DEVELOPMENT AND ON-LINE TESTS OF DIFFERENT TARGET-ION SOURCE UNITS FOR PRODUCTION OF NUCLIDES FAR FROM STABILITY

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1. Introduction

The work carried out has as a general goal the development of a massive (up to 1 kg) uranium carbide target prototype for the third generation ISOL facilities, where two-step reactions will be used to produce intense neutron-rich nuclear beams.

For more than thirty years UC_x targets have been explored for the production of a large set of neutron-rich and neutron-deficient heavy nuclei. The main line of uranium carbide target development was the creation of the target material with a high dispersivity and porosity in order to decrease the release time of the species produced by different reactions in the target material, which is mainly determined by the diffusion and effusion processes. On the other hand, for a more efficient use of a neutron beam after the primary beam converter the density of the target material containing uranium should be as high as possible. Additionally, the target material with a higher density has a higher thermal conductivity that would allow dissipating a higher power introduced into the target by the primary beam, when the target is used for exotic nuclei production in the direct reaction. Hence, for more effective production of short-lived isotopes of the element chosen for the investigation, a special selection of a metal carbide target material is required – concerning not only the target material structure but also its density. At the IRIS (Investigation of Radioactive Isotopes at Synchrocyclotron) facility in Gatchina uranium carbide target materials of different structure and density have been tested.

The first general goal of the work is to find out whether the high density uranium carbide can be competitive with presently used low density UC_x targets in the production of short-lived nuclei far from stability. The yields and release characteristics are reported here to compare properties of tested target materials. The second goal is the development and tests of an effective, high-temperature target unit for the on-line production of short-lived rare nuclides. The peculiarity of the developed target unit is the absence of the ion source [1]. The ionization process happens in the target volume itself, so it can help to avoid an additional delay time due to the effusion inside the transfer tube and the ion source. The combined target-ion source (CTIS) unit could be especially useful for the production of isotopes of hard volatile elements with long sticking time, such as rare-earth elements Gd and Lu, and of many other nuclides with boiling points higher than 3000°C. The use of 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 isotopes. At the same time, a high work function of tungsten ensures a high efficiency of a surface ionization of the species with the ionization potentials lower than 6 eV produced in the target material.

2. Target materials tested on-line

A schematic view of the typical target unit used for different uranium carbide target material tests is shown in Fig. 1. As a first step in that comparison, the isotopes of alkali elements Rb, Cs and Fr have been selected for the yield and release efficiency measurements. The main reason of such a choice was their high ionization efficiency (theoretically up to 100%) on the hot inner surface of the target-ion source unit, that allows to obtain short-lived nuclei far from stability. This is very important for the target material comparison.

The second important point in the selection of alkalis is that the target construction can be very simple, including only the tungsten target cavity (ionizing target) [3], as the ionization process happens inside the target container itself. It allows to avoid some uncertainties concerning possible temperature difference of the tested units, as in that case only one parameter (the target container temperature) should be controlled in the course of the experiment.

