Method for *in situ* measuring the thickness of a lithium layer

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ABSTRACT: This work describes a new method for *in situ* measuring the lithium layer thickness. The method is based on the registration of the yield of 478 keV gamma-quanta of ^{7}Li(p,p'γ)^{7}Li reaction. The results of measuring the radial distribution of the thickness of a lithium layer thermally deposited in vacuum on a cooled copper substrate are presented. The possibility of using this method for certification of lithium targets used for boron neutron capture therapy is noted.

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Contents

1 Introduction

Boron neutron capture therapy (BNCT) is currently considered as a promising technique for treatment of malignant tumors [1]. As a result of the neutron absorption by boron a nuclear reaction ^{10}B(n,α)^{7}Li takes place with a large energy release inside the cell which contains a boron nucleus that leads to the destruction of this cell. The technique has been tested at nuclear reactors and is expected to be introduced into clinical practice in the first five BNCT clinics equipped with neutron sources based on a particle accelerator, and a neutron generating target [2–7]. The threshold reaction ^{7}Li(p,n)^{7}Be is considered to be the best for BNCT due to the huge neutron yield and low neutron energy [8]. The neutron generating target is usually made of three layers [5]: a thin lithium layer to generate neutrons, a thin layer of radiation-resistant metal to absorb protons, and a cooled copper substrate to remove heat. The thickness of the lithium layer is chosen such that the proton energy at the exit from lithium is equal to or slightly below 1.882 MeV, the threshold of the ^{7}Li(p,n)^{7}Be reaction. In case of a thicker lithium layer, the neutron yield will not increase, yet the flux of unwanted 478 keV photons emitted during the reaction of inelastic scattering of a proton on a lithium nucleus ^{7}Li(p,p'γ)^{7}Li will increase. For this reason, it is important to measure the thickness of the lithium layer of the neutron generating target without damaging it.

Measuring the lithium thickness is not an easy task due to the fact that lithium has high chemical activity requiring vacuum or inert atmosphere to work with, and low hardness which does not allow using mechanical measuring devices. Previously, during the development of thermal lithium deposition method, we implemented the technique for measuring the thickness of a lithium layer based on measuring the conductivity of water where we put the copper witnesses that were in good thermal contact with the substrate during the lithium deposition [9]. At present we determine an average thickness of the lithium layer according to the mass of lithium which we measure on the microbalance before the lithium deposition, or according to the conductivity of water which we use to wash off the lithium from the neutron generating target.

The aim of this work is to develop the method for measuring the spatial distribution of lithium layer thickness without damage the neutron generating target.
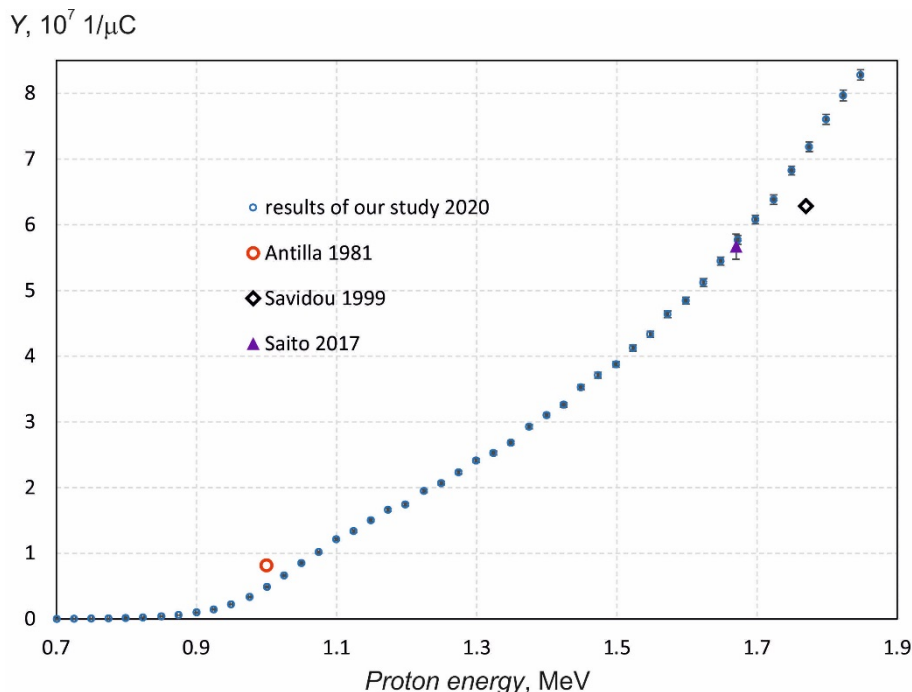
Figure 1. The measured yield of 478 keV gamma-quanta from the thick lithium target in the ^{7}Li(p,p'γ)^{7}Li reaction. The data of measuring are shown for comparison: $\circ$ — [11], $\diamond$ — [12], Δ — [13].

