FIRST ECR-IONIZED NOBLE GAS RADIOISOTOPES AT ISOLDE

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Abstract

The production of light noble gas radioisotopes with high ionization potentials has been hampered by modest ionization efficiencies for standard plasma ion-sources. However, the decay losses are minimal as the lingering time of light noble gases within plasma ion-sources is negligible when compared to its diffusion out of the target material. Previous singly charged ECRIS have shown a higher efficiency but also a lingering time of the order of 1 s and a total weight that prevents remote handling by the ISOLDE robot. The compact MINIMONO efficiently addressed the lingering time and weight issues. In addition, the MINIMONO maintained the high off-line ionization efficiency for light noble gases.

This paper describes a standard ISOLDE target unit equipped with a MINIMONO ion-source and the first tests. The ion-source has been tested off-line and equipped with a CaO target for on-line tests. Valuable information was gained about high current (100-500 μ A) transport through the ISOLDE mass separators designed for <10 μ A. The yields and release curves (merged diffusion and effusion lingering time) of several light gaseous elements produced via irradiation by 1.4 GeV protons of the CaO target (thickness of 4.6 g/cm²) are presented and compared with those obtained under similar condition with a plasma ion-source.

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1 Introduction and motivation

At the ISOLDE/CERN facility [1,2] radioactive ion beams of about 70 elements and almost 700 isotopes are produced with the isotope separator on-line (ISOL) method. Until now ISOLDE makes use of three different types of sources based on surface, resonant laser and plasma ionization [3], each source type being dedicated for a certain category of elements.

The production at ISOLDE of light noble gas radioisotopes with high ionization potentials and short transit-time through the ionizing plasma [4,5] has been hampered by modest ionization efficiencies for standard plasma ion sources. With the cold plasma ion source of FEBIAD type (ISOLDE MK7), the following relatively modest noble-gas ionisation efficiencies have been recorded on-line: He<0.19%, Ne<0.36%, Ar<2.0%, Kr<4.3% and Xe<11% [6]. Moreover, C, N and O are particularly troublesome since also their volatile molecular compounds CO, CO₂, N₂ and O₂ are very reactive in hot enclosures, as found in plasma ion sources. Nevertheless, the decay losses are minimal as the lingering time of light noble gases within plasma ion-sources is negligible when compared to its diffusion out of the target material.

The answer to the above listed problems could be a cold-enclosure ECR ion source with high ionization efficiency for light elements with high ionization potential, and low reaction rate on the cold surfaces. Even fluorine and sulphur ions/molecules could be ionized with some efficiency. In a 2.45 GHz ECRIS electron densities and energies up to 10¹¹ cm⁻³ (plasma cut-off limit) and tens of eV can be obtained.

ECR ion sources for radioactive ions exist already at, for example, TRIUMF [7] and JINR [8]. In collaboration with GANIL we decided to merge the GANIL MINIMONO ECRIS [9] with a standard ISOLDE target unit. This source had demonstrated a high ionisation efficiency and acceptable delay time. The operation at 2.45 GHz permitted the use of a coaxial cable for the RF transmission, which resulted in only minor modifications of the front-end. The low frequency leads, in general, to a less highly charged ions, a smaller transverse beam emittance, and very cheap magnetron systems for the RF generation are available on the market.

2 Technical design

2.1 General comments

One should bear in mind that increased ionisation efficiency should not be obtained at the price of longer atom/ion lingering time within the ionisation volume, as the decay losses reduce the yield. Previous singly charged ECRIS, such as MONO1000 [10] and MONO300 [11], have shown a high efficiency but also a lingering time up to 1 s. Scaling down the size to MINIMONO efficiently addressed the lingering time and weight issues. In addition, the MINIMONO maintained the high off-line ionization efficiency for light noble gases. Investigation of the ionisation efficiencies and times are presented in ref. [9,12]. A summary of the relevant performance values are listed in Table 1.

Element	Off-line [12,13]			On-line [14]
	Ionisation efficiency	T _{response} (50%) (ms)	T _{response} (90%) (ms)	Ionisation efficiency
He	6 %	20	50	N.A.
Ne	13 %	30	150	²³ Ne 1.5 %
Ar	90 %	40	250	³⁵ Ar 14.7 %
Kr	90 %	40	N.A.	⁷³ Kr 13 %

Table 1. Measured ionisation efficiencies and response times for a number of gases. T_{response} (50%) is the transient time from the inlet of a gas pulse until 50% of the atoms that are ionised are extracted. Ditto for T_{response} (90%).

