so the ratio ($\alpha(E_d)$ of retrieved distinct fission ($\sigma_f(E_d)$) and capture ($\sigma_c(E_d)$) cross sections for the distinct neutron energy ($E_d$) from the nuclear data base describe the average values:

$E_d = \bar{E}$ ; $\sigma_f(E_d) = \bar{\sigma}_f$ ; $\sigma_c(E_d) = \bar{\sigma}_c$

Having the average fission and capture cross section values we can evaluate the average neutron flux ($\bar{\phi}$) in the location of the actinide sample using the measured amount of fissioned and captured actinide isotopes.

## 1. Proposed method of actinide sample application as neutron fluency detector.

- The amount of neutron induced fissioned ($N_{yf}$) and neutron captured actinide isotopes ($N_{yc}$) in the actinide sample of volume $V_p$ can be expressed:
- $$N_{yf} = V_p \, \bar{\phi} \, N \, \bar{\sigma}_f \, t \qquad (2)$$
- $$N_{yc} = V_p \, \bar{\phi} \, N \, \bar{\sigma}_c \, t \qquad (3)$$

Where

$V_p$ - actinide sample volume [$cm^3$],

$\bar{\Phi}$ - average neutron flux in the place of actinide sample location [$n/cm^2 \cdot s$],

N – number of actinide isotopes in volume unit [$cm^{-3}$],

$\bar{\sigma}_f$ ; $\bar{\sigma}_c$ -average microscopic cross section for the reactions (n, f) and (n, γ) respectively [barns],

t - irradiation time [s].

# 1. Proposed method of actinide sample application as neutron fluency detector.

- Two different equations for fissioned ($N_{yf}$) and captured ($N_{yc}$) actinide isotopes should give the same average neutron flux value what is a proof for correct measurements.

$$\frac{N_{yf}}{N_{yc}} = \frac{V_p\ \bar{\phi}\ N\ \bar{\sigma}_f\ t}{V_p\ \bar{\phi}\ N\ \bar{\sigma}_c\ t} = \frac{\bar{\sigma}_f}{\bar{\sigma}_c}$$

# 1. Proposed method of actinide sample application as neutron fluency detector.

- It is useful to look closely at the ratios $\alpha = \sigma_f / \sigma_c$ of the capture and fission cross section of the Np.-237 isotope.
- The fission/absorption ratios are consistently higher for the fast-neutron spectrum. Thus, in a fast spectrum, actinides are preferentially fissioned, not transmuted into higher actinides.

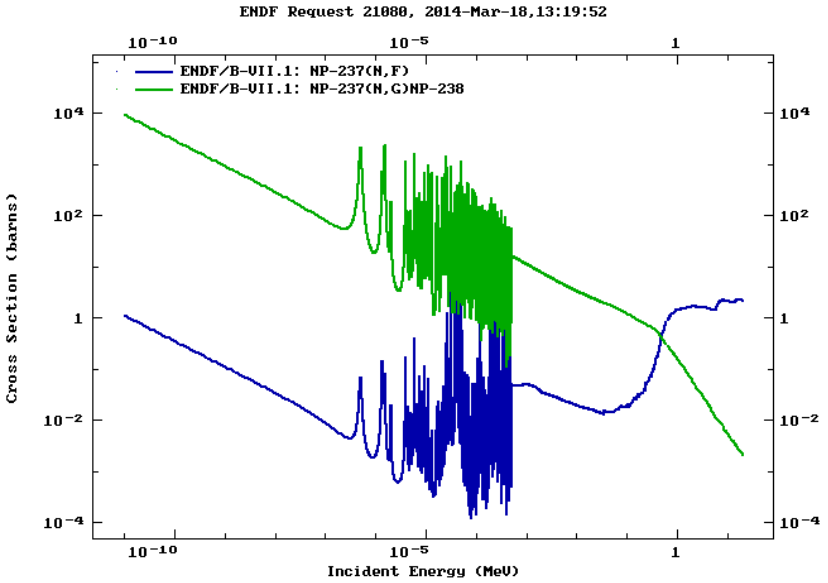


Fig. 1. Cross-sections of Np-237(n,g)Np-238 and Np-237(n,f) reactions.

