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Sub-millisecond atom-to-ion transformation in the TULIP ISOL system.

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TULIP (Target Ion Source for Short-Lived Isotope Production) is an original Isotope Separator On Line project dedicated to the production of short lived neutron deficient ions. It was tested for the first time in March 2022 at GANIL (Grand Accélérateur National d'Ions Lourds) / SPIRAL1 (Système de Production d'Ions Radioactifs Accélérés en Ligne, phase 1) with the production of Rb isotopes using a primary beam of ^{22}Ne at an energy of 4.5 MeV/A. ^{76}Rb and ^{78}Rb isotope rates of $\sim 4 \times 10^3$ and $\sim 6 \times 10^4$ pps were measured for a primary beam intensity of 6×10^{12} pps and may be improved by a factor between 5 to 10.

A remarkably short atom-to-ion transformation (AIT) time close to 200 μs was achieved. This result demonstrates how beneficial the design of the TULIP target ion source system is in term of response time and allows us to confidently look forward $^{74}\text{Rb}^+$ production, which is one of the goal of the TULIP project.

The results obtained on Rb production suggests testing the principle for the production of more sticky elements. Ions from metallic isotopes around ^{100}Sn will be the next objective.

Key words: SPIRAL1, radioactive ion beam, ISOL, neutron-deficient Rb isotope, TULIP project

1. INTRODUCTION

Exploring nuclear matter far from the valley of stability is a major challenge in nuclear physics. It often requires systematic studies of a number of exotic nuclei along isotopic or isotonic chains, or the availability of a given short-lived isotope at a high enough intensity to measure specific observables with a reasonable statistical precision. Several facilities currently operate worldwide on the production of such exotic ions, using two main methods: the Isotope Separator On Line method and the In Flight method [1].

An ISOL system generally uses a reaction target in which radioactive nuclei are produced and stopped. They then effuse up to an ion source where they are ionized before being accelerated. The Target and Ion Source System, referred thereafter as TISS, constitutes the heart of the production device.

The In Flight separation method is based on a beam of stable ions colliding with a thin target. Fragments are produced at a velocity close to the one of the projectile and are selected in a magnetic spectrometer possibly combined to a velocity filter before they reach the experimental setup. Although very short half-lived isotopes can be produced with such a technique, it presents an important drawback especially when relatively low primary beam energies are available: the thickness of the target must be small enough to limit the energy and angular spread of the reaction products, which is detrimental to the final aim of intensity and purity.

Compared to the In Flight method, one advantage of the ISOL technique is the lesser effect of target thickness. All the recoils are stopped in the target, whatever their energy and angular spread. The in-target production can thus be larger. Nonetheless, in ISOL production systems, half-lives of the produced isotopes are generally limited to some tens of milliseconds due to the physical processes involved between the instant the radioactive nuclei are produced in the target and the instant they are transformed into accelerated ions [2,3,4]. Diffusion of atoms out of the target material, effusion in the vacuum chamber including sticking on the chamber walls and ionization take time, depend on the chemical properties of the atoms, and on the TISS materials and temperature. The competition between radioactive decay and these processes limits the atom-to-ion transformation (AIT) efficiency of isotopes with half-lives longer than several tens of milliseconds.

One obvious solution to obtain sufficient ion production rate when AIT efficiency is low consists of increasing in-target production rate. However, in case of short-lived isotopes, this solution must be carefully studied. High in-target production rates are generally obtained using more intense primary beams, involving larger targets to sustain the power deposited by the beam and more complex geometries and target micro-structures to help the power dissipation. Usually, larger sizes and complex geometry lead to a longer release time out of the target material and to a longer effusion time in the target container. They are finally detrimental to short AIT time. To maximize the ion production rate, a compromise needs to be found between the primary beam energy and intensity, the target design, and the AIT efficiency.

Another criteria used to appreciate the quality of a radioactive ion beam (RIB) is its purity. There are often several reactions channels to produce one particular isotope. The higher the energy involved in the reaction, the greater the variety of reaction products. For example, isotopes obtained by spallation or by proton-induced fission can cover large area of the nuclide chart [5]. As a consequence, greater selectivity can be obtained by lowering the energy available for reactions thereby avoiding more complex downstream filters.

The TULIP project [6] aims to develop an innovative TISS addressing these issues. Its goal is the production of short-lived neutron deficient isotopes for the SPIRAL1 ISOL facility operated at GANIL by fostering the AIT efficiency rather than the in-target production yield. To extend the SPIRAL1 panel of more exotic RIB, the production cave and services were upgraded from 2013 to 2018 to host new types of TISS [7,8]. In this technically constrained environment limiting ionization sources to ECR, hot surface and electronic impact devices, with primary beam energies below 100 MeV per nucleon and thick target material lighter than Nb, a Jugaad innovation approach was mandatory to find a solution to efficiently produce new exotic ion beams.

The first aim of the TULIP project is the production of neutron-deficient Rb ion beams, and this is chosen as a case study. The second aim is the production of neutron-deficient Sn isotopes, both being of high scientific interest for the nuclear physics community. This article presents the first results obtained for Rb production.

2. Principle and setup description

In an ISOL system, the minimization of the diffusion time requires reaction products to be implanted at the surface of the releasing material, target or catcher. This requirement is fulfilled by reaction products having a low kinetic energy and stopped in a single thin foil. For high energy reaction products, the stopping range can be spread over a stack of thin foils to let atoms stop close to the material surface. Nonetheless, stacked foils increase effusion time by complicating the path of atoms in the target chamber. A unique thin stopping foil is best. These two considerations lead to favour the production of reactions products at low energy. Added to the constraint of selective in-target production, the fusion-evaporation or deep inelastic transfer reactions seem to be optimal. Once stopped in the catcher material and released from it, reaction products must be transformed into ions as fast as possible. This means that the target chamber should also be the volume where they are ionized. So we considered

merging the target chamber and the ion source. Finally, to speed up their exit out of the chamber, instead of letting them freely effuse to find the exit, ions must be pushed towards the exit, and so the use of an electric field is appropriate.

