Integrated target-ion source unit for on-line production of radioactive short-lived isotopes

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Abstract. A new version of integrated target-ion source unit (ionising target) has been developed for the on-line production of radioactive single-charged ions. The target is able to withstand temperatures up to 2500 °C and acts also as an ion source of the surface and laser ionisation. Off-line and on-line experiments with the ionising target, housing tantalum foils as a target material, have been carried out at the IRIS (Investigation of Radioactive Isotopes on Synchrocyclotron) facility. The off-line surface ionisation efficiency measured for stable atoms of Li, Rb and Cs was correspondingly 6%, 40% and 55% at the target temperature of 2000 °C and 3–10% for atoms of rare-earth elements Sm, Eu, Tm and Yb at a temperature of 2200 °C. The off-line measured values of the ionisation efficiency for stable Gd and Eu atoms by the laser beam ionisation inside the target were 1% and 7%, respectively. The radioactive beam intensities of neutron-deficient rare-earth nuclides from Eu to Lu produced by the integrated target-ion source unit have been measured at a temperature of 2500 °C. The results of the integrated target-ion source unit use for on-line laser resonance ionisation spectroscopy study of neutron-deficient Gd isotopes have been also presented.

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1 Introduction

The objective of this work is to develop an effective, high-temperature target unit for on-line production of short-lived radioactive isotopes. The construction of the target unit allows to avoid cold spots usually arising in the target-ion source assemblies in the places of the target - transfer tube - ion source connection. The peculiarity of the developed target unit is the absence of the ion source [1]: since the ionisation process happens in the target volume itself, it can help to avoid additional delay time due to the effusion inside the transfer tube and the ion source. It could be especially useful for production of isotopes of elements with long sticking time, such as isotopes of rare-earth elements Gd and Lu and of many other nuclides with boiling points higher than 3000 °C. The use of a tungsten container [2] instead of a traditional tantalum one allowed to rise the target working temperature up to 2500 °C, thereby decreasing the delay time for nuclides produced in the target and hence increasing the yield of short-lived ones. A high temperature of the target material and the absence of cold spots in the target volume are crucial for effective production of isotopes of many refractory elements delayed in the target volume by the effusion determined process. In the first off-line and on-line tests the integrated target-ion source unit having the thickness of the target material 3 g/cm² was used [3] to get reference values for on-line production efficiencies of measured isotopes. Small target dimensions allowed to avoid more effectively a possible appearance of cold spots on the target container and heat it up to a higher temperature. The developed target-ion source unit can be used on heavy-ion projectile beams and may be considered as a prototype of a massive, thick target (mass of some hundred grams) for an effective utilization of projectile beams of light particles of high energy.

2 Ionising target construction

The schematic diagrams show the high-temperature ionising target [3] which was used for off-line and on-line tests (fig. 1 and fig. 2). The target container was made of a very tightly rolled multi-layer tungsten foil and had a wall

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Fig. 1. Scheme of the high-temperature ionising target used for off-line tests.

thickness of 0.2–0.3 mm. From both sides it was closed by tantalum plugs with the holes for the ion extraction (right) and laser beam introducing (left) into the target volume. Inside the tungsten outer container a tantalum cylinder containing the target material (tantalum foil of 20 micron thickness) supported by tantalum plugs from both sides was placed. The only difference in the construction of the ionising target (IT) used for off-line ionisation efficiency measurement was the presence of a separately heated oven, which was connected to the IT for a slow evaporation of the sample, consisting of a known amount of atoms of the element being investigated.

For off-line measurements of the surface ionisation efficiency of alkalis Li, Rb, Cs, rare-earth elements Sm, Eu, Tm and Yb and for laser ionisation efficiency of Eu and Gd the same IT unit was used.

Dimensions and geometry of the integrated target-ion source (ITIS) used for off-line and on-line tests were the following ones: length = 45 mm; outer tungsten container diameter = 12 mm; inner tantalum cylinder diameter =10 mm; ion extraction hole diameter = 1.5 mm; diameter of the hole for the passing laser beam = 2 mm. The diameter of the axial hole in the tantalum foils for the passing laser beam was 3 mm. The target container can be heated by DC or AC passing through it. For the resonance laser ionisation tests three laser beams from the IRIS laser installation [4] brought together were led into the target container through the hole in the left tantalum plug and directed along the target central axis. For the first tests a target thickness of 3 g/cm^2 was implemented. Such a value of the target material thickness was selected to ensure its packing fraction to be about 25% that gives a large free space between target material foils for faster release of species having long sticking times, such as Gd and Lu.



Fig. 2. Scheme of the high-temperature ionising target used for on-line tests.

In the course of off-line and on-line experiments the target temperature was varied in the interval 2000–2500 °C.

