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TRANSMUTATION OF RADIOACTIVE WASTE
WITH THE HELP OF RELATIVISTIC HEAVY IONS

Submitted to «Kerntechnik»

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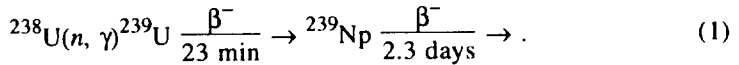
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1. INTRODUCTION

This contribution is the continuation of studies investigating extended targets (> 10 cm in length) irradiated with relativistic heavy ions. In the original work we reported on «enhanced nuclear cross sections» for secondary fragments produced in the interaction of 44 GeV ^{12}C and 72 GeV ^{40}Ar on various copper-block configurations [1–3]. Later on, a related enhancement was observed in the breeding rates induced by secondary neutrons originating from the interaction of 44 GeV ^{12}C with extended copper and lead targets [3–10]. As an example, such enhanced breeding rates could be observed for ^{239}Np , produced according to the reaction:



This enhancement was originally observed by Tolstov and coworkers [4] in the interaction of 44 GeV ^{12}C with large lead targets and interpreted by Bisplinghoff et al. [5]. An independent observation of enhanced neutron-production rates in 44 GeV ^{12}C interactions with extended Pb-targets has been published by Vassilkov et al. [6].

Earlier, we have carried out transmutation studies in extended copper and lead targets irradiated with relativistic ^{12}C -ions. We used radiochemical sensors, copper and lead targets as well as solid-state nuclear track detectors. This work has already been described [5, 7–10].

In this paper these experiments are extended and continued as follows:

1) The first direct experimental transmutation studies on long-lived radioactive waste-nuclei, such as ^{129}I ($T_{1/2} = 1.57 \cdot 10^7$ y) and ^{237}Np ($T_{1/2} = 2.14 \cdot 10^6$ y), using relativistic heavy ions are reported [13]. The suggestions of other scientists, such as Rubbia [11] and Bowman [12], followed. There is an apparent agreement among the participating scientists that mankind has two options handling radioactive waste (including plutonium):

a) One can bury the radioactive waste such as it is. Then one needs final depositories within geological formations stable for millions of years. Everybody is aware of the fact that such stability cannot be guaranteed with any rational degree of certainty.

b) One separates part of the radioactive waste such as actinides (Np, Pu, Am and Cm) plus a few long-lived fission fragments (^{129}I , ^{99}Tc) from the rest of the radioactive fission fragments by a process called partition. This is a series of chemical separation steps. The actinides and fission products (^{129}I , ^{99}Tc) are then transmuted in an accelerator-coupled nuclear assembly into stable and short-lived nuclei, needing only final depositories up to 600 years [11].

2) The efficiency of different targets for the production of secondary neutrons should be studied. The yield of secondary neutrons produced in a primary uranium target is compared to those produced in a primary lead target irradiated with relativistic ions such as 18 GeV ^{12}C -ions and 3.67 GeV protons.

3) The amounts of secondary neutrons produced in the interaction of 3.67 GeV protons in extended lead targets are studied in detailed experiments using a variety of techniques. Then experimental neutron yields are compared with theoretical estimations.

2. EXPERIMENTAL

The experiments are carried out at the Synchrophasotron, Laboratory of High Energies (LHE), Joint Institute for Nuclear Research (JINR), Dubna. Some details of the irradiation are given in Table 1. The different experiments are described consecutively.

Table 1. Experiments at the Synchrophasotron, LHE, JINR (Dubna)

No.	Experiment*	Irradiation time (min)	Flux**
1	3.67 GeV $^1\text{H} \rightarrow \text{I/Np}$	25	$1.25 \cdot E + 13$
2	3.67 GeV $^1\text{H} \rightarrow \text{Pb} + \text{I/Np}$	15	$1.24 \cdot E + 13$
3	18 GeV $^{12}\text{C} \rightarrow \text{Pb}$	416	$7.73 \cdot E + 11$
4	3.67 GeV $^1\text{H} \rightarrow \text{U/Pb}$	27	$1.24 \cdot E + 13$
5	18 GeV $^{12}\text{C} \rightarrow \text{U/Pb}$	472	$8.95 \cdot E + 11$

*The target setups are described later.

**For yield calculations the flux was determined by the monitor reaction: $^{27}\text{Al}(p, 3pn)^{24}\text{Na}$.

2.1. The Transmutation Experiments on ^{129}I and ^{237}Np

The first target used is shown in Fig.1a: a cylinder of 20 cm Pb, composed of 20 disks, each being 8 cm in diameter and 1 cm thick, is irradiated with a well-focused beam of 3.67 GeV protons during 15 minutes with a total intensity of $1.24 \cdot 10^{13}$ protons. The full width at half-maximum of the beam is about 22 mm. The Pb-cylinder is surrounded by a paraffin moderator 6 cm thick. On top of the moderator one places 0.425 g ^{129}I (in form of NaI) and 0.742 g ^{237}Np (in form of

Fig.1. The target setup for transmutation studies on ^{129}I - and ^{237}Np -samples using 3.67 GeV protons: *a*) the configuration using secondary neutrons; *b*) the direct irradiation using spallation reactions. (Details are given in [13])

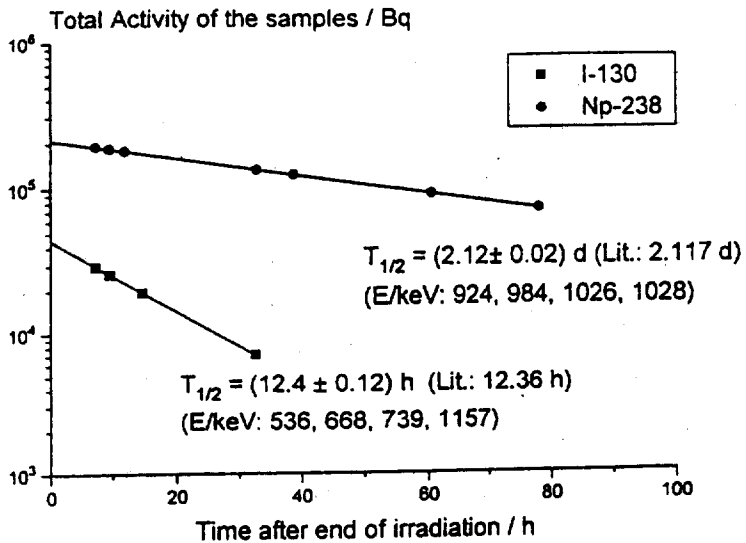
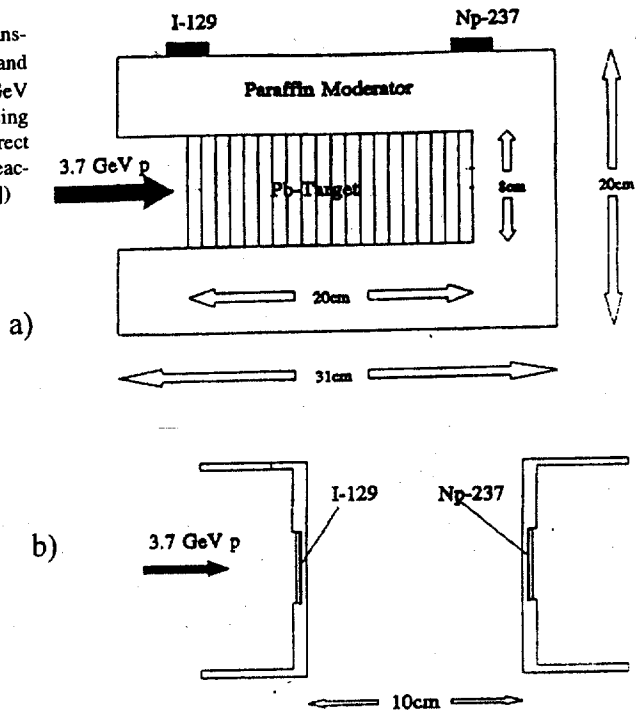


Fig.2. The decay curves of ^{130}I and ^{238}Np , formed in the reactions $^{129}\text{I}(n, \gamma)^{130}\text{I}$ and $^{237}\text{Np}(n, \gamma)^{238}\text{Np}$, using the experimental setup of Fig.1a