A schema of the TULIP principle is given Figure 1. A primary beam hits a solid target foil, at an energy just above the Coulomb barrier to favour nuclear fusion-evaporation reactions. In the case of Rb isotope production, a beam of ^{22}Ne at an energy of 4.5 MeV/A impinges on a 4 μm thick ^{nat}Ni foil. The beam energy and target thickness were chosen to maximize the rate of residuals leaving the target foil and stop then in the first layer of a thick catcher placed behind the target. Using graphite as a material for the catcher, the range of residuals is of the order 10 μm , compared to the total catcher thickness of 200 μm . To mechanically confine the nuclei in the TISS chamber, a thin foil of diamond like carbon is placed between the Ni target and the catcher, the Ni foil alone being unable to support the temperature of the TISS and the beam power deposition. The TISS temperature was maintained at 1300°C to accelerate the diffusion of atoms out of the catcher and to reduce their sticking time on the cavity walls. Ions are then surface ionized and pushed towards the exit of the cavity by an electric field ($\sim 1\text{V/cm}$) induced by the heating current circulating in the cavity walls.

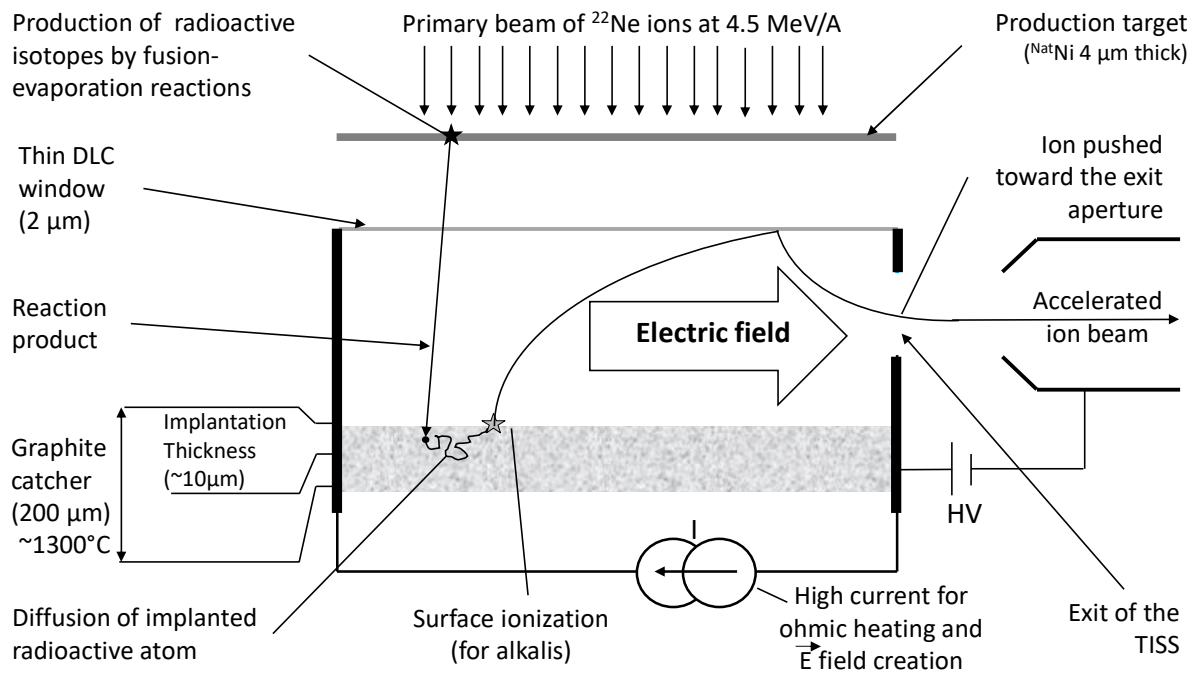


Figure 1: Principle of the TULIP TISS for Rb^+ ion production.

Once out of the TISS, ions are accelerated by means of an extraction electrode, then magnetically separated with respect to their mass to charge ratio. They are then either stopped on an electro-emissive foil coupled to a micro-channel plate (referred to below as micro-channel plate detector, MCPd), or implanted on the movable tape of a gamma ray identification system (GRIs). The MCPd and its electronics allows current measurements with response time longer than ~ 1 ms. Combined with a high detection efficiency (close to 100%), this detector is well adapted to the measurement of AIT time of the order of 20 ms. This was deduced from preliminary tests performed on a test bench, where short pulses of Rb^+ delivered by an ion gun were injected into the TISS. AIT time measurements were performed in transient mode. The period of the square signal driving the primary beam intensity on the target spanned a range between 40 ms and 25 s, ignoring *a priori* the AIT time expected for a given temperature of the cavity. Identification and counting on the GRIs were performed in steady mode (primary beam 100% on the target).

