

Effect of the Residual Gas on the Lithium Layer of a Neutron-Generating Target

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Abstract—An epithermal neutron source based on a tandem accelerator is being constructed at the Budker Institute of Nuclear Physics for use in neutron capture therapy at a cancer clinic. A lithium neutron-generating target obtained by deposition of lithium onto the target substrate is one of the key elements of the facility. The effect of residual gas on the lithium layer composition has been investigated using a secondary-ion mass spectrometer.

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Neutron capture therapy [1] of malignant tumors has passed clinical trials on nuclear reactors, exhibiting a high efficiency [2]. An accelerator-based compact source of epithermal neutrons [3] is currently being constructed at the Budker Institute of Nuclear Physics (BINP) for use in a cancer clinic. Neutrons are generated in the threshold nuclear reaction ${}^7\text{Li}(p, n){}^7\text{Be}$ upon spill of the proton beam with an energy of 2.0–2.5 MeV on a lithium target [4]. Carrying out therapy requires that the neutron flux be stable and controllable; however, a 10% decrease in the neutron yield per unit current has been observed within 3 h after the spill of the proton beam on a lithium target in the experiment on the accelerator in Birmingham (Great Britain) [5]. This decrease may be caused by a change in the lithium layer composition as a result of its interaction with the residual gas, since the neutron yield, e.g., in lithium nitride was a factor of 1.66 lower than in pure lithium.

The distribution of the elemental composition along the thickness of lithium layers exposed under different vacuum conditions was investigated using the secondary-ion mass spectrometry (SIMS) method. In this paper, we present the results of this study and, based on these data, estimate the possibility of a decrease in the neutron yield through interactions between lithium and the residual gas.

EXPERIMENT

Deposition of the lithium layer onto a substrate was performed on a facility created at the BINP. The arrangement for deposition that we developed was a

heated volume with a circular slot, which was placed on a movable valve plate. Heating in vacuum has made it possible to uniformly transfer the whole of the lithium placed in this volume onto a cooled target [6] (on a sample in particular) for subsequent mass spectrometric investigations. A tantalum plate with dimensions of 4×8 mm and a thickness of 1 mm, preliminarily purified in an ultrasonic bath, was used as a sample. Via a thin indium layer, this plate was pressed to the transportation unit (Fig. 1), which was connected to the target (Fig. 2), which had a hole at its center.

A heater with aluminum plates hung on it was attached to the target near the sample. As the temperature of the heater increased, a thin aluminum layer was deposited onto the lithium layer in order to protect it against the effect of air after subsequent unsealing of the arrangement. The heater was produced according to a technique in [7] from five 0.3-mm-diameter tungsten wires twisted into a bunch and then into a coil upon heating by a direct current.

Three samples with preliminarily deposited lithium layer ~ 80 μm thick were examined in our experiment. The samples were held in a vacuum of 2×10^{-5} Torr over different periods of time: the first sample was coated with a protective aluminum layer ~ 1 μm thick immediately after the lithium layer was deposited, while the second and the third were coated 0.5 and 1 h later, respectively. The vacuum chamber was then filled with gaseous argon at an excessive pressure. The piston was moved into the transportation unit, and the unit was removed and closed with a disk while argon was blown through tube 5 (see Fig. 2). Continued blowing of argon

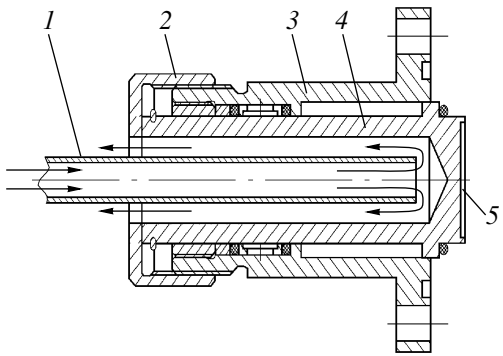
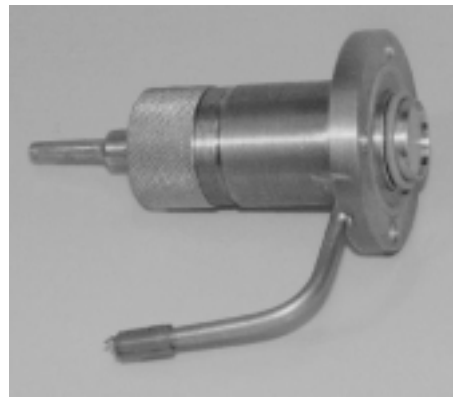


Fig. 1. Sample transportation unit: (1) nozzle, (2) captive nut, (3) case, (4) piston, and (5) space for placing a sample.



has made it possible to substantially reduce interaction between air and the sample as it was transported to the MIQ-256 secondary-ion mass spectrometer (CAM-ECA-RIBER, France).

