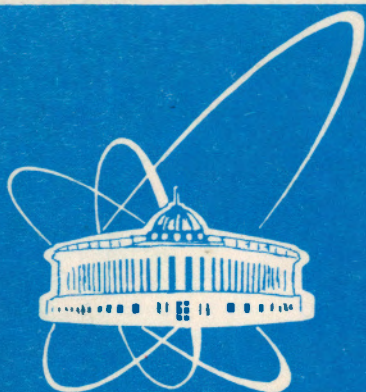


94-116

~~Бислингхофф~~



ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E1-94-116

Bisplinghoff, B.

ON NEUTRON GENERATION
IN MASSIVE Cu-TARGET AT IRRADIATION
WITH 22 AND 44 GeV CARBON IONS

Submitted to «J. of Radioanalytical and Nuclear Chemistry»

1994

B.Bisplinghoff¹, V.Bradnova², R.Brandt¹, K.K.Dwivedi³, V.S.Butsev^{1,2},
E.M.Friedlander^{1,4}, S.Gosh³, Guo Shi-Lun⁵, M.Heck^{1,6}, Jin Huimin^{1,7},
M.I.Krivopustov^{1,2}, B.A.Kulakov^{1,2}, C.Laue¹, L.Lerman^{1,4}, Th.Schmidt¹,
A.N.Sosnin², Wang Yu-Lan⁵

¹Kernchemie, F.B. 14, Philipps-Universität, Marburg, Germany

²Laboratory of High Energies, Joint Institute for Nuclear Research, Dubna, Russia

³Department of Chemistry, North-Eastern Hill University, Shillong, India

⁴Nuclear Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, CA, USA

⁵China Institute of Atomic Energy, Beijing, China

⁶Present address: Gesellschaft für Nuclear Service m.b.H., GNS, Essen, Germany

⁷Visiting scientist from Section of Technical Research for Nuclear Safeguards, Department of Radiochemistry, China Institute of Atomic Energy, Beijing, China

INTRODUCTION

For the last half century there has been deep and practical interest in using accelerators for the production of secondary neutrons. These neutrons can induce all kinds of activation processes. Nearly two decades ago, extended studies were carried out in North America using high-energy proton beams ($E \sim 1$ GeV) for what is called «Electrical- or Accelerator-Breeding» [1]. Afterwards, Vassilkov, Goldanski and Orlov published a very systematic theoretical paper [2]. Furthermore, Bakhmutin et al. [3] reported on experimental results obtained with an extended iron-target irradiated with high-energy protons. In 1991, with increasing interest in the problem of nuclear transmutation of long-lived radioactive wastes from nuclear power reactors, a workshop was held in Obninsk (Russia) [4], and K.D. Tolstov suggested a very interesting idea to introduce a relativistic heavy ion accelerator into the cycle of nuclear power generation for many purposes, some of them are [5]:

1. Generation of electric power in a subcritical nuclear assembly coupled to a relativistic heavy ion accelerator. Such a nuclear power station cannot go beyond control due to an uncontrolled increase in energy production. Power output could be over a range of (10—500) MW (electr.), depending on the safety margin the system operates below a «neutron multiplication factor» k (effective) of ≤ 1.000 .

2. Transmutation of long-lived radioactive wastes. In this case, considerable research and development will be needed before sufficiently large neutron fluxes can be obtained as will be discussed below.

In addition, Tolstov and coworkers [6] carried out some pilot experiments as follows: As shown in Fig. 1a, a rather massive Pb-target $0.5 \cdot 0.5 \cdot 0.8$ m³ in size was irradiated with several types of relativistic heavy ions from the Synchrophasotron at the Laboratory of High Energies (LHE), JINR (Dubna). They used 8 GeV protons, and ²H, ⁴He, ¹²C beams each having an energy of 3.6 GeV/u. The inside of 2.3 tn Pb-target had some holes containing small samples of appr. 1 g uranium metal. These uranium samples were centrally aligned along the beam axis. Then the ²³⁸U(n, γ) reaction was studied through the determination of the yield of ²³⁹Np. The results are given in Fig. 1b. The units on the ordinate are (atoms ²³⁹Np produced) per (1gU-detector) per (single heavy