## 2. Example of neptunium 237 sample experiments .
### 2.1. Quinta assembly – deeply subcritical assembly.

- The Quinta assembly , located at the Joint Institute for Nuclear Research (JINR), Dubna, Russia, has been used for application of fast neutron spectrum to the actinides incineration study. It is an assembly massive uranium target and lead shielding (see Figs 2a,2b).
- In Figs 2a - a view on the uranium target with supporting structures and plastics used for sample placement (detector's plates), and in Fig. 2b - a view on the lead shielding enfolding the target.

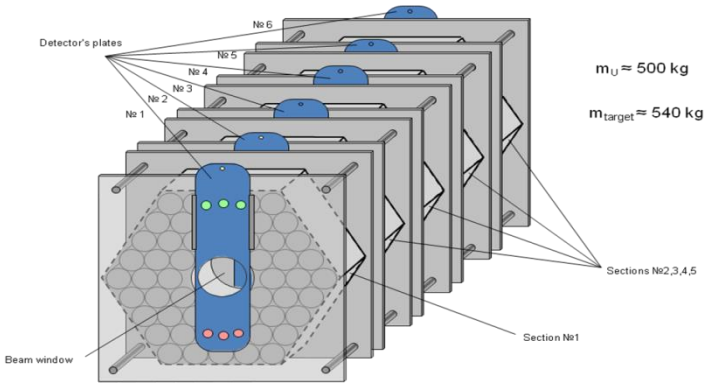


**Fig. 2a**

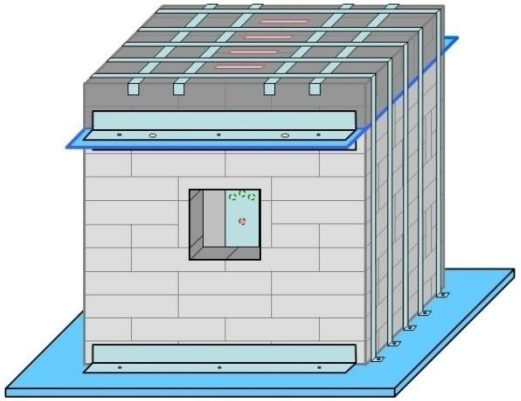

**Fig. 2b**

## 2. Example of neptunium 237 sample experiments .
## 2.1. Quinta assembly – deeply subcritical assembly. – cont.

- In Fig. 2c is presented the Quinta assembly with marked the window for the actinides sample location in the lead shielding .

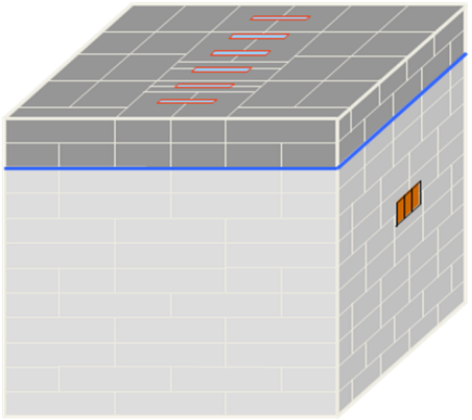

**Fig. 2c**

- In Fig 2d – Top view of actinide experimental samples position in the Quinta assembly (R = 180 mm, Z = 319 mm).

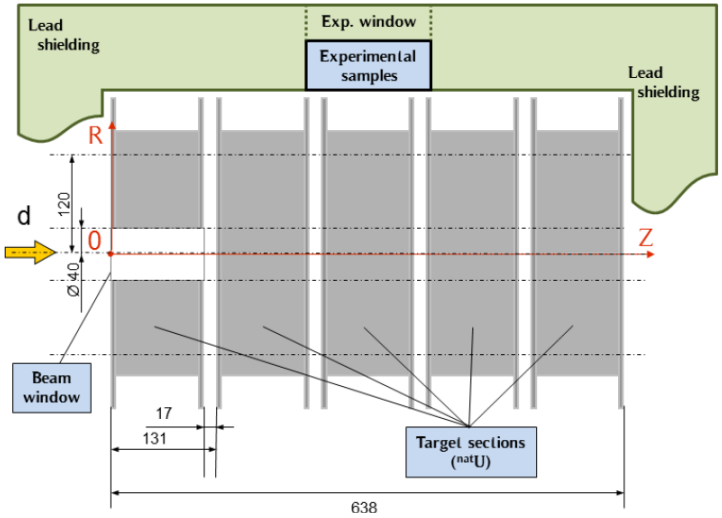


**Fig. 2d**

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## 2. Example of neptunium 237 sample experiments .
## 2.2. Irradiation details – cont.