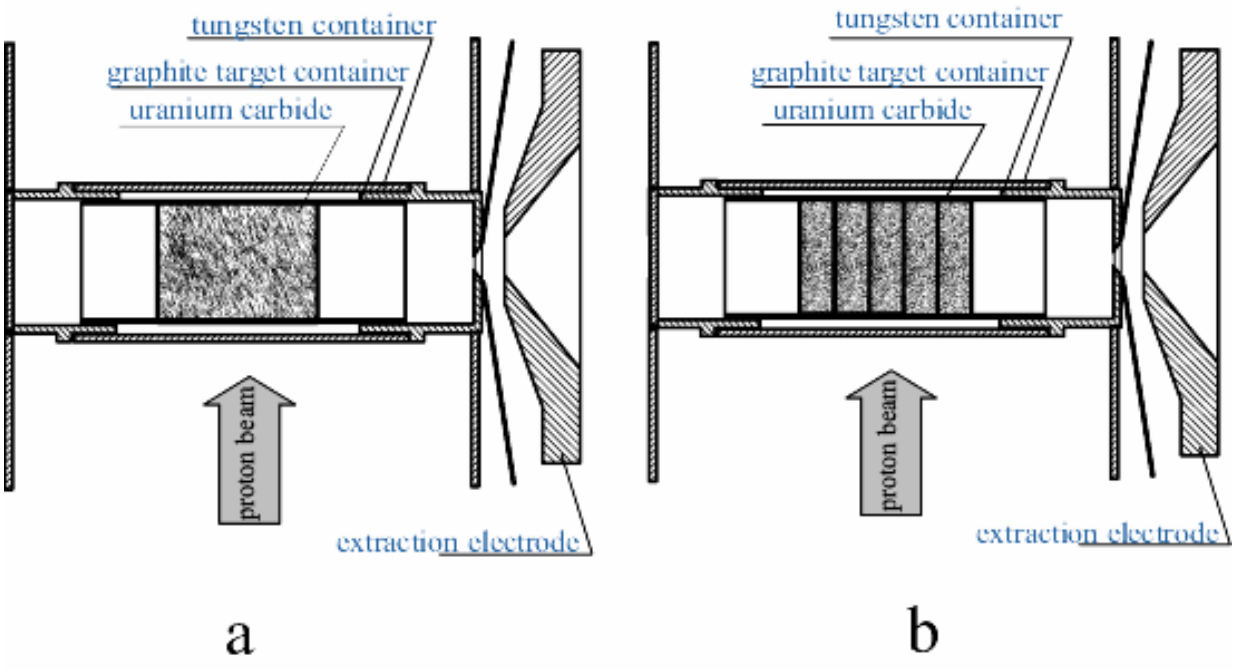

Fig. 1. Schematic drawings of a) high density rod (HDR) uranium carbide target; b) high and low density pellet uranium carbide target

Three kinds of the target materials were studied: a high density UC rod; a low density UC_x target prepared by the ISOLDE method at the IRIS facility (as few pellets placed transversely to the axis of the target); a high density UC powder target material prepared by the method of the powder metallurgy. The high density rod (HDR) target had the following characteristics: uranium rod density 11 g/cm^3, target thickness 6.7 g/cm^2, about 6 mm length and 11 mm in diameter. The grain size was about 200 μm. The low density target (LDT) had: uranium density 2.3 g/cm^3, target thickness 2.8 g/cm^2, about 12 mm length and 11 mm in diameter; 8 pellets, each of 1.2–1.6 mm thickness with the grain size about 20 μm. This target material has been prepared by the Orsay–PNPI collaboration at the IRIS facility, using the ISOLDE technology, a similar graphite powder brought by Orsay group and uranium dioxide from Russian producers. This target is also referred later as ISOLDE like or PARRNe-IRIS. The high density powder (HDP) target had: uranium density 12 g/cm^3, target thickness 6.3 g/cm^2, length 5.25 mm, diameter 11.2 mm; 3 pellets, each of 1.6–1.9 mm thickness, grain size about 20 μm. Targets were operated in the temperature range of 1900–2200°C. The investigated target materials were maintained in the target containers of the same dimensions and geometry. More detailed description of used target construction can be found in Refs. [4, 5]. The uranium fission reaction was produced by 1 GeV proton beam with the intensity of 50–70 nA. Mass-separated isotope beams were collected on a tape station and were either transported to γ-, β-, α-counting stations or directly counted by α-detector to be identified by the characteristic lines. Detailed description of γ-rays measurement and yield determination can be found in Ref. [4]. The yields of Cs isotopes were determined by means of β-counter as well as of γ-detector. Fr isotopes were produced in the same targets by the spallation reaction. The Fr isotope yields were measured using γ-rays and α-particles. To get consistent yield data obtained by α- and γ-measurements, the α-yield data were normalized to the yields obtained by γ-measurement (as the efficiency of the Ge detector is well known), taking as normalizing coefficient the yield ratio for isotopes measured by both methods.