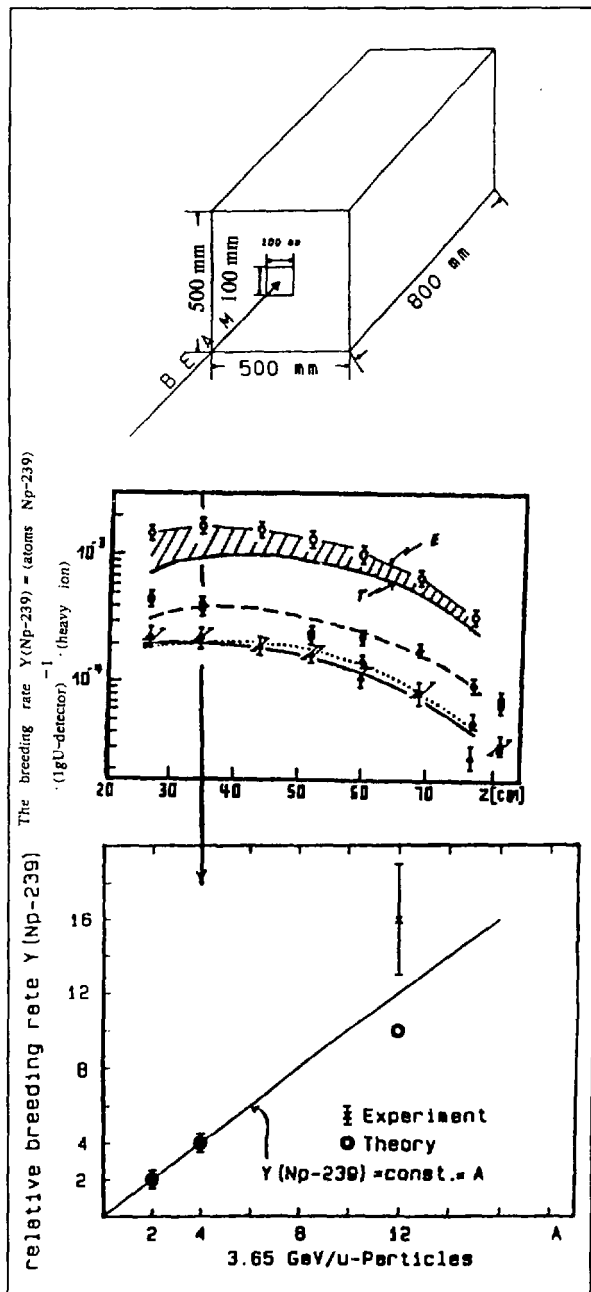


Fig. 1a. The very massive Pb-target ($0.5 \times 0.5 \times 0.8 \text{ m}^3$), irradiated with 3.65 GeV/u ions from the Synchro-phasotron, JINR, Dubna [6]

Fig. 1b. Results for the breeding of ^{239}Np in the very massive Pb-target (fig. 1a), as determined along the central beam axis. Open circles: $3.65 \text{ GeV/u } ^{12}\text{C}$, closed circles: $3.65 \text{ GeV } ^4\text{He}$, open triangle: $3.65 \text{ GeV/u } ^2\text{H}$, closed triangles: $8 \text{ GeV } ^1\text{H}$. For ^4He , ^2H and ^1H , the calculations of Y agree with the experiment. For ^{12}C , the calculations (T) are below the experiments (E), as shown by the hatched area

Fig. 1c. More-than-calculated-breeding (possibility) of ^{239}Np as seen in Fig. 1b. Here we show a «cut» along the line $Z = 35 \text{ cm}$

ion). This unit is strictly defined for the given empirical conditions. Nevertheless, one could calculate the production rates of ^{239}Np and obtain generally a fine agreement between experiment and theory. With only one exception (Fig. 1b): The experimental production rates of ^{239}Np for 44 GeV ^{12}C were larger than the calculated ones. This is demonstrated in Fig. 1c. A «cut» along the $Z = 35$ cm line in Fig. 1b is given here. We show the relative breeding rate of ^{239}Np as a function of the mass of the relativistic heavy ion (with a constant specific energy of 3.65 GeV/u). One can observe a linear correlation between these two parameters for ^2H and ^4He . However, for ^{12}C -ions we observe a discrepancy between experimental and theoretical (n, γ) rates. This discrepancy is $(60 \pm 30)\%$. Such an effect is interesting, but not yet significant. We come back to this observation later.

The American authors have investigated rather similar systems. Bowman et al. [7] published a very systematic theoretical study on «Nuclear Energy Generation and Waste Transmutation Using an Accelerator-Driven Intense Thermal Neutron Source». These authors base their study on the use of an accelerator for approximately 1 GeV protons. They could show that one would need thermal neutron sources of an intensity of approx. 10^{16} neutrons/s cm^2 before obtaining substantial transmutation rates of radioactive wastes. Quite recently, Carminati et al. have presented rather similar ideas in their paper on «An Energy Amplifier for Cleaner and Inexhaustible Nuclear Energy Production Driven by a Particle Beam Accelerator» [8]. It is not the intention of this article to comment on the statements found in Reference 8. But it appears useful to point out that Refs. 2—5, as quoted in this article, contain quite interesting aspects of the work concerned, even if they are not quoted in Ref. 8.

The authors of this contribution wanted to present some results of experiments studying the neutron generation in extended targets irradiated with relativistic ions at Dubna. A «Preliminary Note» of our work has already been published [9] giving the results for (n, γ) studies using La-detectors. In this article we give the additional results of studies using U-detectors and of some auxiliary experiments using solid state nuclear track detectors for the studying hadron-induced fission reactions in the Au-detectors.

EXPERIMENTAL

We used an extended Cu-target in contact with a paraffin moderator and irradiated it with ^{12}C -ions at two different relativistic energies. In the paraffin moderator we activated small samples of La and U. The choice of Cu as a target was motivated by radiochemical experiments using various Cu-target

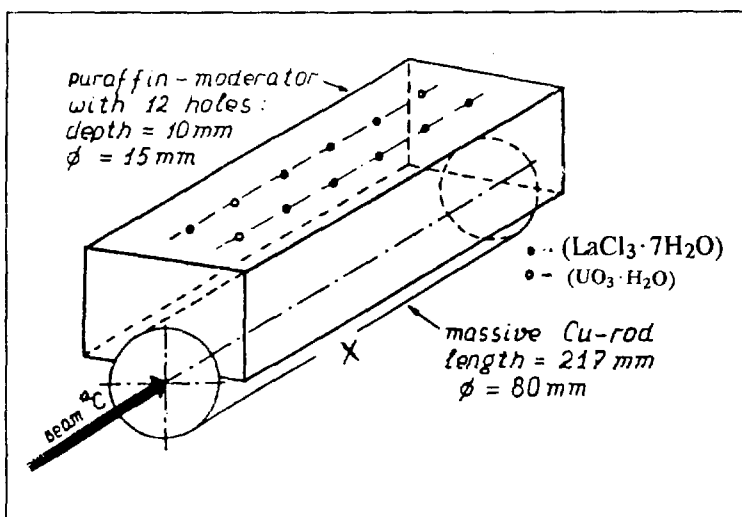


Fig. 2. Drawing of the experimental apparatus: a copper rod ($\varnothing = 8$ cm, $l = 21.7$ cm) is covered on its top with paraffin, having a thickness of 6 cm in the center. This paraffin block contains tiny holes filled with $(\text{LaCl}_3 \cdot 7\text{H}_2\text{O})$ and $(\text{UO}_3 \cdot \text{H}_2\text{O})$ samples. The position X, 9 cm downstream the beam entrance into the copper rod, marks the place, where the auxiliary exposure of a thin Au-layer on mica is carried out on the surface of the copper. Here we study the fission of Au as registered in a solid state nuclear track detector

configurations exposed to different relativistic beams [10—16]. The most significant experiments were carried out with relativistic ^{12}C -beams. In this paper we want to test the suggestion of Refs. 13 and 15: The «breeding rates» of (n, γ) products in such experiments should increase stronger between 22 and 44 GeV ^{12}C than could be calculated theoretically.