NpO₂). The isotopic composition of iodine is 15% ¹²⁷I and 85% ¹²⁹I. The iodine isotope samples were obtained from the Bochvar Institute in Moscow (VNIINM). The radioactive samples of I and Np are well sealed in Al-capsules, which is shown in Fig.1b. These were prepared by the Institute of Physics and Power Engineering (Obninsk, Russia). In addition to the ¹²⁹I- and ²³⁷Np-samples, shown in Fig.1a, two identical samples were irradiated directly in the 3.67 GeV proton beam with 1.25 · 10¹³ ions during a separate irradiation (Fig.1b). After the irradiation, the radioactive samples were placed in front of a HPGe-detector in order to study the gamma activity. As both ¹²⁹I- and ²³⁷Np-samples are fairly radioactive even without any activation, the samples were placed at a distance of 20 cm from the detector. For the ²³⁷Np-sample, a Pb-plate 1 cm thick is paced between the sample and the detector, reducing considerably the low energy gamma activity. We identified in the first target (Fig.1a) the (n, γ)-reaction products ¹³⁰I and ²³⁸Np as shown in the decay curves for these two nuclides in Fig.2. Further analysis of the gamma-spectra is state of the art as described in [2, 7, 8]. The results are expressed in terms of «breeding rates» *B*, defined as follows:

$$B = \frac{\text{number of produced particles}}{(\text{single incident ion}) \cdot (\text{1g sample})} \quad (2)$$

This term *B* is defined strictly in an empirical manner: this means that ¹²⁹I is placed at the geometrical positions given in Fig.1a. This target system is irradiated with 3.67 GeV protons. Part of this work has already been published [13].

The resulting *B*-values are shown in Table 2. In addition, one can compare the corresponding cross sections σ(*n*, γ) for the two reactions studied, using the following approximate equation (we assume that despite some differences in the respective excitation functions equation (3) gives sufficiently correct answers):

$$\frac{B_1}{N_T(1) \cdot \sigma_1(n, \gamma)} = \frac{B_2}{N_T(2) \cdot \sigma_2(n, \gamma)} \quad (3)$$

N_T being the number of target atoms per gram. All the targets are exposed in the same geometrical position and to the same neutron fluence. Assuming σ(*n*, γ) = 6.15 b for thermal neutrons in the reaction ¹²⁷I(*n*, γ)¹²⁸I, one obtains two further σ(*n*, γ)-values, as given in Table 2. The corresponding *B*-value for the reaction ¹²⁷I(*n*, γ)¹²⁸I has been reported in [8a].

Table 2. Observed breeding rates *B* and measured σ(*n*, γ)-values for some transmutation reactions. The *B*-value applies to the geometrical position of the targets shown in Fig. 1a

Reaction	²³⁷ Np(<i>n</i> , γ) ²³⁸ Np	¹²⁷ I(<i>n</i> , γ) ¹²⁸ I	¹²⁹ I(<i>n</i> , γ) ¹³⁰ I
<i>B</i> (10 ⁻⁴)	44 ± 4	1.9 ± 0.2	3.0 ± 0.4
(σ <i>n</i> , γ), b	140 ± 30	6.15 ± 1.23	10 ± 2

Finally we can answer to some extent the question: How long must one expose ^{129}I - or ^{237}Np -samples to neutrons before substantial amounts are transmuted into stable or short-lived nuclei? Taking the setup as shown in Fig. 1a one can estimate that a 10 mA accelerator of 3.67 GeV protons should transmute

- i) approximately 10% of ^{129}I per year,
- ii) approximately 30% of ^{237}Np per month.

Such a transmutation rate for ^{237}Np is large. According to the results of Bowman et al. [12], a flux of thermal neutrons of $1.0 \cdot 10^{15}$ neutrons/cm² · s can achieve such large transmutation rates. This flux is considerably larger than that in a conventional nuclear power station with approximately $3 \cdot 10^{13}$ neutrons (thermal)/cm² · s. (It is obvious that conventional power plants have not been able to transmute ^{237}Np to a substantial degree.) In this estimation we did not consider any technical problem, such as the heat-shielding of the target at 10 mA proton intensity or the stability of the NaI-compound in its containment during the irradiation. Both problems are nontrivial. In this paper we only wanted to give indications of the transmutation capacity of our target system.

In the direct irradiation of ^{129}I and ^{237}Np (Fig. 1b), one could observe a large variety of spallation products as shown in Fig. 3. The experimental results are

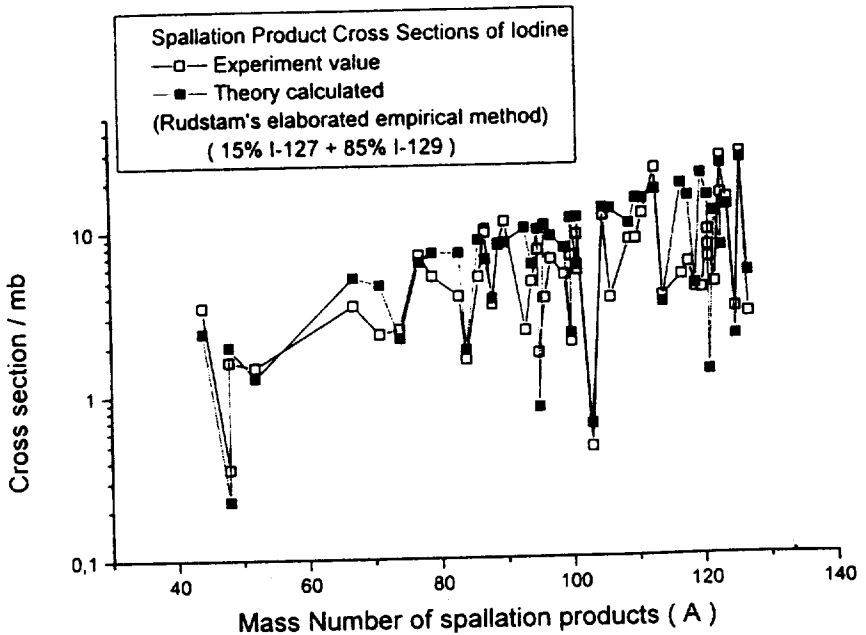


Fig.3. Spallation product cross sections for iodine irradiated with 3.67 GeV protons

compared with the results obtained by the semiempirical formula of Rudstam [14]. This pattern of observed cross sections for spallation products as a function of the mass of the spallation product fits well into the known spallation product behavior [15].

2.2. On the Comparison of Uranium Targets with Lead Targets Irradiated with Relativistic Ions

The target setups are shown in Fig.4. The Pb-target has been already used for I and Np (Fig.4a), and irradiated earlier with a variety of deuteron- alpha-, and ^{12}C -beams [8a, 10]. A new target system was also irradiated, containing two natural uranium rods 3.6 cm in diameter and 10.4 cm long encapsulated in thin Al-foils and placed into the center of a lead target 8 cm in diameter, and surrounded again by a 6 cm paraffin moderator (Fig.4b). Both target setups were irradiated with 3.67 GeV protons, as well as 18 GeV ^{12}C -ions. The irradiation of the pure Pb-target with 18 GeV ^{12}C is a reproduction of a series of preceding experiments [5,8,10]. The small uranium samples on top of the moderator are approx. 1 g depleted uranium oxide (0.42% ^{235}U). After irradiation, the small uranium samples were analyzed for their gamma activity, as already reported [10]. The observed B -values in reaction (1) leading to ^{237}Np are shown in Fig.5 for 3.67 GeV protons, as well as for 18 GeV ^{12}C irradiations. The results are well within expectations, as they are similar to the results of [8,10]: the maximum B -values are always about 5–10 cm downstream the entrance of the relativistic ion into the metallic target. Table 3 presents the average B -values on the surface of the moderator, observed for a variety of nuclear reactions. The results for $B(^{140}\text{La})$ were obtained with 1 g La-samples placed close to the U-samples on the surface of the paraffin moderator.

All the observed B -values fit well ($\pm 10\%$) the known systematics [10,17]. The B -values for the reaction 3.67 GeV $p + \text{Pb}$ are very close to those of the reaction 3.0 GeV $\text{D} + \text{Pb}$, as the total energies of the incoming ions are apparently very similar. The duplication experiment 18 GeV $^{12}\text{C} + \text{Pb}$ gives within their experimental uncertainties the same B -values as the preceding one, only $B(^{239}\text{Np})$ is about $45 \pm 15\%$ larger than reported earlier [10]. The increase of yields for a uranium target as compared to a lead target for low-energy neutron induced reactions is

$$\frac{(\text{yield with U-target})}{(\text{yield with Pb-target})} = 1.7 \pm 0.2, \quad (4)$$

a value similar to the one reported by Hilscher et al. [16] with 1.70 for relativistic protons.

