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TRANSMUTATION OF ^{239}Pu AND OTHER NUCLIDES USING SPALLATION NEUTRONS PRODUCED BY RELATIVISTIC PROTONS REACTING WITH MASSIVE U- AND Pb-TARGETS

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1. Introduction

During recent years some aspects of the nuclear energy fuel cycle have attracted considerable attention: In connection with the introduction of the concept of „energy amplifiers“ by C.Rubbia and his coworkers at CERN, Geneva, Switzerland, in 1993 [1], the coupling of modern proton accelerators with energies of about 1 GeV and high intensities exceeding 1 mA to subcritical nuclear power plants became a feasible technological option. Previously, Tolstov had suggested related technologies at the Joint Institute for Nuclear Reasearch (JINR) in Dubna, Russia [2]. In this paper we are not concerned with the aspect of obtaining energy with such systems, however, we will focus our attention on the aspect of **transmutation**. This term is describing in this context the transmutation of long-lived radioactive fission products into short-lived or finally s+ nuclei, such as the transmutation of ^{129}I ($t_{1/2}$ = 1.6 E+6 years):

$$^{129}\mathrm{I}\ (n,\gamma)\ ^{130}\mathrm{I}\ (\ t_{1/2} = 12\ \mathrm{h}\ ) \Rightarrow\ ^{130}\mathrm{Xe}\ (\ \text{stable}\ ). \qquad (1)$$

The same term transmutation is also used to describe the conversion of long-lived actinides, such as ^{237}Np ($t_{1/2}$ = 2.1E+06 years) or ^{239}Pu ($t_{1/2}$ = 2.4 E+4 years) into short-lived products, such as

$$^{237}\mathrm{Np}\ (n,\gamma)\ ^{238}\mathrm{Np}\ (t_{1/2} = 2.1\ \mathrm{d}\ ) \Rightarrow\ ^{238}\mathrm{Pu}\ (\ t_{1/2} = 88\ \mathrm{a}\ ) \qquad (2),$$

$$^{239}\mathrm{Pu}\ (n,\text{fission}) \Rightarrow \text{relatively short-lived fra} \qquad (3).$$

In addition, the transmutation of ^{238}U into ^{239}Pu is also considered:

$$^{238}\mathrm{U}\ (n,\gamma)\ ^{239}\mathrm{U} \Rightarrow\ \ \Rightarrow\ ^{239}\mathrm{Pu} \qquad (4)$$

The modern aspects of transmutation have been introduced in a theoretical approach by Bowman and coworkers from the Los Alamos National Laboratory, Los Alamos, USA in 1992 [3]. They studied the transmutation capacity of thermal neutrons, produced by 1 GeV protons interacting with a lead target introduced into an extended subcritical nuclear power reactor. Our team has published a series of experimental papers on the transmutation of ^{129}I, ^{237}Np, ^{nat}U, and stable ^{139}La during recent years [4 - 8]. We used a small target system surrounded by 6 cm paraffin as partial moderator irradiated with low-intensity relativistic proton beams of the Synchrophasotron at the Laboratory of High Energies, Joint Institute for Nuclear Research (JINR), Dubna, in Russia. In this way we studied some principle experimental features. The proton energies used so far are 1.5 GeV, 3.7 GeV, and 7.4 GeV.

In this paper, an extension of this research is presented with the following new features:

- As a new transuranium target, two samples of well-sealed ^{239}Pu, each containing 0.449 g of plutonium, were employed.
- The spallation targets were irradiated with 0.53 GeV and 1.0 GeV protons, in addition to a 1.5 GeV run for control purposes. These studies appear to be interesting, as similar energies have been used during earlier experiments by the [9].
- A new radiochemical proton fluence measuring system is used. Two monitor foil-stacks, in contact and the other 35 cm upstream were employed in order to measure in a direct and reliable way the proton fluences, [10].

In this paper the experimental results, as well as a theoretical estimation on the conversion of transmutation rates observed radiochemically into spallation neutron yields, will be presented. Detailed results of auxiliary SSNTD (solid state nuclear track detectors) experiments are described in the Appendix.

2. Experiment

2.1 The proton fluence measurements with a new radiochemical proton fluence monitor system

The preceding publication [6] describes in detail, that the proton fluence measurements at the Synchrophasotron during the previous experiments with 3.7 and 7.4 GeV protons were a rather delicate task. During the experiments in 1998 and 1999 - which are reported in this contribution - we measured proton fluences in two independent manners. The first method was based on conventional electronics methods while in the other a new system of two radiochemical monitors was used as described below.

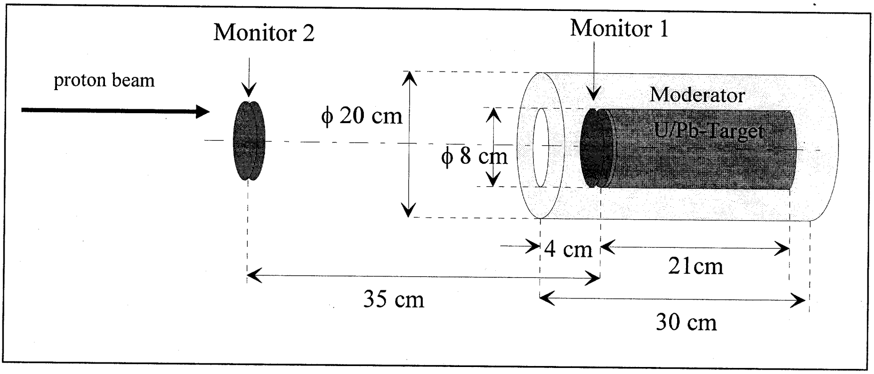

Fig. 1.: The experimental layout of the new radiochemical method to determine the proton fluence with two monitor foil stacks [10]. (Details see in text)

This new system has two stacks, each consisting of one 1 mm thick Cu foil and a stack of three 10μm thick Al foils as shown in Fig. 1. One stack is in contact with the extended target, the other is placed 35 cm upstream. After the proton irradiation the Cu-foil and the middle Al foil were assayed for ^{24}Na activity, the Al foils were investigated in some experiments (1999) in addition for their ^{7}Be and ^{22}Na activities. This technique already has been published [10]. The results of the proton fluence measurements are given in Table 1. In all further evaluations we will consider only proton fluences that are measured by the radiochemical method.

Table 1 shows that the results for ^{24}Na determined in Al-foils of monitor 2 in a distance of 35 cm upstream the massive target give consistent results for the proton fluence with the other 3 monitor reactions: ^{7}Be and ^{22}Na in Al and ^{24}Na in Cu. These latter three reactions are insensitive to low-energy secondaries, as has been shown in Ref. [10]. As it is fairly straightforward to measure accurately ^{24}Na in thin Al-foils, we essentially used the proton fluence determined with ^{24}Na in Al (monitor 2).

Table 1: Details for the proton irradiations[#].

a.) Fluences (E+13) of experiments 1998 in Dubna, measured via ^{24}Na in Cu- and Al-foils.

	Fluence (Cu-Monitor)		Fluence (Al-Monitor)		Final fluence
Experiment	Monitor 1	Monitor 2	Monitor 1	Monitor 2	chemistry*
0.5 GeV p + U				1.17 ± 0.05	1.17 ± 0.05
1.0 GeV p + U	1.22 ± 0.14	1.09 ± 0.13	3.15 ± 0.24	1.23 ± 0.09	1.23 ± 0.09
1.5 GeV p + U	1.41 ± 0.17	1.37 ± 0.17	6.67 ± 0.49	1.41 ± 0.11	1.41 ± 0.11
1.0 GeV p + Pb⁺	1.22 ± 0.14	1.25 ± 0.15	2.19 ± 0.17	1.30 ± 0.11	1.30 ± 0.11
1.5 GeV p + Pb⁺		1.31 ± 0.16	2.57 ± 0.18	1.29 ± 0.09	1.29 ± 0.09

b.) Fluences (E+12) of experiments 1999 in Dubna, measured via ^{24}Na, ^{22}Na and ^{7}Be in Al.

	Monitor 2			Monitor 1		Final fluence
Experiment	^{24}Na	^{7}Be	^{22}Na	^{7}Be	^{22}Na	chemistry*
0.53 GeV p + U	5.65±0.28	5.95±0.42	6.00±0.45	6.32±0.44	5.93±0.43	5.93 ± 0.53
0.53 GeV p + Pb				6.43±0.45	6.34±0.42	6.39 ± 0.43
1.0 GeV p + U	12.1±0.61	12.7±0.84	12.6±0.91	12.5±0.84	12.5±0.91	12.5 ± 0.65
1.0 GeV p + Pb	12.8±0.64	13.6±0.92	12.4±0.90	14.5±0.91	13.1±0.89	13.3 ± 1.08

[#]) Simultaneously, we irradiated ^{129}I, ^{237}Np, and ^{239}Pu samples, as indicated in Table 4.