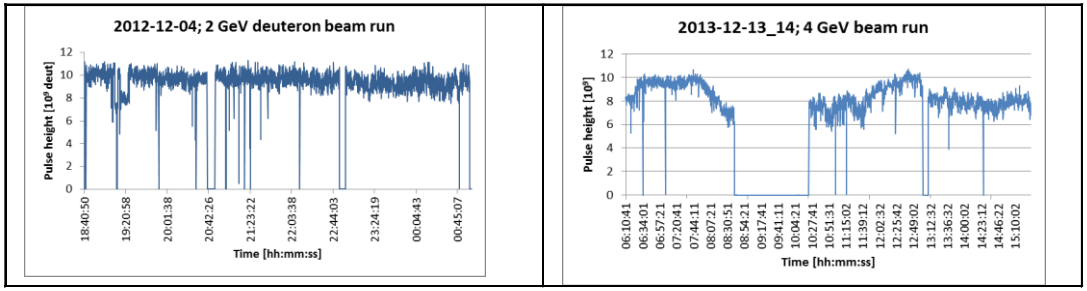



Fig. 3. Distribution of deuteron pulses during the irradiation period as measured with an ionisation chamber.

- The Quinta target was irradiated with a pulsed deuteron beam of energies 2, and 4 GeV. extracted from the Nuclotron accelerator. Total number of deuterons of the two irradiations are equal to 3.02(30) $10^{13}$, and 2.73(27) $10^{13}$ during the time of irradiation equal to 6.27, and 9.35 h respectively. The distribution of deuterons for the experimental session of December 2012 as extracted from the Nuclotron accelerator are shown in Fig. 3. Prior to the irradiation, several polaroid films were placed on the front of Quinta to ensure the deuteron beam was striking in the centre of the beam window.

# 2. Example of neptunium 237 sample experiments .
# 2.2. Irradiation details – cont.

- **Measurement of beam fluency.**

- The total number of deuterons to hit the Quinta target was determined by the activation of aluminium via beam induced $^{27}Al(d,3p2n)^{24}Na$ reactions. Three aluminium foil monitors were placed between 2 and 3 m away from the Quinta target to avoid backscattered neutrons contaminating the measurement.
- Only three experimental values for the $^{27}Al(d,3p2n)^{24}Na$ cross section in the GeV range (at 2.33 , 6.0 and 7.3 GeV are known. Therefore, the cross section at 2, 4 and 8 GeV energies were estimated from interpolating the curve fitted to experimental data in the energy range of 0.1–7.3 GeV.
- The $^{27}Al(d,3p2n)^{24}Na$ cross section was found to be 15.4 ± 1.5 mb, 14.5 ± 1.5 mb, and 14.0 ± 1.4 for 2, 4 and 8 GeV deuterons, respectively.

# 2. Example of neptunium 237 sample experiments .
# 2.2. Irradiation details – cont. .

- **Measurement of beam position.**

- The beam position and beam shape was determined using two separate techniques. -The first involved measuring the yield of reaction products produced by beam induced reactions on natural copper foils (method described in [9]). -The other method utilized an array of fission track detectors measuring beam induced $_{nat}$Pb(d,f) reactions [10].
- Both the copper activation foils and fission track detectors were placed directly in front of the Quinta target on the surface of the lead castle. The beam shape was fitted to a Gaussian and the resulting FWHM (Full Width at Half Maximum) in the vertical and horizontal directions, as well as the positioning of the beam centre is presented in Table 1.