MCPd and GRIs can both deliver a number of counts. The one delivered by the MCPd is expected to be equal to- or higher than the one provided by the GRIs, due to the sensitivity of the MCPd to radioactive as well as stable (contaminating) ions. Owing to the mere hot surface ionization process involved, only alkali ions can be provided. Thus a given isotope of Rb can only be contaminated by other isotopes of Rb. To better evaluate the importance of the contamination observed during the on-line experiment, a specific study was performed off-line one week after the end of the on-line study to get rid of residual radioactive isotopes of Rb. A stable isotopically enriched ^{85}Rb tracer was initially introduced in the TISS. It was still present after the on-line study, with traces of ^{87}Rb ($^{87}\text{Rb}/^{85}\text{Rb}$ relative abundance= 1/400). The beam line and the separator were tuned at masses between 74 and 89 (see Figure 2) while the TISS pressure in the front end chamber connected to the TISS was equal to 10^{-7} mbar. At masses 84 and 86, for which nothing was expected to be measured, ion currents 25% of the ion currents

respectively collected at masses 85 and 87 were observed. These currents were attributed to the detection of $^{85}\text{Rb}^+$ and $^{87}\text{Rb}^+$ ions respectively, slowed down during their extraction of the TISS by collision on the desorption gas. To observe how sensitive the 84/85 current ratio was to the TISS residual gas, the pressure was doubled by increasing the temperature of the TISS. For a pressure of 2×10^{-7} mbar, the 84/85 current ratio reached the value of 1.

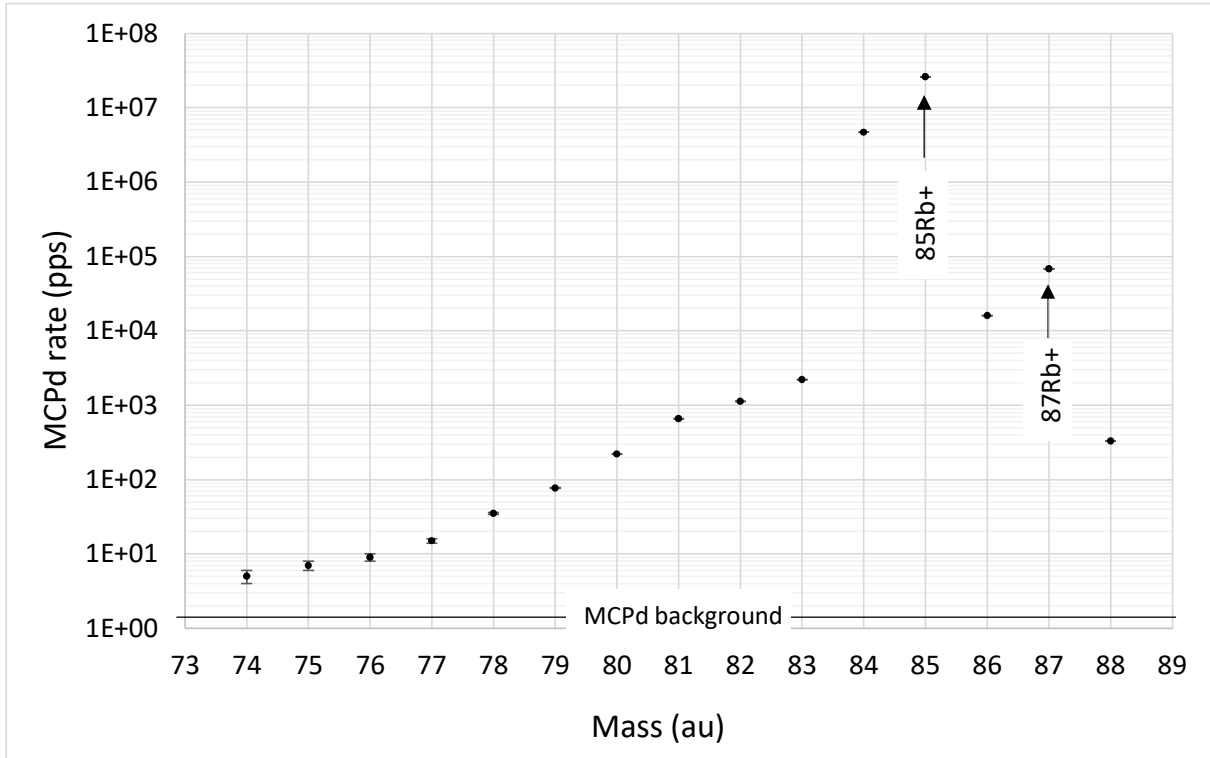


Figure 2: MCPd intensity as a function of the mass selected with the separator

The low current sensitivity of the MCPd allowed to observe a $^{85}\text{Rb}^+$ current up to mass 74, this being the result of the energy spread of $^{85}\text{Rb}^+$ ions induced in their extraction from the TISS. This observation partly explains the presence of unexpected contaminants on the GRIs, corresponding to isotopes of mass A+1 when the separator was optimized on mass A.

The pressure and thermal relaxation time of the TISS are thus important parameters to monitor, and must be taken into account in order to reduce the contamination level of the selected RIB. Due to the short time dedicated to this instrumental study and our ignorance of this contamination source during the on-line experiment, the relaxation time (of the order of 10 minutes at 50%) was not systematically respected during the experiment. When the separator was tuned to observe a given isotope of $^A\text{Rb}^+$, the MCPd counting was thus generally higher than the actual intensity of $^A\text{Rb}^+$ ions.

3. On-line atom-to-ion transformation time measurement

The ^{22}Ne primary beam turns around the beam axis with a wobbling frequency of 20 Hz to spread the power of the beam over the whole surface of the target. To measure the AIT time, the beam was chopped at different periods, with a duty cycle of 50%. Four signals were simultaneously measured (Figure 3).

