93-127

объединенный
институт
ядерных
исследований
дубна

E1-93-127

B.Bisplinghoff[1], R.Brandt[1], V.S.Butsev, E.M.Friedlander[1,2],
M. Heck[1], M.I.Krivopustov, B.A.Kulakov, C.Laue[1],
L.Lerman[1,2], Th.Schmidt[1], A.N.Sosnin

NEUTRON GENERATION IN AN EXTENDED Cu-TARGET IRRADIATED WITH 22 AND 44 GeV CARBON IONS

Submitted to "Isotopenpraxis"

---

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

[2] Nuclear Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, CA, USA

1993

INTRODUCTION

For the last half century there has been a 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 to use high-energy proton beams ($E \sim 1$ GeV) for what is called «Electrical or Accelerator Breeding» [1]. Afterwards, Vassilkov et al. [2] published a very interesting paper. Furthermore, Bakhmutkin et al. [3] reported on experimental results obtained with an extended iron-target irradiated with high-energy protons. Recently, these studies have been reactivated [4] and Tolstov obtained interesting results [5,6]. Moreover, Bowman et al. published a very extended theoretical study on: «Nuclear Energy Generation and Waste Transmutation Using an Accelerator-Driven Intense Thermal Neutron Source» [7]. In principle, the American authors base their study on the use of an accelerator for roughly 1 GeV protons.

The same scientific aims are pursued at the Laboratory of High Energies (LHE), JINR (Dubna) using the relativistic heavy ion accelerator complex SYNCHROPHASOTRON-NUCLOTRON. Some theoretical concepts have been presented at first by Vassilkov et al. [2], later on by Tolstov [5]. Initial experimental work using a Pb-target irradiated with proton (8 GeV) and d, ^{12}He, ^{12}C beams having an energy of 3.6 GeV/n has been published [6].

EXPERIMENTAL

It is the intention of this short communication to report on somewhat similar work, this time using an extended Cu-target in contact with a paraffin moderator and irradiated with one sort of heavy ions, i.e. ^{12}C-ions at two different relativistic energies. In the paraffin moderator we activated small samples of La and U. In this paper we report on some La-investigations, other results will be published in a consecutive note.

The choice of Cu as a target was motivated by radiochemical experiments using various Cu-target configurations exposed to many different relativistic beams [8—12]. It is not the intention to discuss these publications now and their connection to the experiment reported here. This may be the subject of subsequent papers.

Our experimental set-up is shown in Fig.1. It was irradiated axially with well-focussed 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 $5.90 \cdot 10^{11}$ ions. The beam intensity was rather stable during the irradiations.