# 2. Example of neptunium 237 sample experiments .
# 2.2. Irradiation details – cont.

**Table 1**

**Details of the two Quinta irradiations performed at the Nuclotron accelerator. Xc and Yc refer to the coordinates of the beam centre on the x–y plane**

| | | |
|---|---|---|
| • Experimental session in December 2012 | | |
| • Incident ion | Deuteron | |
| • Ion energy | 2 GeV | 4 GeV |
| • Irradiation duration | 6.27h | 9.35h |
| • Deuterons on target | $3.02(30)\times10^{13}$ | $2.73(27)\times10^{13}$ |
| • $X_c$ (cm) | $2.0 \pm 0.2$ | $2.1 \pm 0.1$ |
| • $Y_c$ (cm) | $0.0 \pm 0.1$ | $-0.3 \pm 0.2$ |
| • $FWHM_x$ (cm) | $2.2 \pm 0.3$ | $1.4 \pm 0.2$ |
| • $FWHM_y$ (cm) | $1.5 \pm 0.3$ | $0.9 \pm 0.1$ |

# 2.3. Neptunium 237 transmutation measurement.

## 2.3.1. Evaluation method of the number of Np.-237 atoms which undergo fast-neutron-induced fission and capture.

- After irradiation, the samples were removed and transported away to be analyzed with gamma spectrometry. The actinide samples were measured using a HPGe n-type coaxial detector, manufactured by CANBERRA. Measurements began ~1.5 h after irradiation had stopped, continuing for up to 6 days afterwards. The spectra collection times ranged from 15 min to just over 3 h. All spectra were analyzed using the DEIMOS program. The total yield of observed isotopes per one gram of activated material and one deuteron was calculated $N_{yield}$. The $N_{yield}$ is calculated accordingly to formula:

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

- Where $S_\gamma$ – gamma peak area, $m$ – activation sample mass [g], $\varepsilon_p$ – gamma spectrometer efficiency, $I_\gamma$ – correction for gamma line intensity [%], $\phi$ - deuteron fluence, $COI$ – correction for gamma quanta coincidence, $\lambda$ – isotope decay constant, $t_+$ – cooling time, $t_{ir}$ – irradiation time, $t_{real}$ – real time of measurement, $t_{live}$ – live time of measurement.

## 2.3. Neptunium 237 transmutation measurement.
## 2.3.1. Evaluation method of the number of Np.-237 atoms which undergo fast-neutron-induced fission and capture. – cont.

The procedure of evaluation of actinide fission rate per deuteron and per gram ($I_{f\gamma}$) consists in the calculation of number of produced nuclei $N_{yield}$ for each isotope and then dividing it by isotope production efficiency per 1 fission:

$$I_{f\gamma} = \frac{N_{yield}}{\gamma_f}$$

where

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

Gamma-ray spectrum analysis was carried out in a well established manner. The measured activities of considered isotopes at end of irradiation (EOI) were corrected for decay during the irradiation according to known deuteron beam profile, for decay between EOI and start of counting (cooling time) and for decay during the counting time and also not forgetting about dead time correction ($t_{real}/t_{live}$). Thus one can calculate the relative number of considered produced isotopes at the end of irradiation for each measurement.

## 2.3. Neptunium 237 transmutation measurement.

### 2.3.2. Experimental data of Neptunium 237 measurement.

- From gamma-ray spectrum of the irradiated Np-237 we have selected the following fission products $^{133}$I, $^{135}$I, $^{97}$Zr, $^{132}$I and $^{92}$Sr (see Table 2) which we have used to evaluate the fission rate per one gram Np-237 and per one deuteron applying the equation presented above.

Table 2. Fission fragment data of fast neutron induced fission of Np.-237.

| Isotope | $T_{1/2}$ | $\gamma$ line [keV] | $I_\gamma$ [%] | $\gamma_f$ [%] cumulate |
|---|---|---|---|---|
| Sr-92 | 2.66h | 1383.93 | 90 | 4.01 |
| Zr-97 | 16.744h | 507.64 | 5.03 | 5.35 |
| Nb-97 | 72.1 min | 1024.4 | 1.09 | 5.38 |
| I-132 | 2.295h | 772.6 | 75.6 | 4.39 |
| I-133 | 20.87h | 529.87 | 87 | 4.45 |
| I-135 | 6.57h | 1260.41<br>1131.51 | 28.7<br>22.6 | 4.16 |
| Cs-137 | 30.04y | 661.66 | 85.1 | 5.11 |
| Cs-138 | 33.4 min | 462.8 | 30.7 | 4.81 |
| La-142 | 91.1 min | 641.28 | 47.4 | 4.50 |

