Transmutation of spent nuclear fuel

Summer student program

Dzhelepov Laboratory of Nuclear Problems

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1. Introduction

I was participant of 2015 summer student program in the Joint Institute for Nuclear Research in Dubna for eight weeks. During the practice I visited the reactor building and following laboratories:

- Dzhelepov laboratory of nuclear problems – where I worked
- Veksler and Baldin laboratory of High energy physics
- Frank laboratory of neutron physics
- Flerov laboratory of nuclear reactions

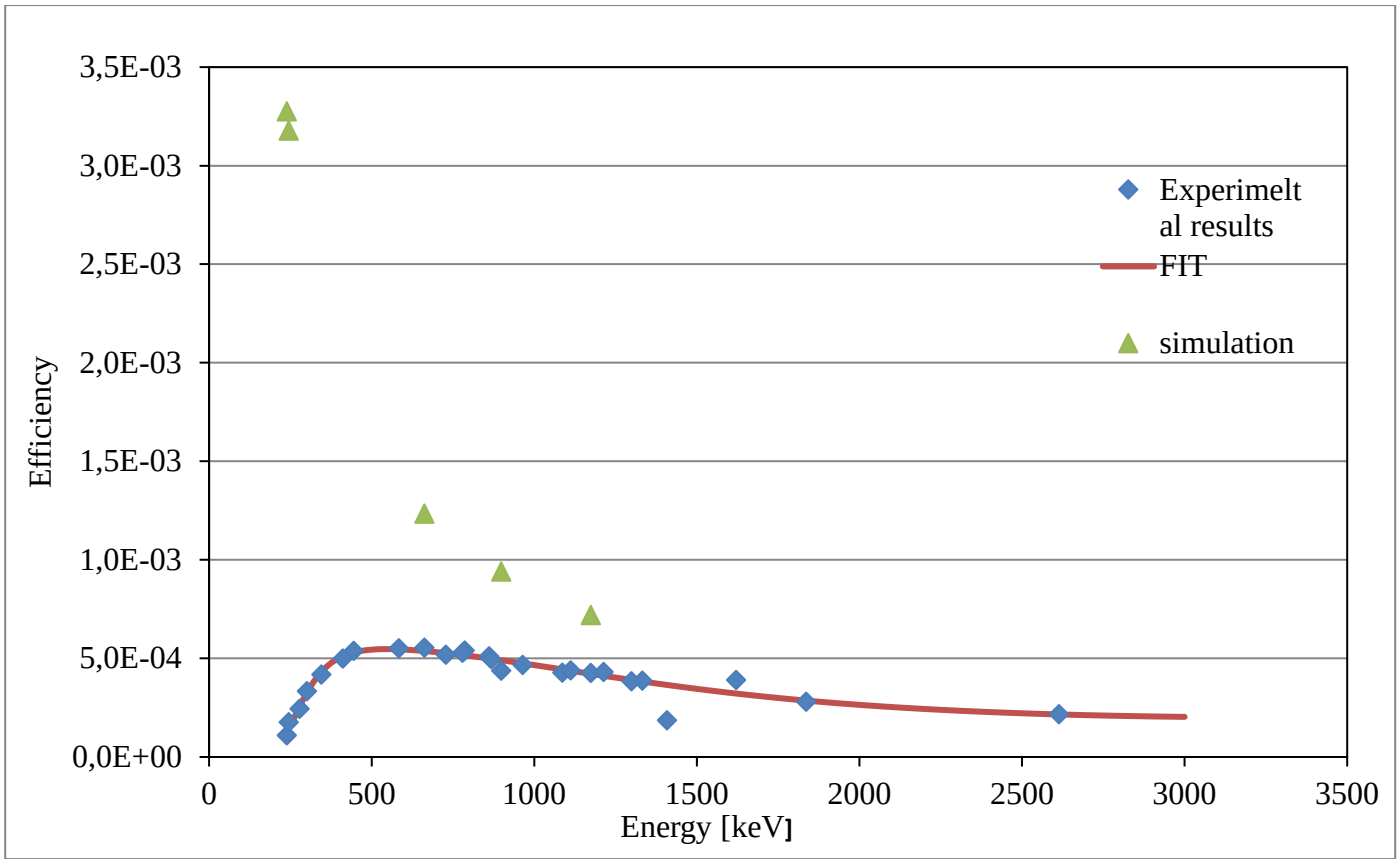
The aim of our project was to investigate the transmutation rates in ^{137}Np and ^{239} Pu in a massive neutron spallation target – QUINTA (m = 512 kg). The target was irradiated with proton beam of kinetic energy 660 MeV provided by the Phasotron accelerator at the Joint Institute for Nuclear Research (JINR) in Dubna. The neutron-induced transmutation of the actinide samples was measured by methods of gamma-ray spectrometry with HPGe detectors. Results of measurement are expressed in the form of both the individual reaction rates and average fission transmutation rates [1].