2 Description of the method

Before the development of the method for measuring the thickness of the lithium layer we have measured the yield of 478 keV gamma-quanta from the thick lithium target under irradiation by protons with the energy from 0.7 to 1.85 MeV [10]. The result of this experiment is shown in figure 1. The energy stability ranged from 0.1 to 0.2% with an average value of 0.14%. The measurements were carried out with an energy step of 25 keV. The relative error of measuring the yield of gamma-quanta did not exceed 1%. The absolute yield of gamma-quanta is measured with an accuracy better than 15%. Figure 1 also shows all data on the yield of gamma-quanta from the thick lithium target measured by other authors in this proton energy range [11-13]. It is obvious that the data is not sufficient and the reliability of some results is doubtful.

The idea of the method for measuring the thickness of the lithium layer is to irradiate the target by protons and to measure by a gamma-spectrometer the count rate of 478 keV gamma-quanta released during an inelastic scattering of the proton on the lithium nucleus. Two lithium targets are used during the measurements: the investigated one and the thick one. The thick target is that in which the protons decelerate to the energy lower than 478 keV — the threshold of the reaction ^{7}Li(p,p'γ)^{7}Li. The thin target is that in which the protons decelerate to the energy higher than 478 keV. Further deceleration of protons occurs in copper, tantalum, molybdenum, or in any other material and is accompanied by a significantly lower intensity of gamma radiation [14].

If the investigated target is not thick, then the yield of 478 keV gamma-quanta from it will be less than from the thick one. It is possible to determine the thickness of lithium h by measuring the ratio A between the count rate of 478 keV gamma-quanta from the investigated and from the
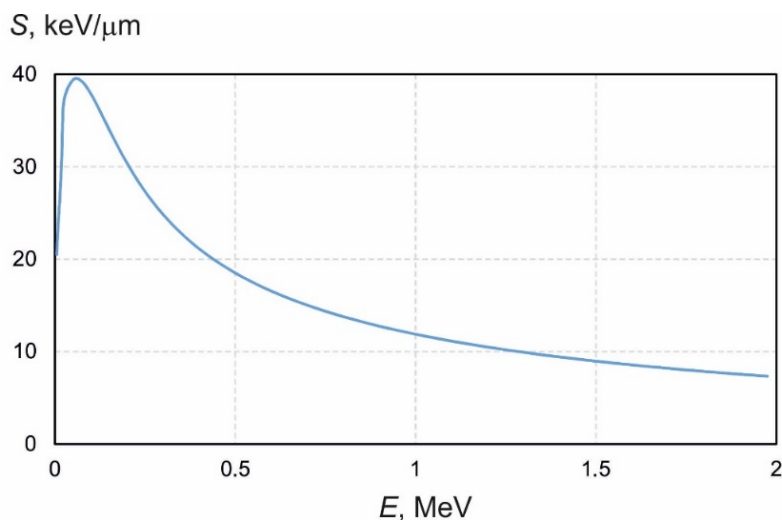
Figure 2. The dependence of the energy loss rate S of a proton in lithium on the proton energy E.
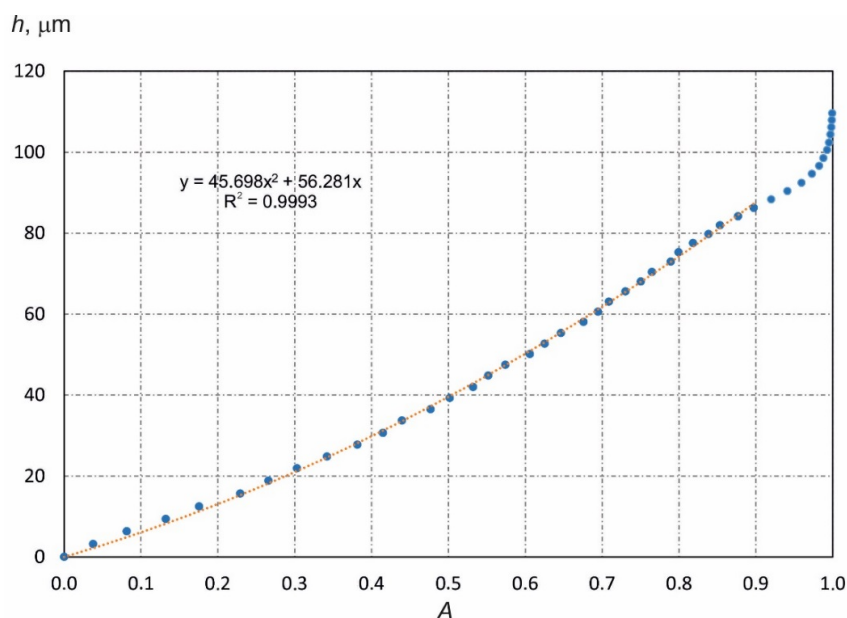

