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Review of ion-source developments for radioactive ion-beam facilities

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Abstract

The ion-sources dedicated to the production of radioactive ion beams (RIB) shall be highly efficient, selective and fast. This efficiency is mandatory since only limited amounts of radionuclides are produced. Chemical selectivity is needed to confine other elements near to the production site and to suppress isobaric contaminants. Eventually, the ion-source shall only delay the radioisotopes by a fraction of their half-life to reduce decay losses. The world wide spread RIB facilities came up with a large variety of solutions to meet part or all of these requirements such as: ion traps, surface, plasma, sputtering, electron cyclotron resonance and laser ion-sources.
In this review, the latest developments are presented and their applications to charge states breeder systems proposed for post-acceleration are discussed.

1 INTRODUCTION

Around the world, 23 radioactive ion beam facilities are currently in operation construction or planned [1-4]. Among them, the new generation facilities aims at more than one order of magnitude increased RIB intensities. Most of these schemes rely on an equivalent increase of the driver beam and on the development of targets able to cope with the deposited energy [4-8]. In Isotope Separation On Line (ISOL) facilities the isotopes are produced and stopped in the target and then transported as atoms by diffusion and effusion processes into the ion source (section 1.2-1.3). In Ion Guide Isotope On Line (IGISOL) facilities, the highly charged fragments recoiling from a thin target are stopped in a noble gas catcher where they rapidly decrease to the 1^+ charge state and keep this for a few milliseconds. In the mean time, they are transferred by the gas flow to the extraction and thus do not require any ion source. To overcome the missing chemical selectivity inherent to the IGISOL principle, the solution chosen by the Leuven group is to let the ions recombine in the gas catcher and then selectively reionize the wanted element (section 2.1).
The energy of the radioactive ion beam is dictated by the physics case requirements. A few eV for mass measurement, keV for solid state physics and atomic physics and many MeV/nucleons to reach and pass the coulomb barrier. The later requires a post acceleration of the low energy extracted ions. Cost effective post acceleration schemes requires a charge to mass ratio (q/M) in the region of ¼, the proposed charge state breeder are described in section 3.

1.1 RIB production mechanisms

The radioisotopes are produced by nuclear reactions between a projectile and a target [9]. The energy involved begins near the coulomb barrier with the fusion-evaporation reactions (complete fusion is at the origin of the super heavy elements). Neutron induced fission of heavy elements produces two neutron rich nuclei. At thermal energies, the isotopic distribution is the classical double peak of nuclear reactor (centered around A=95 and 140), which become broader with increasing neutron energy. The fragmentation of relativistic nuclei on a light target, or of heavy nuclei by relativistic light projectiles (called spallation in the case of relativistic protons) produces radioisotopes over the complete periodic table up to uranium. Obviously, the choices of the projectile, its energy and of the target material are crucial, witnessed by the efforts made in the computation and measurement of cross sections [10,11].

1.2 General criteria for target and ion-sources

The target region will become radioactive, its radio toxicity depends mainly on the proton number of both projectile and target and its activity on the projectile beam intensity and duration. Already on existing facilities, the exchange of target and ion source has to be remote controlled and repair works have to be made in hot cells and with manipulators. In addition, these works are preferably done after a few weeks to allow the short-lived elements to decay. Therefore, targets and ion sources shall be very reliable or designed as consumable items.
The ion source shall be highly efficient since the production rate is limited by the facility and cannot be increased. This is a major difference compared to ion sources producing beams of stable elements that are tuned to maximise the ion current.
The ion source shall be chemically selective to increase the beam purity and to confine the unwanted radioactive species near to the target area.
The emittance and the energy spread have to be small to match the requirements of the first separation stage (often a “simple” dipole magnet) and the acceptance of the post acceleration.

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1.3 Selected ion sources used for the production of radioisotopes

A figure of merit of the 1^+-ion sources for RIB is presented in figure 1 as a function of the ionization potentials. Major deviation from the presented trends is the result of the chemical or metallurgical bounds, which may exist between the ion source material and the element to be ionized. Or simply due to the low mobility of the refractory elements even at very high temperature. Developments of chemical processes currently attempting to circumvent these limitations are described in this section.