2.1. Yield comparison

For the investigated target materials the Cs and Fr yields under similar temperature conditions (T = 2100°C) have been determined. All presented yield values were normalized to a target thickness of 1 g/cm^2 and 0.1 μA proton beam current. The error of the temperature measurement for different targets can reach 50°C. The experimental errors of the obtained yield values are within the limits of 30% for isotopes not far from stability and of 80% for short-lived neutron-rich isotopes. Cs yields from the investigated targets are plotted together with the ISOLDE yields obtained for standard target at the PS-Booster and for graphite cloth target at the synchrocyclotron. As one can see in Fig. 2, there is a good agreement of yields from similar target materials of ISOLDE and PARRNe-IRIS. The yields of short-lived Cs isotopes from a HDR target are 1.5–2 times lower. This fact demonstrates its lower release efficiency than the ISOLDE or PARRNe-IRIS target. It is an expected result, as the grain size of the HDR target is 10 times larger. Therefore the diffusion process should be much slower. The yields measured for Rb isotopes, which are not presented here, confirm the results obtained for Cs. The Cs yield trend from the lately developed HDP pellet target is similar to the fast PARRNe-IRIS target, but the efficiency is about 20 times less. It can be explained by a strong carbonization of the inner surface of the target container that leads to the ionization efficiency decrease, as the HDP target had been outgased much longer time than other targets. The calculated yields based on the experimental cross sections are also presented to show the in-target production rate of the investigated isotopes. The enhancement of the yield of ^{139}Cs from the HDR target respectively to the calculated one can be explained by the decay of the mother ^{139}Xe nucleus that has the production cross section comparable to the Cs daughter nucleus. It is worth stressing that the yield excess in respect to the calculated yield values was measured only for Cs isotopes produced from the HDR target. It can indicate that in the HDR target the Xe precursors may have longer release times, therefore allowing a large probability of decay in the target as an additional production mode. That assumption requires a special verification.

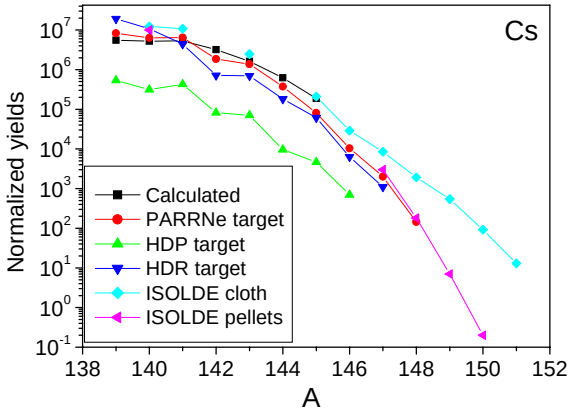

Fig. 2. The yields of Cs isotopes released from a high density rod, high density and low density pellet targets at 2100°C. The yields from ISOLDE UC_x pellet target are presented. The yields calculated on the basis of measured cross section are shown as well

It should be specially pointed out the extraordinary high yields of short-lived Rb and Cs obtained from the ISOLDE graphite cloth UC_x target at the SC. Unfortunately, these high yield data were not reproduced at the PS-Booster, where presently the ISOLDE standard powder-pellet UC_x target is used.

In Fig. 3 the yields of Fr isotopes from the HDR and HDP targets are presented. Fr isotopes have not been measured from PARRNE-IRIS target. The yields of short-lived ^{214}Fr ($T_{1/2}$ = 5 ms), ^{218}Fr ($T_{1/2}$ = 1 ms) and ^{219}Fr ($T_{1/2}$ = 21 ms) were measured by detection their characteristic α-decays. The yields of isotopes with longer half-

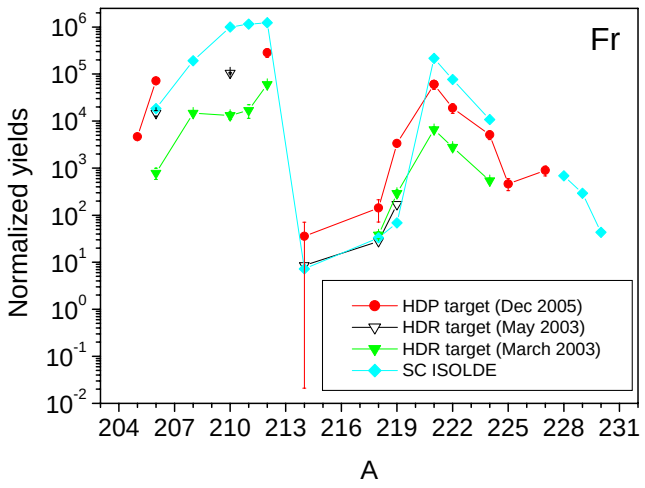

Fig. 3. The yields of Fr isotopes from the HDR, HDP and ISOLDE graphite-cloth targets

lives were determined using both α-particles and γ-rays. The Fr isotope yields obtained at the ISOLDE from the UC_x graphite-cloth target are also shown in the graph. The normalized yields of long-lived Fr isotopes from the ISOLDE graphite-cloth target are higher than from the HDR and HDP targets. The lower HDR and HDP target efficiency, which is the product of the ionization and release efficiencies, may be explained by lower value of the ionization efficiency of the ionizing targets used for experiments, where the ionization process takes place on the strongly carbonized inner surface of the target container. But, at the same time, the normalized yields of short-lived $^{214, 218, 219}$Fr are equal or even higher than the yields of these short-lived isotopes from the ISOLDE target. As there was only one measurement of the Fr yields from a newly developed HDP target, we give here the obtained yield values from that target as preliminary ones.

