doi: 10.56761/EFRE2024.S4-O-052601

Comparison of use of TPU accelerators for nuclear analysis

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Abstract. A comparison was made of the use of ion beams accelerated with the U-120M cyclotron and the TEMP-4M vacuum diode of the Tomsk Polytechnic University for nuclear analysis of light elements. Cyclotron ion beams make it possible to solve a wide range of problems of elemental analysis, including determining the mass thickness and stoichiometry of coatings and their contamination with gaseous impurities, but they are an expensive tool. A cheap and portable TEMP-4M accelerator operating in a Plyutto-Luce diode mode at voltage of 200–350 kV produces nanosecond ion bunches of up to 10^{14} ions with ion energies randomly distributed within the range of 0.5–0.7 MeV/amu from shot to shot. Sufficiently high yields of (d,n) and (d,p) nuclear reactions even at such low energies allows radioactivation analysis of light elements from Li to Si with a reasonable sensitivity. It has been experimentally shown that irradiation with single nanosecond deuteron bunches with a number of up to $2 \cdot 10^{13}$ deuterons per shot and an energy of 1–1.4 MeV allows radioactivation determination of the stoichiometry of thin AlN, BN and B_4C coatings based on the ratios of $^{28}Al/^{15}O$, $^{15}O/^{11}C$ and $^{13}N/^{11}C$ radionuclides, respectively.
Keywords: ion beam analysis, collective ion acceleration, cyclotron, vacuum diode.

1. Radioactivation analysis

When studying mass transfer in solids, stimulated by Intense Pulsed Ion Beams (IPIBs), in most cases it is necessary to obtain information about the concentration of elements in depth and/or the mass thickness of the deposited coating and its elemental composition, e.g., stoichiometry of compounds and gaseous (C, N, O) contaminations. Ion Beam Analysis (IBA) methods are not sensitive to the chemical state of elemental atoms, and therefore they avoid the errors inherent in such methods for determining elements and their depth distributions as SIMS and AES, at least when using IBA as reference methods for certifying reference samples [1].

1.1. Radioactivation with cyclotron ion beams

Beams of accelerated ions from the U-120M cyclotron of TPU have been used to characterize the processing of solids with IPIBs since the 1990s [1–4]. The cyclotron allows acceleration of protons (6.8–10.7 MeV), deuterons (13.6 MeV), alpha particles (27.2 MeV) and heavy ions like ^{12}C (8.5–21 MeV), ^{14}N (10.6–17.5 MeV) and ^{16}O (21 MeV). Reducing the energy of the light ions by foils allows for some matrices to implement nondestructivel options for determination of elements from Li to Fe at elevated levels of their content (10–100 ppm) by using pure Ge Canberra detectors for the registration of the induced radioactivity. Depending on the half-life of analytical radionuclides, irradiation of the series of samples, including reference samples, is carried out simultaneously on a rotating target holder or in turn. In the latter case, the ion flux is determined by the activity of a thin foil (monitor) installed in front of the samples.

The content of the element in the studied samples C_i is determined after introducing corrections for the decay of radionuclides using the following formula:

$$C_i\ (\text{мас.}\%) = C_R \times (A_i/A_R) \times (M_R/M_i) \times (p_i/p_R), \quad (1)$$

where C_R is the element content in reference samples; A_i and A_R, are activities of analytical radionuclides in the studied and reference samples, respectively, while M_i and M_R are activities of analytical radionuclides in the monitor foils, given at the end of irradiation; p_i and p_R are corrections for the range of ions in the studied and comparison samples, respectively.

Thus, using deuteron beams accelerated at the cyclotron, the mass thickness of DLC coatings [3] and contents of C, N and O in W and Au coatings [3, 4] were determined by radioactivation

analysis based on (d,n) nuclear reactions – see Table 1. These samples of W and Au coatings were further used as reference samples to determine the profiles of C, N and O along the depth of the coatings using the AES method with ion etching of layers [3].

Table 1. Deuteron induced nuclear reactions for non-destructive analysis.

#	Nuclear reaction, E_{th} (MeV)	B_C, MeV	Abund., at%	$T_{1/2}$, s	Eγ, keV (intensity, %)
1	^{6}Li(d,n)^{7}Be	0.6	7.6	4589568	477.6 (10.52)
2	^{10}B(d,n)^{11}C	1	19.9	1223	511 (200)
3	^{12}C(d,n)^{13}N, 0.33	1.1	99	598	511 (200)
4	^{14}N(d,n)^{15}O	1.3	99.64	122.24	511 (200)
5	^{16}O(d,n)^{17}F, 1.83	1.5	99.76	64.49	511 (200)
6	^{19}F(d,p)^{20}F	1.6	100	11	1633.6 (100)
7	^{23}Na(d,p)^{24}Na	2	100	53852	1369 (100)
8	^{25}Mg(d,n)^{26m}Al	2.2	10	6.34	511(200)
9	^{26}Mg(d,p)^{27}Mg	2.2	11	567.48	843.7 (71.8)
10	^{27}Al(d,p)^{28}Al	2.3	100	134.4	1779 (100)
11	^{28}Si(d,n)^{29}P	2.5	92.2	4.14	511 (200)
12	^{29}Si(d,n)^{30}P	2.5	4.685	149.9	511 (200)