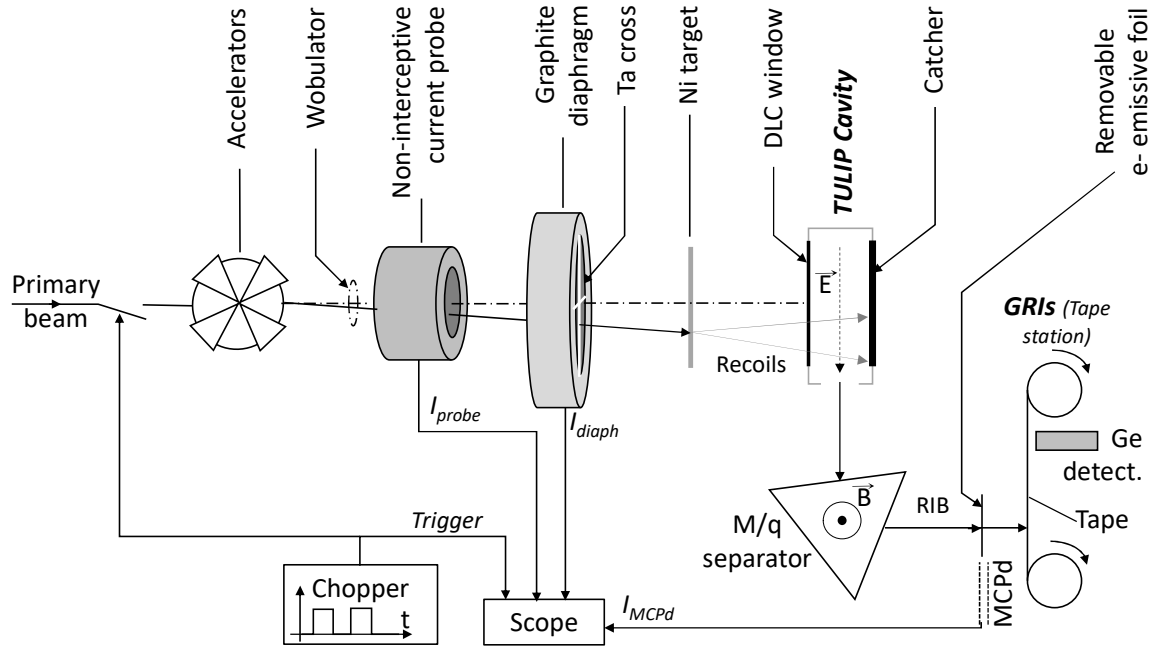


Figure 3: Schema of the setup.

A TTL signal (*Trigger*) serves as a time reference. It triggers the primary beam chopper and the oscilloscope (100 MHz max. frequency, 2 GHz sampling frequency, 8-bit amplitude encoding).

A non-interceptive calibrated probe measures the current I_{probe} of the primary beam, 2 m upstream from the TISS.

A carbon diaphragm is placed at the entrance of the TISS. It is present on all SPIRAL1 TISS and aims at centring the beam on the target by minimizing the current I_{diaph} it collects. For the TULIP TISS, it was equipped with a Ta crosswire, which aims to collect 15% of the beam and determine the instant the beam is sent on the TISS.

The MCPd collects the current I_{MCPd} of Rb^+ ions. All four signals were recorded on the acquisition oscilloscope.

There is no apparent delay between the instant the trigger signal is ON and the instant the currents I_{probe} and I_{diaph} start increasing (Figure 4). This means that the delay induced by the trigger signal transmission up to the primary beam chopper, the chopper commutation, the acceleration of primary beam ions in the cyclotrons and the beam transport is negligible on the millisecond time scale. The rise time of I_{probe} is due to the rise time of the electronic chain. The rise time of I_{diaph} is due to the RC time of the cable and connection between the diaphragm and the electronics. The 1.75 ms measured (Figure 4) is close to the 1.5 ms calculated. The rise time of the beam current on the TISS target is thus of the order of 0.25 ms.