High temperature surface ion sources: Saha and Langmuir described the ionizing properties of a hot surface. Positive ions are produced when the minimal energy needed to remove an electron from a surface (its work function) is larger than the ionization potential and negative ions are produced when the work function is smaller than the electron affinity of the atom impacting on the surface at thermal energies. The amplification effect (multiple hits on the surface) and the trapping after themalisation in a hot cavity (density $\approx 5\times10^{14}/cm^3$) filled with Xe were demonstrated by Kirchner [12,13]. Both techniques increase the efficiencies when compared to the prediction of the Saha-Langmuir model. Surface ionization remains the most efficient ionization scheme for low ionization potential radioisotopes (alkalis and some lanthanides) that are currently produced with W, WO_3 and Re surfaces. Negative chlorine, bromine, iodine and astatine have been efficiently produced on LaB_6 Surfaces [14]. The work function (and thus the ion source efficiency) depends on crystal orientation, temperature and cleanliness [15,16].

Electron Cyclotron Resonance (ECR) ion sources are known to produce very high currents of highly charged stable elements (e.g. 100eμA of Pb^{27+} for LHC [17]) and to very efficiently ionize gaseous elements (H_2, N_2, O_2, Cl_2 and noble gases). The extension of ECR to the condensable or reactive radioisotopes is demonstrated in the following examples: The production-ionisation of condensable B and Be was achieved via a volatile fluorine molecular side band (BF^+ and BeF_2^+) [18]. The 2.2% efficiency obtained for $^7BeF_2 \rightarrow {}^7BeF_2^+$ is one order of magnitude below the results of the same ion source for gaseous elements (12% to 48% for 1^+ states) [19]. Using a volatile fluoromethane to transport ^{18}F, produced via the $^{18}O(p,n)^{18}F$ reaction, to the ECR, an efficiency of 0.3% was obtained for $^{18}F^+$ and 0.12% for $^{18}F^{2+}$ [20]. These successful applications of ECR ion sources to produce highly condensable or reactive materials also reveal the efficiency drop, tribute to the chemical nature of these elements.

Forced Electron Beam Induced Arc Discharge (FEBIAD) [21,22] are known for their efficient ionisation of gaseous and condensable elements (up to refractory and highly reactive like F and O) the ionisation occurs in a high temperature cavity by collision with 100-200eV electrons. The electrons are produced by a hot cathode and accelerated via a grid into a typically $1cm^3$-plasma chamber under low magnetic field. The production of 2×10^7 $^{17}F^+$/s via the $^{16}O(d,n)^{17}F$ reaction was achieved via a AlF molecular side band obtained by Al vapors diffusing through the HfO target and ionized with a FEBIAD type ion source. The efficiencies were 5%, 1.3% and 4% for the ^{19}F, ^{18}F (109.7m) and ^{17}F (64.8s) isotopes respectively. These values include the decay losses in the target [23].

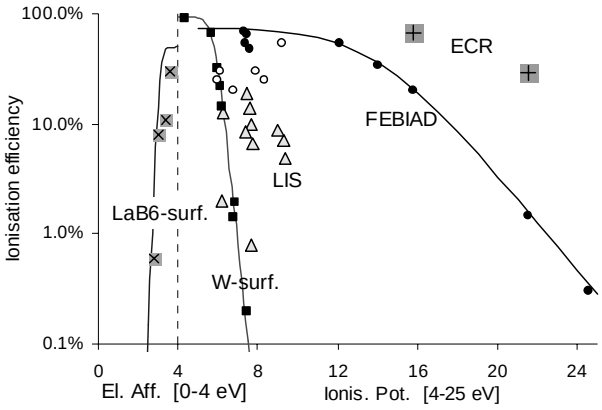

Figure 1 : 1^+ Ionization efficiencies measured with surface (black squares), plasma-FEBIAD (circles), laser (triangles) and ECR ion sources. The lines drawn to guide the eye represent the cases for which no chemical reaction occurs between the ion source materials and the element to be ionized. i.e. full and empty circles show the difference between the high and low temperature FEBIADs for condensable and volatile elements.

