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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Summary. 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 were carried out at the Synchrophastron of the Laboratory for High Energies at the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. The radioactive targets (I, Np and Pu) were contained in weld-sealed aluminium holders produced by the Institute of Physics and Power Engineering, Obninsk, Russia. Spallation neutrons were produced by relativistic protons with energies in the range of 0.5 GeV $\leq E_p \leq$ 1.5 GeV interacting with 20 cm long uranium or lead target stacks. The metallic targets were surrounded by 6 cm thick paraffin moderators. The uranium and lanthanum samples were positioned on the outside of the moderator surface and typically contained approximately 0.5 to 1.0 gram of uranium or lanthanum. The highest fluence of spallation neutrons was observed in the region of 5 to 10 cm downstream the entrance of the primary beam into the metallic target, rather independent of the target material or the proton energy. The results obtained by nuclear chemistry methods were supplemented by SSNTD (Solid State Nuclear Track Detector) studies. Consistent and systematic results of B-values and spectral distributions for neutrons have been found. From the experimentally observed transmutation rates one can 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 within one month in one gram of sample about 3 mg ^{129}I, 21 mg ^{237}Np, 3.3 mg ^{238}U, and 200 mg ^{239}Pu. Rather similar results have been found by another group for ^{129}I and ^{239}Pu. Observations show that the transmutation rates increase almost linearly with the proton energy in the energy interval 0.5 GeV up to 7.4 GeV. These findings are largely confirmed by model calculations using the LAHET- and DCM/CEM-codes.

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 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 of $\geq$ 1 mA to subcritical nuclear assemblies 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 producing energy with such systems; we will rather focus our attention on the aspect of transmutation. The term describes in this context the *transmutation* of long-lived radioactive fission products into short-lived or finally stable nuclei, such as the transmutation

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of ^{129}I ($t_{1/2} = 1.6 \times 10^{+6}$ a):

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

The same term transmutation is also used to describe the conversion of long-lived actinides, such as ^{237}Np ($t_{1/2} = 2.1 \times 10^{+6}$ a) or ^{239}Pu ($t_{1/2} = 2.4 \times 10^{+4}$ a) into short-lived products, such as

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

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

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

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

Modern aspects of transmutation were introduced in a theoretical approach by Bowman *et al.* 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 within an extended subcritical nuclear power plant. Our team has backed their theoretical approach with experiments and published a series of papers on the transmutation of ^{129}I, ^{237}Np, ^{nat}U, and stable ^{139}La in recent years [4–8]. We used a small metallic 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 principal physical features. The proton energies used so far were 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 0.45 g of plutonium, were employed.
- The spallation targets were irradiated with 0.53 GeV and 1.0 GeV protons, in addition to another 1.5 GeV run for control purposes. These studies appear to be interesting, as similar energies have been used at CERN in their experiments [9].
- A modified classical proton fluence measuring system is used. Two monitor foil-stacks, one in contact with the lead target and the other 35 cm upstream were employed in order to measure in a direct and reliable way the proton fluences [10].

The experimental results as well as a theoretical estimation of the conversion of radiochemically observed transmutation rates into neutron yields will be presented. Detailed results of auxiliary SSNTD experiments are also described.

2. Proton fluence measurements

The proton fluence measurements were carried out with a proton fluence monitor system using activation techniques. This new system has two stacks, each consisting of one 1 mm thick Cu foil and a stack of three 30 μm thick Al foils, as shown in Fig. 1. One stack is in contact with the target (monitor 1), the other one is placed 35 cm upstream. After the proton irradiation, the Cu-foil and the middle Al foil were assayed for ^{24}Na activity and the Al foils were

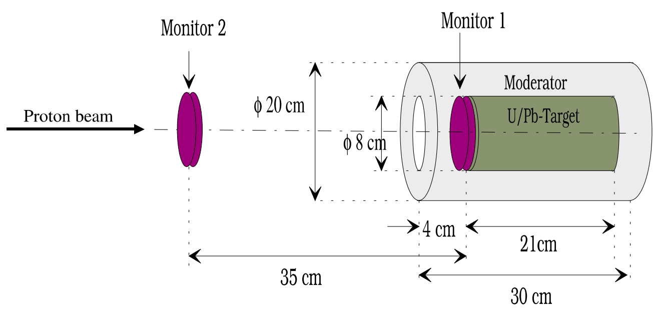

Fig. 1. The experimental layout of the new activation method to determine the proton fluence with two monitor foil stacks [10], for details see text.

Table 1. Details of the proton irradiations. (The fluence gives the total number of protons impinging onto the entire metallic target surface during the irradiation.)

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

Experiment	Fluence (Cu-Monitor) Monitor 1	Fluence (Cu-Monitor) Monitor 2	Fluence (Al-Monitor) Monitor 1	Fluence (Al-Monitor) Monitor 2	Final fluence
0.5 GeV $p+\mathrm{U}^a$				1.17 ± 0.14	1.17 ± 0.14
1.0 GeV $p+\mathrm{U}$	1.22 ± 0.22	1.09 ± 0.21	3.15 ± 0.38	1.23 ± 0.15	1.23 ± 0.15
1.5 GeV $p+\mathrm{U}$	1.41 ± 0.27	1.37 ± 0.27	6.67 ± 0.80	1.41 ± 0.17	1.41 ± 0.17
1.0 GeV $p+\mathrm{Pb}^b$	1.22 ± 0.24	1.25 ± 0.25	2.19 ± 0.27	1.30 ± 0.16	1.30 ± 0.16
1.5 GeV $p+\mathrm{Pb}^b$		1.31 ± 0.26	2.57 ± 0.31	1.29 ± 0.16	1.29 ± 0.16

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

Experiment	Monitor 2 ^{24}Na	Monitor 2 ^{7}Be	Monitor 2 ^{22}Na	Monitor 1 ^{7}Be	Monitor 1 ^{22}Na	Final fluence
0.53 GeV $p+\mathrm{U}$	5.65 ± 0.72	5.95 ± 0.90	6.00 ± 0.90	6.32 ± 0.95	5.93 ± 0.89	5.93 ± 0.72
0.53 GeV $p+\mathrm{Pb}^a$				6.43 ± 0.45	6.34 ± 0.42	6.39 ± 0.96
1.0 GeV $p+\mathrm{U}$	12.1 ± 1.50	12.7 ± 1.92	12.6 ± 1.90	12.5 ± 1.91	12.5 ± 1.90	12.5 ± 1.50
1.0 GeV $p+\mathrm{Pb}^a$	12.8 ± 1.54	13.6 ± 2.01	12.4 ± 1.86	14.5 ± 2.18	13.1 ± 1.97	13.3 ± 1.56

a: Simultaneously we irradiated ^{129}I, ^{237}Np, and ^{239}Pu samples, as indicated in Table 4;
b: In these two Pb-target irradiations, **no** moderator was installed (“blank” targets).

also investigated in some experiments for ^{7}Be and ^{22}Na activities. The details of this technique have already been published [10]. The results of the proton fluence measurements are given in Table 1. Uncertainties of about ±12% are due to statistical uncertainties in the measurements and systematic uncertainties in the monitor reaction cross-sections. Fluences based on ^{24}Na determinations in Al-foils of monitor 2 at a distance of 35 cm upstream of 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 [10]. As it is fairly straightforward to accurately measure ^{24}Na in thin Al-foils, we essentially used the proton fluence determined with ^{24}Na in Al (monitor 2). These fluences are experimentally most accurately determined.

