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SPATIAL DISTRIBUTION OF NEUTRONS IN PARAFFIN MODERATOR SURROUNDING A LEAD TARGET IRRADIATED WITH PROTONS AT INTERMEDIATE ENERGIES

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Introduction

As a result of the continued operation of commercial nuclear power plants and other nuclear facilities, the world has accumulated increasing amounts of highly radioactive waste (R.W.). These large quantities of R.W. constitute a very serious problem for mankind both from a commercial and as well as from an ecological point of view. Recently, several researching teams suggested the development of „subcritical nuclear assemblies" [1 - 3] fired by relativistic accelerators in order to transmute - and in this way destroy - long-lived R.W. efficiently into stable or short-lived nuclei. The essential technical step is the production of large amounts of neutrons during spallation reactions in heavy target materials induced by relativistic protons. The energy spectrum of neutrons within such a Setup extends from thermal energies up to few handred MeV. The exact form of this neutron energy spectrum is depends on a wide range of parameters, such as position within the target assembly, the target composition, etc. Studies of these associated problems have also been carried out at the Laboratory of High Energies (JINR, Dubna, Russia) during recent years [4 - 11]. The present work is a continuation of these investigations describing a technical details of the neutron fluence determination close to the the moderator surface of our standard experimental Setup, called "Gamma-2", as described in the next section.

For the determination of the neutron distribution at different depth inside a paraffin moderator surrounding a lead target the experimental Setup "Gamma 2" was used. The neutron fluence was measured with radiochemical sensors. In our previous work we employed small radiochemical sensors, such as 1 g samples of natural uranium or lanthanum (^{139}La). In this work ten ^{139}La sensors in the form of lanthanum-chloride ($LaCl_3 \cdot 7H_2O$) were distributed on top of paraffin moderator. Thermal neutrons interact with ^{139}La nuclei leading to production of ^{140}La, which is a β - emitter with a suitable half live of 40.2744 h [12]

$$^{139}La + n \rightarrow {}^{140}La \xrightarrow{\beta^-} {}^{140}Ce\,.$$

The decay scheme [12] for ^{140}La has several gamma lines, the most intense lines are in the energy range from 300 keV up to 1600 keV. In our studies we used up to eight lines to calculate the experimental production rate or activity of the La sensors. Table 1 contains a list of gamma lines with intensities per decay (percent).

Table 1. Gamma ray energies and intensity per decay for ^{140}La

Energy, keV	Intensity per decay % (Error)	Energy, keV	Intensity per decay% (Error)
328.762(8)	20.3(3)	867.846(20)	5.50(7)
487.021(12)	45.5(6)	919.550(23)	2.66(3)
751.637(18)	4.33(4)	925.189(21)	6.89(7)
815.772(19)	23.3(4)	1596.21(4)	95.4

The Experimental Setup

Several experiments [4 - 11] were carried out during the last decade using at the JINR in Dubna the transmutation Setup "Gamma 2". Nuclotron or Synchrophasotron accelerators provided beams of relativistic particles within energy range from 0.5 GeV up to 8 GeV.

Fig.1 gives the top view of this experimental Setup "Gamma 2" together with its complete monitoring system. In order to control the beam five fast scintillation detectors C1xC2 (for measuring the direct beam) and C3xC4xC5 (to measure the amount of secondary particles) were used. Scattering targets with thickness of 1 g/cm^2 were used for dispersion of a part of the initial beam and detection of the scattered particles with appropriate detectors and electronics. Polaroid films were used for the definition of position and the size of the beam on the target at different places. The initial information about the beam intensity was obtained using instrumentation at the accelerator beam-exit. Al activation foils were used to determine the integral proton flux under well-defined conditions at the exact target position.

In November 2001, the Nuclotron accelerator delivered to this experiment relativistic protons with energies 1.5 GeV, 1.0 GeV and 0.65 GeV. The proton beam interacted with 20 lead discs as target. Each disk was of dimensions of 8 cm diameter and of 1 cm thickness. Lead target was installed inside paraffin cylinder with inner diameter 8 cm and outer diameter 20 cm used as a moderator. The total target arrangement is shown in Fig. 2.

