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Studies of release properties of ISOLDE targets*

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Abstract

Off-line release rates of Be, Mg, S, Mn and Kr from refractory materials were studied. Mn yields were determined from a ZrO_2 target and Kr yields from a SrO and ZrO_2 targets. A Monte Carlo code to optimize ISOLDE targets was introduced.

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1. Introduction

The Isotope Separator On-Line (ISOL) technique [1] has been in use at CERN already more than 30 years and allows to make radioactive ion-beams of 70 % of the chemical elements. To maintain the attractiveness of CERN/ISOLDE and as preparation for the EURISOL project continuous development of the targets is required. This development concentrates presently on creating a more solid understanding of the performance of the target in order to reduce the decay losses and make beams of the remaining elements available. These studies have a growing importance since all chemically “easy” beams are already in use and the experiments are pursued far from the valley of beta-stability, where often one has to work with extremely short half-lives and low production cross-sections.

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In this article we concentrate onto the three different topics related to the target and ion source development. First new results from off-line target material studies are presented. The release of Be, Mg, S, Mn and Kr is discussed. Secondly, previously unpublished Mn and Kr yields are presented. As a third topic, a short introduction is given to a new Monte Carlo code that allows to optimize the intrinsic delay time as function of target and ion-source geometry. The release of Kr from a Nb foil target is used as an example.

2. Off-line studies

At ISOLDE activation and radioactive tracer implantation are used to produce the samples of interest for the off-line material studies. Activation is done using the well known pneumatic transport system "rabbit" [2, 3]. Both 1 and 1.4 GeV protons are employed. The fractional activity after heating of the samples in vacuum ($p<3\times10^{-5}$ mbar) is determined by radioassay with a gamma-ray spectrometer [2, 3]. Temperature equilibrium between the sample and the oven is reached in less than one minute [2]. Each sample is only heated once. Details of the irradiated materials are given in Table 1.

A rapid release of implanted ^{7}Be from a Ta foil was observed (60, 160 and 260 keV implantation energies were used corresponding to about 800, 2200 and 3400 Å projected ranges of Be in Ta [5]). The measured fractional activities are consistent with the used implantation energies (see Fig. 1a).

The fastest release of Mg was observed from a Ti foil, see Fig. 1b. For Mg also fast desorption from Ti surfaces is expected (the Eichler-Miedema model [6, 7] predicts a sticking time of the order of 1 ns at 1600 ^{o}C).

S was observed to have a fast release from TiO_2 powder and felt and also from the VC powder (see Fig. 1c) (fast release from VC was reported earlier in [2]). To avoid long effusion delay times S should form immediately after the release a volatile molecule for instance with CO.

Mn is released most quickly from a Zr foil (see Fig. 1d). For Mn a slower desorption from Zr surfaces is expected (the Eichler-Miedema model [6, 7] predicts a sticking time of the order of 50 μs at 1800 ^{o}C).

Kr is releasing from all the examined materials i.e. from $SrZrO_3$, Y_2O_3, Zr, ZrO_2, Nb and Mo (see Fig. 1e). As an example the release of Kr is faster from Y_2O_3 felt than from ZrO_2 felt and also the release of Kr is at least as fast from a Zr foil as from a Nb foil. A more extensive and complete study of the performed off-line measurements will be presented elsewhere [8].

3. On-line studies

The yields of Mn nuclei from a ZrO_2 felt target were determined using the ISOLDE spectroscopy station [9] (this gamma-detector set-up was mainly used to determine the yields of $^{48, 49}$Mn) and the monitoring tape-station [10] (beta counting) set-ups. The pulsed proton beam energy was 1.4 GeV. The Resonance Ionization Laser Ion Source (RILIS) [11] was used for Mn ionization. Compared to the former measurement with a Nb foil target at the General Purpose Separator (GPS) [12], the laser power focused to the High Resolution Separator (HRS) front-end was about a factor two lower which halfed the ionization efficiency. Further details are given in Table 2.

The yields of Mn from a Nb foil [12] and ZrO_2 felt targets are presented in Fig. 2a. The yields are generally lower from the ZrO_2 target due to the lower target thickness. Only for the most short-lived isotopes the faster release from ZrO_2 overcompensates this reduction.

