U-238 fission and Pu-239 production in subcritical assembly

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Abstract. The project touches upon an issue of U-238 fission reactions and Pu-239 production reactions in subcritical assembly. The experiment took place in November 2014 at the Dzhelepov Laboratory of Nuclear Problems (JINR, Dubna) using PHASOTRON. Data of this experiment were analyzed in Laboratory of Information Technologies (LIT). Four MCNPX models were considered for simulation: Bertini/Dresnen, Bertini/Abla, INCL4/Dresnen, INCL4/Abla. The main goal of the project was to compare the experimental data and simulation results. We obtain a good agreement of experimental data and computation results especially for detectors placed besides the assembly axis. In addition, the U-238 fission reactions are more probable to be observed in the region of a higher particle energy spectrum, located closer to the assembly axis and the particle beam as well and vice versa Pu-239 production reactions were dominant in the peripheral region of geometry.

1 Introduction

The electromagnetic method of neutron generation using a spallation reaction in Accelerator Driven System (ADS) has been studied by many scientists worldwide [1-9]. The efficiency of the neutron production based on spallation reaction depends on many parameters such as: beam energy, kind of particles and kind and size of target [3, 4, 6]. These neutrons can be employed for transmutation of the long-lived isotopes existing in nuclear waste [5, 7, 9] and for nuclear energy production from thorium or spent uranium fuel [1, 2, 8, 9]. The main aim of this work is to analyze the U-238(n,f) and U-238(n,g) reactions in QUINTA assembly [7, 9]. This work presents a comparison of experimental data and calculated results predicted by MCNPX code [10].

2 The Experiment

The experiment was employed QUINTA assembly run with a PHASOTRON in Dzhelepov Laboratory of Nuclear Problems in JINR, Dubna (see [8] for more details). Experimental data come from experiment run by Professor Voronko and his group of scientists from Ukraine.

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The research described in this paper was a part of experimental collaboration "Energy and Transmutation of Radioactive Waste" as well as the experiment which is the source of data. Research connected with particle beam profile was run by Lukas Zavorka from Czech Republic. Experimental data were obtained for activation detector of natural uranium. Activation detectors were located on six aluminum holders at the distance R from assembly axis equal to 0, 4, 8, and 12 cm. These holders were placed between all sections and before the first and after the last section (Z = 0, 12.3, 25.4, 38.5, 51.6, 64.7 cm). In this experiment were measured the fission products: Zr-97, I-131, I-133 and Ce-143 by yield of gamma-lines 743.36 keV, 364.49 keV, 529.9 keV, and 293.3 keV respectively. The number of the fission reactions was determined by measured value of fission product and following neutron fission yield: Zr-97- 5.7%, I-131- 3.6%, I-133- 6.3%, Ce143-4.3%[8]. It means that, experimental data of the number of the fission reactions include neutron induced fission reaction only. The number of Pu-239 production was determined by measuring number of decay reactions Np-239(β-). It was assumed that the axis of the assembly and the beam are parallel. This experiment employed a of 0.66 GeV proton beam.

3 Calculation results and experimental data

This section gives a graphical presentation of computation results vs. experimental data. On every plot black, bold lines and full symbols depict experimental data. On Fig.1 (left) shows the spallation neutron yield vs. proton energy for natural uranium target compared to calculations. The Bertini/Dresnen model is in the best accordance to experimental data (Fig.1 left). The number of neutrons produced by spallation reaction depends on the initial particle energy and this magnitude rises with the energy.[3, 4] The results of Bertini/Dresnen model shows that the number of neutrons occurring due to spallation reaction induced by proton with energy E=0.66 GeV is approximately 20 neutrons/proton but when the energy of a particle is rised to E=1.5 GeV the amount of neutrons also rises to approximately 60. For energy levels smaller or equal to 600 MeV models give results less than experimental data about 17%. The radial distribution of plutonium production reaction U-238(n,g) is a decreasing function of distance R from the assembly axis (Fig.1, right), compare [1, 2]. Calculation for Z=123 turned out to be more accurate comparing to experimental data. The axial distributions of U-238(n,g) and U-238(n,f) reactions have local maximum for Z=254 for different distance from assembly axis R=40, 80, 120 (Fig.2). The number of reactions decreases when the radial location increases. This effect was expected because of the geometry in the middle of section there is the smallest escape probability of the particles inducing the both reactions Please note, that agreement of calculation results and experimental data for fission reactions are significantly better than for capture reaction (Fig.2). However the radial distribution of fission reactions show a big disagreement for detectors placed on the assembly axis i.e. for R=0 (see Fig.3 left). This figure presents comparison number of fission reactions induced by neutron only for both experimental data and calculation results. If we take into account proton and neutron induced fission reaction in calculation results we obtain better agreement (Fig.3 right). Important magnitude useful for this research is a spectral index defined by the following formula:

$$SI = \frac{N[U-238(n, g)]}{N[U-238(n, f)]}$$ (1)

Where:
SI- Spectral Index
N[U-238(n,g)] - number of U-238(n,g) capture reactions
Figure 1. Spallation neutron yield for uranium target (left; experimental data from [4, 6]) and radial distribution of U-238(n,g) reaction (right).

Figure 2. Axial distribution of plutonium production (left) uranium fission (right).
Figure 3. Radial distributions of U-238 fission reactions before (left) and after (right) considering fission reactions induced by protons.

Figure 4. Radial distribution of Spectral Index (left) and cross section of U-238(n,f) and U-238(n,g) reactions - the evaluated data come from JEDL library. (right).
$N[\text{U-238(n,f)}]$ – number of U-238(n,f) fission reactions

Spectral index is often used because it does not depend on normalizing method of neutron flux. In other words, it is not sensitive on normalizing errors. The spectral index value is an increasing function of radius (Fig.4 left). The reason is simple-the average neutron energy is decreasing function of radius because neutron loses its energy during scattering reactions when it moves away from the neutron beam radially. Additionally radially decreasing neutron energy cause decreasing U-238(n,f) fission cross section and increasing U-238(n,g) capture cross section (see Fig.4 right). That is why the denominator decreases and the nominator increases of Eq.(1) simultaneously and as the whole fraction increases.(compare [1, 2]).

4 Conclusions

The calculation results for U-238 fissions reactions are in good accordance to experimental data besides detectors placed on the assembly axis (compare Fig.3 left and right). The reason for this discrepancy was that there was not taken into account an experimental number of fission reaction induced by high energy particles of beam and not enough precisely determined the beam parameters. Our recent studies confirm that taking into account proton and neutron induced fission reaction in experimental data and correcting the angle between the assembly and the beam axis, one can obtain good accordance for all detectors. For energy levels smaller or equal to 660 MeV the models give results less than experimental data about 17% (Fig.1). Maximal difference between the presented models is equal to about 20% (Fig.1). The difference between experimental data and calculation results for U-238(n,g) reactions (Fig.2) is greater than experimental and calculation errors. This result suggests that the low energy fraction of the neutron flux in the experiment was greater than in calculation simulation. The reasons this situation can be in that simulation model not take into account all materials used in experiment for instance: pieces of paper or scotch tape or plastic box around detectors. As we know these materials have a very low mass but they are very good moderators and are placed directly on the detectors.

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