*) For further evaluations we used a 15% uncertainty in the proton fluence determination.

+) In these two Pb-target irradiations, **no** moderator was installed („naked“ targets).

2.2 Transmutation studies using ^{nat}U- and stable ^{139}La-sensors: the calibration of the experimental set-up

The two principle experimental set-ups used by our group since many years [4 - 8] are shown

- in Fig. 2: a massive Pb-target, consisting of 20 Pb-discs, 8 cm ∅ and 1 cm thick, is surrounded by 6 cm paraffin moderators and called Pb-target.
- in Fig. 3: a massive uranium target consisting of two uranium rods, 3.6 cm ∅ and 10.5 cm long, is surrounded by 2 cm lead and 6 cm paraffin moderators and called U/Pb-target.

The surface of the moderator is spiked with several La-sensors (La1 - La10), each containing approximately 1 g La in the form of ($LaCl_3 \cdot 7H_2O$). Spallation neutrons induce the nuclear reaction:

$$^{139}\mathrm{La}(n,\gamma)^{140}\mathrm{La} \qquad (5).$$

The product nuclei ^{140}La has a convenient half-life of 40 hours, its activity is easily determined with gamma spectrometry.

Additionally, we have placed also several 1 g ^{nat}U (U1 - U5) samples on top of the moderator. This allows the study of the transmutation of natural uranium:

$$^{238}\mathrm{U}\ (n,\gamma)\ ^{239}\mathrm{U} \Rightarrow \mathbf{^{239}Np} \Rightarrow {}^{239}\mathrm{Pu} \qquad (6)$$

In this reaction only the activity of ^{239}Np is determined radiochemically, as this nuclide has a convenient half-life of 2.3 days to be measured by gamma spectrometry.

The reaction

$$^{nat}\mathrm{U}\ (n,\mathrm{fission}) \Rightarrow \text{fission fragments} \qquad (7),$$

was also investigated, however, these results are not presented here. These results can be found in [8], they are strictly analogous to those of the ^{239}Np-production rates [5 - 8].

The beam profiles at 0.53 GeV and 1 GeV proton energy have been determined with the SSNTD technique: a Lavsan track detector is placed upstream a 8 cm ∅ thin Pb foil in front of the massive metallic target. (Only high-energy particles induce fission in Pb. The fission fragments are registered in Lavsan). Afterwards, the foil is etched and scanned with an optical microscope. Details of this technique are described in [11]. Typical beam profiles are shown in Figs. 4 and 5. The beam profile at 1.0 GeV proton is characteristic for experiments of this kind and has been reported earlier [5 - 8]. The entire beam is essentially covered by the 8 cm ∅ metallic target and the beam is to a large extent attenuated within the 20 cm long target. The 0.5 GeV proton beam is not completely focused onto the metallic target, however, we consider this not as very serious, as low-Z material, such as paraffin, are quite inefficient to produce spallation neutrons. On the other hand, this low-energy proton beam is completely stopped within the 20 cm metallic target.

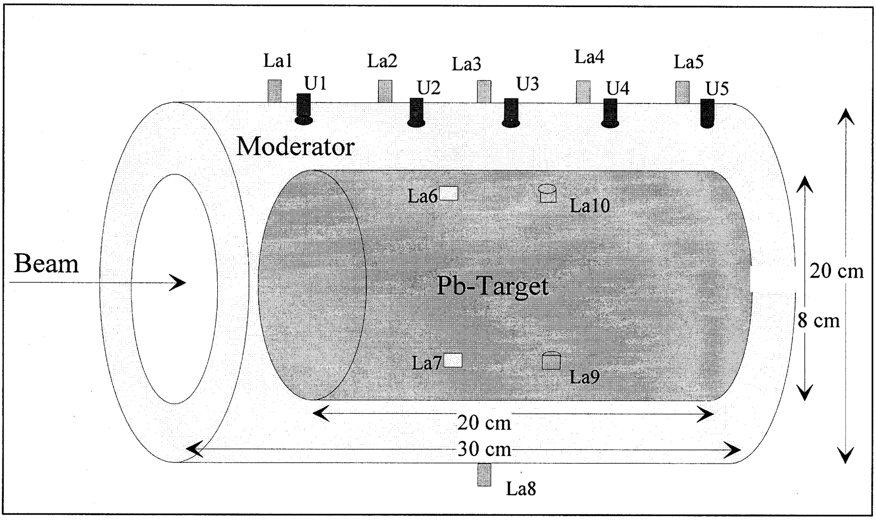

Fig. 2: Lanthanum (La-1 up to La-10) and uranium (U1 up to U5) sensors on top of the surface of the massive *Pb-target* surrounded by 6 cm paraffin moderator.

After the irradiations, the sensors and parts of the metallic lead targets were transported to a laboratory to investigate the gamma activity with HP Germanium counting systems using well-established procedures [12]. This allowed the determination of decay rates of produced radioactive nuclides. One could obtain the "equilibrium" decay rate for ^{140}La in the La-sensors. Then it is a convenient to calculate a "transmutation rate" B(^{140}La), defined as follows:

$$\mathbf{B(^{140}La) = (\text{ number of } ^{140}La \text{ formed }) / [(\,1\text{ g La }) (\,1 \text{ primary proton })]} \quad (8).$$

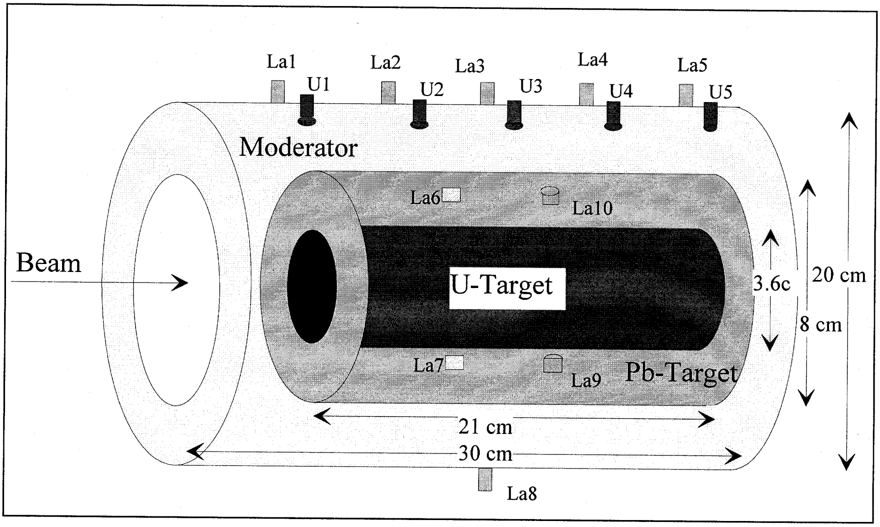

Fig. 3: Lanthanum (La1 - La10) and uranium (U1 - U5) sensors on top of the massive ^{nat}U target, surrounded by 2 cm Pb and 6 cm paraffin. This system is called *U/Pb-target.*

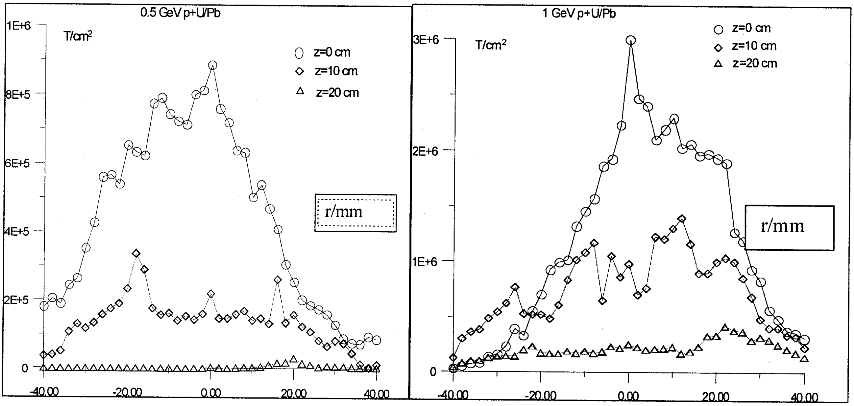

Fig. 4: The beam profile for 1 GeV p (1998). Fig. 5: The beam profile for 0.53 GeV p (1999). (The track density is given in T/cm^2. The z-values in Figs. 4 and 5 are the beam profiles measured horizontally at different depth of the target: Z = 0 cm is in front of the target, Z = 10 cm is in the middle, Z = 20 cm at the end of the target).

