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REPORT

Transmutation of Spent Nuclear Fuel

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ACKNOWLEDGEMENT

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ABSTRACT

Following experiment was conducted: neutron-induced transmutation reactions in ^{237}Np and ^{239}Pu at the massive natural uranium spallation target. Results of experiment were expressed in the form of both the individual reaction rates and the average fission transmutation rates. After calculations a comparison with previously obtained data was conducted and conclusions were drawn.

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INTRODUCTION

From the beginning of the nuclear era the presence of long-lived actinides and fission products in the spent nuclear fuel stands as great disadvantage of nuclear energy. The disposal of spent nuclear fuel is crucial, yet demanding, if concerning the usage of nuclear power plants for producing electricity. The spent nuclear fuel is classified as the high level radioactive waste and must therefore be handled properly.

There are several different ways of management of spent nuclear fuels.
One approach is to 'simply' bury it deep underground as it is, using, of course, several types of barriers as copper casks, thick concrete shielding and granite rocks (Swedish model[1]).
The other way is to reuse the spent nuclear fuel since it still contains energy (95% uranium and 1% plutonium, which can be used to produce electricity). This is a kind of recycling, where fractions of uranium and plutonium are recovered and then used for production of new nuclear fuel (e.g. MOX)[2]. The rest of the spent fuel with long-lived actinides and fission products has to be stored properly and for long time.
The third and the most interesting method of management of spent nuclear fuel is transmutation. Transmutation is a 'modern alchemy', but in contrast to the ancient pseudoscience, transmutation works. By definition it is a conversion of one isotope to another. Thanks to this process the negative effects of radioactive wastes can be reduced. Namely, the most dangerous long-lived actinides will transmute into other, less dangerous isotopes. Transmutation can be done in light water reactors, fast neutron reactors and in sub-critical reactors. Unfortunately, transmutation in light water reactors (widely used in the power engineering industry) is not effective and is not considered to be the future of transmutation. That is why there is an urge to develop other methods, like fast neutron reactors and sub-critical reactors.

In this report I will focus on the sub-critical reactors, and to be more precise on neutron-induced transmutation reactions in ^{237}Np and ^{239}Pu at the massive natural uranium spallation target.

[1] http://www.skb.com/
[2] http://www.areva.com/

AIM

The purpose of the experiment was to study transmutation rates – in particular the neutron-induced fission rates – in the actinide samples located in the secondary neutron field generated in the spallation process by the 660 MeV proton beams irradiating the massive uranium spallation target QUINTA. Exclusively, transmutation rates of ^{237}Np and ^{239}Pu were investigated.

EXPERIMENTAL SETUP

Figure 1 – QUINTA target

The QUINTA target consists of 512 kg of metallic natural uranium in aluminum envelope and 10 cm thick lead shielding 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.

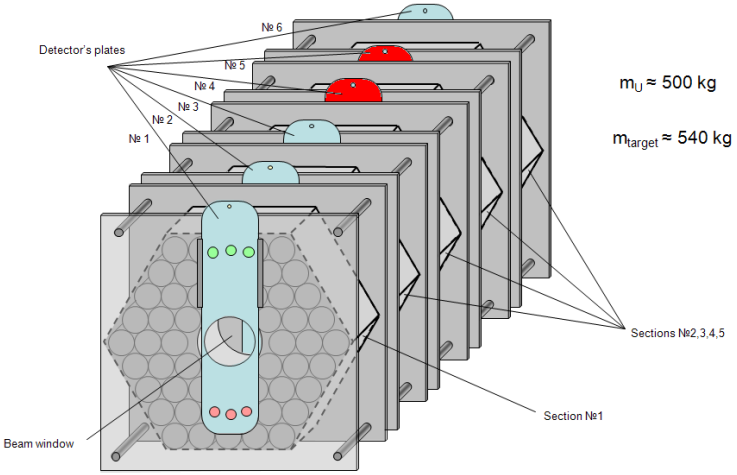


Figure 2 – My new friend high-purity germanium (HPGe) detector and I

The neutron-induced transmutation of the actinide samples was measured by methods of gamma-ray spectrometry with high-purity germanium detector (HPGe). For ^{137}Np sample 5 measurements has been conducted over the time and for ^{239}Pu samples the number of measurements was equal to 6.

METHOD

In this chapter subsequent steps that led to the final results will be described.