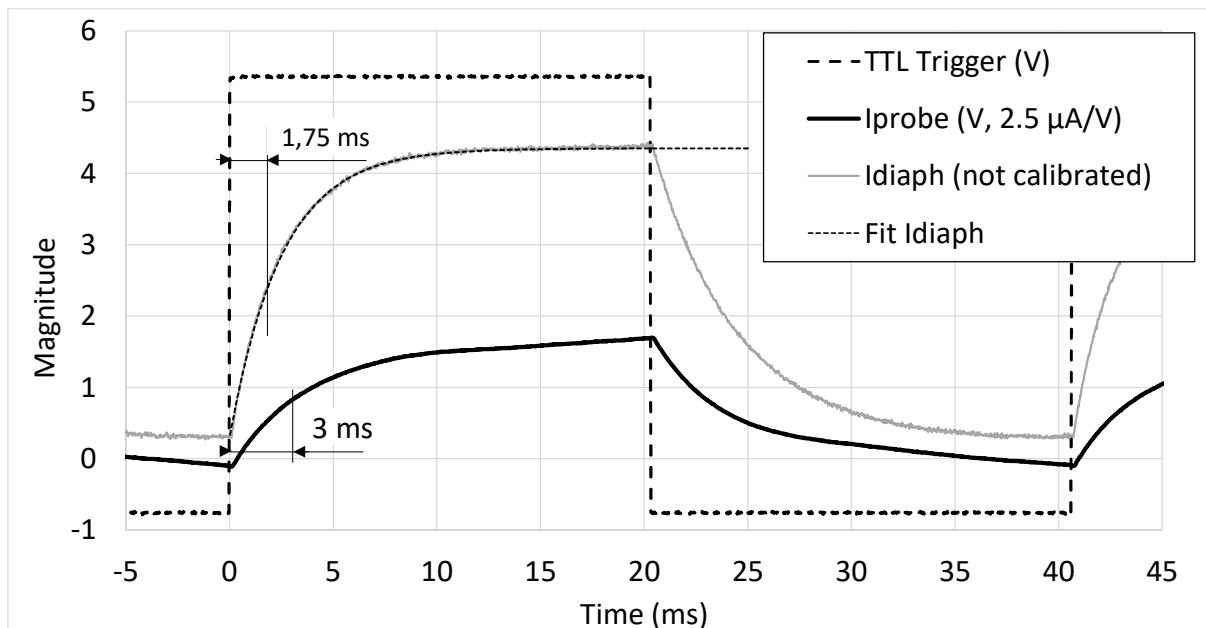


Figure 4: Chopper driving signal, primary beam current and current collected on the diaphragm versus time

The response time of the MCPd for the collection of $^{76}\text{Rb}^+$ is shown Figure 5. It increases almost immediately after the chopper signal is ON at $t = 0$, and reaches 50% of its steady magnitude after only $\sim 650 \mu\text{s}$.

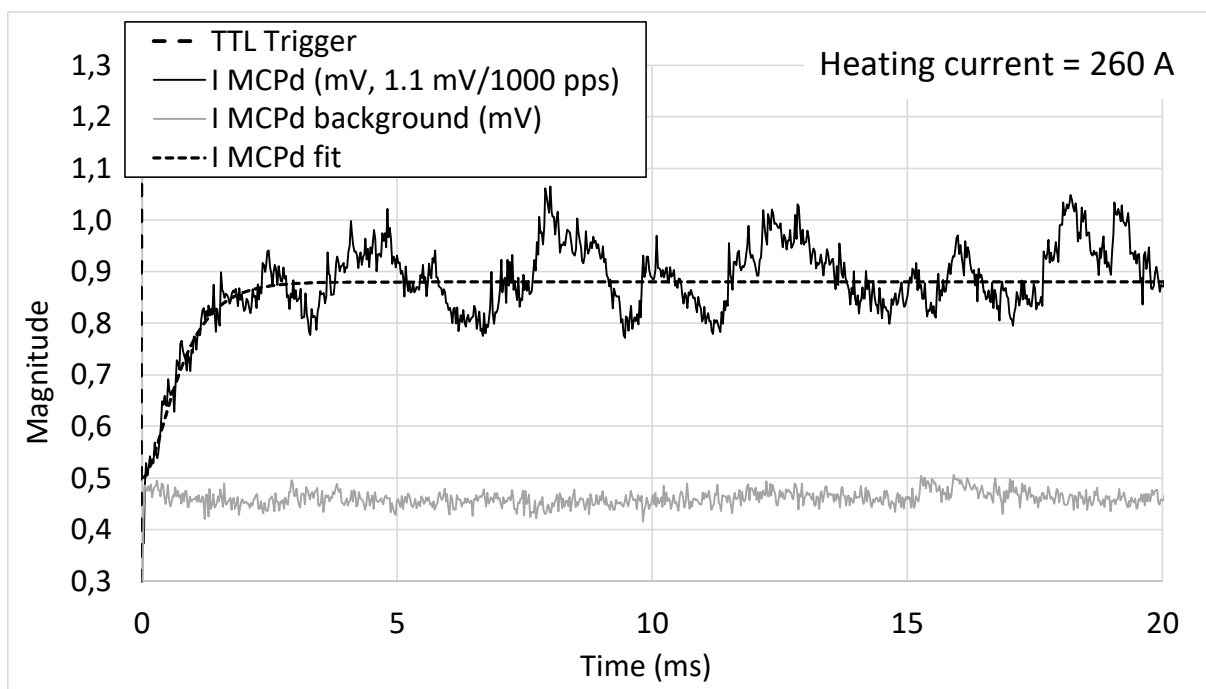


Figure 5: Chopper driving signal, $^{76}\text{Rb}^+$ signal on MCPd, MCPd background and MCPd $^{76}\text{Rb}^+$ intensity fit versus time.

To extract the AIT time from the MCPd response, the latter must be corrected for the intrinsic response time of the MCPd, which depends on the MCPd itself and on its associated electronics (fast amplifier, discriminator and NIM to TTL signal conversion). The electronics is expected to have an intrinsic response time well below 1 ms. The output signal from the MCPd is a logical signal which frequency is equal to the number of ions per second impinging the electro-emissive foil. This logical signal is injected into an analogic frequency to voltage converter (FVC) connected to a channel of the oscilloscope triggered by the chopper signal. The slowest part of the chain is the FVC, designed while expecting an AIT time of the order of 20 ms. The FVC was thus rather slow, and not adapted to response times shorter than 1 ms. To measure *a posteriori* precisely its response time, a TTL signal with a frequency ranging from 4 kHz to 40 kHz, and with a 200 ns width identical to the signal delivered by the MCPd electronics was injected into the FVC (see Figure 6).

The electronic response time varied from $360 \mu\text{s}$ to $435 \mu\text{s}$ for a frequency ranging from 4 to 40 kHz.