3. Transmutation studies using ^{nat}U- and ^{139}La-samples: Calibration of the experimental set-up

The two principal experimental set-ups used by our group for many years [4–8] are shown:

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

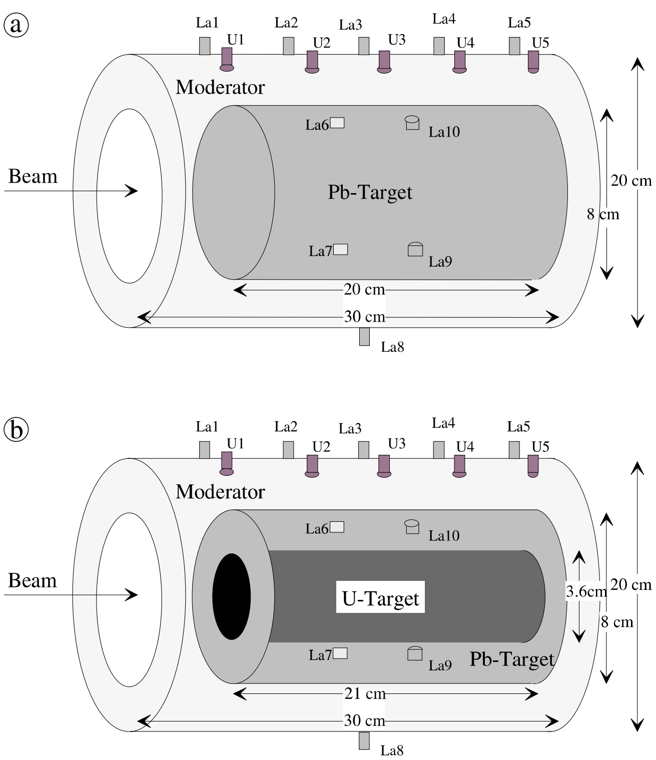

Fig. 2. (**a**) Small lanthanum (La-1–La-10) and uranium (U1–U5) samples *on the surface* of the 6 cm paraffin moderator surrounding the massive Pb-target. (**b**) Small lanthanum (La1–La10) and uranium (U1–U5) samples on the surface of the 6 cm paraffin surrounding the massive ^{nat}U target and the 2 cm Pb. This system is called U/Pb-target.

Several La-samples (La1–La10) each containing approximately 1 g La in the form of ($LaCl_3 \cdot 7H_2O$) are located on the surface of the moderator or in exactly 10 mm deep holes [5]. Spallation neutrons are produced in the metallic target some of which are thermalized within the 6 cm paraffin moderator. The actual neutron spectra on the surface of the moderator have been calculated and published [5, 6]. Neutrons of all energies may induce nuclear reactions, however, the following reaction is most sensitive to thermal neutrons [5, 6]:

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

The product nucleus ^{140}La has a half-life of 40 hours and can easily be determined *via* gamma spectrometry.

In adition we have placed simultaneously five samples of approximately 1 g ^{nat}U (U1–U5) on the top surface of the moderator. This allows the study of the transmutation of natural uranium into ^{239}Np:

$$^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U} \Rightarrow {}^{239}\mathrm{Np}\,. \tag{6}$$

In this publication the results for activity determinations of ^{239}Np are reported; this nuclide has a convenient half-life of 2.3 days and it is easily measured by gamma-ray spectrometry. The reaction

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

was also investigated, however, these results can be found in [8]. They are strictly analogous to those of the ^{239}Np-production rates [5–8].

The beam profiles have been determined with the SSNTD technique: Lavsan track detectors in contact with a thin Pb foil were placed upstream in front of the massive metallic target, at 10 cm inside the metallic block as well as behind the target. Essentially only high energy particles above approximately 50 MeV induce fission in Pb. The excitation function for hadron induced fission in lead is well-known, see for example [11]. These details are not of major importance for the purpose of the beam profile determination. The fission fragments were registered in Lavsan. The foil was etched after the irradiation and scanned with an optical microscope. Details of this straightforward technique are described in [12]. Typical beam profiles are shown in Fig. 3a,b for irradiations at 0.53 GeV and 1 GeV proton energy. The beam profile at 1.0 GeV proton is characteristic of experiments at this energy and higher energies [5–8]. The entire beam essentially matches the 8 cm ∅ metallic target and the beam is attenuated within the 20 cm long target, though it is not completely stopped. The 0.5 GeV proton beam is not sufficiently well focussed onto the metallic target and some protons hit the paraffin, but we consider this not to be very serious as low-Z materials, such as paraffin, are quite inefficient in producing spallation neutrons. The 0.5 GeV proton beam is completely stopped within the 20 cm metallic target as can be seen in Fig. 3a.

After the irradiations, the activated La- and U-samples and parts of the metallic lead targets were transported to

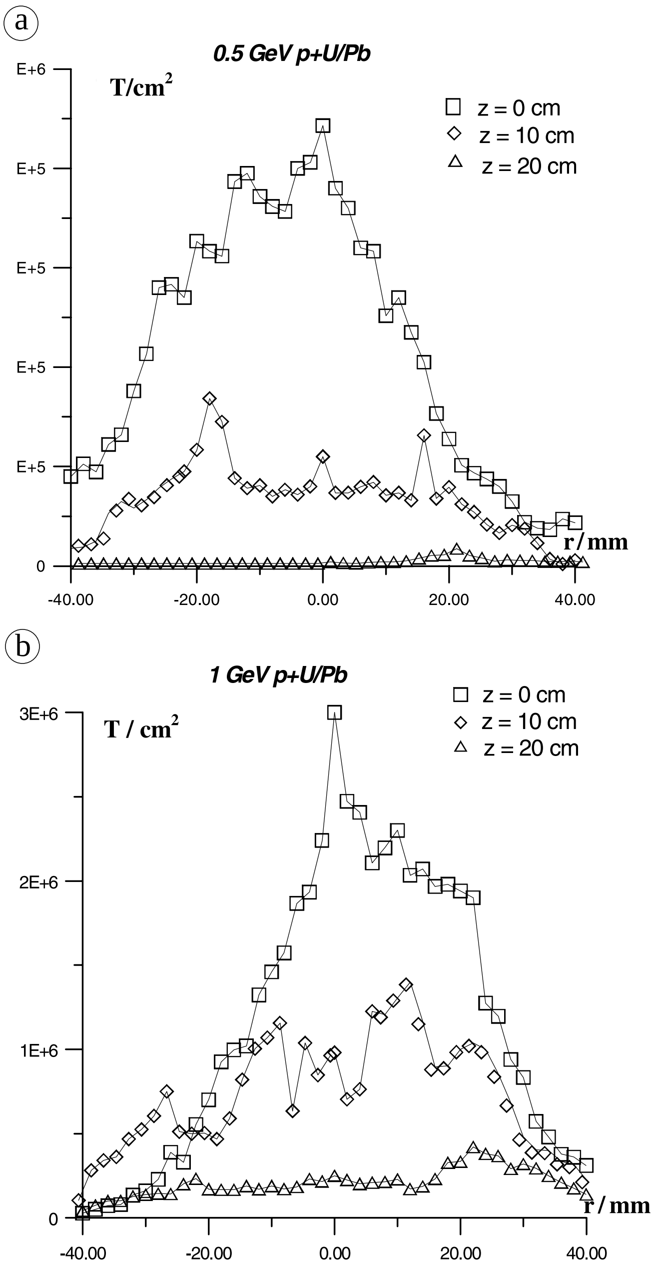

Fig. 3. (**a**) The beam profile for 0.53 GeV p (1998). The track density (number of fission tracks, T, per unit area) is given in T/cm^2. The z-values denote the beam profiles measured horizontally at different depths of the target: $z = 0$ cm is in front of the target, $z = 10$ cm is in the center, $z = 20$ cm at the end of the target. (**b**) The beam profile for 1.0 GeV p (1999). The track density (number of fission tracks, T, per unit area) is given in T/cm^2. The z-values denote the beam profiles measured horizontally at different depths of the target: $z = 0$ cm is in front of the target, $z = 10$ cm is in the center, $z = 20$ cm at the end of the target. uncertainties of these data are on the order of ±8%, the magnitudes of systematic uncertainties are not known.

a laboratory to investigate the gamma activity with HP Germanium counting systems using well-established procedures [13]. This allowed the determination of decay rates of radioactive nuclides produced. Activities at the end of bombardment were converted into production rates of these nuclei. As an example, we calculated the production rate of ^{140}La nuclei in the La-targets. In the literature, describing transmutation experiments of this kind, it is considered convenient to calculate a "transmutation rate" B which for the product nucleus ^{140}La is defined as follows:

$$B\left(^{140}\mathrm{La}\right) = \left(\text{number of } ^{140}\mathrm{La} \text{ nuclei formed}\right) / \{(1 \text{ g La target})\ (1 \text{ primary proton})\}\ [\mathrm{g}^{-1}]\,. \quad (8)$$

The 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 a specific target. As the reaction leading to ^{140}La is essentially sensitive only to thermal neutrons, the measurement of several $B(^{140}\mathrm{La})$-values for various geometrical positions around the target system gives a measure of the distribution of thermal neutrons around this target during the irradiation. In particular, the determination of "azimuthal" $B(^{140}\mathrm{La})$-values (samples La-3 plus La-6 to La-10 in Fig. 2) allows for the correction of azimuthal variations in the fluence of secondary neutrons. Details can be found in Refs. [5, 8]. Typical results for $B(^{140}\mathrm{La})$-values on the *top surface* of the moderator (La1–La5) are shown in Fig. 4a for the U/Pb target, corrected for azimuthal variations [5, 8]. Similar results for $B(^{239}\mathrm{Np})$-values in uranium-samples (U1–U5) on the top surface of the U/Pb target are shown in Fig. 4b.