Figure 3. Graph for determining the thickness of the lithium layer h from the measured ratio A of the radiation intensities of 478 keV of gamma-quanta from the investigated and the thick target at the proton energy of 1.85 MeV. The dashed line shows the trend line drawn through the experimental points in the A range from 0 to 0.9. The equation of the trend line and the value of the approximation reliability R^2 are also given.

thick target, since the rate of the energy loss of a proton in lithium is known and depends on its energy as follows [15]: $S = \frac{S_{\text{low}} \cdot S_{\text{high}}}{S_{\text{low}} + S_{\text{high}}}$ eV/(10^{15} atoms/cm^2), where $S_{\text{low}} = 1,6E^{0,45}$, $S_{\text{high}} = \frac{725,6}{E} \ln(1 + \frac{3013}{E} + 0,04578E)$. The dependence of the energy loss of a proton in lithium on the energy value is presented in figure 2.

The most optimal regime of experiments is the energy slightly below the threshold of ^{7}Li(p,n)^{7}Be reaction, for example, 1.85 MeV. Such energy value is chosen because there will

be no neutrons able to damage the gamma-spectrometer and no additional gamma-quanta flux, also the yield of 478 keV gamma-quanta will be maximum. The graph in figure 3 is built at the energy 1.85 MeV to determine the lithium thickness h depending on the measured ratio between the radiation intensities of 478 keV gamma-quanta from the investigated and from the thick target A. This graph is build according to the data shown in figure 1. Firstly, we find the ratio $A_i = Y_i/Y_{1.85}$ for every energy value E_i, where Y_i — the yield of gamma-quanta at the energy E_i, $Y_{1.85}$ — the yield of gamma-quanta at the proton energy 1.85 MeV, that is, we found a discrete strictly monotone function $A = f(E)$. Then with the use of the above formula for the rate of energy loss by a proton in lithium, we find the thickness h_i at which the proton loses energy from 1.85 MeV to E_i, that is, we found a discrete strictly monotone function $h = f(E)$. Knowing the values of A_i and h_i for each value from the discrete set of energies E_i, we determine the function $h = f(A)$ which is shown in figure 3.

In the A range from 0 to 0.9 the experimentally measured values of $h(A)$ are well approximated by the expression $h \,(\mu m) = 45.698 \cdot A^2 + 56.281 \cdot A$ (figure 3) which is suitable for practical use.

Figure 3 shows that at the proton energy lower than the threshold of neutron generation the proposed method allows measuring the lithium thickness up to 100 μm. To measure the greater thickness the proton energy must be higher and, consequently, the gamma-spectrometer must be resistant to neutron flux.

3 Experimental setup

The proposed method for measuring the lithium thickness was practically implemented on the accelerator-based epithermal neutron source in the Budker Institute of Nuclear Physics [7]. The experimental scheme is shown in figure 4. A part of the beam of protons with the energy from 0.7 to 2.1 MeV is directed from the tandem-accelerator 1 to the lithium target 7 through the cooled collimator 3 with 2 mm aperture.

The neutron-generating target is a copper disk with 144 mm diameter and 8 mm thickness. One side of this disk is covered by a thin, visually uniform layer of lithium of crystalline density thermally deposited on 82 mm diameter area in vacuum [16]. The average thickness of the layer is from 1 to 300 μm. For water cooling on the opposite side of the copper disk four double-turn spiral channels are made inside 122 mm diameter [17]. The neutron generating target connected to the installation through bellows 5 is shifted in the horizontal plane relative to the setup axis with an electric linear actuator Bohua (China) at a distance of up to 5 cm in both directions. The positioning accuracy of the actuator is 0.5 mm. The displacement of the target provides its scanning along the diameter with a 2 mm proton beam.