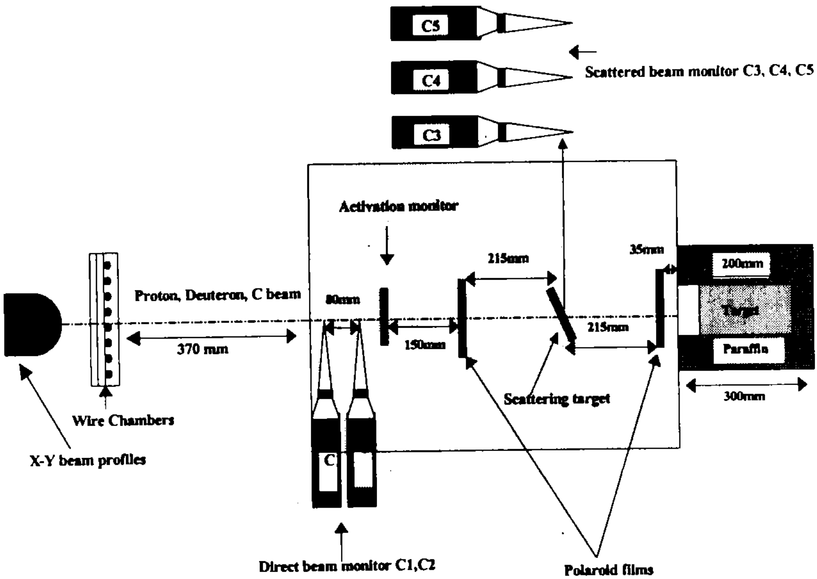

Fig.1. Top view of the experimental Setup Gamma 2

Ten La sensors contained in small plastic vessels with approximately 1 g stable La were installed at the top surface of the paraffin moderator. For proton energy 1.0 GeV the first five La samples (1-5) were placed onto the surface of moderator (0 mm) with the distance 46.5 mm, 100mm, 147.5 mm, 199 mm, 249 mm measured from the front edge of paraffin moderator. The other five samples (6-10) were arranged in a straight line separated by about 50 mm and divided into two groups, the first group consists of three La samples (6, 8, 10) installed at depth 9.3 mm 10.1 mm and 8.4 mm with distance 46.5 mm 147.5 mm and 249 mm respectively, and the second group consists of two La samples (7, 9) placed at depth 15.9 mm and 16.6 mm with the distance 10 mm and 19.9 mm. From the front of paraffin, respectively. Fig.2 shows the lead target including La sensors positions. Detailes for other proton energies are tabulated in table 3. This arrangement was irradiated with relativistic protons and after the end-of-bombardment, the induced gamma-activity was studied with HPGe gamma detectors in a standard manner.

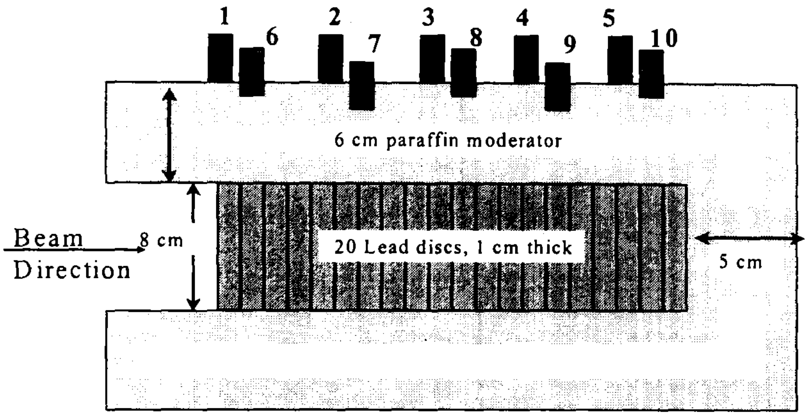

Fig.2 Schematic presentation of the lead target assembly surrounded by a paraffin moderator and the placement of 10 La sensors on top of the moderator, as described in the text.