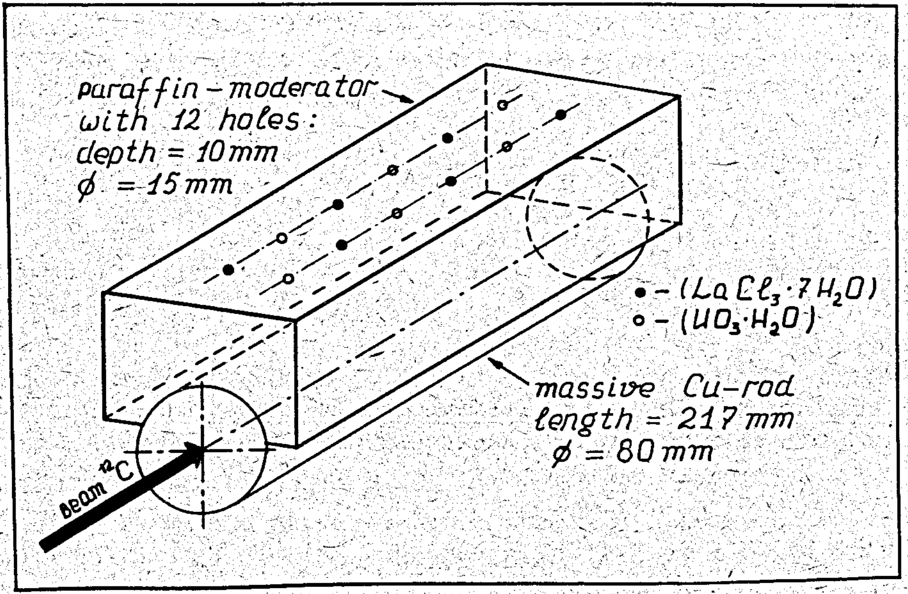

Fig.1. Drawing of the experimental apparatus: a copper rod (Ø = 8 cm, l = 21,7 cm) is covered on its top with paraffin (6.0 cm). This paraffin block contains tiny holes filled with ($LaCl_3 \cdot 7H_2O$) and ($UO_3 \cdot H_2O$) samples

The target, relevant for the experiment reported here, consisted of a copper rod with a diameter of 8 cm and a length of 21.7 cm. The front plate of 2 cm thickness taken off the target after the 44 GeV irradiation was replaced by a new one for the 22 GeV run. The beam diameter was smaller for 44 GeV ^{12}C than for 22 GeV ^{12}C. In this first note we use the measurement of a beamspot as determined by a polaroid-film with one burst (appr. $1.3 \cdot 10^{8}$ ions).

We measured beam-spot sizes of roughly 21 mm for 44 GeV ^{12}C and 29 mm for 22 GeV ^{12}C.

A more precise beam profile was measured with a nuclear emulsion plate, its result will be incorporated in the next publication. The copper rod was covered on top with paraffin $(-CH_2-)_n$ intended to moderate the emitted secondary neutrons. The thickness of the paraffin amounted to 6 cm the width of the block being 15 cm. Twelve holes, as indicated in Fig.1, were bored into the top layer of the moderator. These small holes were filled with samples of approximately 1g La (as $LaCl_3 \cdot 7H_2O$) or 1g U (as $UO_3 \cdot H_2O$). Secondary neutrons induced in these samples the following reactions:

1) $^{139}La(n,\gamma)\,^{140}La \xrightarrow[40.2h]{\beta^-}$

2) a) $^{238}U(n,\gamma)\,^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow[56h]{\beta^-}$

b) $^{235}U(n,f) \longrightarrow$ two fussion fragments + neutrons + energy

After the irradiations, the samples (front Cu 2 cm in thickness, 6 samples of La, 6 samples of U — obtained at each energy) were transported to the Kernchemie, University Marburg (Germany), and counted there. The counting started appr. 60 hours after the end of the 44 GeV ^{12}C experiment and appr. 47 hours after the end of the 22 GeV ^{12}C experiment. The details of the gamma-counting procedure and data handling can be found in Refs. [8—10], the used technique is state-of-the-art radiochemistry [10].

RESULTS AND DISCUSSIONS

In this first note we present the experimental results for the activation of the La-targets. Since the gamma-counting of fission fragments in the U-target is still going on, we will present their results later.

1. Measurement of the Heavy Ion Flux

The heavy ion flux has been measured by the operators of the SYNCHROPHASOTRON in the conventional way. The uncertainty in the ratio of the total fluxes at 44 and 22 GeV energies is estimated by the operators of the accelerators to be approximately 7%. Then we measured the ^{24}Na-activity radiochemically in the 1 cm thick front Cu-plates and obtained the following activity ratios normalized to the same heavy ion flux (the statistical uncertainty in this ratio includes only the uncertainty of the radiochemical activity measurements):

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

A^{∞} are radiochemical «equilibrium» decay rates to be defined later. As the cross sections for both monitor reactions are known:

$$\sigma\,(44\ \text{GEV}\ ^{12}C + Cu \rightarrow {}^{24}Na + X) = (12.3 \pm 1.8)\ \text{mb}\ [10\,],$$

$$\sigma\,(18.5\ \text{GeV}\ ^{12}C + Cu \rightarrow {}^{24}Na + X) = (9.7 \pm 1.0)\ \text{mb}\ [13\,],$$

one could calculate the same ratio normalized again to the same heavy ion flux (the uncertainty in this ratio includes here the uncertainty in the monitor reaction cross sections):

$$R(^{24}Na)_{\text{monitor}} = \frac{A^{\infty}(44\ \text{GeV}\ ^{12}C + Cu \rightarrow {}^{24}Na)}{A^{\infty}(18.5\ \text{GeV}\ ^{12}C + Cu \rightarrow {}^{24}Na)} = \frac{12.3 \pm 1.8}{9.7 \pm 1.0} = 1.27 \pm 0.23.$$

