

EURISOL JRA WP 14 First Periodic Scientific Report

Steering Committee

A steering committee was put in place with the following members:

- Alberto Andrichetto (INFN – LNL Legnaro)
- Piotr Bednarczyk (IFJ Krakow)
- Yorick Blumenfeld (IN2P3-IPNO, WP leader)
- Manssour Fadil (GANIL)
- Przemysław Gmaj (HIL Warsaw)
- Maher Cheikh Mhamed (IN2P3-IPNO)
- Fredrik Wenander (CERN, deputy WP leader)

Milestones and Deliverables

No milestones or deliverables were planned for the first reporting period

Meetings

The following meetings were organized during the reporting period

Kickoff Meeting: June 17, 2016 at Orsay, France; 10 participants

CRIBE (task 3) meeting: October 19, 2016 at Leuven, Belgium; 8 participants

BeamLab (task 2) meeting: March 20, 2017 at Legnaro, Italy; 11 participants

ICBT (task 1) meeting: March 30, 2017 at CERN, Geneva; 8 participants

Publication

“The TwinEBIS setup: Machine description”, M. Breitenfeldt, R. Mertzig, J. Pitters, A. Shornikov, F. Wenander, Nucl Instrum Meth A, Volume 856, 1 June 2017, Pages 139-146, <https://doi.org/10.1016/j.nima.2016.12.037>

Conference Presentations

“EURISOL in ENSAR2” presented by Yorick Blumenfeld at the EPS Divisional Conference: Towards EURISOL Distributed Facility, Leuven, Belgium, 18-21 Oct. 2016

“Charge breeding techniques for European RIB facilities” presented by Pierre Delahaye at the ARIS Conference in Colorado, USA 27 May – 2 June 2017

I ICBT task

Scientific coordination: Fredrik Wenander (CERN)

Introduction

In this section we report on the status and progress of the Innovative Charge Breeding Techniques (ICBT) task of the EURISOL Joint Research Activity (JRA), in the framework of the ENSAR-2 program. The progress of the ICBT JRA will be benchmarked against declared deliverables and milestones and their respective timelines. There are 2 deliverables and 1 milestone related to ICBT:

Deliverables

- D14.1 Report on performances of the EBIS debuncher (Month 24)
- D14.3 Conceptual design report of a new generation charge breeder (Month 36)

Milestones

- MS14.2 Experiments to find the optimal breeder configuration (Month 24)

As we see, none of the deliverables/milestones were due by month 17. Nevertheless, we will take a more detailed look to each point in order to demonstrate and discuss the present status and progress made towards the final goals.

Progress report on individual deliverables and milestones

D14.1 Report on performances of the EBIS debuncher (Month 24)

By month 15 the EMILIE debuncher is being prepared at LPC Caen for further commissioning tests. After a successful initial commissioning proof-of-the-principle campaign with Na⁺ ions [1] the program planned for the autumn of 2017 focuses on experimental studies that are better representing the future operational conditions.

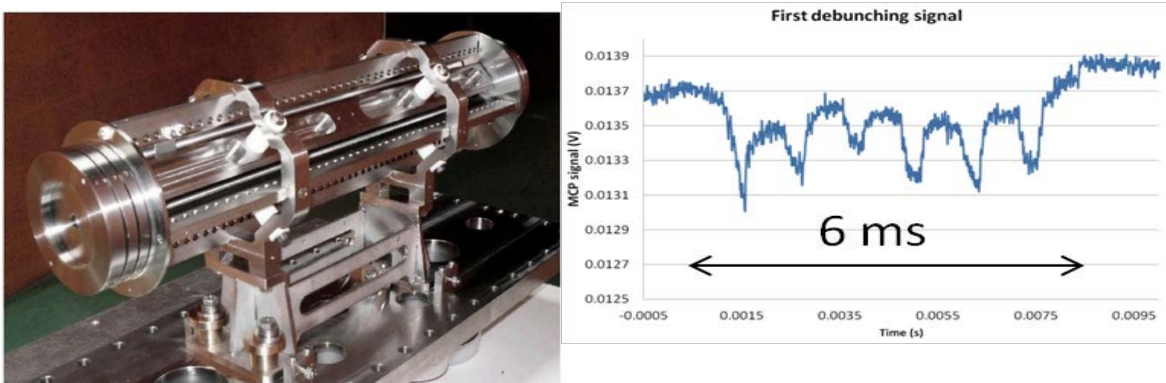


Figure 1. EMILIE debuncher (left) and debunched Na⁺ ion beam (right).

The preparation campaign has started for the new tests. The first important difference is the use of Li ions having an A/q -ratio typical for Highly Charged Ions (HCI), thus having similar particle dynamics in the trap. This modification will allow the estimation and optimization of the storage efficiency of HCI. Proof-of-principle tests demonstrated that substantial improvements can be made in storage

efficiency and uniformity in time of the extracted beam. In order to improve the storage efficiency the position of the injection quadrupole triplet was recently adjusted. The uniformity of the extracted beam pulse will be improved using slow gradual extraction from the individual trap segments, similar to slow extraction from EBISes where the potential barrier is gradually lowered. A Li source was ordered and delivered and is presently under conditioning.

D14.3 Conceptual design report of a new generation charge breeder (Month 36)

As was shown in previous studies [2] [3] [4] the new generation charge breeders (CB) require substantial leap in several technologies, first of all in the technology of high-compression electron beams for Electron Beam Ion Sources (EBIS). While many of the EBIS sub-systems such as drift tube, main magnet or high-power collector can be based on the existing devices, the high-compression electron beam optics for required parameters have no fully functioning prototype. The feasibility of such is vital as several earlier projects failed to deliver the design specifications for reasons not manifesting themselves in numerical studies. Thus, a safe conceptual design should be based on a solid prototype, and use only scaling which relaxes constraints that have already been proven achievable. Following this approach we pay great attention to early prototyping and tests.

As an example of a technical specification for a new generation EBIS-based CB we can take parameters considered in the HIE-ISOLDE design study recently carried out at CERN for the upgrade of ISOLDE facility [5]. For the CB part of the study atomic, plasma and accelerator physics aspects were taken into consideration. The technical specifications matching the demands of future radioactive ion-beam facilities were weighed against technological risks. The key electron-beam optics parameters are listed in Table 1.

Table 1. Approximate electron beam parameters of a new generation EBIS charge breeder

Parameter	
Electron beam energy, keV	20-150
Electron beam current, A	2.7
Electron current density A/cm ²	>1x10 ⁴

For the new generation EBIS charge breeder there are two projects which can serve as early prototypes. The first is the HEC2 electron gun project, jointly started by BNL and CERN, which conclusion is an important part of the ICBT task. The other project is the development of MEDeGUN electron optics at CERN. While most of the work is funded from an independent source, it is partly based on HEC2 proceeds and profits from participation of the ICBT manpower, as the results are of great interest and importance for ICBT. In this section we will describe how the progress of HEC2 and MEDeGUN prepare the ground for an experimentally-backed concept of a new generation EBIS charge breeder.

The HEC2 project was started in parallel with the preparation phase of the ENSAR-2 application and was supported by other means in 2013-2015. The construction and tests performed at this stage are reported elsewhere [6]. While demonstrating proof-of-principle performance and allowing us to better understand the high compression electron optics, the HEC2 first only achieved an electron current of about 1.5 A out of a design value of 10 A. Even if an ideal beam compression performance predicted by Herrmann theory [7] was achieved, the 1.5 A full current in a 3.3 T magnetic field only a maximum current density of 3.5×10^3 A/cm² could be anticipated.