The stoichiometry of BN and AlN was determined with accuracy of less than ±1% using cyclotron ion beams via nuclear reactions ^{10}B(p,α)^{7}Be, ^{14}N(p,n)^{14}O, ^{10}B(^{12}C,α)^{18}F, ^{14}N(d,n)^{15}O and ^{27}Al(d,p)^{28}Al. The mass thickness of AlN and TiN coatings deposited with a thickness of 2–5 μm by magnetron sputtering on silicon substrates was determined by irradiating the samples on a rotating target holder with a cyclotron proton beam exposed to air, followed by recording the induced γ-activity of radionuclides formed in samples and foil monitors [5]. All samples analyzed using cyclotron ion beams were subsequently used as reference samples to test a new radioactivation method using nanosecond deuteron beams collectively accelerated in a compact Plyutto-Luce diode.

1.2. Radioactivation with nanosecond ion bunches

A cheap and portable TEMP-4M accelerator operating in a Plyutto-Luce diode mode at voltage of 200–350 kV produces nanosecond ion bunches of up to 10^{14} protons and $2\cdot10^{13}$ deuterons [6, 7] with specific ion energies randomly distributed within the range of 0.5–0.7 MeV/amu from shot to shot. Deuteron energies of 1–1.4 MeV are enough to perform radioactivation analysis of light elements (up to Si) via (d,n) and (d,p) nuclear reactions with a reasonable sensitivity. Ion energy is determined in each individual shot of the diode by measuring drift velocity of a virtual cathode (VC) with a ToF spectrometer of electrons emitted by the VC normally to the axis of the VC propagation from anode to target [8]. The random "scanning" from shot to shot across ion energies in the range of 0.5–0.7 MeV/amu enables the analysis of thin coatings.

The main problem when analyzing thin coatings is the possible overheating and destruction of the coating by nanosecond bunches of ions, which is aggravated by the inevitable capture of protons in the collective acceleration, the number of which in a bunch is usually 5 times greater than the number of deuterons. The contribution to the absorbed energy from protons is on average 2.5 times higher than from deuterons, which can increase the temperature of coatings and substrates when irradiated with single shots to 400–450 °C. The use of Teflon anodes instead of anodes made of deuterated polyethylene makes it possible to reduce the parasitic contribution of protons by an order of magnitude – down to $4\cdot10^{12}$ per shot [9], however, this requires the use of a special pulsed gas source of deuterium or collective acceleration of deuterons from the residual deuterium atmosphere

[7]. The use of a residual helium atmosphere also makes it possible to significantly reduce the number of protons in the bunch [10], thereby reducing the thermal effect on the coating structure.

It has been experimentally shown that irradiation with single nanosecond deuteron bunches with a number of up to $2 \cdot 10^{13}$ deuterons per shot and an energy of 1–1.4 MeV allows radioactivation determination of the stoichiometry of thin AlN coatings on Si substrates, based on the ratio of $^{28}Al/^{15}O$ radionuclides with an error of no worse than ±4% [11]. Thanks to random "scanning" of deuteron energy from shot to shot, three samples of AlN films with average thicknesses of 4.2, 4.6 and 5.7 μm gave three branches of solutions from the "trunk" of the 28Al/15O ratios for thick stoichiometric samples – see Fig.1. Based on the ratios of radionuclides $^{15}O/^{11}C$ and $^{13}N/^{11}C$, the stoichiometry of the BN and B_4C plates was also determined – see Fig.2.

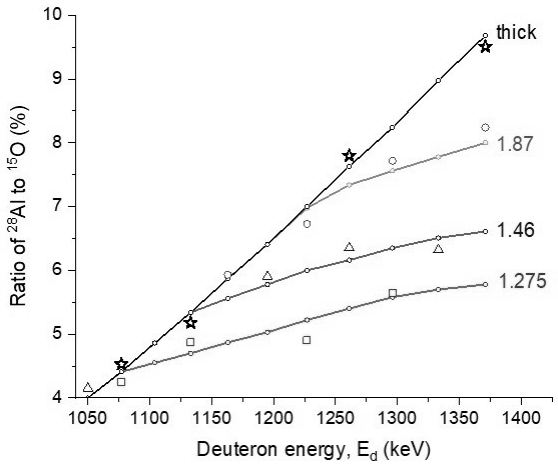

Fig. 1. Ratios of $^{28}Al/^{15}O$ activities in the AlN thin coatings and thick plates.

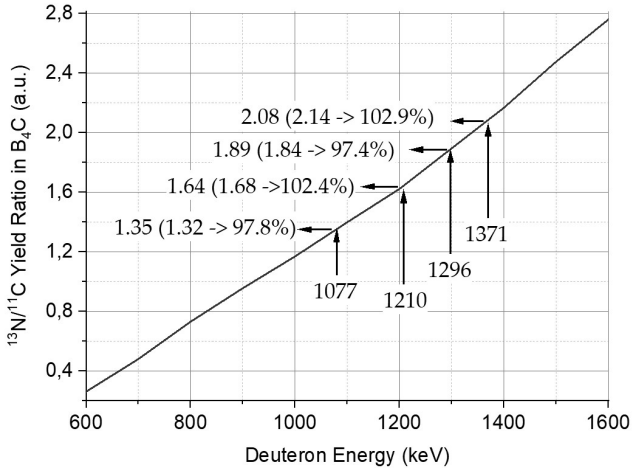

Fig. 2. Ratios of $^{13}N/^{11}C$ activities in a B_4C ceramic plate.