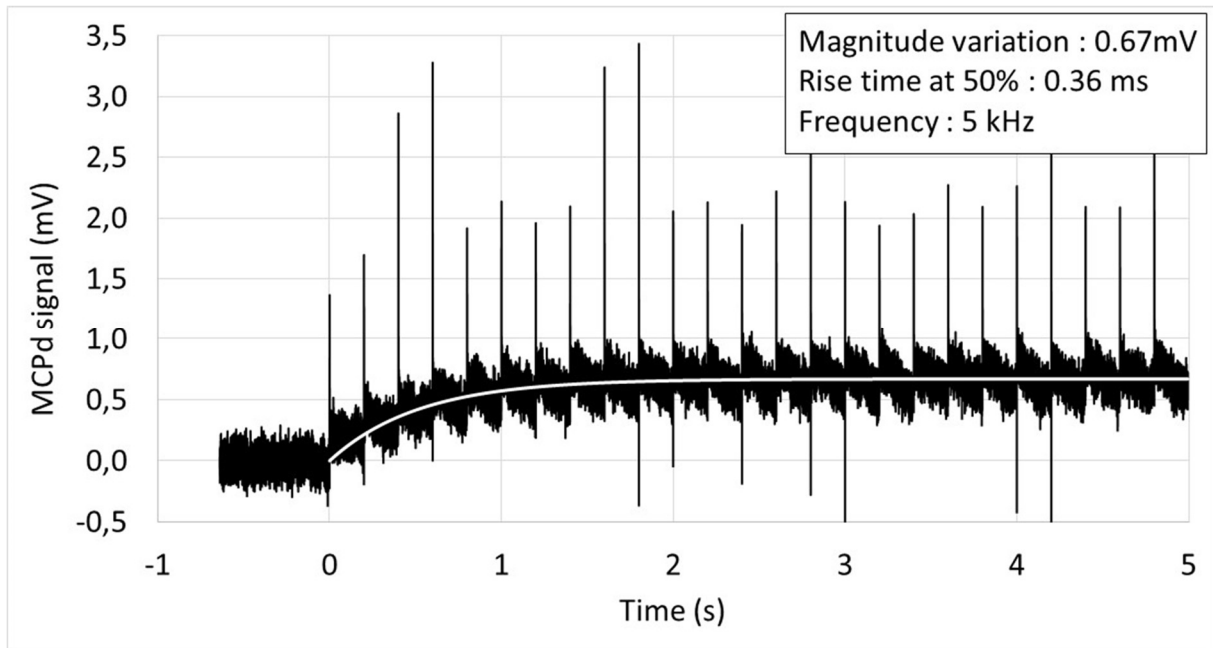


Figure 6: Response time of the FVC submitted to a sudden 5 kHz frequency at time $t = 0$. White line corresponds to an exponential fit.

Given the MCPd and FVC electronics were in series with the TISS, the overall response time is taken to be a simple combination of two exponential functions. Their deconvolution allowed deriving an AIT time of $\sim 200 \mu\text{s}$. The simplicity of the measurement makes this result reliable but the AIT time is so short compared to the minimum time of some tens of millisecond usually observed in ISOL systems that it had to be confirmed through complementary measurements.

4. Off-line analysis

After irradiation, the TISS was dismantled in a glove box. The only damage observed was a trace of the primary beam impact on the surface of the target (see Figure 7), and a hole presumably induced by a localized excessive beam power deposit. Their curvature indicates a beam rotation diameter of $\sim 18 \text{ mm}$, which corresponds to the desired wobbling diameter. The beam was clearly misaligned. According to the red circle shown in Figure 7, it passes alternatively on the target, on the diaphragm situated between the two dot circles, and on the chamber.

The misalignment is due to the presence of the Ta crosswire added to the diaphragm for the purpose of the experiment, which prevented the diaphragm from fully playing its role as the crosswire always collects part of the beam. In presence of the crosswire, centring the beam on the target by minimizing I_{diaph} has proven to be unreliable.

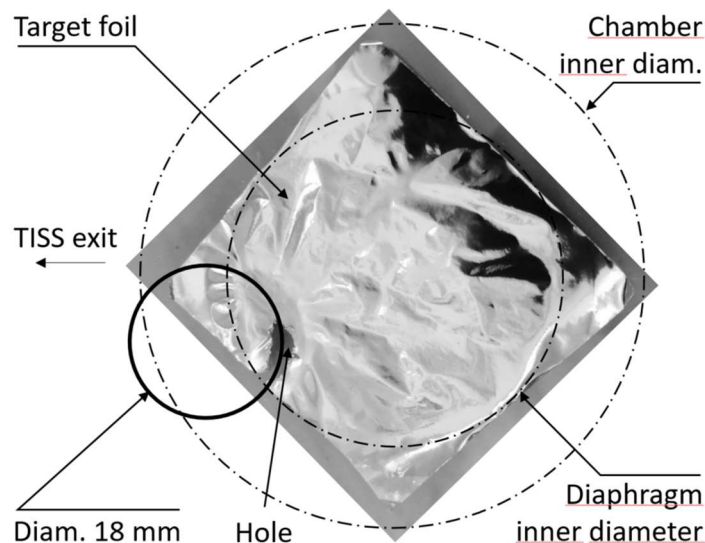


Figure 7: Picture of the target after irradiation

With the combined width beam and its rotation at 20 Hz, the beam current induces a modulation on the mean current collected by the diaphragm (Figure 8). 20 Hz modulated signal is also visible on the current of $^{78}\text{Rb}^+$ ion beam collected on the MCPd. Two 20 Hz sinusoidal waves have been superimposed on these signals to roughly determine the time difference between the instant the primary beam current is maximum on the diaphragm (blue) and the instant the $^{78}\text{Rb}^+$ ion intensity is minimum on the MCPd (red).