At this point the CERN participation in the project ceased with the lack of funds and was only resumed under ENSAR-2 support. With the lessons learned from the first tests the design of the HEC2 gun underwent several iterations. In its final form the HEC2 gun achieved 3.1 A, outperforming the revised design specification of 2.7 A, and making it the first experimentally tested prototype for a future EBIS CB. The limitations of this test come from the fact that no direct measurement of the electron beam compression was possible during the tests, and no future tests will be conducted due to other priorities at the BNL EBIS test stand. In this situation we have to use estimations based on numerical simulations to characterize the beam current density. We know from the experiment that only minor beam losses (~20 mA) occurred when passing the magnetic field gradient from low field at the gun to full magnetic field strength. Low losses indicate that the electron beam has an acceptable quality in terms of transverse emittance, and can therefore be approximated with simulations. From those simulations we should anticipate a current density of about $6.9 \times 10^3 \text{ A/cm}^2$ for the used test conditions (3.3 T magnetic field, 35 keV electron energy). The TestEBIS magnet is rated for 4.8 T but was used at 3.3 T for safety reasons. Assuming the beam quality would also be sufficiently good to inject into the full field of 4.8 T, the achievable current density would reach up to $10.1 \times 10^3 \text{ A/cm}^2$.

The incompleteness of the HEC2 results (in terms of direct current density measurements) can be overcome with the first experimental results coming from the MEDeGUN electron gun that is being built [8] and prepared for tests on the recently re-commissioned TwinEBIS test stand at CERN [9]. Being smaller in electron current and energy than future charge breeder demands, MEDeGUN nevertheless puts stricter constraints on the beam quality. Both HEC2 and MeDeGUN have area compression factor of 10^4 , assuming a 5 T field and Herrmann theory, but MEDeGUN is designed for a substantially lower electron energy (7.5-10 keV vs 35-50 keV). The stricter limitations at lower energies come from two major sources: First, any beam divergence at the source will be more pronounced at lower beam energies after acceleration. Such divergence may exceed the acceptance angle of the high-field solenoid magnet and may cause beam reflections due to magnetic bottle effect. This is especially important for the outermost part of the beam, where fringe field effects disturb beam trajectories and creates a low-quality halo around otherwise high-quality beam. The second source is the space-charge effects, as the beam space-charge field will be stronger for lower beam energies at the given beam current. Therefore, beam optics performing well at lower energies should perform even better at higher energies, provided that technological aspects related to higher voltage, sparks and discharges are taken care of. Thus, it is a valid prototype for the future charge breeders as the up-scaling to higher energy will relax the requirements on beam quality and manufacturing tolerances. By the time of the writing of this report MEDeGUN is being commissioned.

As summarized in Table 2 both HEC2 and MEDeGUN have design parameters close enough to the demands of the next generation charge breeder to serve as valid prototypes. Should the MEDeGUN tests be successful we will have an early-phase solid base for the key component of the new generation charge breeders. Having such a base greatly reduces future risks, cost, contingency budgets and development time as the rest of the components do have prototypes in other projects.

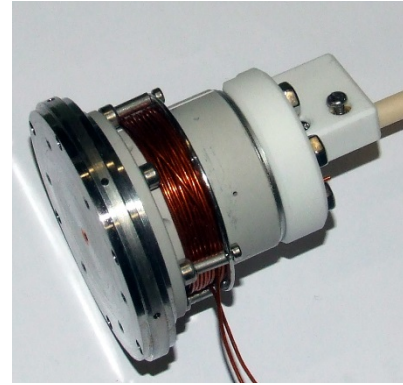
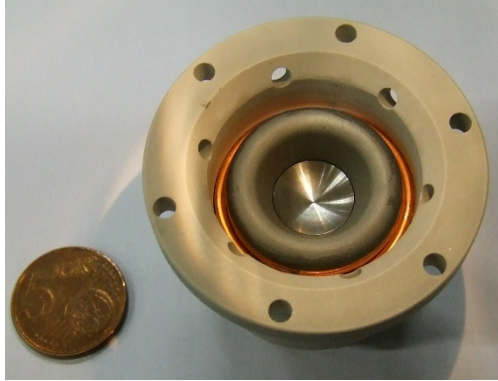
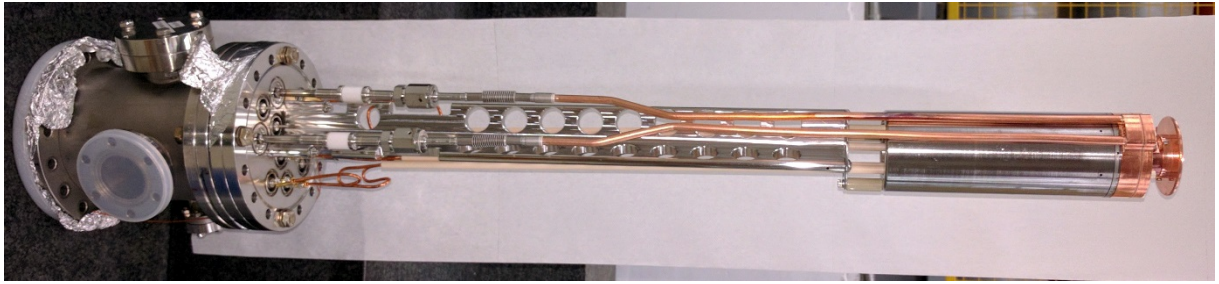


Figure 2. MEDeGUN assembly mounted (top) and its core unit with cathode, single isolator base piece and film-deposited on isolator focusing electrode (bottom left). Assembled MEDeGUN interior with cathode unit mounted on the front magnetic shield and with interior coil wound around the isolator (bottom right).

Table 2. Comparison of next generation EBIS CB characteristics to its prototypes HEC2 and MEDeGUN.

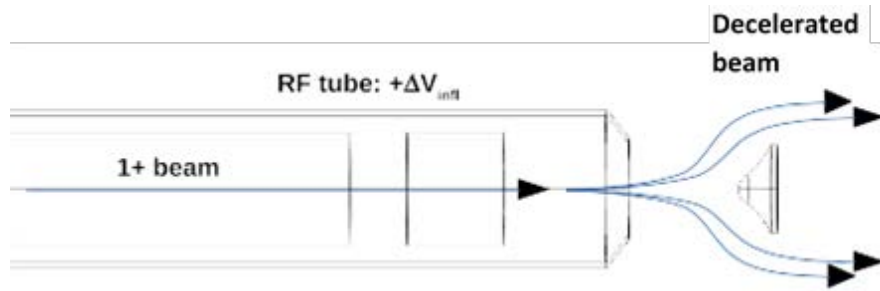
Parameter	Next generation charge breeders	HEC2 (experimental)	MEDeGUN (design)
Electron beam energy, keV	20-150	35	10
Electron beam current, A	2.7	3.14	1
Electron current density A/cm ²	>1x10 ⁴	6.9x10 ³ (estimated) in 3.3 T field	3.1/7.5 x10 ³ in 2T/5T

MS14.2 Experiments to find the optimal breeder configuration (Month 24)

In this milestone we will give an overview of several experimental activities aiming to optimize CB performance. Unlike in previous sections, here we will refer to both types of charge breeders, both ECR- and EBIS-based. While in the longer term more and more facilities tend to favour EBIS-CB in order to achieve higher charge states and cleaner beams, ECR-CB will remain important particularly if the beam contamination issue is addressed.

We start with research activities aiming to improve the performance of an ECR-based CB at HIL (Warsaw). In this milestone we concentrate on numerical modelling of efficient 1+ beam injection into an existing ECR ion source. The simulation results will be used to design and build a 1+ injection system, where one has the possibility to control the energy of the injected 1+ beam. The injection system will be terminated with a deflection electrode which allows us to change the angle of the injected beam respect to the axis of the plasma chamber, i. e. to the magnetic field lines of the ion trap. Due to the angle the ion path will be increased through the ECR plasma and consequently the ions will spend more time inside the hot electron plasma more time during the injection phase.