Over the short period of time (≤ 5 min) it took for the samples to be removed from the transportation unit and placed on the mass spectrometer's platform, the samples were in contact with air. Though the lithium was coated with an aluminum layer, the samples became darker, which indicated that lithium nitride was formed at their surfaces. Afterward, the platform with the sample was placed in the MIQ-256 facility with an operating vacuum of 10^{-10} Torr. In this case, the samples were held in a vacuum of $< 10^{-3}$ Torr for 10 min or less and in vacuum of $< 10^{-8}$ Torr for 20 min.

In our experiments, a beam of primary O_2^+ ions with an energy of 10 keV bombarded the surface of the analyzed sample at an angle of 45° . To reach the desired depth, sputtering was effected by the beam with a current of $1.2 \mu A$, which scanned an area of $250 \times 250 \mu m$. The mass spectrum in the range of 0–60 m/e was recorded at a beam current of $0.05 \mu A$ and a scanning area of $100 \times 100 \mu m$. The characteristic dependence of the yield of secondary negative ions on mass-to-charge ratio m/e is shown in Fig. 3.

EXPERIMENTAL RESULTS AND THEIR DISCUSSION

It is a well known fact [8] that, at a temperature of $200^\circ C$, lithium mostly interacts with nitrogen, forming lithium nitride Li_3N —a substance of black color. Other compounds can also be produced, in particular, at temperatures of > 200 and $> 400^\circ C$, these are lithium oxide Li_2O and lithium hydride LiH , respectively; at higher temperatures, lithium carbide Li_2C_2 .

Under the experimental conditions at the accelerator in Birmingham, the temperature was $120^\circ C$; therefore, possible changes in the properties of the lithium layer

might be primarily caused by formation of Li_3N . Taking into account that the lithium film is ~ 1 mm thick and that protons with an initial energy of 2.8 MeV are decelerated to a threshold neutron generation energy of 1.882 MeV upon passing through a $137\text{-}\mu m$ -thick lithium layer [9], a 10% decrease in the neutron yield requires that 25% of lithium in the neutron-generating layer be replaced by Li_3N [4].

If the above considerations hold true, it was the Li_3N formation process in our experiment that had to manifest itself most clearly in the height of peaks corresponding to production of nitrogen-containing negatively charged clusters (complexes). As the measurements showed, the mass spectra of all the three samples are almost identical; therefore, in what follows, we ana-

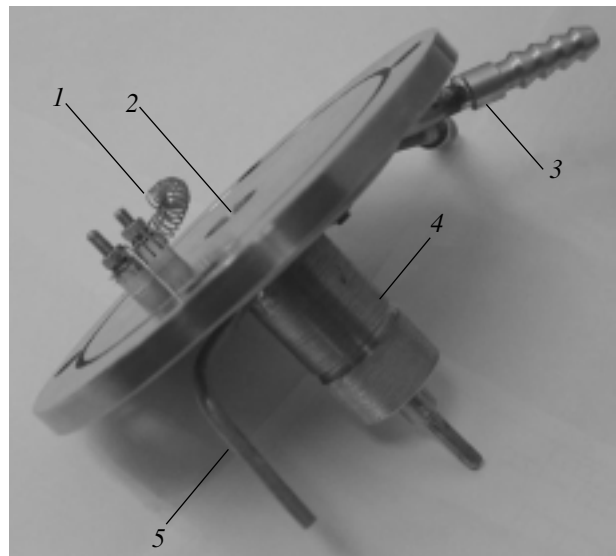


Fig. 2. Target with the heater and the sample transportation unit: (1) heater for aluminum deposition, (2) space for a sample, (3) inlet of water cooling, (4) sample transportation unit, and (5) argon feeding tube.

