First experiments with a large uranium blanket within the installation “energy plus transmutation” exposed to 1.5 GeV protons


The first experiment using a large U/Pb assembly (lead target plus 103.2 kg natural uranium blanket) exposed to 1.5 GeV proton beams was carried out in November 1999. It is a step towards the construction of a full scale setup to study two aspects of accelerator driven systems (ADS) at the Synchrophasotron/Nucleotron complex at the Laboratory of High Energies, Joint Institute for Nuclear Research (JINR), Dubna, Russia: The energy production and transmutation of radioactive waste using relativistic protons. Some experimental results are presented obtained with calorimetric, radiochemical and solid state nuclear track detector (SSNTD) techniques.


1 Introduction

Within the framework of world-wide attempts to solve the problems of CO2-free generation of energy using nuclear techniques together with simultaneous attempts to solve the problem of long-lived radioactive waste from conventional nuclear power plants the following study was undertaken: A large scale research program was initiated by the Laboratory of High Energies (LHE) within the Joint Institute for Nuclear Research (JINR) in Dubna, Russia, and named “Investigation of physical aspects of energy production and study of radioactive waste transmutation using relativistic beams at the Synchrophasotron/Nucleotron accelerator complex of the LHE”. Within this program, a more limited program has actually been started and called “Energy plus Transmutation”. Its principle features are presented in Figs. 1 and 2 and the underlying research program can be summarized as follows (details are given in Refs. [1, 2]):

- The major components of this system include the accelerators, the lead target, and the natural uranium blanket without any low-Z moderator.

![Fig. 1. Schematics of the installation “Energy plus Transmutation”](image-url)
The uranium fission calimeter includes the following detector systems on relativistic beams from the JINR synchrophasotron/nucleotron:

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Methods of heat measurements:
- microthermocouples, quartz resonator, platinum sensors and detectors of infrared radiation
- distribution of heat generation over the volume of the uranium blanket and lead converter-target
- specific heat generation in the uranium blanket

Neutron spectrometers (Berger spheres, activation detectors):
- specific neutron yields
- neutron energy spectrum
- space distribution of neutrons in the uranium blanket

![Fig. 2. Program for the experimental studies within the frame of the project “Energy plus Transmutation” [1, 2]](image)

- A solid metal construction is supporting the massive target and blanket structures.
- Various detection systems for beam monitoring, dosimetric controls and radiation safety are included.
- Several electronic devices for radiation detection, such as neutron- and gamma-spectrometers are employed, simultaneously with SSNTD (solid state nuclear track detectors) for fission fragment registration. These sensors are used to study the reaction products from the interaction of relativistic primary protons and their secondary particles within the lead target and the surrounding uranium blanket.
- Auxiliary systems for experimental automation, data acquisition and data processing are described additionally.

A multitude of experiments has been carried out at the 660 MeV Synchrocyclotron and the GeV-range Synchrophasotron at the JINR during the last decades using extended targets of medium or heavy elements, such as iron, copper, tungsten, lead, bismuth, or uranium [3-10]. Nuclear processes have been studied in these targets. The present installation has a more complex structure: The primary proton beam with an energy of typically 1.5 GeV is interacting essentially only within the massive lead target producing spallation neutron of energy in the range of some MeV. These neutrons leave the lead target and interact directly - almost without any moderation - with the blanket. This blanket contains uranium, but one could also employ thorium or arbitrary mixtures of fissile materials within the blanket. The main reason for the introduction of the uranium blanket into the assembly is the desire to increase significantly the total neutron fluence during the proton irradiation as well as to amplify significantly the energy production due to neutron induced nuclear fission within the blanket. In this manner the present installation differs from previous experiments using extended targets that are mostly aimed at the study of generation of spallation neutrons.

The first experiment at the LHE using the new set-up is described here. 1.5 GeV protons from the Synchrophasotron were used as projectiles to study physical aspects of the electronuclear method of energy production. Some first results will also be presented.

2 Experimental techniques

The schematic presentation of the experimental set-up (scale 1:30) is shown in Fig. 3 with its central lead target surrounded by the uranium blanket and shown together with the positions of the activation-foil targets, SSNTDs and other detectors. The uranium blanket consisted of two sections, each containing 30 natural uranium rods, the total weight of natural uranium was 103.2 kg. The positions of the thermocouples and thermocouples for calorimetric measurements are also indicated.

The direction and center of the 1.5 GeV proton beam is along the horizontal symmetry axis of the entire system. The beam intensity in each pulse was up to \(1 \times 10^{23}\) protons. Each pulse lasted 0.35 s, the pulse repetition rate was 0.9 s. The use of the \(\alpha\)-activation foil for fluence determinations together with SSNTDs for measuring the beam profile has already been tested and described in earlier reports [6-11].The proton beam was properly centered onto the Pb-target at the beginning of the irradiation using sensitive Polaroid films and ionization chambers.