2 LASER ION SOURCES

Laser ion sources (LIS) are the last born of the RIB ion sources. The excitation energies are a unique fingerprint of each element. This is the origin of the high selectivity of resonant laser ionization methods [24]. Two LIS applications are described in the following sections.

2.1 Ion Guide Laser Ion Source (IGLIS)

The IGLIS developed at the LISOL separator [25] relies on the resonant laser ionization in a noble gas cell of the thermalized and neutralized recoil products [26]. The thin target sits in a gas cell filled with 500mbar helium or argon. Fission recoils have a wide energy distribution (up to 100MeV) which reduces the nearly 100% stopping efficiency down to 50%. The constant flow of the carrier

gas transports the radionuclides within typically 40ms from their stopping point to the region of ionization that is located in front of the opening hole. The purity of the gas is mandatory to suppress molecular sidebands. As a matter of fact, this method would allow detection of impurities in nobles gases at the ppt level. The release time-constant after laser ionization measured with stable ^{58}Ni is 10ms with a He carrier. Off line tests with argon showed that a fraction of the Ni^+ ion recombine before reaching the outlet, the effect is correlated with the observation of $(NiAr)^+$, $(NiAr_2)^+$ and $(NiArH_2O)^+$ charged molecules [27]. A major improvement in IGLIS systems was the replacement of the skimmer electrode used to separate the ions from the carrier gas by a sextupole ion guide (SPIG) [28]. The efficiency increased by a factor of 2 and the resolving power of the LISOL separator stepped from 300 up to 1450. Furthermore, a SPIG surrounded by a set of annular electrodes that are set at different potentials delivered ion bunches after 10ms confinement with an off line efficiency of 7% [29].

The IGLIS coupled to a SPIG or SPIG-trap is the system showing the least dependence on the chemistry of the recoiled element. The short time needed for transport and ionization essentially depends on the gas carrier. It had its first milestone with the successful production and ionization of the $^{54\text{-}55}$Ni (^{54}Ni: $T_{1/2}$ = 106ms [30]) and $^{68\text{-}74}$Ni isotopes and opens the door for the production of elements for which the laser ionization scheme is known (Ti, Ni, Co and Rh isotopes are currently produced). The IGISOL-SPIG combination is an elegant production scheme for all refractory elements (ionization potentials between 6.8 and 9.1eV).

2.2 Laser ion-sources in hot cavities

Stepwise resonant laser ionization in hot cavities is used for RIB production in ISOLDE [31] and ISIS [32]. The 2 to 3 laser beams are collinear to the axis of the metallic tube where the atoms effuse out of the target container. Dye lasers pumped by copper vapor lasers produce them. The UV light required for some transitions is obtained via frequency doubling or tripling of the dye laser light in non linear BBO crystals [33]. The lasers are pulsed at a frequency of 10 kHz, which corresponds to the typical 100μs needed for an atom of mass 10 to be pumped through the cavity. The electrons emitted from the cavity surface generate a negative potential well, which confines the positive ions. The ions then drift along the tube towards the extraction hole thanks to the electrical field generated by the ohmic heating of the cavity. The width of laser ionized bunches is typically 40μs FWHM for m=100amu. LIS-ions trapped in the high temperature cavity are confined in a 3×30mm size W- or Nb-tube at temperatures between 2100 and 2500K and released even 100 μs after ionisation [34]. Very short $^{139}La^+$ ion bunches of 0.4μs duration were produced [35] which seems to correspond to LIS ionization restricted to the ions localized in the extraction field region. The very high selectivity of the resonant laser ionization towards other elements is somewhat perturbed by the surface ionized elements. The choice of a low work function cavity like Nb and the micro gating of the LIS ion bunch effectively improves the selectivity.

Table 1. Off-line LIS ionisation efficiencies in high temperature cavities obtained by integrating the ion current of a typically 10μg sample. The sample was vaporised in a Ta-oven sealed to the rear of the LIS cavity. Radioisotope beams of each of these elements (but Mg) were produced.