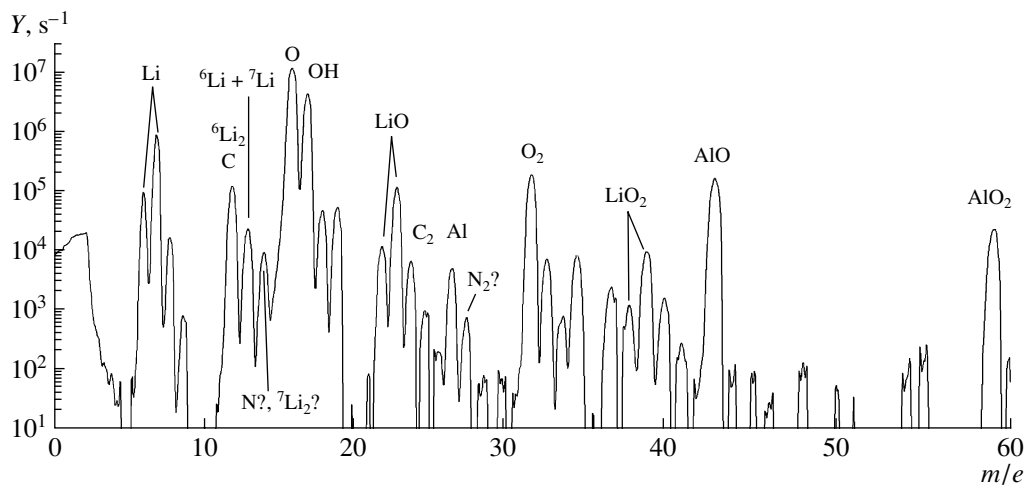


Fig. 3. Characteristic dependence of the secondary negative ion yield on the ratio of mass m to charge e .

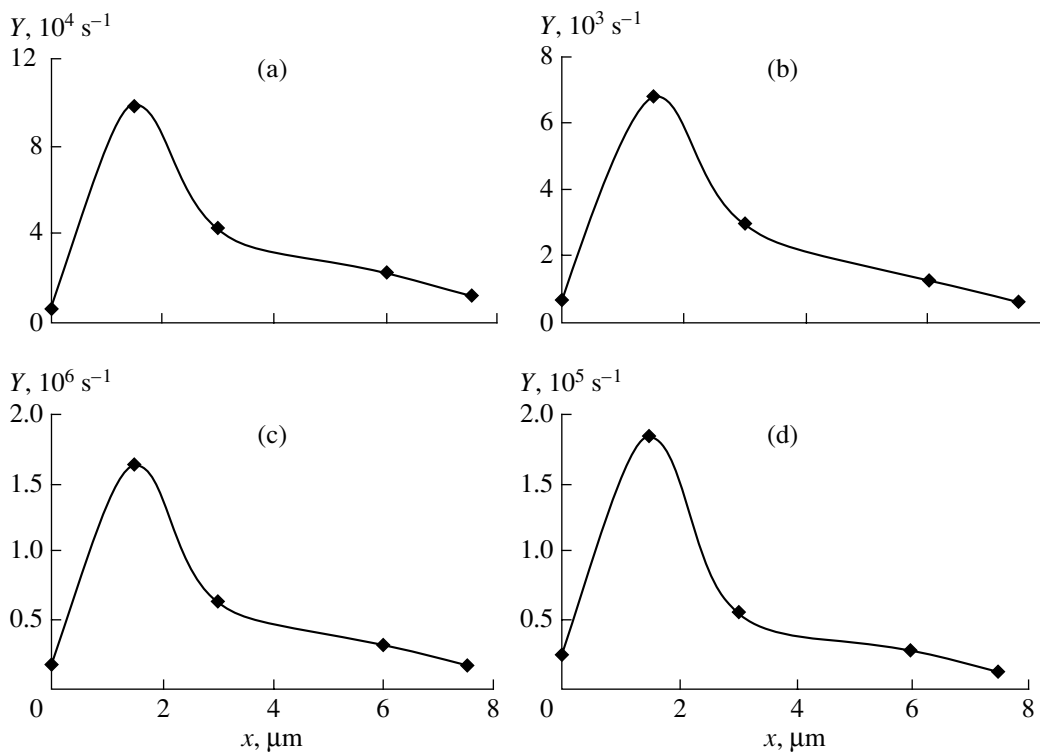


Fig. 4. Dependence of the secondary negative ion yield on the layer depth for ions with (a) $m/e = 27$, (b) 34, (c) 43, and (d) 59.

lyze in detail the results obtained with the second sample (held for 30 min in a vacuum of 2×10^{-5} Torr).

The dependences of the secondary ion yield on the depth of the layer are shown in Figs. 4–6. The depth at which the composition of the layer is analyzed has been determined by referring the etch rate of silicon measured on reference samples to the etch rate of lithium. The calculations made with the aid of the SRIM-2006

program showed that the etch rate of lithium is a factor of 1.2 lower than that of silicon (and a factor of 1.1 lower than that of aluminum). This estimate is fully acceptable, since, for the effect to be revealed, exact knowledge of the depth is not needed. It should also be noted that, in this case, there was no necessity for use of the standard microstandards technique (e.g., see [10]); it was sufficient that only the relative yield of negative ions be measured.

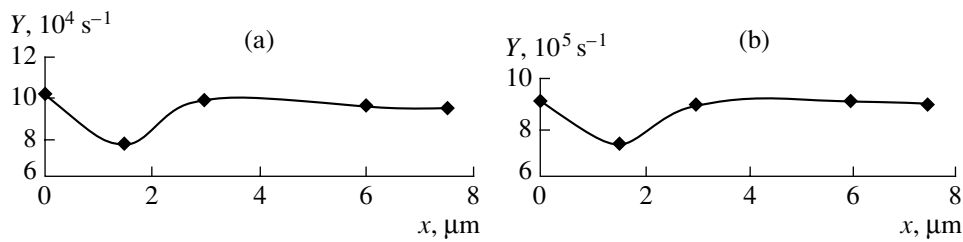


Fig. 5. Dependence of the secondary negative ion yield on the layer depth for ions with (a) $m/e = 6$ and (b) 7.