2.2. Comparison of the target release properties

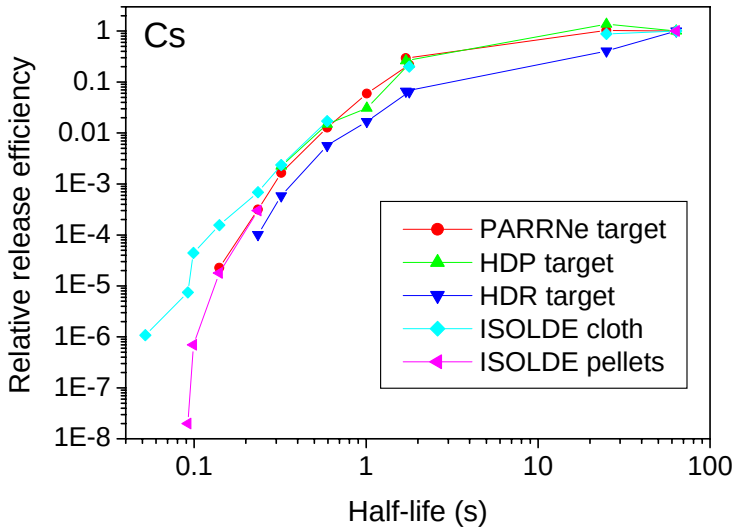

Fig. 4. Comparison of the Cs yield trends from HDR, HDP, LDP PARRNe IRIS and ISOLDE targets. The yields are normalized at the yield point of ^{140}Cs with the half-life $T_{1/2}$ = 63.7 s

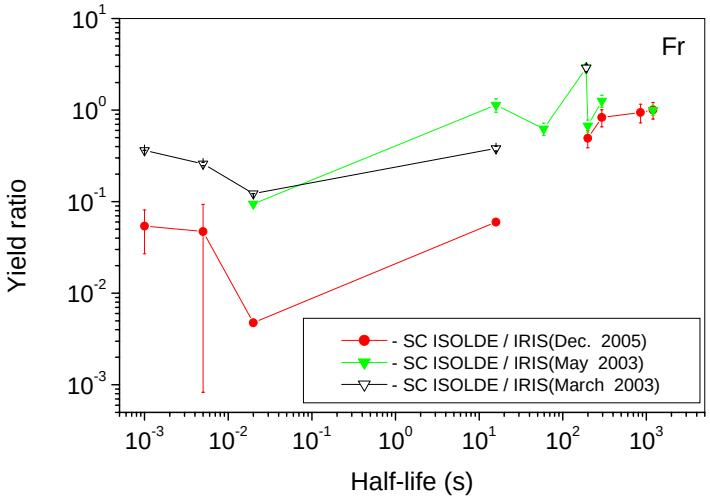

Fig. 5. Ratio of the yields of Fr isotopes from ISOLDE graphite-cloth target to the yields of the same isotopes from HDR (triangles) and HDP (circles) targets, plotted as a function of the half-lives. The obtained ratios are normalized at the point ^{212}Fr with the half-life $T_{1/2}$ = 20 min

The comparison of the relative release efficiencies of different targets for produced isotopes has been derived by normalizing the obtained yield values to the yield of a long-lived isotope ^{140}Cs ($T_{1/2}$ = 63.7 s) which is produced with the efficiency close to unity. In Fig. 4 the relative release efficiencies of Cs isotopes are presented as functions of their half-lives. The trend of the normalized yield values demonstrates that the ISOLDE and PARRNe-IRIS targets in general are slightly faster than the HDR target. A rather promising result is that the trend of the relative efficiency curve for the newly developed HDP target coincidences with one of the ISOLDE target.