Efficiency calibration

If interpreting the data from the gamma-ray spectroscopy there is a need of conducting of an efficiency calibration of the HPGe detector. There are several ways of how the term efficiency can be defined. In general, it is the ratio between the response of a detector to the real quantity of incidents a phenomenon measured. For the sake of this experiment, this section will focus on the two of them, namely: *full energy peak efficiency* and *total efficiency*.

Full energy peak efficiency

The full energy peak efficiency is conducted to relate the peak area in investigated spectrum to the amount of the radioactivity it represents. It relates the peak area, at a specific energy, to the number of gamma-rays emitted by the source.

Total efficiency

The total efficiency relates the number of gamma-rays emitted by the source to the number of counts detected anywhere in the spectrum. This considers not only the full energy peak but also incorporates all incomplete absorptions due to the Compton scattering and pair production phenomena.

To obtain both above spectra from measurements of 10 radioactive sources were used, namely: ^{22}Na, ^{54}Mn, ^{60}Co, ^{65}Zn, ^{88}Y, ^{113}Sn, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{228}Th. For each specific, known and high intensity energy in those spectra calculation of energy efficiency was conducted. Then a proper fit of a trend line was applied. As a result the full energy peak efficiency and total efficiency as a functions of energy were obtained.

Deimos32

For analysis of all obtained spectra Deimos32 program was used. This software applies the energy calibration based on postulated points and energies. An energy calibration is necessary due to the fact that the data from the detector come as the number of counts (incidents recorded) in the channel function. To obtain these data in the function of energy, the energy calibration is needed. It is performed by the investigation of spectra of a known radionuclide with a known activity and specific energies of gamma-rays emitted. Then, by knowing the channel positions of such peaks and its energies, a calibration can be conducted.

The Deimos32 program gives also possibility to investigate each spectrum in details. It is possible to determine the area of each peak, which is then necessary for further

calculations. In general, knowing the energy of the peak, it is possible to identify the radionuclide, knowing the area of a peak it is achievable to approximate the quantity of the radionuclide. Having those date measured at different time, it is possible to determine the reaction rate, so the rate at which the quantity of the radionuclide changes over time.

Ruby programming

Ruby is a programming language that was designed not to be easy, but natural. This language was used when creating the scripts for further examination of data obtained with the use of the Deimos32 program. A pack of previously created Ruby scripts consisted of:

- TailCor.rb

  Makes corrections of so called peak tails. Sometimes. At high energies one peak can be interpreted by the Deimos32 software as two peaks. This program sums the areas of both of them and recalculates the energy of the peak.

- TimeConst.rb

  Creates input file RBScript.INP from Deimos32 program outputs, which is used by the following programs.

- Nonlin64.rb

  Applies the correction for non-linearity of the data.

- Puregam.rb

  Filters out the background radioactivity of the measurement. The background radiation data is measured for a certain detector without any artificial source inside or around it.

- SepDepe.rb

  Eliminates the single escape peak (SEP) and the double escape peak (DEP) that arise due to the fact that at high gamma-ray energies the escape of annihilation radiation may occur.

- EffCor.rb

  Performs the full energy peak efficiency correction.

- AttCor.rb

  Determines the self-absorption of gamma-rays correction coefficient.

- TrueCoin.rb

  Applies the correction for the coincidence process.

- MidLit7.rb

Identifies isotopes in the investigated data.

- BeamCor.rb

Applies the correction for any fluctuations of the beam, if such occurred.

- TransCs9.rb

Calculates the number of residual nuclei, the reaction rate and their uncertainties for each gamma line identified.

- SigmaJ7.rb

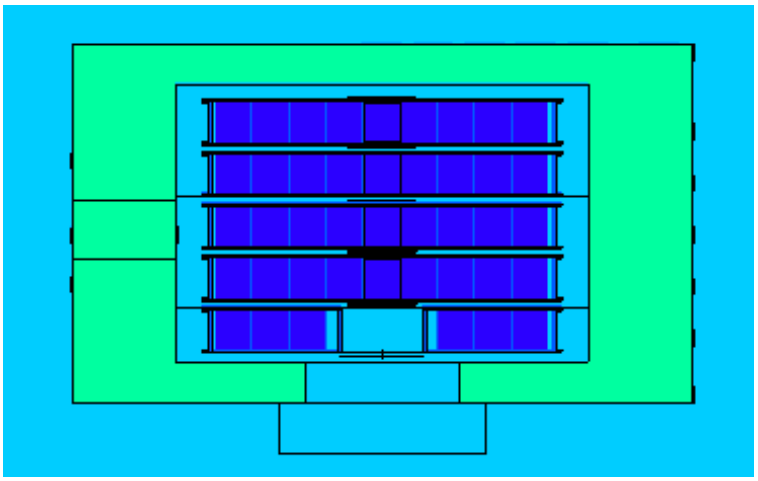
Reads all the output data sorting isotopes by name and subsequently by their energies.

