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EXPERIMENTAL STUDY OF TRANSMUTATION OF SELECTED RADIOACTIVE WASTE USING THE BEAMS OF RELATIVISTIC PROTONS

Speciality: 01.04.16 — Physics of Atomic Nucleus
and Elementary Particles

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Status of the Thesis

Since the end of 1945 the countries, which started to build up nuclear technologies have generated large amounts of nuclear wastes. Highly radioactive spent fuel elements are continuously being unloaded from power reactors, typically 25-30 ton per year from a 1000 MW light water reactors, which is the most widely used reactor type. Now there are about 400 commercial nuclear power plants in operation world wide including ~120 GW nuclear electric capacity operational in Western Europe and 45 GW operational in the ex-USSR and East European countries. Although nuclear program have been slowing down throughout the world, spent fuels are continuously stored, mainly in reactor pools, waiting for reprocessing or for final deep-underground disposal. According to OECD/NEA, by the end of last century (1992) one expects about 150000 tons of spent fuel unloaded from the commercial reactors through out the QECD countries.

When the nuclear fuel is loaded in usual light water reactors, it contains freshly enriched uranium at around 3.5% in ^{235}U. Spent fuel contains almost all the radioactivity produced inside the reactor by fission, neutron capture and radioactive decay modes. This radioactivity is characterized by long-lived, highly active and highly radiotoxic nuclides. The most important fission fragments, fissile material and long-lived actinides are given in Ref. [1].

These amounts of radioactive wastes make very serious problems both from economic and ecological points of view. Since spent fuel elements contain very long-lived isotopes they must be stored at least for 10^5 years by isolating them physically from the biosphere using successive barriers at a suitable depth in the ground before they decay to the level of natural uranium ore. There are two parallel ways to reduce these amounts of radioactive wastes. The first one is the transmutation or the transformation of radioactive LLFP such as ^{99}Tc, ^{129}I and ^{135}Cs by neutron capture and following β decay into stable or short-lived ones. The second methods is incineration or burning up of transuranic nuclides by means of nuclear fission induced in nuclear reactors causing energy release and emission of secondary neutrons. At present time one can consider rather commonly accepted idea of complex solution of the following mutually related problems: ecologically safe and commercially competitive production of energy with the simultaneous transmutation and incineration of created radioactive waste using so called accelerator driven subcritical reactors. Now one can consider that the best way for transmutation of such hazardous material to stable or short-lived nuclides needs to employ intense and highly intensive thermal neutron fluxes (~10^{16} n/cm^{2}s) and for this purpose a beam of high-energy particles interacting with extended heavy targets (Pb, U, Bi or other compositions) should be used [H3]. During the last decade intense investigations of the radiation waste transmutation (RWT) are conducted at many

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[1] J.P. Schapira, NIM A280 (1989) 568-582

laboratories and first reliable estimation have been obtained for the yield of some concrete processes for the major long-lived nuclides: ^{129}I, ^{237}Np and ^{239}Pu (for example experiments carried out at the LHE, JINR, Dubna, using Synchrophasotron or Nuclotron accelerators with the small scale relatively inexpensive experimental set-up Gamma-2. The present work is aimed at further continuation of this experimental approach and contains the results of the investigation of Gamma-2 set-up and transmutation of long-lived radioactive isotopes ^{129}I, ^{237}Np and ^{239}Pu using spallation neutrons generated in massive targets as a result of interaction between target material and relativistic proton beams at different energies.

The experimental production rate (B-value) is defined as the total number of atoms formed during the irradiation of one gram of sample for one incident particle. It is used in this work as well as in [2] to estimate the distribution of the neutron flux in the target-moderator volume.

$$B(^{A}X) = \frac{Number\ \ of\ \ atoms\ \ formed\ \ ^{A}X}{(1gram\ \ of\ \ samples)\ * \ (one\ \ bombarding\ \ ion)} \tag{1}$$

B-value is a strictly empirical value, valid only for a given set-up, target, projectile, its energy and the exact position of the target.

Main Results Obtained in This Research

1-Investigation of the experimental set-up Gamma-2:

a-With monitor system and distribution of ^{139}La and uranium sensors for the experiments carried out at Nov.1999.

b-With monitor system and distribution of ^{139}La sensors at different depth for the experiments carried out at the Nov.2001.

c-With transmutation sample positions for both experiment (Nov.1999, Nov.2001).

2-Determination of beam position and size for both irradiations.

3-Determination of proton fluence using activation methods (^{27}Al).

4-Determination of the experimental production rate for different sensors and transmutation samples.

5- Distribution of the experimental production rate using radiochemical sensors.

6- Transmutation rate for some long-lived radioactive isotopes.

Scientific Novelty

1-Transmutation of fissile material ^{239}Pu was done for the first time using spallation neutrons generated in lead target by proton beams at energies 0.53 and 1.0 GeV

2-New energies were used to study the transmutation of ^{129}I and ^{237}Np using Pb target at proton energy 1.0 GeV and U/Pb target at proton energies 0.53 and 1.0 GeV.