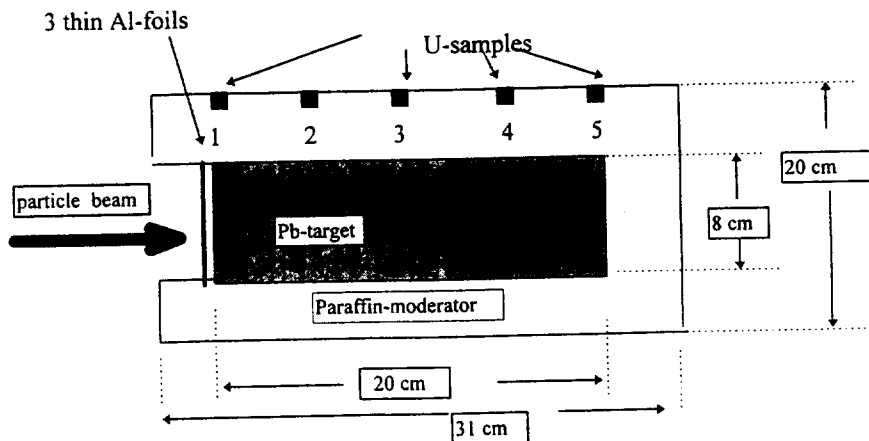


Fig.4a. Experimental setup for Pb-target. In addition to the U-samples shown, small La-samples were also placed on the top of the moderator. Details are given in [10]

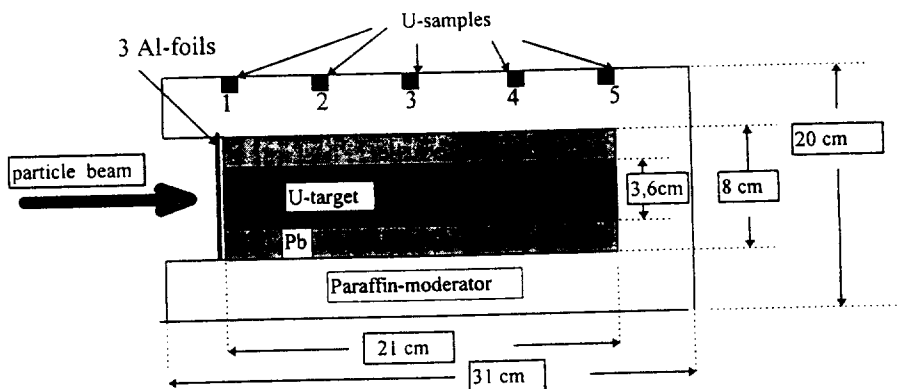


Fig.4b. Experimental setup for U(Pb)-target. In addition to the U-samples shown, small La-samples are also added, as remarked in Fig.4a

Finally, it is interesting to compare our $B(^{239}\text{Np}) = (5.7 \pm 0.5) \cdot 10^{-4}$ (Fig.5a), as observed 10 cm off axis and 10 cm downstream from the entrance of 3.67 GeV protons into the uranium target, with $B(U(n, f)) = (7 \pm 1) \cdot 10^{-4}$, as found by Andriamonje et al. [17] in about the same geometrical position for 2.75 GeV protons [2]. Details will be discussed later. This similarity is quite remarkable, as the two uranium target setups are different. Complete model calculations for U-

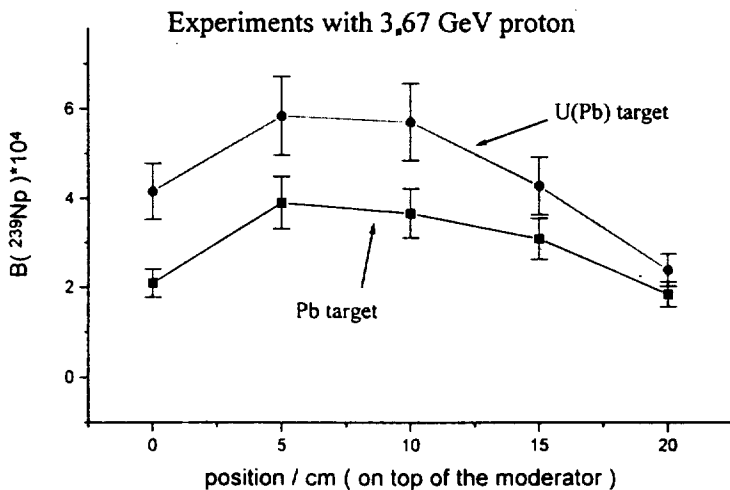


Fig.5a. $B(^{239}\text{Np})$ distribution in experiments with 3.67 GeV protons bombarding Pb- or U(Pb)-target, shown in Fig.4

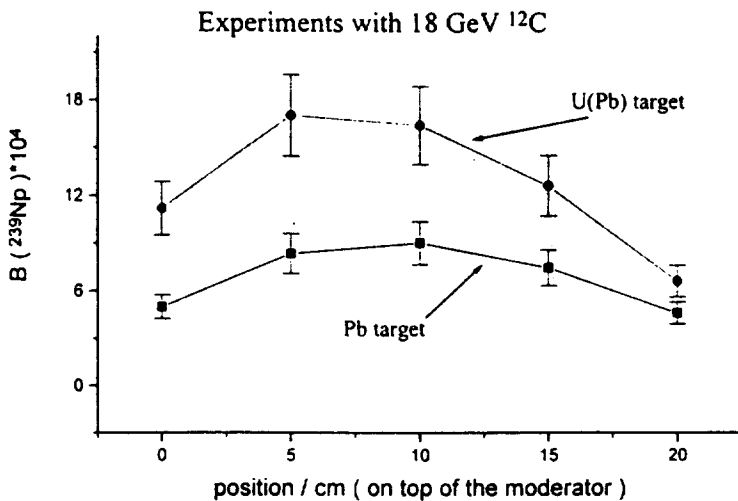


Fig.5b. $B(^{239}\text{Np})$ distribution in experiments with 18 GeV ^{12}C bombarding a Pb- or U(Pb)-target, shown in Fig.4

Table 3. Average B -values on the surface of the moderator (all uncertainties are $\pm 15\%$)

Reaction	3.67 GeV $p + \text{Pb}$	3.67 GeV $p + \text{U(Pb)**}$	3.67 GeV p : U/Pb***
$^{139}\text{La}(n, \gamma)^{140}\text{La}$	$6.04 \cdot E-4$	$10.5 \cdot E-4$	1.73
$^{238}\text{U}(n, \gamma) \dots ^{239}\text{Np}$	$2.93 \cdot E-4$	$4.48 \cdot E-4$	1.52
$^{238}\text{U}(n, 2n)^{237}\text{U}$	$9.64 \cdot E-6$	$11.5 \cdot E-6$	1.19
$^{235}\text{U}(n, f)^{91}\text{Sr}^*$	$9.69 \cdot E-5$	$14.3 \cdot E-6$	1.47
	18 GeV $^{12}\text{C} + \text{Pb}$	18 GeV $^{12}\text{C} + \text{U(Pb)}$	18 GeV ^{12}C : U/Pb
$^{139}\text{La}(n, \gamma)^{140}\text{La}$	$1.24 \cdot E-3$	$2.48 \cdot E-3$	2.00
$^{238}\text{U}(n, \gamma) \dots ^{239}\text{Np}$	$6.88 \cdot E-4$	$12.8 \cdot E-4$	1.86
$^{238}\text{U}(n, 2n)^{237}\text{U}$	$2.07 \cdot E-5$	$2.62 \cdot E-5$	1.26
$^{235}\text{U}(n, f)^{91}\text{Sr}^*$	$2.40 \cdot E-5$	$4.16 \cdot E-5$	1.73

*This applies to depleted uranium with 0.42% ^{235}U .