Results and discussion

Determination of the full energy peak efficiency of a HPGe detector

The HPGe detector system was calibrated using a well-defined ^{152}Eu-source, which has several gamma lines ranging from 122 keV up to 1408 keV. The calibration curve was calculated using the following equation:

$$\varepsilon_{\gamma,A} = \frac{S_A}{(A_o e^{-\lambda t_w}) \cdot I_{\gamma,A} \cdot T_m} \tag{1}$$

where $\varepsilon_{\gamma,A}$ is the registration efficiency at a gamma energy γ_A
S_A is the peak area at energy γ
A_o is the initial activity of the Eu source in Bq.
λ is the decay constant for ^{152}Eu

T_m is the measuring time in seconds
t_w is the *waitting time in seconds*
$I_{\gamma,A}$ is the intensity for the gamma line γ_A.

Fig.3 shows the resulting calibration curve used in this work. The experimental values are fitted with a fourth order polynomial function.

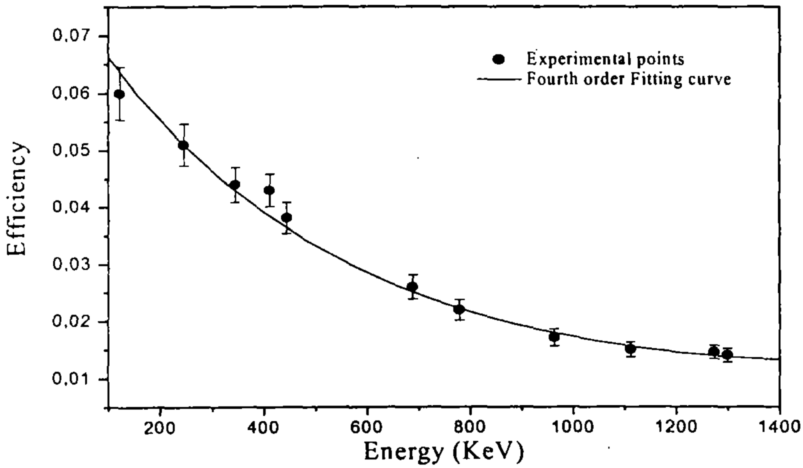

Fig.3 Efficiency curve for HPGe detector used for measuring gamma rays in the decay of ^{140}La samples

Radiochemical determination of the proton fluence using Al monitor foils

Aluminum foils of 8 cm diameter were irradiated at proton energies 1.5 GeV, 1.0 GeV and 0.65 GeV and used as conventional monitor foils to determine the integral proton fluence during the irradiation. The activity of ^{24}Na in the Al foils was calculated using eq. 2.

$$A = \frac{\lambda S_{\gamma} e^{\lambda t_w} t_{m,real}}{I_{\gamma} \varepsilon_{\gamma} \; (1 - e^{-\lambda t_{m,real}}) \; (1 - e^{-\lambda t_{irr}}) \; t_{m,live}} \qquad (2)$$

where A is the activity of ^{24}Na
S_{γ} is the peak area at energy γ
λ is the decay constant for $^{24}Na = 1.2835*10^{-5}\ s^{-1}$
$t_{m,real}$ is the real measuring time in second
$t_{m,live}$ is the live measuring time in second

t_w is the waiting time between the irradiation and measurements
t_{irr} irradiation time
I_γ is the intensity for the gamma line γ.

Then the integral proton flux was calculated using eq.3

$$F = \frac{A \cdot t_{irr}}{\sigma N_T} \tag{3}$$

where N_T is density of the Al foil in [atoms/cm^2]
σ interaction cross section for proton in Al producing ^{24}Na .

The results for the proton fluences are given in Table 2.

Table 2. Integral proton fluence calculated using the monitor reaction in Al foils (E13)

Experiment	Time of Irradiation (min)	Total fluence (10^{13})	Cross section (mb) [13, 14]
1.5 GeV p+Pb	416.8	1.159±0.050	10.0±0.5
1.0 GeV p+Pb	509.8	0.764±0.034	10.5±0.5
0.65 GeV p+Pb	1123	1.024±0.050	10.8±0.5

Experimental production rate (B-value) for ^{140}La

The gamma ray spectra of the 10 La sensors in each run were collected after the end of irradiation and investigated in a standard manner for their gamma activity using HPGe detector systems. Each La sample was measured three or four times during a period of three or four days. In order to obtain gamma spectra with good statistical accuracy each spectrum was collected for at least two hours. Fig. 4 shows a typical ^{140}La spectrum measured with the high-resolution HPGe detector system.