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[2] J.-S. Wan et al., Kerntechnik Vol. 63(1998) 167-177

3-Investigation of the paraffin thickness on B-Values using ^{139}La sensors irradiated by spallation neutrons generated in Pb target by proton beams at 0.65, 1.0 and 1.5 GeV.

4-Determination of the experimental production rate B(^{140}La) for ^{139}La sensors irradiated by spallation neutrons generated in Pb and U/Pb targets by proton beams at energies 0.53 and 1.0 GeV:

a-Radial distribution for the experimental production rates.

b-Azimuthal distribution for the experimental production rates.

c-Distribution of the experimental production rates on the top surface of paraffin.

5-Determination of (B-values) for uranium fission fragments produced by spallation neutrons generated in Pb and U/Pb targets by proton beams at energies 0.53 and 1.0 GeV.

The Aim of the Research

1-Investigation of the experimental production rates B(^{140}La) for La sensors irradiated by spallation neutrons generated in Pb and U/Pb targets on beams at energies 0.53 and 1.0 GeV:

I-Distribution of B(^{140}La) at different positions on the top surface of paraffin.

II-Azimuthal distribution for the experimental production rates on top surface of paraffin moderator at 15 cm from the front of paraffin moderator.

III-Radial distribution of spallation neutron at different radii inside the paraffin moderator (4.5, 6.5, 8.5 and 9.7 cm measured from the center of the target).

2-Investigation of the impact of moderator thickness on B(^{140}La) for ^{139}La irradiated by spallation neutrons generated in Pb target at proton energies 0.65, 1.0 and 1.5 GeV.

3-Modeling calculation using Monte Carlo simulation on the effect of the moderator thickness on production rates for ^{139}La irradiated by spallation neutrons generated in Pb target by proton beams at energies 0.65, 1.0 and 1.5 GeV.

4-Modeling calculations of neutron spectra generated in targets by proton beams at energies 0.65, 1.0 and 1.5 GeV at different thickness using Monte Carlo simulation with DCM-CEM code.

5-Transmutation studies for some long-lived radioactive isotopes using spallation neutrons generated in massive targets at different proton energies:

I-Transmutation of ^{129}I and ^{237}Np:

A-At proton energies 0.5 and 1.0 GeV with U/Pb targets.

B-At proton energies 1.0 GeV with Pb targets.

II-Transmutation of ^{239}Pu using spallation neutrons generated in Pb target by relativistic protons at energies 0.53 and 1.0 GeV

6-Determination of the neutron production per unit energy of the incoming proton for each sample

$$R = \frac{B - value}{Proton\ energy\ Ep} \qquad (2)$$

Practical value of the research is determined by the possible use of the results in:

-development of nuclear data libraries necessary to develop prototype electronuclear installations and nuclear waste transmuters;
- to check and modernize computer programs used to calculate nuclear processes and parameters of experimental installations;
- to prepare future experiments on transmutation using beams from high energy accelerators.

Publications

There are 6 articles published basing on the results of this work.

Conferences and seminars

1). 5th Conference for Young Scientist and Specialists, (4-10 Feb., 2001, Dubna, Russia)
2). XVI International Baldin Seminar on High Energy Physics Problems (10-15, June 2002, Dubna, Russia).
3). VII International School-Seminar for Heavy Ion Physics, (May 27-June 1, 2002, Dubna, Russia).
4). 3rd Conference on Nuclear and Particle Physics (20-24 Oct., 2001, Cairo, Egypt)

Contents of the Thesis

Chapter 1: contains introduction to the problems related to the topic and overview of the previous research activities as well as the purpose of the present work. Main experiments on nuclear waste transmutation carried out elsewhere are discussed. Place of the JINR research activities in this field is shown. Basic data to be obtained in this research are listed.

Chapter 2: In this chapter the Experimental set-up Gamma-2 is explained in details. This set-up consists of a massive target such as Pb and U/Pb surrounded by 6 cm paraffin CH_2 as a moderator [H1]

I-Lead target: The experimental set-up for Pb target consists of 20 natural Pb disks each of them has 8 cm diameter and 1 cm thickness.

II-The Uranium-Lead target: Experimental set-up for U/Pb target consists of two U rods each of them with 3.6 cm diameter and 10.4 cm length, uranium rods were placed inside 21 Pb rings each of them has 1cm thickness, 3.6 cm as inner diameter and 8 cm as outer diameter Activation technique was used to determine the integral proton flux and Polaroid films were used to define the position and size of a beam. These last issues are of crucial importance in determination of the experimental production rates (B-values).

Chapter 2 is divided into sections. In section A, four experiments performed in Nov. 1999 are discussed. In these experiments two sets of monitors were used to measure the proton fluence. Each set of monitor was composed of high purity Al foil sandwiched between two other Al foils with the same diameter. The first group of monitors (monitor 1) was placed in close contact with the target and the second group (monitor 2) was placed at 35 cm upstream from the target [H1]. In section B, three experiments carried out in Nov.2001 are discussed. In these experiments only one group of ^{27}Al monitor of 20 cm diameter was placed at 85 cm [H2]. ^{27}Al monitor was divided into several rings then activity of two rings with 8 cm diameter was determined.