The experimental results for $B(^{140}\mathrm{La})$ and $B(^{239}\mathrm{Np})$ using the Pb-target system are omitted in this paper. Re-

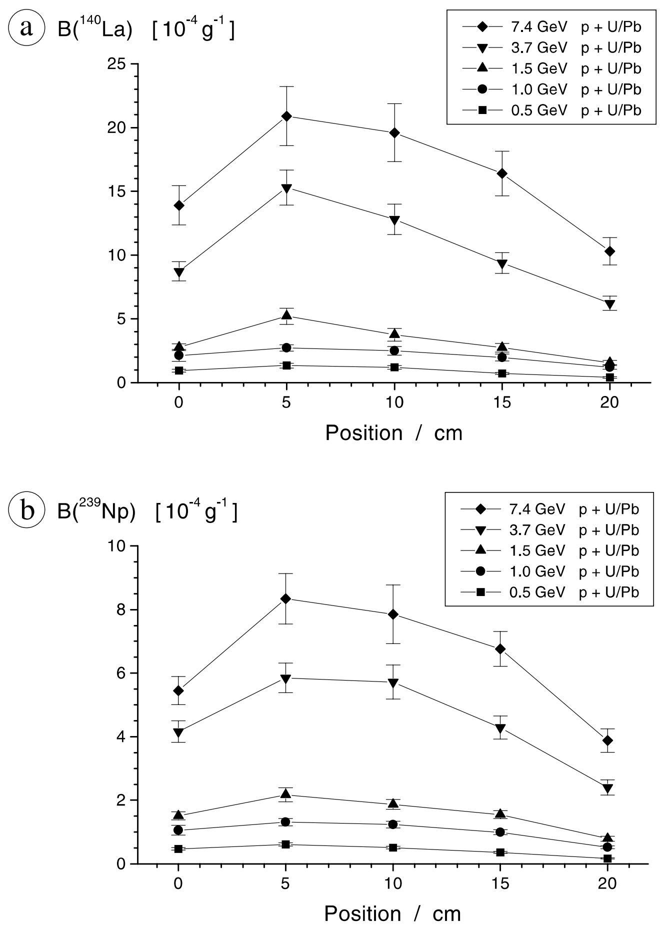

Fig. 4. (**a**) $B(^{140}\mathrm{La})$ in La-samples (La1–La5) distributed on the surface of the moderator for experiments with U/Pb targets. Results for 1.5, 3.7 and 7.4 GeV are from [6]. (**b**) $B(^{239}\mathrm{Np})$ in U-samples (U1–U5) distributed on the top of the surface of the moderator in experiments with U/Pb-targets. Results for 3.7 and 7.4 GeV from [6].

sults have been presented before: *B*-values from the Pb-target are reduced by a factor 1/1.7 as compared to U/Pb-targets [5–8].

The distribution of neutrons, measured *via* ^{140}La or ^{239}Np on top of the surface of the moderator is also known from previous experiments [5–8]: The maximum induced radioactivity is always found about 5 cm behind the entrance of the protons into the massive uranium target, rather independent of 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 block. Induced radioactivities always decrease from the middle towards the back-end of the target. Details for $B(^{140}\text{La})$-values from the experiments are tabulated in Refs. [8, 14].

The experimental average $B_{av}(^{140}\text{La})$-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 is calculated as the arithmetic mean value. As the geometry of our geometrical arrangement is very well-defined, it is not surprising that these values are reproducible and they can be compared to theoretical estimations. Various $B_{av}(^{140}\text{La})$-values for all experiments in this series are compiled in Table 2. It is also of interest to show in the same table the *relative transmutation rate* $R(^{140}\text{La})$, defined as follows:

$$R(^{140}\text{La}) = B_{av}(^{140}\text{La})/E_p\ [\text{g}^{-1}\ \text{GeV}^{-1}]\,. \tag{9}$$

These values $R(^{140}\text{La})$ are of interest, as they give a measure of the relative neutron production efficiency for a certain nuclear reaction in a given arrangement with respect to the proton energy. It is a relative measure for the number of spallation neutrons produced per unit proton energy in the primary interaction. $R(^{140}\text{La})$ is rather *energy independent within the proton energy range* studied. The same phenomenon is presented graphically for the average $B_{av}(^{239}\text{Np})$- and $R(^{239}\text{Np})$-values in Figs. 5 and 6 for U/Pb targets. Again, the values $R(^{239}\text{Np}) = B_{av}(^{239}\text{Np})/E_p$ are rather independent of the energy E_p. A preliminary account of this observation has already been published [15].

Last but not least, we want to report briefly on the result of experiments using the Pb-target without a paraffin moderator (see Table 1: "blank" Pb-targets). The La and U sensors were placed on top of the lead target and the resulting activities were induced by the whole spectrum of spallation neutrons that leak out of the metallic target. Transmutation rates are compared for experiments with and without paraffin moderator:

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

$$B_{av}(^{239}\text{Np})_{\text{moderator}}/B_{av}(^{239}\text{Np})_{\text{blank}} = 12 \pm 3\,. \tag{11}$$

These ratios are understood on a qualitative basis: due to the lack of moderator, the number of low energy and of thermal neutrons is considerably smaller around a "blank" lead target as compared to the outer surface of the moderator. As the ^{140}La production is practically only sensitive to thermal neutrons, the above ratio is rather large. For ^{239}Np, however, epithermal neutrons are also quite effective, and these are produced within the metallic "blank" target with reasonable cross sections. This results in a smaller *B*-value ratio.

Table 2. $B_{av}(^{140}\text{La})$ for the U/Pb target and the corresponding $R(^{140}\text{La}) = B_{av}(^{140}\text{La})/E_p$.

System studied	$B_{av}(^{140}\text{La})$ [10^{-4} g^{-1}]	$R(^{140}\text{La})$ [10^{-4} g^{-1} GeV^{-1}]
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 ± 1.58	2.84 ± 0.43
7.4 GeV p + U/Pb	16.2 ± 2.43	2.19 ± 0.32

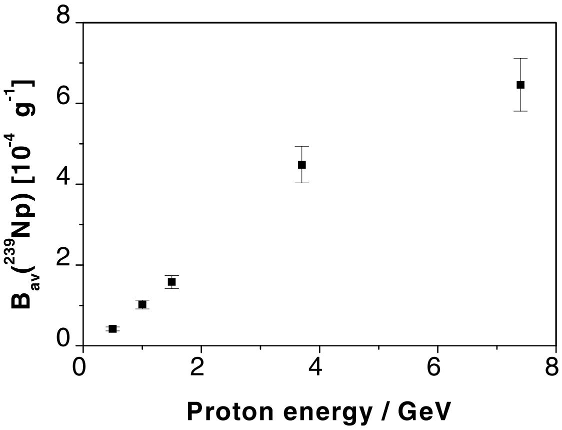

Fig. 5. Dependence of $B_{av}(^{239}\text{Np})$ in U/Pb-targets on the energy of the incoming proton.

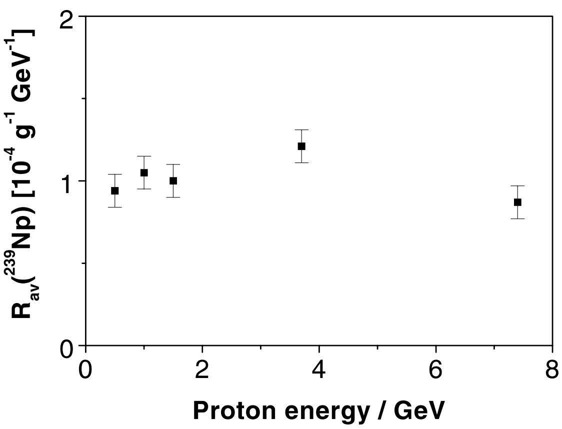

Fig. 6. Dependence of $R(^{239}\text{Np}) = B_{av}(^{239}\text{Np})/E_p$ in U/Pb-targets on the energy of the incoming proton.