**The note U(Pb) stands for the uranium target shown in Fig.4b.

***U/Pb gives the ratio for uranium to lead target.

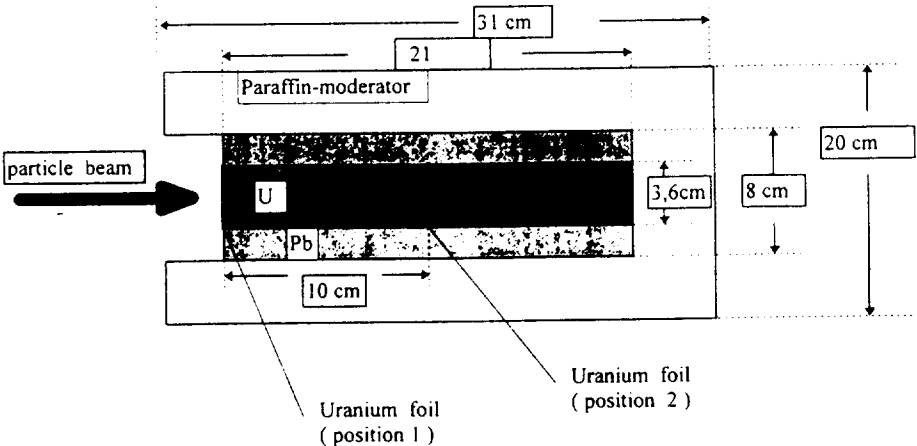


Fig. 6. Experimental setup for U-foils for IN-beam irradiations. (Foils are natural U, 50 μm thick, 3.6 cm in diameter)

target setups have not yet been carried out. However, we studied simultaneously also thin natural uranium foils of thickness 50 μm and 3.6 cm in diameter irradiated in front of and in the center of the uranium target, as shown in Fig.6. These thin uranium foils were suitable for the gamma-spectrum analysis after their irradiation. The observed B -values are given in Table 4.

As one would expect, this direct irradiation of uranium gives larger B -values than on the surface of the paraffin moderator (Table 5). The ratios given therein

Table 4. B -values for reaction products in natural uranium foils, irradiated IN-beam (Fig.6) (uncertainties are $\pm 15\%$)

Experiment Nuclide	3.67 GeV $p + U(Pb)$		18 GeV $^{12}C + U(Pb)$	
	position 1: 0 cm $\times 10^{-4}$	position 2: 10 cm $\times 10^{-4}$	position 1: 0 cm $\times 10^{-4}$	position 2: 10 cm $\times 10^{-4}$
^{91}Sr	0.789	1.40	1.62	3.14
^{97}Zr	0.916	1.48	1.82	3.42
^{132}Te	0.663	1.07	1.33	2.40
^{133}I	1.02	1.60	1.88	3.57
^{237}U	2.89	3.63	4.33	6.64
^{239}Np	13.2	21.7	25.8	52.1

Table 5. Ratio of B -values as observed in thin uranium foils IN-beam (Fig. 6) and compared to uranium samples exposed to secondary neutrons on the surface of the paraffin moderator (Fig. 4b) (uncertainties are $\pm 15\%$)

Experiment Nuclide	3.67 GeV $p + U(Pb)$		18 GeV $^{12}C + U(Pb)$	
	$\frac{B(\text{IN-beam pos. 1})}{B(\text{paraffin mod.})}$	$\frac{B(\text{IN-beam pos. 2})}{B(\text{paraffin mod.})}$	$\frac{B(\text{IN-beam pos. 1})}{B(\text{paraffin mod.})}$	$\frac{B(\text{IN-beam pos. 2})}{B(\text{paraffin mod.})}$
	^{239}Np	3.0	4.8	2.0
$^{91}Sr^*$	3.1	5.7	2.2	4.4
^{237}U	25	32	16	25

*Normalized to natural uranium with 0.72% ^{235}U .

range from 3 to 6 for low-energy neutron induced reactions and are around 25 for high-energy neutrons, as observed for $^{238}U(n, 2n)^{237}U$. This suggests a strong focusing for the energy deposition into a relatively small volume during these «electrical breeding» reactions. This may very well be one of the major technological challenges during the construction of large-scale subcritical reactors. The B -values for fission are a little larger IN-beam, as compared to B -values for ^{239}Np -production in the same geometrical position. As this is, to our knowledge, the first experimental description of several B -values in various geometrical positions in «electrical breeding» experiments the rates

$$R = \frac{B(^{239}Np)}{B(U(n, f))} \quad (5)$$

are given in Tables 6, 7.

As can be seen, the R -value are slightly lower with $R = 0.95 \pm 0.05$ for IN-beam positions (Fig.6) as compared to 10 cm off-axis positions (Fig.4) with $R = 1.10 \pm 0.10$ for uranium sensors. (The results are normalized to natural uranium with 0.72% ^{235}U .) This is interesting, since one has a considerable high-energy particle flux in the directly irradiated volume, which induces predomi-

Table 6. Ratios of $^{238}\text{U}(n, \gamma)$ to $^{235}\text{U}(n, f)$ for IN-beam natural U-foils (Fig. 6) (uncertainties are $\pm 15\%$)

Experiment		3.67 GeV $p + \text{U}(\text{Pb})$		18 GeV $^{12}\text{C} + \text{U}(\text{Pb})$	
Nuclide	Yield*	$R[(n, \gamma)/(n, f)]$ position 1 (0 cm)	$R[(n, \gamma)/(n, f)]$ position 2 (10 cm)	$R[(n, \gamma)/(n, f)]$ position 1 (0 cm)	$R[(n, \gamma)/(n, f)]$ position 2 (10 cm)
^{91}Sr	0.0588	0.98	0.91	0.98	0.98
^{97}Zr	0.0598	0.86	0.88	0.85	0.92
^{132}Te	0.0425	0.85	0.86	0.83	0.93
^{133}I	0.0661	0.85	0.90	0.91	0.95
Average R		0.90	0.94	0.93	1.00

*From NEA Data Bank 12, Yssy-Les-Moulineaux, France, 1994. Yield (fission product) is the fractional fission yield.

Table 7. Ratios of $^{238}\text{U}(n, \gamma)$ to $^{235}\text{U}(n, f)$ for U-samples on the surface of the moderator (Fig. 4) (uncertainties are $\pm 15\%$; depleted uranium with 0.42% ^{235}U was used)

Experiment		3.7 GeV $p + \text{Pb}$	3.7 GeV $p + \text{U}(\text{Pb})$	18 GeV $^{12}\text{C} + \text{Pb}$	18 GeV $^{12}\text{C} + \text{U}(\text{Pb})$
Nuclide	Yield	$R[(n, \gamma)/(n, f)]$	$R[(n, \gamma)/(n, f)]$	$R[(n, \gamma)/(n, f)]$	$R[(n, \gamma)/(n, f)]$
^{91}Sr	0.0588	1.79	1.85	1.69	1.82
^{97}Zr	0.0598	1.75	1.89	1.72	1.79
^{132}Te	0.0425	2.17	2.27	2.08	2.03
^{133}I	0.0661	1.64	1.79	1.64	1.69
Average R		1.82	1.92	1.75	1.82

nantly fission reactions in uranium. Nevertheless, the $B(^{239}\text{Np})$ -values are quite remarkable in all the geometrical positions studied. Such information is of some practical importance for proliferation considerations, further details are given in [18].

2.3. Neutron Fluences $\Phi(n)$ in the Pb-Target Irradiated with 3.67 GeV Protons: Measurements and Calculations

Finally, we are interested to estimate total neutron fluences $\Phi(n)$ on various surfaces of the Pb-target system irradiated with 3.67 GeV protons. This system is of the largest practical importance and the most complete sets of model calculations are available. We are aware of the fact that in this case we carry out a rather crude approximation on the determination of neutron numbers. In order to

estimate the secondary neutron fluence $\Phi(E)$ from empirical data, one has to use equation (2) and modify it according to elementary considerations:

$$B = \Phi(E) \cdot N_T \cdot \sigma(n, \gamma), \quad (5a)$$

where N_T is the number of target atoms in 1g of the target [1/g]; $\Phi(E)$ is the spectrum of neutrons per one incident 3.67 GeV proton;

$$\Phi(E) = B / (N_T \cdot \sigma(n, \gamma)) \text{ (neutrons per one 3.67 GeV proton per cm}^2\text{)}. \quad (5b)$$

This equation holds strictly only for a monoenergetic neutron fluence and gives the fluence $\Phi(E)$ only for the surface for which B has been determined experimentally.