## 2.3. Neptunium 237 transmutation measurement.
## 2.3.2. Experimental data of Neptunium 237 measurement – cont.

Fig. 4a

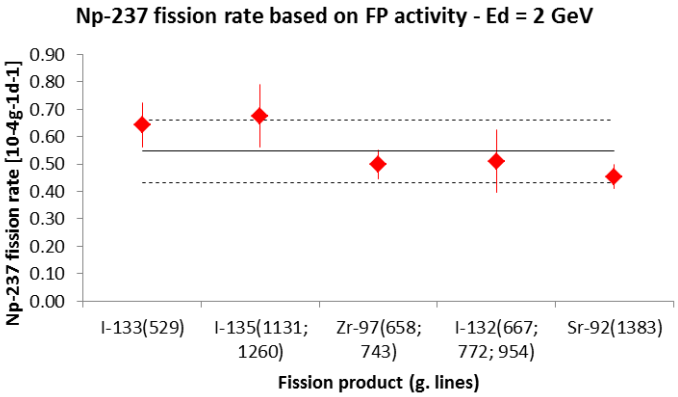
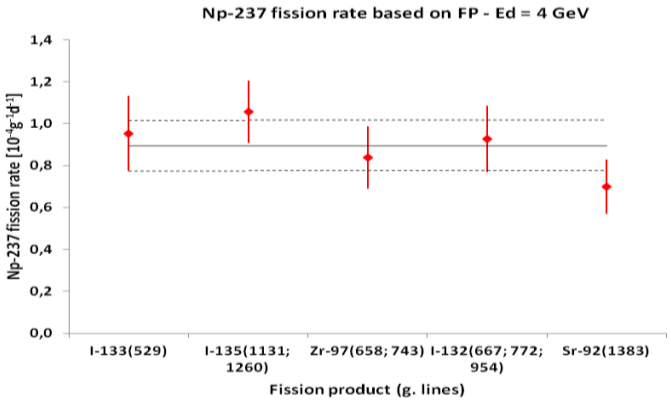



Fig. 4b




- The resulting normalized fission rate $I_{f\gamma}$ values are presented in Fig.4a and Fig. 4b for deuteron beams of 2 and 4 GeV respectively:
- Fig. 4a. Fission rate for different fission products of Np.-237 sample of 0.987 g weight for deuteron beam of 2 GeV energy.
- Fig. 4b. Fission rate for different fission products of Np.-237 sample of 1.115 g weight for deuteron beam of 4 GeV energy.

## 2.3. Neptunium 237 transmutation measurement.
## 2.3.2. Experimental data of Neptunium 237 measurement – cont.

- Moreover, considering the two different actinide neptunium 237 samples after irradiation we have focused on the neutron capture (n,γ) reaction leading to formation of Np-238. From gamma-ray spectrum of the irradiated Np-237 we have selected five gamma ray lines shown in Table 3, which we have used to evaluate the capture rate per one gram of Np-237 and per one deuteron applying the equation presented above.

Table 3. Gamma line data of Np.-238 formed in Np-237(n,γ)Np-238 reaction.

| Isotope | $T_{1/2}$ | γ line [keV] | $I_\gamma$ [%] |
|---|---|---|---|
| Np-238 | 2.117d | 984.45 | 27.8 |
| | | 1028.54 | 20.38 |
| | | 1025.87 | 9.65 |
| | | 923.98 | 2.869 |
| | | 962.77 | 0.702 |

- Neutron capture (n,γ) reaction was used for transmutation and γ-rays of the product nuclei was measured:
- $^{237}Np(n,\gamma)\ ^{238}Np$ ($\beta^-$ , $T_{1/2}$ = 2.12 d ) ⇒ short lived decay chain.

