On-line yields obtained with the ISOLDE RILIS

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Abstract

The ISOLDE resonance ionization laser ion source (RILIS) allows to ionize efficiently and selectively many metallic elements. In recent yield surveys and on-line experiments with the ISOLDE RILIS we observed $^{23-34}$Mg, $^{26-34}$Al, $^{98-132}$Cd, ^{149}Tb, $^{155-177}$Yb, $^{179-200}$Tl, $^{183-215}$Pb and $^{188-218}$Bi. The obtained yields are presented together with measured release parameters which allow to extrapolate the release efficiency towards more exotic (short-lived) nuclides of the same elements.
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1. Introduction

The resonance ionization laser ion source (RILIS) [1] provides an efficient, fast and selective way to ionize many metallic elements. The elements ionized with the ISOLDE RILIS were reviewed recently [2–4]. In this article we will discuss on-line

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results, i.e. release curves and yields measured in recent target tests, nuclear spectroscopy experiments and on-line collections.

The measured release curves were fitted with the empiric four-parameter-formula described in [5,6]. Table 1 shows a summary of the release parameters: t_r the rise time, t_f the fall time of the fast component, t_s the fall time of the slow component and α the relative intensity of the fast component. Parameters which had to be fixed to obtain a stable fit are marked in parentheses.

The release parameters were used to determine for each radio-isotope the fraction which is released before it decays. The on-line efficiency of the RILIS cannot be easily monitored as in the case of a plasma ion source where the support gas is injected at a well-defined flowrate. Still a rough estimate can be made by scaling the off-line measured efficiency with the laser power available in the on-line run. Also the ion yield of a long-lived isotope with known production cross-section gives an indication of the ionization efficiency. Correcting the ion yields for the released fraction and the ionization efficiency allows to deduce the in-target production rate. In some of the following graphs both values are plotted: the *ion yield*, showing the presently available radioactive ion beam intensity for the user, and the *in-target production rate* showing the theoretical limit that could be obtained with an ideal target (no decay losses) of the same thickness and dimensions coupled to an ideal ion source (100% efficiency).

The ratio between both curves shows the overall efficiency of the present target and ion source unit.

2. Magnesium

Neutron-rich Mg beams have been produced formerly at ISOLDE SC [7] from a UC_x/graphite target with a chemically unselective "hot plasma" ion source (MK5) [8]. Although this ion source is optimized for the production of singly charged ions, it also produces with less efficiency multiply charged ions. Hence, at a mass separator setting $A/q = 31$ in addition to most $A = 31$ isobars also doubly charged ions of mass 62 (e.g. $^{62}Mn^{2+}$), triply charged ions of mass 93 (e.g. $^{93}Kr^{3+}$), quadruply charged ions of mass 124 (e.g. $^{124}Sb^{4+}$) and molecules (e.g. $^{12}C^{19}O^{+}$) may appear. In particular the contamination from abundantly produced fission products may be disturbing. Special ion source tuning can reduce this kind of background, but it cannot suppress it completely.

With the RILIS there is much less background of multiply charged ions. The mainly occurring beam contaminants are the surface ionized Na and Al isobars, the intensity of the former being about one order of magnitude lower compared to Mg. The measured Mg ion yields and the deduced in-target production rates are shown in Fig. 1.

The pure RILIS Mg beams were used for precision mass measurements with the RF spectrometer MISTRAL [9]. In a former beamtime with a

Table 1

Overview of target characteristics, operation conditions, release parameters and estimated ionization efficiencies during the different on-line runs

Element	Ion. pot. (eV)	Target			Ionizer		Ionization efficiency (%)	Release			
			g/cm²	°C		°C		α	t_r (ms)	t_f (ms)	t_s (ms)
Mg	7.65	UC_x/gr.	50	2020	W	2200	10	0.98	66	190	860
Al	5.99	UC_x/gr.	49	2070	W	2250	1–10	(0)	(100)	–	22,000
Cd	8.99	UC_x/gr.	50	≈2000	Nb	≈2000	10	0.98	≈60	≈300	≈3000
Tb	5.86	Ta foil	42	2200	W	1900	?	–	–	–	–
Yb	6.25	Ta foil	42	2150	W	2000	≤15	0.91	(300)	100	35,000
Tl	6.11	UC_x/gr.	50	2000	Nb	2100	5	0.98	45	600	5700
Pb	7.42	UC_x/gr.	50	2050	Nb	2100	3	0.96	70	580	4700
Bi	7.29	UC_x/gr.	50	2050	Nb	2100	1	0.86	45	540	8000