Independently of the injection angle, change should also be able to tune the energy of the injected



beam by changing 1+ ion source potential.

Figure 3. Principal scheme of the injection with deflecting electrode.

The injection system will be mounted to the RF coupler tube, which is axially movable so its position with respect to the plasma chamber can be changed in the range of 25 mm.

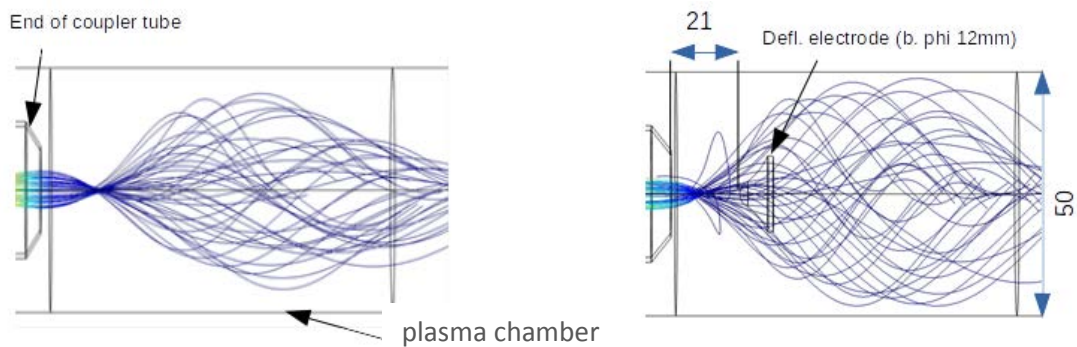
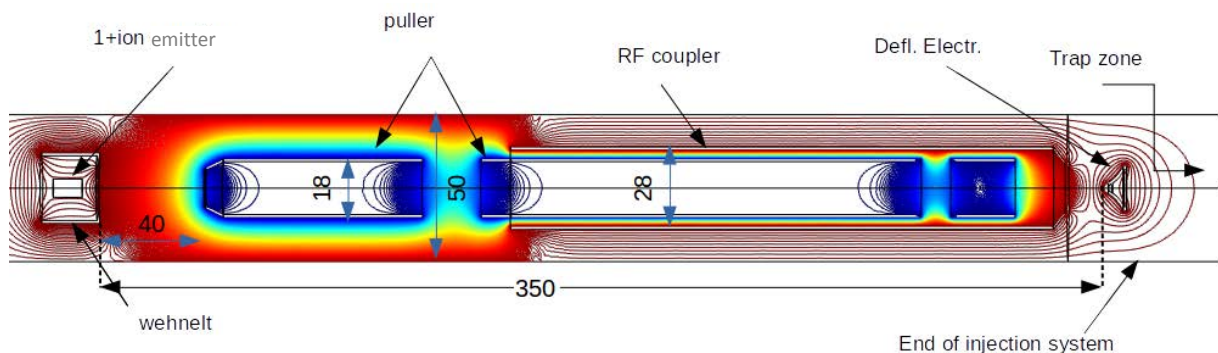


Figure 4. Trajectories of Ca^{+} ions without (left) and with (right) deflecting electrode at the end of the injection system (dimensions in mm).

The simulations describe so far the transport through the injection system, deceleration and entering into the plasma chamber, while the capture process is not yet included. The magnetic field input for



the calculation was taken from measurements on the ECRIS.

Figure 5. Equipotential lines for the 1+ ion injection system mounted in the extension of plasma chamber. The two gaps in the puller (extraction electrode from 1+ source) give better optical condition for the incoming beam (dimensions in mm).

The simulations were performed with dimension set to match the already existing ECRIS system. The deflecting electrode and beam outlet are placed just inside of the first magnetic field maxima of the trapping structure. A few test simulation runs were performed for the same injection potential setup but for different ions, see Fig. 6.

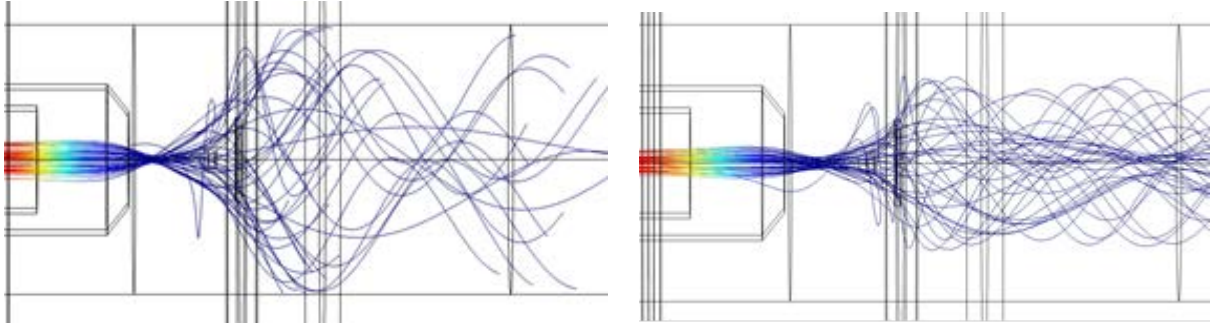


Figure 6. Beam of Al^{1+} with injection energy 150 eV and deflector potential +80 V (left). Beam of B^{1+} with injection energy 150 eV and deflector potential +80 V (right).

Lighter ions have a higher speed than heavier and their trajectories are less dispersed. By simultaneously adapting the position of the deflecting electrode with respect to magnetic trap and the 1^+ beam energy, we expect to be able to minimize the fraction of the beam that strikes the surface of the deflector.

Let us continue with the performance optimization experiments carried out on EBIS-based charge breeders. The most important advances here are related to the equipment preparation phase for upcoming tests and commissioning of new EBIS-prototype devices. After the cooperation program with BNL was concluded, development of new EBIS charge breeders faced a serious challenge as there was no full-scale EBIS test stand available to experimentally test new beam-optics concepts. Additionally, the high-compression electron-beam design under commissioning at MSU-NSCL was abandoned for a less risky immersed optics option. The lack of research infrastructure for R&D on high-compression EBIS optics became the main concern for the ICBT task. Thus, in parallel to other activities, an offline EBIS test-stand, the so-called TwinEBIS was re-commissioned at CERN [9]. The name Twin reflects on the fact that the core of the machine is a replica of the on-line charge breeder REXEBIS at the ISOLDE facility. By the time of this report being written the TwinEBIS is being prepared for ion extraction.