# 2.3. Neptunium 237 transmutation measurement.
## 2.3.2. Experimental data of Neptunium 237 measurement – cont.

**Fig. 5a**

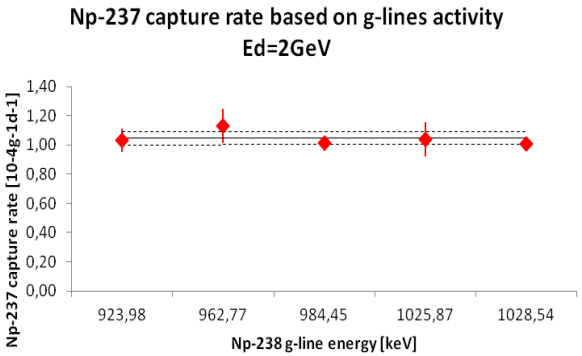
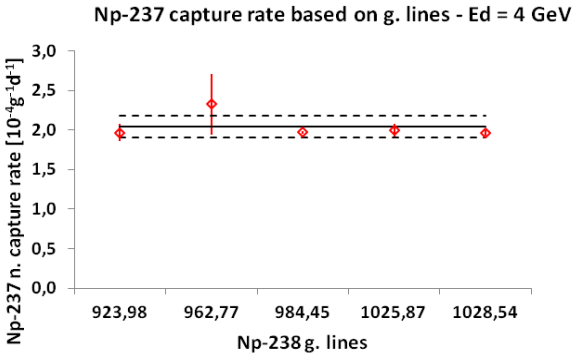



**Fig. 5b**




- The resulting normalized capture rate values for different gamma lines of Np-238 are presented in Fig.5a and Fig. 5b for deuteron beams of 2 and 4 GeV respectively:
- Fig. 5a. Capture rate for different gamma lines of Np-238 for deuteron beam of 2 GeV energy for Np.-237 sample of 0.987 g weight .
- Fig. 5b. Capture rate for different gamma lines of Np-238 for deuteron beam of 4 GeV energy for Np.-237 sample of 1.115g weight .

# 2.3. Neptunium 237 transmutation measurement.
## 2.3.2. Experimental data of Neptunium 237 measurement – cont. .

- Fission rate and capture rate of Np.-237 results for two deferent samples and two deferent deuteron energy are presented in Fig. 6 beside.

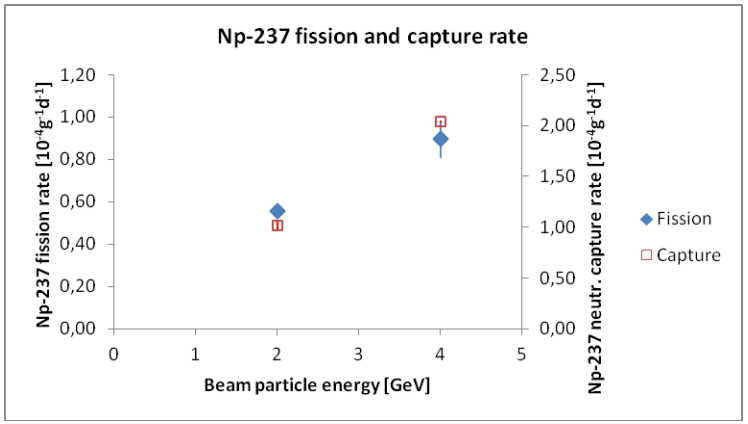



**Fig. 6**

- Using the experimental data from Fig. 6 we could evaluate Fission/Capture ratio of Np-237 versus deuteron energy in fast neutron spectrum of Quinta assembly (see Fig. 7 beside).