Note the uncertainty in the determination of the ionization efficiency: about a factor two. The UC_x/graphite targets contain about 10 g/cm² of carbon in addition to the ≈50 g/cm² ^{238}U.
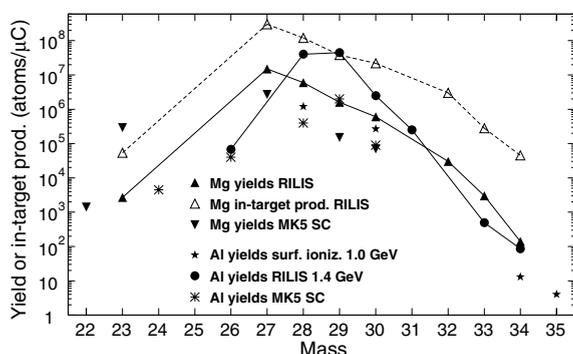
Fig. 1. Ion yields and in-target production rates of magnesium and aluminum isotopes. For ^{26}Al only the yield of the 6.3 s isomeric state was measured. The lines are drawn to guide the eye.

hot plasma ion source ^{32}Mg could already be measured [10], but the strong background had to be reduced with narrow slit settings which compromised the transmission and hence the statistics. With the RILIS beams the slits could be opened wider and the statistics was significantly improved. Thus also the mass of the $N = 21$ isotope ^{33}Mg could be measured for the first time [11].

3. Aluminum

Al is moderately well surface ionized. Neutron-rich Al beams up to ^{35}Al [12] were already studied at ISOLDE from a UC_x/graphite target with W surface ionizer. These yields obtained with a 1.0 GeV proton beam are shown in Fig. 1 compared to yields from a MK5 ion source at ISOLDE SC.

Off-line tests had shown that the RILIS can increase the Al ionization efficiency by up to a factor 70 compared to surface ionization in a 2200 °C hot W cavity [3]. This was obtained by simultaneous excitation of the two fine structure levels of the ground state with two frequency doubled dye laser beams (308.2 and 309.3 nm, respectively). For the on-line tests (dedicated for another purpose) only the 308.2 nm beam was used to excite the less populated fine structure level. Moreover the second step laser power focused to the HRS front-end was lower, giving a "laser on"/"laser off" ratio of only about three. There is no doubt that the shown

yields will be tripled when using both laser beams simultaneously. Even more could be gained by increasing the power focused to the target. Fig. 1 shows the yields measured with the 308.2 nm beam only.

Due to long desorption times, elemental Al is released very slowly. [1] Adding CF_4 will considerably speed up the release of Al, then as AlF molecule [13]. To allow for ionization with the RILIS (tuned to excite atomic Al) the AlF molecule needs to be dissociated in the ionizer cavity. A successful combination of fluorination and RILIS would increase the beam intensity of short-lived Al isotopes enormously and would allow one to produce beams well beyond ^{36}Al.

4. Cadmium

Neutron-deficient Cd nuclides were one of the first beams separated at ISOLDE [14]. Concerning beam intensity and purity, these beams from a molten tin target are still showpieces at ISOLDE. A drawback of the molten targets is their rather slow release time of the order of tens of seconds to minutes [15]. Thus, considerable decay losses occur for the isotopes below ^{102}Cd.

A LaC_x/graphite target (molar ratio La:C $\approx$ 1:4) combined with the RILIS was tested. [2] Due to a reduced target density (≈ 0.22 mol/cm^2 La versus 1 mol/cm^2 Sn) and smaller cross-sections the yields of $^{100+x}$Cd are lower than from molten Sn targets, see Fig. 2. For ^{98,99}Cd they are comparable, but the most short-lived isotope ^{97}Cd, where the faster release should give the biggest advantage, was not observed in this test [16]. With an efficient and selective identification method (e.g. detection of βp) the RILIS could be used to perform atomic spectroscopy directly in-source [4], i.e. to measure magnetic moments and nuclear charge radii of the

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[1] With the available tracer isotopes no indication for a fast release component was observed, hence $\alpha = 0$ was assumed for the fit.

[2] This run was performed with 1.0 GeV protons, most other yields shown in this paper were measured with 1.4 GeV protons.
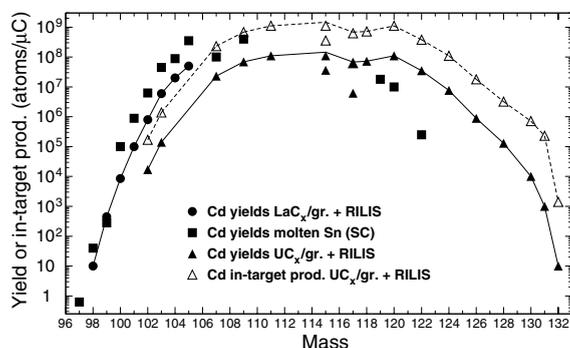
Fig. 2. Ion yields and in-target production rates of cadmium isotopes. For ^{115,117}Cd the $11/2^-$ isomers are more abundantly produced than the $1/2^+$ ground state.

very neutron-deficient isotopes $^{97+x}$Cd around the $N = 50$ shell closure.

