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PHYSICS AND TECHNIQUE OF ACCELERATORS

Study of the Accumulation of Impurities in a Thin Lithium Target by Ion Scattering Spectroscopy

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Abstract—An accelerator-based epithermal neutron source (VITA) has been created and is now operating at the Budker Institute of Nuclear Physics. VITA includes an original tandem electrostatic accelerator (vacuum-insulated tandem accelerator), which produces a monoenergetic beam of protons or deuterons with energies from 0.3 to 2.3 MeV and currents up to 10 mA, and a thin lithium target that generates a powerful neutron flux in ^{7}Li(p, n)^{7}Be and Li(d, n) reactions. The facility is used to develop boron neutron capture therapy and many other applications. Heavy impurities contained in a lithium target are known to significantly decrease the neutron yield and make the target unusable. The purpose of this study was to examine the accumulation of impurities in a thin lithium target. The elemental composition of the sample was determined by ion scattering spectroscopy. The composition of the lithium layer immediately after sputtering on a copper substrate was determined in the experiments. The interaction of lithium with air and the effect of the impurities on the neutron yield were studied. The accumulation of impurities during proton beam irradiation of the target at a beam power density of 1 kW/cm^2 and up to 3.4 kW/cm^2 was investigated.

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1. INTRODUCTION

Neutrons are required by a wide variety of applications . Neutrons are generated in nuclear reactors or using charged particle accelerators by directing a beam of high-energy ions at a neutron-generating target. One of these targets is lithium, which provides an intense flux of neutrons with relatively low energy in the ^{7}Li(p, n)^{7}Be reaction for neutron capture therapy of malignant tumors [1, 2] or an intense flux of fast neutrons in the ^{6}Li(d,n) ^{7}Be reaction for radiation testing of promising materials. A thin lithium target for efficient neutron generation has been proposed and developed at the Novosibirsk-based Institute of Nuclear Physics [3–7]. This lithium target is used in the VITA accelerator neutron source at the Institute of Nuclear Physics for scientific research [8], at the BNCT center in Xiamen (China) for the treatment of patients with malignant tumors using the boron neutron capture therapy (BNCT) [9]; they will also be used at the National Blokhin Medical Research Center for Oncology (Moscow) to conduct BNCT. Determining the service life of a lithium target and developing technology for supplying a lithium target from the manufacturer to the consumer are now urgent tasks.

The purpose of this work is to determine the presence of impurities in a lithium target that can reduce the yield of neutrons, study the dynamics of their accumulation during long-term irradiation with an ion beam, determine the service life of the target for efficient generation of neutrons, and investigate the interaction of the lithium target with air to develop a technology for delivering lithium targets from manufacturer to consumer.

2. EXPERIMENTAL SETUP

The elemental composition of the lithium layer of the target and its changes during irradiation with a proton beam were studied at the VITA accelerator neutron source (Fig. 1) operated by the Institute of Nuclear Physics [8]. A tandem accelerator with vacuum insulation produces a stationary monoenergetic beam of protons with an energy ranging from 0.3 to 2.3 MeV and a current of up to 10 mA, which is directed to a lithium target. The developed lithium target consists of three layers: a thin layer of pure lithium metal for generating neutrons, a thin layer of material for absorbing protons, and a thin copper substrate for efficient heat removal. The copper substrate is a copper disk with a diameter of 144 mm and a thickness of 8 mm. On the proton beam side, a thin layer of crystalline density lithium with a diameter of 84 mm is thermally sputtered onto the copper disk.

