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OF ACCELERATORS

Methods for Determining the Thickness and Elemental Composition of Thin Layers Irradiated by an Ion Beam in Measuring Nuclear Reaction Cross-Sections

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Abstract—VITA, a neutron accelerator based on an accelerating tandem with vacuum isolation, has been created and is now operated at Budker Institute of Nuclear Physics. VITA includes a tandem electrostatic particle accelerator with a unique design (accelerator tandem with vacuum insulation) intended for generating a monochromatic proton or deuteron beam with an energy of up to 2.3 MeV and the current up to 10 mA. The facility is equipped with γ , α , and neutron spectrometers and dosimeters. The accelerator is used for the development of boron neutron-capture therapy of malignant tumors, radiation testing of advanced materials, and more recently for measuring cross-sections of nuclear reactions. Reliable determination of the linear thickness of atomic nuclei, interacting with a charged particle beam and impurities and affecting the charged particle braking rate, is of uttermost importance for measuring cross sections of nuclear reactions. The difficulty in determining these parameters may explain the significant variation in the data on cross-sections of nuclear reactions provided by various groups of researchers. The study presents the results of the measurements of cross-sections of some nuclear reactions. Attention is focused on the description of the methods used to measure the thickness and elemental composition of thin investigated layers irradiated by a proton or deuteron beam. The applicability of these methods and their accuracy are discussed; the results obtained are compared. Proposals for the development of the diagnostic for further measurements of cross-sections of nuclear reactions are discussed.

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1. INTRODUCTION

The measurement of nuclear reaction cross sections is currently of great interest to many fields of research. The reliability of the results is determined by the accuracy of the experimental parameters. The main contribution to the error in the result obtained is usually due to the determination of the thickness of the target used. It is desirable not only to enhance the accuracy to the maximum possible extent, but also to ensure the use of more than one independent methods of thickness control.

Reaction cross sections were measured at the VITA accelerator neutron source at the Institute of Nuclear Physics in Novosibirsk (Russia) using γ , α , and neutron spectrometers.

The aim of this study is to present various methods for determining the thickness of the examined thin layer using the example of lithium and boron targets interacting with proton and deuteron beams.

2. INSTALLATION DIAGRAM

The study was conducted at the VITA accelerator neutron source at the Institute of Nuclear Physics in Novosibirsk (Russia) [1, 2]. The experimental setup diagram is shown in Fig. 1. Vacuum Insulated Tandem Accelerator 1 is used to obtain a monoenergetic beam of protons or deuterons with energy ranging from 0.1 to 2.3 MeV (stability 0.1%), and current, from 1 nA to 10 mA (stability 0.4%). The beam current is measured and controlled by a non-destructive direct cur-

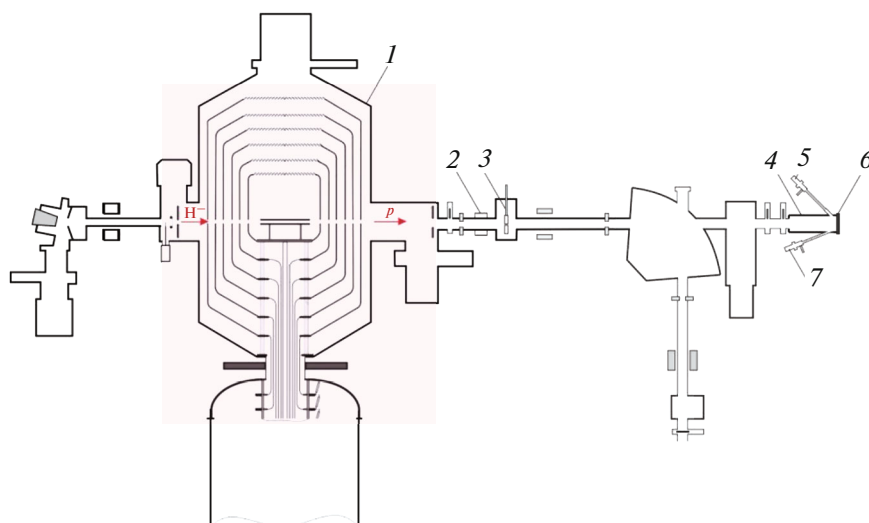


Fig. 1. Experimental setup: (1) tandem accelerator with vacuum insulation, (2) non-destructive DC transformer, (3) collimator, (4) target unit, (5) α spectrometer at an angle of 135° , (6) target, (7) α spectrometer at an angle of 168° .

rent transformer NPCT (Bergoz Instrumentation, France) 2. The beam is directed at the target 4 through 1 mm collimator 3.