In order to study the distribution of spallation neutrons (which is approximately proportional to the distribution of the experimental production rates) at different targets and different proton energies, radiochemical sensors were used.

In Nov. 1999 experiments ^{139}La and ^{nat}U sensors were used to investigate surface, radial and azimuthal distributions of B-values (spallation neutrons) [H1].

I-Natural Uranium Sensors: Five U sensors were arranged in one row at the surface of paraffin moderator with an angle 20° within the vertical plane.

II-^{139}La Sensors:

A- Five ^{139}La sensors Nos. (1, 2, 3, 4, 5) were placed at the surface of paraffin moderator normal to the horizontal plan.

B- Four ^{139}La sensors Nos. (16, 17, 18, 3) were placed inside paraffin moderators at different radii (4.5, 6.5, 8.5 and 9.7 cm) at 15 cm from the front of paraffin

C- Six La sensors Nos. (3, 6, 7, 8, 9, 10) were installed around the paraffin at 15 cm from the front of paraffin, with angle 60° between them. Fig.1 shows vertical view for the Gamma-2 set-up with sensor positions.

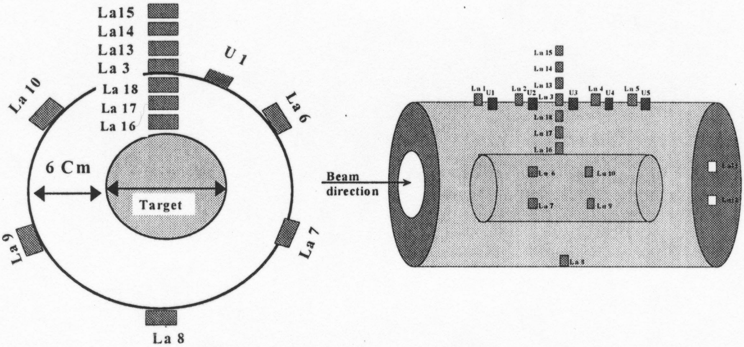

Fig. 1 Vertical veiw (side and front)for the experimental set-up Gamma-2 with U and La sensors for experiments carried out in (Nov. 1999)

During the 2001 experiments new arrangement for ^{139}La sensors was employed to investigate the effect of the moderator thickness on neutron yield in the lead target at different proton energies [H2, H3].

I- Ten La sensors were installed at the surface of the moderator:

A-Five ^{139}La sensors Nos. (1, 2, 3, 4, 5) installed at depth 0 mm.

B-Five ^{139}La sensors Nos. (6, 7, 8, 9, 10) divided into two groups, the first group consists of 3 sensors Nos. (6, 8, 10) placed at depth about 10 mm, and the second group consists of two samples Nos. (7, 9) placed at depth about 20 mm.

One ^{139}La sample No. 11 was encapsulated in a plastic container with inner diameter 21 mm. Experimental set-up Gamma-2 was employed to transmute radioactive wastes ^{129}I, ^{237}Np and fissile material ^{239}Pu. Each sample was encapsulated in Al container and placed together with La sample No. 11 at the surface of the paraffin moderator between 10 and 15 cm from the front plate of paraffin moderator, as it is shown in Fig.2 [H1].

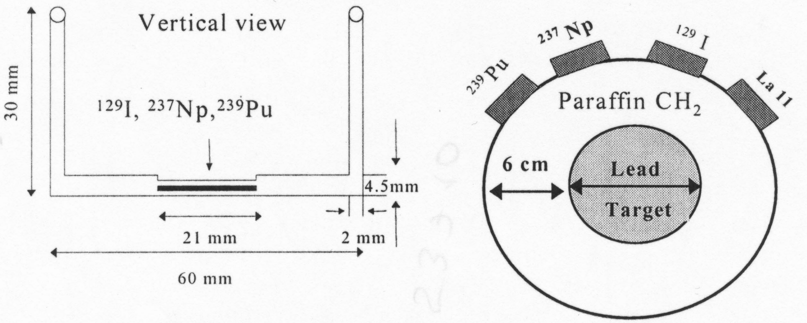

Fig. 2 Vertical view of the Al container used for ^{239}Pu, ^{237}Np and ^{129}I and scheme of the distribution of transmutation samples

In Chapter 3 the following points are discussed:

1-Determination of the integral proton fluence by means of activation foils:

Foil activation (^{27}Al) is a well-known technique used to measure the fluence of the high-energy proton beam. ^{27}Al foils interact with relativistic protons via the following reactions [3]:, ^{27}Al(p, 3pn)^{24}Na, ^{27}Al(p, x)^{22}Na and ^{27}Al(p, x)^{7}Be. These three reactions have high cross section for interaction with protons, and the interaction products ^{24}Na, ^{7}Be and ^{22}Na have acceptable half-life's and high intensity gamma ray lines. Al foils were installed in front of target. After the irradiation of activation foils (^{27}Al), they were taken out and analyzed with gamma detectors. Gamma rays due to the ^{24}Na, ^{22}Na and ^{7}Be were measured using HPGe detector system for sufficient time to obtain spectra with good statistics. Then the integral proton fluence was determined.