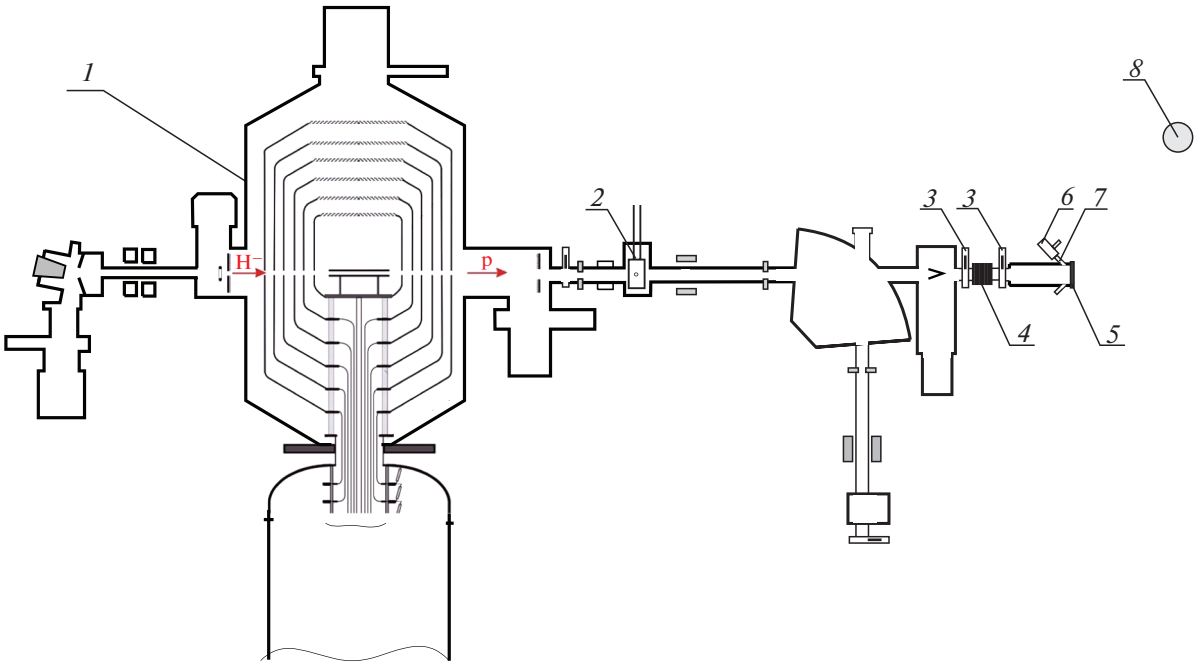

Fig. 1. Schematic view of the experimental setup: (*1*) tandem accelerator with vacuum insulation, (*2*) cooled collimator with an aperture of 1 mm, (*3*) gates, (*4*) bellows, (*5*) video camera, (*6*) α-spectrometer, (*7*) lithium target, (*8*) neutron radiation dosimeter.

To determine the elemental composition of a sample, the ion scattering spectroscopy technique is used [10]. The method consists in irradiating a target with a beam of protons and measuring the energy spectrum of backscattered protons that lose energy as a result of elastic or inelastic scattering on atomic nuclei of the target. To determine the depth distribution of the elemental composition of the sample under study, the SIMNRA v.7.03 program (Max Planck Institute for Plasma Physics, Germany) is used [11]. To measure the intensity and energy of backscattered protons, an α spectrometer is installed via a gate on one of the nozzles of the target unit oriented at an angle of 135° to the beam axis. The α-radiation spectrometer consists of a silicon detector PDPA-1K and a digital spectrometric device TsSU-1K (Institute of Physical and Engineering Problems, Dubna, Russia).

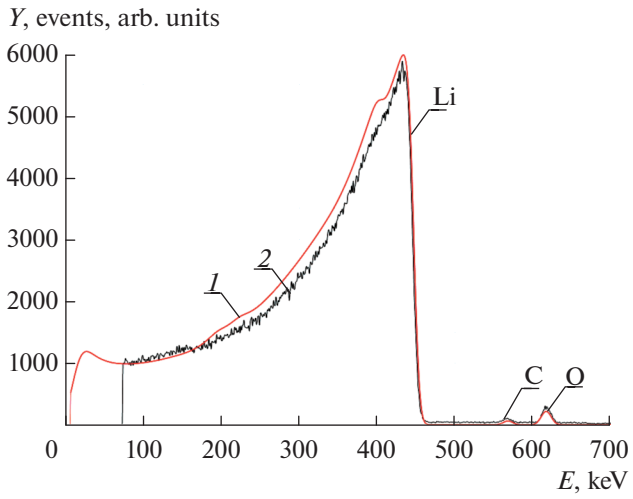

Fig. 2. Spectrum of backscattered protons for a layer of freshly sputtered lithium at a proton energy of 1 MeV: (*1*) experimental data, (*2*) calculation using the SIMNRA program, Li—signal from protons reflected from lithium, C—from carbon, O—from oxygen.

3. RESULTS AND DISCUSSION

3.1. Studying the Purity of Freshly Deposited Lithium Layer

To study the purity of a freshly deposited lithium layer, lithium targets are prepared in a standard way, and the energy spectrum of backscattered protons is measured. The characteristic spectrum of backscattered protons obtained at a proton energy of 1000 ± 2 keV, a current of 2.6 ± 0.3 μA, and a lithium layer thickness of 30 μm, is presented in Fig. 2. The figure also shows the spectrum of the backscattered protons modelled using the SIMNRA program for a given thickness of the main lithium layer of 30 μm and a layer of impurities with a thickness of 37 nm, presumably consisting of lithium carbonate and lithium oxide. The thickness of the layer covering lithium, which is often controlled during the manufacture of targets, varies from 10 to 50 nm (40×10^{15}–300×10^{15} at/cm^2).