Both values, $R(^{24}Na)_{\text{meas.}}$ and $R(^{24}Na)_{\text{monitor}}$, agree remarkably well with each other. Within the uncertainties of the experiment it is reasonable to assume that the monitor cross section does not vary appreciably between 18.5 GeV and 22 GeV for ^{12}C-ions. We also measured ^{22}Na in the two 1 cm thick Cu-disks and obtained the same activity ratio $R_{\text{meas.}}$ as for ^{22}Na. This is the evidence that the radiochemical monitor cross sections give results for heavy ion flux ratios consistent with the measurements of the operators of the SYNCHROPHASOTRON. As the monitor cross sections have rather large uncertainties, we decided to use in subsequent calculations only the ^{12}C-flux measurements and their uncertainties as given by the above-mentioned operators.

2. Measurement of ^{140}La-Acitivities

As can be seen in Fig.1, we have placed 6 small vials, each containing approximately 1g La (in the form of $LaCl_3 \cdot 7H_2O$), in the paraffin moderator. The vials were numbered and aligned along the 21.7 cm beam axis. After two

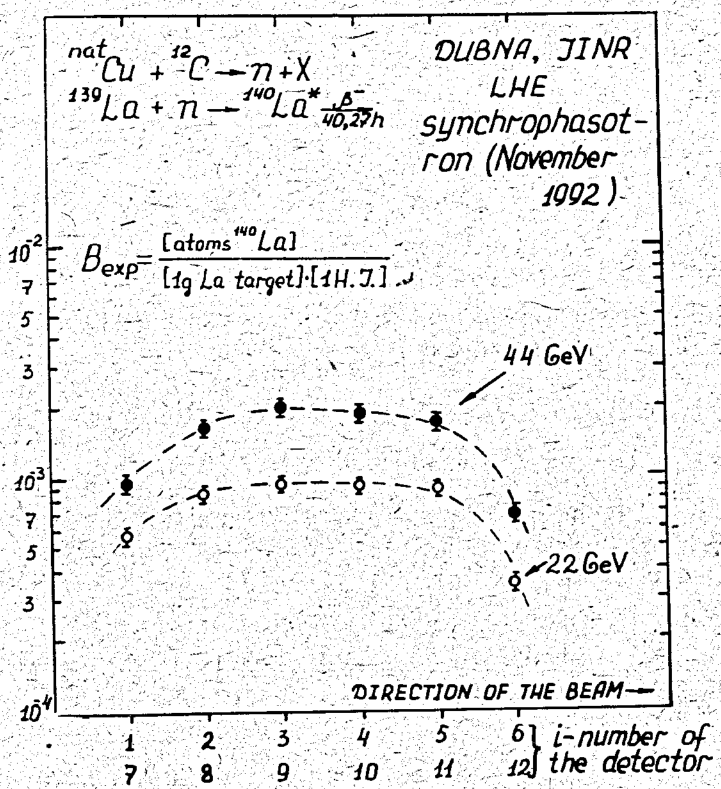

Fig.2. Production rates B_{exp} as observed in La-samples shown in Fig.1. Detector numbers $1 \leq i \leq 6$ have been obtained at 44 GeV; $7 \leq i \leq 12$ at 22 GeV

irradiations, two sets of 6 vials were counted for their gamma-activity on the top of a calibrated HPGe detector for several days until the 40.27 hours ^{140}La activity had decayed. The spectrum analysis was quite straightforward as practically only the decay of ^{140}La was observed. For our analysis we used gamma lines having a relative decay probabilibty larger than 2%. We determined in each sample at first the decay rate at the end of bombardment A_{eob}. Then taking the total irradiation time, one could calculate the so-called «equilibrium» decay rate of ^{140}La theoretically obtained after an infinite irradiation time of sample i