During the program, I had several task to do.

My first task was to obtain HPGe detector efficiency. Five different radio nuclides are used as calibration sources – ^{88}Y, ^{60}Co, ^{252}Eu, ^{137}Cs, ^{228}Th. The efficiency was measured with spectroscopy filter (1 mm Pb, 0.5 mm Cd 0.5 mm Cu) at position 10 (at 15.14 cm from the detector) in the spectrometry system. With special nuclear spectroscopy software, the measured spectra are analyzed. After the evaluation of the spectrum an output file is obtained, consisting important data to calculate the full-energy-peak efficiency rate. The formula [2], used for the calculations is

$$\mathrm{eff} = \frac{S\,\lambda\,\frac{t\,real}{t\,live}}{A_o e^{-\lambda t decay}\,(1-e^{-\lambda t real})\,I\gamma} \qquad (1)$$

The dependence of the efficiency on the energy is presented in the Graph. 1



Graph. 1 Efficiency of HPGe detector at measurement position 10

My second task was to create a model of the HPGe detector and simulate the full-energy-peak-efficiency curve. [3]
Using Monte-Carlo method I simulated the detector and the efficiency. During the creation of the input file I used VisedX24 program to visualize the model.

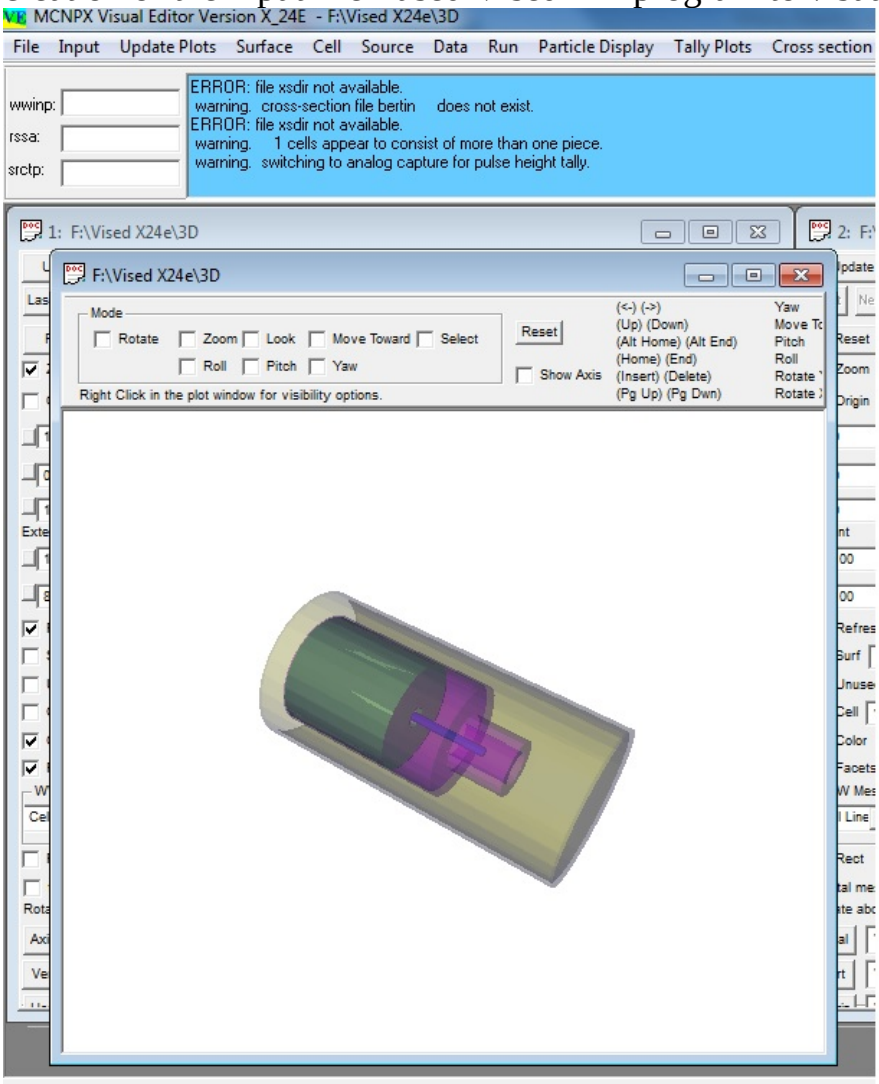


Figure 1 3D model of the detector

In the Graph 1 there is presented the result of the simulation without considering the spectroscopy filter. It can be seen its importance as regards cutting the low energy gamma-rays and the difference between simulated and experimental data because of that.