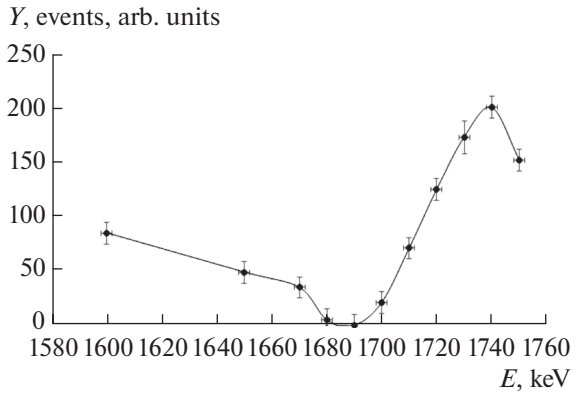

Fig. 3. Dependence of carbon peak intensity Y on proton energy E.

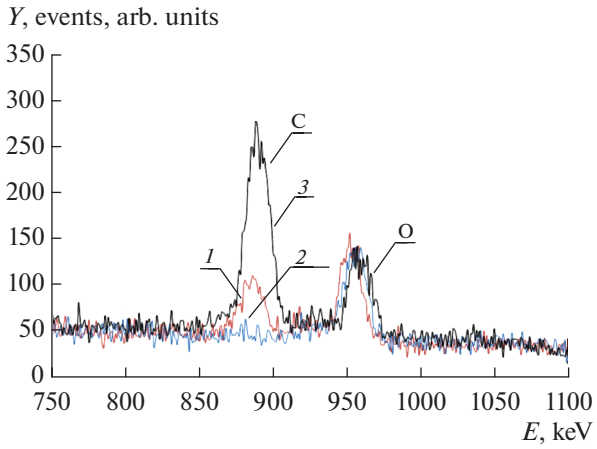

Fig. 4. Signal from protons backscattered from carbon (C) and oxygen (O) at proton energy (*1*) 1.65, (*2*) 1.69, and (*3*) 1.74 MeV.

It can be seen that the signals due to the scattering of protons on oxygen and carbon have the form of isolated narrow peaks. This circumstance indicates that these layers are located on the surface of lithium or in the bulk of lithium as a thin layer. If oxygen or carbon penetrated lithium, the protons scattered on these atomic nuclei would lose energy as they pass through the lithium layer, resulting in broadening of their energy spectrum.

To confirm that the carbon layer is located on the surface of lithium, a narrow resonance in the cross section of elastic scattering of a proton on a ^{12}C atomic nucleus was used. Carbon-12 very efficiently scatters 1.74-MeV protons and virtually does not scatter 1.69-MeV protons. Such a well-pronounced resonance is only present in the cross section of the elastic interaction of a proton with carbon. The proton energy was varied from 1.6 to 1.75 MeV, and the spectrum of protons backscattered by carbon atomic nuclei was measured (Fig. 3). If the proton energy is 1.74 MeV, the carbon peak signal is maximum, while at 1.69 MeV it is minimum (Fig. 4). This implies that a layer containing carbon nuclei is located on the surface of the lithium. If carbon were in the bulk of lithium, the scattered proton would lose some energy when passing through the lithium layer, so that the extrema in the signal intensity would be shifted to lower energies relative to those in the cross section. Thus, it has been established that the surface of the lithium layer, when sputtering is carried out on a stand, is covered with a thin layer of impurities containing nuclei of carbon, oxygen, and lithium.

3.2. Studying the Influence of Air on the Purity of the Lithium Layer

Lithium is a reactive metal, so usually a lithium target is manufactured, stored, and used only in a vacuum. We studied the resistance of the target to atmospheric air. The spectrum was measured immediately after lithium was deposited while maintaining a vacuum in the target unit of $\leq 10^{-3}$ Pa. Next, air flowed into the target assembly and was evacuated after 1 minute. Then measurements were carried out again using the α-spectrometer. The experiment was repeated for the duration of exposure to air of 10 minutes, 1 hour, 24 hours, and a week. Air temperature and humidity controlled using a DHT11 sensor were 23 ± 1°C and 50 ± 20%, respectively. The measurement results are presented in Fig. 5. An analysis of the spectra yields that with increasing duration of exposure to air, both oxygen and carbon accumulate. The longer the target is in contact with air, the greater the amount of oxygen and carbon deposited on the surface, the oxygen accumulation rate being higher. Over time, the spectrum of protons reflected from oxygen becomes flatter. This implies that oxygen moves deeper into the target material. By the end of the