In order to obtain a more realistic approximation, such as based on Eqs.5a,b for monoenergetic neutrons alone [10], the following must be considered carefully.

i) One calculates an energy-dependent neutron spectrum $\Phi(E)$ for the total surface of the paraffin moderator, O_S , such as estimated with the LAHET code for the reaction 3.67 GeV $p + \text{Pb}$ (Fig.7a). The LAHET code is a high-energy transport code using intranuclear cascades based on the Bertini model, followed by the pre-equilibrium model and then by the Rutherford-Cameron-Cook-Ignatyuk model, giving level densities. In addition, we used the DCM-CEM model developed in Dubna on rather similar principles. The DCM-CEM code has been used earlier [1,2]. Both codes (LAHET and DCM-CEM) give rather similar results within $\pm 10\%$ deviations.

ii) The «effective» cross section $\sigma_{\text{eff}}(n, \gamma)$ is defined in the conventional manner:

$$\sigma_{\text{eff}}(n, \gamma) = \frac{\int_{E_1}^{E_2} \Phi_{\text{the}}(E) \cdot \sigma_{n, \gamma}(E) \cdot dE}{\int_{E_1}^{E_2} \Phi_{\text{the}}(E) \cdot dE}. \quad (6)$$

This «effective» cross section is defined for an energy interval from E_1 (lower energy limit) to E_2 (upper energy limit). For this calculation one needs the excitation function $\sigma_{n, \gamma}(E)$. An example is given for the reaction $^{139}\text{La}(n, \gamma)^{140}\text{La}$ in Fig.7b.

iii) Using the known neutron energy spectrum $\Phi_{\text{the}}(E)$ and the «effective» cross section $\sigma_{\text{eff}}(n, \gamma)$, one can calculate a breeding rate B_{cal} as follows:

$$B_{\text{cal}} = N_T \cdot \sigma_{\text{eff}}(n, \gamma) \int_{E_1}^{E_2} \Phi_{\text{the}}(E) dE. \quad (7)$$

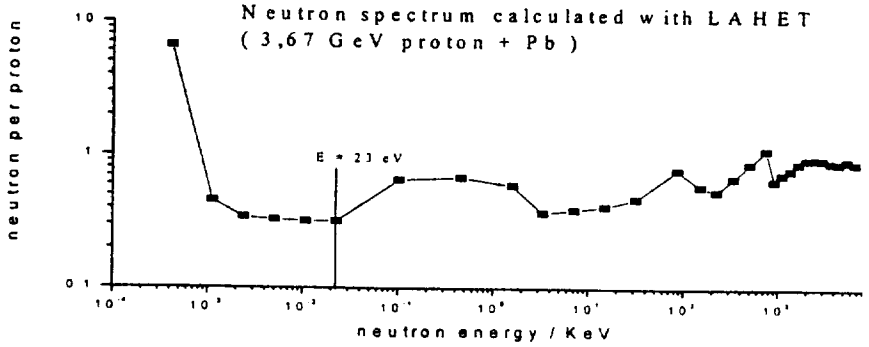


Fig.7a. The neutron spectrum calculated with LAHET for the surface of the moderator, 10 cm off center

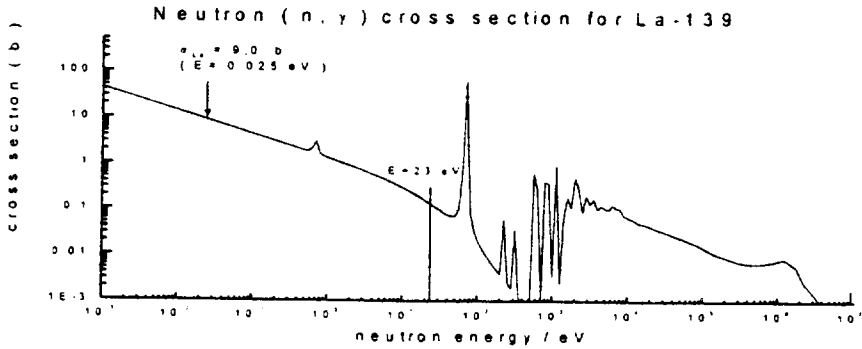


Fig.7b. Excitation function for the reaction $^{139}\text{La}(n, \gamma)^{140}\text{La}$

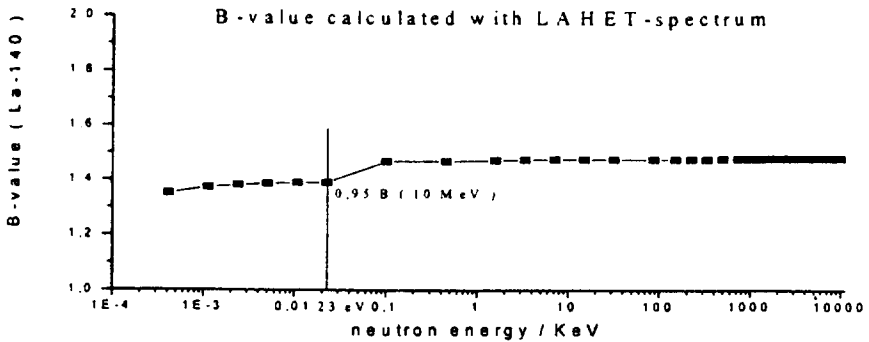


Fig.7c. The calculated breeding rate $B_{\text{cal}} (\cdot 10^{-4})$ for ^{140}La . The cut at 23 eV is explained in the text

As an example, B_{cal} is shown for the reaction $^{139}\text{La}(n, \gamma)^{140}\text{La}$, starting from $E_1 = 0$ eV up to $E_2 = E_{(n)}$, in Fig.7c. Its value is obviously increasing, but from a practical point-of-view is nearly constant at $E_{(n)} > 23$ eV, since neutrons with larger energies do not contribute to B_{cal} substantially. To find a practical cut in the number of neutrons contributing to B_{cal} , we choose arbitrarily a limit of $0.9 \cdot B_{\text{cal}}$ (10 MeV). The value is found at 10 MeV neutron energy. Only those neutrons below this limit are considered contributing to breeding rates B . This «cut» is also indicated in Figs.7a-c. The calculated B_{cal} (10 MeV) = $1.5 \cdot 10^{-4}$ is considerably smaller than the observed breeding rate $B_{\text{exp}} = (6.0 \pm 0.9) \cdot 10^{-4}$ (Table 3). To compare experimental observations with calculated values, we choose to present neutron fluences $\Phi_{\text{exp}}(E)$, or neutrons on the surface of the moderator (per an incoming 3.67 GeV proton), rather than B -values. For this, one must transform Eq. (7) as follows:

$$\Phi_{\text{exp}} = \frac{B_{\text{exp}} \cdot O_s}{N_T \cdot \sigma_{\text{eff}}(n, \gamma)} \quad (\text{per one 3.67 GeV proton}). \quad (8)$$

This experimental neutron number $\Phi_{\text{exp}}(E)$ (on the surface of the moderator, O_s) has been calculated for the same energy interval, for which $\sigma_{\text{eff}}(n, \gamma)$ has been determined, as shown graphically in Figs.7a-c. The fission reactions $\sigma(n, f)$ and the $(n, 2n)$ reactions have been determined in an analogous manner. The results are given in Table 8. As is seen, the experimental neutron fluence depends strongly on the technique employed:

i) At low energy ($E_{(n)} < 100$ eV) all fluences are in the range 17-39. (The exception of $B(n, \alpha) \rightarrow 128$ neutrons is ignored at present.) This large variation of $\Phi_{\text{exp}}(E)$ around 28 neutrons is not surprising, considering our quite approximate method of calculating $\Phi(E)$ from experimental breeding rates B_{exp} . Nevertheless, all empirical $\Phi_{\text{exp}}(E)$ -values are substantially larger than the calculated neutron fluences with $\Phi_{\text{cal}}(E) = 8 \pm 1$ low-energy neutrons. The reasons for this discrepancy of a factor of about 3 are not known with certainty [1,2].

ii) The excess of experimental neutron fluences over calculated ones is observed for all the sensors studied, up to about $E_{(n)} = 15$ MeV.

iii) Apparently, the neutron fluence increases quite rapidly inside the paraffin moderator. Using the solid-state nuclear track detector (SSNTD) technique, one observes 20 neutrons on the surface of the moderator, however, 120 neutrons only 3 cm inside the moderator. This effect has been observed earlier [19] and yet it

Table 8. Experimental and theoretical neutron fluences Φ on the mantel surface of the moderator (Fig. 4) for a Pb-target irradiated with 3.67 GeV proton (the LAHET code was employed)

Nuclear reaction	Energy range	$\sigma_{\text{eff}}(n, \gamma)$ (b)	d ¹⁾ (cm)	Φ_{exp} ²⁾	Φ_{the} ³⁾	method ⁴⁾
¹³⁹ La(n, γ) ¹⁴⁰ La	0–23 eV	7.2	9.4	39 ⁵⁾	8.3	R
²³⁸ U(n, γ)... ²³⁹ Np	0–100 eV	13.6	9.4	17	9.0	R
²³⁵ U(n, f), fragm.	0–1 eV	560	10.0	20	7	SSNTD
		560	7.0	120	–	SSNTD
^{nat} U(n, f)... ⁹¹ Sr	0–1 eV	0.25	9.4	30	7.1	R
^{nat} U(n, f)... ⁹⁷ Zr	0–1 eV	0.25	9.4	31	7.1	R
^{nat} U(n, f)... ¹³² Te	0–1 eV	0.18	9.4	25	7.1	R
¹²⁹ I(n, γ) ¹³⁰ I	0–26 eV	10 ⁷⁾	10.1	37 ⁶⁾	8.4	R
²³⁷ Np(n, γ) ²³⁸ Np	0–11 eV	173	10.1	29 ⁶⁾	8.1	R
$B(n, \alpha)$ recoil	thermal	–	10.1	128 ⁸⁾	7.3	SSNTD
CR-39 recoil p	0.3–3 MeV	–	10.1	16	12.5	SSNTD
²³⁸ U($n, 2n$) ²³⁷ U	8–15 MeV	1.0	9.4	7.3	3.2	R

¹⁾Perpendicular distance d from the beam center.