$$A^{\infty}(^{140}\mathrm{La})_{\mathrm{sample\ i}}\,.$$

The result was used to calculate an "experimental production rate" B which is defined in a self-evident way as follows:

$$B_{\mathrm{exp}} \equiv \frac{(\text{number of }^{140}\text{La atoms formed})}{(\text{1g La target})\cdot(\text{single heavy ion})}.$$

This value, B_{exp}, is an experimental number and has been also used by Voronko et al. [6]. It is specified in each experiment for:

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

The results of our experiment are given in Fig.2. The actual numbers observed are similar to those observed in Ref. [6].

3. Comparison of Experimental Results with Theoretical Estimations

In this section we want to investigate two questions:

3.1. How does the experimental production rate B_{exp} increase with the energy of the ^{12}C-ion?

We sum up the values of A^{∞}(La) for samples $1 \leq i \leq 6$, i.e. all the activities observed at 44 GeV. Then we do the same for samples $7 \leq i \leq 12$, i.e. all the activities observed at 22 GeV. Taking the ratios, one obtains (normlaized to the same heavy ion flux, H.I.F.):

$$R(^{140}\mathrm{La})_{\mathrm{exp}} = \frac{\sum\ (i=1,\ldots,6)\ A^{\infty}(^{140}\mathrm{La})/\mathrm{H.I.F.}}{\sum\ (i=7,\ldots,12)\ A^{\infty}(^{140}\mathrm{La})/\mathrm{H.I.F.}} = 1.97 \pm 0.15\ \text{(uncorrected)}.$$

The uncertainty is due to the ratio in the uncertainties of the heavy ion fluxes (± 7%) and the uncertainties in the ^{140}La determinations (±2%). It can be seen that the most important influence on this uncertainty of our result is due to the flux measurements. As mentioned previously, it turned out that the 44 GeV ^{12}C-beam was focussed better than the 22 GeV ^{12}C-beam. Only once the exact beam geometry is known and after proper corrections applied, we'll be in a position to state what the «correct» $R(^{140}$La) value really is.

3.2. How is the increase in B_{exp} compared with theoretical estimations?

For this we carried out some calculations for relative values of B due to ^{12}C ions at 44 GeV and 22 GeV. The ratio between the two values of B gives us a