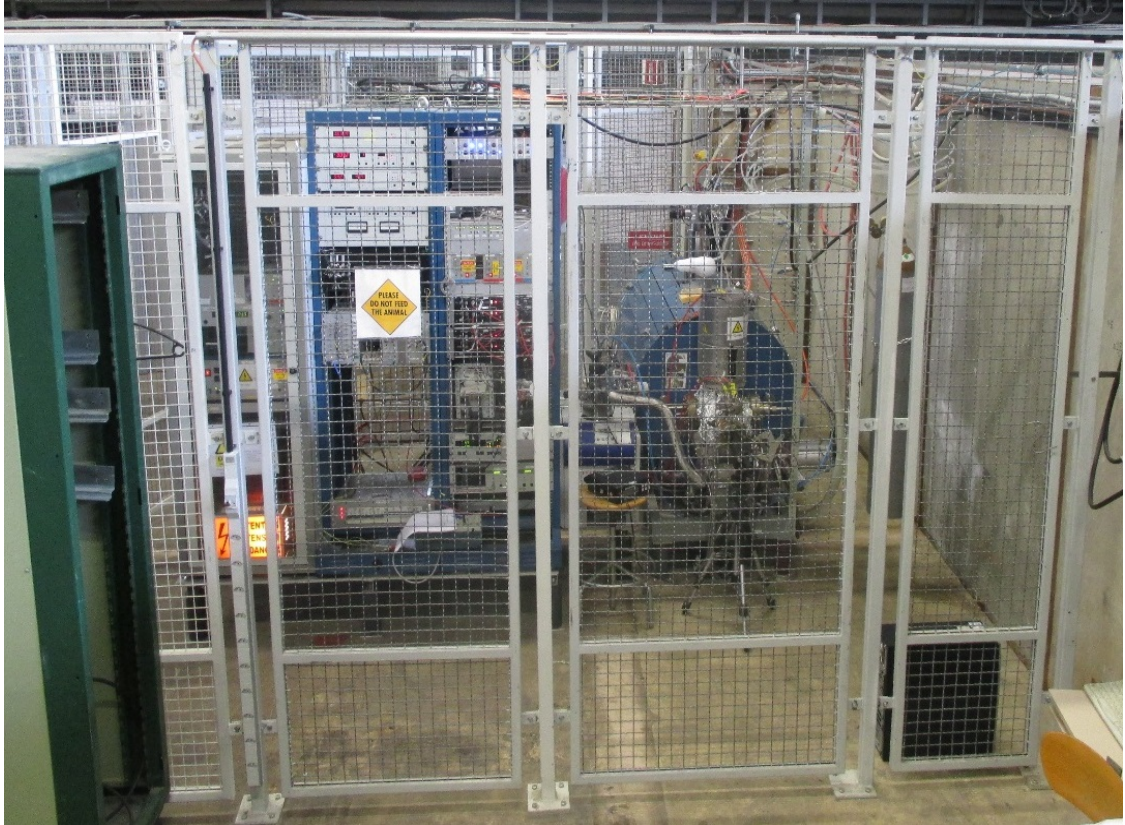


Figure 7. Re-commissioned TwinEBIS and related equipment installed in a dedicated lab at CERN.

As both TestEBIS used for HEC2 and TwinEBIS have single main solenoids which do not permit to view the fully compressed electron-beam from the side, the only experimental method to measure the electron beam current density is to perform controlled injection, charge breeding extraction and analysis of the charge state distribution in the extracted beam. For this purpose already at the time of the HEC2 program the construction of the analytical Reflection type Time of Flight Mass Spectrometer (ToF MS) was started. While this device had not been finished before the end of the HEC2 program, the said ToF MS is now being prepared to assess performance of MEDeGUN and provide solid experimental data concerning electron current density and ion production.

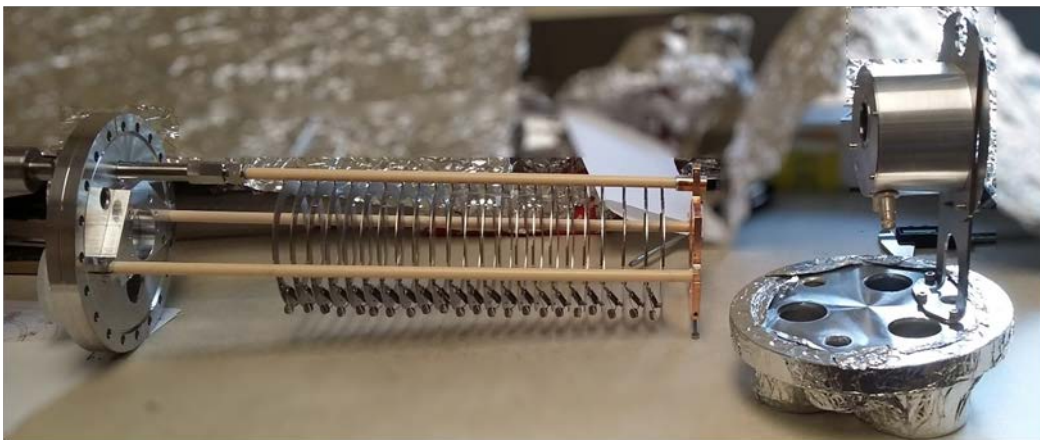


Figure 8. ToF MS internal structure and fast beam chopper prepared for installation.

Due to the potential risk of ion-heating caused by inelastic collisions and/or collective plasma effects there is a need to assess the emittance of the extracted ion beam, and if needed be prepared to proceed with the R&D on ion-cooling. While the ion-cooling technique is known in general, it was not used before for similar high intensities and very good vacuum conditions. Therefore, it is important to identify at the earliest stage whether the transverse beam emittance is of concern with the new high-compression optics. In order to measure the emittance of the ion-beam extracted from the CB the construction of a pepperpot-type emittance meter (PPEM) was started within the HEC2 program. After some mechanical adjustments and improvements of the control software, the emittance meter has now been finished and tested within ICBT on the REXEBIS charge breeder. It will be installed at TwinEBIS when an extraction line is available.



Figure 9. Assembled PPEM (left) and PPEM in the vacuum chamber prepared for tests (right).

Conclusion

The results presented in this report cover the first 15 months of the ICBT task. In spite of none of the deliverables being due, we can nevertheless present significant advances, which is also reflected in the first published paper within ICBT [9].

The debuncher part of the project achieved the first operational results and is being prepared for optimization tests. The EBIS CB part has addressed the electron beam launching, which is the most critical risk for future EBIS upgrades. Thanks to early planning the access to R&D infrastructure for EBIS CB is guaranteed without relying on partners outside of ICBT (such as it used to be with BNL).

Moreover, the HEC2 program demonstrated performance figures close to the requirements of the next generation EBIS CB. With MEDeGUN built and awaiting tests we can take full advantage of research infrastructure created in several projects. TwinEBIS, MEDeGUN and diagnostic equipment based on the HEC2 legacy are all ready for operation and using them together during the remaining 24 months period allows to perform in-depth study of the EBIS high-compression optics and outline a safe prototype-based conceptual design of the future EBIS CB. Regarding the optimization of ECR CB at HIL the numerical calculations needed to develop the final design of the test equipment are in progress.

II BEAMLAB –TASK02

Scientific Coordination: M. Cheikh-Mhamed (CNRS – IPNO)

Subtask #2.1: EFFICIENT ION SOURCES FOR DIFFICULT ISOL BEAMS

INVOLVED LABORATORIES: CERN, LNL-INFN, IPNO, GANIL

The LNL-INFN contribution for subtask 2.1 is planned between 2018 and 2019. Most of the activities will be focused on the optimization of the SPES Plasma Ion Source (FEBIAD ion source based on the ISOLDE MK5 design), concentrating on its thermal-structural behavior, on thermionic emission and beam extraction. The aforementioned work will be developed making use of both numerical codes and dedicated experimental tests at the SPES off-line front-end. The final version of the SPES Plasma Ion Source will be opportunely characterized in terms of ionization efficiency and transversal emittance.

The proposed work by IPNO will be in relation to the optimization of the IRENA ion source prototype. These optimization studies will be achieved, mainly, by computer simulation with the Lorentz-3EM code. Operating parameters such as Thermionic emission and beam extraction will be optimized and the final optimized design will be qualified by ionization efficiency measurements at an off-line mass separator. The realization of this task is planned for 2018 and 2019 with an estimated consumable costs of 25 K€ to achieve all the required developments and characterization measurements. The BeamLab Post-doc works will be shared with this work.

At GANIL, mainly two types of ion sources are available: the Nanogan III Electron Cyclotron Resonance Ion Source (ECRIS) and the VADIS Forced Electron Beam Induced Arc Discharge (FEBIAD) source adapted from ISOLDE to be coupled to the SPIRAL 1 targets. While the first one mainly produces gases, the other one should produce mainly metallic elements. The FEBIAD source has been commissioned on-line in 2013 producing numerous radioactive isotopes of different metallic elements. The use of the FEBIAD should be tested in 2018 on-line for the production of the reactive P and S elements. The formation of PH_x and SH_x molecules to ease the effusion towards the ion source will be investigated.