For typical fission fragments such as as ^{132}Te one observes the same $B(^{132}\text{Te})$ value on top of the moderator and on the "blank" metallic surface, because neutrons of all energies induce fission in uranium [8]. The $B_{av}(^{140}\text{La})_{\text{blank}}$- and $B_{av}(^{239}\text{Np})_{\text{blank}}$-values measured at proton energies of 1.0 and 1.5 GeV are essentially identical at both energies. Further details are given in [8].

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

The experimental determination of $B_{av}(^{140}\text{La})$-values is certainly of practical interest; however, direct comparisons with model calculations are very difficult. One reason is that the 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 requires complex algorithms. Some attempts in this direction have been made in earlier publications without being completely convincing [5, 6]. In this paper another approach will be presented, based on a recent calibration experiment [16] and detailed theoretical calculations of Hashemi-Nezhad *et al.* [17]. In the calibration experiment a Pu/Be source emitting 8.1×10^6 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. 7). This setup is similar to the target shown in Fig. 2b but without a uranium insert. In this way the spallation neutron production in the accelerator-experiments was simulated quite realistically. On the outer surface of the paraffin moderator La-samples were placed, identical to those used in the transmutation experiments in Dubna. Additionally, "LR-115 2B" SSNTD strips were irradiated as shown in Fig. 7. One set of strips was bare, thus being exposed to a broad energy spectrum of neutrons, the other set of strips was covered with 0.75 mm thick Cd foils in order to absorb thermal neutrons. After the irradiation both the radiochemical and SSNTD sensors were analysed. The results for $B(^{140}\text{La})$-values along the longitudinal direction of the paraffin moderator are shown in Fig. 8a together with a model calculation using the MCNP-4B2 neutron transport code. Fig. 8b shows the same experimental result, this time after converting $B(^{140}\text{La})$ into neutron fluences using the same standard computational procedures already used in [5] and described in detail in [16]. The agreement between experiment and the model calculation in this calibration experiment is good. Similarly good results have been obtained in the analysis of the SSNTD calibration [16]. Using these data for calibration, the $B_{\text{av}}(^{140}\text{La})$-value observed for 1 neutron emitted from the spallation neutron source for La-samples placed into 1 cm deep holes in the moderator of the Dubna experiments could be determined as follows:

$$B_{\text{av}}(^{140}\text{La})_{\text{for 1 spallation neutron}} = 9.7\times10^{-6}\ [\text{g}^{-1}]\ (\pm13\%)\,. \tag{12}$$

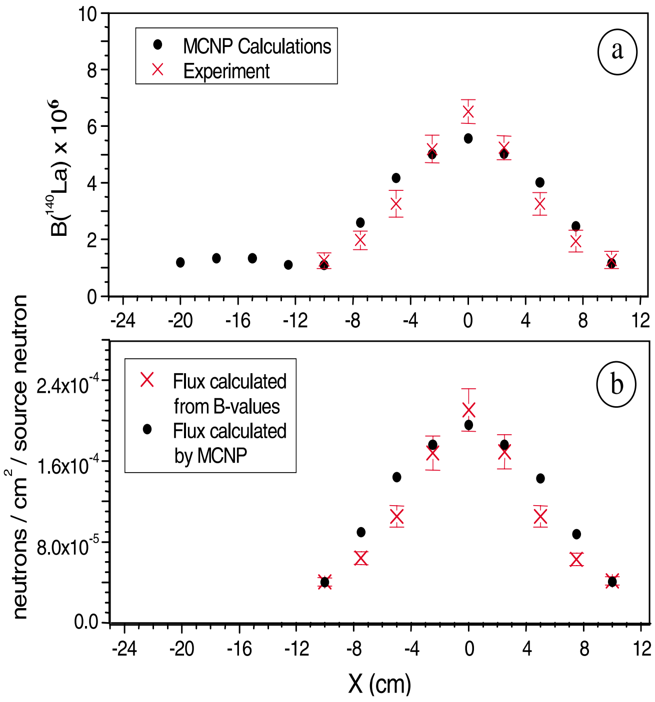

Fig. 8. Variation of the activation rate of ^{139}La, measured as $B(^{140}\text{La})$, and neutron fluence at the La sample locations, as a function of longitudinal distance on the moderator surface. The figures show both the experimental results and those obtained by MCNP-4B2 simulations. (**a**) activation rates and (**b**) slow neutron fluence. For details see [14].

The uncertainty of this conversion is mainly due to experimental uncertainties as described in [5]. Geometrical considerations have been taken properly into account. Eq. (12) can be used to estimate the number Y of spallation neutrons from a $B_{\text{av}}(^{140}\text{La})$-value for spallation reactions at the Synchrophasotron irradiations. Calculations 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 yields spallation neutrons with an average energy of 3.98 MeV. These average neutron energies are sufficiently close to allow a conversion of the $B_{\text{av}}(^{140}\text{La})$-value into the number of spallation neutrons Y(experimental) according to Eq. (12). Results of this conversion are shown in Table 3 together with results of a detailed theoretical cal-

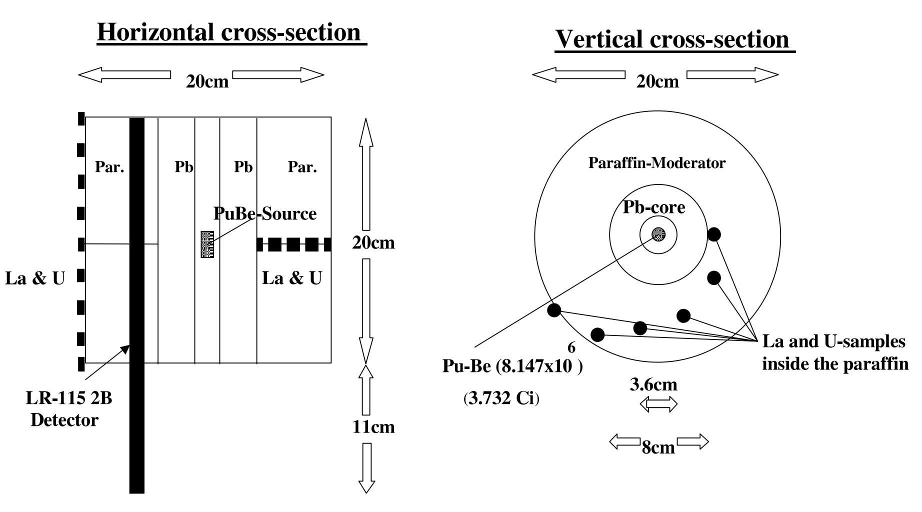

Fig. 7. The experimental set-up for the absolute calibration of $B(^{140}\text{La})$ using a well defined Pu/Be neutron source emitting 8.1×10^6 neutrons/second. For details see [16].

Table 3. Experimental $B_{av}(^{140}La)$ values for Pb-targets and their conversion into experimental spallation neutron number Y(experimental) together with theoretical model calculations. The results for the U/Pb-targets are shown for illustrative purposes, as the calibration has only been carried out for the Pb-target. The uncertainties in Y(experimental) are approx. ±20%.

Reaction	$B_{av}(^{140}La)$ Radiochemistry [g^{-1}]	Y(experimental) Based on Eq. (12) [neutrons/proton]	Y(theoretical) LAHET [neutrons/proton]	Y(theoretical) DCM/CEM [neutrons/proton]
0.53 GeV p + Pb	5.9×10^{-5}	6.1	7.7	9.3
1.0 GeV p + Pb	11.4×10^{-5}	11.7	15.3	17.4
1.5 GeV p + Pb	17.0×10^{-5}	17.5	19.8	23.6
3.7 GeV p + Pb	60.4×10^{-5}	62.3	38.7	43.9
7.4 GeV p + Pb	74.0×10^{-5}	76.3	61.1	70.8
0.5 GeV p + U/Pb	9.3×10^{-5}	9.6	10.4	17.2
1.0 GeV p + U/Pb	21.1×10^{-5}	21.7	23.5	35.1
1.5 GeV p + U/Pb	32×10^{-5}	33	32.4	47.2
3.7 GeV p + U/Pb	105×10^{-5}	108	65.4	93.6
7.4 GeV p + U/Pb	162×10^{-5}	167	112.1	151

culation for the number Y(theoretical) of spallation neutrons using the geometry of the target systems employed in this work and reported in detail in [17]. These calculations are based on two model codes: LAHET from the Los Alamos National Laboratory and DCM/CEM from the JINR, Dubna.

