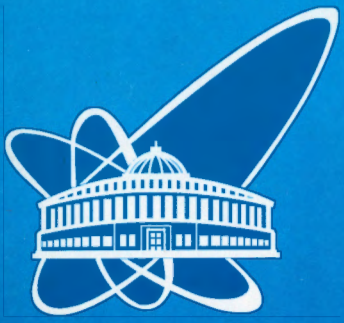

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FIRST RADIOCHEMICAL STUDIES ON THE TRANSMUTATION OF ^{239}Pu WITH SPALLATION NEUTRONS

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1. Introduction

An Accelerator Driven System (ADS) as a subcritical device using lead as the spallation target has the potential to transmute long-lived fission products by neutron capture and destroy all actinide elements by fission. The advantages are well-known:

1) Such systems are not critical, i.e. they keep the chain reaction at all times in a subcritical condition ($k_{eff}<1$).
2) Such systems can strongly reduce the burden of radioactive waste for future generations. Conventional nuclear power stations have produced - so far - about 1400 tons of plutonium. This plutonium is practically not used at present. When this plutonium together with the other minor actinides is placed into any geological final depository, it may constitute a serious danger to the biosphere for millions of years[1].
3) When the long-lived radwaste is destroyed by transmutation, final depositories need to be separated from the biosphere for only about 600 years[1].

For designing a subcritical system to transmute long-lived nuclear waste from nuclear power reactors, it is important to optimize the efficiency of the neutron capture and fission process. Most of the published ADS designs for transmutation of long-lived nuclear waste in subcritical systems are based on theoretical simulations. Only a few groups published experimental results, among them is a group in CERN[2-4] and our collaboration[5-9]. We have studied the neutron production in a massive uranium lead target (20 cm long, 8 cm in diameter) bombarded with proton beams of energies from 0.5 GeV to 7.4 GeV and the related transmutation of long-lived nuclear waste nuclei, such as ^{129}I and ^{237}Np. These last two long-lived waste nuclides can be transmuted as follows:

$$^{129}I\ (T_{1/2}=1.57\ E{+}07\ a) + n \Rightarrow {}^{130}I\ (T_{1/2}=12.4\ h) \Rightarrow {}^{130}Xe\ (\text{stable})$$

$$^{237}Np\ (T_{1/2}=2.14\ E{+}06\ a) + n \Rightarrow {}^{238}Np\ (T_{1/2}=2.12\ d) \Rightarrow {}^{238}Pu\ (T_{1/2}=88a)$$

It is known that a subcritical system is attractive not only due to its transmutation capacity, but also because of its potential to produce energy out of waste transuranium nuclei, in particular plutonium. However, until now there are no published experimental results concerning the efficiency of neutron induced fission for ^{239}Pu in a subcritical system except one recent study on fission of μg amounts of ^{239}Pu, using solid state nuclear track detectors[2,3]. The present work reports on the transmutation of gram amounts of ^{239}Pu with spallation neutrons generated by relativistic protons in a subcritical system. Radiochemical techniques for product quantification are used.

2. Experimental

We used 0.53 GeV and 1.0 GeV proton beams to irradiate massive lead targets, consisting of 20 lead discs, each 1 cm thick and 8 cm in diameter, and surrounded by 6 cm paraffin moderators as shown in Figure 1. The Synchrophasotron of the Laboratory for High Energy at the Joint Institute for Nuclear Research, Dubna, Russia delivered the relativistic protons. The ^{239}Pu samples were placed on top of the moderator (see Fig.1) 5 cm downstream from the entrance point of proton into the lead target and irradiated with spallation neutrons generated in the target. Two Pu samples were used, each contained 449 mg metallic ^{239}Pu. The samples were well sealed in Al-capsules (see Fig.2) and obtained from the Institute of Physics and Power Engineering in Obninsk, Russia.

The experimental parameters of two irradiations are given in Table 1.