GANIL focuses its efforts towards the optimizations of the VADIS for production at SPIRAL 1. As shown in Figure 1, the FEBIAD efficiencies are different according to the version used at different places. Interestingly, the VADIS used at GANIL [11] does not reproduce in routine operation the

efficiencies measured at ISOLDE [10]. These efficiencies have only been obtained so far in a transitory regime in the early commissioning of the ion source. Investigations are going on to uncover the conditions which can permit to establish a permanent regime with the nominal efficiencies.

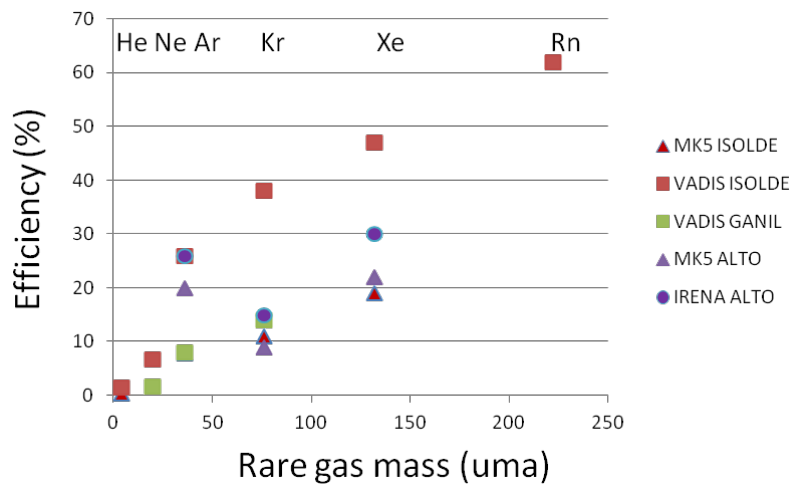


Figure 10: FEBIAD efficiencies as measured in different places [10-12].

In the FEBIAD ion source, ions are either originated from the plasma created in the anode volume, or from hot surfaces which can surface ionize alkali and alkali earth elements. The resulting energy spread can be very different. This energy spread will be measured by the fall of 2017 using a precise energy analyzer originally developed by LPC Caen for characterizing the performances of a RFQ cooler. If too large, the beam energy spread may eventually affect the SPIRAL 1 charge breeder efficiencies, whose acceptance is only of the order of a few eVs [13]. The precise measurement of the mean energy and energy spread of beams originated from the anode should additionally give some insights of the plasma characteristics and regime.

At longer term, GANIL is planning to adapt the FEBIAD source to the fusion-evaporation target (see report of subtask 2.4). Some optimizations concerning the effusion rapidity of atoms to the ion source volume will be considered for such adaptation.

At CERN, activities are focused on the optimization of the VADIS ion source for the Boron beam productions [14]. For the extraction of exotic radioactive boron beams as BF_x molecules (subtask 2.3) offline tests had to be conducted on the survivability and ionization of such molecules once formed. Thermodynamical modelling with excess F comparing to B was conducted. This was done in order to assess the reactivity of such molecules with the target/transfer line/ion source structural materials (Ta, Re, Mo and Cu) which can form stable compounds (e.g. borides). From 500 °C and 1900 °C there is no predicted reaction of Ta, Re, Mo with the BF_3 molecules. The Cu is normally operated close to room temperatures (VADIS cold line). From 2000°C the thermodynamics predicts that the BF_3 molecule converts into BF_2 . Nonetheless in environments with excess F, fluoride formation with boron is favoured instead of borides with the refractory target structural materials.

During the experimental tests done with the VADIS, BF_2^+ was seen in greater abundance than BF_3^+ as seen in Figure 2 - left. This is due to the higher dissociative ionization cross section from BF_3 to BF_2^+

than the direct ionization cross section of BF_3^+ . It was also noticed that both BF_2^+ and BF_3^+ peak at a maximum target temperature of 1500 °C, even though this wasn't predicted by the thermodynamical models (Figure 2). The BF_2^+ has been seen to increase linearly with the amount of SF_6 injected. The total efficiency (molecule formation x transport x ionization) was found to be 1.5%. With a VADIS anode scan (Figure 2 – right) it was found that, higher anode voltages brought higher ionization efficiencies for BF_2 . However, for lower anode scan voltages, the curve did not follow the normalized theoretical cross section likely due to the potential variance inside the VADIS and also due to the dissociative ionization mechanism happening at the same time.

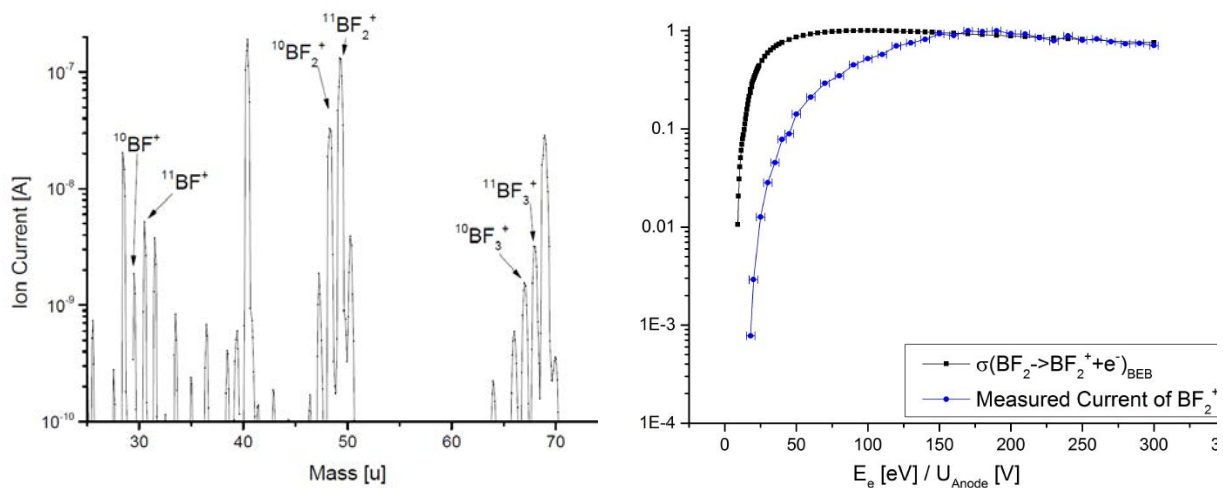


Figure 11: Left - Mass scan showing the peaks of boron fluorides molecular ions from a VADIS ion source. Right – Comparison of normalized ion current of BF_2^+ coming from a VADIS ion source with the normalized ionization cross section of BF_2 .

Subtask #2.2 (COORDINATION: A. ANDRIGHETTO - LNL-INFN): MATERIAL COMPATIBILITY IN REACTIVE GAS ATMOSPHERES

INVOLVED LABORATORIES: GANIL, IJF-PAN, IPNO, LNL-INFN

At IPNO, the work will focus on the optimization of the thermal behavior of the transfer line device for beam production at ALTO. The optimization work will be achieved by means of thermal simulations with The ANSYS code and temperature measurements will be done on the new thermal test bunch. The design of the thermal test bunch is taken in charge by the IPNO design office and the final design will be approved in few months. The estimated global acquisition cost of such equipment will be of 30 k€. The vacuum system has already been purchased. The optimization simulation work has been already launched and a full modelization of the actual target oven, the transfer line tube and surface ionization ion source assembly was achieved (see Figure 3). Thermal studies are still going on in the frame work of a Master internship. We note the great collaboration of the LNL team in this task and future work will continue with the help of a post-doctoral fellow hired in the framework of ENSAR2.