The biological shielding is of major importance during this irradiation. Extensive neutron and gamma fluxes are generated around this target set-up. Therefore, the container used in a previous experiment [8, 9] was modernized and is shown in its present form in Fig. 4. The entire uranium-lead assembly is placed into the center of the container which holds also the moderator-shielding, consisting of granulated polyethylene with boron carbide, lead walls and plates of cadmium. The cadmium absorbers in the internal walls of the container towards the U/Pb assembly significantly reduce the number of thermal neutrons reaching the inner volume. The container has dimensions of \((100 \times 106 \times 111)\) cm and a total weight of
about 950 kg. It is placed on top of a mobile platform which is moved over rails from a closed storage into the irradiation position “F3” in the Experimental Hall of the LHE.

3 Uranium-lead assembly

The detailed technical design of this lead target/uranium blanket assembly (Fig. 3) was carried out by the All-Russian Institute of Nuclear Energy Machine Building (VNIAEAM) in Moscow. The manufacturing of the steel structure was performed at the mechanical workshop of the LHE. The details on parts of the uranium-lead assembly are described in the following:

- **The beam monitoring systems** including activation foils and SSNTDs has already been described in Ref. [11].
- **The multi-section lead target** is constructed from lead disks having 8.4 cm diameter, a target length of 52 cm and total weight of 43 kg.
- **The two sections of the uranium blanket** are manufactured from uranium rods with 3.6 cm diameter and 10.4 cm length, each having a weight of 1.72 kg. They are made of natural uranium hermetically sealed into aluminum cladding. Each of the two sections contains 30 such uranium rods with a weight of 51.6 kg per section. The steel container (Fig. 5) fixes rigidly the position of each rod. This is essential for providing reliable contact of the rod surfaces to thermometric sensors placed at the front and back ends of the rods.

![Fig. 4. Technical drawing of the container for the Pb-target and the U-blanket together with the biological shielding as placed on a mobile platform: (a) side view, (b) front view. Details are given in the text.](image-url)
The three fiberglass plates are positioned in front, in the center and at the end of the assembly as shown in Fig. 3. Their dimensions are 30 cm × 30 cm × 0.3 cm. These plates are used to mount the activation foils and SSNTDs. The total number of such sensors varies between 30 up to 60 per plate. Further details are given in Section 4.1.

The thermometric sensors are placed outside and inside of each section of the lead targets, as well as on the side and end surfaces of uranium rods in the two uranium blanket sections. They perform on-line measurements of the time-space distribution of the temperature. This allows the determination of the heat generation inside the uranium blanket and inside the lead target during the proton irradiations. Further details are given in Section 4.2.

This method of simultaneous use of activation foils and SSNTDs during the determination of calorimetric properties in electronuclear studies using relativistic protons is referred to as activation tomography and fragment radiography [8, 9].

4 Measurement of nuclear reactions within the U-blanket and Pb-target

The energy production due to fission reactions within the uranium blanket, just as the neutron production due to spallation within the Pb-target, is caused by nuclear interactions. A large variety of experimental techniques are employed to investigate these phenomena. The essential features of the experimental techniques together with the diagnostic and data handling methods are described shortly [12–15]:

4.1 Activation sensors, foils and SSNTDs

A typical example for the location of activation sensors and SSNTDs on fiberglass plates is shown in Fig. 6 and described in detail below:

- SSNTDs of different kinds are employed as strips covered with uranium foils. The uranium is either depleted, natural or 30% enriched in 235U. As shown in Fig. 6, detectors in directions A, B, F, and H are unshielded while those in directions C, D, I, and J are covered with cadmium filters, to exclude thermal neutrons entering those detectors. The choice of angles allows the mounting of a large number of detectors in an axially symmetric manner. Some information about the neutron spectrum can be obtained with this set-up [16].

- Uranium activation sensors in the form of metallic natural uranium pellets with 8 mm diameter, 1 mm thickness and about 0.9 g weight are placed in the direction K at intervals of 1 cm starting from the centre of the symmetry axis and extending up to 16 cm. The aim is to study (n, γ), (n, 2n), and (n, f) reactions in 235U using gamma spectrometry. Particular attention is given to the study of the (n, γ) reaction as it yields via 239Np formation the important nuclear fuel 239Pu. Other sets of detectors could also be placed in analogous positions later.

- Many other nuclear interactions using the radiochemical activation methods were studied additionally. The following 23 targets are positioned in a circular arrangement at a distance of 75 mm from the center (Fig. 6): C, F, Mg, Si, S, Al, Ti, Mn, Fe, Co, Ni, Cu, Zn, Mo, Rh, Cd, In, Au, Hg, Pb, Bi, and Np. The gamma activity of these samples were measured after the irradiation using HPGe detectors with good energy resolution and a large counting efficiency. Results of these measurements will be presented in a future publication.

The SSNTD method was used for the detailed investigation of the neutron spectrum in the center of the U/Pb assembly via the measurements of fission rates relative to 235U for
The positions of the highly sensitive thermocouples are shown in Fig. 7. The sensors are positioned so as to extract maximum information on the heat generation in the lead target and uranium blanket. This technique has been developed at the VNIIAM (Moscow) and has been described in detail in our earlier report [9]. The system allows the determination of temperature changes with an accuracy of 0.17–0.03 degrees.

Special highly-sensitive semiconductor thermoresistors were tested in the present experiment, they appear to be particularly useful for low beam power irradiations.