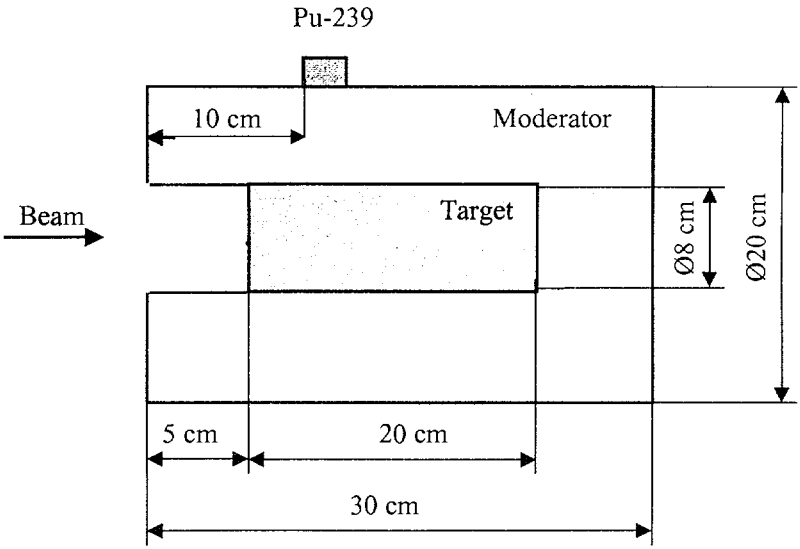

Fig. 1 : Experimental set-up for transmutation experiments of ^{239}Pu.
Lead target: ∅ = 8 cm, L=20 cm, surrounded by 6 cm paraffin.

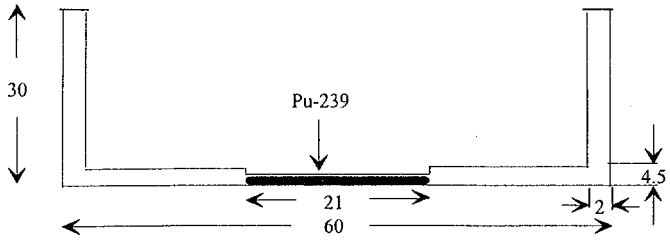

Fig. 2 : Sample of 449 mg metallic ^{239}Pu sealed in a Al-capsule,
as produced by the Institute of Physics and Power Engineering, Obninsk, Russia.
Dimensions are shown in mm.

Table 1 : Experimental parameters for the transmutation experiments. The statistical uncertainties in the fluences are 7%, the total uncertainties in the fluences are 15%. (Details see text).

Experiment	irradiation time / min	fluence
0.53 GeV p + Pb	227	6.39E+12
1.00 GeV p + Pb	220	1.33E+13

In order to determine the center of the proton beam with respect to the target, Polaroid films were used. They were placed in front of the targets and irradiated with one beam pulse (about 10^9 protons). The photographs showed that the beam spot was 12 cm in the vertical and 4 cm in the horizontal direction for 0.53 GeV and 5 cm by 2 cm for 1 GeV protons. The exact beam profile was determined with the Solid State Nuclear Track Detector technique. [10]

The radiochemical determination of the proton fluence was carried out with 30 µm thin Al monitor foils using the nuclear reactions of $^{27}Al(p, 3pn)^{24}Na$, $^{27}Al(p,3p3n)^{22}Na$ and $^{27}Al(p,X)^{7}Be$ employing our new monitor system[11]: Two monitor Al foil stacks with 8 cm diameter were used during the irradiation, one stack (Al-A) was placed 40 cm upstream from the proton entrance point into the lead target, the other one (Al-B) was placed upstream in contact with the thick lead target. The cross sections measured with Al-B were strongly influenced by secondary particles generated in the thick lead target. The major reaction, disturbing the reaction of $^{27}Al(p, 3pn)^{24}Na$, is the well-known $^{27}Al(n, \alpha)^{24}Na$ reaction. However, we have not seen any such influence in the reactions $^{27}Al(p,3p3n)^{22}Na$ and $^{27}Al(p,X)^{7}Be$ in both Al foils, indicating no major influence from secondary neutrons. Therefore, the fluences given in Table 1 are the averages from the experimental results for ^{24}Na in the Al foil Al-A and of ^{22}Na and ^{7}Be in both Al foils (A, B). The size of the Al foils was 8 cm in diameter, just as the diameter of the Pb target. We did not consider the fluences given by the machine operators. This will be discussed in a forthcoming and more detailed paper [17].