3. RESULTS AND DISCUSSION

3.1. Lithium Target

The developed lithium target is a thin layer of pure metallic lithium thermally deposited on a thin copper substrate. Natural lithium produced by the Novosibirsk Chemical Concentrates Plant is used for spraying. The content of the lithium-7 isotope in natural lithium varies from 92.41 [3] to 92.58% [4]; it is assumed below assume that the content of lithium-7 in natural lithium is equal to the average value, 92.5%.

Thermal spraying of lithium onto a target in a vacuum is carried out on a separate stand [5]. The stand is a vacuum chamber with vacuum pumping, in which a titanium cup with a flat ceramic infrared heating element driven by the input stepper motor. Before spraying, the required amount of lithium is weighed with an accuracy of 0.1 mg on an OHAUS laboratory microbalance (USA) inside a glove box. The spraying method provides uniform distribution of lithium on the copper substrate. In this way, the weighed amount of lithium is easily converted into the thickness of the lithium layer on the target with an accuracy of $\sim 10\%$.

Another way to estimate the thickness of the lithium layer of the target is to measure the conductivity of water after removing lithium from the target. Of course, it should be taken into account that this method destroys the target and is only applicable after the main experiment is completed. The accuracy of this method is $\sim 10\%$.

It is preferable to use in situ methods not destructing the target to measure lithium layer thickness. For example, to determine the thickness and elemental composition of the sample, the method of ion scattering spectroscopy is used. The essence of the method is that a target is irradiated with a beam of protons or deuterons and the energy spectrum of back-scattered protons or deuterons, which lose energy as a result of elastic or inelastic scattering on the target's atomic nuclei, is measured. The spectrum is analyzed using the SIMNRA v.7.03 program (Max Planck Institute for Plasma Physics, Germany) [6]. To measure the intensity and energy of back-scattered protons, an α -spectrometer based on a PDPA-1K silicon detector and a TsSU-1K digital spectrometric device (Institute of Physical and Technical Problems, Dubna, Russia) is installed on one of the nozzles of the target unit [7]. Figure 2 displays an example of a spectrum obtained from a lithium target of thickness $\sim 30 \mu\text{m}$ under irradiation with a proton beam, measured using an α spectrometer. The spectrum also provides information about the composition of the target; it is clear that it contains heavy impurities in the form of oxygen and carbon. The accuracy of this method is $\sim 10\%$.

The study of the chemical composition of the upper layer of the target ($\sim 2 \text{ nm}$) was studied by X-ray electron spectroscopy at the Institute of Inorganic Chemistry, Siberian Branch, Russian Academy of Sciences (Novosibirsk), using a FleXPS (SPECS) photoelectron spectrometer. The target surface was found to consist of lithium carbonate (Li_2CO_3).

Our group has proposed and developed an in situ method that significantly enhances the accuracy of measuring the thickness of the target [5]. The method is based on a comparison of the yield of 478 keV photons (1 in Fig. 3) from the lithium layer under study

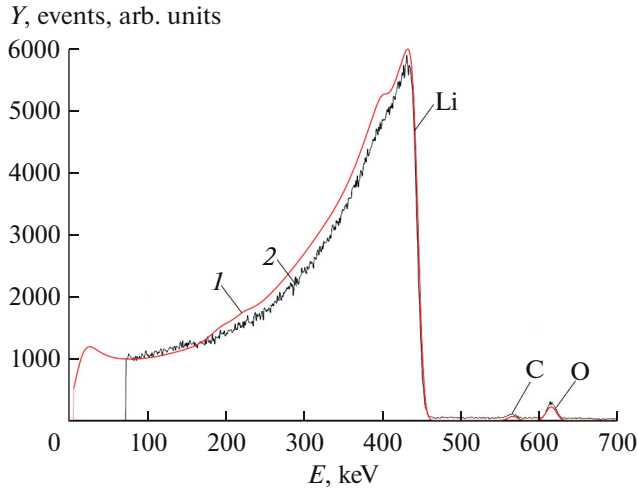


Fig. 2. Spectrum of back-scattered protons for a lithium layer at a proton energy of 1 MeV: (1) experimental data, (2) data calculated using the SIMNRA program, Li denotes the signal from protons reflected from lithium, C, from carbon, and O is for signal from oxygen.