The yields of Kr nuclei from SrO and ZrO_2 targets were determined using the monitoring tape-station set-up. Both targets were connected to an ISOLDE type FEBIAD ion source using a water-cooled transfer line (MK7) [14]. The ionization efficiency of Kr was about 3 % for both ion-sources. The proton beam energy was 1.0 GeV. Further details are given in Table 2.

The yields of Kr from a SrO and ZrO_2 felt targets are presented in Fig. 2b. Observed low ^{73}Kr yield from a SrO target is probably due to the fluctuation of the measurement. Fig. 2b together with Table 2 demonstrates that the Nb foil target (foil thickness 25 μm) [15] is still a competitive choice for the production of short-lived and n-deficient Kr isotopes like ^{69}Kr ($T_{1/2}$=32(10) ms [16]).

4. Monte Carlo code

The new Monte Carlo code admits flexible target and ion source geometries, with no limitation to the size or complexity of the system (other than CPU time consumption). Like some transport codes [17], the geometry is defined in terms of cells, enclosed by second order surfaces. Their equation coefficients and respective temperatures are defined in the input file, allowing variable temperature profiles and permitting all kind of changes without any new compilation. Moreover, it becomes easy to compose large input files from several modules (target, container, transfer line and ion source) by simply stacking one on top of the following in the input file, and by linking the interfaces. Concerning the particle generation, the user can specify in the input file the primary energy and type of effusing particles and their geometrical distribution on creation, including gaussian-like profiles.

On execution the user is offered to use several variance reduction methods. The output file includes various average figures like the number of collisions and, if wanted, individual delay times. Additionally, a complementary file is printed which expresses a compressed form of the connectivity matrix of the input geometry. It is a useful file in geometry verification. Moreover, the program uses the matrix to speed up computations.

The code has been tested in several situations where the effusion delay curve can be calculated analytically with the kinetic theory. The code yielded accurate results within the statistic error of reasonably long simulations.

At ISOLDE 1 GeV protons are delivered in 2.4 μs long pulses. Usually in the release studies the time difference between neighboring pulses is 14.4 s. The squares in the inset of Fig. 3

represent the measured on-line release function of ^{73}Kr ($T_{1/2}$=29.0(10) s [18]) from the Nb roll foil target (Nb foil thickness 25 μm) at a temperature of 2000 ^{o}C. The shape of the curve (N in the Eq. 1) can be reproduced in the calculations by combining effusion (f), diffusion (h) and radioactive decay (d) data (see circles in the inset of Fig. 3 (N×constant)):

$$N(k\Delta t) = d(k\Delta t)\sum_{i=0}^{k} f((k-i)\Delta t)h(i\Delta t)\Delta t + d(t_{SCY} + k\Delta t)\sum_{i=0}^{SCY} f(t_{SCY} + (k-i)\Delta t)h(i\Delta t)\Delta t \quad (1)$$

In Eq. 1 $f(t)$ is the effusion delay time distribution for particles released from the target into a target container at t = 0 s. $h(t)$ determines the relative amount of effusing particles released into the target volume as a function of time. $h(t)$ term originates from the diffusion process. The $d(t)$ term corrects for decay losses. The contribution of the previous proton pulse is also included (one proton pulse per super cycle (t_{SCY} = 14.4 s or 16.8 s, Δt = 1 ms) is assumed [10]). Since the N, d and h terms are known experimentally, Eq. 1 can be used to determine the shape of the Kr effusion delay time distribution $f(t)$. Solid line in Fig. 3 ($f(t)$) together with the circles in the inset of Fig. 3 (N×constant) represent the outcome of this iteration process. The diffusion coefficient 9×10^{-9} cm^2/s was taken from [19].