²⁾The uncertainty of the experimental neutron fluence is typically 15% based solely on the uncertainty in B_{exp} .

³⁾This neutron fluence was calculated with LAHET-code for the moderator surface at $d = 10$ cm.

⁴⁾R — radiochemical sensor, SSNTD — solid-state nuclear track detector.

⁵⁾The La-sensors had a larger volume than the U-sensors: 1.4 cm high in their vials as compared to 0.5 cm for U-sensors. The La-results were properly corrected (i.e. a 20% increase was applied).

⁶⁾Proper correction due to their geometrical position on top of the moderator were carried out (i.e. a 50% increase was applied).

⁷⁾The experimental value (Table 2) was used.

⁸⁾This neutron number was obtained by a different method: the standard technique of determination of neutron numbers using this SSNTD technique was employed.

should be studied further, possibly using radiochemical sensors, as shown in the Appendix.

Finally, it is interesting to compare the total neutron fluences Φ for the 3.67 GeV $p + \text{Pb}$ reaction with two different model calculations, as shown in Table 9. The agreement in the calculated total neutron fluences between the two model calculations is remarkable.

One should remember that the calculated fluences are not identical with the total number of neutrons emitted from the lead target into the moderator mantel. It is calculated with the DCM-CEM code that only 41 neutrons are generated in the lead target; due to scattering of neutrons between lead and paraffin the

Table 9. Neutron fluence Φ in a pure Pb-target (Fig.4a) as estimated using model calculations

Model	Neutron fluence Φ	
	on the outer mantel surface of the moderator, 10 cm off axis	on the mantel surface of the Pb-target, 4 cm off axis
LAHET	34.3 neutrons per 3.67 GeV proton ($E(n) < 3.67$ GeV)	105 neutrons per 3.67 GeV proton ($E(n) < 3.67$ GeV)
DCM-CEM	34.8 neutrons per 3.67 GeV proton ($E(n) < 10.5$ MeV)	

resulting effective fluence is 105 neutrons crossing the lead-paraffin boundary surface. Our sensors measure actually the resulting effective fluence. The observed large decrease of the neutron fluence by a calculated factor of 3 within the 6 cm paraffin is in approximate agreement with the Dubna experiments and comes, on the outer mantel surface of the moderator, close to 34 neutrons, as the reflection of neutrons from the surrounding air into the moderator can be neglected. This implies that approximately 17% of all the neutrons are absorbed in the moderator.

4. COMMENTS ON OTHER RELATED EXPERIMENTS

C.Rubbia's group of CERN carried out related experiments [17].

As has been mentioned in the preceding sections, the $B(^{239}\text{Np})$ values at a depth of 10 cm to the «center of cascade interactions» (the term defined in [17]) in this experiment (3.67 GeV $p + \text{U(Pb)}$) agree surprisingly well with $B(\text{U}(n, f))$ as found during the interaction 2.75 GeV $p + \text{U}$ [2]. This result is shown again in Fig.8 taken from [17] (Fig.6) and modified so as to include also the experimental results from this work. The decrease of $B(^{239}\text{Np})$ with the «corrected distance» is shown in Fig.8. (Details are given in [10].) The stronger decrease of B -values observed in this experiment as compared to C.Rubbia's experiment is a direct consequence of the lower total uranium content of our entire system ($3.4 \cdot 10^3$ g U as compared to CERN's $3.7 \cdot 10^6$ g U). The corresponding k -values are indicated (k is the neutron multiplication factor).

Then there could be a problem with neutron yields in both experiments:

i) As shown earlier, we are having difficulties to understand our results within well-accepted model calculations.

ii) The neutron balance of another experiment has been given by Calero et al. [20]:

1) Assuming a calculated «cost» for the generation of 1 neutron in a uranium target of 24+30 MeV, one can calculate that at most 41 neutrons are produced with one 1 GeV proton.

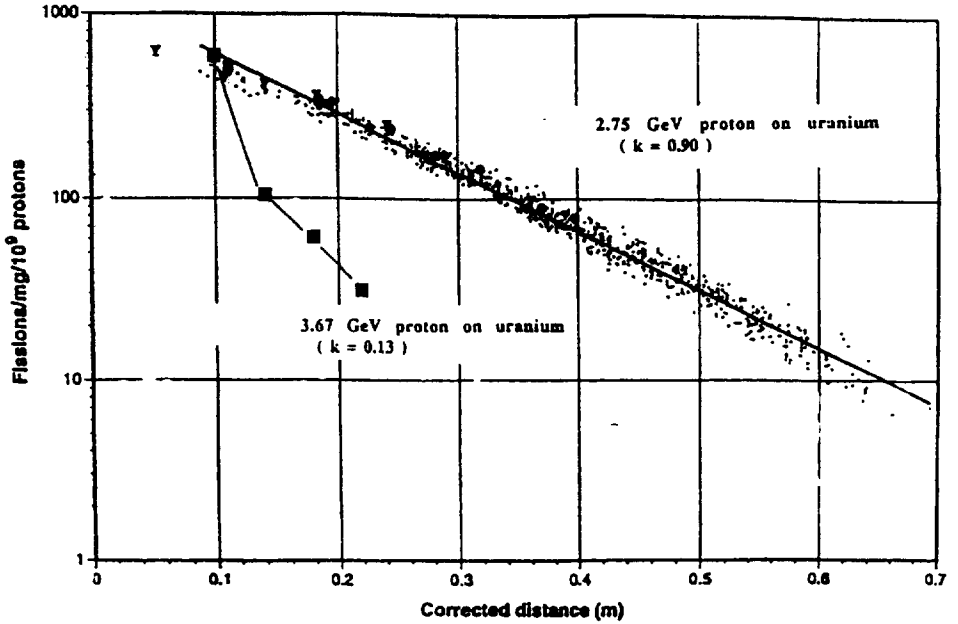


Fig.8. Reaction rates (i.e. fissions in [17] and (n, γ) reactions in this paper) for uranium targets as a function of the distance from the «center of cascade» (see the text). In [17] the solid/liquid target system extends to more than 0.7 m, in our experiment the paraffin moderator extends along the radius up to 0.1 m, afterwards one has open air. The original of this figure can be found in [17] (without our results)

2) Rubbia's experiment has a measured neutron multiplication factor $k = 0.90 \pm 0.01$, consequently the total number of neutrons per a 1 GeV proton is

$$N_{\text{tot}} = \frac{\leq 41}{1 - k} \leq 410 \text{ neutrons.}$$

3) The measured «energy amplification» is 30 ± 1 , consequently a 1 GeV proton generates 30 GeV «heat» through nuclear fission in this setup. As the energy released in fission is about 200 MeV, one needs

$$150 \text{ fissions} \rightarrow 150 \text{ neutrons}$$

in this case. Here the authors stop their evaluation [20].

4) We know from the present paper that $\sigma(n, f)$, as compared to $\sigma(n, \gamma)$ for natural uranium, is 10 cm off axis in our system (this holds for ^{238}U -targets alone):

$$\sigma(n, \gamma) = (1.1 \pm 0.1) \cdot \sigma(n, f) \rightarrow 165 \pm 15 \text{ neutrons.}$$

In addition, we have for ^{235}U an extra neutron absorption, $\sigma(n, \gamma) = 0.2\sigma(n, f)$, using again approximately 30 neutrons. This estimation is based on the conservative assumption that $\sigma_E(n, \gamma) = 0.2\sigma_E(n, f)$ for ^{235}U , independent of the neutron energy E . Altogether we have already used about 345 neutrons.