3 Off-line measurements of the ionisation efficiencies for Li. Sm. Eu. Tm and Yb

Off-line measurement of the surface ionisation efficiencies for Li and rare-earth elements were carried out by using samples of lithium fluoride and oxides of pointed out rareearth elements weighing from 10 to 20 μ g placed into the oven connected with the IT volume. The current of each evaporated sample was measured by a Faraday cup placed in the focal plane of the mass-separator magnet and connected with the current integrator. In the course of the off-line efficiency measurements some samples were used to observe the current of molecular ions of investigated species. There have not been measured noticeable traces of the lithium fluoride ions at the mass position 25 and 26 at the target temperature 2000 °C and at higher temperatures. As to rare-earth element oxides, only molecular ions of gadolinium have been observed. The measured ratio of GdO^+ and GdO^{2+} to Gd^+ current was correspondingly 25% and 10% at a temperature of 2500 °C.

For the element being investigated, the measured integrated current is easily put into relation with the number of singly charged ions which generated it; the ratio of this number to the amount of atoms of the same element contained initially in the sample gives the lower limit of the ionisation efficiency. Since the measured current belongs only to a single isotope, the isotopic ratio should

Table 1. The efficiencies of ionisation for Li, Rb, Cs, Sm, Eu, Gd, Tm and Yb measured off-line with IT and LT. The error of the ionisation efficiency measurement was 20% for Rb and Cs and up to 40% for other elements.

Element	Temperature (°C)	Surface ionisation efficiency (%)	Laser ionisation efficiency (%)
Li	2000	6	
Rb	2000	40	
Cs	2000	55	
Sm	2200	7	
Eu	2200	10	7
Gd	2500	2	1
Tm	2200	3	
Yb	2200	5	

be taken into account when calculating the ionisation efficiency. The process of the laser ionisation efficiency measurement of Eu and Gd atoms inside the target volume was carried out simultaneously with the surface ionisation measurement. For that purpose three laser beams tuned to the resonance with the appropriate transitions in Eu and Gd were introduced into the ITIS. We indicate this method as the laser target (LT) [5], in order to emphasize that it differs from the method employing the resonance ionisation in the laser ion source (LIS) [4].

In table 1 the efficiencies of surface ionisation measured off-line for Li, Rb, Cs, Sm, Eu, Gd, Tm, Yb and resonance laser ionisation for Eu and Gd, making use of the integrated target-ion source unit are presented. During that measurements and on-line tests the containers of both target units were heated by the AC. For the ion source heating the DC has been used. For ionisation of the pointed out elements the temperature of the IT was kept in the interval of 2000–2500 °C. The oven temperature was increased very slowly to avoid a fast uncontrolled evaporation of the sample being investigated.

4 On-line measurements of the beam intensities of neutron-deficient rare-earth isotopes from the ionising target

For on-line testing of the IT and LT the construction shown in fig. 2 has been used: a 1 GeV proton-beaminduced spallation reactions in the target material (tantalum 20 μ m foils, target thickness = 3 g/cm²), producing nuclides of neutron-deficient isotopes of rare-earth elements. After surface or laser ionisation and extraction from the target, the ions were separated by a massseparator and implanted into a moving tape installed at one of the three beam lines of the IRIS facility. Identification of the radioactive implanted specimens and radioactive beam intensity measurements were performed by means of appropriate γ -line measurements by a coaxial high-purity germanium detector. The proton beam inten-

Tab	le 2.	On-lin	e rad	ioactive	beam	intensiti	es of	some	rare-
earth	n isoto	opes fro	m the	e IT at 2	$500 ^{\circ}\mathrm{C}$	(proton	beam	intens	sity =
0.05	μA ; t	arget t	hickn	ess = 3	g/cm^2).			

Nuclide	$T_{1/2}$	Radioactive beam
		intensities, s^{-1}
¹³⁸ Eu	$12.1 \mathrm{~s}$	2.4×10^3
¹³⁹ Eu	$17.9 \mathrm{\ s}$	1.6×10^4
$^{141}\mathrm{Eu}$	41.4 s	4.0×10^5
$^{142}\mathrm{Eu}$	1.22 min	4.1×10^5
$^{143}\mathrm{Eu}$	$2.57 \min$	2.0×10^6
$^{139}\mathrm{Sm}$	$2.57 \min$	2.8×10^5
$^{143}\mathrm{Sm}$	66 s	4.4×10^5
$^{139}\mathrm{Pm}$	4.15 min	3.1×10^5
$^{160}\mathrm{Tm}$	9.4 min	5.0×10^5
$^{164}\mathrm{Tm}$	$5.1 \mathrm{min}$	9.6×10^4
160 Yb	4.8 min	1.0×10^6
^{160}Lu	40 s	2.2×10^4
¹⁶⁸ Lu	$6.7 \min$	1.3×10^6
$^{143}\mathrm{Gd}$	39 s	6.7×10^4
$^{143\mathrm{m}}\mathrm{Gd}$	$1.87 \min$	
$^{145}\mathrm{Gd}$	23.0 min	6.7×10^5
$^{145\mathrm{m}}\mathrm{Gd}$	$85.2~{\rm s}$	

sity was 0.05 μ A. The radioactive beam intensities were calculated according to the well-known equation [6]. In table 2 the beam intensities of the neutron-deficient rareearth isotopes produced on-line have been presented. For ¹⁴³Gd and ¹⁴⁵Gd the total values of the beam intensities are given summarizing the measured isomeric and ground states.