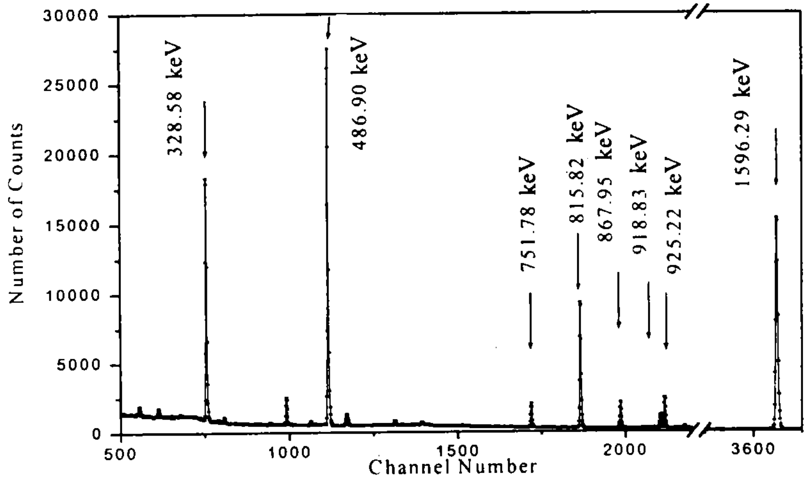

Fig. 4 La spectrum with the most intense gamma lines measured using high resolution HPGe detector

The measured spectra were analyzed and the net peak area was calculated using well-known procedures [15]. The experimental production rates (B-value) for ten La sensors were calculated using eq.4:

$$B = \frac{N\left({}^{140}La\right)}{M \cdot I_p} \qquad (4)$$

where $N\left({}^{140}La\right)$ is the number of ^{140}La atoms formed during irradiation of the ^{139}La target with mass M (grams) normalized to 1 incident proton (I_p - fluence of the proton beam).

We had several spectra (three spectra for each La samples position) and several gamma lines were used to calculate the experimental production rate B. The weighted average [16] over the number of gamma lines and over the number of spectra was calculated for each individual determined B-value and is tabulated in Table 3. The relation between the experimental B-values for all La sensors as a function of depth at proton energies 0.65 GeV, 1.0 GeV and 1.5 GeV were plotted in Fig.5a, b, c for different position on the top surface of paraffin moderator (distances are measured from the front plate of the lead target).

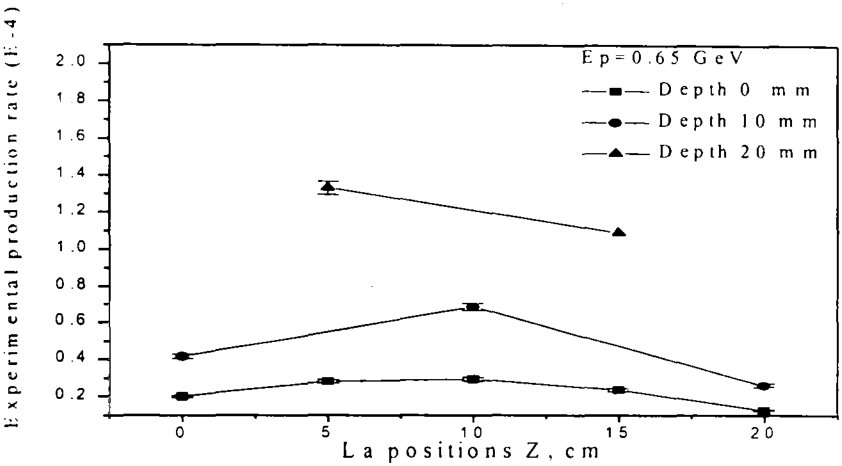

Fig. 5-a Experimental production rate for La sensors as a function of the sample position. Distances along Z axis are measured relative to the front plate of the lead target