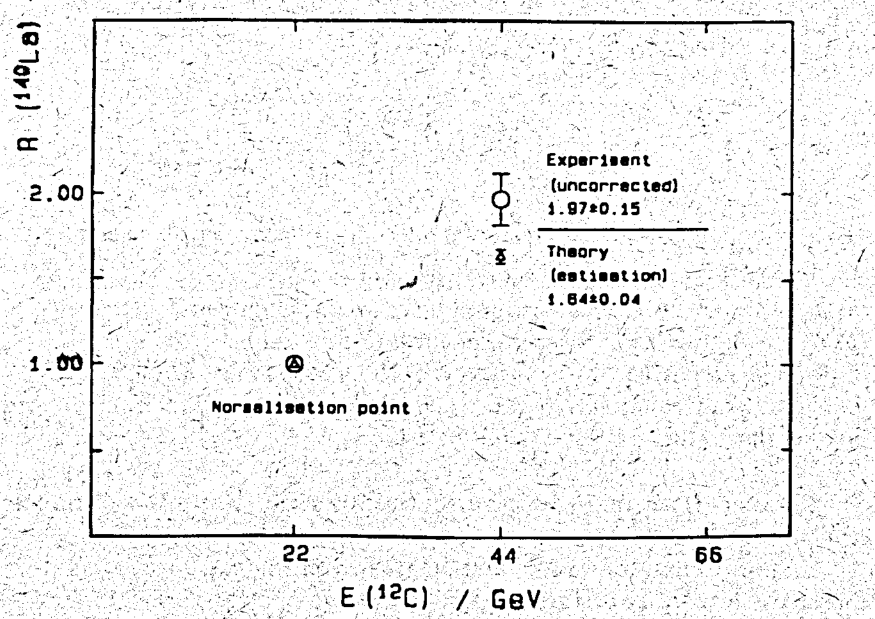

Fig.3. Preliminary results, as described in the text. The experimental and calculated results are normalized to $R(^{140}\mathrm{La}) = 1$ for 22 GeV carbon ions. The values $R(^{140}\mathrm{La})_{\mathrm{exp}}$ — (uncorrected) and $R(^{140}\mathrm{La})_{\mathrm{theor.}}$ — (extimation) have been described in the text

calculated value of R. The calculations are based in principle on the well-known Dubna Cascade Model, however its details have been slightly modified (as published recently by Polanski et al. [14]). In the calculation for the problem presented here, only preliminary results will be shown. We considered only neutrons with energies above 10.5 MeV escaping the well-defined Cu-target (Fig.1), integrating over energy and the target surface and assuming a Gaussian beam distribution with $\sigma = 0.43$ cm.

The experimental and theoretical rates R are normalized to unity at 22 GeV. Then one obtains for 44 GeV ^{12}C:

$$R(^{140}\mathrm{La})_{\mathrm{exp}} = 1.97 \pm 0.15 \text{ (uncorrected)}, \; R(^{140}\mathrm{La})_{\mathrm{theor}} = 1.64 \pm 0.04 \text{ (estimation)}$$

as shown in Fig.3. Statistically, the difference between $R(^{140}\mathrm{La})_{\mathrm{exp}}$ and $R(^{140}\mathrm{La})_{\mathrm{theor}}$ amounts to two standard deviations. Further work on systematic effects must be carried out in this field before unambiguous conclusions can be drawn. However, it is interesting to note that similar discrepancies between experimental and theoretical values of R have been observed for 44 GeV ^{12}C on Pb-targets (and not for 3.6 GeV/n,d, ^{4}He-beams on the same Pb-target) [6].

ACKNOWLEDGEMENT

We want to thank the excellent operating crew at the SYNCHROPHASOTRON, LHE (Dubna) for fine irradiations, Academician A.M.Baldin, Drs. A.I.Malakhov and I.B.Issinsky for their continued support of this work. The Marburg team is grateful to their Russian colleagues for their cordial hospitality at the JINR in Dubna.

One of the authors (M.I.K) thanks the DAAD, Deutscher Akademischer Auslandsdienst (Bonn, Germany) for the support of his stay at the Kernchemie, Philipps-University (Marburg, Germany).

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

REFERENCES

1. Proceedings of an Information Meeting on Accelerator Breeding, Upton, New York, January, 1977.
2. Vassilkov B.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, (July 1991, Obninsk, Russia), Institute of Nuclear Power Engineering, Obninsk and Institute of Theoretical and Experimental Physics, Moscow, in particular: V.S.Bychenko et al., p.273.
5. Tolstov K.D. — JINR, 18-89-778, Dubna, 1989; 18-92-303, Dubna, 1992.
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, A320, p.336 (containing a rather extensive list of references).
8. Brandt R. et al. — Proc. of the IX International Seminar on High Energy Physics Problems (June 1988, Dubna), D1,2-88-653 V.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.
9. Brandt R. et al. — Isotopenpraxis, 1989, 25, p.434.

10. Brandt R. et al. — Phys. Rev., 1992, C45, p 1194.
11. Brandt R. et al. — Science Intern. (Lahore), 1992, 4, p.3.
12. Heck M., Sosnin A.N. — Proc. XI Intern. Seminar on High Energy Physics Problems (Sept. 1992, Dubna), in print.
13. Cole C.D., Porile N.T. — Phys. Rev., 1982, C25, p.244.
14. Polanski A. et al. — JINR E2-91-562, Dubna, 1991.

Received by Publishing Department
on April 9, 1993.