After above processing, the data was further examined in order to draw valid conclusions.

Monte Carlo simulations

Monte Carlo methods are used for simulations of nuclear processes. It is possible to model any experiment using this software. Once it is incorporated in the safety analysis of nuclear reactors or experiments like this, once the experiments are used to valid and upgrade the software. The first step to perform any simulation is to create geometrical model of an examined set-up. The Monte Carlo dedicated software possesses its own language for creation of models. Further, during the simulation, the reconstruction of the reality takes place using statistical Monte Carlo methods. As a result of it is possible to receive highly probable results.

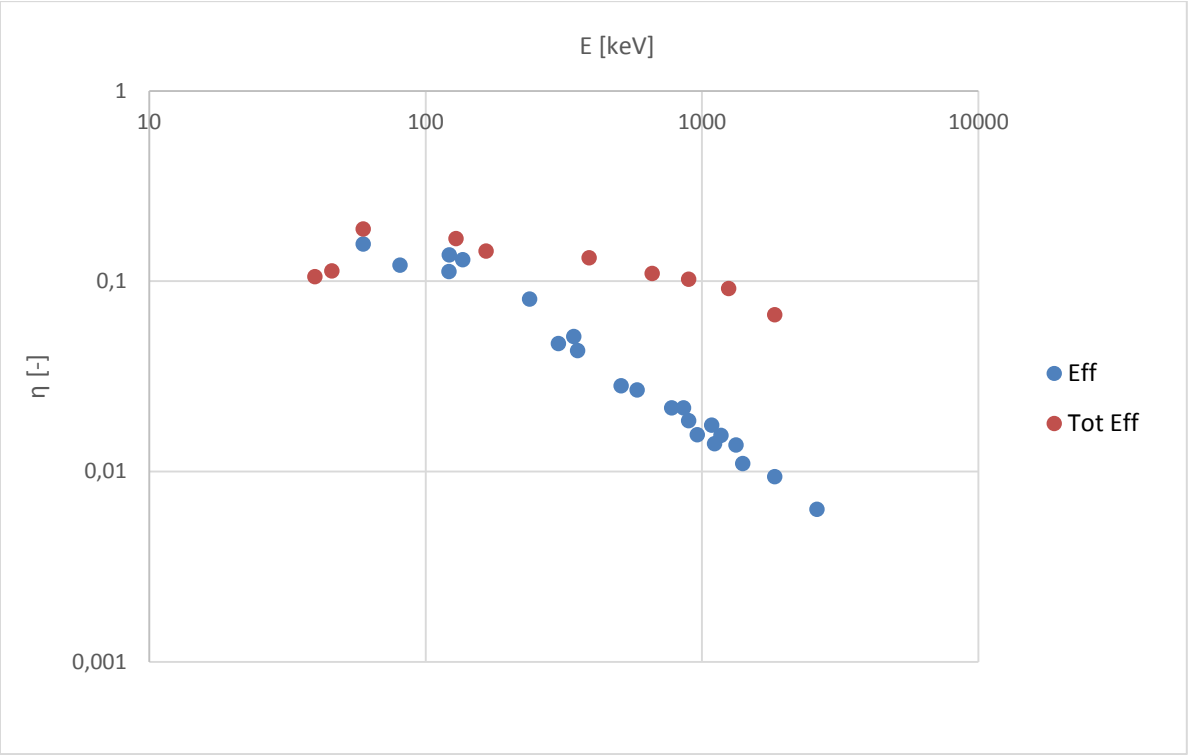
Figure 3 – Monte Carlo model of the QUINTA target



RESULTS

Full energy peak efficiency and total efficiency graphs were obtained.

Figure 4 – Full energy peak and total efficiency functions with regard to the energy



The reaction rates for residual nuclei for both samples were obtained.