2-Interaction of spallation neutrons with ^{139}La, $^{nat.}$U, ^{129}I, ^{238}Np and ^{239}Pu:

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[3] J.S.Wan et al., NIM, B155 (1999) 110 and JINR preprint E13-99-189, Dubna (1999)

In the present work two types of radiochemical sensors (natural uranium and stable ^{139}La) were used to measure the space distribution of spallation neutrons around the paraffin moderator. Stable ^{139}La interacts with neutrons through the capture of neutrons followed by β^- decay to ^{140}Ce (stable), while natural uranium interacts with spallation neutrons through the (n, γ), (n, 2n) and (n, f) reactions according to the energy of incident neutrons. Three of the most important long-lived radioactive isotopes were investigated in this work (^{129}I, ^{237}Np, ^{239}Pu).

I-^{129}I interacts with spallation neutrons through the capture of neutrons, transforming into the short-lived isotopes ^{130}I following by β^- decay to stable isotope ^{130}Xe.

II-The second radioactive waste material is ^{237}Np, which transmutes into ^{238}Np.

III- Fissile isotope ^{239}Pu was transmuted through fission process activated by spallation neutron generated in massive target.

3-Determination of the experimental transmutation (production) rate: After the end of irradiation each sample was measured several times during the period of approximately one week or ten days. Then the Experimental Production Rates ("B-value") were determined, (see Equation 1 page 2).

Chapter 4 contains the results of the experiments carried out during Nov. 1999 and Nov. 2001.

1-Gamma Ray Measurements: Several gamma ray detectors were used to measure gamma spectra from the irradiated samples. Each sample was measured several times through the period of ten days. The investigation of transmutation of long-lived radioactive isotopes including ^{239}Pu ^{129}I and ^{237}Np was performed by the following manner: they were irradiated together on top of the Gamma-2 set-up then the activity of each sample was measured simultaneously using three HPGe detectors with different filters.

2-Detection Efficiency: The efficiency curve for each detector was measured using standard calibration source ^{152}Eu under the same conditions as those used during the measurement of the samples.

3-Determination of Beam Position and Size: Before each experiment Polaroid films were used to determine experimentally the centre of the proton beam with respect to the target. They were irradiated with one pulse of particle beam.

4- Determination of Integral Proton Fluence using Activation Foils: Integral proton fluxes were determined using reaction ^{27}Al(p, 3pn) ^{24}Na for each experiment.

5-Experimental Production Rates (B-values) for ^{140}La, U fission fragments and ^{239}Np: In order to investigate spallation neutron distribution around the experimental set-up Gamma-2 at different positions, different targets and different proton energies, B-values for radiochemical sensors were determined.

I- Distribution of B-values at the surface of paraffin moderators using La sensors is shown in Fig.3.

II- Distribution of spallation neutrons at the surface of the paraffin moderator using natural uranium sensors: Uranium sensors were measured at least three times during a period of one week then half-life of each fragments was determined. The experimental production rates for fission fragments ^{97}Zr, ^{91}Sr, ^{133}I and ^{239}Np were determined at 5, 10, 15, 20 and 25 cm, measured from the front plate of the paraffin moderator. The results are given in Table 1a, b, c, d [H4, H6].

Table 1a B-value for ^{97}Zr at different positions at the surface of the paraffin moderator

Sensor Position Z(cm)	Ep=0.53 GeV (U/Pb)* 10^{-6}	Ep=0.53 GeV (Pb)* 10^{-6}	Ep=1.0 GeV (U/Pb)* 10^{-6}	Ep=1.0 GeV (Pb)* 10^{-6}
5	1.98±0.15	0.93±0.06	2.93±0.22	1.23±0.10
10	2.58±0.22	1.34±0.09	3.34±0.26	1.75±0.14
15	2.01±0.17	1.18±0.10	3.14±0.20	1.90±0.15
20	1.61±0.14	0.85±0.09	2.12±0.16	1.39±0.12
25	0.93±0.08	0.74±0.06	1.42±0.11	0.98±0.08

Table 1b B-value for ^{91}Sr at different positions at the surface of the paraffin moderator

Sensor Position Z(cm)	Ep=0.53 GeV (U/Pb)* 10^{-6}	Ep=0.53 GeV (Pb)* 10^{-6}	Ep=1.0 GeV (U/Pb)* 10^{-6}	Ep=1.0 GeV (Pb)* 10^{-6}
5	2.00±0.29	0.94±0.12	2.76±0.37	1.34±0.19
10	2.29±0.32	1.49±0.19	3.46±0.49	1.61±0.21
15	1.96±0.28	------	2.89±0.35	1.97±0.25
20	1.44±0.20	------	2.12±0.26	1.36±0.22
25	0.87±0.13	1.21±0.16	1.29±0.16	0.91±0.15

Table 1c B-value for ^{133}I at different positions at the surface of the paraffin moderator