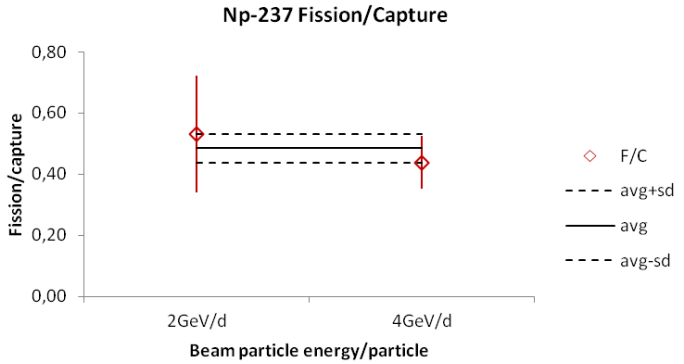



**Fig. 7**

# 2.3. Neptunium 237 transmutation measurement.
## 2.3.2. Experimental data of Neptunium 237 measurement – cont.

- Comparing the Fission/Capture ratio of Np-237 versus deuteron energy in fast neutron spectrum of Quinta assembly presented in Fig. 7 with the ratio in SFR (Sodium Fast Reactor) equal to about 0.25 (see Fig. 8) we can see that the incineration of Np-237 is twice higher in fast neutron spectrum of ADS (accelerator driven system) – Quinta assembly than in SFR.
- Fig. 8 is copied from the paper of M. Salvatores a, G. Palmiotti ;Radioactive waste partitioning and transmutation within advanced fuel cycles: Achievements and challenges; Progress in Particle and Nuclear Physics 66 (2011) 144–166.

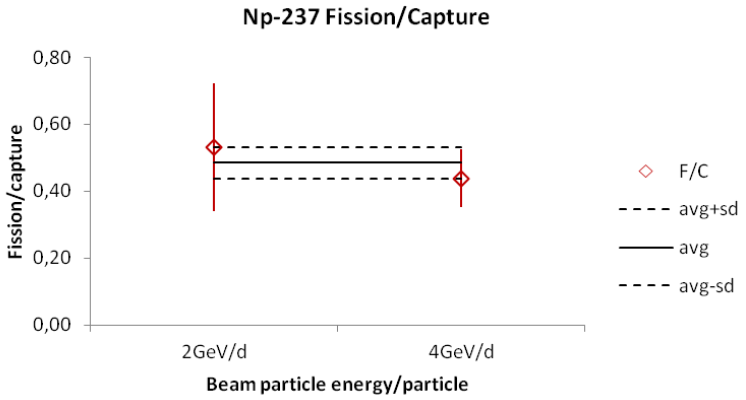



**Fig. 7**

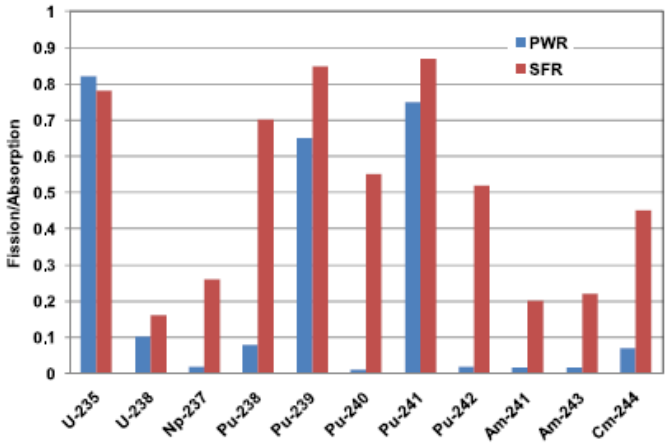



**Fig. 8**

## 2.3. Neptunium 237 transmutation measurement.

## 2.3.2. Experimental data of Neptunium 237 measurement – cont.

Table 4. Compilation of incineration rate and capture rate of Np.-237 results.

| Deuteron beam energy | Ed = 2 GeV | Ed = 4 GeV |
|---|---|---|
| Deuteron fluency | $3.02(10)\times 10^{13}$ | $2.73(10) \times 10^{13}$ |
| Mass of Np.-237 sample [g] | 0.987 | 1.115 |
| Number of Np-237 fissions per gram sample | $(1.12\pm0.393)\times10^{9}$ | $(1.69\pm0.698)\times10^{9}$ |
| Number of neutron captured Np-237 atoms per gram sample | $(2.13\pm0.\ 271) \times10^{9}$ | $(4.73\pm0.609) \times10^{9}$ |
| Fission rate per gram sample and deuteron | $(3,72\pm1,3)\times10^{-5}$ | $(6.20\pm2,56)\times10^{-5}$ |
| Capture rate per gram sample and deuteron. | $(7.064\pm0.897) \times10^{-5}$ | $(1.73\pm0.223 \times10^{-4}$ |
| Fission/Capture rate | 0.53±0.195 | 0.36±0.154 |

# 3. Application of the proposed method for assignment the neutron fluency and average neutron energy in the place of location the neptunium 237 sample.