Experimental Y-values with an uncertainty of ±20% agree to some extent with model calculated Y-values. The discrepancies are mostly below 50% and never more than 100%. The discrepancy between experimental and theoretical values calculated with the LAHET code seems to be largest for U/Pb targets irradiated with 7.4 GeV protons. This appears to be state-of-the-art for this type of investigation, as published in a “Special Issue on Accelerator Driven systems” (Plendl, H. S., Ed.), Nucl. Instrum. Methods A **463**, No. 3 (2001).

Finally, we could study theoretically the neutron fluence within a very large heavy element experimental setup, used as a “subcritical nuclear assembly”. The CERN group published results of a related investigation, called the TARC experiment and using a $3.3 \times 3.3 \times 3\,m^3$ lead target. They found that 35.6 spallation neutrons are produced by one 1 GeV proton in this extended target [18–20]. Using these data we may compile an approximate energy balance for the production of spallation particles in a large accelerator driven transmutation setup. It is well known that the number of spallation and fission neutrons increases by (70 ± 10)% [5–8] when the target material is uranium rather than lead. This will enhance the neutron number from 35.6 to (61 ± 3) neutrons in an extended uranium target. According to calculations of Hashemi-Nezhad *et al.* [17] the abundance of spallation protons is about 20%, so the number of spallation particles is (76 ± 4) nucleons altogether. The incoming proton is calculated to lose about 150 MeV in the target through Coulomb interactions, thus the approximative energy balance for particle production in an extended U-target may read: 1 GeV proton energy *minus* 150 MeV Coulomb losses produce (76 ± 4) nucleons, *i.e.* one needs (11.2 ± 0.6) MeV of proton beam energy to produce one spallation nucleon. This estimate is not a real energy balance because minor contributions from other ejectiles (pions, deuterons, tritons, ... *etc.*) as well as some amount of energy released in uranium fission are not accounted for. It is, however, suitable to calculate that a 10 mA beam of 1 GeV protons will produce 4.7×10^{18} spallation particles per second in an extended uranium target.

5. Supplementary experimental estimates of neutron fluences using various SSNTD systems

Our collaboration has published several papers in which activation techniques and SSNTD techniques were independently used to determine neutron fluences at various geometrical positions in target setups under a variety of different experimental conditions. The last of these more detailed reports was presented by Wan *et al.* [6] using target setups shown in Fig. 2a,b and irradiated with 1.5, 3.7, and 7.4 GeV protons. The experimental SSNTD results were described in detail discussing their relative merits. 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 is sufficient to report here only briefly on the various SSNTD experiments with emphasis on the pertaining results.

5.1 Determination of thermal and epithermal neutrons on top of the moderator

Zamani *et al.* [21] carried out experiments with LR115(B)-Kodak, covered with $^6Li_2B_4O_7$ convertors for thermal neutrons. This allowed a direct determination of the fluence of thermal neutrons. More energetic neutrons were determined with CR-39 SSNTD with Cd-shielding which allowed the quantification of neutrons with energy of $300\,keV < E_n < 3\,MeV$ through the measurement of recoil-proton tracks. Five targets were distributed on top of the moderator. The targets were exposed to neutrons originating from a few beam pulses from the Synchrophasotron with a total fluence of approximately 10^{11} protons. In this case the proton beam fluence was taken from the machine operators, it could not be checked independently with radiochemical methods. The results are given in Tables 4 and 5. The distribution of neutrons along the upper surface of the target system for the Pb-targets is compatible with the distribution of $B(^{140}La)$ and $B(^{239}Np)$ in the same geometrical positions as shown in Fig. 4 (a and b). However, the radiochemical results show some decrease of the fluence downstream the target whereas SSNTD data do not.

The average values for thermal neutron fluence per primary proton on the outer surface of the moderator is calculated from both tables and shown in Table 6 together with theoretical estimations, as described earlier in [16]. The calculated numbers of low energy neutrons with ($E_n < 1$ eV) are also given. The results of Table 6 show a reasonable

Table 4. Number of neutrons per primary proton on the outer surface of the moderator in the experiment (0.53 GeV protons + Pb), as measured with SSNTD. The detectors are evenly distributed on top of the moderator. Energetic neutrons have energies E_n of 300 keV < E_n < 3 MeV. For details see text.

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

Table 5. Number of neutrons per primary proton on the outer surface of the moderator in the experiment (1.0 GeV protons + Pb), as measured with SSNTD. The detectors are evenly distributed on top of the moderator. Energetic neutrons have energies E_n of 300 keV < E_n < 3 MeV. For details see text.

Distance	2.5 cm	5.5 cm	8.5 cm	11.5 cm	14.5 cm
Thermal neutrons [neutrons/proton]	4.3 ± 9	4.7 ± 10	5.8 ± 12	6.7 ± 13	6.5 ± 1.3
Energetic neutrons [neutrons/proton]	2.6 ± 0.5	4.1 ± 0.8	4.1 ± 0.8	5.1 ± 1.0	5.1 ± 1.0

Table 6. Number of thermal neutrons, N, as observed experimentally with SSNTD and comparison with model calculations. For details see text.

Reaction	N (thermal) SSNTD experiment [neutrons/proton]	N ($E < 1$ eV) LAHET-code [neutrons/proton]	N ($E < 1$ eV) DCM/CEM-code [neutrons/proton]
0.53 GeV p + Pb	3.6 ± 1.2	1.4	2.4
1.0 GeV p + Pb	5.6 ± 1.5	2.75	4.3

agreement between the radiochemical results and the corresponding model calculations.

5.2 Further SSNTD experimental results

Debeauvais *et al.* investigated the production of thermal neutrons with thin ^{235}U targets on Lexan SSNTDs and the production of energetic ($E_n > 2$ MeV) neutrons with thin ^{232}Th targets on Lexan SSNTDs *inside the 6 cm thick paraffin moderator* [22]. With this method they could show experimentally that in the middle of the 6 cm thick moderator the fluence of thermal neutrons is nearly 4 times larger than on the outer surface of the moderator for Pb- and U/Pb-targets when irradiated with protons of energies E_p in the range 1.5 GeV $\leq E_p \leq$ 7.4 GeV. This same behavior was also observed in two irradiations during recent experiments, as shown in Table 7. Further experimental details can be found in [6, 22].

The results of Table 7 also show that at proton energies studied the fluence of thermal neutrons is a factor of 2 to 3 larger in the middle of the moderator at $d = 7$ cm, *i.e.* inside the moderator, than at the moderator surface. This implies that the corresponding B-values measured close to the moderator surface at $d = 10$ cm would increase correspondingly in the middle of the moderator for the cases where B is essentially determined by thermal neutrons. B-values predominantly caused by high-energy neutrons do not show such a strong distance dependence. This is demonstrated for energetic neutrons with $E_n > 2$ MeV which induce fission in ^{232}Th: here the B(fission)-value decreases slowly with increasing distance d.

Experiments *inside the paraffin moderator* have also been performed with small ^{139}La-targets, similar to the ones described above. The results for observed $B(^{140}\text{La})$ values measured inside the moderator are given in [14]. Radiochemical results are more accurate; however, the SSNTD data give additional information on the effect of thermal neutrons in comparison to energetic ($E_n > 2$ MeV) neutrons.

Dwivedi *et al.* measured the fluence of very high energy neutrons ($E_n > 30$ MeV) using fission reactions in gold [6].

Table 7. Number of neutrons per proton inside the paraffin moderator at a distance d from the central axis. Uncertainties are about ±20%. For details see text.

Reaction	$d = 4$ cm	$d = 5.5$ cm	$d = 7$ cm	$d = 8.5$ cm	$d = 10$ cm
0.53 GeV p + U 235-U target	2	4.8	4.1	5.4	1.4 theor: 1.4–2.4
0.53 GeV p + U 232-Th target	2.3	–	0.5	–	0.3
1.0 GeV p + Pb 235-U target	14	19	23	19	8.6 theor: 2.75–4.3
1.0 GeV p + Pb 232-Th target	9	–	2.3	–	1.3

Notes: The distances d are measured radially from the axis of the target system: $d = 4$ cm is between the metallic target and the moderator, $d = 10$ cm is at the surface of the moderator. The SSNTDs were placed 10 cm downstream at the longitudinal middle of the target set-up and they had an active surface of about 1 cm^2. Theoretical estimates at $d = 10$ cm are from [17], giving the range of values from two models used for low energy neutrons with $E_n < 1$ eV.