Our experimental set-up is shown in Fig. 2. A Cu-rod was irradiated axially with well-focused external beams at the Synchrophasotron, LHE, JINR (Dubna) for some extended periods of time. We used a 44 GeV ^{12}C -beam for 14.7 hours yielding in total $1.03 \cdot 10^{12}$ ions. This was followed by irradiation with a 22 GeV ^{12}C -beam for 11.3 hours yielding in total $5.9 \cdot 10^{11}$ ions. The beam intensity was rather stable during the irradiations.

The target, relevant for the experiment reported here, consisted of a copper-rod, 8 cm in diameter, 21.7 cm long. The front-plate 2 cm thick was taken off the target after the 44 GeV irradiation and replaced by a new one for the 22 GeV run. The beam-diameter was smaller for 44 GeV than for 22 GeV ^{12}C .

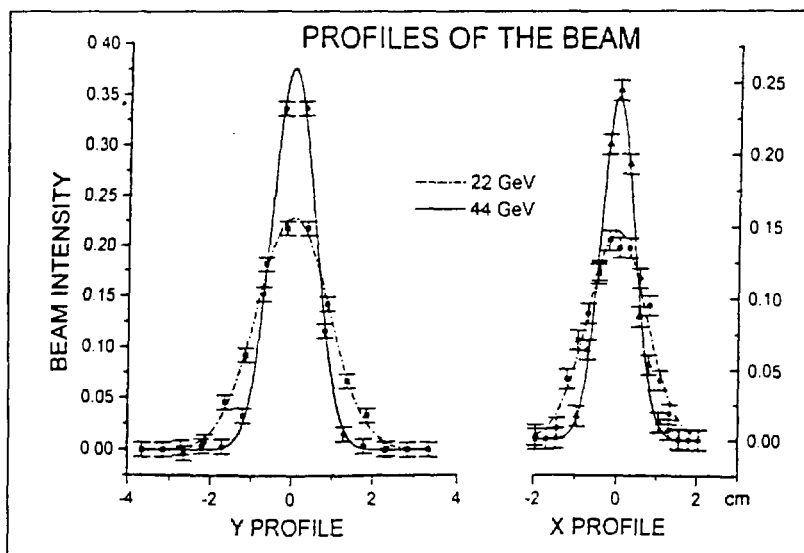
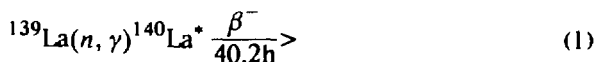


Fig.3. The beam profiles at 22 GeV and 44 GeV as observed with nuclear emulsions

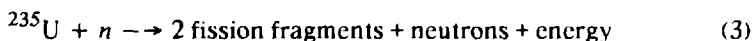
To measure the profile of the beam, emulsion films (MP 20 μ) were placed in front of and behind the Cu-target for perpendicular irradiations.

The measurements were done with and without the target. For measuring the beam profile a densitometer (Macbett TR-924) was used. The integral intensity of the beam for the irradiation of the films was $1 \cdot 10^8$ ^{12}C ions. The beam profiles for the 22 and 44 GeV carbon beams are given in Fig.3. We used the projection of the beam density on the X and Y axes.

The copper rod was covered on top with a paraffin block, $(-\text{CH}_2-)_n$, intended to moderate the emitted secondary neutrons. The thickness of the paraffin amounted to 6 cm, the width of the block being 15 cm (Fig.2). Twelve holes, as shown in Fig.2, were bored into the top layer of the moderator. Small holes were filled with samples of approximately 1 g La (as $\text{LaCl}_3 \cdot 7\text{H}_2\text{O}$) or 1 g U (as $\text{UO}_3 \cdot \text{H}_2\text{O}$). The following reactions were induced by secondary neutrons in these samples:



$${}^{238}\text{U}(n, \gamma) {}^{239}\text{U} \xrightarrow{20\text{min}} \beta^- > {}^{239}\text{Np} \xrightarrow{56\text{h}} \beta^- , \quad (2)$$



(here we measured only ${}^{132}\text{Te}$ and ${}^{140}\text{Ba}$ as fission fragments)

After the irradiation, some samples (front Cu 2 cm thick, 6 La samples, and 6 U samples obtained at each energy) were transported to the Kernchemie, University of Marburg (Germany) and counted there. The counting started appr. 60 hours after the end of the 44 GeV ${}^{12}\text{C}$ irradiation and appr. 47 hours after the end of the 22 GeV ${}^{12}\text{C}$ irradiation. As we used rather depleted uranium activation detectors (0.4% ${}^{235}\text{U}$) and started counting the gamma-activity of fission fragments rather late, we can report only marginal results on the reaction (3) (${}^{235}\text{U} + n \rightarrow \text{fission}$) for lack of activity. Otherwise, the gamma-counting procedure and data handling are state-of-the-art radiochemistry as reported in Refs.11 and 12.

In addition we present the results for an auxiliary experiment using solid state nuclear track detectors: A thin Au-layer (appr. 1 mg/cm^2) evaporated onto mica (2.2 cm^2) was placed in direct contact with the outside of the Cu rod (position X in Fig.2). There Au was exposed to all secondary hadrons leaving the Cu rod 9 cm downstream.