As the design of the MINIMONO has been described elsewhere, only a short summary will be given here with the main parameters listed in Table 2. The magnetic field formation is done by NdFeB permanent magnets, creating a min-B field with closed surfaces up to 2000 Gauss. The microwave power, typically <100 W, is injected radially into the cavity through the coaxial vacuum transition. The microwave generator is situated on the high voltage platform, and the RF coupling to the plasma is capacitive, in the form of a 10 mm sphere. The cavity, also plasma chamber, is a stainless steel cylinder with a length and inner diameter of 39 and 68 mm, respectively. The exchangeable plasma electrode has an aperture of 2.5 or 4 mm. No mechanical cavity tuning is implemented. Therefore, for a given antenna, varying the support gas pressure and the RF power is the only available tuning to match resonance of the cavity with operating plasma to the microwave frequency.

2.2 ISOLDE target unit integration

Size, weight and limited connection possibilities were the main constraints when merging the MINIMONO with an ISOLDE target. Modifications of the front-end to which the target is connected were constrained by a standardized connection interface with other target ion-source types and a high radiation level preventing manual work. No maintenance is possible once the device has gone on-line, it is a consumable. The lifetime is determined by the release out of the target material. Thus, a simple and cheap design was chosen. The MINIMONO could successfully be fitted within a standard ISOLDE target unit (see Figure 1). An N-type quick-coupling was introduced to transmit the RF to the source. A total target unit weight of only 22 kg enables remote robot handling.

Total weight	22 kg
Microwave frequency	2.45 GHz
Microwave power	<100 W
Cavity dimensions	length 68 mm, diameter 39 mm
Magnetic mirror ratio, $B_{\text{ext}}/B_{\text{res}}$	2.0
Plasma electrode aperture	2.5 or 4 mm
Pressure within cavity	$3 \cdot 10^{-4}$ to $2 \cdot 10^{-3}$ mbar
Extraction voltage	tested up to 60 kV

Table 2. Main characteristics of the MINIMONO and ISOLDE target unit.

The ion-source is enclosed in a vacuum vessel with a tantalum oven containing a thick CaO target. From the target a cold transfer line leads the atoms to the ionizing volume. The transfer line is water-cooled to $\sim 15^\circ\text{C}$, and efficiently prevents any condensable elements from reaching the source. To produce helium or neon ions, or even to ignite the plasma when the vacuum in the extraction region is better than $\sim 1 \cdot 10^{-6}$ mbar, a support gas with a low first ionisation potential is required. The support gas, introduced into the transfer line, passes via a fixed leak installed in the target unit. The leak is a modified compressed powder leak with a $6 \pm 1 \cdot 10^{-3}$ mbar·l/s (air, 293 K, 1013 mbar) leak rate. By varying the pressure on the high pressure side between 0.05 and 2 bar the leak rate can be changed. A 25 cm³ gas vessel is incorporated in the target and filled with stable tracers of the radioactive element to be produced. The gas flows via a calibrated leak ($\sim 1 \cdot 10^{-6}$ mbar·l/s for air, 293 K, 1013 mbar) into the transfer line. Therefore, the ionization efficiency can be monitored while tuning the ECRIS.

3 Experimental setups

The target and ion-source unit was first tested with stable beams at the ISOLDE off-line. Thereafter it was taken on-line to the General Purpose Separator [1], for stable and radioactive beam tests. Both separators use electrostatic focusing elements with double focusing separator magnets, and are designed for low currents without space-charge effects (<10 μA). Special attention had to be paid to the non-cooled Faraday cups and wire-grids diagnostics, as the total extracted current from the ECRIS could easily amount to 0.5 mA, corresponding to 30 W. We were reminded of this potential heat load by damaging our tape collection system while tuning the beam transmission with a 50 μA Ar beam.

The CW microwave field was generated with a commercial magnetron system with an adjustable power level. The operation output power was typically 40-150 W, yielding 20-75 W at the MINIMONO due to cable and connector losses.

Two types of extraction electrodes were used. The obtuse angled standard ISOLDE puller and a specially designed puller type represented in Figure 1. In both cases the optimal electrode position was around 40 mm from the ion source outlet (for 60 kV). Two types of plasma electrodes with extraction apertures of 2.5 and 4 mm were used.

Figure 1. The MINIMONO ECRIS merged with a standard ISOLDE target unit.