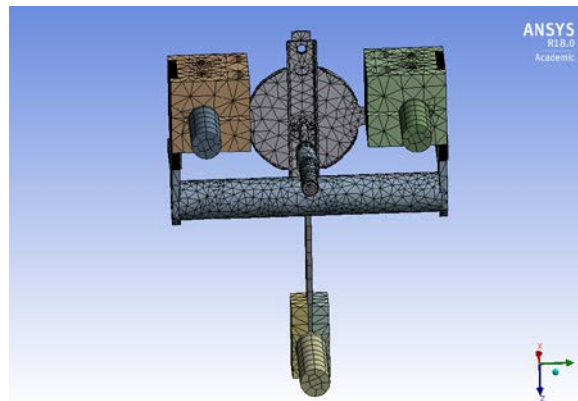


Figure 12: Fully modelization of the target oven, transfer tube and surface ionization ion source assembly with the ANSYS code

At GANIL, a setup for measuring the emissivity and electrical conductivity of thin foils of different materials is being designed. This setup should be used for characterizing the materials used for the fusion-evaporation targets at SPIRAL 1. It could additionally be used for measuring properties of oxidized material. Such study would be complementary to the contribution of IFJ-PAN for the tantalum material.

The proposed work by IFJ-PAN is in relation to the oxidation kinetics of Tantalum in oxygen at atmospheric pressure and the kinetics of oxidation were investigated by the thermogravimetry. The experimental setup has already been designed and achieved (see Figure 4). A summary of the used experimental protocol is described as the following: Tantalum plates, cut from 100 μ m Ta foil, were washed in tetrachloromethane and acetone using an ultrasonic cleaner and then were oxidized between for 5 to 60 minutes at temperatures between 788 and 988 K. A flow rate of oxygen - 3.5 mL/min was controlled by Brooks Mass Flow Meter.

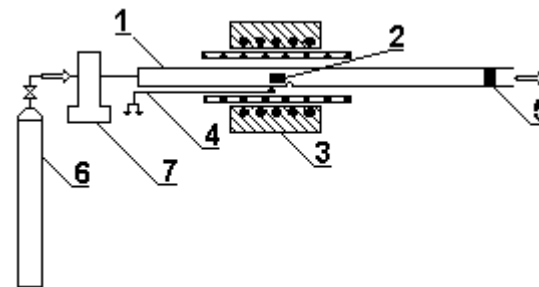


Figure 13: Apparatus for oxidation of Ta foil in atmospheric pressure: 1 - quartz column (inside diameter 11 mm); 2 - Ta plate (10x10 mm); 3- resistance oven; 4 - thermocouple; 5 - quartz plug; 6 - O₂ gas; 7- Brooks Mass Flow Meter.

Obtained results of the thermogravimetry experiments were plotted as (weight gain/area)² vs. oxidation time and presented in Figure 5.

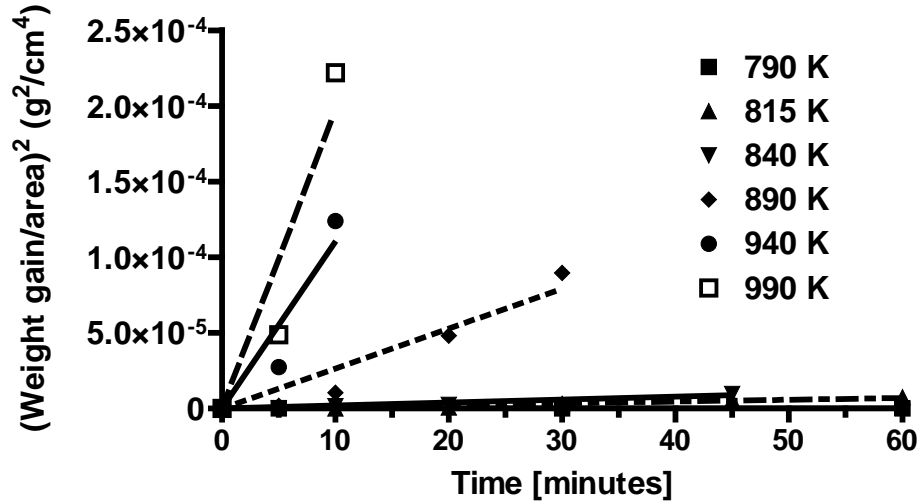


Figure 14: (Weight gain/area)² vs. oxidation time for different temperatures.

These plots show an approximately straight-line behaviour indicative of parabolic oxidation kinetics, Eq. 1:

$$\left(\frac{\Delta W}{A}\right)^2 = k_g t \quad (1)$$

Where ΔW is weight gain, A is the sample surface area, k_g is the parabolic rate constant, and t is the oxidation time. The obtained parabolic oxidation rate constants, k_g , at temperature range of 815 to 990 K were used for determination of an activation energy $-Q$ by Arrhenius equation- Eq. 2 (Figure 6):

$$k_g = k_o \exp\left(\frac{-Q}{RT}\right) \quad (2)$$

Where R is the gas constant, T is the temperature and k_o is the pre-exponential factor.

The activation energy of Ta foil for oxidation in oxygen at flow conditions is 213 kJ/mol*K over the temperature range from 815 to 990 K.

It is clearly seen on Figure 5 that the oxidation kinetics of Ta foil is faster from 890 K. Up to 800 K the oxidation of Ta foil is very slow. The activation energy describes two processes: chemical reaction on

the tantalum surface (a chemisorption process) and a diffusion of ionic oxygen through the oxide layer which will affect the oxidation kinetics.

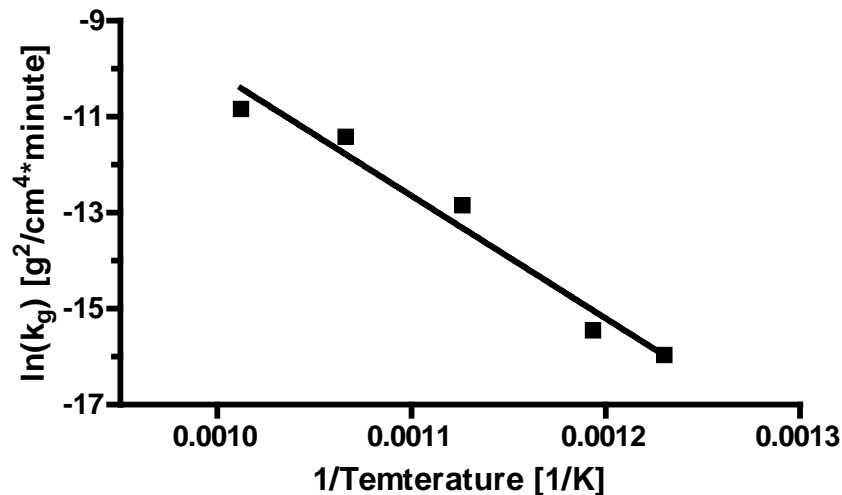


Figure 15: $\ln(\text{parabolic rate constant, } k_g) \text{ vs. } 1/\text{temperature}$ for Ta from 815 to 990 K.

Subtask #2.3 (COORDINATION: T. STORA-CERN): NEW MOLECULAR BEAMS

INVOLVED LABORATORIES: CERN, GANIL, LNL-INFN, IPNO

[Exotic Boron beams and refractory metal carbonyl beams at ISOLDE](#)

Despite the manifold new developments introduced to ISOL target units within the last 60 years, the beam extraction of elements with very high boiling points (refractory elements) remains still among the most challenging topics. Due to their vanishingly low volatility, the radionuclides generated by the driver beam are captured within the target and suffer from hampered release.