My third task was to measure actinide samples with HPGe detectors, to identify residual nuclei and to obtain both reaction and transmutation rates.

Deimos32.exe software is used in an interactive mode to find and fit gamma peaks in measured spectra. The program finds a possible peak, fits it with a gamma function and determines the gamma peak area $S_\gamma(E)$ and the energy peak position E. The fitted are of the peak $S_\gamma(E)$ is the number of fully registered gamma-quanta with energy E. The energy peak position E is determined using precise calibration points E_1, E_2. Three output files are obtained *.prn *.rem and *.dsk containing different information.

Six spectra for ^{237}Np sample were analyzed and five for ^{239}Pu. After the results were stored the output files were put on following procedure. Using Ruby software package corrections were made in the obtained files. Correction for:

- Peak tails
- Integral beam intensity
- Background radioactivity
- Single and double escape peak
- Full energy peak efficiency correction
- Self-absorption
- Beam fluctuation

For both samples, placed in position 10 in the spectrometry system, output files were obtained, consisting information about residual nuclei, their energies and reaction rates.

The data are presented in following tables:
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Table 1 Reaction rates for residual nuclei in the ^{237}Np sample

Np		
Isotope	R	dR
91Sr	2,256E-28	3,616E-29
92Sr	2,110E-28	1,474E-29
95Zr	3,189E-28	3,010E-29
97Zr	3,129E-28	1,173E-29
103Ru	1,835E-28	4,216E-29
133I	3,333E-28	1,923E-29
135I	3,335E-28	1,232E-29
238Np	1,063E-26	1,239E-28

Table 2 Reaction rates for residual nuclei in the ^{239}Pu sample

Pu		
Isotope	R	dR
88Kr	1,167E-28	1,259E-29
91Sr	3,074E-28	1,349E-29
92Sr	3,283E-28	1,605E-29
95Zr	4,980E-28	1,785E-29
97Zr	6,075E-28	2,069E-29
103Ru	7,888E-28	1,609E-29
105Ru	6,983E-28	1,567E-29
115Cd	7,799E-29	8,597E-30
131I	3,503E-28	1,414E-29
133I	8,161E-28	1,217E-29
135I	7,202E-28	8,200E-30
140Ba	6,219E-28	3,864E-29
143Ce	4,985E-28	1,080E-29
238Np	2,825E-29	2,035E-30

In order to obtain average fission rate in the actinide samples, the values of individual reaction rates were divided by the cumulative fission yields from the ENDF/B-VII.1 library [4], for the particular energies: 0.025 eV, 0.500 MeV, 2 MeV, and 14 MeV.

Table 3 Average reaction rate divided by the cumulative fission yield

Sample	0,0253 eV	500 keV	2 MeV	14MeV
Reaction rate	10^{-27} atom^{-1}proton^{-1}	10^{-27} atom^{-1}proton^{-1}	10^{-27} atom^{-1}proton^{-1}	10^{-27} atom^{-1}proton^{-1}
^{237}Np	4,853±0,341	5,010±0,158	-	5,863±0,446
^{239}Pu	11,02±0,38	11,210±0,398	11,365±0,454	0,969±0,638

Finally, I selected the results with the minimum relative uncertainty and divided them by unit beam energy:

For ^{237}Np sample: $(7{,}59 \pm 0{,}24) * 10^{-27}$ atom^{-1}proton^{-1} GeV^{-1}
For ^{239}Pu sample: $(16{,}07 \pm 0{,}57) * 10^{-27}$ atom^{-1}proton^{-1} GeV^{-1}

The results will be compared with the results of Kamila Wilczyńska and published in a join publication in future.

From the obtained results you can see that plutonium sample shows that it is more sensitive to the transmutation than the neptunium sample.

In conclusion, during the summer program, I got familiar with the transmutation problem. I learned how to create the efficiency of a detector and got familiar with the Monte-Carlo method and DEIMOS program for spectra analysis.

Acknowledgement

I want to thank my supervisor Lukáš Závorka for the opportunity to work in his group and for all the things that I learned from him, and also all the colleagues for their help.

References

[1] L.Zavorka et al. *Neutron-induced transmutation reactions in ^{237}Np, ^{238}Pu, and ^{239}Pu at the massive natural uranium spallation target,* NIMB 349, 2014
[2] Debertin K. and Helmer R. *Gamma - and X-ray spectrometry with semiconductor detector*
[3] http://newuc.jinr.ru/img_sections/file/project/2015/Student_practice_2015_ZVT.pdf
[4] M.B. Chadwick et al., Nucl. Data Sheets 112 (2011) 2887.