Table 1 – Reaction rates with uncertainty for ^{239}Pu sample

Isotope	Reaction rate	Uncertainty
^{87}Kr	1.454E-28	3.832E-29
^{88}Kr	2.313E-28	1.705E-29
^{91}Sr	5.171E-28	3.468E-29
^{92}Sr	4.807E-28	2.328E-29
^{95}Zr	9.214E-28	1.985E-29
^{97}Zr	1.049E-27	3.282E-29
^{103}Ru	1.299E-27	2.477E-29
^{105}Ru	1.149E-27	2.635E-29
^{127}Sb	1.066E-28	2.131E-29
^{129}Sb	2.694E-28	1.310E-29
^{131}I	5.374E-28	7.059E-29
^{133}I	1.519E-27	5.293E-29
^{135}I	1.246E-27	1.624E-29
^{140}Ba	1.021E-27	2.124E-29
^{143}Ce	7.998E-28	1.964E-29
^{147}Nd	6.269E-28	3.649E-28
^{238}Np	2.506E-29	4.593E-30

Table 2 – Reaction rates with uncertainty for ^{237}Np sample

Isotope	Reaction rate	Uncertainty
^{88}Kr	6.766E-29	4.590E-30
^{91}Sr	1.830E-28	2.905E-29
^{92}Sr	1.268E-28	5.818E-30
^{95}Zr	1.653E-28	5.595E-29
^{97}Zr	2.101E-28	1.127E-29
^{103}Ru	2.111E-28	4.028E-29
^{105}Ru	1.130E-28	7.142E-30
^{129}Sb	2.339E-27	1.394E-27
^{133}I	2.003E-28	5.237E-30
^{135}I	1.912E-28	3.472E-30
^{140}Ba	3.194E-28	6.092E-29
^{238}Np	6.923E-27	1.010E-28

Then, to obtain average fission transmutation rates of the actinide samples, the values of individual reaction rates were divided by the cumulative fission yields for the particular energies: 0.025 eV, 500 keV, 2 MeV, and 14 MeV.[3]

Table 3 – Average fission rates with uncertainty

Sample	**Reaction rate ×10^{-26} [per atom per proton]**			
	0,0253 eV	500 keV	2 MeV	14MeV
^{237}Np	0.32±0.03	0.32±0.03	-	0.43±0.02
^{239}Pu	-	1.97±0.03	2.01±0.05	2.20±0.16

Finally, for each sample the results with the lowest value of the relative uncertainty were selected. The fission transmutation rate per beam energy unit is as follows:

- for ^{237}Np sample: $(0.65 \pm 0.34) \times 10^{-26}$ [atom^{-1}proton^{-1} GeV^{-1}],
- for ^{239}Pu sample: $(2.99 \pm 0.52) \times 10^{-26}$ [atom^{-1}proton^{-1} GeV^{-1}].

[3] M.B. Chadwick et al., Nucl. Data Sheets 112 (2011) 2887

CONCLUSION

The fission transmutation rates in the actinide samples of ^{237}Np and ^{239}Pu were experimentally investigated in the field of secondary spallation and fission neutrons. For ^{237}Np sample the fission transmutation rate per beam energy unit is equal to (0.647 ± 0.342) ×10^{-26}, whereas for ^{239}Pu sample equals (2.99 ± 0.0521) ×10^{-26} [atom-^{1}proton^{-1} GeV^{-1}].

From these results first conclusion can be drawn, transmutation of ^{239}Pu is 5 times more probable than the transmutation of ^{237}Np. The reason for that are, obviously, different cross sections to fission and its dependency on energy.

Moreover, by comparison of this data to previous experiments, where the target was bombarded with deuterons, it is possible to draw more conclusions. In the mentioned experiment the fission transmutation rate per beam energy unit for ^{237}Np sample is equal to (0.81 ± 0.08) ×10^{-26}, whereas for ^{239}Pu sample equals (4.0 ± 0.04) × 10^{-26} [atom-^{1}deuteron^{-1} GeV^{-1}]. The beam of deuterons was between 1 and 8 GeV. In connection with the above it can be concluded that the change from deuterons to protons is an effort worthy change. Unfortunately, as a result the transmutation rate decreases, but the energy of protons used is significantly lower. One should also consider a different geometric arrangement of the experiments.

FURTHER WORK

In order to maximize the scientific outcome of this experiment there is some more work to do, namely:

1. The Monte Carlo simulation will be performed (with the use of the previously obtained models) in order to analyze the approprability of the results and to review the accuracy of the simulation codes.
2. The results of this part of the experiment will be compared with the results obtained by Milena Stoyanowa and Lukáš Závorka in one collaborative scientific paper. The aim is to draw the most suitable and useful conclusions.

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