Table 8. Measurement of the high energy neutron component (E_n > 30 MeV) with SSNTD using "gold on mica" as the target system.

Reaction	T [tracks/cm^2]	W [10^{-3} g Au/cm^2]	$T/\Phi \cdot W$ [g^{-1}]
0.53 GeV p + U/Pb	2.42×10^4	1.56	1.3×10^{-9}
1.0 GeV p + U/Pb	3.50×10^4	1.62	1.8×10^{-9}
1.5 GeV p + U/Pb	4.20×10^4	1.57	1.9×10^{-9}
1.5 GeV p + U/Pb [6]	0.53×10^4 [Ref. 6]	1.46 [Ref. 6]	2.4×10^{-9} [Ref. 6]

Notes: "T" is the track density in [cm^{-2}], "W" is the target thickness of gold sample in (10^{-3} g Au/cm^2), Φ is the total proton fluence obtained during the entire proton irradiation and given in Table 1. The product $T/\Phi \cdot W$ is a measure of the "effective fission rate", its uncertainty is about ±20%.

The number of fission fragments was measured as tracks in mica-detectors. A SSNTD target was placed between the metallic target and the moderator, approximately in the middle of the cylinder during the entire proton irradiation. Details of the technique have been described elsewhere [6]. The results are shown in Table 8; the agreement between the latest (1.5 GeV p + U/Pb) experiment and a previous irradiation at the same energy is good [6]. The "effective fission rate" as defined in Table 8 decreases slightly with proton energy. SSNTD experiments here yield essential auxiliary information and they agree with the respective model estimations: about 1.1 very high energy ($E_n > 30$ MeV) neutron is produced per proton at 1.5 GeV energy [6].

Guo *et al.* [23] measured the fluence of energetic neutrons ($E_n > 10$ MeV) using 20 cm long, 1 cm wide and 1 mm thick CR39 slides, placed along the metallic target surface and parallel to the axis. In this way, they obtained additional evidence for the proton energy. They could show in this experiment, that the decrease of track density along the beam direction is much more rapid for 0.53 GeV protons, as compared to 1.0 GeV and higher proton energies, as shown in Fig. 9 [23].

Zhuk *et al.* [24] carried out experiments with sandwiches containing artificial mica or Lavsan as SSNTDs and thick target foils with U^{natural}, U-6.5% (6.5% enrichment of ^{235}U) and ^{232}Th as sensors, thus allowing the determination of the fission rates for ^{235}U and ^{232}Th as well as the spectral index $\overline{\sigma}_f^{232Th}/\overline{\sigma}_f^{235U}$. The fission process of ^{235}U is initiated by neutrons of all energies (however, mainly thermal neutrons in our setups with paraffin moderator) and the fission process in ^{232}Th is sensitive only to fast neutrons because the fission threshold of ^{232}Th is 1.5 MeV. Seven sandwiches were placed along the upper moderator surface of the cylinder for the Pb- and U/Pb-targets. These SSNTD systems were exposed to 7 to 10 beam pulses from the Synchrophasotron with a total fluence of approximately 10^{12} protons. The track density T [in tracks per cm^2] on the surface of SSNTDs for U-6.5%, ^{nat}U and ^{232}Th sensors was 10^6, 10^5 and 10^4, respectively. Additional SSNTDs with ^{232}Th sensors were irradiated at the same positions during the entire proton irradiation and used to link with the full proton fluence determined by radiochemical methods. The fission rates of ^{235}U and ^{232}Th and spectral indices $\overline{\sigma}_f^{232Th}/\overline{\sigma}_f^{235U}$ at the seven positions placed along upper surface of the cylinder for the Pb-and U/Pb-targets were determined. Details of this analysis are given in Refs. [6, 24]. Spectral characteristics of the neutron flux along upper moderator 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 proton energy of 1.0 GeV are given in Table 9 and in Fig. 10.

The neutron spectrum of the U/Pb-target is softer as compared to the Pb-target: the ratio $\overline{\sigma}_f^{232Th}/\overline{\sigma}_f^{235U}$ in the target center is larger by a factor of approximately 2 for Pb-targets as compared to U/Pb-targets. At the ends of the paraffin moderator, the neutron spectrum becomes more energetic ("hard") for both setups. The spectral index $\overline{\sigma}_f^{232Th}/\overline{\sigma}_f^{235U}$ is larger at the beginning and at the end of the surface of the paraffin moderator by a factor of about 2.4 and 8, respectively, as compared to the central part. It should be noted that in the central region over more than 15 cm length there is a rather constant neutron-energy spectrum on the surface of the paraffin, where the value

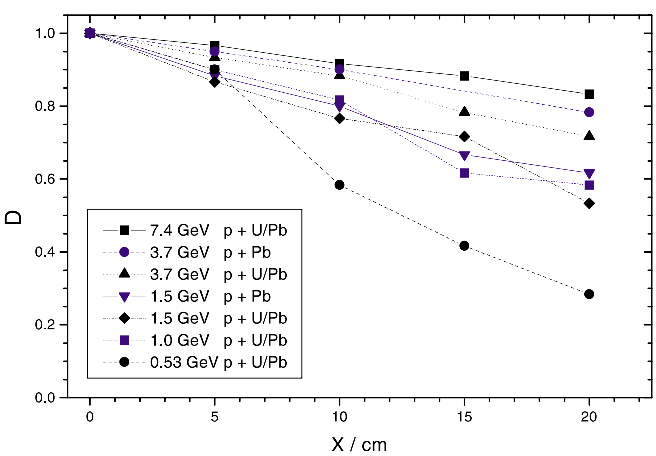

Fig. 9. Normalized track yields D [tracks per cm^2] at different positions of the inner CR39 detectors placed in contact with the Pb-target for different proton energies. The measurement is carried out along the central axis [X] of the metallic target. For details see text and [23].

Table 9. Distribution of the spectral index $\overline{\sigma}_f^{232Th}/\overline{\sigma}_f^{235U}$ along the top of the cylinder surface for U/Pb- and Pb-targets with paraffin moderator for 1.0 GeV protons.

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

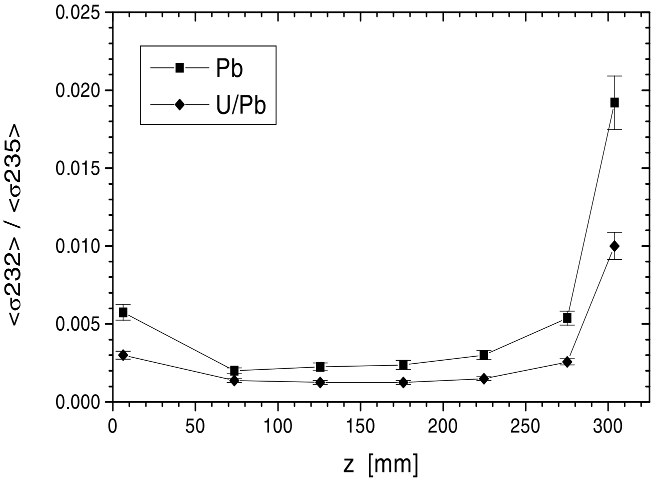

Fig. 10. Distributions of spectral index $\overline{\sigma}_f^{232\text{Th}}/\overline{\sigma}_f^{235\text{U}}$ along the upper moderator surface of U/Pb- and Pb- targets for 1.0 GeV protons.

$\overline{\sigma}_f^{232\text{Th}}/\overline{\sigma}_f^{235\text{U}}$ remains constant within the limits of experimental uncertainty.

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

Here we give a survey on our studies of the transmutation of long-lived radioactive waste and/or transuranium nuclides such as ^{129}I, ^{237}Np and ^{239}Pu. The target systems used are shown in the preceding sections. Radioactive samples were placed on top of the moderator, as shown in Fig. 11 a. The highly radioactive samples of ^{129}I, ^{237}Np, and ^{239}Pu were produced especially for these transmutation studies by the Institute of Physics and Power Engineering in Obninsk/Russia. Sample weights were 0.43 g for ^{129}I, 0.74 g for ^{237}Np and 0.45 g for ^{239}Pu. The target construction is shown in Fig. 11 b and the samples were placed on the outer surface of the paraffin moderator. We cannot quantify the influence of neutrons reflected from surrounding material, however, one must expect some influence of these neutron fluences, in particular when one takes into account the high cross-sections for neutron induced reactions with transuranium nuclides. The irradiations and the data analysis were carried out the same way as already described. The results for transmutation B-values of the three long-lived radwaste nuclides as well as the corresponding B/E_p values are shown in Table 10.