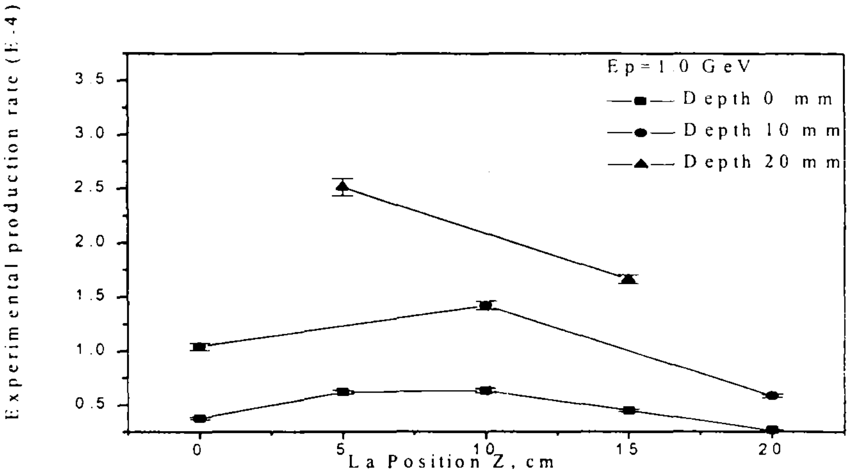

Fig. 5-b Experimental production rate for La sensors as a function of the sample position. Distances along Z axis are measured relative to the front plate of the lead target

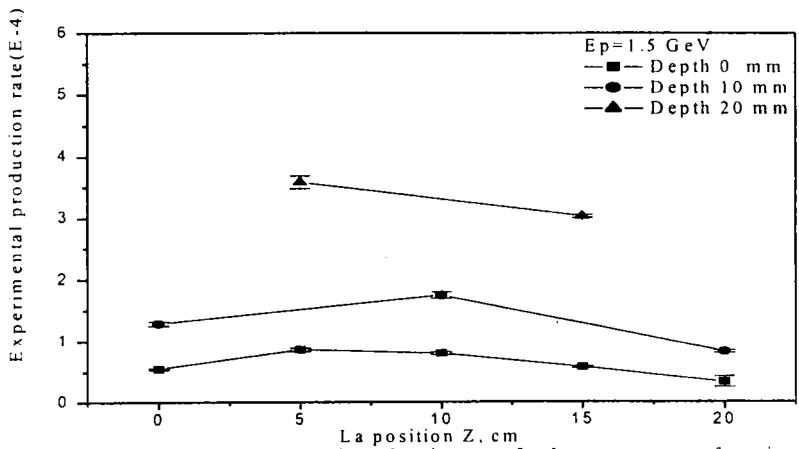

Fig. 5-c Experimental production rate for La sensors as a function of the sample position. Distances along Z axis are measured relative to the front plate of the lead target

The comparison of experimental B-values with calculated B-values as based on the LAHET-code [17] is given also in Table 3. The calculation procedure is described in our earlier publications (see e.g. [18, 19]). The agreement between the experimental and calculated B-values generally is satisfactory. The error values for the calculated B-values given in the Table 3 refer to the statistical errors only and possible systematic errors are not taken into account.

Table 3 Experimental B(^{140}La) multiplied by 10^5 for La sensors at three proton energies and their comparison with theoretical calculations. Distances are measured relative to the front plate of the paraffin moderator.

Proton Energy 0.65 GeV			
Sample depth in paraffin (cm)	Sample position, Z (cm)	Experimental B-values*10^{-5}	Calculated B-values*10^{-5}
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
0.88	15	7.86±0.50	10.11 ± 0.42
0.85	24.75	2.70±0.17	4.52 ± 0.32
1.44	10.15	14.7±0.9	15.52 ± 0.59
1.6	19.8	11.74±0.7	13.7 ± 0.56

Table 3 continued

Proton Energy 1 GeV			
Sample depth in paraffin (cm)	Sample position, Z (cm)	Experimental B-values*10^{-5}	Calculated B-values*10^{-5}
0	4.65	3.67±0.23	3.85 ± 0.31
0	10	6.14±0.38	6.26 ± 0.34
0	14.75	6.41±0.39	7.02 ± 0.39
0	19.9	4.49±0.28	5.18 ± 0.32
0	24.9	2.71±0.17	3.37 ± 0.24
0.93	4.65	10.75±0.66	7.94 ± 0.56
1.01	14.75	14.43±0.88	16.35 ± 0.78
8.4	24.9	5.87±0.36	6.58 ± 0.53
1.59	10	26.19±1.67	25.78 ± 0.12
1.66	19.9	18.95±1.16	20.48 ± 0.98