Sensor Position Z(cm)	Ep=0.53 GeV (U/Pb)*10^{-6}	Ep=0.53 GeV (Pb)*10^{-6}	Ep=1.0 GeV (U/Pb)* 10^{-6}	Ep=1.0 GeV (Pb)* 10^{-6}
5	2.31±0.19	1.31±0.10	3.64±0.3	1.62±0.14
10	2.39±0.22	1.66±0.12	4.36±0.37	2.21±0.19
15	2.16±020	1.39±0.12	3.90±0.27	2.21±0.19
20	1.75±016	1.37±0.15	2.94±0.24	1.65±0.15
25	1.01±0.10	0.90±0.10	1.85±0.15	1.11±0.10

Table 1d B-value for ^{239}Np at different positions at the surface of the paraffin moderator

Sensor Position Z(cm)	Ep=0.53 GeV (U/Pb)*10^{-6}	Ep=0.53 GeV (Pb)*10^{-6}	Ep=1.0 GeV (U/Pb)*10^{-6}	Ep=1.0 GeV (Pb)*10^{-6}
5	6.15±0.72	3.42±0.39	10.70±1.40	4.57±0.61
10	7.58±1.02	4.69±0.53	13.40±1.76	6.43±0.85
15	6.31±0.86	4.22±0.55	11.50±1.28	6.59±0.87
20	4.65±0.63	3.16±0.51	8.66±1.12	5.00±0.68
25	2.36±0.32	1.72±0.20	4.57±0.59	3.05±0.42

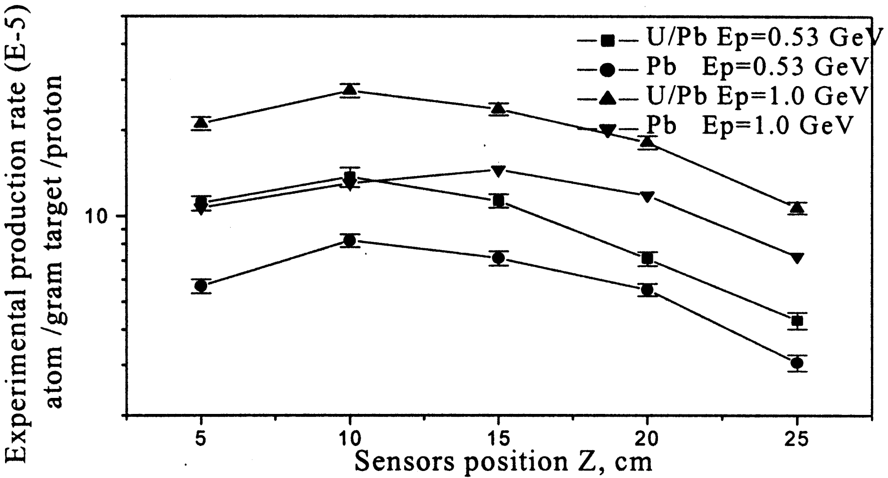

Fig.3. Experimental production rate for La sensors at different proton energies and different targets

III-Radial distributions of B-values inside the paraffin moderator were measured and the results are plotted in Fig.4 [H1].

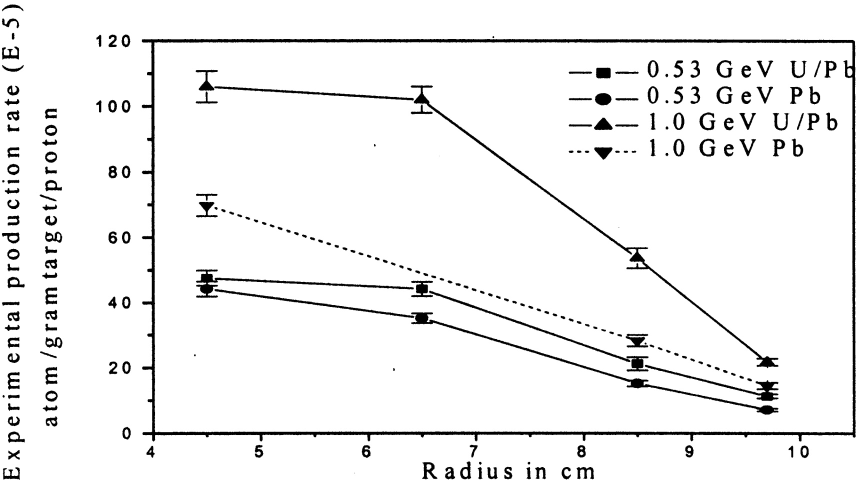

Fig.4 B(^{140}La) a function radius at proton energies 0.53 and 1.0 GeV with Pb and U/Pb targets

IV- Azimuthal distribution of spallation neutrons around the surface of the paraffin moderator was investigated by using six La sensors Nos. 3, 6, 7, 8, 9 and 10. The experimental results are given in the following Table.