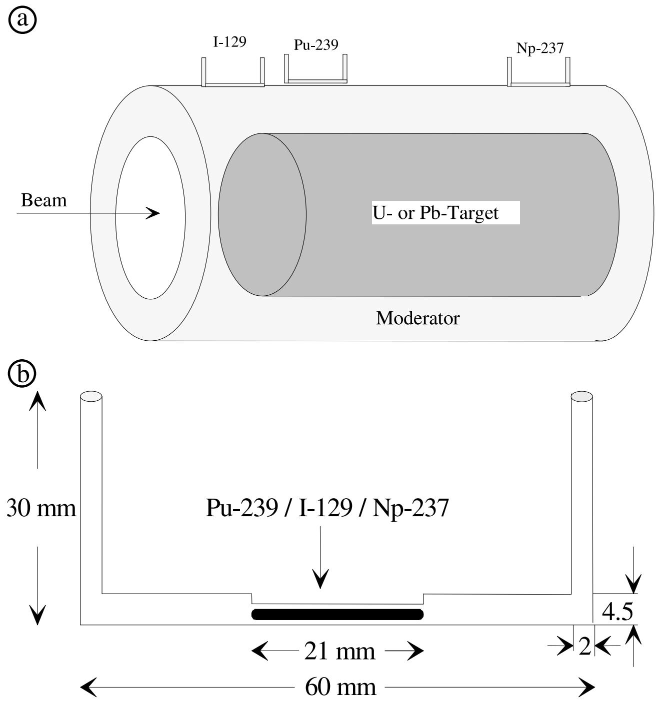

Fig. 11. (**a**) Experimental setup for transmutation studies of ^{239}Pu, ^{129}I and ^{237}Np samples. (**b**) Transmutation samples of ^{239}Pu, ^{129}I and ^{237}Np are weld-sealed in Al-capsules.

For transmutation of ^{239}Pu some additional remarks are necessary: 11 fission fragments were identified in the analysis of the gamma-ray spectra: ^{91}Sr, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{129}Sb, ^{132}Te, ^{133}I, ^{135}I, and ^{143}Ce. Knowing the relative fission yields for thermal/epithermal neutron induced fission of ^{239}Pu it is possible to extract $B(^{239}\text{Pu})$ which may be called the "transmutation rate *via* fission" [15].

From Table 10 one can conclude:

- As it was shown in Table 2 and Fig. 6, the $R(^{140}\text{La}) = B(^{140}\text{La})/E_p$ and $R(^{239}\text{Np}) = B(^{239}\text{Np})/E_p$ values are nearly constant within the experimental uncertainty of $\pm 15\%$ in the energy interval $0.5\,\text{GeV} \le E_p \le 7.4\,\text{GeV}$.

Table 10. Experimental transmutation yields B and B/E_p for three long-lived radwaste nuclides under study. Uncertainties are about ±25%.

Proton $E+$ target	$B(^{129}\text{I})$ $[10^{-4}\,\text{g}^{-1}]$	$B(^{129}\text{I})/E_p$ $[10^{-4}\,\text{g}^{-1}\,\text{GeV}^{-1}]$	$B(^{237}\text{Np})$ $[10^{-4}\,\text{g}^{-1}]$	$B(^{237}\text{Np})/E_p$ $[10^{-4}\,\text{g}^{-1}\,\text{GeV}^{-1}]$	$B(^{239}\text{Pu})$ $[10^{-4}\,\text{g}^{-1}]$	$B(^{239}\text{Pu})/E_p$ $[10^{-4}\,\text{g}^{-1}\,\text{GeV}^{-1}]$
0.5 GeV + U	1.30	2.60	3.1	6.2	–	–
1.0 GeV + U	2.61	2.61	4.8	4.8	–	–
1.5 GeV + U	2.33	1.55	8.2	5.4	–	–
7.4 GeV + U	15.8	2.14	50.4	6.8	–	–
0.53 GeV + Pb	–	–	–	–	21	39
1.0 GeV + Pb	0.81	0.81	3.3	3.3	31	31
1.5 GeV + Pb	0.90	0.60	7.5	5.0	–	–
3.7 GeV + Pb	3.0	0.81	30	8.1	–	–
7.4 GeV + Pb	3.9	0.53	34	4.6	–	–

Notes: The experiment (1 GeV p + U/Pb) was carried out twice. The experiments at (0.53 GeV p + Pb), (0.5 GeV p + U) and (1 GeV p + Pb) are from this work, the other results are from [6, 8].

These measurements were carried out in well-shielded positions *inside* 1 cm deep holes on the surface of the paraffin moderator. The *B*-values for long-lived radwaste nuclides were measured under slightly different geometrical conditions. The samples (Fig. 11 b) were placed on the upper surface of the moderator. An exact determination of the uncertainty of these *B*-values was not intended in this experiment, however, we estimate the experimental uncertainty to be approximately ±25%.

- The neutron spectra at the positions of ^{129}I, ^{237}Np, and ^{239}Pu on top of the moderator are different at various geometrical positions along the axis of the target system, as has been shown by the Minsk-group [6, 24]. Neutron spectra become "harder" the further downstream along the target, as also shown in Fig. 10. This means that the ^{237}Np-target is exposed to a somewhat "harder" neutron spectrum as compared to the ^{129}I- and ^{239}Pu-target. Consequently, a direct and strict comparison of the *B*-values for the three isotopes studied is difficult and outside the scope of this paper. However, for illustrative purposes this comparison is shown in Table 11 together with some other *B*-values.

From Table 10 and [8] the macroscopic transmutation rates of long-lived radwaste nuclei using a Pb-target with paraffin moderator and irradiated with 10 mA proton beams of 1 GeV energy can be estimated. Covering the whole moderator surface of our small setup with radioactive target material as thin as it is in our samples, calculated transmutation rates are as follows:

85 g of ^{239}Pu are transmuted in one month,
1.4 g of ^{238}U are transmuted in one month,
9 g of ^{237}Np are transmuted in one month,
1.3 g ^{129}I are transmuted in one month.

Of course the target shown in Fig. 11 a cannot be irradiated with 10 mA protons of 1 GeV as it would immediately melt. This implies that our actual "transmutation rates" given for a 10 mA/1 GeV proton beam are indicative. It should also be noted that sample positions *inside* the 6 cm thick paraffin moderator yield considerably larger transmutation rates. This shows that our results have a high predictive value, and that, before such a transmutation process could become a technological reality, one needs considerably more research and development in this field. 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 (Table 11) also show satisfactory agreement, in particular when we consider that they employed a higher proton energy and a different Pb-target without any moderator which may result in a "harder" neutron spectrum [18–20]. The good agreement of the Pu-transmutation rates between the two different laboratories as compared to the not-so-good agreement between the corresponding ^{129}I transmutation rate is not surprising. The neutron energy spectra in the two different experimental setups are probably very different – and they have not been determined experimentally. We want to repeat the statement that most experiments to date are indicative only. However, it is clear that before such transmutation processes can become a technological reality on large scale, considerably more advanced and detailed experimental investigations are mandatory.