Boron is one of these elements, which is not only refractory, but also forms stable bonds with various materials the target unit is made of. Motivated by the strong interest in ^8B beams we developed and tested a target unit, which allows the extraction of ^8B by in-situ volatilization of boron as fluoride. Sulfur hexafluoride served as fluorinating agent and was injected through a calibrated gas leak into the target container filled with pressed pills of multiwall carbon nanotubes. The latter were chosen as target material due to their favorable diffusion behavior, which was investigated in preparatory experiments. Non-volatile reaction products were trapped in the water cooled transfer line, which connects target container and ion source.

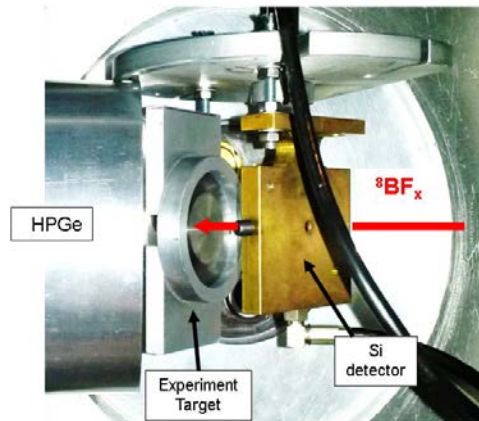


Figure 16: In-Beam detection setup used for ^8B .

Online tests of the prototype unit were conducted in 2014, 2015 and 2017. In addition to the ISOLDE tape station, which is equipped with a plastic scintillator and a high-purity germanium detector, an in-beam detection setup was used. It consisted of a silicon and a high-purity germanium detector, capable of detecting alpha particles in coincidence with gamma radiation and therefore tailored to the $\alpha 2\beta^+$ decay mode of ^8B .

As predicted, we were able to measure activity having the signature of ^8B on the corresponding masses of $^8\text{BF}_x$, where $x = 0$ to 3. In agreement with offline tests, the highest yield was found on the mass of BF_2 . In 2017 intense boron beams have been delivered for the first time to a physics experiment, in which the electron capture of ^8B into highly excited states in ^8Be was studied at the ISOLDE decay station (IS633).

Another group of elements, which is so far difficult to extract by the ISOL-technique, is the group of refractory transition metals. We propose here an advanced target design for ISOL facilities, in which diffusion through condensed matter can be fully avoided. Instead of diffusion, we make use of the recoil momentum, which allows fission fragments to propagate through and emerge from thin uranium foils. The recoils are thermalized in a reactive gas and form volatile compounds *in-situ*. Subsequently, the reactive gas is removed by cryogenic gas separation, as shown in Figure 8. The system is evacuated while the carbonyl compounds are retained on a cooling trap. After removal of the excess gas, the volatile compounds are fed into the ion source by warming up the cooling trap. A compound class that appears well suited for *in-situ* volatilization is that of metal carbonyl complexes.

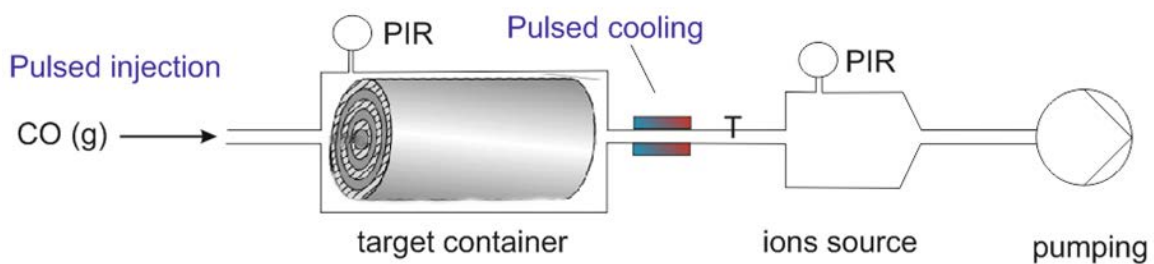


Figure 17: Gas separation concept

1																	2
H																	He
3	4											5	6	7	8	9	10
Li	Be											B	C	N	O	F	Ne
11	12											13	14	15	16	17	18
Na	Mg											Al	Si	P	S	Cl	Ar
19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
55	56	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
Cs	Ba	La...	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn

Figure 18 - Periodic Table of elements showing available beams at ISOLDE (green), Non-available Elements, which form transition metal carbonyls (red background) and carbonyl chalcogenides (red frame).

Already in 1970, the extraction using carbonyl complexes was proposed and recently, it was shown that volatile carbonyl complexes form readily at ambient temperature and pressure by thermalizing fission fragments of suitable elements in a carbon monoxide-containing atmosphere. The ISOLDE Periodic Table of Elements shown in Figure 9 demonstrates the potential of the “carbonyl method”. Nine out of fifteen transition metals, which are not yet available, form volatile carbonyl compounds.

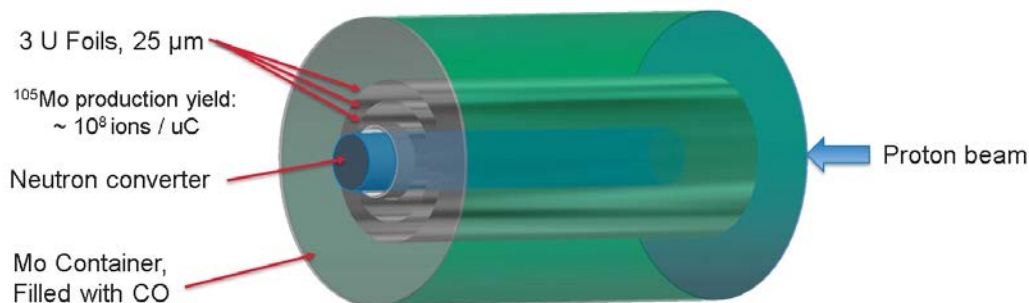


Figure 19 - Design of the model target

While carbonyl compounds open up new perspectives in the development of refractory beams, they are delicate compounds. In contrast to the relatively stable BF₃ compounds, the organometallic carbonyl compounds decompose on hot surfaces, by electron beam impact, in plasmas and upon exposure to UV-light, which needs to be considered not only in the choice of the ion source, but also in the target design. The intense proton driver beam at ISOLDE is expected to induce a plasma in the gas filled target container. The direct interaction of the charged beam with the reactive gas atmosphere should therefore be avoided. While a proton to neutron converter is readily available at ISOLDE, it needs to be further improved to reduce proton spill and the accompanying formation of a beam induced plasma. See below for a discussion of the proton-to-neutron converter project.

Simulations have been conducted aiming at the estimation of attainable yields (design of the model target shown in Figure 10). Several different ion sources were tested, including a FEBIAD-type VADIS source and cold plasma RF ion sources. Finally, the laser-induced break-up of carbonyl complexes as essential preparatory step to resonant laser ionization was also experimentally addressed.