The effusion of Kr was also computed using the Monte Carlo code described earlier. Zero sticking time on surfaces was assumed. The starting distribution of effusing particles was derived from the radial structure of the impinging proton pulse. In the simulations 12 Nb target foil rolls (length of 1.5 cm) were placed in a Ta target container (length 19.5 cm) with 1 and 0.1 mm roll spacing. With 1 mm even roll spacing we have 2 mm gap at both ends of the target container while with 0.1 mm spacing we have 7 mm end gaps. For both geometries about 10000 particles were computed. Fig. 3 represents the outcome of these simulations; up triangles correspond to the 1 mm roll spacing and down triangles to the 0.1 mm spacing (note that all curves in Fig. 3 are normalized to the same maximum amplitude instead of equal area). These results indicate that the roll spacing is one of the important tuning

parameters of the ISOLDE foil targets. The shape of the experimental curve would be possible to reproduce simply by tuning the spacing between the target rolls. A more extensive study will be presented elsewhere [20]. The average number of wall collisions before the exit with 1 mm roll spacing is about 765 000 (corresponding to about 150 m traveled distance) and with 0.1 mm spacing about 3 450 000 (corresponding to about 615 m traveled distance). The suggested faster effusion of Kr out of the target and ion source unit with the 1 mm Nb foil roll spacing would for example double the earlier measured ^{69}Kr yield ~3×10^{-4} at/μC [21].

5. Conclusions

Even though the ISOL technique has already a long history, off-line material studies are still relevant. They provide a fairly inexpensive way to probe the properties of materials and, as was shown, they are still able to produce valuable information for the target and ion source data-base. Real on-line yields and release curves like the ones presented now are always very important since the whole operation of ISOLDE is based upon them. The introduced Monte Carlo code showed immediately its capability. In the next phase it will be extended to include diffusion etc. In addition, this code will be cross-checked with the dedicated off-line effusion delay time measurements.

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Table 1. Studied materials.

Material	Melting point [^{o}C]	Characteristic size [μm]	Crucible material	Supplier
Ti foil	1657	30	Ta	Johnson Matthey
TiO_2 powder pellet (2.4 g/cm^3) [a]	1850	<50	Re	Fluka [b]
TiO_2 felt	1850	~6	Re	Home made [4]
V foil	1890	30	Ta	Goodfellow
VC powder pellet (3.8 g/cm^3) [a]	2810	<44	C	Alfa [c]
$SrZrO_3$ powder pellet (3.4 g/cm^3) [a]	2750	<10	Ta	Aldrich [d]
Y_2O_3 powder pellet (3.2 g/cm^3) [a]	2410	<25	Ta	Fluka [e]
Y_2O_3 felt	2410	~9	Ta	Zircar Zirconia
Zr foil	1852	127	Ta	Ventron
ZrO_2 powder pellet (2.9 g/cm^3) [a]	2700	50-100	Ta	Serva
ZrO_2 felt	2700	~9	Ta	Zircar Zirconia
Nb foil	2470	127	Ta	Johnson Matthey
Mo foil	2624	50	Ta	Goodfellow
Ta foil	2996	50	Ta	Goodfellow

[a] Made by pressing the powder.
[b] Product No. 89490.
[c] Stock No. 12141.
[d] Catalogue No. 39,616-8.
[e] Product No. 95832.

Table 2. Overview of target characteristics, operation conditions, release parameters (release is proportional to $(1-\exp[-\ln 2 t/t_r])(\alpha \exp[-\ln 2 t/t_f]+(1-\alpha)\exp[-\ln 2 t/t_s])$ [10]) and ionization efficiencies during the different on-line runs.

Element	Target			Line		On-line eff. %	Release			
		g/cm^2	^{o}C		^{o}C		α	t_r ms	t_f ms	t_s s
Mn	ZrO_2	5.8	1800	W	2150	10	0.72	20	256	11.2
Mn	Nb	50	2050	W	2450	19	0.9996	42	31000	95
Kr	SrO	18	1460	MK7	2080	3	0.78	67	397	3.89
Kr	ZrO_2	8	1850	MK7	2020	3	0.71	90	938	12.7
Kr	Nb	42	1900	MK7	1900	9	0.82	75	829	23.1

Figure captions

Fig. 1. Release of species from materials as a function of temperature (a, c) and heating time (b, d, e).

Fig. 2. (a) The yields (▲, ■) and in-target production yields (Δ, □) (see definition [13]) of Mn nuclei from a Nb foil and ZrO_2 felt targets. (b) The yields (●, ■, ▲) and in-target production yields (○, □, Δ) of Kr nuclei from a SrO, ZrO_2 felt and Nb foil targets.

Fig. 3. Experimental (solid line) and simulated (up and down triangles) effusion delay time distributions for Kr from a Nb foil target. In the inset measured (squares) and calculated (circles) on-line release curves for ^{73}Kr from a Nb foil target.