The current of protons irradiating the target is measured by a resistive voltage divider connected to the target unit, electrically isolated from the setup.

The intensity of gamma radiation is measured with a spectrometer of gamma radiation 11 (JSC IPTP, Russia) based on a semiconductor detector made of ultrapure germanium. The sensitive part of the spectrometer is located at 2 m distance from the lithium target at an angle of $110°$ to the axis of the accelerator. The spectrometer is placed inside a lead collimator 12 with an outer diameter of 27 cm, a length of 50 cm, and a wall thickness of 5 cm. The spectrometer and collimator are protected from the accelerator radiation by the wall 10 of 23 cm thickness, built of concrete blocks.
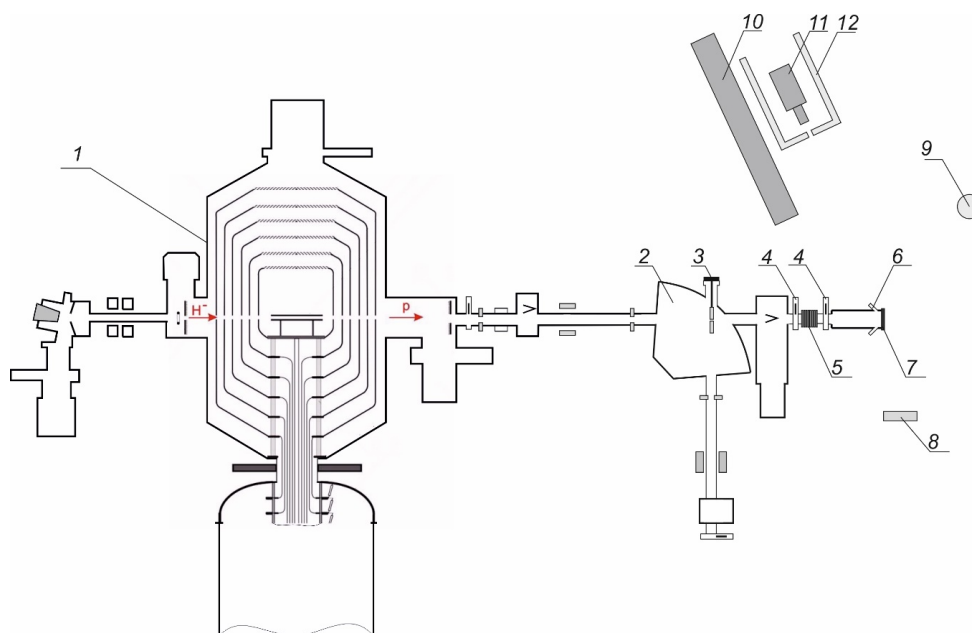
Figure 4. Experimental scheme: 1 — tandem accelerator with vacuum insulation, 2 — bending magnet, 3 — cooled collimator with 2 mm aperture, 4 — gate valve, 5 — bellows, 6 — observation window, 7 — lithium target, 8 — gamma radiation dosimeter, 9 — neutron radiation dosimeter, 10 — concrete wall, 11 — gamma radiation spectrometer, 12 — lead collimator.

The placement of the concrete wall, collimator and spectrometer is shown in figure 4 schematically; in reality, they are located in the horizontal plane behind a vacuum chamber with the lithium target.

The gamma radiation spectrometer is calibrated for full sensitivity by a closed-type Cs-137 radionuclide photon source with an activity of 1.6×10^8 Bq (10% confidence) in the 661.657 keV emission line. During the calibration, the radionuclide source was placed in the center of the copper disk of the target - as close as possible to the place of gamma-quanta emission from lithium. The energy dependence of the sensitivity of the spectrometer was calibrated with the following standard sources of photon radiation (Ritverc, Russia): Na-22, Mn-54, Co-60, Ba-133, Cs-137, Eu-152, and Bi-207.

When the energy of protons is higher than 1.882 MeV the dose rate of neutron radiation is measured by the BDMN-100-07 detection unit (OOO Doza, Russia) 9 located on the wall of the radiation-protected room at a distance of 5.4 m from the target at an angle of 22° to the direction of the proton beam.

In this configuration the surface of the lithium target is scanned with a proton beam of 2 mm diameter. The proton beam is visually controlled by a Hikvision DS-2CD4026 video camera with an 11–40 mm F1.4 lens (Hikvision Digital Technology Co., Ltd, Hangzhou, China) mounted on the window 6 with the fused quartz glass. The video camera registers the nonthermal glow of the target caused by the luminescence of lithium under the influence of high-energy protons. Examples of images which are obtained by moving the target relative to the proton beam are shown in figure 5. The images clearly show the glow in the form of a light oval spot. The shape of the spot is caused by that the video camera is directed to the target at an angle of 45°.