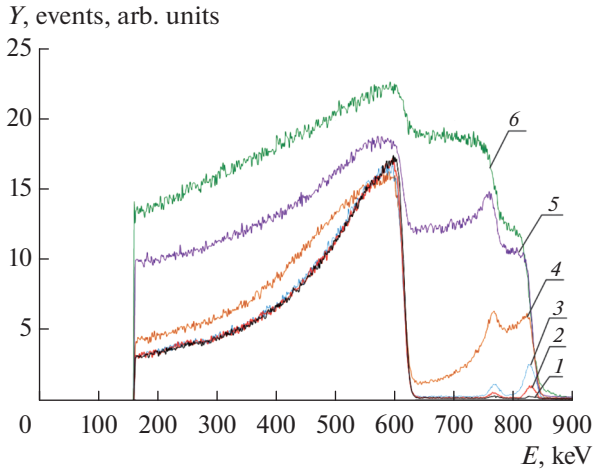

Fig. 5. Lithium target spectra recorded by an α-spectrometer under irradiation with 1-MeV protons. Air exposure time: (*1*) 0, (*2*) 1, (*3*) 10 min, (*4*) 1, (*5*) 24 h, (*6*) a week.

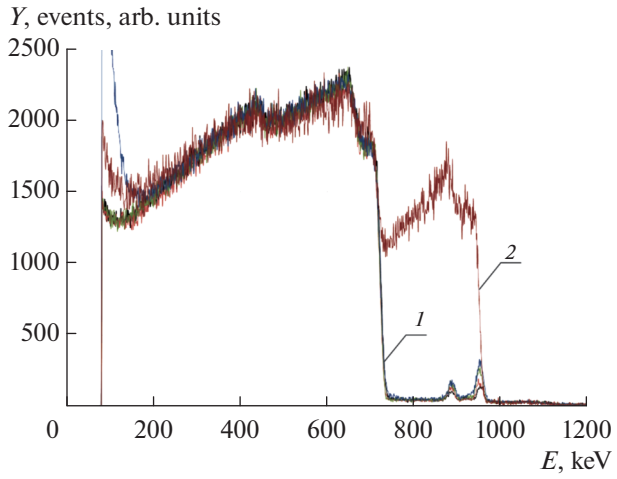

Fig. 6. Lithium target spectra recorded by an α-spectrometer under irradiation with 1.65-MeV protons. Air exposure time: (*1*) a day or less, (*2*) a week.

experiment, the amount of carbon increased by 25 times, and oxygen by 1300 times. Due to accumulation of impurities the neutron yield significant decreases, i.e., the target becomes unusable.

A similar experiment was carried out in winter, when the outside temperature was below zero (Celsius scale), and heating was turned on in the laboratory room, so that the air humidity was 10%. Having processed experimental results, we concluded that after 20 hours of exposure of the lithium target to low-humidity air, the thickness of the layer containing oxygen nuclei increased from 38 to 140 nm, and that of the carbon layer, from 2 to 7 nm (Fig. 6). This accumulation of impurities has virtually no effect on the neutron yield. Consequently, short-term exposure of a lithium target to dry air does not deteriorate its neutron-generating properties, which can be taken into account in developing a technology for supplying lithium targets from manufacturer to consumer.

3.3. Studying the Purity of the Lithium Layer Irradiated with a Proton Beam

Next, we tested a lithium target under irradiation with a proton beam. A target with a lithium thickness of ~30 μm was irradiated for 4 days with a proton beam with an energy of 2 MeV, a current of 1 mA, and a power density of 1.1 kW/cm^2, and the elemental composition of the target was measured using an α-spectrometer before and after irradiation. Based on the results of the experiment, it was found that the amount of oxygen and carbon increases during irradiation and reaches saturation (Fig. 7). The content of carbon on the target surface increased fourfold and that of oxygen, fivefold. The presence of impurities reduces the neutron yield by 0.86%. Accumulated impurities have