This B-value is strictly an empirical number, defined for a specific geometrical set-up, a specific transmuted isotope and a specified proton energy impinging onto the target. As the reaction (8) is essentially sensitive only to thermal neutrons, the measurement only to thermal neutrons, the measurement of several B(^{140}La)-values for various geometrical positions around the target system gives a measure for the distribution of thermal neutrons around this target during the irradiation. In particular, the determination of „azimuthal“ $B(^{140}La)$-values (sensors La- 3 plus La-6 to La-10 in Fig. 2) allow for the correction of azimuthal variations in the fluence of secondary neutrons. Details can be found in Ref. [5]. Typical results for $B(^{140}La)$-values of La-sensors on top of the moderator (La1 -La5) are shown in Fig. 6 for the U/Pb target corrected for azimuthal variations, details are given in Ref. (8). Similar typical results for $B(^{239}Np)$-values in uranium-sensors (U1 -U5) using the U/Pb target are shown in Fig. 7.

The experimental results for $B(^{140}La)$ and $B(^{239}Np)$ using the Pb-target system are omitted from this paper. Results have been presented quite often: the Pb-target gives B-values reduced by a factor 1/1.7 as compared to U/Pb-targets [5-8].

The distribution of secondary neutrons, measured via ^{140}La or ^{239}Np on top of the 20 cm long surface of the moderator shows a behavior known from previous experiments [5-8]: the maximum induced radioactivity is always concentrated about 5 cm behind the entrance of the protons into the massive uranium target, rather independent on the energy of the incoming proton. For a lead target this maximum is about 10 cm behind the entrance of the protons into the massive lead target. All induced radioactivity is decreasing drastically when one determines them at the back-end of the target, as shown previously [5-8]. Details for $B(^{140}La)$-values during the 1999 experiment are given in the tables of the Appendix together with the *average* $B_{av}(^{140}La)$*-values* for the La-sensors (La1 - La5) on top of the moderator inside the 1 cm deep holes on the surface of the moderator for a given target and a given proton energy E(p).

It is interesting to compare the different $B_{av}(^{140}La)$-values for all the experiments in this series, (Table 2). It is also of interest to show in the same table the *relative transmutation rate* $R(^{140}La)$, defined as follows:

$$R(^{140}La) = B_{av}(^{140}La) / E(p). \qquad (9)$$

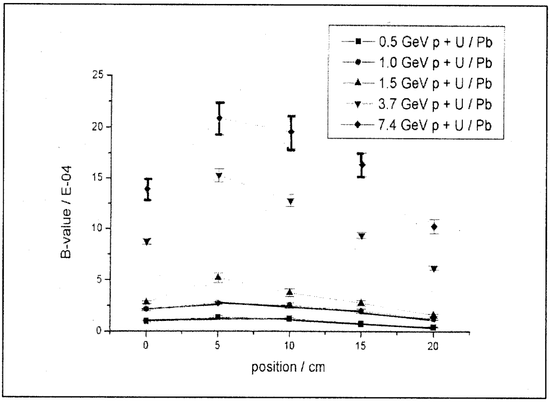

Fig. 6: $B(^{140}La)$ in La-sensors (La1 - La5) distributed on the surface of moderator for experiments with U/Pb targets. Results for 1.5, 3.7 and 7.4 GeV are from Ref. [6].

These values $R(^{140}La)$ are of interest, as they give a measure of the relative neutron production efficiency for a certain nuclear reaction with respect to the proton energy. It is a relative measure for the number of spallation neutrons. R(^{140}La) is rather energy independent within the proton energy range under study. The same phenomenon is presented graphically for the average B_{av}(^{239}Np)-values in U/Pb targets in Figs. 8 and 9. Again, the values R(^{239}Np)=B_{av}(^{239}Np)/E(p) are rather independent on the energy E(p).

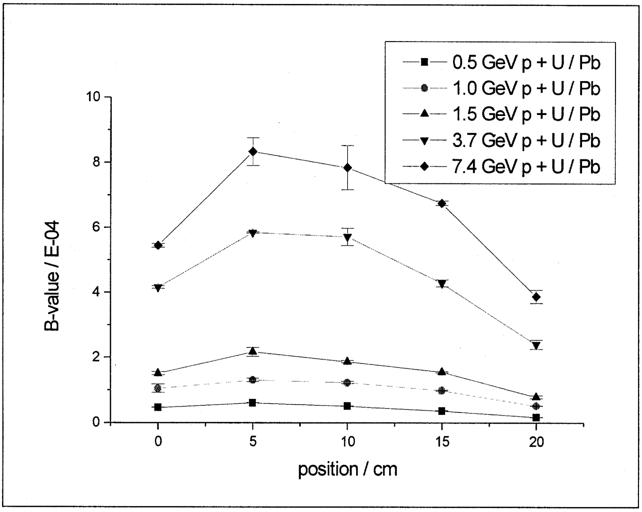

Fig. 7: B(^{239}Np) in U-sensors (U1 - U5) distributed on the top of the surface of moderator in experiments with U/Pb-targets. Results for 3.7 and 7.4 GeV are taken from Ref. [6].

Last but not least we want to report shortly on the result of using the Pb- target without the paraffin moderator (see Table 1: *"naked" Pb-targets*). The standard La and U sensors were placed on top of the lead target and the resulting activities are induced by the whole spectrum of spallation neutrons that leak out of the target. Their yields are compared to experiments with paraffin moderator:

$$B_{av}(^{140}La)_{moderator} / B_{av}(^{140}La)_{naked} = 128 \pm 27, \qquad (10)$$

$$B_{av}(^{239}Np)_{moderator} / B_{av}(^{239}Np)_{naked} = 12 \pm 3. \qquad (11)$$

These numbers can be easily understood: Due to the lack of moderator, the number of low energy/thermal neutrons is considerably smaller around a "naked" lead target as compared to the outer surface of the moderator in Fig. 2. As ^{140}La is practically only sensitive to thermal neutrons the above ratio is rather large. For ^{239}Np also epithermal neutrons are effective and one can expect here relatively more epithermal neutrons, as compared to thermal neutrons.
This results in a smaller experimental ratio. For typical fission fragments, such as ^{132}Te, one observes the same B(fission-fragment) value on top of the moderator and the "naked" exposure as neutrons of all energies induce fission in uranium [8]. The B_{av}(^{140}La)$_{naked}$ - and B_{av}(^{239}Np)$_{naked}$ -values are energy independent at the proton energies of 1.0 and 1.5 GeV (see Table 1). Further details are given in Ref. [8].

Table 2: $B_{av}(^{140}La)$ for the U/Pb target and the corresponding $R(^{140}La)=B_{av}(^{140}La)$ / E(p) in units of [1/g*1/GeV].

System studied	$B_{av}(^{140}La)$ (E-4)	$R(^{140}La)= B_{av}(^{140}La)$ (E-4)/ E(p)
0.53 GeV p + U/Pb	0.93 ± 0.14	1.86 ± 0.28
1.0 GeV p + U/Pb	2.11 ± 0.31	2.11 ± 0.31
1.5 GeV p + U/Pb	3.21 ± 0.48	2.14 ± 0.31
3.7 GeV p + U/Pb	10.5 ± 1.61	2.84 ± 0.43
7.4 GeV p + U/Pb	16.2 ± 2.48	2.19 ± 0.32

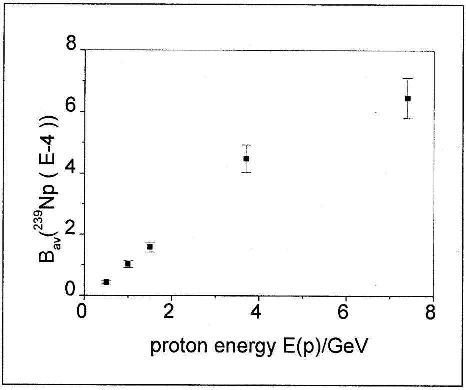

Fig. 8: Dependence of $B_{av}(^{239}Np)$ in U/Pb-targets on the energy of the incoming proton

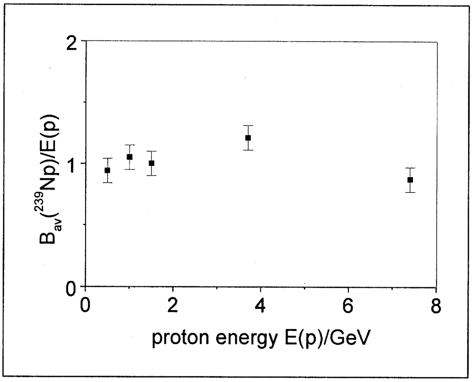

Fig. 9: Dependence of $R(^{239}Np)=B_{av}(^{239}Np)/E(p)$ [units of (E-4)] in U/Pb-targets on the energy of the incoming proton.