After the irradiation, the plutonium samples together with the Al monitor foils were taken to the Laboratory for Nuclear Problems, JINR, Dubna for gamma measurements using High Purity Germanium detectors. Only the central Al foil out of the three Al foil stack was investigated. Gamma detector efficiency calibration was carried out using an ^{152}Eu-standard sample. Each plutonium sample was measured 9-10 times for counting times ranging from 3000 s to 15000 s.

3. Experimental Results

The ^{239}Pu samples were exposed during the irradiations to spallation neutrons generated in the lead target. Out of the copious plutonium fission products in the present work we have determined quantitative data for 11 nuclides: ^{91}Sr, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{129}Sb, ^{132}Te, ^{133}I, ^{135}I and ^{143}Ce. These nuclides were selected according to the following criteria:

1. If all major photo peaks of a nuclide appear in the gamma spectra, (see Fig. 3), it is chosen as a candidate.
2. The analyzed photo peaks shall only belong to the interesting nuclide. There should be no other interfering nuclides having the same gamma energy.
3 The decay curve of this photo peak must follow the half-life of this interesting nuclide. Fig. 4 shows the decay curve for the fission product ^{133}I as an example. Only those photo

peaks, where experimental half-life agrees within 10% to the reference value, are chosen for analysis.

Analysis of γ-spectra was carried out with the GAMMAW program[12]. After obtaining the activity at the end-of-bombardment A_{eob} for each nuclide in each sample, we calculated an "experimental production/destruction rate" B, which is defined in a self-evident manner as follows[5-9]:

B = (number of ^{239}Pu atoms fissioning) / (1 g ^{239}Pu sample) / (1 primary proton)

This parameter B is strictly an empirical parameter specified in each experiment for

- a) a given nuclear reaction induced by the incident particle or by secondary neutrons;
- b) a well-defined experimental set-up, for example as shown in Fig. 1; and
- c) a given particle and energy, such as 1.0 GeV protons.

The experimentally determined B values for ^{239}Pu-fission measured via different fission products are listed in Table 2. For the calculation of the total Pu fission rate from the experimentally determined fission fragment B values, it is necessary to know the yield of a certain fission products with respect to the total fission cross section. However, there are no suitable fission yield data available, therefore the fission yields used here are averages of the thermal and the epithermal yields from JEF-PC[13]. According to those fission yields, the B values for individual fission products are converted to plutonium fission B values. The experiments show that on the outer surface of our set-up (Fig.1) there are 0.0021 fissions of ^{239}Pu in a one gram plutonium sample when one proton of 0.53 GeV energy hits the lead target, and 0.0031 fissions of ^{239}Pu when one uses 1.0 GeV protons. The close agreement of the average fission B values of ^{239}Pu calculated from the B values as determined for individual fission products suggests, that essentially only thermal and epithermal neutrons contribute to the ^{239}Pu fission in our setup. Such high fission rates (B values) for plutonium can been used for a very fast burn up during ADS operation, as will be shown later.

Figs. 5 shows the B values for individual fission products uncorrected for cumulative yields as a function of product mass. Since we did not measure the production of stable, very short-lived and very long-lived nuclides, we could not calculate the corrected mass yields. Therefore, these plots just give an impression of the fission yield distribution for ^{239}Pu under the prevailing experimental conditions.