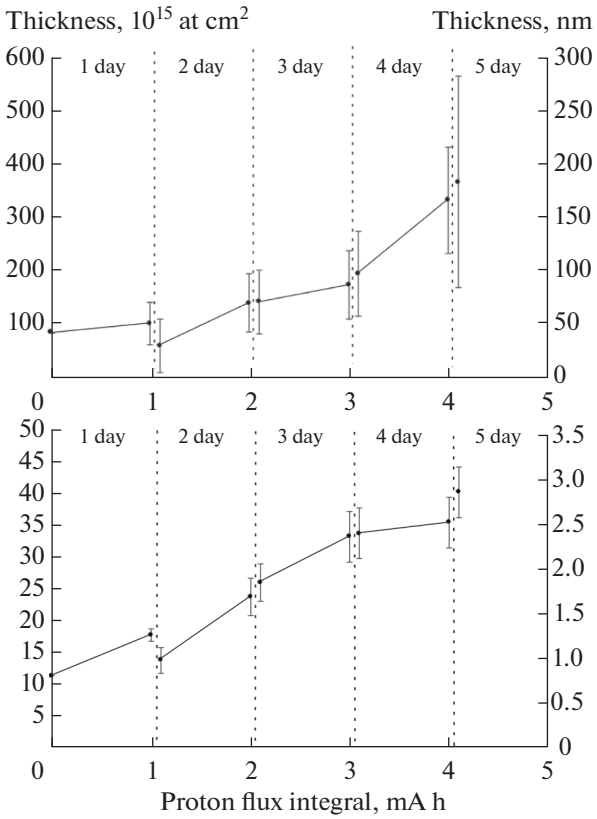

Fig. 7. (a) Dependence of oxygen concentration on the total current; (b) dependence of carbon concentration on the total current.

little effect on the neutron yield. Moreover, they perform as a film for the lithium target protecting against penetration, for example, of nitrogen.

As a continuation of the experiment, the target was irradiated at an increased power density of the proton beam of 3.4 kW/cm^2. At this beam power density, the temperature of the copper target substrate, measured by a thermal resistance, was 240°C, and the lithium under the beam became liquid (the melting point of lithium is 182°C). Although the surface of the lithium target is vertical, the lithium does not flow off. Observation of the target surface using a video camera suggests that, undoubtedly, the lithium became liquid, but it was trapped between the copper substrate of the target and a solid layer of impurities. A heavier impurity appears in the target composition (Fig. 8). Most likely, these are copper flakes formed as a result of radiation blistering of copper in the process of proton implantation [12] and lifted from the surface of the copper substrate by convection of liquid lithium. Further irradiation of the target with a proton beam with such a power will lead to the accumulation of copper in the bulk of lithium and reduce the neutron yield, making the target unusable.

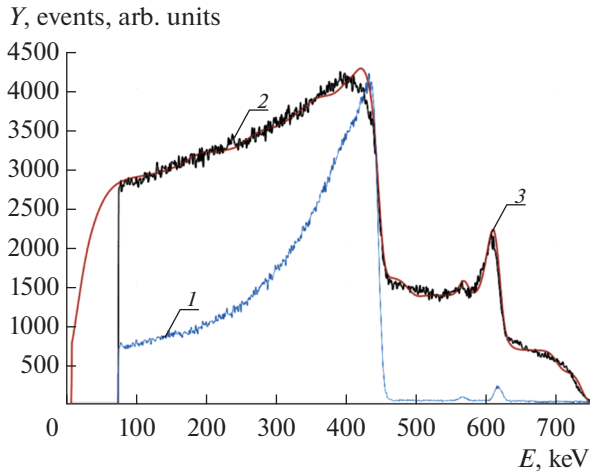

Fig. 8. Spectrum of protons backscattered from a layer of pure lithium at a proton energy of 1 MeV: (*1*) signal of protons backscattered from a pure target, (*2*) signal of protons backscattered from the target after irradiation with a proton beam with a power of 3.4 kW/cm^2, (*3*) simulation using the SIMNRA program.

4. CONCLUSIONS

To generate neutrons, a thin lithium target is used, in which a neutron-generating layer of metallic lithium with a thickness of 30 to 100 microns is thermally sputtered in a vacuum onto a cooled copper substrate. The following results were obtained using the ion scattering spectroscopy method.

(1) In the process of thermal sputtering of lithium in a vacuum onto a copper substrate, the formed lithium layer is covered with a film with a thickness of 10 to 50 nm containing lithium, oxygen (40×10^{15}—300×10^{15} at/cm^2), and carbon (5×10^{15}–20×10^{15} at/cm^2).

(2) The film formed during the deposition of lithium, containing lithium, oxygen, and carbon, protects it from interaction with dry air.

(3) During long-term irradiation of a lithium target with a proton beam with a power density of 1 kW/cm^2 the thickness of the film containing lithium, oxygen and, carbon increases in severalfold, without a noticeable decrease in the neutron-generating properties of the target.

(4) During long-term irradiation of a lithium target with a proton beam with a power density above 3 kW/cm^2 the lithium layer becomes liquid, and copper flakes formed as a result of radiation blistering during proton implantation penetrate into it, which leads to deterioration of the neutron-generating properties of the target.

The results obtained may be of use for developing a technology for delivering a lithium target from manufacturer to consumer and for determining the service life of the lithium target.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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