2.3. Conversion of $B_{av}(^{140}La)$-values into spallation neutron numbers and comparison with model calculations

The experimental determination of $B_{av}(^{140}La)$-values is certainly of practical interest, however, direct comparisons with model calculations are very difficult. One reason is that a B-value is only one single number, but it is caused by neutrons having a very broad energy spectrum with energy-dependent (n,γ) cross-sections. The exact calculation needs to carry out complex calculations. Some attempts in this direction have been made in earlier publications of the group without being completely convincing [5,6]. In this paper another approach will be presented, based on a recent calibration experiment of Hashemi-Nezhad, et al. [14]. Essentially, a Pu/Be source emitting $8.1 \cdot 10^6$ neutrons/s with an average neutron energy of about 4 MeV is placed into a Pb-shell surrounded with a 6 cm thick paraffin moderator (see Fig. 10). This setup is similar to the target shown in Fig. 3 without uranium insert. In this way, the spallation neutron production in the Dubna-experiments was simulated as realistic as we could do. On the outer surface of the paraffin moderator La-sensors were placed, identical to those used at the Synchrophasotron in Dubna. Additionally LR-115 2B SSNTD strips were irradiated as shown in Fig. 10. One set of strips was bare, being sensitive to a broad energy spectrum of neutrons, the other set of strips was covered with 0.75 mm Cd foils, in order to keep thermal neutrons off the SSNTD. After sufficient irradiation time, both the radiochemical and SSNTD sensors were treated. The results for $B(^{140}La)$-values along the longitudinal direction of the paraffin moderator are shown in Fig. 11(a) together with the model calculations using the MCNP-4B2 neutron transport code. The same experimental result is shown in Fig. 11(b), this time after having converted $B(^{140}La)$ into neutron fluences using the same standard computational procedures, already employed in Ref. [5] and described in detail in Ref. [14]. As one can see, the agreement between experiment and model calculation in this calibration experiment is good. Similarly good results have been obtained in the analysis of the SSNTD calibration [14]. In this way, the $B_{av}(^{140}La)$-value observed for 1 neutron emitted from the calibrated Pu/Be neutron source for La-sensors placed into tiny holes of the moderator during the Dubna experiments could be determined as

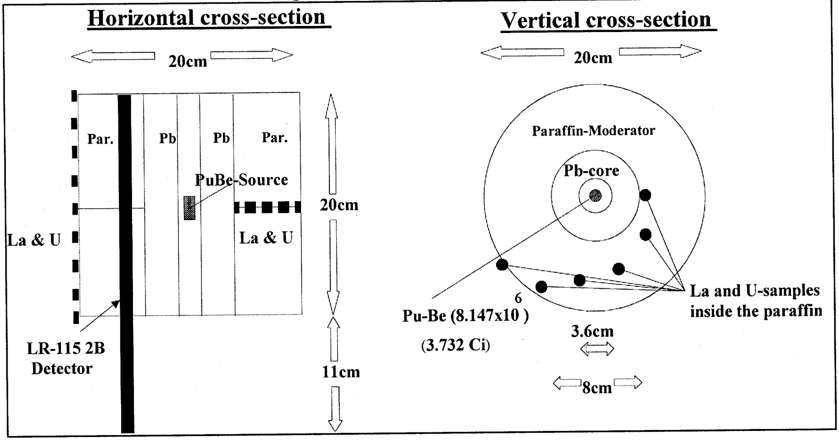

Fig. 10.) The experimental set-up for the absolute calibration of $B(^{140}La)$ using a well defined Pu/Be neutron source emitting $8.1\ 10^6$ neutrons/second. From Ref. [14].

follows:

$$\mathbf{B_{av}(^{140}La)_{for\ 1\ neutron\ from\ Pu/Be} = 9.7 \cdot 10^{-6}\ (\pm 20\%)} \qquad (12)$$

The value $B_{av}(^{140}La)$ for 1 neutron from Pu/Be is defined as the average of $B(^{140}La)$ on the 20 cm long surface of the moderator in Fig. 10. The uncertainty in this determination is based on experimental results reported in Ref. 5: The $B(^{140}La)$-value close to the outer surface of the moderator is very dependent on the *exact geometrical position of the sensor*: Acc. to Ref. 6, $B(^{140}La)$=20.4 E-6 *inside* the holes, as during the Dubna runs, and one measured $B(^{140}La)$=6.5 E-6 for the same sensor when positioned on the *surface* of the moderator. The results in equation (12) are uncertain by about 20%. This equation (12) can be used to estimate the number Y of spallation neutrons, produced in the MeV-energy range during the spallation reactions at the Synchrophasotron irradiations, from the $B_{av}(^{140}La)$-value. Detailed calculations by one of us [15a] have shown that the mean kinetic energy of neutrons emitted from the Pu/Be-source is 4.30 MeV. The lead target irradiated with 1 GeV protons gives average spallation neutron energy of 3.98 MeV. These two neutron energies are sufficiently close to allow a conversion of the $B_{av}(^{140}La)$-value into the number of spallation neutrons Y according to equation (12). The results of this conversion are shown in Table 3, together with a theoretical estimation of the number Y of spallation neutrons, as reported in Ref. [15b]. These calculations are based on two model codes mentioned earlier [5,8]: LAHET from the Los Alamos National Laboratory and DCM/CEM from the JINR, Dubna.

The experimental Y-values with their uncertainty of 20% agree reasonably well with model calculated Y-values. The discrepancy is mostly below 50% and never more than 100%. This appears to be state-of-the-art in this type of investigations. Simultaneously, Table 3 indicates a *linear increase* of the observed and calculated values for the number of spallation neutrons Y within the energy range studied. This is in agreement with independent neutron number measurements of Zucker, et al. [16] who observed a *linear* increase in neutron numbers using a similar small lead target irradiated with protons in the energy interval 0.4 GeV < E(p) < 1.4 GeV. Both experiments used similarily small targets, certainly smaller targets than those that were used by the CERN-group, as published by Andriamonje, et al. [9]. They observed a *very strong decrease of B, measured as Energy Amplification, in this energy interval* going from 1.0 GeV down to proton energy of 0.6 GeV. The reason for this apparent discrepancy has been explained by Hashemi-Nezhad, et al. [15b]: The target in Ref. [9] had dimensions of about 1 m, considerably larger than our targets. In such large targets, it is calculated [15b], that the number of neutrons increases *more than linearly* with the proton energy around the crucial proton energy of about 1 GeV. Of course, one should remember, that both groups used different experimental techniques to determine the equivalent to the "energy amplification": They measured at CERN the produced energy mostly in a direct manner with thermometers, in this work one measured only indirectly the number of spallation neutrons. In addition, it must be pointed out again, that the proton beam fluence measurements in this work had a rather large experimental uncertainty of 15%, thus reducing somewhat the statistical significance of the stated discrepancy.

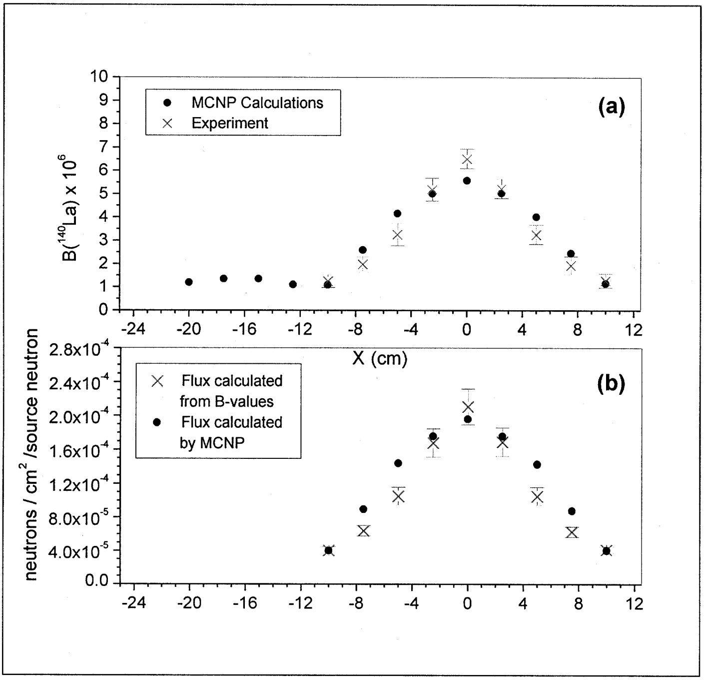

Fig. 11. Variation of the activation rate of ^{139}La, measured as B(^{140}La), and neutron fluence at the La sample locations, as a function of longitudinal distance on the moderator surface. The figures show both the experimental results and those obtained by MCNP-4B2 simulations. (a) activation rates and (b) slow neutron fluence. For details see Ref. [14].