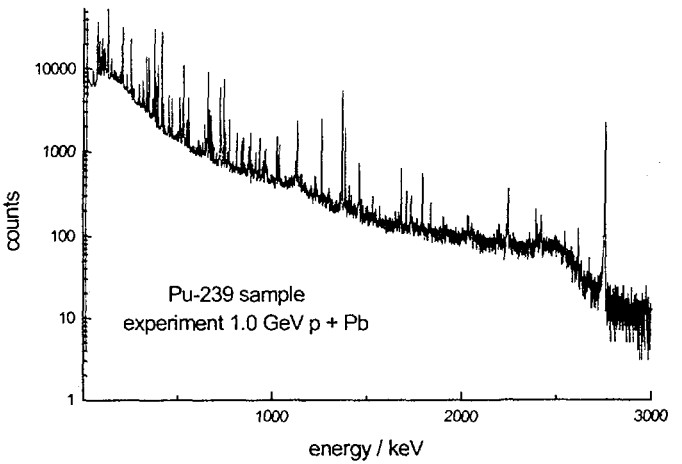

Fig. 3.: γ-spectrum of the ^{239}Pu sample irradiated on the outer surface of moderator in the experiment 1.0 GeV p + Pb. The measuring time was 6000 s.

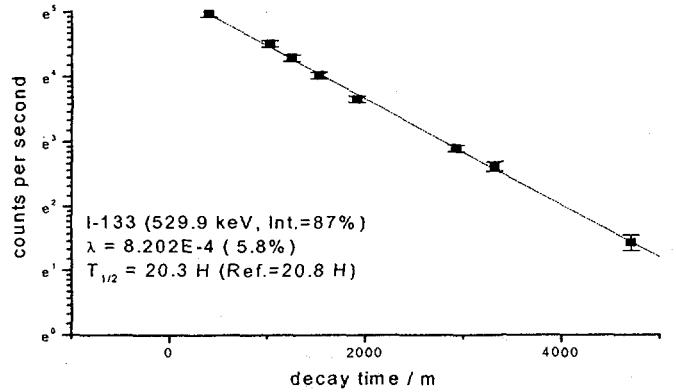

Fig. 4 : Decay curve of ^{133}I at 529.9 keV observed in the ^{239}Pu sample after the irradiation: (0.53 GeV p + Pb). The fitted half life is (20.3 ± 1.2) hours.

Table. 2 : B-values for ^{239}Pu-fission, as determined via different fission products, details are given in the text.

nuclides	Yield[13]	B(exp.: 0.53 GeV p + Pb)	B(exp.: 1.0 GeV p + Pb)
^{91}Sr	0.02504	2.08 ± 0,18·E-3	3.07 ± 0.26·E-3
^{92}Sr	0.02998	1.99 ± 0.17·E-3	2.83 ± 0.23·E-3
^{97}Zr	0.05033	2.13 ± 0.18·E-3	3.10 ± 0.28·E-3
^{99}Mo	0.06009	1.85 ± 0.16·E-3	2.83 ± 0.34·E-3
^{103}Ru	0.06786	2.03 ± 0.17·E-3	2.89 ± 0.23·E-3
^{105}Ru	0.05526	2.28 ± 0.23·E-3	3.22 ± 0.27·E-3
^{129}Sb	0.01175	2.34 ± 0.21·E-3	3.51 ± 0.29·E-3
^{132}Te	0.05125	1.79 ± 0.21·E-3	2.83 ± 0.25·E-3
^{133}I	0.06953	2.10 ± 0.18·E-3	3.31 ± 0.43·E-3
^{135}I	0.06378	2.16 ± 0.20·E-3	3.01 ± 0.33·E-3
^{143}Ce	0.04372	2.10 ± 0.31·E-3	3.15 ± 0.39·E-3
average		**2.08 ± 0.16·E-3**	**3.07 ± 0.22·E-3**