Further technical details about all the different techniques described will be presented in a forthcoming paper giving also the experimental results together with their interpretation.

5 Preliminary experimental results

Some preliminary experimental results obtained after the first proton irradiation in the year 1990 with an energy of 1.5 GeV are given below.

5.1 Radiochemical results from uranium sensors

A large number of radiochemical sensors were irradiated with secondary neutrons in the target-blanket system. As mentioned earlier, uranium sensors were placed along the radial direction K (Fig. 6). The decay of the induced gamma activity was measured with standard counting and data handling systems. First results on relative amounts of three typical product nuclei identified through their gamma activities are presented in Fig. 8. The results are presented as intensity in units of ("relative activity" "radial distance from central axis").

\[
^{209}\text{Bi}, ^{232}\text{Th}, ^{233}\text{U}, ^{234}\text{U}, ^{238}\text{U}, \text{ and } ^{239}\text{Np} \text{ isotopes. All measured fission reactions – except for } ^{233}\text{U} \text{ and } ^{238}\text{U} – \text{ have a finite energy threshold. Lavsan- } \text{ and artificial mica-SSNTD foils with fissogenic material sandwiched between them were employed in the standard manner. These targets were placed at the radial positions along } G (225^\circ) \text{ and } E (135^\circ). \]

4.2 Measurement of thermal heat

One of the two major goals of the experiment is to study the thermal heat production within a subcritical system. The energy gain G in this subcritical system is defined as follows:

\[
G = \frac{P_{\text{neut}}}{P_{\text{beam}}} = \frac{E_{\text{neut}} \gamma_{\text{e}} k_{\text{eff}}}{E_{\text{p}} \gamma_{\text{i}} (1 - k_{\text{eff}})}
\]

G is the ratio of the power generated in the uranium blanket due to neutron induced fission within (\(P_{\text{neut}}\)) to the power introduced into the lead target by the proton beam (\(P_{\text{beam}} = 1 - E_{\text{p}}\)). \(E_{\text{neut}}\) is the proton beam intensity of energy \(E_{\text{p}}\), \(E_{\text{neut}}\) is the number of spallation neutrons produced in the lead target per incoming proton, \(k_{\text{eff}}\) is the number of neutrons emitted per fission, \(E_{\text{neut}}\) is the energy released in fission and \(k_{\text{eff}}\) is the effective neutron multiplication factor of this subcritical nuclear system. Both methods were used to measure the temperature fields in the Pb-target and in the U-blanket: (a) Thermocouples and (b) semiconductor resistors.
5.2 Calculated neutron spectrum within the blanket

The Dubna Cascade Model in its version DCM-CEM [17] was employed to calculate the neutron spectrum at different positions of the target system. Three geometrical angles along the beam direction were chosen for this calculation at a circle in 7.5 cm distance from the central axis. The dependence of the neutron spectra on the angle $\theta$ (see schematic insert in Fig. 9), in the upstream position of the first section of the blanket, was calculated for three values of the traversed distance $I$. The calculated neutron spectra are shown in Fig. 9 in absolute terms of n/cm$^2$/proton. One observes in this set-up an exceedingly hard neutron spectrum as practically no low-Z moderating materials have been included.

5.3 Distributions of $^{235}$U fission densities

Distributions of $^{235}$U fission densities along the perpendicular radius of the U/Pb assembly are presented in Fig. 10. Fission rates decrease with the radial distance from the central axis, as already known from other experiments under different experimental conditions [7].

5.4 Thermometric measurements

The first results indicating a definite increase in temperature within the entire target set-up during an irradiation with 1.5 GeV protons with an intensity up to $10^{10}$ protons/second are shown in Fig. 11: Before the irradiation the temperature in the system was constant at 13.0°C, small changes in the environmental temperature have been taken care of. When the target is irradiated the temperature increases up to 0.8°C during about 40 min depending on the location of the thermometer. All thermometers have an "identification number", which is given in Fig. 7 together with their exact geometrical position. Thermometers "24" and "25" which are placed in front of the target into the proton beam, showed all the time the same environmental temperature, thus indicating that they work properly in the high radiation field of the direct beam.

Further publications will give a more complete and also quantitative analysis of the results of this experiment.
6 Conclusions

The first experiments with the model U/Pb are described, and technical arrangements are presented. This is an important step in our efforts to construct a full scale subcritical installation at the Laboratory of High Energies, JINR, to study the aspects of electronuclear methods for the energy production and transmutation of radioactive wastes using relativistic beams from the JINR Synchrophasotron/Nuclotron. We called the project "Energy plus Transmutation". The first qualitative results obtained during 1999 using this 1.5 GeV proton irradiation are presented. A set of radiochemical activation sensors, SSNITDs and thermometric sensors have been employed. All individual parts of the complex experimental system worked according to expectation regarding their reliability, accuracy and effectiveness. The future analysis of this experiment shall give quantitative results on

- the energy gain and neutron multiplication,
- the values for the effective neutron fission and neutron capture integrals in natural uranium,
- neutron spectra and energy balance in the volume of the lead target and uranium blanket.

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