Table 3.: Experimental $B_{av}(^{140}La)$ values for Pb-targets and their conversion into experimental spallation neutron number Y(experimental) and the comparison with theoretical model calculations. The experimental uncertainties are 20%. (It should be noted, that the determination of the proton beam intensity at 3.7 GeV and above was not yet measured with the reliable radiochemical method given in Fig. 1 and used at lower E(p)).

Reaction	$B_{av}(^{140}La)$ Radiochemistry	Y(experimental) Based on equ. 12	Y(theoretical) LAHET	Y(theoretical) DCM/CEM
0.53 GeV p + Pb	5.9 E-5	6.1	7.7	9.3
1.0 GeV p + Pb	11.4 E-5	11.7	15.3	17.4
1.5 GeV p + Pb	17.0 E-5	17.5	19.8	23.6
3.7 GeV p + Pb	60.4 E-5	62.3	38.7	43.9
7.4 GeV p + Pb	74.0 E-5	76.3	61.1	70.8
0.5 GeV p+ U/Pb	9.3 E-5	9.6	10.4	17.2
1.0 GeV p+ U/Pb	21.1 E-5	21.7	23.5	35.1
1.5 GeV p+ U/Pb	32 E-5	33	32.4	47.2
3.7 GeV p+ U/Pb	105 E-5	108	65.4	93.6
7.4 GeV p+ U/Pb	162 E-5	167	112.1	151

Finally, we could study theoretically the neutron fluence for a very large setup using natural uranium in water as a „subcritical reactor“. (Some considerations along this line have been published by Calero, et al. [17]). The CERN group published the results of the TARC experiment using a $3.3\times3.3\times3\ m^3$ lead target. They found that 35.6 spallation neutrons are produced by one 1 GeV proton in this extended target [18,19]. It is well-known, that by changing the target from lead to uranium of the same size the number of spallation neutrons increases by 70% [5-8]. This brings the number of spallation neutrons within such an extended uranium target up to 61. Conseqently, one can define an average "electric power" cost to produce 1 spallation neutron in an extended uranium setup and calculate it as follows:

$$\mathbf{E(n)_{electric\ power} = 1000\ MeV : 61\ n = 16.4\ MeV / n} \qquad \mathbf{(11)}$$

In order to carry out a first-order estimation of the total energy balance in such an extended uranium target, the following considerations are added: Hashemi-Nezhad, et al. [15b] have calculated that not only neutrons are emitted during spallation reactions in extended targets, however, one must count also in addition about 20% spallation protons with slightly larger energy. Altogether one observes in such an extended uranium target 73 spallation nucleons. It has been calculated that one 1 GeV proton looses about 150 MeV due to Coulomb interactions. This leaves only 850 MeV for the generation of 73 spallation nucleons. This allows estimating the *production cost of one spallation nucleon within an extended uranium target as:*

$$\mathbf{E(nucleon) = 850\ MeV : 73\ (nucleon) = 11.6\ MeV / (nucleon).} \qquad \mathbf{(12)}$$

It is well known, that the nuclear binding energy of nucleons in uranium is 7.6 MeV, the kinetic energy of spallation neutrons has been already mentioned (4.0 MeV). The considerations for a total energy balance in this system must be continued: in the present work we are not having all the relevant experimental facts included (i.e. proton emission energy, pion production, etc.), nor have we been taking into account the uncertainties of the experimental results.

2.4. Transmutation of ^{129}I, ^{237}Np, and ^{239}Pu with spallation neutrons

The complete presentation of transmutation studies on long-lived radioactive waste and/or transuranium nuclides, such as ^{129}I, ^{237}Np and ^{239}Pu, is presented here. The target systems used are shown in the preceding sections. The radioactive samples were placed on top of the moderator, as shown in Fig. 12.

The highly radioactive samples of about 0.5 g up to 1 g of ^{129}I, ^{237}Np, and ^{239}Pu were produced by the Institute of Physics and Power Engineering in Obninsk/Russia especially for these transmutation studies at the JINR, Dubna/Russia. Their construction is shown in Fig. 13. As the samples were placed outside the paraffin moderator, one must expect some influence of the neutron fluences around the entire target setup, in particular, when one takes into account the high neutron reaction cross-sections of transuranium nuclides. The irradiations and the data analysis have been carried out the same way as already described. The results are shown in Table 4.

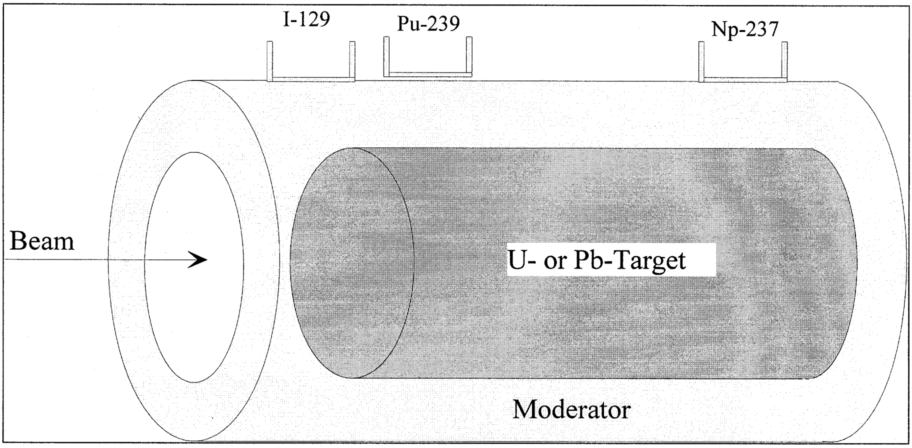

Fig.12: Experimental set-up for transmutation studies of ^{239}Pu, ^{129}I and ^{237}Np samples.

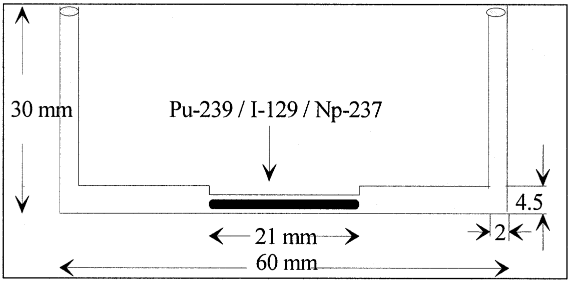

Fig. 13.: Transmutation samples of ^{239}Pu, ^{129}I and ^{237}Np well-sealed in Al-capsules. The amount of radioactivity in these samples was in the range of 0.5 g up to 1 g each, as given in Table 4.

For the case of transmutation of ^{239}Pu, some additional remarks are necessary: 11 fission fragments were identified through the analysis of the gamma spectra: ^{91}Sr, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{129}Sb, ^{132}Te, ^{133}I, ^{135}I, and ^{143}Ce. With the knowledge of the relative fission yields for thermal/epithermal neutron induced fission of ^{239}Pu it is possible to extract the "transmutation rate via fission" B(^{239}Pu) [13]. The results for all the transmutation B-values of the three long-lived radwaste nuclides as well as the corresponding B/E(p) values are shown in Table 4,.

- As it was shown in Table 2 and Fig. 9, the R(^{140}La)=B(^{140}La)/E(p) and R(^{239}Np)=B(^{239}Np)/E(p) values are constant within ±10% within the energy interval 0.5 GeV $\leq$ E(p) $\leq$ 7.4 GeV. (The values at 3.7 GeV deviate slightly more, most likely due to uncertainties in the fluence determinations at this energy, as described in [6]). These determinations were carried out in essentially well-shielded positions *inside* the paraffin moderator. As one observes variations of order of 30% in the B/E(p) values in Table 4, we assume a 30% systematic uncertainty in these B-values themselves.
- The neutron spectra at the positions of ^{129}I, ^{237}Np, and ^{239}Pu on top of the moderator are

different, as has been shown by the Minsk-group [6,20]. The neutron spectra become "harder" the more downstream is their position. Consequently a direct and strict comparison of the B-values for the three isotopes studied is not possible. However, for heuristic purposes this comparison is shown anyway in Table 5, together with some other B-values.

One can estimate from Table 4 and Ref. [8] the macroscopic transmutation rates of long-lived radwaste nuclei using a Pb-target set-up, as shown in Fig. 10 and irradiated with 10 mA proton beams at 1 GeV. Using 1 g samples these transmutation rates are:

6.7 mg of ^{239}Pu is transmuted per day,
3.3 mg of ^{238}U is transmuted in one month,
21 mg of ^{237}Np is transmuted in one month,
3 mg ^{129}I is transmuted in one month.