Be	7.0%	[24]	Ag	14.0%	[39]
Mg	9.8%	[36]	Cd	8.8%	[34]
Mn	19.2%	[37]	Sn	8.5%	[40]
Ni	0.8%	[38]	Tm	2.0%	[41]
Cu	6.6%	[34]	Yb	12.5%	[41]
Zn	4.9%	[34]			

Hyperfine splitting (HFS) of the energy levels may exceed the line width of the laser (typically 5GHz) and reduce the efficiency. On the other hand, by reducing its line width, the LIS can be used to select between the spin states of a given isotope. This was experimentally shown on the ^{107m,g}Ag [42] and used for physics of the ^{70m,g}Cu [43], ^{155}Yb and ^{154}Tm [44,45] isotopes.

The fine-tuning of the laser frequencies shall also account for the isotope shift. Therefore, measurement of the isotope shift (thus on the mean square charge radius of heavy nuclides) is done by scanning the laser frequencies while recording the radioisotope production.

The isotope shift of all Be isotopes [42] and of Yb and Tm isotopes [44] were measured with this technique. The efficiencies of hot cavity laser ion sources are given in table 1.

3 CHARGE STATE BREEDING $1^+/N^+$

Post acceleration schemes require charge states with q/A of the order of ¼. In GANIL, in the framework of the SPIRAL [46] project, an ECR ion sources is foreseen for the ionization of high charge state radionuclides of noble gases, oxygen and nitrogen. However, for condensable elements, the charge state breeding of low energy 1^+-ion beams is a natural thought which is foreseen by TRIUMF, GANIL and ISOLDE. The $1^+/n^+$ technique would dramatically reduce the activity deposited in the ECR or EBIS (Electron Beam Ion Source) breeders and thus ease their maintenance.

3.1 ECR based charge state breeding

The first experimental proof of an ECR based $1^+/n^+$ scheme [47] showed the high sensitivity of the breeding efficiency on the energy of a condensable Rb^+ ion beam.

A Rb^{+} beam must be decelerated from 18kV to 15±3eV in order to be captured in the ECR plasma, while for noble gases, the ionisation is at a flat maximum between 30 and 90eV. The losses resulting from the mismatch of the 1^{+} beam emittance to the breeder acceptance were estimated as the ratio of the n^{+} ionization efficiencies of the $1^{+}/n^{+}$ to the *neutral*/n^{+} schemes and yields 50% for Argon [8]. The injection efficiency of the 1^{+} beam into the ECR plasma at very low velocities was investigated with the $Cr^{1+} \rightarrow Cr^{11+}$ breeding. The emittance of the 18kV 1^{+} beam was set from 40 to 6 πmm.mrad thus also reducing the total 1+ion current from 210 to 13 nA. The efficiency increased from 0.7% to 3.5% [48] as shown in figure 2. This result sets stringent requirement on the 1^{+} beam emittance which shall be of the order of 5 πmm.mrad at 18kV.

The ECR Ion Trap (ECRIT) [49,50] operating in the after glow mode [51] is under development to bunch the high charge state ions as required by pulsed post acceleration schemes. The storage half-life of confined ions is typically $\tau_{1/2}$=360ms. The after glow ion bunch has a 1ms peak followed by a 5ms tail (pulse duration ~20ms). The efficiency measured for Rb^{15+} is a factor 2 lower than that of an ECR in continuous mode. 10^{11} Rb ions were simultaneously trapped in the ECRIT (full charge spectrum).

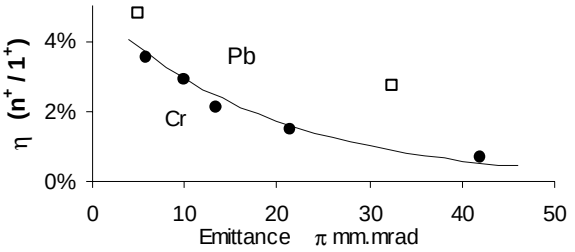

Figure 1. Cr^{11+} and Pb^{24+} Breeding efficiency (ECR continuous mode) vs. 1^{+} ion beam emittance (18kV)[49].