Table 3 continued

Proton Energy 1.5 GeV			
Sample depth in paraffin (cm)	Sample position, Z (cm)	Experimental B-values*10^{-5}	Calculated B-values*10^{-5}
0	5.28	5.45±0.43	4.02 ± 0.27
0	10.1	8.40±0.67	6.92 ± 0.35
0	14.95	8.10±0.64	7.1 ± 0.39
0	19.95	5.71±0.45	5.4 ± 0.32
0	24.9	3.25±0.26	3.41 ± 0.24
0.89	5.28	12.96±1.04	10.1 ± 0.61
0.9	14.9	18.54±1.47	18.47 ± 0.84
0.85	24.9	8.75±0.69	8.88 ± 0.59
2	10.1	38.59±3.06	40.12 ± 1.44
1.9	19.95	30.12±2.39	40.92 ± 1.43

Conclusions

The results of this study on the production of ^{140}La can be summarized as follows:

1. The distributions of all B(^{140}La)-values on top of the moderator at the three proton energies studied reproduces the feature, which has been observed so far for Pb-targets sytems using La-sensors placed in holes of 10 mm depth: They all have their maximum at Z-values (measured along the beam direction from the front of the Pb-target) between 5 cm to 10 cm. The position of this maximum is not dependent on the proton energy. This conclusion is valid only considering the geometry and composition of the setup used in the experiments.
2. The B(^{140}La)-values at a given geometrical position increase approximately linearily with the proton energy - again a feature, observed in previous experiments [4-10].
3. 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 coresponding low-energy neutron fluence, which is essentially producing ^{140}La, increases strongly when one goes deeper into the paraffin moderator.
4. 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.
5. Compared to specific previous experiments [9,10] using the Pb-target setup irradiated with 1.5 GeV protons, the results for the depth of 10 mm proton energy are reasonably well reproduced.

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внутри парафинового замедлителя, окружающего свинцовую мишень,
облучаемую протонами промежуточных энергий

Изучалось распределение нейтронов, генерируемых релятивистскими протонами с энергиями 0,65, 1,0 и 1,5 ГэВ, которые взаимодействовали со свинцовой мишенью. Эксперименты проводились на пучках нуклотрона на установке «Гамма-2». Распределение нейтронов внутри парафинового замедлителя исследовалось в реакции $^{139}La\,(n,\gamma)\,^{140}La$ с помощью образцов $LaCl_3 \cdot 7H_2O$, которые помещались на глубину 0, 10 и 20 мм. Анализ экспериментальных данных показывает, что при заданной энергии протонов активность образцов ^{140}La зависит от глубины, на которой они размещаются. При заданной глубине активность возрастает с увеличением энергии налетающих протонов.

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Adam J. et al. E1-2002-233
Spatial Distribution of Neutrons
in Paraffin Moderator Surrounding a Lead Target Irradiated
with Protons at Intermediate Energies

The distribution of neutrons emitted during the irradiation with 0.65, 1.0 and 1.5 GeV protons from a lead target ($\varnothing$=8 cm, ℓ=20 cm) and moderated by a surrounding paraffin moderator of 6 cm thick was studied with a radiochemical sensor along the beam axis on top of the moderator. Small ^{139}La-sensors of approximately 1 g were used to measure essentially the thermal neutron fluence at different depths near the surface: i.e., on top of the moderator, in 10 mm deep holes and in 20 mm deep holes. The reaction $^{139}La\,(n,\gamma)\,^{140}La$ ($\tau_{1/2}$ = 40.27 h) was studied using standard procedures of gamma spectroscopy and data analysis. The neutron induced activity of ^{140}La increases strongly with the depth of the hole inside the moderator, its activity distribution along the beam direction on top of the moderator has its maximum about 10 cm downstream the entrance of the protons into the lead and the induced activity increases about linearily with the proton energy. Some comparisons of the experimental results with model estimations based on the LAHET code are also presented. The experiments were carried out using the Nuclotron accelerator of the Laboratory of High Energies (JINR).

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

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