Of course the target shown in Fig.12 cannot be irradiated with 10 mA protons at 1 GeV, as it immediately would melt. This shows that our results have a certain indicative value, however, before such a transmutation process could become a technological reality, one needs considerably more research and development in this field. Nevertheless, these transmutation yields for ^{239}Pu agree with those estimated by Bowman et al. [3] on a theoretical basis. The comparison with the results from the CERN group shows satisfactory agreement, in particular considering, that they employed a higher proton energy and a different Pb-target without any moderator, resulting in a considerably “harder’ neutron spectrum [18-20].

Table 4: Experimental transmutation yields B and B/E(p) for the three long-lived radwaste nuclides under study. Uncertainties are about 30%. B-values are given in units of (E-4).

Proton E + target	B(^{129}I)	B(^{129}I) /E(p)	B(^{237}Np)	B(^{237}Np) /E(p)	B(^{239}Pu)	B(^{239}Pu) /E(p)
0.5GeV+U	1.30	2.60	3.1	6.2	-	-
1.0GeV+U	2.61	2.61	4.8	4.8	-	-
1.5GeV+U	2.33	1.55	8.2	5.4	-	-
7.4GeV+U	15.8	2.14	50.4	6.8	-	-
0.53GeV+ Pb	-	-	-	-	21	39
1.0GeV +Pb	0.81	0.81	3.3	3.3	31	31
1.5GeV +Pb	0.90	0.60	7.5	5.0	-	-
3.7GeV +Pb	3.0	0.81	30	8.1	-	-
7.4GeV +Pb	3.9	0.53	34	4.6	-	-
Weight of sample	0.43 g	-	1 g	-	0.449 g	-

Notes: The experiment (1GeV p + U/Pb) was carried out twice.

The experiments at (0.53GeV p + Pb), (0.5GeV p + U) and (1 GeV p + Pb) are from this work. The other results are from [6,8].

Table 5:_B-values for various sensors obtained on the outer mantel of the Pb-target and irradiated with 1 GeV protons. The uncertainties are about 15%. Two results from the CERN group obtained at similar geometric positions, but with very different neutron spectra are also given [18,19] .

Nuclear reaction	B value
^{239}Pu→fission (destruction of Pu)	3.1 E-03
^{238}U→^{239}Np (breeding of Pu)	5.5 E-05
^{237}Np→^{238}Np (destruction of Np)	3.3 E-04
^{139}La→^{140}La (practical sensor)	1.1 E-04
^{129}I→^{130}I (destruction of I)	8.1 E-05
^{239}Pu→fission (CERN) (with SSNTD, μg Pu)	5 E-03 (for 2.6 GeV protons)
^{129}I→^{130}I (CERN) (radiochemistry, 64,7 mg ^{129}I)	5 E-04 (for 2.6 GeV protons)

Conclusion

The present work gives a summary of transmutation studies carried out during recent years at the Synchrophasotron of the Laboratory for High Energies at the Joint Institute for Nuclear Research in Dubna, Russia, with major emphasis on experiments with 0.5 and 1.0 GeV protons yielding the following results:

- A new beam monitoring device based on nuclear chemical methods was tested and employed successfully. Two stacks of monitor foils were employed, one in contact with the massive metallic target, the other one placed 35 cm upstream into the proton beam.
- The transmutation yields of stable lanthanum into radioactive ^{140}La, as well as the corresponding yields for natural uranium into the nuclide ^{239}Np, increase on the surface of the relatively small target systems employed linearly within the entire proton energy range studied, ($0.5 \text{ GeV} \leq E(p) \leq 7.4 \text{ GeV}$). This behavior could be interpreted sufficiently well with model calculations based on the LAHET-code from Los Alamos, as well as with the DCM/CEM-code from Dubna.
- Using highly radioactive samples of about 0.5 up to 1 g of ^{129}I, ^{237}Np, resp. ^{239}Pu, one could measure for the first time their transmutation yields in a target system irradiated with high-energy protons. The results confirm theoretical estimations, that such transmutation processes can become practically feasible, when high-intensity (≥ 100 μA) accelerators for high-energy protons (≥ 0.5 GeV) may become easily available technically.
- Solid State Nuclear Track Detectors (SSNTD's) yield very valuable auxilary information, such as on the confirmation of the proton beam energy, the amount of thermal neutrons on the surface of the target system employed and on the fact, that inside our 6 cm thick paraffin moderator surrounding the metallic target of lead or uranium the neutron fluences are several times larger than on the moderator surface.

All these results show, that ADS-systems can be used in principle for large scale transmutation work. However, one will need considerable more "Research and Development" before this technology can become a technological reality.

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Appendix

A I.) Experimental estimations of neutron fluences using various SSNTD systems

This collaboration has published over the years many papers, where radiochemical techniques together with SSNTD techniques were independently used to determine neutron fluences at various geometrical positions in our target set-ups under a variety of different experimental conditions. The last of these more detailed reports was presented by Wan, et al. [6] using the target set-ups shown in Figs. 2 and 3, irradiated with 1.5, 3.7, and 7.4 GeV protons. Several experimental SSNTD results were described in detail and their relative merits have been evaluated. As this paper is a continuation of such work extended into the lower energy regimes of 0.5 and 1.0 GeV energy, with 1.5 GeV energy for control purposes, it suffices to report here only shortly on the various SSNTD experiments, concentrating on the pertaining results. Further details can be obtained from Ref. [6].

1.a.) Determination of thermal and epithermal neutrons on top of the moderator

Zamani and coworkers [21] carried out experiments with LR115(B)-Kodak, covered with $^6Li_2B_4O_7$ converter for thermal neutrons. This allowed a direct determination of the fluence of thermal neutrons. The energetic neutrons were determined with Cr-39 SSNTD 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 AI,1 and AI,2. The distribution of neutrons along TOP of the cylinder surface for the Pb- and U/Pb-targets is compatible with the distribution of B(^{140}La) and B(^{239}Np) in the same geometrical positions as shown in Figs. 6 and 7.

Table A I,1.: Neutron fluences per primary proton on the outer surface of the moderator in the experiment *(0.53 GeV protons + Pb),* as measured with SSNTD. The targets are evenly distributed on top of the moderator. Details are discussed in the text.

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 A I,2.: Neutron fluences per primary proton on the outer surface of the moderator in the experiment *(1.0 GeV protons + Pb),* as measured with SSNTD. The targets are evenly distributed on top of the moderator. Details are discussed in the text.

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

The average values for thermal neutron fluences per primary proton on the outer surface of the moderator can be calculated from both tables (and unpublished data) as shown in Table AI,3 and compared to theoretical estimations of Ref. 15b, where the numbers of low energy neutrons with (E < 1 eV) are given.

Table AI,3.: Number of thermal neutrons, N, as observed experimentally with SSNTD and its comparison with model calculations. Details are discussed in the text.

Reaction	N(thermal) SSNTD experiment	N (E < 1 eV) LAHET-code	N (E < 1 eV) DCM/CEM-code
0.5 GeV p + Pb	3.6 ± 1.2	1.4	2.4
1.0 GeV p+ Pb	5.6 ± 1.5	2.75	4.3
1.5 GeV p + Pb Ref. [6]	6 ± 2 Ref. [6]	3.7 Ref. [15b]	5.8 Ref. [15b]
0.5 GeV p + U/Pb	11.6 ± 3.6	2.3	4.3
1.0 GeV p + U/Pb	26 ± 6	5.5	9.1
1.5 GeV p + U/Pb, Ref.[6]	14 ± 3 Ref. [6]	7.6 Ref. [15b]	12.3 Ref. [15b]

The results of Table AI,3 show a general agreement with the radiochemical results and the corresponding model calculations, only the experiments at 0.5 and 1.0 GeV proton energy using the U/Pb target set-up give a factor of about (2-3) too large numbers. This is most likely due to the inherent difficulties to measure the very low total primary proton fluences for SSNTD exposures at the Synchrophasotron [6]. The experiments at 1.5 GeV agree rather well with the model calculation.

1.b.) Some further SSNTD experimental results

I.) *Debeauvais* and coworkers have investigated over the years the production of thermal neutrons - with thin ^{235}U sensors on Lexan SSNTD's - and the production of energetic (E[n] > 2 MeV) neutrons - with thin ^{232}Th sensors on Lexan SSNTD's - well *inside the 6 cm thick paraffin moderator* [22] . In this way they could show experimentally that in the middle of the 6 cm thick moderator the fluence of thermal neutrons is about 4 times larger than on the outer surface of the moderator for Pb- and U/Pb-targets used by us and irradiated with protons of energies E(p) in the range 1.5 GeV ≤ E(p) ≤ 7.4 GeV. This behavior was also observed in two irradiation during our present experiments, as shown in Table A I.4. Further experimental details can be found in Refs. [6] and [22].