Table 2 Charge state breeding efficiencies: $\eta(n^{+}/1^{+})$ including the injection losses of the 1^{+} beam. The confinement time τ in the breeder is given. The trapping efficiency of the penning trap is not included in the EBIS measurement (*).

	$\eta(n^{+}/1^{+})$	τ *ms*			$\eta(n^{+}/1^{+})$	τ *ms*	
Rb^{9+}	2.8%	50	[47]	Pb^{24+}	4.2%	200	[51]
Kr^{9+}	6.5%	60	[47]	Rb^{15+}	5.0%	200	[51]
Xe^{14+}	4.0%	70	[47]	Cr^{11+}	3.5%	200	[49]
Kr^{9+}	9.2%	100	[51]	$*N^{7+}$	30.0%	40	[53]
Ar^{8+}	9.0%	100	[51]	$*Ar^{14+}$	9.4%	40	[53]
Zn^{9+}	3.5%		[51]				

3.2 EBIS based charge state breeding

The REX-ISOLDE [52] experiment aims to accelerate the low energy ion beams of the ISOLDE facility up to 2.2MeV/u and perform in its first phase nuclear structure studies of the neutron rich isotopes of Na, Mg, K and Ca. EBIS provides high charge states, excellent beam quality and tunable time structure [53]. At a repetition rate of 50Hz, the EBIS is designed to deliver 10μs ion bunches of up to 2×10^{7}ions. The emittance of the 60kV ISOLDE ion beams, typically 30π·mm·mrad has to be reduced to match the design acceptance (3π·mm·mrad) of the EBIS. A penning trap [54] working with low noble gas pressure of 0.01mbar was designed to accept, thermalize the ISOLDE beam decelerated to 70eV and deliver the ions as bunches with an emittance below 3π·mm·mrad. The trapping losses shall be overcompensated by the improved capture in the EBIS. The lifetime of trapped ions mainly depends on the purity of the gas, for Cs trapped in Ar, the lifetime yields 400ms independently of the total Ar pressure between 0.2 and 2.6 10^{-4} mbar [55]. The 1^{+} confinement time in the REX trap is of the order of 10ms. The chemistry of the trapped element and its ability to bind to the rest gas traces should affect its trapping lifetime. The noble gas of the trap is therefore purified with a set of Zr-V-Fe non-evaporable getters. A summary of the available breeding data is shown in figure 3.

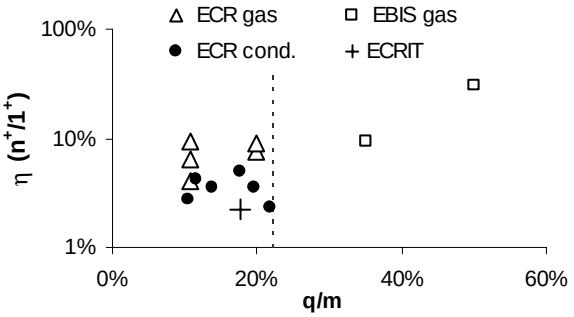

Figure 3 Charge state breeding efficiencies (including the injection losses) as a function of the charge to mass ratio. The values from table 2 are represented with triangles for gaseous, full dots for condensable or reactive elements ionized in an ECR breeder and squares for gaseous elements tested in the DIONE EBIS (Orsay). The q/m=1/4.5 dotted line is the specification of the REX-ISOLDE experiment.

4 CONCLUSION / SUMMARY

Complementary production mechanisms are available for the enthusiastic RIB community. The fast, efficient, and selective ion sources that exist for most elements are now tuned to specifically match the desired isotope. Beside the physics of ion sources and high temperature chemistry which rules the selectivity of ISOL ion sources, the ion guide and trap technologies [56-59] are

now a growing part of the equipment of RIB facilities. They adapt the phase space of high emittance ion beam to the acceptance of charge state breeder and linear accelerators. Last but not least, the intersection of nuclear medicine, biophysics, fundamental interactions studies, solid state, atomic, nuclear and astrophysics is research with radioactive ion beams and the origin of these fruitful developments.

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