Neutron-rich Cd isotopes up to ^{130}Cd had already been separated at the ISOLDE SC with a UC$_x$/graphite cloth target and a plasma ion source [17]. Some chemical selectivity was provided by a quartz transfer line at 600 °C, suppressing part of the indium and cesium isobars but leaving (on mass 130) still a molecular beam of ^{40}Ca^{90}Br$^+$ as background. To detect the heavier Cd isotopes with still shorter lifetimes the decay losses have to be minimized by keeping the full target and ion source unit at a sufficiently high temperature. Using the chemical selectivity of the RILIS, the short-lived neutron activities of the isotopes $^{130-132}$Cd could thus be identified [18].

Fig. 2 shows the yields of Cd isotopes obtained with the RILIS at ISOLDE PSB. These yields were measured with four different targets, fluctuating [3] by up to an order of magnitude, probably due to a varying on-line ionization efficiency. Using a "neutron converter" [19], the yields are lower than in direct bombardment, but the isobaric background of neutron-deficient Cs is suppressed considerably.

The RILIS provides now beams free of molecular contaminations but leaves most of the In and Cs isobars in the beam. The ultimate beam purity could be obtained by combining a chemically selective transfer line (suppressing Cs and In) with the hot cavity RILIS (suppressing most molecules).

5. Terbium

Neutron-deficient Tb isotopes are abundantly produced by spallation of a Ta target and moderately well surface-ionized. The 4.1 h isomer ($1/2^+$) of ^{149}Tb has a 17% branching ratio for alpha decay [20] and is therefore considered for radiotherapeutic applications [21]. A recent in vivo experiment at the University Hospital of Geneva using ^{149}Tb marked antibodies gave very promising results for the treatment of lymphoma [22]. To obtain sufficient quantities of ^{149}Tb for therapeutic purposes any possible increase in yield is important. Therefore we tested on-line the prospects of the RILIS for Tb ionization. We used an ionization scheme which had been developed in Troitsk [23]: three visible transitions (579.6, 551.7 and 618.3 nm) leading to an autoionizing state. Due to the presence of many surface ionized isobars and oxides in the lanthanide region it is difficult to obtain a clean Tb signal by simple counting of atoms or beta-decays. Two collections at mass 149, one with surface ionization (laser off) and one with additional use of the RILIS (laser on) were performed and analysed by γ-spectroscopy. The latter increased the ^{149}Tb yield from 1.2×10^7/μC to 2.1×10^7/μC. This 70% increase is much smaller than expected, [4] indicating that a dominant part of the off-line measured Tb is indeed produced indirectly as decay daughter of ^{149}Dy$^+$ and ^{149}Ho$^+$. Hence, using the RILIS to ionize rather Dy than Tb could give a much higher cumulative ^{149}Tb yield. A suitable RILIS scheme for dysprosium still needs to be developed.

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[3] Also the fitted release times of Cd fluctuate by up to a factor two when comparing the different targets at different "age". Table 1 shows an average.

[4] Also the absolute yields of Tb were very low since target and ion source were rather cold during this experiment.

6. Ytterbium

Yb was the first element tested off- and on-line at the ISOLDE RILIS [1]. Neutron-deficient Yb isotopes down to [153]Yb were ionized and used for in-source atomic spectroscopy with the RILIS of the IRIS facility in Gatchina [24]. Here we report the yields obtained at ISOLDE with a standard Ta foil target and W ionizer. [5] With the RILIS the Yb yields were enhanced by a factor of about 20 against surface ionization. The now measured on-line efficiency was probably below the off-line measured 15% [1]. Note that the W ionizer is "optimized" to create isobaric background from other lanthanides and lanthanide oxides. Hence only the most abundant isotopes (down to [155]Yb) could be clearly identified by measurements using a $4\pi\beta$ detector, Faraday cup, laser on/laser off and half-life checks, respectively, see Fig. 3. Before irradiation of the target no contamination with stable Yb was observed, i.e. all shown yields are radiogenic. [168]Yb shows a significantly higher yield than the neighboring isotopes. [6] This shows that 6 min [168]Lu which accumulates from the decay of the refractory (and thus not released) higher-Z isobars is not significantly released before decaying towards [168]Yb. Thus the release time of lutetium is under these target conditions well above 1000 s! From our measurement we cannot exclude that also Yb has an additional very slow (>100 s) release component.