(a)

(b)

Figure 5. Luminescence recorded by the video camera during the irradiation of the lithium target by a proton beam with 2 mm diameter: (a) when the beam is directed to the edge of the target, (b) to the center.

4 Measurement results and discussion

The measurements were carried out for three targets on which a visually uniform layer of lithium of 82 mm diameter with an average thickness of 200 μm, 68 μm, and 7.4 μm was deposited. The average thickness of the lithium layer is determined from the mass of lithium weighted before the process of the deposition, taking into account 2% losses which were previously determined experimentally.

Just after the deposition of the lithium layer the target unit is transferred with a residual vacuum of about 0.5 Pa from the deposition stand to the experimental setup and irradiated with a proton beam of 1.85 MeV energy and with an average 11–14 μA current passing through the collimator.

Protons with an initial energy of 1.85 MeV at a length of 126 μm (according to the above formula for the dependence of the energy loss by a proton in lithium [15]) are decelerated in lithium to 478 keV, the threshold of the ^{7}Li(p,p'γ)^{7}Li reaction. Therefore, one of the targets with an average lithium thickness of 200 μm is considered thick, and the other two are investigated.

During the process of proton irradiation, the target is moved horizontally using an actuator with a step of 0.5 cm in the range of ±5 cm relative to the accelerator axis.

The gamma spectrometer measures the counting rate of 478 keV gamma quanta generated in the ^{7}Li(p,p'γ)^{7}Li reaction after each movement of the target. The 478 gamma rays were measured for 2 minutes at each points, so that the error in measuring the count rate does not exceed 0.5%. The obtained values are normalized to the average proton current. Further, A value is calculated — the ratio between the normalized counting rate of gamma quanta emitted from the investigated target and the normalized counting rate of gamma quanta emitted from the thick target. Then, using the formula $h\,(\mu m) = 45.698 \cdot A^2 + 56.281 \cdot A$ the thickness of lithium is determined.
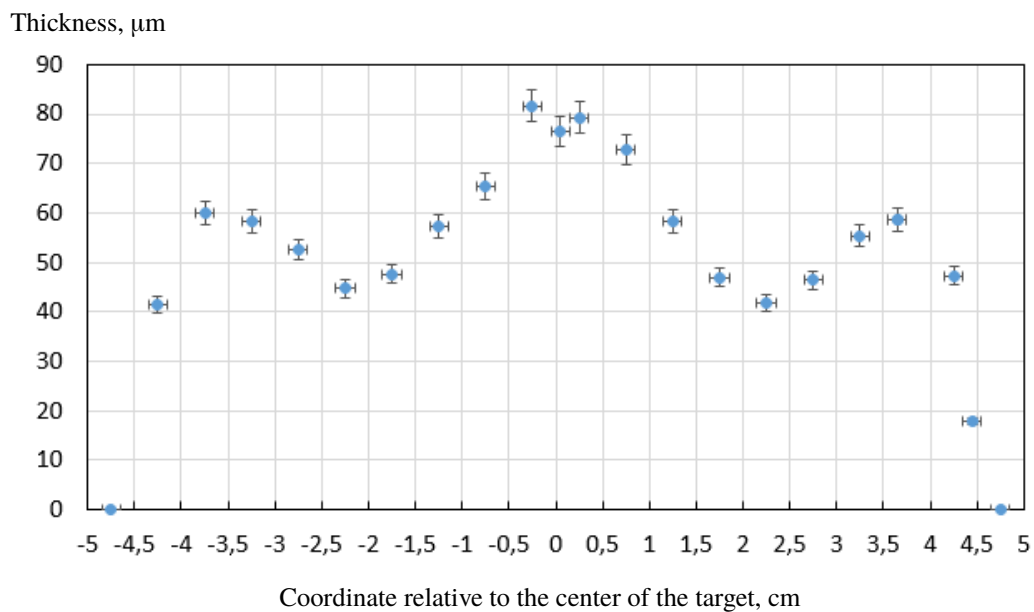
Figure 6 shows the results of measuring the radial distribution of the lithium thickness of the investigated targets. The measurement accuracy along the x-axis is determined by the size of the proton beam on the target, in this case it was 2 mm, along the y-axis — mainly by the stability of the proton beam current, in this case from 15 % to 17 %. In figure 6(a), it can be seen that the distribution of lithium thickness is double-humped: the maximum in the center and at a diameter of 7 cm. This distribution is obtained because lithium is placed on the heater in the form of a ring with a diameter of 4 cm and from above at a distance of 0.5 cm it is covered with a reflector made in the form of a disk 5.4 cm with a hole 2.3 cm in diameter. At the same time, with a small amount of lithium, as seen in figure 6(b), only the central part of the target is deposited. It is clear that the developed technique for measuring the lithium thickness will make it possible to study in detail the process of lithium deposition and find the mode and conditions under which a uniform deposition of a thin lithium layer will be ensured.