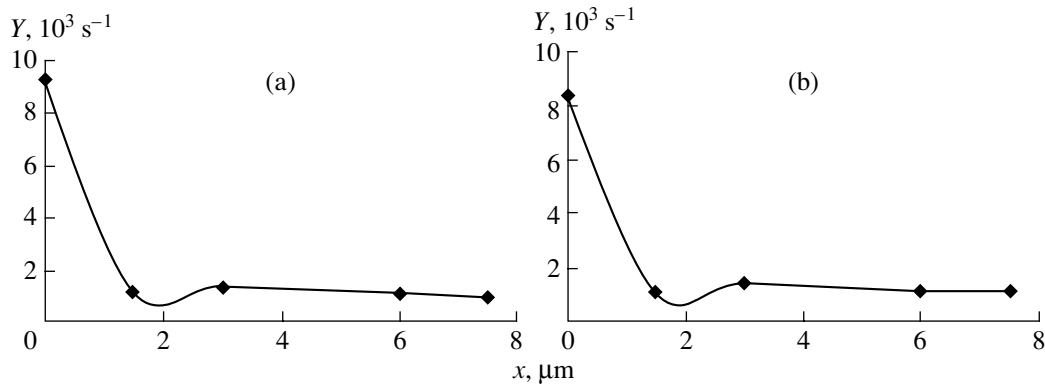


Fig. 6. Dependence of the secondary negative ion yield on the layer depth for ions with (a) $m/e = 8$ and (b) 35.

Analysis of Fig. 4 shows that the protective aluminum coating is unambiguously identified by signals due to ions with ratios $m/e = 27$ (Al), 34 (AlLi), 43 (AlO), and 59 (AlO₂) and that the thickness of this layer is $\sim 1 \mu\text{m}$, as was predicted before the deposition.

The yield of secondary negative ions with ratios $m/e = 6$ and 7 versus the layer thickness is shown in Fig. 5. The signals correspond to two lithium isotopes: ⁶Li and ⁷Li. Analysis of these curves shows that lithium is also present in the protective aluminum layer applied to the surface. The most probable explanation to this fact is that lithium penetrates into the aluminum layer or is trapped during deposition. According to Fig. 5, as lithium isotopes move through the aluminum deep into the target material, the intensity of their signals changes only slightly (within the limits of 20%). This allows us to assume that sputtering of lithium instead of aluminum has little or no effect on the yield of secondary negative lithium ions.

At the same time, the intensity of the signals corresponding to such lithium compounds as lithium hydride (LiH, $m/e = 8$) and nitride (Li₃N, $m/e = 35$) varies along the depth of the aluminum layer from its surface to its inner boundary with the lithium layer by almost an order of magnitude (Fig. 6) and then remains almost unchanged until the depth becomes as great as $7.5 \mu\text{m}$. Such a behavior of the signals can be attributed to the ratio of concentrations, provided that the use of lithium

as a sputtered material instead of aluminum affects the yield of secondary negative ions only slightly for LiH ($m/e = 8$) and Li₃N ($m/e = 35$) compounds, as well as for negative secondary lithium ions (see Fig. 5). In other words, if these compounds have no specific features from the standpoint of electron affinity, such a signal behavior is related to the difference in the concentration of LiH and Li₃N in the corresponding sputtered layers.

Lithium hydride and nitride are most likely to be produced at the surface in the period of time when the sample is removed from the container and placed in the mass spectrometer. It is in this 5-min period of exposure to air that lithium compounds are produced on the sample surface, since, as was ascertained, lithium either penetrates or is trapped into the protective aluminum coating during deposition. The observed blackening of the sample during exposure to air is indirect corroboration of this fact.

Thus, if the whole of the lithium in an $\sim 2\text{-}\mu\text{m}$ -thick upper layer is considered to be transformed into Li₃N compound, the proportion of lithium fixed in nitride makes $\sim 10\%$ at large depths. Hence, it follows that $<10\%$ of lithium could be transformed into Li₃N over 30 min in which the sample was held in a vacuum of 2×10^{-5} Torr. It is therefore highly improbable that $\sim 25\%$ of the lithium was transformed into Li₃N in a $100\text{-}\mu\text{m}$ -thick layer over 3 h in a considerably higher

vacuum of 6×10^{-7} Torr during the Birmingham experiment.

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