Table 2 Azimuthal distributions for B(^{140}La) at the surface of the paraffin moderator at different proton energies and for different target material [H1]

Sensor Position	Ep=0.53 GeV (U/Pb)*10^{-5}	Ep=0.53 GeV (Pb) *10^{-5}	Ep=1.0 GeV (U/Pb) *10^{-5}	Ep=1.0 GeV (Pb) *10^{-5}
3 (0°)	11.3 ±0.60	7.11±0.40	21.7±1.06	14.4±0.98
6 (60 °)	10.5±4.0	9.08±0.43	23.9±2.34	12.7±1.07
7 (120°)	13.7±0.60	7.20±0.39	21.5±1.03	17.0±1.08
8 (180°)	14.3 ±0.82	10.6±0.46	34.9±2.58	18.9±0.87
9 (240°)	10.9±0.57	8.61±0.53	29.5±1.50	10.6±0.44
10 (300°)	12.5±0.50	7.46±0.38	21.9±1.05	13.6±0.63

V-In this section the effect of moderator thickness was investigated both theoretically and experimentally for lead target at proton energies 0.65, 1.0 and 1.5 GeV:

A-Experimental investigations were carried out with the help of ^{139}La sensors installed at different depths inside the paraffin moderator [H2].

B- Modeling calculations using Monte Carlo simulation with LAHET code[4].

The experimental production rates B(^{140}La) at proton energies 0.65 are tabulated in table.3.

Table 3 Experimental production rates for ^{139}La sensors at proton energy 0.65 GeV compared with theoretical calculations done by LAHET [H2]

Sensors positions (cm)		Experimental (B±ΔB)*10^{-5}	Calculations (B±ΔB)*10^{-5}
Depth in Paraffin	Position (Z)		
0	5.2	2.12±0.13	3.17 ± 0.24
0	10.15	3.16±0.20	4.83 ± 0.34
0	15	3.13±0.19	4.42 ± 0.25
0	19.8	2.55±0.20	3.57 ± 0.25
0	24.75	1.37±0.20	2.23 ± 0.21
0.79	5.2	4.44±0.28	4.96 ± 0.31
1.0	10	--------	9.93±0.60

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[4] R.E.Prael and Henry Lichtenstein, "User Guide to LCS: The LAHET Code System", Los Alamos National Laboratory, report LA-UR-89-3014 (Sept, 1989)

Table 3 continued

Sensors positions (cm)		Experimental $(B \pm \Delta B) * 10^{-5}$	Calculations $(B \pm \Delta B) * 10^{-5}$
Depth in Paraffin	Position (Z)		
0.88	15	7.86±0.50	10.11 ± 0.42
1.0	20	--------	7.88±0.48
0.85	24.75	2.70±0.17	4.52 ± 0.32
2.0	5	--------	16.73±1.17
1.44	10.15	14.7±0.9	15.52 ± 0.59
2.0	15	------	24.49±0.98
1.6	19.8	11.74±0.7	13.7 ± 0.56
2.0	25	--------	11.87±0.59

6- Modeling calculation of neutron spectra for Pb target with paraffin moderator using Monte Carlo simulations with DCM-CEM [H5]

Usually the rate at which radioactive materials are transmuted is rather dependent on the neutron spectrum. Some of the isotopes need to be transmuted in the field of thermalized neutrons, some of the isotopes need to use fast neutron field. Usually the "hardness" of the neutron spectrum depends on the space position of the sample inside the setup. In our case we could study experimentally the flux of thermalized neutrons in just 5 points. To analyze in detail the development of the neutron field inside the setup one therefore needs to undertake computer simulation of the nuclear processes taking place in the assembly. Neutron spectra were calculated using Monte Carlo simulation with DCM-CEM [5] for Pb target with paraffin moderator at proton energies 0.65, 1.0 and 1.5 GeV and different depth inside the paraffin moderator (0, 1 and 2 cm) which corresponds to the experimental conditions. Modeling calculations at proton energies 1.5 GeV at the surface of paraffin are plotted in Fig. 5 [H5].

It is seen from Fig.5 that the distribution of the neutron flux escaping the surface of the moderator along the beam line shows some flat maximum at about 10 – 15 cm. Maximal value exceeds the values of the neutron flux at the periphery by a factor of 1.5 – 2. The "hardness' of the spectrum does not vary much with the displacement along the beam line.

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[5] A.Polanski, A.N.Sosnin, V.D.Toneev. JINR Preprint E2-91-562, Dubna, 1991.

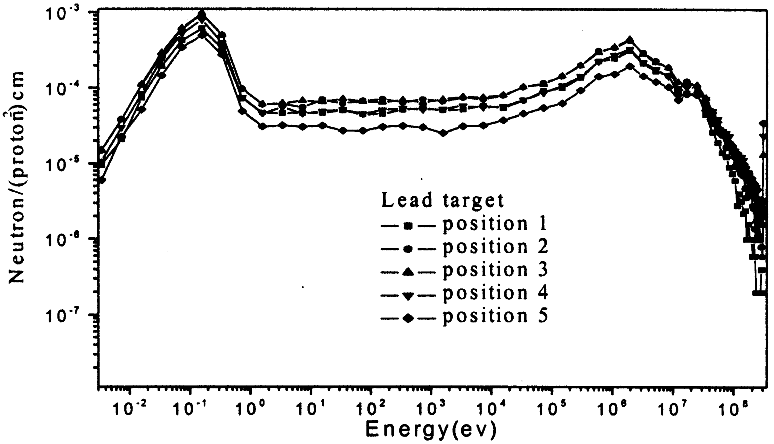

Fig. 5 Calculated neutron spectra at the surface of paraffin moderator with proton energy 1.5 GeV at different position