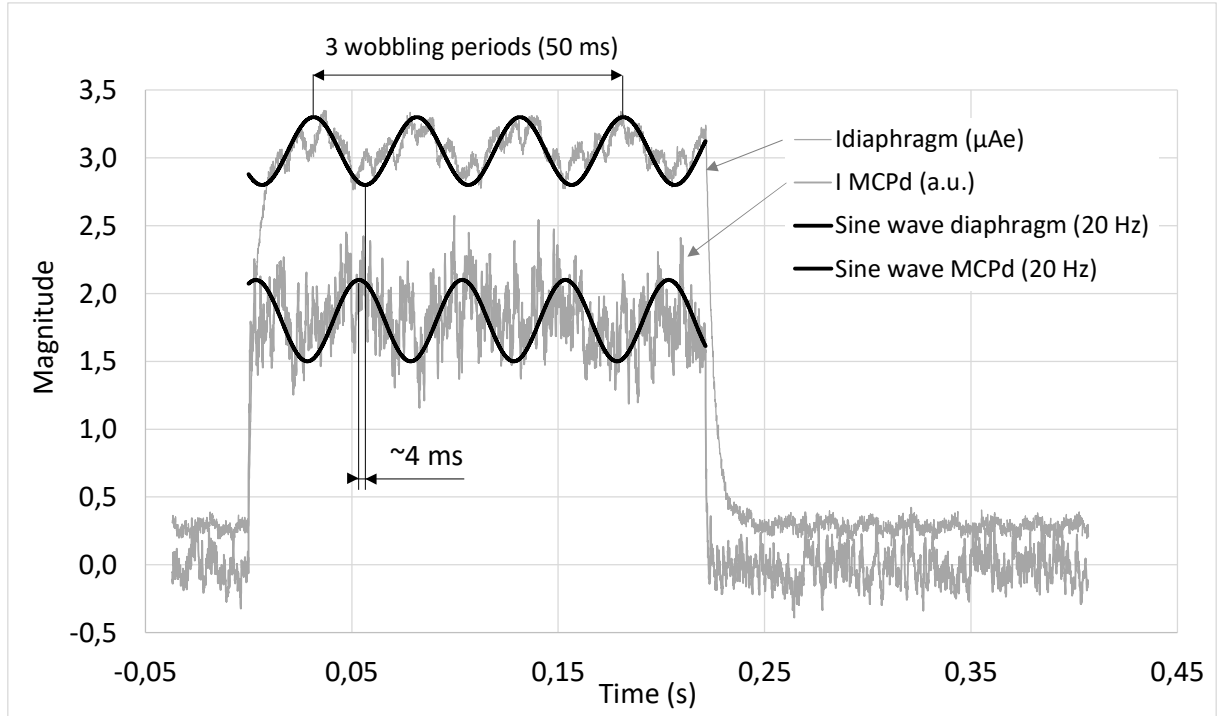


Figure 8: 20 Hz modulations of the primary beam current measured by the diaphragm and of the current of $^{78}\text{Rb}^+$ ions collected on the MCPd.

This time difference is of the order of 4 ms. The maximum MCPd current is slightly ahead of the diaphragm current due to the impedance of the electronic readout of the diaphragm current, which induces a delay on I_{diaph} of 3.9 ms (calculated) at 20 Hz. This leads to an AIT time of the order of 0.1 ms, and is consistent with the AIT time deduced from the direct measurement (0.2 ms, see paragraph 3).

All measurements performed at different temperatures of the TISS led to AIT times ranging from 0 to 400 μs . The AIT time is two orders of magnitude shorter than that (20 ms) obtained during off-line measurements with stable tracers. This can be partly explained by the region of the target irradiated by the primary beam, close to the exit hole of the cavity. Once the reaction products are stopped in the catcher, close to the exit hole, they diffuse out of the catcher material, are immediately surface ionised and extracted by the inner electric field of the cavity. During off-line tests, stable Rb^+ ions were injected into the centre of the cavity. They had first to effuse and stick several times on the cavity walls before leaving the cavity. With a primary beam correctly centred on the target during the on-line experiment, the average AIT time would probably have been longer than 1 ms.

Noteworthy is the fact that since the AIT time includes the release time of Rb atoms out of the graphite catcher, and their sticking time in the cavity. These times are necessarily shorter than 0.2 ms.

5. Radioactive ion production rates

Due to the misalignment of the primary beam on the target, about 10% of the beam actually hit the target, and corresponds to a current of 0.107 μA (6.7×10^{11} pps) of ^{22}Ne . With this intensity, mean production rates of $\sim 4 \times 10^3$ pps and $\sim 6 \times 10^4$ pps of $^{76}\text{Rb}^+$ and $^{78}\text{Rb}^+$ have been obtained respectively (see Table 1). They could be improved with a beam properly centered on the target.

To take into account that the primary beam current varied between 0.587 μA and 1.07 μA depending on the measurements and to make measured rates comparable, the results presented in Table 1 are normalized to a current of 1.07 μA ($\sim 0.107 \mu\text{A}$ impinging on the target).

Table 1: Production rates of $^{76}\text{Rb}^+$ and $^{78}\text{Rb}^+$ deduced from gamma ray decays recorded on the GRIs, normalized to a primary beam current of 1.07 μA . The heating current is indicated.

Rb ⁺ isotope	Heating current (A)	Gamma ray (keV)	Rate (pps) normalized to 1.07 μA of primary beam intensity
76	370	355	4.19 (6) × 10 ³
76	370	424	3.3 (2) × 10 ³
76	250	424	4.0 (3) × 10 ³
76	250	424	4.4 (4) × 10 ³
78	250	455	6.7 (3) × 10 ⁴
78	250	693	5.0 (2) × 10 ⁴
78	250	1148	5.0 (2) × 10 ⁴

The measured production yields of ⁷⁶Rb⁺ and ⁷⁸Rb⁺ are reported in Figure 9. The ⁷⁸Rb⁺ yield measured with the GRIs corresponds to that measured with the MCPd. The ⁷⁶Rb⁺ GRIs yield is lower than that of the MCPd one, and is consistent with a contamination of mass 76 by mass 77 (see paragraph 2). An example of contamination of mass A by mass A+1 is the presence of ⁷⁹Rb⁺ at mass 78, which represented ~1/4 of the yield measured at mass 78.