RESULTS AND DISCUSSIONS

1. Measurement of a Heavy Ion Flux

A heavy ion flux has been measured at the Synchrotron in the standard way using electronic counters. The uncertainty in the total flux was less than 3%. If the flux measurement for the 22 GeV ${}^{12}\text{C}$ experiments is compared with that for the 44 GeV ${}^{12}\text{C}$ experiment, the uncertainty in the ratio of the two total ion fluxes is 4%, as one uses the same apparatus in both cases. Then we measured the ${}^{24}\text{Na}$ -activity radiochemically (via the 1369 keV gamma line) in the 2 cm thick front Cu-plate and obtained the following activity ratio $S({}^{24}\text{Na})$. The statistical uncertainty in this ratio includes only the uncertainty of the radiochemical activity measurements. The variation of the heavy ion flux during the irradiation has been taken care of in the conventional mathematical manner. The heavy ion flux is given in particles per unit time; HIF(44) and HIF(22) are the heavy ion fluxes at 44 GeV and 22 GeV, respectively

$$S(^{24}\text{Na})_{\text{meas.}} = \frac{A^\infty(44 \text{ GeV}^{12}\text{C} + \text{Cu} \rightarrow ^{24}\text{Na} + X) / \text{HIF}(44)}{A^\infty(22 \text{ GeV}^{12}\text{C} + \text{Cu} \rightarrow ^{24}\text{Na} + X) / \text{HIF}(22)} = 1.21 \pm 0.03,$$

A^∞ are radiochemical "equilibrium" decay rates, to be defined later. The cross-sections for both monitor reactions are known as follows:

$$\sigma(44 \text{ GeV}^{12}\text{C} + \text{Cu} \rightarrow ^{24}\text{Na} + X) = (12.3 \pm 1.8) \text{ mb (Ref.}^{11})$$

$$\sigma(18.5 \text{ GeV}^{12}\text{C} + \text{Cu} \rightarrow ^{24}\text{Na} + X) = (9.7 \pm 1.0) \text{ mb (Ref.}^{17}).$$

The ratio for the monitor cross-sections agrees remarkably well with the experimentally determined $S(^{24}\text{Na})_{\text{meas.}}$. This gives evidence that the radiochemical monitor cross-section ratio yields results for the heavy ion flux ratio consistent with the measurements at the Synchrophasotron. We decided to use in subsequent calculations only the ^{12}C -flux measurements and their uncertainties as obtained by conventional beam measurements using on-line electronic counters.

Additionally, we also measured the long-lived nuclide ^{22}Na ($T_{1/2} = 2.6 \text{ a}$) in the two 1 cm thick Cu-disks and obtained the same activity ratio $S(^{22}\text{Na})_{\text{meas.}}$ as for the isotope ^{24}Na ($T_{1/2} = 15 \text{ h}$).

2. Measurement of a ^{239}Np -Activities

As can be seen in Fig.2, we have 12 small plastic vials, half of them containing uranium. The vials were numbered and aligned along the 21.7 cm beam axis. Their gamma-activity was measured on the top of a calibrated HPGe (High Purity Germanium) detector for several days until the ^{239}Np activity ($T_{1/2} = 56 \text{ h}$) had decayed. The spectrum analysis was quite straightforward as the decay of ^{239}Np was dominant for the uranium activation detector. For our analysis we used the 277.6 keV gamma line (a branching ratio of 14.1% in ^{239}Np -decay) and the 209.8 keV gamma line (a branching ratio of 3.2%). At first we determined in each sample individually the decay rate at the end of bombardment, $A_{\text{cob.}}$ (i). Then we collected all 6 vials, exposed at one C-energy, counted their activity together and obtained the corresponding value for $A_{\text{cob.}}$ (coll.). We analysed the results twofold.

3. Analysis of Individual U-Samples:

After having obtained $A_{\text{eob.}}(i)$ for each sample i , one took the total irradiation time and calculated the so-called «equilibrium» decay rate of ^{239}Np theoretically obtained after an infinite irradiation time of sample i :

$$A^{\infty}(^{239}\text{Np})_i.$$

The result was used to calculate an «experimental production rate» B_{exp} defined in a self-evident manner as follows:

$$B_{\text{exp.}} = \frac{(\text{number of } ^{239}\text{Np}\text{-atoms formed})}{(1 \text{ g U-detector}) \cdot (\text{single heavy ion})}$$

The results are given in Fig.4. The absolute numbers are rather similar to those shown in Fig.1. But this may be accidental as we have used copper as a

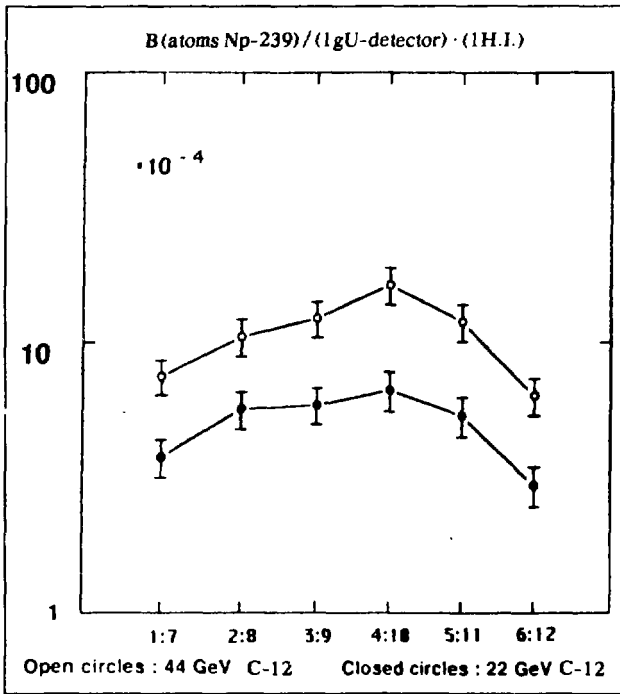


Fig.4. Production rates B_{exp} as observed in U-samples shown in Fig.2. Vial numbers $1 \leq i \leq 6$ have been obtained at 44 GeV, $7 \leq i \leq 12$ at 22 GeV ^{12}C ion energy