5) Only no more than 65 neutrons ($\leq 15\%$ of all the neutrons) are left for all the other reaction channels within a large subcritical assembly of dimensions of approximately 1m^3 , in particular for the neutron absorption in water. This relatively small loss of neutrons is not necessarily in contradiction to our observations that about 17% of all the neutrons are lost in 6 cm paraffin, as mentioned earlier.

iii) Last but not least, one may look at [17] from a more general point of view. Recently, several authors have shown that there are some operational limits for accelerator-driven subcritical nuclear systems [10,21]:

1) The input of the energy P_I into the subcritical system is given by the beam intensity I and the proton energy E_p :

$$P_I = I \cdot E_p. \quad (9)$$

2) The production of the energy P_F due to fission of uranium nuclei ($E_{\text{fiss}} \approx 200 \text{ MeV}$) inside the subcritical system is given by

$$P_F = \frac{E_{\text{fiss}} \cdot k_{\text{eff}} \cdot I \cdot n_{\text{sp}}}{(1 - k_{\text{eff}}) \cdot \nu}, \quad (10)$$

with k_{eff} being the «effective» neutron multiplication factor ($k_{\text{eff}} < 1$ in subcritical systems), $\nu = 2.5$, i.e. the neutron observed per fission event and n_{sp} , the average number of spallation neutrons produced within the entire target system per one incident proton.

3) Consequently, the «energy amplification» (EA) of the entire system can be calculated as follows:

$$\text{EA} \equiv \frac{P_F}{P_I} = \frac{n_{\text{sp}} \cdot k_{\text{eff}} \cdot E_{\text{fiss}}}{\nu \cdot (1 - k_{\text{eff}}) \cdot E_p}. \quad (11)$$

When a system with a given E_p has a measured value for EA and k_{eff} , then the value n_{sp} can be determined uniquely.

For the CERN experiment one finds experimentally [17]

$$\text{EA} = 30 \pm 1 \text{ for } 1 \text{ GeV} \leq E_p \leq 2.75 \text{ GeV},$$

$$k_{\text{eff}} = 0.90 \pm 0.01.$$

This determines for 1 GeV protons in a unique manner

$$n_{sp} = (41.7 \pm 1.3) \text{ neutrons/proton in uranium.} \quad (12)$$

This value agrees with the largest value for $n_{sp} \leq 41$, as suggested by Calero et al. [20]. (In this case, the cost for the production of 1 neutron is 24 MeV, as has already been mentioned.)

On the other hand, n_{sp} has been determined via electronic counter-experiments in lead targets:

$$n_{sp} = 17 + 24 \text{ neutrons/proton in lead for 1 GeV protons}$$

(Hilscher et al. [16]: 24 ± 1 neutrons/proton; Vassilkov et al. [6]: 21.5 ± 0.5 neutrons/proton; Zucker et al. [22]: 17.5 ± 0.3 neutrons/proton)). Replacing an extended lead target with an extended uranium target leads to an increase in secondary neutrons by 70%, as found by Hilscher (lit. cit.) and in this paper experimentally, yielding

$$n_{sp} = 29 + 41 \text{ neutrons/proton in uranium for 1 GeV protons.} \quad (13)$$

Comparing the two independent determinations of n_{sp} in uranium, one has only small problems using the experimental results of Hilscher [16] with $n_{sp} = 35.3$. However, there is a problem with the result of Zucker et al. [22] who measured $n_{sp} = 17.5 \pm 0.3$. Such a low value generates difficulties: a value of 17.5 neutrons/proton measured by Zucker for (1 GeV $p + \text{Pb}$) would lead to 30 neutrons/proton for (1 GeV $p + \text{U}$). This gives in total only 300 neutrons — not enough to use 150 neutrons for fission reactions and 195 neutrons for (n, γ) reactions in uranium.

A further difficulty is the observation by Zucker et al. [21] of a linear relation obtained experimentally between n_{sp} and E_p (Eq.11) in the proton energy interval from 0.2 to 1.4 GeV. This leads to a constant energy amplification EA — in contrast to the experimental observation [17], finding EA decreasing strongly below 1 GeV proton energies. This shows that we have not reached understanding of the neutronics in the subcritical systems under investigation. Further experimental and theoretical work is needed. It is interesting to note that Voronko et al. [23] have already suggested rather low energy costs for production of one neutron in an extended Pb-target: 32 ± 5 MeV leading to about 19 MeV/neutron in a large extended U-target.

5. CONCLUSIONS

The essential results of this study can be summarized as follows:

1) It is possible to transmute the long-lived fission product, ^{129}I , into stable ^{130}Xe via (n, γ) reactions using the «electrical breeding» method. However, a

10 mA accelerator of 3.7 GeV protons will transmute only 10% of this nuclide per year under the given experimental conditions. Higher proton intensities may be needed.

2) It is possible to transmute the long-lived minor actinide, ^{237}Np , into short-lived ^{238}Pu via (n, γ) reactions using a 10 mA accelerator of 3.7 GeV protons: approx. 30% of this nuclide are transmuted per month under the given geometrical conditions.

3) Comparing massive Pb-targets 20 cm long with massive uranium targets 21 cm long, one observes approx. 70% larger secondary neutron fluxes in uranium as compared to lead. This holds for 3.67 GeV protons, as well as for 18 GeV ^{12}C . The fission and (n, γ) transmutation rates within the directly irradiated uranium target are substantial and about a factor of 3 larger than in a paraffin moderator about 10 cm off axis.

4) In the estimations of total neutron fluences per 3.67 GeV protons using the Pb-target one has a problem: preliminary estimations show the observed neutron fluences are possibly up to a factor of 2–3 larger than the calculated neutron fluences. We used the LAHET and DCM-CEM codes.

5) A related and somewhat similar experiment [17] using 2.75 GeV protons on uranium gave rather similar results as compared to 3.67 GeV protons on uranium in this paper, at least in close proximity to the «central part of interactions» inside the extended uranium target. However, it was not possible to understand both experiments from our theoretical point of view completely.

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APPENDIX: Simulation experiments on the neutron moderation within the Pb-target used, employing a Pu-Be neutron source

The neutron moderation within the Pb-target used (Fig.4a) could be studied experimentally using a mock-up Pb-target together with a $1.4 \cdot 10^{11}$ Bq Pu/Be neutron source emitting $8.2 \cdot 10^6$ neutrons/s with an average energy of about 4 MeV. The Pb-target is shown in Fig.A-1. In this way, a direct calculation of the neutron moderation was obtained together with a direct measurement of the experimental *B*-value for La- and U-sensors for a given neutron source. The radiochemical sensors (La, U) are the same as those used in the accelerator exposures. The activation experiment lasted always about 24 hours. The data analysis has been the same as during the accelerator experiments.

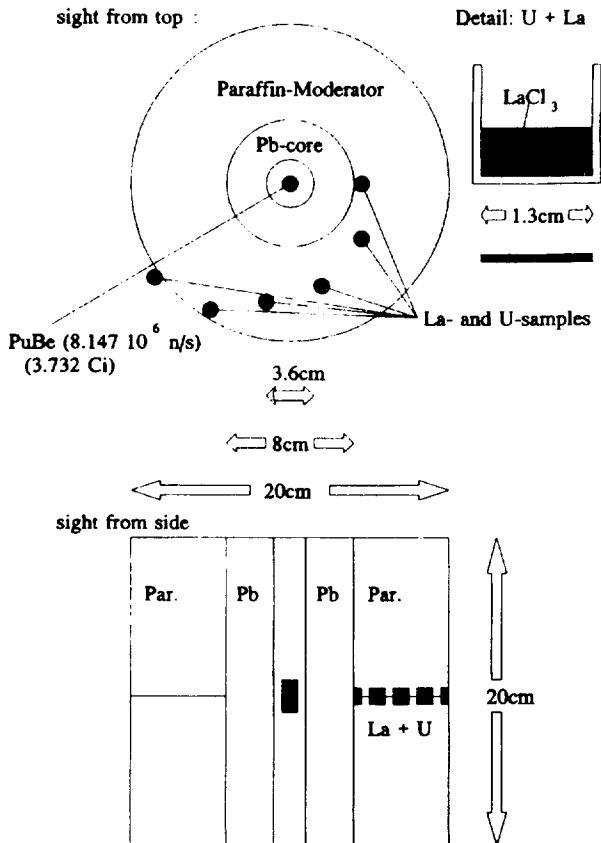


Fig.A-1. Setup for the neutron-calibration experiment

The results are given in Figs. A-2, A-3 and Table A-1. The experimental B -values decrease quite drastically within the 6 cm paraffin moderator. This result was also observed with solid-state nuclear track detectors (SSNTD) in a preliminary manner during earlier Dubna experiments [10]. Considering that neutrons are emitted spherically, one must multiply B -values by R^2 (R being the distance from the n -source to the sensor) in order to obtain a feeling how the total measured neutron fluence decreases with R . This result is also shown in Table A-1. The values $B \cdot R^2$ are fairly constant for $5 < R < 9$ cm, however they decrease rather drastically towards the edge of the paraffin moderator. As we present here only experimental results, we cannot determine to what extent «lack of reflection» of neutrons at the edge of the moderator and/or simple neutron absorption in hydrogen is responsible for this decrease. In any case, this neutron calibration experiment confirms

i) the strong decrease of the neutron fluence in 6 cm paraffin within our target system as calculated with the LAHET code (Table 9),

ii) the strong decrease of the experimental neutron fluence at $R = 7 + 10$ cm, as observed with SSNTD studying the fission fragment track density due to ^{235}U (n, f) fragment (Table 8).