and from the thick layer irradiated with 1.85 MeV protons (Fig. 3). A thick layer is a layer of lithium whose thickness exceeds the proton range in lithium up to the threshold energy of the ${}^7\text{Li}(p, p'\gamma){}^7\text{Li}$ reaction, equal to 478 keV. The thickness of lithium is selected using the formula for the rate of proton energy loss S in lithium as a function of its energy E [8]:

$$S = \frac{S_{\text{low}} S_{\text{high}}}{S_{\text{low}} + S_{\text{high}}} \text{ eV}/(10^{15} \text{ at}/\text{cm}^2), \quad (1)$$

where $S_{\text{low}} = A_1 E^{0.45}$, $S_{\text{high}} = \frac{A_2}{E} \ln\left(1 + \frac{A_3}{E} + A_4 E\right)$, $A_1 = 1.6$, $A_2 = 725.6$, $A_3 = 3013$, $A_4 = 0.04578$, and E is measured in keV. Using this formulas for the rate of proton energy loss in lithium and taking into account the almost rectilinear propagation of the proton in lithium, we find the penetration depth, which is 145 μm for a 1.85 MeV proton and 17 μm for a 0.478 MeV proton. Therefore, protons with an initial energy of 1.85 MeV generate 478 keV photons down to a depth of 128 μm from the lithium surface. Knowing the ratio of the radiation intensity of 478 keV photons per current unit from the lithium layer under study and from the thick one [8], we obtain the thickness of the lithium layer under study. For example, we measured the thickness of a thin lithium target made for an experiment to measure the cross section of the ${}^7\text{Li}(p, \alpha){}^4\text{He}$ nuclear reaction [9]; the measurement error was 3%, and $l = 0.422 \pm 0.013 \mu\text{m}$.

3.2. Boron Target

The thin boron target is a thin layer of boron ($\sim 1 \mu\text{m}$) deposited on a copper substrate using a magnetron at the Institute of High Current Electronics

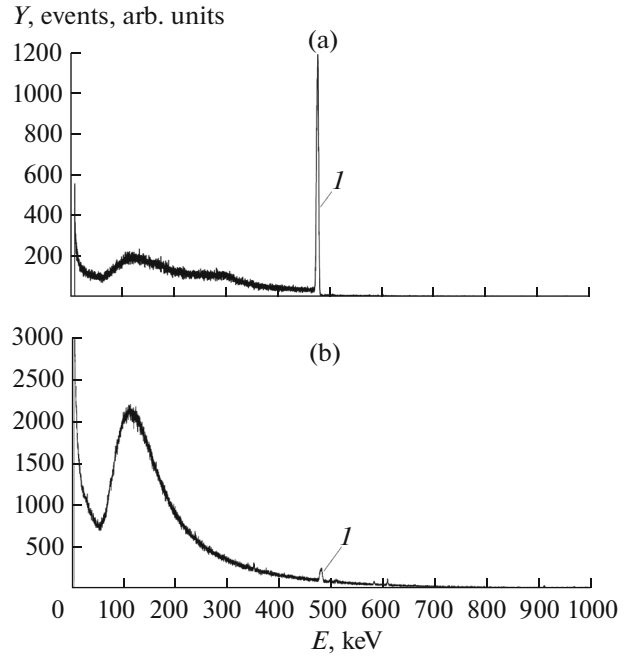


Fig. 3. Spectrum from the HPGe γ spectrometer: (a) thick lithium layer; (b) thin lithium layer.

(Tomsk). The sputtering was carried out in an atmosphere of high-purity nitrogen (99.9%), so the target contains not only natural boron of crystalline density, but also boron compounds with nitrogen. To determine the rate of boron deposition on copper and aluminum substrates, microinterferometry was carried out using the MII-4 device at five points on the surface of the substrate from which the coating was removed. The average rate coating application under experimental conditions is 8.4 nm/min, and the expected coating heterogeneity is 10%. To determine the composition of the target, the micro-X-ray spectral analysis was used. The coating was found to primarily contain boron and nitrogen in proportion of 84.5 and 15.5 at %, respectively (Fig. 4).