4 Results

4.1 Stable beam operation at off-line

Due to a pressed schedule for on-line tests, the source could not be fully characterized with stable beam off-line. Ionization efficiencies of 5%, 19% and 55% (corrected for transport losses) for He^+ , Ar^+ and Xe^+ were recorded. The transverse emittance for non-mass-separated beams was measured to be $33 \pi \cdot \text{mm} \cdot \text{mrad}$ (95%) for an extraction voltage of 30 kV, an outlet hole of 2.5 mm, and an extracted current of 150 μA . The phase-space recording is presented in Figure 2.

Figure 2. Transverse phase-space recording of a non-separated beam produced at the 30 kV ISOLDE off-line separator.

4.2 Stable beam operation at on-line

To the ECRIS unit a target container charged with 4.6 g/cm^2 CaO powder was connected. The unit has been tested three times on-line, second time with protons. After out-gassing, 60 kV extraction voltage could be applied without discharges, for pressures up to $3 \cdot 10^{-6}$ mbar in the extraction region. The total extracted current varied between 50 and 500 μA depending on operation conditions. The optimal operation was around 80-100 μA . This should be compared with the usual few μA from a plasma ion source and nA from a surface ionizer. The beam transport efficiency from the source through the separator magnet ranged from 25% to 80%, the higher value for currents below 100 μA . The guard ring voltages (tested with -1000 V) around the electro-static lenses that should keep the beam space-charge compensated in between the lenses had an effect on the beam. Stable beams of Ar, Xe, C, CO/N₂, O, O₂, OH, H₂O among others were produced. The 60 kV switching off at each proton pulse impact had no measurable effect on the plasma.

Encouraging observations from the run was the ease of igniting the ECRIS and the good base pressure ($< 1 \cdot 10^{-6}$ mbar) in the front-end at which the source could still operate. Worrisome is the need for retuning of the beam line settings when the extracted current changes, as it does with varying buffer gas pressure, RF power etc. The source could be operated in two current modes. Either at a low (50-100 μA) or with a high (300-400 μA) total extracted current. We could not confirm if the high current mode had higher ionization efficiency as the lifetime of this mode was only kept for 30 minutes, thus not permitting the completion of the mandatory tuning of space-charge dominated beam through the mass separator. The high current mode was ignited at around $4 \cdot 10^{-3}$ mbar·l/s (for 2.5 mm plasma electrode aperture).

The efficiency tuning of the ECRIS was found to be very dependant on the support gas pressure within the cavity. The time constant of our gas systems made it a real challenge especially in view of the well observable effect of the out-gassing of the target material acting as a non controlled additional support gas. The switch between the ionisation modes observed during the out-gassing of the target is presented in Figure 3. The injected microwave power had less effect on the ionization efficiency.

Figure 3. During the stepwise heating up of the CaO target to nominal temperature, a constant RF-power and a stable argon flow were injected into the ion-source. The out-gassing of mainly oxygen from the CaO matrix was sufficient to act as a support gas for the ECR plasma. The pressure measured in the accelerator tank is indicated and the Ar^+ beam intensity is a figure of merit of its ionization efficiency. This is shown to critically depend on the residual gas pressure as a fluctuation of a factor 2 in pressure affects the argon beam by one order of magnitude.

The ionization efficiencies for stable Ar and Xe were measured to be $>22\%$ and $>29.5\%$, respectively, in good agreement with off-line measurements. The gases were let in via the calibrated leak. If the separator beam transport and the mass slit transmission are included the efficiencies go down to 7% and 9%. The $\text{Ar}^{2+}/\text{Ar}^+$ and $\text{Xe}^{2+}/\text{Xe}^+$ particle ratios were ~ 0.05 , to be compared with 0.1 for an MK7 FEBIAD source. Due to large measurement uncertainties, and a possibility of internal gas leaks, a reservation for the efficiency values has to be made.

The performance of the permanent magnets seemed to be unchanged after approximately $1 \cdot 10^{16}$ 1.4 GeV protons on the target. The axial magnetic field along the symmetrical axis was measured after proton irradiation and compared with a field from a newly built, not yet irradiated, source. The curves are plotted in Figure 4. The field difference is within the margin of the statistical error. The number of accumulated protons is a factor 100 lower than for a normal run. The radiation hardness of MINIMONO's permanent magnets is a potential concern that triggered the development of a coil-based ECR ion-source described in ref. [15].

4.3 Radioactive beam yields of He, Ne and Ar

^{35}Ar , ^{18}Ne and ^6He are radioactive noble gas isotopes that are routinely used at ISOLDE to monitor the functionality of new target and ion-source units. They were used to compare the yields (Table 3) and effusion properties of the MINIMONO to the standard ISOLDE ion-source (MK7). The half-life dependent decay losses are computed from the release curve of these isotopes [16] and presented in Figure 5. The production rates were computed using the Silverberg and Tsao cross sections [17,18].