It is possible to measure the relative thickness of lithium according to the neutron yield for the target with an average thickness of the lithium layer of 7.4 μm. To implement this, the proton energy is increased to 2.05 MeV and the dose rate of neutron radiation is measured with a dosimeter. At an energy of 2.05 MeV, protons lose energy in lithium with the rate of 0.14 keV/μm and decelerate to 50 keV at the thickness of 7 μm. In this proton energy range, the cross section of the ^{7}Li(p,n)^{7}Be reaction is constant with the good accuracy (from 295 mb at 2.00 MeV to 309 mb at 2.05 MeV); therefore, the neutron yield is proportional to the thickness of the lithium layer. Figure 6(b) shows the result of measuring the radial distribution of lithium thickness obtained by this method. As it is seen, the measurement results are in a good agreement.
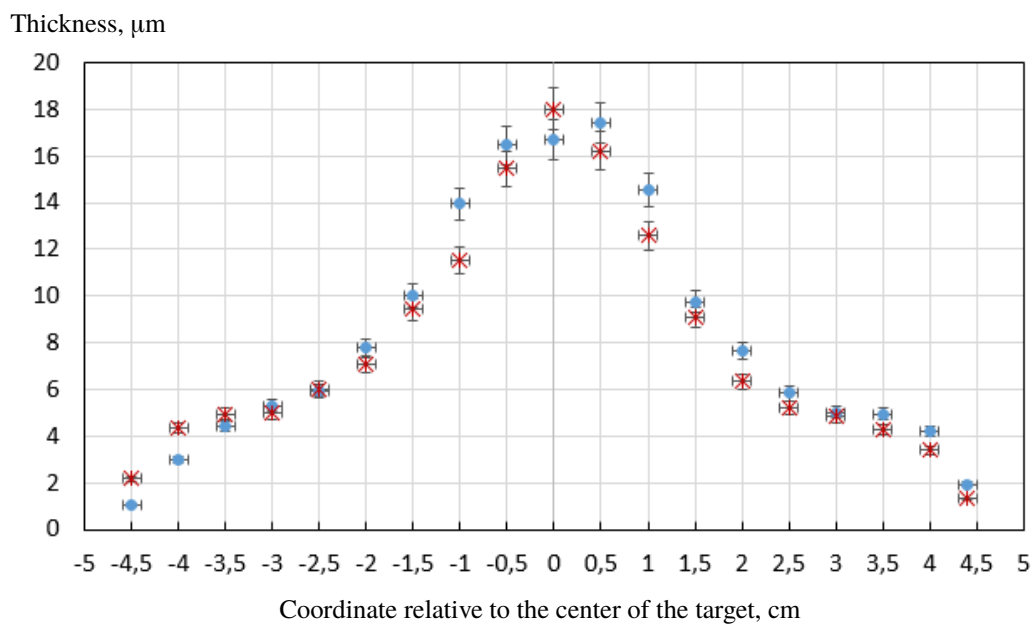
5   Conclusion

In this study the method for *in situ* measuring the thickness of the lithium layer is proposed. The method is based on comparing the radiation intensity of 478 keV gamma-quanta from the investigated lithium layer and from the thick one. The studies carried out at the accelerator-based neutron source of the Budker Institute of Nuclear Physics demonstrated the possibility of measuring the thickness of the lithium layer of up to 100 μm.

The method of non-destructive control of the thickness of the lithium layer is helpful for the certification of lithium targets applied for the boron neutron capture therapy of malignant tumors.

Thickness, μm

Coordinate relative to the center of the target, cm

(a)

Thickness, μm

Coordinate relative to the center of the target, cm

(b)

Figure 6. Radial distribution of lithium thickness: (a) the targets with an average lithium thickness of 68 μm, (b) the targets with an average lithium thickness of 7.4 μm, ∘ — measured by the yield of gamma-quanta, x — measured by the neutron yield.

Acknowledgments

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