Rb isotope yields have also been measured for masses ranging from 74 to 80 with the MCPd. They are only indicative of the order of magnitude, due to their potential contamination. It is presently not possible to give a precise production yield for 75, 77, 79 and 80 Rb⁺ ions. A new measurement with the GRIs is necessary.

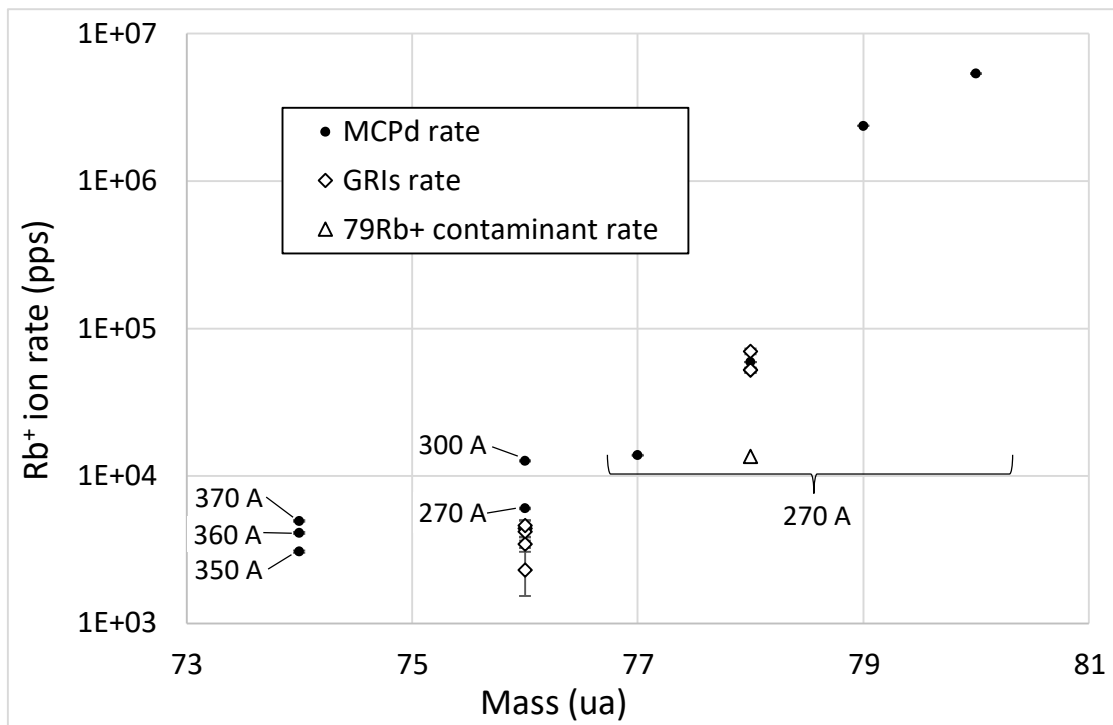


Figure 9: Rates measured on the MCPd (black dots) and on the GRIs (square empty dots with error bars). The triangle corresponds to the intensity of ⁷⁹Rb⁺ at mass 78. The heating current is indicated.

The production of ⁷⁴Rb⁺ is one important aim of the TULIP project, as a proof of principle of the system to deliver short lived neutron deficient ion beams. Estimates from this analysis show realistic production rates of the order of 10³ pps. This estimate is based on the extrapolation of the ⁷⁶Rb⁺ and ⁷⁸Rb⁺ rates (Figure 9), multiplied by a factor of ten to compensate the misalignment of the primary beam, and on the AIT efficiency, which is not impacted by the half-life of ⁷⁴Rb (64.76 ms).

6. Conclusion

Sub-millisecond atom-to-ion transformation has been obtained in an ISOL system for the $^{76}\text{Rb} \rightarrow ^{76}\text{Rb}^+$ transformation. The TULIP TISS was initially designed to reach Rb AIT times of ~ 20 ms. The outcome of the online measurement performed at GANIL is far better than the initial aim as the AIT time obtained is of the order of some hundreds of micro-seconds. The setup was designed for time measurements of some tens of milliseconds and must be adapted for sub-millisecond AIT time measurement.

For Rb, the short AIT time makes the efficiency quasi-independent of the half live of isotopes provided it is larger than 1 ms.

The yields of ^{76}Rb and ^{78}Rb isotopes produced during the experiment are of $\sim 4 \times 10^3$ and $\sim 6 \times 10^4$ pps respectively, with $\sim 1 \mu\text{A}$ ^{22}Ne @ 4.5 MeV/A primary beam and a 4 μm thick $^{\text{nat}}\text{Ni}$ target. Higher yields can certainly be obtained by using a ^{20}Ne primary beam in place of ^{22}Ne , an isotopically enriched ^{58}Ni target and a beam properly centred on the target. This will probably lead to increased AIT times of residuals stopped in the cavity further away from the exit of the TISS. After technical improvements, production yields and AIT times will be measured during new on-line production tests expected in 2023.

Based on the present results, a TULIP version equipped with an electron beam for ionization by electronic impact will also be tested. Its goal is the production of metallic neutron deficient isotopes in the region of ^{100}Sn .

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