Transmutation of Fissile Material ^{239}Pu and Radioactive Wastes (^{129}I and ^{237}Np) Using Spallation Neutrons

I- Incineration studies of ^{239}Pu were carried out at the Synchrophasotron and Nuclotron at LHE, JINR, Dubna using proton beams with energies of 0.53 and 1.0 GeV with lead target and paraffin moderator. The transmutation of ^{239}Pu and the associated production of fission products ^{91}Sr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{129}Sb, ^{132}Te, ^{133}I, ^{135}I, and ^{143}Ce were studied in the present work. Half-life for each fragment was obtained and the experimental production rate for ^{239}Pu fragments B(^{239}Pu fragments) was determined. The results were plotted in Fig.6, then the experimental fission rate for ^{239}Pu was found using fission yield. The results are collected in Table 4 [H1].

II- Transmutation of ^{237}Np was investigated through the capture of neutrons ^{237}Np(n, γ) ^{238}Np, The experiments were carried out using Gamma-2 set-up with U/Pb and Pb targets at different proton energies. Gamma quanta were measured several times then half-life for ^{238}Np and B(^{238}Np) were determined. The results are tabulated in Table 4 [H1].

III-Transmutation of ^{129}I was studied through the capture of neutrons ^{129}I(n, γ) ^{130}I, using Gamma-2 set-up with U/Pb and Pb targets at different proton energies. Decay curve and B(^{130}I) were determined. The results are tabulated in Table 4 [H1].

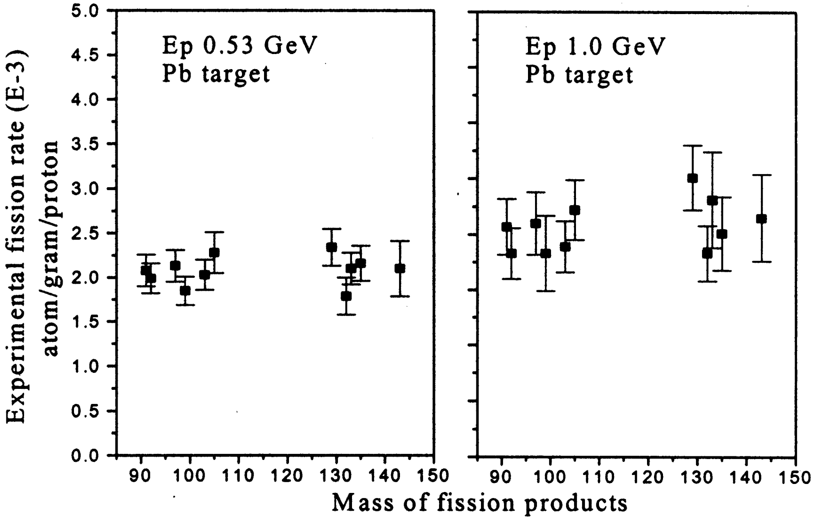

Fig. 6 Total experimental fission rates B(^{239}Pu) determined for different fission products at proton energies 0.53 GeV and 1.0 GeV with Pb target

Table 4 Experimental production rates B(^{238}Np), B(^{130}I) and total experimental fission rates B(^{239}Pu) [H1]

Experiment p+ target	B(^{238}Np) $*10^{-4}$	R (^{238}Np) $*10^{-4}$	B(^{130}I) $*10^{-4}$	R(^{130}I) $*10^{-4}$	B(^{239}Pu) $*10^{-3}$	R(^{239}Pu) $*10^{-3}$
0.5 GeV U/Pb	3.1	6.2	1.30	2.60	----	----
0.53 GeV Pb	----	----	----	----	2.08±0.20	3.9±0.4
1.0 GeV U/Pb	4.8	4.8	2.61	2.61	----	----
1.0 GeV Pb	3.3	3.3	0.81	0.81	3.07±0.31	3.1±0.4

Conclusions

In the present work several parameters for the experimental set-up Gamma-2 were investigated using radiochemical sensors (^{139}La and natural uranium). These parameters included the influence of different target materials at different proton energies. The experimental production rates for ^{140}La and uranium fission fragments (^{97}Zr, ^{133}I, ^{91}Sr) and ^{239}Np were determined for different targets, positions on the target and proton energies. Simultaneously with these investigations the transmutation of ^{129}I, ^{237}Np and ^{239}Pu was carried out using Pb and U/Pb targets irradiated by proton beams at energies 0.53 and 1.0 GeV. The results are summarized in the following points.