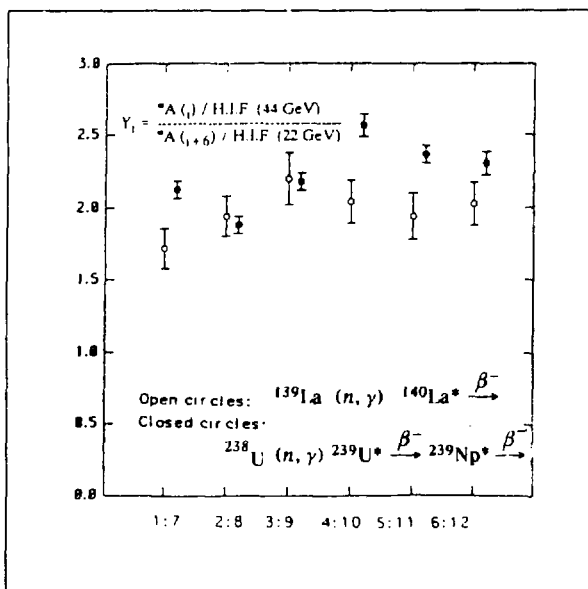


Fig.5. The ratio Y_1 for the reactions $^{139}\text{La} (n, \gamma)$ and $^{238}\text{U} (n, \gamma)$. Details are given within the figure and text

target plus moderator instead of lead. The production rate of Np was measured on the surface of a copper-plus-paraffin target as compared to the center of a pure metallic Pb-target (Ref.6). The parameter B_{exp} introduced by Voronko et al. [6] is a strictly empirical parameter specified in each experiment for

- a given nuclear reaction induced by secondary neutrons;
- a well-defined experimental set-up as shown in Fig. 1a or Fig. 2;
- for a given particle and its energy such as 44 GeV (or 22 GeV) ^{12}C .

The uncertainty in the values of B (Fig. 3) is about 15% and essentially due to uncertainties in the counting efficiencies. A strong decrease of the «edge» values of B (vials numbered 1;7 and 6;12) appears to be not connected with any fundamental aspect of this work. But it can merely reflect that these plastic vials were placed very close to the edge of the large paraffin block (appr. 17 mm from this edge) and did not experience a rather large «neutron sea» within the center of the paraffin block.

In addition, it seems interesting to compare the values of A^∞ obtained for U- or La-activation detectors within the same position of this paraffin block, but exposed to 44 GeV or 22 GeV carbon beams. The results for the ratio Y are

shown in Fig.5 for U- and La-activation detectors. The value of Y is measured as follows:

$$Y_i = \frac{A^\infty(i) / \text{HIF}(44)}{A^\infty(i+6) / \text{HIF}(22)},$$

HIF(44), HIF(22) are the ion fluxes at 44 GeV and 22 GeV, respectively. This flux is measured in ions/hour.

The given uncertainties are caused by the uncertainties in the ratio of heavy ion fluxes (4%) and in the activity ratios ((3–4) %).

It is interesting to note that the two independent (n, γ)-reactions studied with our system give rather similar results for Y. However, the Np-production appears to yield slightly larger values of Y. The comparison with theory will be given below.

4. Analysis of Combined U-samples for ^{239}Np , ^{140}Ba and ^{132}Te Activities

When we measure all 6 U-vials exposed at one energy simultaneously on the HPGe detector, we obtain results for combined activities. Now one can compare the activity ratios at 44 GeV and 22 GeV as follows:

$$R(^{239}\text{Np}) = \frac{\sum A_{1-6}^\infty(^{239}\text{Np}) / \text{HIF}(44)}{\sum A_{7-12}^\infty(^{239}\text{Np}) / \text{HIF}(22)} = 2.30 \pm 0.12,$$

HIF(44) and HIF(22) are the heavy ion fluxes at 44 GeV and 22 GeV, respectively.

The reasons for uncertainty in this value of $R(^{239}\text{Np})$ have already been given. Fig.6 shows the increase in this value of R from 22 GeV (normalized to $R = 1.000$) up to 44 GeV for La- and U-activation detectors.

In this paper we can only present marginal results on fission studies in uranium (reaction (3)) due to a low activity of fission fragments in the samples studied. The results for $R(^{140}\text{Ba})$ and $R(^{132}\text{Te})$ are:

$$R(^{140}\text{Ba}) = \frac{\sum A_{1-6}^\infty(^{140}\text{Ba}) / \text{HIF}(44)}{\sum A_{7-12}^\infty(^{140}\text{Ba}) / \text{HIF}(22)} = 2.0 \pm 0.3,$$

$$R(^{132}\text{Te}) = \frac{\sum A_{1-6}^\infty(^{132}\text{Te}) / \text{HIF}(44)}{\sum A_{7-12}^\infty(^{132}\text{Te}) / \text{HIF}(22)} = 2.0 \pm 0.3.$$

The uncertainties in these values are essentially due to those in the counting rates, i.e. the 487 keV gamma line for ^{140}Ba and the 772 keV gamma line for ^{132}Te .

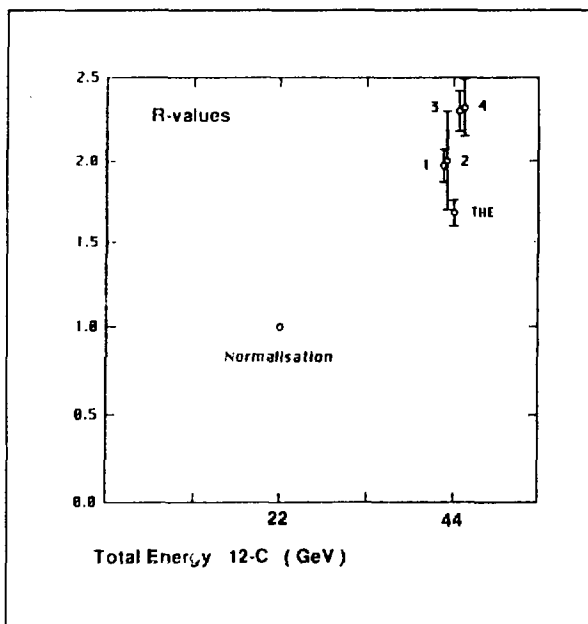


Fig.6. R-values for 44 GeV as compared to 22 GeV ^{12}C -ions (normalization). The numbers stand for the following experiments:

1. $^{139}\text{La} (n, \gamma) ^{140}\text{La}^* \xrightarrow{\beta^-}$; $R = 1.97 \pm 0.10$,

2. $^{235}\text{U} (n, f) ^{140}\text{Ba}^* \xrightarrow{\beta^-}$; $R = 2.0 \pm 0.3$,

3. $^{228}\text{U} (n, \gamma) ^{239}\text{U}^* \xrightarrow{\beta^-} ^{239}\text{Np}^* \xrightarrow{\beta^-}$; $R = 2.30 \pm 0.12$,

4. $\text{Au} + (\text{f.a.rons}, E > 30 \text{ MeV}) \rightarrow \text{fission}$; $R = 2.32 \pm 0.17$.

Details are described in the text. (THE) stands for theoretical estimation; $R = 1.68 \pm 0.08$

In addition, we have calculated the values of B averaged for all six vials at one energy for the production of one fission fragment, ^{140}Ba , in our set-up. We can expect that uranium is exposed to a somewhat thermalized neutron flux:

$$B_{\text{exp}}(\text{U} \rightarrow ^{140}\text{Ba})_{44\text{GeV}} = (4.2 \pm 0.6) \cdot 10^{-5} \frac{\text{atoms } ^{140}\text{Ba}}{(1 \text{ g U}) \cdot (\text{single heavy ion})}$$

$$B_{\text{exp}}(\text{U} \rightarrow ^{140}\text{Ba})_{22\text{GeV}} = (2.1 \pm 0.3) \cdot 10^{-5} \frac{\text{atoms } ^{140}\text{Ba}}{(1 \text{ g U}) \cdot (\text{single heavy ion})}$$

The results for the fission fragment ^{132}Te are rather similar. As we know that the branching ratio for ^{140}Ba in thermal neutron induced fission is 6.2%, we calculate the values of B for fission in «our» uranium as follows:

$$B_{\text{exp}}(\text{U} \rightarrow \text{fission})_{44\text{GeV}} = (6.9 \pm 1.0) \cdot 10^{-4} \frac{\text{fission}}{(1 \text{ g U}) \cdot (\text{single heavy ion})}$$

$$B_{\text{exp}}(\text{U} \rightarrow \text{fission})_{22\text{GeV}} = (3.3 \pm 0.5) \cdot 10^{-4} \frac{\text{fission}}{(1 \text{ g U}) \cdot (\text{single heavy ion})}$$

Now we compare these values of B_{exp} with those of Fig.4, i.e., for the reaction $^{238}\text{U}(n, \gamma)$. We find the value of B_{exp} for the production of ^{239}Np larger than for the reaction $^{235}\text{U}(n, f)$.

$$\frac{B_{\text{exp}}(\text{U} \rightarrow ^{239}\text{Np})_{44\text{GeV}}}{B_{\text{exp}}(\text{U} \rightarrow \text{fission})_{44\text{GeV}}} = 1.7 \pm 0.3$$

and

$$\frac{B_{\text{exp}}(\text{U} \rightarrow ^{239}\text{Np})_{22\text{GeV}}}{B_{\text{exp}}(\text{U} \rightarrow \text{fission})_{22\text{GeV}}} = 1.5 \pm 0.3.$$

Finally, we took an uranium sample (the same as that used in all the experiments of this paper) and exposed it in a thermal column of a research reactor for 2 minutes to a flux of thermal neutrons of $7 \cdot 10^{11} \text{ n/cm}^2 \cdot \text{s}$. We analysed the γ -activity in the same manner as above and found:

$$\frac{B_{\text{exp}}(\text{U} \rightarrow ^{239}\text{Np})_{\text{n-thermal}}}{B_{\text{exp}}(\text{U} \rightarrow \text{fission})_{\text{n-thermal}}} = 2.09 \pm 0.07.$$

This shows, that the neutron spectra inducing (n, γ) - and (n, f) -reactions in our target are shown to be close to the neutron-spectrum in a thermal column of a research reactor (i.e., TRIGA, Institute für Kernchemie, Mainz, Germany), but the neutron spectra are not identical. This result shows that we need improved experimental and theoretical considerations before complete understanding the system studied here. But we have considered the publication of the marginal values of $B_{\text{exp}}(\text{U} \rightarrow \text{fission})$ to be essential as these results are the first of this kind ever published. The results cannot be found in Ref.6.

5. Results on the Auxiliary Experimental Using Solid State Nuclear Track Detectors (SSNTD's) in Fission Studies of Gold

The mica detector (2.2 cm^2) covered with appr. 1 mg/cm^2 Au was exposed to a secondary hadron beam emitted from the surface of the Cu-rod 9 cm downstream the beam entrance (Fig.2). It is well known that Au is fissioning only with medium energy hadrons (protons, alpha, neutrons, etc.) above appr. 30 MeV [18]. Consequently, our auxiliary target is a tool to measure the flux of medium energy hadrons emitted at the position X in Fig.2. Au-fission fragments are emitted into mica, where they can be revealed as single tracks after the end of bombardment by proper etching (20% H_2F_2 for 30 min.). Then one can determine the track density on the mica surface using an optical microscope. The results are given in the table:

^{12}C -beam energy GeV	track-density I on mica ($\pm 2\%$) (cm^{-2})	target thickness T mg/cm^2 ($\pm 3\%$)	beam intensity B 10^{12} ions ($\pm 3\%$)	relative track density $W = I/T \cdot B \cdot 10^4$ ($\pm 5\%$)
44	$1.17 \cdot 10^4$	0.83	1.03	1.37
22	$0.42 \cdot 10^4$	1.20	0.59	0.59

The relative track density ratio at the two carbon energies equal to

$$R(\text{Au} \rightarrow \text{fission}) = \frac{W(44\text{GeV})}{W(22\text{GeV})} = 2.32 \pm 0.17,$$

is again in agreement with the preceding values for U-detectors.