7. Thallium

Tl is moderately well surface ionized and clean Tl beams were already available at ISOLDE SC. Off-line it was shown that the RILIS allows to increase the Tl ionization efficiency by about a factor 20 compared to surface ionization in a 2200 °C hot W cavity. In the on-line experiment the
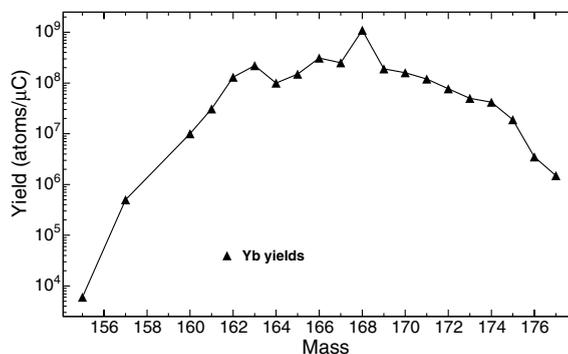

Fig. 3. Ion yields of ytterbium isotopes from a Ta foil target.

laser power was lower, but the surface ionization efficiency in the 2100 °C hot Nb cavity was also reduced, providing again a selectivity of about 20.

Fig. 4 shows the measured yields of neutron-deficient Tl isotopes from a W surface ionizer and the RILIS, respectively. In cases of several detected isomers the sum yield is shown. The yields of [179–181]Tl were estimated from the observed alpha emission rate and assumed branching ratios for α decay of 80% ([179g]Tl), 10% ([180]Tl) and 20% ([181g]Tl). For comparison are shown the yields of surface ionized Tl isotopes which were obtained at the IRIS facility by 1.0 GeV proton induced spallation of a special 10 g/cm^2 ThC$_x$ target (produced by pyrolysis of a thorium diphtalocyanine precursor) coupled to a W/Re surface ionizer [25,26].
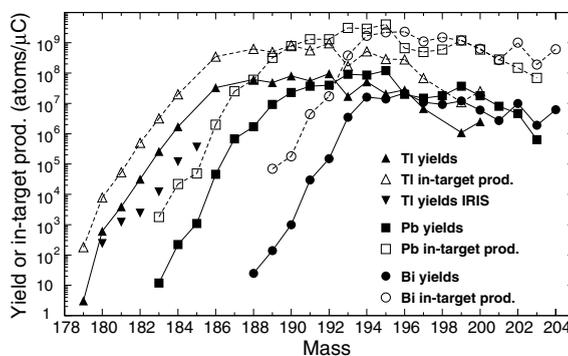

Fig. 4. Ion yields and in-target production rates of neutron-deficient thallium, lead and bismuth isotopes. The yields of [215]Pb, [215m]Bi and [218]Bi (not plotted) were 0.3, 1400 and 44 per μC, respectively.

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[5] This particular target had generally low yields. Thus the given yields can be regarded as a lower limit for standard conditions.

[6] The dip at [164,165]Yb may just be due to a fluctuation of the laser power during the measurement.

8. Lead and bismuth

Pb and Bi beams, ionized with an unselective hot plasma ion source, were available before at ISOLDE [7]. However, the admixture of many isobars (Hg, Tl, Po, At, etc.) made them difficult to use for nuclear spectroscopy and other applications. The use of the RILIS allows now to provide rather pure beams of Pb and Bi, the remaining isobaric contaminations being the surface ionized Tl and Fr. The former is mostly on an acceptable level, but the latter is intense enough to be disturbing for most applications at $A > 206$. Only for masses 215–218 the Fr isotopes are so short-lived that this background can be suppressed completely by a delayed opening of the electrostatic beamgate [27].

In cases of several detected [7] isomers the sum yield is shown, see Fig. 4. In most measured cases the yield of the high-spin isomer was higher. The yields of $^{188-192}$Bi were determined from the alpha decay rates, assuming $\approx 100\%$ branching ratios for α decay of ^{188g,188m,189g,189m}Bi. Note that the laser power in the Bi run was lower than in the former off-line experiment.

The RILIS can also be used to produce isomerically pure beams by tuning the wavelength of the resonantly exciting laser to the hyperfine structure of one or another isomer [28]. For the heavier elements Tl, Pb and Bi the Doppler broadening is rather small compared to the hyperfine splitting and this technique works particularly well. It was recently used to perform nuclear and atomic spectroscopy on two isomers in ^{185}Pb [29].

9. Summary

This article summarizes release functions of eight different elements and the yields of over hundred isotopes which were measured with the ISOLDE RILIS in the last years. Detailed data of the individual experiments are shown in Table 1.

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[7] Not always could all isomers be detected. This causes the fluctuations between neighboring isotopes, as apparent in the plotted yield curve.

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