I- Two target materials were investigated at proton energies 0.53 and 1.0 GeV through the determination of the experimental production rates B(^{140}La) from ^{139}La target and B(^{97}Zr), B(^{133}I), B(^{91}Sr) and B(^{239}Np) from $^{nat.}$U targets. From the analysis of the experimental data we can find:

a) In case of surface detectors (five ^{139}La sensors and five uranium sensors, see fig.1.) the experimental production rates (B-values) have larger values in U/Pb target as compared to Pb target by about 60% to 70% at a given energy.

b) At a given energy and target, in the case of surface detectors the experimental production rates reach maximum values between 10 and 15 cm from the front plate of paraffin moderator.

c) For a given target, the experimental production rates B(^{140}La) increases with increasing proton energy. The experimental production rates has (100%±10%) larger values at proton energy 1.0 GeV compared to 0.53 GeV for U/Pb and Pb target.

II- La sensors were used to investigate the effect of depth inside the paraffin moderator at different proton energies with U/Pb and Pb targets. We used two experimental set-ups:

1) ^{139}La sensors were installed at different radii (4.5, 6.5, 8.5 and 9.7 cm) inside the paraffin moderators at 15 cm from the front plate of paraffin moderator for U/Pb and Pb targets at proton energies 0.53 and 1.0 GeV

2) Ten La sensors were installed at about 0, 10 and 20 mm (see table.6.) inside the paraffin moderator. Spallation neutrons generated in Pb targets by proton beams with energies 0.65, 1.0 and 1.5 GeV activated the La sensors. We observed:

The B(^{140}La)-values at a given geometrical position increase strongly with the depth of the hole, into which the La-sensor was placed. This is a very important result as it demonstrates that the corresponding low-energy neutron fluence, which is essentially producing ^{140}La, increases strongly when one goes deeper into the paraffin:

a-At all proton energies B(^{140}La) has 400%-600% larger values at depth 45 mm compared to the surface of paraffin moderators.

b-At all proton energies B(^{140}La) has 300%-360% larger values at depth about 20 mm compared to the surface of paraffin moderators.

c-At all proton energies B(^{140}La) has 100%-200% larger values at depth about 10 mm compared to the surface of paraffin moderators.

III-Modeling calculations using Monte Carlo with LAHET (see footnote in page 10) were carried out at the same depth (See II-2) and the same proton energies with lead target. The results show that:

The agreement between experimental and calculated B(^{140}La)-values is quite satisfactory. It is interesting to note that at all proton energies the calculated values appear to be a little larger, as compared to the experiments.

IV-Transmutation of long-lived radioactive waste ^{129}I and ^{237}Np and fissile material ^{239}Pu:

1-^{129}I and ^{237}Np were transmuted using secondary neutrons generated by interaction of proton beams at energies 0.53, 1.0 GeV with U/Pb target and 1.0 GeV with Pb target:
a- During the irradiation of ^{129}I the value for B(^{130}I) was measured to be $1.30*10^{-4}$ (atom /gram target/proton) and $2.61*10^{-4}$ (atom /gram target/proton) at proton energies 0.5 and 1.0 GeV using U/Pb target and $0.81*10^{-4}$ (atom /gram target/proton) at proton energy 1.0 GeV using Pb target.
b- During the irradiation of ^{237}Np the value B(^{238}Np) was measured to be $3.1*10^{-4}$ (atom /gram target/proton), $4.8*10^{-4}$ (atom /gram target/proton) at proton energies 0.5 and 1.0 GeV using U/Pb target and $3.3*10^{-4}$ (atom /gram target/proton) at proton energy 1.0 GeV using Pb target.
2-The transmutation of ^{239}Pu was carried out by spallation neutrons generated in Pb target interacting with proton beams at energies 0.53 and 1.0 GeV. The experimental production rates for several individual ^{239}Pu fission products were determined, then total fission rates B(^{239}Pu) were calculated with the help of well-known fission yield. The fission rates were $2.08*10^{-3}$ (atom /gram target/proton) and $3.07*10^{-3}$ (atom /gram target/proton) at proton energies 0.53 and 1.0 GeV respectively.

From table 4 one can calculate that the macroscopic transmutation rates for 1g samples of long-lived radioactive nuclei using the Pb target irradiated with 10 mA proton beams at 1 GeV. Assuming that the conditions of the irradiation are kept unchanged and similar to those, which we have used in our experiments, one can estimate the following transmutation rates.

6.7 mg of ^{239}Pu is transmuted per day (0.7%),
21 mg of ^{237}Np mg is transmuted in one month (2.1%),
3 mg of ^{129}I is transmuted in one month (0.3%).

Any practical transmutation set-up using 10 mA beam of 1 GeV proton must use a considerable more complex target structure than we have used due to the increased heat dissipation of a 10 mA proton beam.
V- Neutron spectra at 0, 10 and 20 cm depths inside the paraffin moderator were calculated using Monte Carlo simulation with DCM-CEM (see footnote in page 10) for Pb target at proton energies 0.65, 1.0 and 1.5 GeV respectively:
1- The number of low energy neutrons increase with increasing depth at a constant energy and position by a factor of 1.5 (when energy is changing from 0.65 to 1.0 GeV) and 1.4 (when energy is changing from 1.0 to 1.5 GeV), which is confirmed by the experimental B-values obtained.
2-At a given energy and depth the number of neutrons have within a certain energy interval a flat maximum between 10 and 15 cm from the front of paraffin moderator.

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