The results from Table A I.4 show, that the fluence of thermal neutrons is a factor of (2 - 3) larger in the middle of the moderator at d = 7 cm also at relatively low proton energies. This implies, that the corresponding B-values of Table 5, measured at the moderator surface at d = 10 cm would increase correspondingly in the middle of the moderator for the cases, where B is caused predominantly by thermal neutrons. B-values predominantly caused by higher energy neutrons do not show such strong dependences. This is demonstrated for energetic neutrons with E(n) > 2 MeV, able to induce fission in ^{232}Th: here the B(fission)-value decreases slowly with increasing distance d. This confirms the earlier statements, that our actual „transmutation rates“ given for a 10 mA/1GeV accelerator (Table 5) are only indicative. Much more "research and development" (R&D) is needed, before such a transmutation system will become operative. On the other hand, the intensity of energetic neutrons with E(n) > 2 MeV, able to induce fission in ^{232}Th, decreases slowly with increasing distance d.

Table A I.4.: Effective neutrons per proton inside the paraffin moderator at a certain distance d from the central axis. Uncertainties are about 20%. Details are discussed in the text.

reaction	d = 4 cm	d = 5.5 cm	d = 7 cm	d = 8.5 cm	d = 10 cm
0.53GeVp+U 235-U sensor	2	4.8	4.1	5.4	1.4 theor:2.3-4.3
0.53GeVp+U 232-Th sensor	2.3	------	0.5	---	0.3
1.0GeVp+Pb 235-U sensor	14	19	23	19	8.6 theor:2.75-4.3
1.0GeVp+Pb 232-Th sensor	9	-----	2.3	---	1.3

Notes: The distances d are measured along the radius from the axis of the target system: d = 4 cm is between the metallic target and the moderator, d = 10 cm is the surface of the moderator. These SSNTDs were placed 10 cm downstream into the longitudinal middle of the target set-up. The SSNTD targets had a surface of about 1 cm^2. The theoretical estimations at d = 10 cm are again from Ref. 15b, giving the range of values from the two models used for low energy neutrons with E(n) < 1 eV. These values have already been given in the preceding Table A I.3.

These experiments *inside the paraffin moderator* have also been performed with ^{139}La-sensors, similar to the ones described earlier in this paper. The results for observed B(^{139}La) values measured *inside* the moderator are given in Table AII,3. We can confirm that the radiochemical results are more accurate, however, the SSNTD give additional information on the effect of thermal neutrons in comparison with energetic (E[n] > 2 MeV) neutrons.

II.) *Dwivedi* and coworkers have measured over the years the fluence of very high energy neutrons (E(n) > 30 MeV) with the fission reaction in gold [6]. The number of fission fragments was measured as „tracks in mica detectors“. A SSNTD target was placed in between the metallic target and the moderator, approximately in the middle of the cylinder during the entire proton irradiation. The technique has been described in Ref. [6]. The results are shown in Table AI.5.

The result of Table AI.5 is, that at (1.5 GeV p + U/Pb) the present experiment agrees with the previous determination published in Ref. [6]. The „effective fission rate“ decreases with the proton energy, however, not quite as linear as one would expect. SSNTD experiments yield here essentially auxiliary information. It is shown in [6], that these experiments agree with the respective model estimations of about 1.1 very high energy (E[n] > 30 MeV) neutrons per proton at 1.5 GeV.

III.) *Guo* and coworkers [23] have measured over the years the fluence of energetic neutrons (E(n) > 10 MeV) using 20 cm long, 1 cm wide and 1mm thick CR39 slides, placed along the axis of the metallic target. In this way they supplied additional evidence on the proton energy. They could show in this experiment, that the decrease of track density along the beam direction is much more rapid for O.53 GeV protons, as compared to 1.0 GeV protons, as shown in Fig. A1 (from Ref. [23])

Table A I.5.: Measurement of the high energy neutron component (E(n) > 30 MeV) with SSNTD using „gold on mica“ as sensor.

Reaction	D(tracks/cm^2)	T/(mg Au/cm^2)	D/Φ·T
0.53GeVp+U/Pb	2.42 E4	1.56	1.3 E-9
1.0 GeVp + U/Pb	3.50 E4	1.62	1.8 E-9
1.5 GeVp + U/Pb	4.20 E4	1.57	1.9 E-9
1.5 GeVp + U/Pb [6]	0.53 E4 [6]	1.46 [6]	2.4 E-9 [6]

Notes: D is the track density in (tracks/cm^2), T is the gold sample thickness in (mg Au/cm^2), Φ is the total proton fluence measured radiochemically and obtained during the entire proton irradiation. The product D/Φ·T is a measure of the „effective fission rate“; its uncertainty is about 20%.

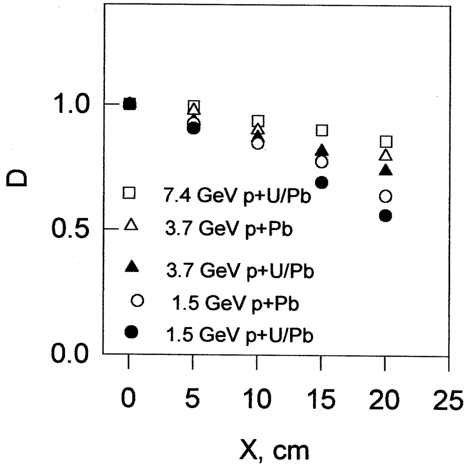

Fig. A 1.: Normalized track yields D at different positions of the inner CR39 detectors placed in contact with the Pb-target for different proton energies. Details are given in Ref. [23].

IV.) Zhuck and coworkers [20] carried out experiments with sandwiches containing artificial mica or lavsan as SSNTDs and thick foils with U- natural, U-6.5% (6.5% enrichment as to ^{235}U) and ^{232}Th as sensors allowing to determine the fission rates of ^{235}U and ^{232}Th as well as spectral index $\overline{\sigma}_f^{232Th} / \overline{\sigma}_f^{235U}$. The fission process of ^{235}U characterises neutrons of all energies (mainly thermal for set-ups with paraffin moderator) and the fission process ^{232}Th - only fast neutrons (with energies higher than the fission threshold of ^{232}Th ~1.5 MeV). Seven sandwiches were placed along the TOP of the cylinder surface at the Pb- and U/Pb-targets. The SSNTDs and sensors were exposed to 7-10 beam pulses from Synchrophasotron with a total fluence of proton approximately 10^{12}. The tracks density on the surface of SSNTDs for U-6.5%, ^{nat}U and ^{232}Th sensors was 10^6, 10^5 and 10^4, correspondingly. Additional SSNTDs with ^{232}Th sensors were irradiated at the same positions during the full time and used for linkage with the full protons fluence determined radiochemically. The fission rates of ^{235}U and ^{232}Th and the spectral index $\overline{\sigma}_f^{232Th} / \overline{\sigma}_f^{235U}$ at the seven positions placed along the TOP of the cylinder surface at the Pb-and U/Pb-targets were determined. The details are given in Refs.[6] and [20]. Spectral characteristics of the neutron flux along the TOP of the cylinder surface for the Pb- and U/Pb-targets were determined by measuring distribution ratios of average cross-section of ^{232}Th fission to average cross-section of ^{235}U fission ($\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$). The results of measurements for protons energy of 1.0 GeV are given in Table AI.6 and in Fig A2.

Table A I.6: Distribution of spectral index $\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$ along the TOP of the cylinder surface of U/Pb- and Pb-targets with parrafin moderator for 1.0 GeV protons.

Z, mm	$\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$	
	U/Pb-target	Pb-target
6.5	(3.01±0.30) ×10^{-3}	(5.80±0.58) ×10^{-3}
75	(1.31±0.12) ×10^{-3}	(1.96±0.18) ×10^{-3}
125	(1.24±0.11) ×10^{-3}	(2.15±0.19) ×10^{-3}
175	(1.26±0.11) ×10^{-3}	(2.45±0.22) ×10^{-3}
225	(1.49±0.13) ×10^{-3}	(3.05±0.27) ×10^{-3}
275	(2.62±0.24) ×10^{-3}	(5.40±0.49) ×10^{-3}
303.5	(1.01±0.10) ×10^{-2}	(1.94±0.19) ×10^{-2}

The addition of uranium (U/Pb-targets) softens the neutron spectrum: the ratio $\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$ is in the target centre larger by a factor of approximately 2 for Pb-targets as compared to U/Pb-targets. At the ends of paraffin moderator the neutron spectrum becomes harder for all set-ups. The value $\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$ is the beginning and at the end of the surface of paraffin moderator larger by a factor of about 2.4 and 8 as compared to the central part. It should be marked that there is the central region with more than 10 cm length with a rather constant neutron spectrum on the surface of the moderator, where the value $\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$ remains constant within the limits of experimental errors.