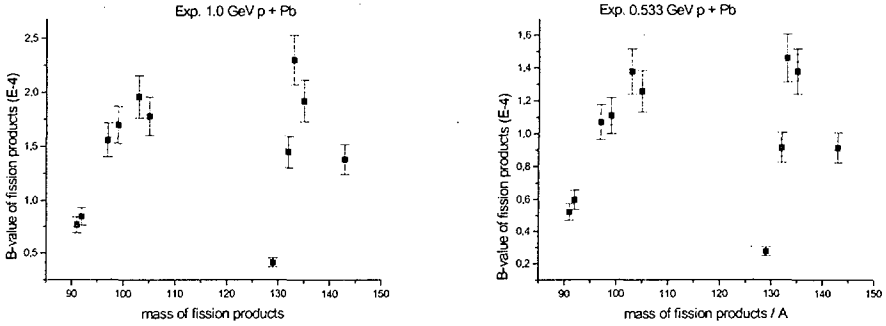

Fig.5. Experimental B value distribution for plutonium fission products, uncorrected for cumulative yields observed in Pu targets during experiments with (1.0 GeV p + Pb) and (0.53 GeV p + Pb). These mass-yields are typical for low-energy neutron induced fission reactions in ^{239}Pu.

4. Modelling and discussion

Two model calculations were performed by means of Monte Carlo simulations based on intranuclear cascades. The Cascade Evaporation Model, CEM[9, 14, 15], based on the Dubna Cascade Model, DCM, and developed in Dubna, Russia, was employed as well as the related program LAHET, as originally developed in Los Alamos, USA, however, used in detail here by one of us[16]. Neutron generation in the lead target is accomplished via multi-stage processes induced by primary particles interacting with the lead target. Details of the calculations have been described in preceding publications [5,7-9,18].

The results of the calculations and their comparison with the experimental results are shown in Table 3. The DCM/CEM model gives slightly larger results whereas the LAHET code calculates slightly lower than experimental results. However, the energy dependence is in all cases remarkably similar. This finding will be discussed in greater detail in a

forthcoming publication[17]. The experimental ratio for B-fission of Pu at 1.0 GeV proton energy compared to B-fission at 0.53 GeV is (1.45 ± 0.32). This value is well within the calculated ratios using both DCM/CEM or LAHET. Thus, for our setup fission rates scale almost linearly with the proton energy within the range of proton energies studied. This is at variance with the experimental and theoretical results obtained for another setup [2].

Table 3.: Comparison of experimental and model-calculated B-fission for Pu target. The experimental uncertainty is approximately 15%.

Proton energy	Experimental B-fission	DCM/CEM calc. B-fission	LAHET calc. B-fission
1.0 GeV	(3.2±0.5) E-3	4.3 E-3	2.7 E-3
0.53 GeV	(2.2±0.3) E-3	2.5 E-3	1.65 E-3
Ratio for 1.0 GeV / 0.53GeV	1.45±0.32	1.72	1.69

To our knowledge the present paper describes the first radiochemical experiment on plutonium fission caused by secondary neutrons in a small subcritical ADS setup. There are no other data yet to compare with. For ^{239}Pu fission, there is only one recent work [3] where ^{239}Pu fission was measured in the TARC-experiment with Solid State Nuclear Track Detectors (SSNTD) using μg Pu targets. The TARC experiment [3] cannot give any information about individual fission products. In the TARC experiment the fission B value for ^{239}Pu was found to be 0.005 [same units as ours] for 2.75 GeV protons in a similar geometric position as ours.

It is also interesting to study theoretically the neutronics of our experimental setup using the DCM/CEM model. The total neutron distribution on the outer surface of the moderator is shown in Fig. 6. The maximum neutron fluence appears on the paraffin moderator at a position about 5 - 10 cm downstream from the front of the lead target for both proton energies under study. Rather similar distributions were calculated in earlier publications[5-11].