Also, the Institute of High Current Electronics conducted an analysis of the roughness of boron films using a Solver P47 atomic force microscope. It has been established that the heterogeneity of the coating thickness at the macroscopic level is associated with the specific features of the magnetron discharge operations. The data presented in Fig. 5 show that the roughness R_a of the surface of a $1 \times 1 \mu\text{m}$ boron coating segment with a thickness of 1 μm is about 0.2 nm, which is three orders of magnitude greater than the coating thickness. Thus, the coating reproduces the relief of the substrate surface, and its surface is quite uniform at the microscopic level.

An additional measurement of the boron film thickness was carried out using the Linnik MII-4 microinterferometer at the Institute of Nuclear Physics (Novosibirsk). With this method, two arrays of rays irradiate the boron film and are reflected: the first rays

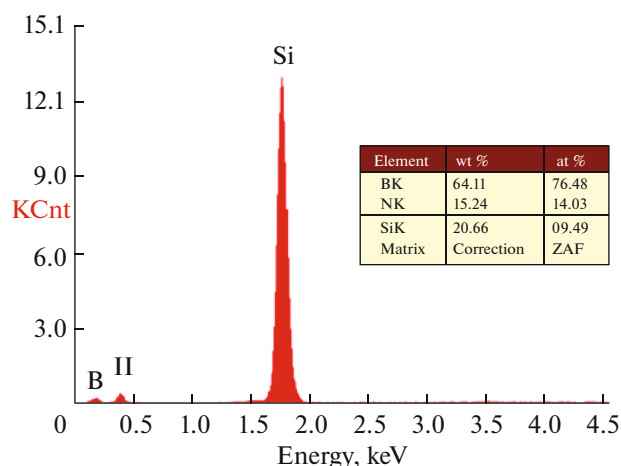


Fig. 4. Analysis of the composition of the boron coating deposited on the KEF-25 silicon substrate with the (100) crystal orientation carried out using a Philips SEM 515 scanning electron microscope with an EDAX ECON IV microanalyzer attachment.

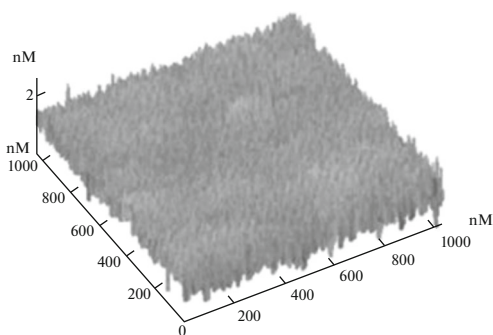


Fig. 5. Morphology and surface characteristics of a 1 μm thick boron coating deposited in a nitrogen atmosphere on a silicon surface, obtained using a Solver P47 scanning atomic force microscope with an NSG-01 cantilever.

is reflected from the surface of the film, while the second passes through it and is reflected from the copper substrate (Fig. 6). There is no phase shift between the beams, since the beams do not interfere with each other, thus, using Snell's law of refraction and the

refractive index of boron, equal to three, we obtain a film thickness of 1.1 μm with an accuracy of 20%.

In situ methods for determining the thickness of the boron are similar to those described above for lithium. We now consider in more detail the method of comparing the yield of α particles from a thin and thick boron target [5, 10]. First we measured the yield of alpha particles from thin and thick targets. Similar to the lithium target, the thick boron target is a target in which protons are slowed down to an energy below the particle generation reaction threshold. Knowing the dependence of the particle yield on the proton energy and measuring the particle yield from thin and thick targets, the proton energy and, as a consequence, the thickness of this layer can be determined. A boron carbide plate was used as a thick target, and the aforementioned boron target was used as a thin target. The measurements were carried out at proton energies of 0.5 and 0.7 MeV; the α spectrometer was installed at an angle 168° . The signal generated by the α spectrometer, normalized to the proton flux, is shown in Fig. 7. The spectrum to the left of the 400–700 channels corresponds to back-scattered protons, and the spectrum to the right of these channels corresponds to α -particles in the $^{11}\text{B}(p, \alpha_1)^8\text{Be}^*$ reaction. It can be noted that the number of detected α particles from a thick target is greater than that from the thin one by a factor of 2.10 at an energy of 0.5 MeV and 3.95 at an energy of 0.7 MeV. Using Eq. (1), we calculated the energy loss of the proton. Taking into account the content of carbon in the thick target and nitrogen in the thin target, determined after processing the spectrum from the α spectrometer, we found that the linear density of boron in the thin boron target is $8.48 \times 10^{18} \text{ at/cm}^2$ (0.65 μm of crystalline-density boron) with an accuracy of 10%. This estimate is in good agreement with the results of the previously presented methods.