Table 11. *B*-values for various sensors obtained on the outer mantle of the Pb-target and irradiated with 1 GeV protons. The uncertainties are about ±25%. 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* [10^{-5} g^{-1}]
^{239}Pu → fission (destruction of Pu)	310
^{238}U → ^{239}Np (breeding of Pu)	5.5
^{237}Np → ^{238}Np (destruction of Np)	33
^{139}La → ^{140}La (monitor reaction)	11
^{129}I → ^{130}I (destruction of I)	8.1
^{239}Pu → fission (CERN) (with SSNTD, µg Pu)	500 (for 2.6 GeV protons)
^{129}I → ^{130}I (CERN) (radiochemistry, 64.7 mg ^{129}I)	50 (for 2.6 GeV protons)

7. Conclusions

The present work is 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. The major emphasis lies on new experiments with 0.5 and 1.0 GeV protons which yield the following results:

- A new beam monitoring device based on nuclear chemical methods was tested and employed successfully. Two stacks of monitor foils were used, one in contact with the massive metallic target, the other one placed 35 cm upstream in 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 linearly with energy on the surface of the relatively small target systems employed. This holds for the entire proton energy range studied (0.5 GeV $\leq E_p \leq$ 7.4 GeV). The finding can be interpreted 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 to 1 g of ^{129}I, ^{237}Np, and ^{239}Pu, we could measure for the first time transmutation yields in a target system irradiated with high-energy protons. The results confirm theoretical estimates that transmutation can become practically feasible when high-intensity ($\gg$ 100 µA) accelerators for high-energy protons ($\geq$ 0.5 GeV) are technically available.
- Solid State Nuclear Track Detectors (SSNTDs) yield valuable auxiliary information; *e.g.* the amount of thermal neutrons on the surface of the target system and details on the finding that inside the 6 cm thick paraffin moderator the neutron fluences are several times larger than on the moderator surface.

All results show, that accelerator-driven systems (ADS) can be used in principle for large scale transmutation. However, one will need considerable more "Research and Development" before ADS can become a reality.

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References

1. Carminata, F., Klapisch, R., Revol, J. P., Roche, C., Rubio, J. A., Rubbia, C.: An energy amplifier for cleaner and inexhaustible nuclear energy production driven by a particle beam accelerator. CERN, Geneva, print CERN/AT/93-47 (ET), November 1 (1993).
2. Tolstov, K. D.: Some aspects of accelerator breeding. JINR-preprint, 18-89-778 (1989), Dubna, Russia.
3. Bowman, C. D., *et al*.: Nuclear energy generation and waste transmutation using an accelerator-driven intense thermal neutron source. Nucl. Instrum. Methods A **320**, 336 (1992).
4. Krivopustov, M. I., *et al*.: First experiments on transmutation of iodine-129 and neptunium-237 using relativistic protons of 3.7 GeV. JINR-preprint E1-97-59 (1997), Dubna, Russia and J. Radioanal. Nucl. Chem. **222**, 267 (1997).
5. Wan, J.-S., *et al*.: Transmutation of radioactive waste by means of relativistic heavy ions. JINR-preprint E1-97-349 (1997), Dubna, Russia and Kerntechnik **63**, 167 (1998).
6. Wan, J.-S., *et al*.: Transmutation studies on ^{129}I and ^{237}Np using 1.5 GeV, 3.7 GeV and 7.4 GeV protons. JINR-preprint E1-99-1 (1999), Dubna, Russia and "Special Issue on Accelerator Driven systems" (Plendl, H. S., ed.) Nucl. Instrum. Methods A **463**, 634 (2001).
7. Brandt, R., *et al*.: Transmutation studies using SSNTD and radiochemistry and the associated production of secondary neutrons. Radiat. Meas. **31**, 497 (1999).
8. Wan, J.-S.: Studies on spallation in massive targets and the transmutation of ^{129}I and ^{237}Np in a subcritical system. Ph.D Thesis, Department of Chemistry, Philipps University Marburg, Germany, published: ISBN 3-89703-310-0 (1999), Görich & Weiershäuser, Marburg (1999).
9. Andriamonje, S., *et al*.: Experimental determination of the energy generated in nuclear cascades by a high energy beam. Report, CERN/SPSLC95-17 (1995) and Phys. Lett. B **348**, 697 (1995).
10. Wan, J.-S., Ochs, M., Vater, P., Song, X. P., Langrock, E.-J., Brandt, R., Adam, J., Bamblevski, V. P., Kulakow, B. A., Krivopustov, M. I., Sosnin, A. N., Modolo, G., Odoj, R.: Monitor reactions in Al-foils for high energy proton beams bombarding a thick target. JINR-preprint E1-99-189 (1999), Dubna, Russia and Nucl. Instrum. Methods B **155**, 110 (1999).
11. Brandt, R., Carbonara, F., Cieslak, E., Piekarz, H., Piekarz, J.: Study of nuclear fission induced by high-energy protons. CERN-print 71-2 (1971), Geneva, Switzerland and Rev. Phys. Appl. **7**, 243 (1972).
12. Perelygin, V. P., Kulakow, B. A., Krivopustov, M. I., Petrova, R. I., Abdullaev, I. G., Bradnova, V., Knjazeva, G. P., Brandt, R., Ochs, M., Wan, J.-S., Vater, P.: SSNTD Studies of lead nuclei fission induced by relativistic *p*, *d*, He and ^{12}C projectiles inside massive Pb and U targets. Radiat. Meas. **31**, 567 (1999).
13. Westmeier, W.: *Benutzerhandbuch des Gamma-Spektrenanalyse-programmes GAMMAW Version 10.32*. Gesellschaft für Kernspektrometrie mbH, 35085-Ebsdorfergrund-Mölln, Germany.
14. Adam, J., *et al*.: Transmutation of ^{239}Pu and other nuclides using spallation neutrons produced by relativistic protons reacting with massive U- and Pb-targets. JINR-Preprint E1-2001, 136 (2001), Dubna, Russia
15. Wan, J.-S., *et al*.: First radiochemical studies on the transmutation of ^{239}Pu with spallation neutrons. JINR-preprint E1-2000-169 (2000), Dubna, Russia and J. Radioanal. Nucl. Chem. **247**, 151 (2001).
16. Hashemi-Nezhad, S. R., Brandt, R., Westmeier, W., Westmeier, H., Wan, J.-S., Vater, P.: Determination of slow neutron flux and spatial distribution using LR-115 2B detectors and comparison of the results with Monte Carlo simulations. Radiat. Meas. **34**, 319 (2001).
17. Hashemi-Nezhad, S. R., Brandt, R., Westmeier, W., Bamblevski, V. P., Krivopustov, M. I., Kulakov, B. A., Sosnin, A. N., Wan, J.-S., Odoj, R.: Monte Carlo analysis of accelerator-driven systems: Studies on spallation neutron yield and energy gain. Kerntechnik **66**, 47 (2001).
18. Abanades, A., *et al*.: Results from the TARC experiment: spallation neutron phenomenology in lead and neutron-driven nuclear transmutation by adiabatic resonance crossing. Nucl. Instrum. Methods A **478**, 577 (2002).
19. TARC-collaboration: Neutron-Driven Nuclear Transmutation by Adiabatic Resonance Crossing. Final Report to the European Commission, EUR 19117, 1999, Office for Official Publications of the European Communities, ISBN 92-828-7759-0.
20. Arnould, H., *et al*.: Experimental verification of neutron phenomenology in lead and transmutation by adiabatic resonance crossing in accelerator driven systems. Phys. Lett. B **458**, 167 (1999).
21. Adloff, J. C., Brandt, R., Butsev, V. S., Debeauvais, M., Fernandes, F., Kulakov, B. A., Krivopustov, M. I., Ochs, M., Sosnin, A. N., Zamani, M.: High flux neutron production from ^{12}C beams on heavy targets. Radiat. Meas. **28**, 313 (1997); Zamani-Valasiadou, M., *et al*., Radiat. Meas. **26**, 87 (1996).
22. Adloff, J. C., Brandt, R., Debeauvais, M., Fernandes, F., Krivopustov, M. I., Kulakov, B. A., Sosnin, A. N., Zamani-Valasiadou, M.: Secondary neutron production from thick Pb target by light particle irradiation. Radiat. Meas. **31**, 551 (1999).
23. Guo, S. L., Li, L., Shen, Q.-B., Shi, Y.-Q., Brandt, R., Vater, P., Wan, J. S., Ensinger, W., Kulakov, B. A., Krivopustov, M. I., Sosnin, A. N., Bradnova, V.: Measurements of neutron yields and spallation distributions in U/Pb, Pb and Hg thick targets bombarded by 0.5 and 1.0 GeV protons. Radiat. Meas. **34**, 301 (2001).
24. Zhuk, I. V., Lomonosova, E. M., Boulyga, S. F., Kievitskaia, A. I., Rakhno, I. L., Chigrinov, S. E., Bradnova, V., Krivopustov, M. I., Kulakov, B. A., Brandt, R., Ochs, M., Wan, J. S.: Determination of spatial and energy distributions of neutrons in experiments on transmutation of radioactive waste using relativistic protons. Radiat. Meas. **31**, 515 (1999).