Figure 4. Measured axial B-field along the symmetrical axis of the source before and after irradiation of the adjacent CaO target with $1 \cdot 10^{16}$ 1.4 GeV protons.

The release curves presented in Figure 6 were measured by successive collections of the radioactive ion beams onto an aluminized mylar tape, the samples are moved into a 4π plastic scintillator where their β -count rates are recorded. The plot of the samples activities as a function of the time elapsed from the last proton pulse is a figure of merit of the diffusion out of the target material folded with the effusion to the ion-source. The so-called release curves were measured with pulses of $1 \cdot 10^{13}$ or $2 \cdot 10^{13}$ 1 GeV protons, at a pulse repetition rate of 14.4 s. The target temperature was 1250°C, within the precision of its calibration identical to the one of the reference unit (CaO-105). This is confirmed by the overlap of the release curves of ^{35}Ar from both target units presented in Figure 6.

While the resonating high tension specific to pulsed proton beam systems did not perturb the plasma, the gas burst released by the target at proton beam impact switched the ion-source mode into its low efficiency mode. Therefore, all these measurements presented in this section were completed while the ion-source was operating in the low efficiency mode described above and illustrated in Figure 3. More work is required to investigate this issue. Other radioactive isotopes (^8He , ^{17}Ne , ^{19}Ne , ^{16}N) and molecules ($^{10}\text{C}^{16}\text{O}$, $^{11}\text{C}^{16}\text{O}$, $^{13}\text{N}^{14}\text{N}$) were produced and identified, their production has to be optimized via specific support gas and ion-source tuning.

Figure 5. Release fraction of the He, Ne and Ar as a function of their half life computed from the data presented in Figure 6.

Figure 6. Release curves of ^{35}Ar , ^{18}Ne and ^6He produced with a CaO target equipped with MINIMONO. The horizontal bar extends over the period of implantation of the radioisotope ion beam onto the aluminized mylar tape. After the typically 900 ms transport time, the β -count rate is measured during 1 s. The data are normalized to a proton pulse intensity of $1 \cdot 10^{13}$ protons and decay loss corrected. The release of ^{35}Ar measured with a standard ISOLDE MK7 is scaled with the ion-source efficiency ratio (little diamonds). The difference visible in the first 100 ms is the signature of a slightly slower effusion and the perfect match after 100 ms (governed by the diffusion process) confirms the identical target parameters settings.

[1/ μC]	S&T	MK7	MINIMONO	Half live	Released fraction
^6He	3.6E+8	2.8E+5	6.6E+4	806.7 ms	81%
^{18}Ne	1.7E+8	1.5E+5	1.6E+4	1.672 s	90%
^{35}Ar	1.3E+9	1.7E+6	1.0E+6	1.775 s	82%

Table 3. Yields of ^{35}Ar , ^{18}Ne and ^6He from a 4.6 g/cm^2 thick CaO target equipped with a MINIMONO ion-source are compared to the yields of a standard noble gas ISOLDE ion-source (MK7). However, following the proton pulse generated gas burst, the efficiency of the MINIMONO dropped by one order of magnitude. The release fractions of the isotopes ionized with MINIMONO are all above 80%. The production rates (S&T – Silverberg and Tsao [17]) are given.

5 Conclusions and Outlook

The first results of radioactive beam production in an ISOLDE target equipped with a MINIMONO ECRIS have been obtained. The yield of light radioactive noble gas isotopes was low in comparison with a target connected to a FEBIAD MK7. Some improvements concerning ionization and transient time still remain until the expectations are achieved. To start with, the high stable beam ionization efficiency values presented in ref. [12] should be reproduced at the ISOLDE off-line. Secondly, the long-term stability with respect to pressure and gas composition variations due to target out-gassing, and achievable ionization performance on-line has to be studied. The permanent magnets showed no field degradation for $1 \cdot 10^{16}$ integrated protons, but it remains to verify the field and the source performance also after a full run with $1 \cdot 10^{18}$ protons and full heating power on the target. The electrostatic transport and separation in the analyzing magnet of total beam currents exceeding 100 μA has to be improved and the causes of losses identified. Finally, if the tests are successful, either one, or both of the tested monoECRIS (MINIMONO or MECRIS [15]) will be included within the ISOLDE standard ion source gallery.

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