6. Model Estimation on the Observed R Values, Including the Study of Effects Due to Different Beam Profiles

Recently a computer program has been developed to calculate the propagation of relativistic heavy ions and all their secondary fragments through a rather massive target as shown in Fig.2 [19]. Using this program, one can calculate the entire neutron flux generated in the massive copper rod. The main purpose of our study here (using the above-mentioned Monte Carlo codes) is to count the number of neutrons inside the moderator volume as it is volume the radiochemical detectors are located in. The process of ^{239}Np generation in uranium detectors, as well as ^{140}La production in lanthanum detectors is strongly dependent on the neutron spectrum especially at energies far below 14 MeV. Thus, we have considered both the overall neutron flux and the neutron flux below 14 MeV. (Our program did not allow calculating the actual neutron flux

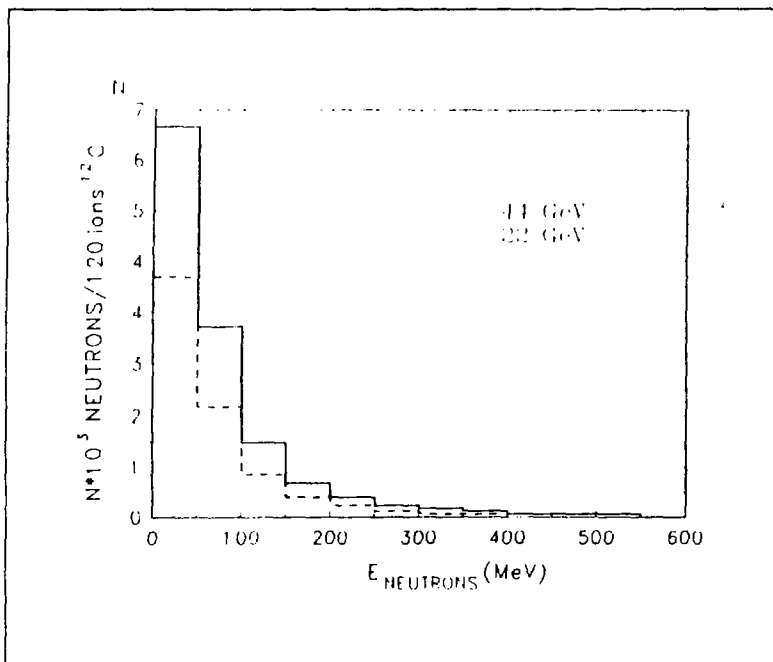


Fig. 7. Neutron spectra of secondary neutrons emitted from the cylindrical surface of the copper target rod as shown in Fig. 2

below 1 MeV down to thermal energies.) The calculations have been performed for incident carbon ions at 44 GeV and 22 GeV, respectively. This beam profile was considered as shown in Fig. 3, it is wider along the vertical axis as compared to the horizontal axis and wider again for 22 GeV as compared to 44 GeV. In order to take this properly into account, we investigated the neutron emission into a $\pm 45^\circ$ sector (as compared to the central vertical plane) in our detection system (Fig. 2). We studied two kinds of neutron emission spectra: one for the entire target, the other for the $\pm 45^\circ$ top sector. For our calculation we considered the entire Cu-target to be surrounded by a 6 cm thick cylindrical moderator $(-\text{CH}_2)_n$. The resulting neutron spectrum emitted from the entire Cu-target is shown in Fig. 7. One can see easily, that the shapes of both neutron spectra are identical for 22 GeV and 44 GeV — within the limits of their statistical accuracy. As only low energy neutrons ($E < 14$ MeV) are of any practical importance, we consider the ratio of neutrons with $0 < E < 14$ MeV emitted from the Cu-target as being equivalent to the calculated ratio of (n, γ) products, as follows:

$$R_{\text{theoretical}} = \frac{(\text{neutrons generated with 44 GeV})}{(\text{neutrons generated with 22 GeV})} = 1.68.$$

The uncertainty in this number is 2% due to the limitations of the available computers. As the calculated influence of the beam profile on $R_{\text{theoretical}}$ for the $\pm 45^\circ$ top sector is less than 5%, we assign a 5% uncertainty to the estimated $R_{\text{theoretical}}$ value and obtain finally

$$R_{\text{theoretical}} = 1.68 \pm 0.08.$$

Now one can consider this result in other words: The intensity of the neutron flux (as shown in Fig.7), i.e. the integral number of neutrons emitted from the cylindrical surface of the copper rod increases only by $(68 \pm 8)\%$ when the ^{12}C ion energy doubles from 22 to 44 GeV. This should only lead to $(68 \pm 8)\%$ increase in the R values, as shown in Fig.6. It is interesting to note that the experimental R value in Fig.6, is up to $(37 \pm 9)\%$ larger than the calculated value.

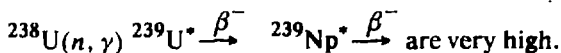
This increase of R values at 44 GeV ^{12}C beyond theoretical calculations can be explained by anomalies observed in relativistic heavy ion interactions above a total energy of appr. (30—35) GeV. Such anomalies are described in Refs. 10—16. In these articles we described radiochemical experiments using various Cu target configurations exposed to different relativistic heavy ion beams. Much of the time was spent studying the production of ^{24}Na ($T_{1/2} = 15$ h) from copper targets, and the anomalies observed were an excess of ^{24}Na production over theoretical estimates under certain well-defined experimental conditions.

After having carried out further experiments we will be able elucidate this point in future publications.