For comparison of the Fr release efficiencies, the yields from the ISOLDE cloth target were divided by the measured yields of the same isotopes from the HDR and HDP targets and normalized to the yield ratio of ^{212}Fr. The obtained ratio values are shown in Fig. 5 as functions of the half-lives; as the Fr isotopes were not studied with the PARRNe target, only the ratios for the HDR and HDP targets are presented. The result for the HDP target is preliminary. As one can see, the HDR and HDP targets has considerably higher release efficiency for short lived-isotopes of ^{214}Fr ($T_{1/2}$ = 5 ms), ^{218}Fr ($T_{1/2}$ = 1 ms) and ^{219}Fr ($T_{1/2}$ = 21 ms). This can be explained by the fact that the sticking and flight time of species produced in the IRIS tested target prototypes is about six time less than in the ISOLDE target due to the ratio of the volumes of the target containers. To confirm or to reject the

assumption of a much shorter effusion time some additional on-line tests on the short-lived Fr isotope production from a HDP target will be carried out at the IRIS facility.

3. Ionizing targets for production of nuclides with high ionization potentials

For the production of nuclides with high ionization potentials three types of combined targets have been developed and on-line tested:

1. The ionizing foil target of the surface ionization for the Li and rare-earth isotope production.
2. The laser foil target for the production and resonance laser spectroscopy investigation of neutron-deficient rare-earth isotopes.
3. The combined target-ion source unit of the electron beam ionization with uranium carbide as a target material for the production of neutron-rich isotopes of the elements with the ionization potentials higher than 6 eV.

3.1. Off-line measurements of the ionization efficiencies for Li, Sm, Eu, Tm, and Yb

The schematic diagram shows the high-temperature ionizing target [6] used for on-line tests (Fig. 6). The only difference in the construction of the ionizing target (IT) used for off-line ionization efficiency measurements 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.

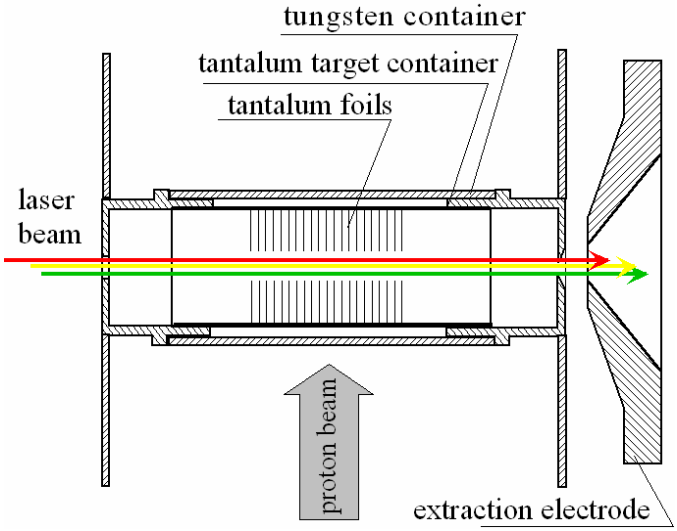

Fig. 6. Scheme of the combined target-ion source unit for Li, rare-earth isotope production and laser spectroscopy experiments

Off-line measurements of the surface ionization efficiencies for Li and rare-earth elements was carried out by using samples of LiF and oxides of pointed out rare-earth elements weighting from 10 to 20 μg 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. The process of the laser ionization efficiency measurement of Eu and Gd atoms inside the target volume was carried out simultaneously with the surface ionization measurement. For that purpose three laser beams tuned in resonance with appropriate transitions in Eu and Gd were introduced into the CTIS. We indicate this method as the laser target (LT) [7], in order to emphasize that it differs from the method employing the resonance ionization in the laser ion source (LIS) [8].

Table 1

The efficiencies of ionization measured off-line with IT and LT

Element	Temperature, °C	Surface ionization efficiency, %	Laser ionization 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	

In Table 1 the efficiencies of surface and laser ionization measured off-line for some alkalis and rare-earth elements are presented. The error of the ionization efficiency measurements was 20% for Rb and Cs and up to 40% for other elements.

3.2. On-line measurements of the yield of neutron-deficient rare-earth isotopes

A 1 GeV proton beam induced spallation reactions in the target material (tantalum foils, target thickness 3 g/cm^2) producing nuclides of neutron-deficient isotopes of rare-earth elements. After surface or laser ionization and extraction from the target, the ions were separated by a mass-separator and implanted into a moving tape installed at one of the three beam-lines of the IRIS facility. An identification of the radioactive implanted specimens and yield determination were performed by means of appropriate γ-line measurements by a coaxial high-purity germanium detector. The proton beam intensity was 0.05 μA. In Table 2 the yields of the neutron-deficient rare-earth isotopes produced on-line, the yields calculated by the EPAX code and the values of the efficiency of the ionizing target are presented.