The calculated neutron spectra at a position 5 cm downstream the proton entrance into the lead target are given in Fig. 7 for the two experiments. No back scattered neutrons are considered. The influence of the thin Al capsule sealing off the radioactive plutonium sample was neglected.

As a summary of our experimental transmutation results, Table 4 shows the B values for the transmutation of ^{239}Pu where for comparison we have also included other data for ^{129}I, ^{237}Np and the chemical sensors La and U for 1 GeV protons onto lead targets. Details on the determination of the other product nuclides will be given in publication to follow [17], where further results for ^{129}I, ^{237}Np and the chemical sensors La and U, exposed to thermalized neutrons are given. As a matter of record, one should recall that the (n, γ) transmutation rate in uranium is the production rate of ^{239}Pu. For comparison, the TARC experimental results for transmutation of ^{239}Pu (SSNTD, μg Pu)[3] are also listed in table 4. One can see that the transmutation rate of ^{239}Pu via fission is much higher than for the (n,γ) transmutation of ^{129}I and ^{237}Np. This comparison is only indicative of the effectiveness of the fission channel as it neglects the different energy spectra of neutrons at various positions of our target.

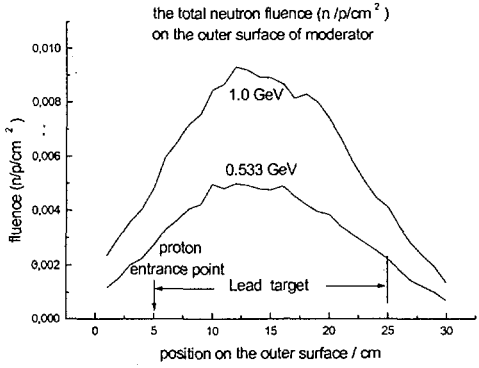

Fig.6: Neutron fluence (n / p / cm^2) on the outer surface of moderator, simulated with DCM/CEM.

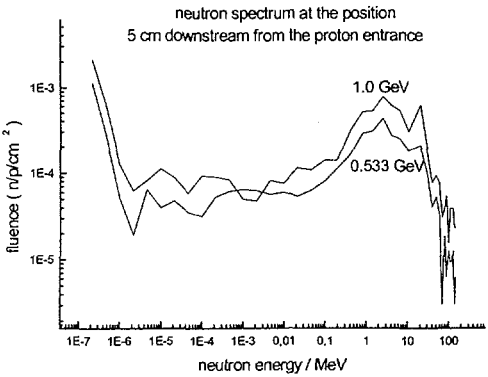

Fig.7: Neutron spectrum on the outer surface of moderator at the position 5 cm downstream the proton entrance into the target, simulated with DCM/CEM.

Table 5.: B values for the 1 GeV protons onto the Pb-target, as shown in Fig.1. (15% statistical uncertainties).

Nuclear reaction	B - value	Reference
$^{239}Pu \rightarrow$ fission	3.2E-03	This work
$^{238}U \rightarrow ^{239}Np$	5.5E-05	[17]
$^{237}Np \rightarrow ^{238}Np$	3.3E-04	[17]
$^{139}La \rightarrow ^{140}La$	1.1E-04	[17]
$^{129}I \rightarrow ^{130}I$	8.1E-05	[17]
$^{239}Pu \rightarrow$ fission (with SSNTD, μg Pu)	5.E-3 (for 2.75 GeV protons)	[3]

6. Conclusions

Plutonium destruction rates via neutron induced fission (B-values) were experimentally determined in a small spallation system for proton beam energies of 0.53 and 1.0 GeV. According to our knowledge, this is the first radiochemical determination for an ADS setup driven by relativistic protons. The database is useful for upcoming applications of accelerator driven nuclear waste transmutation and energy amplification (subcritical reactor). The results imply that a subcritical system can very effectively destroy plutonium. The plutonium fission rate is particularly high. For example: using a 10 mA beam of 1.0 GeV protons under the same experimental conditions as ours, the transmutation rate of ^{239}Pu via (n,f) is estimated as 0.6% per day, (or 3 mg out of 449 mg samples). The corresponding number for 0.53 GeV it is 0.4% per day (or 2 mg out of 449 mg samples). The technical and material problems of a spallation target exposed to 10 mA beam of 1 GeV protons are considerable and they have not been touched in this work.