7. Summary and Outlook

Our experiments confirm the results of the preceding experiments and considerations from Russia (Refs.5,6) as follows:

1. The production of secondary neutrons in extended metallic targets irradiated with relativistic heavy ions is quite remarkable. The rates for the production of (n, γ) products, as measured in the reaction



2. We observe (at 44 GeV) ^{12}C production rates for ^{239}Np due to (n, γ) reactions which are $(37 \pm 9)\%$ more than our theoretical estimations.

And the thing we want to mention:

When rather extended uranium targets (say: larger than appr. 1 ton U) are irradiated with relativistic heavy ions, one must watch out for the extent of the generated neutrons. Such a system may emit more secondary neutrons than one can reasonably estimate. Let us consider an example

— 1 ton uranium irradiated with

— 10^{12} ions/s heavy ions ($A \sim 200$) with $E \sim 100$ GeV/u

— and a fission energy of 160 MeV, a specific heat of uranium (0.03 cal/g), and

— the Np-production rates equal to the fission rates (such an assumption is conservative as the fission rate will be larger than the (n, γ) rate within the center of a metallic block, possibly by orders of magnitude):

$$\frac{(\text{atoms}^{239}\text{Np})}{(1\text{g U}) \cdot (1\text{H.I.})} = \frac{(\text{fission events})}{(1\text{g U}) \cdot (1\text{H.I.})} = 0.5,$$

(1 H.I.) stands for: «single heavy ion».

This value is a linear extrapolation of the values observed in a Pb-target [6] (see Fig. 1). Then the heat of the uranium block increases through fission energy alone by $0.1^\circ\text{C}/\text{second}$. This is a non-trivial number. Considering the enhanced production of secondary neutrons over theoretical estimates, this increase may be even larger. As we have absolutely no information what this «enhancement-effect» could amount to quantitatively, we must say that the irradiation of extended uranium targets with very heavy ions of very high energy is a non-trivial issue.

But, of course, such an experiment can be carried out even under these conditions quite safely: One should study the production of fission fragments and Np from uranium in an extended uranium target at first at low beam intensities, for example, 10^5 ions/second, then slowly increasing the intensity and determining experimentally to what beam intensity such a system could operate safely.

ACKNOWLEDGEMENT

We want to thank the excellent operating crew at the Synchrophasotron, Laboratory of High Energies, Joint Institute for Nuclear Research in Dubna (Russia) for the irradiations and Academician A.M. Baldin as well as Professors A.I. Malakhov and I.B. Issinsky for their continued support of this work. The Marburg team is grateful for the opportunity to train young scientists at the LHE and to enjoy the cordial hospitality of their Russian colleagues in Dubna. Some authors (B.A.K., M.I.K., V.S.B.) thank some German Funding Agencies for financial support during their stay at the Kernchemie, Philipps-Universität Marburg.

The authors thank Prof. Yu.A.Panebratsev, Drs. S.S.Shimansky, and Yu.S.Averichev for their support in carrying out the irradiations of the targets at the F3 focus of the DISK installation.

REFERENCES

1. Proceeding of an Information Meeting on Accelerator Breeding. Upton, New York, January 1977.
2. Vassilkov R.G., Goldanski V.I., Orlov V.V — Uspekhi Phys. Nauk (Russian), 1983, 139, p.435.
3. Bakhmutkin S.V. et al. — Atomnaja Energija (Russian), 1987, 62, p.59.
4. "Workshop on Nuclear Transmutation of Long-Lived Nuclear Power Radiowastes" , (Obninsk (Russia), July 1—5, 1991). Institute of Nuclear Power Engineering, Obninsk and Institute of Theoretical and Experimental Physics, Moscow, in particular: V.S.Buchenkov, et al., p.273.
5. Tolstov K.D. — JINR Preprints 18-89-778, 1989; 18-92-303, 1992, Dubna.
6. Voronko V.A. et al. — Atomnaja Energija (Russian), 1990, 68, p.449; 1991, 71, p.563.
7. Bowman C.D. et al. — Nuclear Instruments and Methods in Physics Research, 1992, A 320, p.336. (containing a rather extensive list of references)
8. Carminati F. et al. — CERN Preprint AT/93-47 (ET), Geneva, Nov.1.93.
9. Bisplinghoff B. et al. — Preprint JINR, E1-93-127, Dubna, 1993; ISOTOPENPRACTICE , 1994, in print.
10. Brandt et al. — Proc. of the IX International Seminar in High Energy Physics Problems (June 1988, Dubna), D1,2-88-652 vol.2, p.687 and Proc. of the X. Intern. Seminar on High Energy Physics Problems (Sept. 1990, Dubna), Ed. A.M Baldin, World Scientific, Singapore. 1991, p.405.
11. Brandt R. et al. — ISOTOPENPRACTICE, 1989, 25, p.434.
12. Brandt R. et al. — Phys. Rev., 1992, C45, p.1194.
13. Brandt R. et al. — Science Intern. (Lahore), 1992, 4, p.3.
14. Heck M., Sosnin A.N. — Proc. XI. Intern. Seminar on High Energy Physics Problems (Sept. 7—12, 1992, Dubna). Ed. A.M.Baldin et al., "World Scientific Publishing" (Singapore), in print.
15. Brandt R. — "Nuclear Tracks and Radiation Measur.", 1993, 21, p.34.
16. Brandt R. et al. — "Nuclear Tracks and Radiation Measur.", 1994, in print.
17. Cole C.D., Porile N.T. — Phys.Rev., 1982, C25, p.244.

18. See, for example, E.K.Hyde, *The Nuclear Properties of the Heavy Elements, Part III, Fission Phenomena*, Prentice Hall, Englewood Cliffs, New Hersey, 1964, Chapter 10; and A.Khodai-Joopari, Ph.D.-Thesis, University of California, Berkeley, 1966 (UCRL-16489).
19. Polanski A. et al. — JINR Preprint E2-91-562, Dubna, 1991.

Received by Publishing Department on April 5, 1994.