Table 2

On-line yields and production efficiencies of the IT at 2500°C

Nuclide	$T_{1/2}$	Yields measured, s^{-1}	Yields EPAX, s^{-1}	Production efficiency by surface ionization, %	Production efficiency by laser ionization, %
^{138}Eu	12.1 s	2.4×10^3	9.5×10^4		
^{139}Eu	17.9 s	1.6×10^4	4.9×10^5		
^{141}Eu	41.4 s	4.0×10^5	5.6×10^6		
^{142}Eu	1.22 min	4.1×10^5	1.2×10^7		
^{143}Eu	2.57 min	2.0×10^6	2.1×10^7	9	
^{139}Sm	2.57 min	2.8×10^5	8.4×10^6		
^{143}Sm	66 s	4.4×10^5	2.3×10^7		
^{139}Pm	4.15 min	3.1×10^5	2.6×10^7		
^{160}Tm	9.4 min	5.0×10^5	3.5×10^6		
^{164}Tm	5.1 min	9.6×10^4	1.6×10^7		
^{160}Yb	4.8 min	1.0×10^6	1.6×10^7	6	
^{160}Lu	40 s	2.2×10^4	1.5×10^7		
^{168}Lu	6.7 min	1.3×10^6	4.1×10^6		
^{143}Gd ^{143m}Gd	39 s 1.87 min	6.7×10^4	3.5×10^6		
^{145}Gd ^{145m}Gd	23.0 min 85.2 s	6.7×10^5	1.7×10^7	4	2

For ^{143}Gd and ^{145}Gd the total values of the yields summarizing the measured isomeric and ground states are given. The efficiency of the laser target for ^{145}Gd production is pointed out as well. The combined target-ion source unit efficiency, which is a product of the release and ionization efficiency, was obtained as the ratio of the measured yields to the yields calculated on the base of cross sections supplying by the EPAX code. That efficiency estimation was obtained only for isotopes with respectively long half-lives and which production cross sections are close to the maximum of the cross section curve (^{143}Eu, ^{145}Gd and ^{160}Yb). As one can see in Table 2, the on-line production efficiencies for pointed out long-lived isotopes are in a good agreement with the off-line measured values (Table 1).

3.3. The CTIS use for on-line laser spectroscopy experiment

The developed combined target-ion source unit has been used for a laser resonance spectroscopy investigation of neutron-deficient Gd isotopes [7]. The scheme of the CTIS used for the laser spectroscopy experiments is shown in Fig. 6.

For the comparison of two methods (LIS and LT), the ratio R of the photo-ion current to the thermal ionization background has been measured for two types of the target system: the first one was a traditional construction (target connected to the ion source) [8] and the second one was the laser target. In Table 3 the

beam intensities of ^{145m}Gd and ^{145g}Gd from both tested target systems at the equal temperature conditions are presented. During the tests the targets having an identical material thickness were used, which were irradiated in both experiments by the same proton beam intensity. 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 for both targets, we have obtained the ratio of ^{145m}Gd ($T_{1/2}$ =1.87 min) to ^{145g}Gd ($T_{1/2}$ = 23 min) almost 7 times higher from the laser target than from the usual target-ion source unit (see Table 3 "laser off"). The enhancement of ^{145m}Gd having a considerably shorter half-life than a long-lived ^{145g}Gd demonstrates that the integrated target-ion source is faster than the usual target-ion source unit.

Table 3

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

	^{145m}Gd beam intensity, s^{-1}		^{145g}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	$1.64(8)\times10^{5}$	$3.21(11)\times10^{5}$				
Laser off	$1.55(6)\times10^{5}$	$2.30(8)\times10^{5}$	$2.02(20)\times10^{6}$	$4.40(16)\times10^{5}$	$7.7(1.1)\times10^{-2}$	$5.20(37)\times10^{-1}$
R	0.06(6)	0.40(6)				

3.4. The uranium carbide CTIS for the production of neutron-rich isotopes

A schematic drawing of the CTIS for the production of nuclides with high ionization potentials is shown in Fig. 7. The electron beam was introduced into the target volume by a cathode shaped for that purpose and placed to the side of the extraction electrode. So the electron beam ionization took place inside the target container [3]. The yields of neutron-rich Ag and Sn isotopes are shown in Fig. 8. The production efficiency values obtained as a ratio of measured and calculated yields are, correspondingly, 4% and 2% for long-lived Ag and Sn. With a high value of probability it is equal to the ionization efficiency of that species, as the release times of these elements are shorter than their lifetimes.