7. Acknowledgements

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Appendix

Determination of thermal and epithermal neutrons on top of the paraffin moderator using independently the SSNTD technique [19].

An experiment was carried out with CR39 SSNTDs, covered with ^{6}LiF as described by Zamani et al. [19] This allowed a direct determination of the fluence of thermal neutrons and energetic neutrons at the proton energies employed. The energetic neutrons were determined with Cd-shielding which allowed the quantification of neutron numbers with energy of 300 KeV < E(n) < 3 MeV through the measurement of recoil protons tracks. Five targets were evenly placed on top of the moderator. The targets were exposed to a few beam pulses from the Synchrophasotron with a total fluence of approximately 10^{11} protons. In this case the proton beam flux was taken as given by machine operators, it could not be checked independently with radiochemical methods. The results for each experiment are given in Tables A.

Table A1.: Neutron fluences per primary proton on the outer surface of the moderator in the experiment 0.53 GeV protons on Pb as measured with CR39 SSNTD. The targets are evenly distributed on top of the moderator. The results at distances of 8.5 cm and 11.5 cm from the front plate of the moderator are printed in bold characters, as they are close to the ^{239}Pu target (Fig. 2).

Distance	2.5 cm	5.5 cm	**8.5 cm**	**11.5 cm**	14.5 cm
Thermal	2.6 ± 0.8	2.9 ± 1.0	**3.7 ± 1.2**	**4.2 ± 1.3**	4.5 ± 1.4
Energetic	0.6 ± 0.2	1.6 ± 0.5	**1.4 ± 0.3**	**1.8 ± 0.4**	1.9 ± 0.4

Table A2.: Neutron fluences per primary proton on the outer surface of the moderator in the experiment 1.0 GeV protons on Pb, as measured with CR39 SSNTD. The targets are evenly distributed on top of the moderator. The results at distances of 8.5 cm and 11.5 cm from the front plate of the moderator are printed in bold characters, as they are close to the ^{239}Pu target (Fig. 2).

Distance	2.5 cm	5.5 cm	**8.5 cm**	**11.5 cm**	14.5 cm
Thermal	4.3 ± 9	4.7 ± 10	**5.8 ± 12**	**6.7 ± 13**	6.5 ± 13
Energetic	2.6 ± 0.5	4.1 ± 0.8	**4.1 ± 0.8**	**5.1 ± 1.0**	5.1 ± 1.0

Taking the observed radiochemical value for B [^{239}Pu (n,f)] = 0.0022 fissions/(1g ^{239}Pu)/(1 proton of 0.53 GeV), one can estimate the fluence of thermal neutrons through the ^{239}Pu target induced by 1 primary proton of 0.53 GeV energy as 0.0012 thermal neutrons per cm^2 or **2.4 thermal neutrons** on the entire surface (approximately 1900 cm^2) of the paraffin moderator. We assume the fission cross-section of 752 barns for ^{239}Pu for thermal neutrons and we have neglected neutrons of higher energies [20] . This neutron number is compatible with the independent thermal neutron fluence measured using SSNTD in Table A1.

The corresponding radiochemically determined fluence of thermal neutrons on the surface of the moderator for 1 GeV proton induced interactions is **3.5 thermal neutrons**, again compatible with the SSNTD results in Table A2. The fluence of both thermal and energetic neutrons scales - as expected - with the total proton energy for both proton energies under investigation. Similar SSNTD results have been observed by us several times earlier[5,7,18]. We consider the overall agreement between radiochemical and SSNTD results as being satisfactory in our experiments.

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