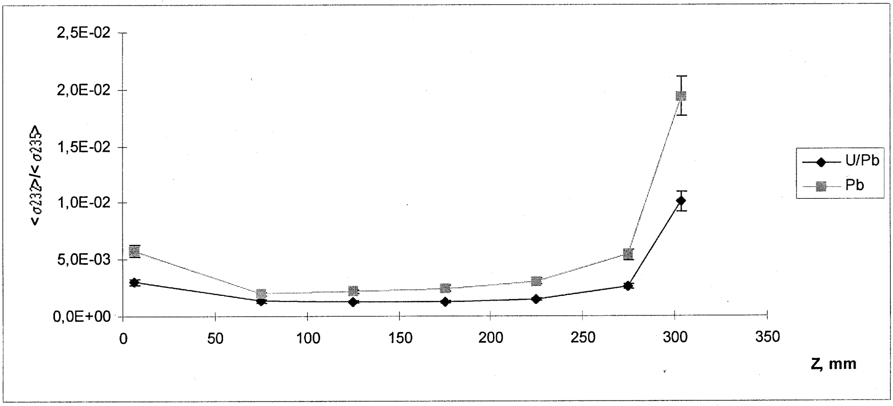

Fig. A 2.: Distributions of spectral index $\overline{\sigma_f}^{232Th}/\overline{\sigma_f}^{235U}$ along the TOP of the cylinder surface of U/Pb- and Pb- targets with paraffin moderator for 1.0 GeV protons.

A II.) Tables for B(^{140}La) for the sensor ^{139}La(n, γ)^{140}La.

(only statistical uncertainties are given, the total uncertainty is about 15%).

(These tables give detailed results for the 1999 experiments. Similar tables for the 1998 experiments are given in Ref. [8])

Table A II,1 : B(^{140}La) × (E-05) for ^{139}La(n, γ)^{140}La in TOP-sensors.

TOP Sensors	0.53 GeV p + U / Pb	0.53 GeV p + Pb	1.0 GeV p + U / Pb	1.0 GeV p + Pb
1 (0 cm)	11.1 (1±5.8%)	5.69 (1±5.7%)	21.1 (1±5.4%)	10.7 (1±6.3%)
2 (5 cm)	13.7 (1±7.8%)	8.22 (1±5.2%)	27.5 (1±5.5%)	13.0 (1±6.6%)
3 (10 cm)	11.3 (1±5.3%)	7.12 (1±5.6%)	21.7 (1±4.9%)	14.5 (1±6.8%)
4 (15 cm)	7.09 (1±5.1%)	5.52 (1±5.0%)	18.1 (1±5.4%)	11.8 (1±5.4%)
5 (20 cm)	4.31 (1±6.8%)	3.06 (1±6.5%)	10.7 (1±5.4%)	7.2 (1±4.7%)
Average	9.48 (1±6.8%)	5.92 (1±5.7%)	20.2 (1±5.5%)	11.4 (1±6.4%)

Table A II,2 : B(^{140}La) × (E-05) for ^{139}La(n, γ)^{140}La in RING-sensors.
(Azimuthal variations.)

RING Sensors	0.53 GeV p + U / Pb	0.53 GeV p + Pb	1.0 GeV p + U / Pb	1.0 GeV p + Pb
3 (0^0)	11.3 (1±5.3%)	7.11 (1±5.6%)	21.7 (1±4.9%)	14.4 (1±6.8%)
10 (60^0)	10.5 (1±4.2%)	9.08 (1±4.4%)	23.9 (1±9.8%)	12.7 (1±8.5%)
9 (120^0)	13.7 (1±4.4%)	7.20 (1±5.5%)	21.5 (1±4.8%)	17.0 (1±6.4%)
8 (180^0)	14.3 (1±5.7%)	10.6 (1±4.3%)	34.9 (1±7.4%)	18.9 (1±4.6%)
7 (240^0)	10.9 (1±5.2%)	8.61 (1±6.2%)	29.5 (1±5.1%)	10.6 (1±4.1%)
6 (300^0)	12.5 (1±4.0)	7.46 (1±5.1%)	21.9 (1±4.8%)	13.6 (1±4.7%)

Table A II,3 : B(^{140}La) • (E-05) for ^{139}La(n, γ)^{140}La in radial-sensors inside the paraffin moderator below the position of sensor 3, as shown in Figs. 2 and 3.
R is the distance from the beam center to the sensor.

Radikal Sensors	0.53 GeV p + U / Pb	0.53 GeV p + Pb	1.0 GeV p + U / Pb	1.0 GeV p + Pb
R=4.5 cm	47.5 (1±4.9%)	44.2 (1±5.1%)	106 (1±4.5%)	69.8 (1±4.7%)
R=6.5 cm	44.2 (1±4.9%)	35.1 (1±4.3%)	102 (1±3.9%)	
R=8.5 cm	21.2 (1±9.2%)	15.2 (1±5.6%)	53.6 (1±5.7%)	28.2 (1±6.2%)
R=9.7 cm	11.3 (1±5.3%)	7.12 (1±5.6%)	21.7 (1±4.9%)	14.5 (1±6.8%)

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Трансмутация ^{239}Pu и других нуклидов с использованием спалляционных нейтронов, генерируемых релятивистскими протонами в массивных урановых и свинцовых мишенях

Трансмутация некоторых долгоживущих ядер, таких как ^{129}I, ^{237}Np и ^{239}Pu, а также природного урана и лантана (два последних элемента использовались в качестве активационных детекторов) изучена экспериментально на пучке синхрофазотрона Лаборатории высоких энергий ОИЯИ (Дубна). Спалляционные нейтроны генерировались релятивистскими протонами с энергией в области 0,5 ГэВ $<E(p)<$ 1,5 ГэВ, которые взаимодействовали с урановыми и свинцовыми мишенями длиной 20 см. Мишени были окружены парафиновыми замедлителями толщиной 6 см. Радиоактивные образцы располагались на внешней поверхности замедлителя и содержали от 0,5 до 1 грамма долгоживущего изотопа. Образцы были изготовлены в Физико-энергетическом институте (Обнинск, Россия) и упакованы в герметичные алюминиевые оболочки. Из полученных экспериментальных данных по скорости трансмутации можно экстраполировать, что в подкритической электроядерной энергетической сборке такого типа при использовании пучка протонов, падающих на свинцовую мишень при энергии 1 ГэВ и токе 10 мА, можно трансмутировать 3 мг ^{129}I, 21 мг ^{237}Np, 3,3 мг ^{238}U и 200 мг ^{239}Pu в образцах радиоактивных изотопов массой 1 г, которые облучались в течение месяца при сохранении тех же геометрических размеров. Результаты показывают, что способность системы трансмутировать радиоактивные изотопы линейно возрастает с энергией протонов в рамках изученного энергетического интервала.

Работа выполнена в Лаборатории высоких энергий ОИЯИ.

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Adam J. et al. E1-2001-136

Transmutation of ^{239}Pu and Other Nuclides Using Spallation Neutrons Produced by Relativistic Protons Reacting with Massive U- and Pb-Targets

Experimental studies on the transmutation of some long-lived radioactive waste nuclei, such as ^{129}I, ^{237}Np, and ^{239}Pu, as well as on natural uranium and lanthanum (all of them used as sensors) were carried out at the Synchrophasotron of the Laboratory for High Energies (JINR, Dubna). Spallation neutrons were produced by relativistic protons with energies in the range of 0.5 GeV $\le E(p) \le$ 1.5 GeV interacting with 20 cm long uranium or lead target stacks. The targets were surrounded by 6 cm paraffin moderators. The radioactive sensors mentioned above were positioned on the outside surface of the moderator and contained typically approximately 0.5 up to 1 gram of long-lived isotopes. The highly radioactive targets were produced perfectly well-sealed in aluminum containers by the Institute of Physics and Power Engineering, Obninsk, Russia. From the experimentally observed transmutation rates one can easily extrapolate, that in a subcritical nuclear power assembly (or «energy amplifier») using a 10 mA proton beam of 1 GeV onto a Pb-target as used here, one can transmute in one gram samples of the isotope within one month about 3 mg ^{129}I, 21 mg ^{237}Np, 3.3 mg ^{238}U, and 200 mg ^{239}Pu under the same geometrical conditions. Our observations show, that the transmutation ability of our system increases linearly with the proton energy within the energy interval studied.

The investigation has been performed at the Laboratory of High Energies, JINR.

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