2. Possibilities for the nuclear depth profiling

The electrostatic generators allow a well-controlled and precise scanning by proton energy, thus providing the best depth resolution. The electrostatic generator EG-2 (up to 2 MeV) of TPU was intensively used for depth profiling via the $^{27}Al(p,\gamma)^{28}Si$ resonant reaction of Al ions implanted in different solids with the MUK-M diode [1–3]. Typical depth profiles of Al ions implanted in $Cd_xHg_{1-x}Te$ and Si plates, obtained by energy scanning EG-2 at the resonance of 991.8 keV, are shown in Fig.3–4: PIGE and SIMS depth profiles of Al in the same Si sample look similar.

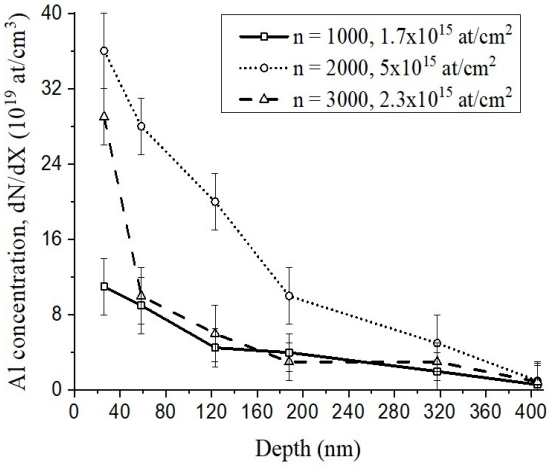

Fig. 3. Depth profiles of ^{27}Al ions implanted in $Cd_xHg_{1-x}Te$.

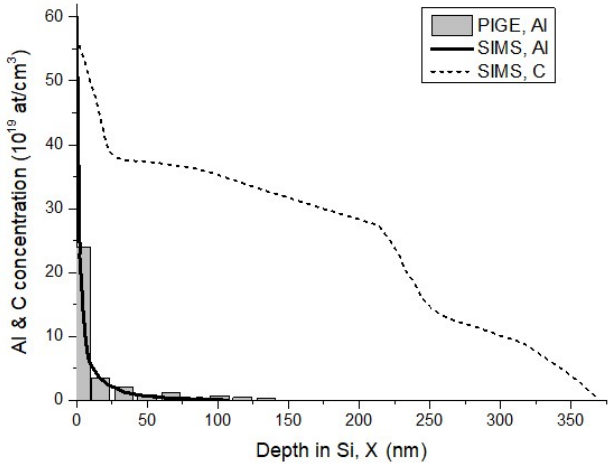

Fig. 4. PIGE and SIMS depth profiles of ^{27}Al and ^{12}C ions implanted in Si.

Due to the random "scanning" of proton energy from shot to shot in the Plutto-Luce diode, it is interesting to use such a simple energy "scanning" for depth profiling of light elements. The prompt γ-rays can be registered with two fast plastic scintillators each filtered by lead layers having different thickness, thus providing two gradations of γ-ray energy, low and high [12]. A reliable control of the number of protons in each diode shot still remains the main problem of a reliable depth profiling of light elements with the nanosecond proton bunches collectively accelerated in a Plyutto-Luce diode.

For the depth profiling of hydrogen, the use of resonances of the $^{1}H(^{19}F,\alpha\gamma)^{16}O$ nuclear reaction, characterized by high yields (10^{-6} and higher), is of great interest. In this regard, bunches of ^{19}F ions must be accelerated to an energy of up to 16 MeV (~0.8 MeV/amu), although "scanning" at an energy of about 6.8 MeV (~0.36 MeV/amu) may allow obtaining some semi-quantitative results for determining hydrogen depth profiles. Collective acceleration of fluorine ions in a Luce diode is possible both from the near-anode plasma when using a Teflon anode, and from the residual atmosphere of freons like carbon tetrafluoride (CF_4). In both cases, carbon ions are also captured in a collective acceleration [13], i.e. together with fluorine ions, thus reducing their number and/or energy. For this reason, it is important to search for optimal parameters for the collective acceleration of fluorine ions in the presence of a significant admixture of carbon ions. This problem is still under our study.

3. Conclusion

The use of a compact, radiation-safe Plyutto-Luce diode for Ion Beam Analysis look as a promising tool for non-destructive determination of light elements. It can greatly expand the capabilities of small laboratories and opens up new opportunities for students to acquire practical skills in the field of experimental nuclear physics.

Acknowledgement

The work was supported by a grant Russian Science Foundation № 23-19-00614, https://rscf.ru/en/project/23-19-00614.

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