Table 5. The fission/absorption ratios in function of neutron energy for neptunium 237.

| Neutron energy [MeV] | Fission cross section σ(n,f) [barn] | Capture cross section σ(n,γ) [barn] | σ(n,f)/ σ(n,γ) |
|---|---|---|---|
| 0.3 | 0.073 | 0.681 | 0.107 |
| 0.37 | 0.142 | 0.594 | 0.239 |
| 0.38 | 0.152 | 0.579 | 0.262 |
| 0.39 | 0.169 | 0.564 | 0.299 |
| 0.4 | 0.183 | 0.548 | 0.333 |
| 0.41 | 0.211 | 0.533 | 0.395 |
| 0.42 | 0.239 | 0.519 | 0.461 |
| 0.43 | 0.268 | 0.504 | 0.531 |
| 0.44 | 0.297 | 0.491 | 0.605 |
| 0.45 | 0.325 | 0.478 | 0.679 |
| 0.46 | 0.348 | 0.464 | 0.750 |
| 0.50 | 0.447 | 0,411 | 1.087 |

## 3. Application of the proposed method for assignment the neutron fluency and average neutron energy in the place of location the neptunium 237 sample – cont.

- If we assume that fission/absorption ratio of cross section is equivalent to the fission to capture ratio measured for Np-237 equal to 0.53, we can state that it corresponds to the average neutron energy 0.43 MeV,
- This energy we can define as an average neutron energy of neutron spectrum in location of actinide in the QUINTA assembly.

## 3. Application of the proposed method for assignment the neutron fluency and average neutron energy in the place of location the neptunium 237 sample – cont.

- So, applying the equation presented above we have evaluated the average neutron flux in the location of the actinide sample Np-237 for the case of deuteron beam equal 2 GeV using once the measured amount of fissioned actinide atoms and once the neutron captured actinide isotopes.
- The results are very alike:
- the average neutron flux 7.29 $10^7$ n/cm$^2$·s and 7.37 $10^7$ n/cm$^2$·s respectively.
- In the next slide, compilation of average neutron flux and neutron fluency in the location of the actinide neptunium 237 in the QUINTA assembly using for evaluation the measured number of fissions and captures in the samples for two experiments are presented.

# 3. Application of the proposed method for assignment the neutron fluency and average neutron energy in the place of location the neptunium 237 sample – cont.

Table 6. Compilation of average neutron flux and neutron fluency in the location of the actinide neptunium 237 in the QUINTA assembly using for evaluation the measured number of fissions and captures in the samples for three experiments

| **Deuteron beam energy** | **2 GeV** | **4 GeV** |
|---|---|---|
| Average neutron flux using for evaluation the number of fissions in the sample [n/cm$^2$·s] x10$^7$ | 7.29 | 10.08 |
| Average neutron flux using for evaluation the number of captures in the sample [n/cm$^2$·s] x10$^7$ | 7.37 | 10.09 |
| Neutron fluency using for evaluation the number of fissions in the sample [n/cm$^2$] x10$^{12}$ | 1.64 | 3.393 |
| Neutron fluency using for evaluation the number of captures in the sample [n/cm$^2$] x10$^{12}$ | 1.66 | 3.396 |
| **Average neutron energy [MeV]** | **0.43** | **0.4** |

## 4. Conclusions

- In conclusion of this analysis it is obvious that actinide samples can be used as average neutron flux detectors especially in the high neutron energy range what is difficult to measure.
- The proposed method let us to evaluate six parameters namely neutron fluence, average neutron flux, average neutron energy, average fission csross section, average capture cross section and fission spectral index using the two measured data: - amount of fissioned ($N_{yf}$) and captured ($N_{yc}$) actinide isotopes .
- Given the importance of high energy neutron measurement the actinide average neutron flux detectors could be a very useful tool.

- Thank you for the attention.