Finally, during the Dubna experiments one could determine B -values for ^{140}La and ^{239}Np in sensors placed at $R = 9.4$ cm. For the reaction $3.67 \text{ GeV } p + \text{Pb}$ it was observed (Table 3) that

$$B(^{140}\text{La}) = (6.0 \pm 0.9) \cdot 10^{-4}, \quad (\text{A-1})$$

$$B(^{239}\text{Np}) = (2.9 \pm 0.5) \cdot 10^{-4}. \quad (\text{A-2})$$

It is evident from Table 8 that about 30 ± 10 thermal neutrons are inducing those two reactions. Table A-1 gives the effect of converting 1 neutron into ob-

Table A-1. Experimental results of the neutron calculation

Distance R^* (cm)	Medium	$B(^{140}\text{La})^{**}$		$B(^{239}\text{Np})^{**}$		$\frac{B(^{140}\text{La})}{B(^{239}\text{Np})}$
		exper. $\times 10^{-6}$	exper. $R^2 \times 10^{-4}$	exper. $\times 10^{-6}$	exper. $R^2 \times 10^{-4}$	
4.8	paraffin	108.5	25.0	42.1	9.7	2.58
5.8	—	104.0	35.0	34.1	11.5	3.05
7.3	—	79.7	42.5	26.8	14.3	2.97
8.3	—	60.2	41.5	20.4	14.1	2.95
9.3	—	20.4	17.6	10.6	9.2	1.92
10.8	air	6.5	7.8	—	—	

*Distance from center-axis to center of La- or U-sensor.

**All uncertainties are $\pm 13\%$.

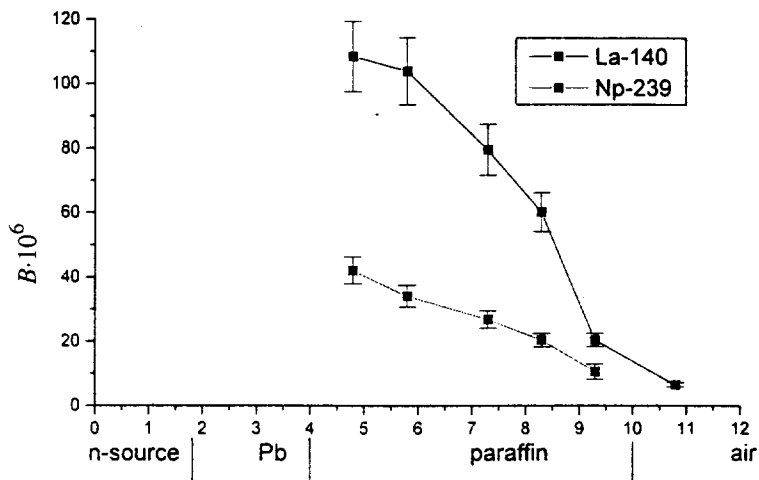


Fig.A-2. B -values for the neutron calibration experiment (abscissa in cm)

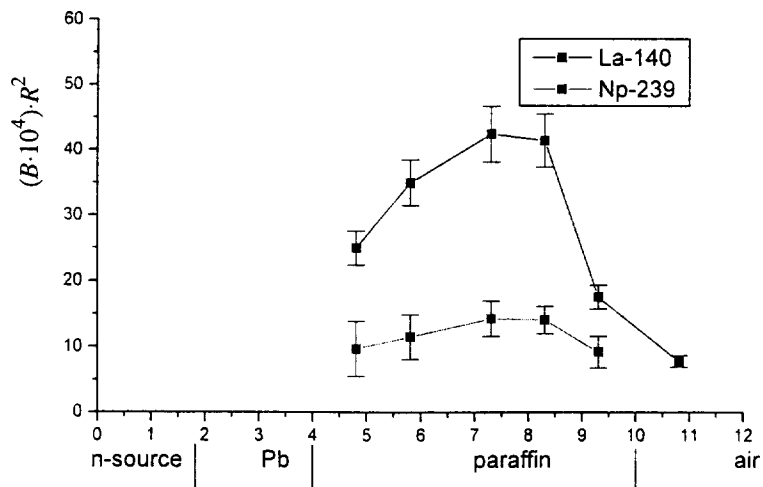


Fig.A-3. B -values for the neutron calibration experiment corrected for radius (abscissa in cm)

servable ^{140}La - or ^{239}Np - B -values: $20.4 \cdot 10^{-6}$ and $10.6 \cdot 10^{-6}$ respectively. This neutron calibration experiment gives accordingly

$$B(^{140}\text{La}) = (6 \pm 2) \cdot 10^{-4}, \quad (\text{A-3})$$

$$B(^{239}\text{Np}) = (3 \pm 1) \cdot 10^{-4}. \quad (\text{A-4})$$

The agreement between the two pairs of B -values indicates an internal consistency of our measurements.

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Трансмутация радиоактивных отходов на пучках релятивистских ядер

Выполнена серия экспериментов на синхрофазотроне Лаборатории высоких энергий ОИЯИ (Дубна) с использованием протонов с энергией 3,67 ГэВ и ядер ^{12}C с энергией 18 ГэВ. Облучены две массивные мишени из свинца и естественного урана, окруженные парафиновым замедлителем, на поверхности которого размещались La- и U-активационные детекторы и твердотельные ядерные трековые детекторы. Для свинцовой мишени обе методики независимо показали, что поверхность замедлителя покидают около 28 низкоэнергетических нейтронов на один протон. Теоретические оценки, проведенные на основе компьютерных программ ЛАНЕТ (Лос-Аламос) и DCM/CEM (Дубна), дают более низкие значения: 7–9 нейтронов с энергией менее 0,1 кэВ при общем значении 34–35 нейтронов. Подобная проблема замечена в аналогичных экспериментах в ЦЕРН. В ходе первых экспериментов на пучках ускорителей высоких энергий по трансмутации долгоживущих радиоактивных отходов ^{129}I ($T_{1/2} = 1,57 \cdot 10^7$ лет) и ^{237}Np ($T_{1/2} = 2,14 \cdot 10^6$ лет) по измерениям реакций радиационного захвата были определены сечения их трансмутации, которые составляют (10 ± 2) и (140 ± 30) б соответственно. Получены оценки скорости трансмутации этих радиоактивных отходов на ускорителе протонов с энергией 3,67 ГэВ.

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Transmutation of Radioactive Waste with the Help of Relativistic Heavy Ions

A series of experiments was carried out at the Synchrophasotron, LHE, JINR, Dubna, using 3.67 GeV protons and 18 GeV ^{12}C ion beams. Two massive lead and uranium targets surrounded by paraffin moderator were irradiated. The outer surface of the moderator was some small U- and La-sensors, to be studied by radiochemistry activation techniques, and also by solid-state nuclear track detectors. Both experimental techniques independently give approximately 28 low energy neutrons on the outer surface of the moderator per 3.67 GeV proton hitting the Pb-target. Theoretical estimations based on ЛАНЕТ and DCM/CEM computer codes give considerable smaller fluences: approximately 7–9 low energy neutrons (<0.1 keV) and 34–35 neutrons at all energies in the same geometrical positions. A similar problem seems to appear in analogous experiments at CERN. In addition, long-lived radioactive waste nuclides, such as ^{129}I ($T_{1/2} = 1.57 \cdot 10^7$ y) and ^{237}Np ($T_{1/2} = 2.14 \cdot 10^6$ y) were placed as well-sealed targets (approximately of 1 g each) into different geometrical positions during the 3.67 GeV proton irradiations. The short-lived transmutation products ^{130}I ($T_{1/2} = 12.4$ h) and ^{238}Np ($T_{1/2} = 2.4$ days), could be identified radiochemically as well as other spallation products. The transmutation rates are substantial: a 10 mA accelerator of 3.67 GeV protons could transmute at least 30% of ^{237}Np and 1% of ^{129}I per month under the given geometrical conditions.

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

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