4. CONCLUSIONS

Using the implemented in situ method for measuring the thickness of a thin target with high accuracy and a number of additional techniques, our group has measured 14 cross sections of nuclear reactions in the interaction of lithium and boron with proton and deu-

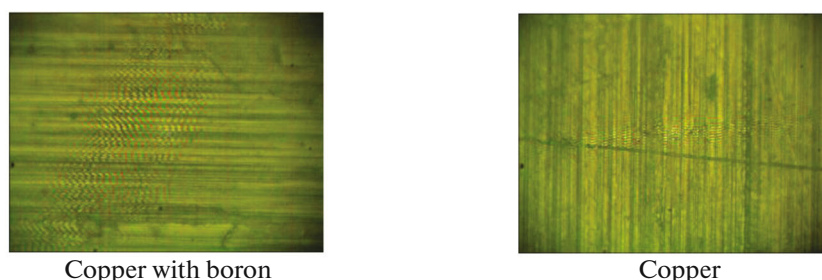


Fig. 6. Images of a boron target made using a Linnik microinterferometer.

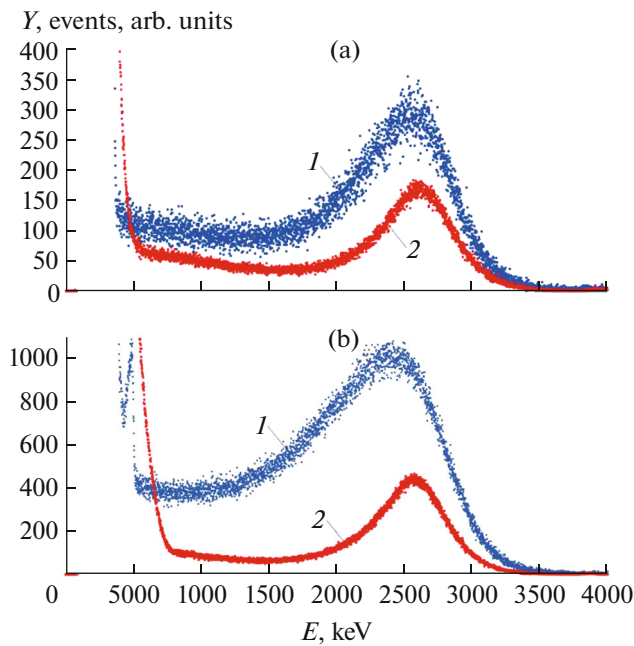


Fig. 7. Spectrum from the α -spectrometer at a proton beam energy of 0.5 (a) and 0.7 MeV (b): (1) thick target, (2) thin target.

teron beams [9–13]: ${}^7\text{Li}(p, p'\gamma){}^7\text{Li}^*$, ${}^7\text{Li}(p, \alpha){}^4\text{He}$, ${}^6\text{Li}(d, \alpha){}^4\text{He}$, ${}^6\text{Li}(d, p){}^7\text{Li}$, ${}^6\text{Li}(d, p){}^7\text{Li}^*$, ${}^7\text{Li}(d, \alpha){}^5\text{He}$, ${}^7\text{Li}(d, n\alpha){}^4\text{He}$, ${}^{11}\text{B}(p, \alpha_0){}^8\text{Be}$, ${}^{11}\text{B}(p, \alpha_1){}^8\text{Be}^*$, ${}^{10}\text{B}(d, \alpha_0){}^8\text{Be}$, ${}^{10}\text{B}(d, \alpha_1){}^8\text{Be}^*$, ${}^{10}\text{B}(d, p_2){}^9\text{Be}^*$, ${}^{11}\text{B}(d, \alpha_0){}^9\text{Be}$, and ${}^{11}\text{B}(d, \alpha_2){}^9\text{Be}^*$.

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CONFLICT OF INTEREST

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

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