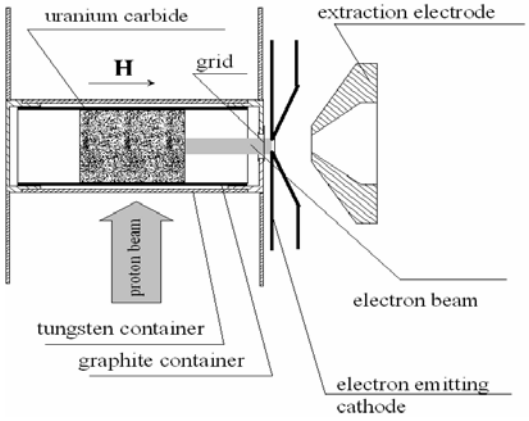

Fig. 7. Scheme of the electron beam ionizing UC target for production of neutron-rich isotopes

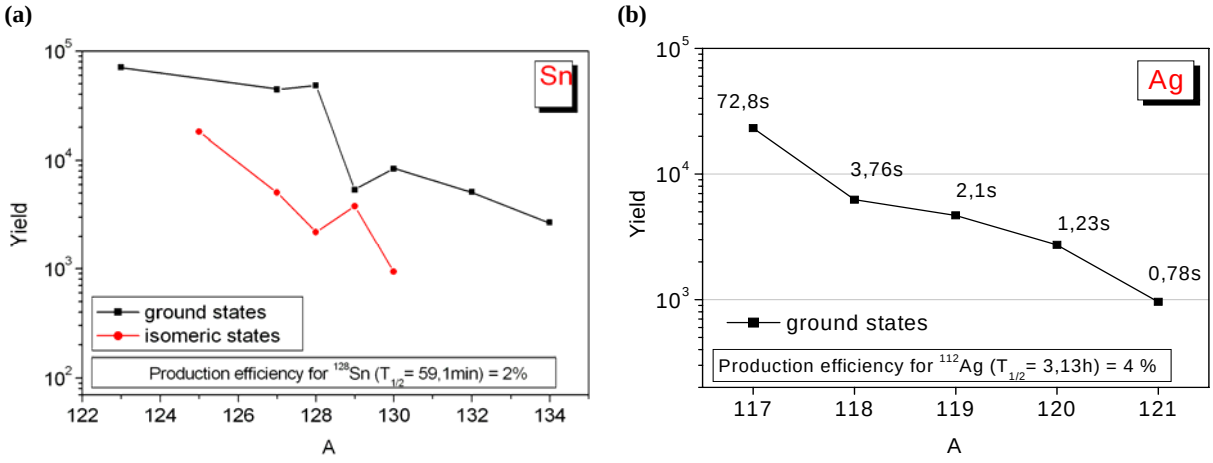

Fig. 8. Sn (a) and Ag (b) isotope yields from the electron beam IT

4. Conclusion

The yields of on-line mass-separated Cs, Rb and Fr isotopes have been studied at the IRIS. Properties of high density UC targets, in form of rod and powder, and a low density UC_x target prepared by the ISOLDE method similar to the PARRNe target are compared. According to the measured yield trends, the release efficiency of an ISOLDE like target for short-lived Cs isotopes is 1.5–2 times higher than that of a HDR (high density rod) uranium carbide target which was studied at the IRIS initially. It is rather encouraging result, as the grain size of the HDR UC target was ten times larger than the ISOLDE like target.

It should be pointed out that the on-line measured production efficiency and release properties of the ISOLDE like target specially prepared at the IRIS for that comparison tests coincide with the ISOLDE original target within the experimental errors (≤ 40%).

To compare target materials having different densities but a similar dispersivity, a new developed, high density pill (HDP) UC target with the grain size of 20 μm have been on-line investigated. According to the measured Cs yield trends, the release efficiency of the new HDP target coincides with the ISOLDE like target within the limits of the measurement errors.

The comparison of the yields of short-lived Fr isotopes from the HDR and HDP targets with the yields from the ISOLDE graphite-cloth target exhibits a rather unexpected result: the normalized yields of extremely short-lived Fr isotopes from high density targets are equal or even higher than those from the graphite-cloth target.

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

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

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