5 The ITIS use for on-line laser spectroscopy experiment

The developed integrated target-ion source unit has been used for a laser resonance spectroscopy investigation of neutron-deficient Gd isotopes [5]. The scheme of the ITIS used for the laser spectroscopy experiments has been shown in fig. 2.

For the comparison of two methods (LIS and LT) the ratio R of the photoion current to the thermal ionisation background has been measured for two types of the target system: the first one was a traditional target-ion source assembly (target connected to the ion source) [4] and the second one was the target with the laser ionisation inside its volume (laser target). In both experiments radioactive sources were collected on the tape (during 30 s) and then gamma spectrum was measured during the same interval. This cycle was repeated 5 times. In table 3 the beam intensities of ^{145m}Gd and ^{145g}Gd from both tested target systems at the equal temperature conditions is presented. During the tests the targets having an identical material

	^{145m} Gd beam	h intensity, s^{-1}	$^{145\mathrm{g}}\mathrm{Gd}$ beam intensity, s^{-1}		Ratio $^{145m}Gd/^{145g}Gd$	
Type of target-source unit used	Target with ion source	Laser target	Target with ion source	Laser target	Target with ion source	Laser target
Laser at resonance	$1.64(8) \times 10^5$	$3.21(11) \times 10^5$				
Laser off	$1.55(6) \times 10^5$	$2.30(8) \times 10^5$	$2.02(20) \times 10^6$	$4.40(16) \times 10^5$	$7.7(1.1) \times 10^{-2}$	$5.20(37) \times 10^{-1}$
R	0.06(6)	0.40(6)				

Table 3. Beam intensities of ^{145m}Gd and ^{145g}Gd produced by different target-source units at a temperature of 2500 °C.

thickness were used (3 g/cm^2) , which was irradiated in both experiments by the same proton beam intensity $0.05 \ \mu$ A. Also we were trying to keep the same temperature conditions for the tested targets and the ion source. The temperature was 2500 ± 50 °C that ensured a rather fast escape of Gd radioactive atoms for which the effusion is considered to be the main delay dominating process. In spite of similar conditions of radioactive Gd production from both targets, we got from the laser target by the surface ionisation the yields 1.5 times higher than from the usual target-ion source unit (see table 3 "laser off"). But, if the concentration of radioactive Gd neutrals would be the same in both cases, we should get from the LT the Gd ion current 4 times lower than from the usual construction of target-ion source. The reason of a lower surface ionisation efficiency in the volume of the laser target is due to the fact that the ionisation mainly is happening on the surface of the target material (tantalum foil) which have a work function $\varphi = 4.19$ eV, but in the case of the target-ion source unit the surface ionisation is inside the tungsten ion source ($\varphi = 4.53$ eV). Therefore the Gd ionisation efficiency (according to Saha equation) inside the LT should be about 4 times lower than in the tungsten ion source of the usual target construction. So, comparing only surface ionisation currents of radioactive ^{145m}Gd from both targets we come to conclusion that we have an enhancement of the concentration of Gd radioactive neutrals in the case of the LT about $1.5 \times 4 = 6$ times. If there is an enhancement of an isotope with its half-life $(T_{1/2} = 1.87 \text{ min in the case } {}^{145 \hat{\text{m}}}\text{Gd})$ in the pointed out experimental conditions, it can signify that the integrated source-target is faster than the usual target-ion source unit. One of the natural explanations of a lower density of neutrals inside the tungsten ion source is that Gd radioactive neutrals "are dying" sticking with cold places of the target-transfer tube-source connections, as it is well known that Gd has one of the longest sticking times among all rare-earth elements. This consideration is confirmed by the fact (as one can see from table 3) that the ratio of ion currents of ^{145m}Gd and ^{145g}Gd $(T_{1/2} = 23 \text{ min})$ from the laser target is almost seven times higher than from the target of a usual construction.

6 Discussion

Off- and on-line tests of ionising targets have demonstrated that they can be used for the on-line production of a large set of nuclides using surface and resonance laser ionisation. As one can see from table 1, the ionisation efficiency measured off-line for surface and laser ionised different species is comparable with those published in the literature [2,7]. A low ionisation efficiency value for Li can be explained by a low temperature of the ionising target. To ensure a higher Li ionisation efficiency, a target temperature should be at least 2200 °C, or even higher. The ionisation efficiency for the tested elements can be increased in our construction almost two times by decreasing the diameter of the hole for the laser beam from 2 to 1 mm, as the laser beam diameter is about 0.5 mm.

As is seen from table 2, the target can be successfully used for production of neutron-deficient isotopes of all rare-earth elements. The ionisation efficiency may be higher about 3 times, if the ionising target is used only in surface ionisation mode, when the hole for the laser beam introduction is eliminated.

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