For fluorinated Lanthanides beams, IPNO has performed the first systematic study on fluorination of lanthanides in surface ionization (SI) and FEBIAD (MK5) ion sources. The off-line tests carried out using graphite pellets doped with stable lanthanides showed that adding CF₄ in the target and ion source system increases the Ln release by a factor of 10 to 100 and that most of the lanthanides are preferentially observed as di-fluorinated ions except Sm, Eu and Yb that appear as mono-fluorinated ions. These features are more pronounced with the MK5 ion source than with the SI one, which seems to indicate that the processes involved in fluorination differ between the two ion sources and are simpler within the MK5 ion source. These measurements showed that the release properties of lanthanides from thick uranium carbide pellets are similar to that obtained from graphite pellets. With the SI ion source the Ln ionization efficiency is found to be higher by a factor of 50 to 100 compared to that obtained with the MK5 ion source (see Figure 11). The off-line tests and the first on-line experiments allowed us to determine the experimental requirements for producing lanthanide radioactive isotopes: the target temperature is really a key parameter and has to be as high as possible ($T_{\text{target}} \geq 2050$ °C); moreover we have to use the SI (Surface Ionization) ion source in order to achieve maximum efficiency. Before performing a new on-line fluorination experiment, it remains to improve the gas-inlet system using a well-chosen fixed calibrated leak in order to better control the injected gas flow into the TIS. The future tests are scheduled for 2018 with costs of consumable and the upgrade of the gas system estimated at 18 K€. This work is done with the collaboration of the IPNO nuclear physics group “Nester”.

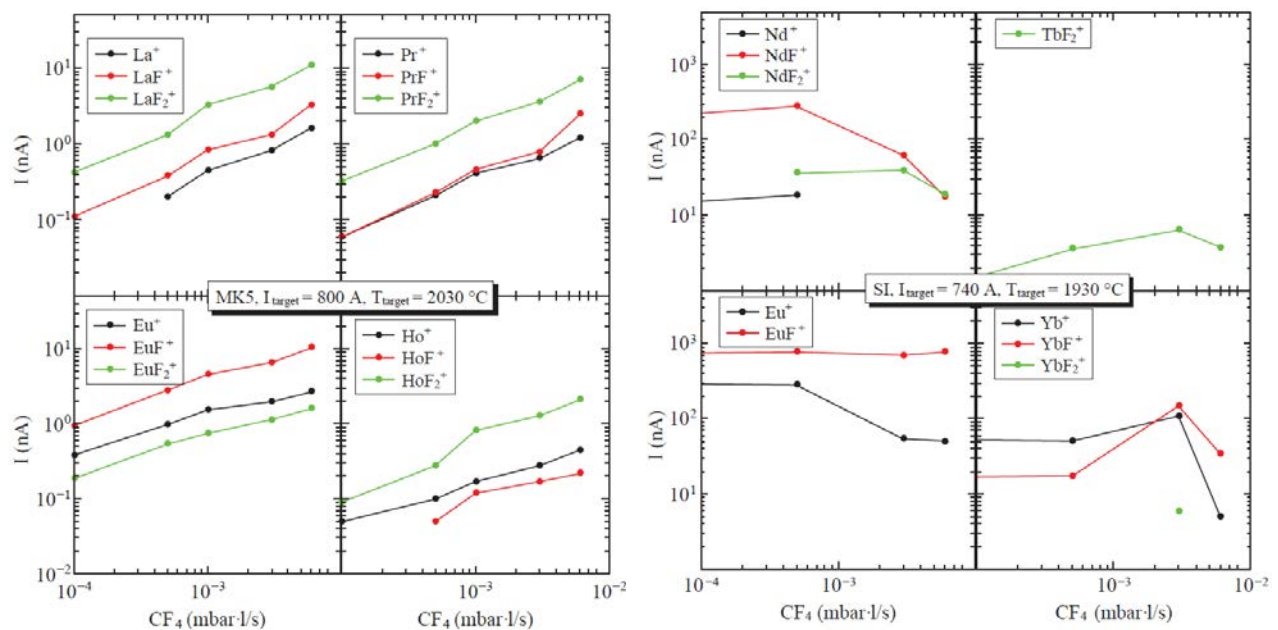


Figure 20: Ion beam intensity measured during the off-line tests as a function of the quantity of gas injected for various lanthanides in their elemental and molecular forms using MK5 or SI ion source.

At LNL-INFN, and taking as a reference the techniques developed at ORNL [15], the LNL-SPES group is preparing the upgrade of all the Plasma Ion Source auxiliary components required for the production of pure beams of germanium and tin isotopes starting from specific molecular beams (SnS^+ and GeS^+). In particular, the first tests will be done combining Sn with S powder, both inserted in a dedicated tubular oven (first samples ready to be tested and illustrated in the Figure 12). Then, in a second step, S will be introduced in a gaseous form (SF_6 and/or H_2S) inside the ion source. The LNL-INFN contribution for this subtask 2.3 is planned between 2018 and 2019.

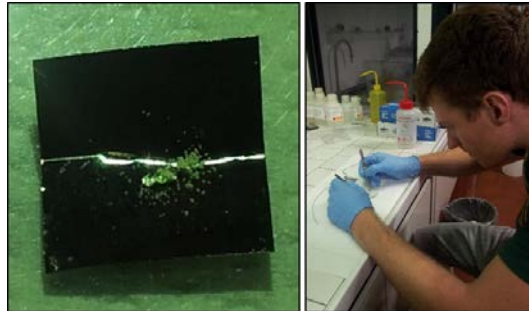


Figure 21: First Sn/S samples preparation at LNL-INFN

For GANIL, the SPIRAL 1 upgrade commissioning is presently going with stable beam. A radioactive beam commissioning period is foreseen in the first half of 2018, during which tests will be conducted in order to produce S and P beams in a molecular form. Such beams have been requested by the GANIL community for different astrophysics experiments. A calibrated leak of CH_4 will be introduced into the target volume in order to attempt producing SH_x and PH_x molecules, which were already observed using different ECR ion sources at GANIL [16,17].

Subtask #2.4 (COORDINATION: M. CHEIKH-MHAMED-IPNO): SPECIFIC TARGETS DESIGNS FOR NON-VOLATILE ELEMENTS

INVOLVED LABORATORIES: CERN, GANIL, LNL-INFN, IPNO

The proposed work by IPNO was to optimize the photofission target of the ALTO Facility. This work is intended to optimize the geometry shape of the UCx target design and its tantalum oven for the beam production at ALTO. The new design should enhance the final beam intensities produced at ALTO and reduces the nuclear wastes. The optimization of the geometry with the FLUKA MC code has already begun and the first preliminary results with a shorter geometry show that the total fission rate is preserved (see Figure 13). Nevertheless, this work continues to improve this solution. A second part of this work will consist in optimizing the homogeneity of the temperature distribution in the tantalum furnace. Temperature measurements will be carried out with the thermal bench once the thermal optimization simulations have been completed. On-line production measurements will be achieved once the new design has been finalized. This work will be carried out by a post doctoral fellow and planned to be achieved by 2019.

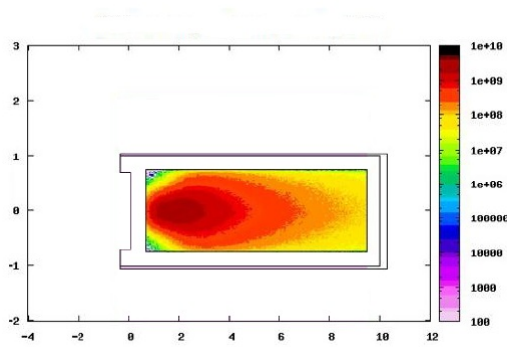


Figure 22: Fission Yield distribution with an UCx target of 88 mm of length (preliminary results)

At LNL-INFN, The LNL-SPES group is planning to complete within the first half of 2019 the design and the high temperature testing of the target that will be used for the first radioactive ion beam production at SPES. MonteCarlo simulations (FLUKA code) for production and power deposition will be performed, together with detailed coupled field electrical-thermal-structural analyses (ANSYS code). The aforementioned high temperature testing will investigate the behavior of the target and of all the temperature sensors that will be used for the machine protection system.

At GANIL, works started by investigating the use of two different types of new targets for enlarging the production of non-volatile elements at SPIRAL1: this concerns the use of new fragmentation targets such as Nb, and thin targets for fusion – evaporation such as Ni. The Nb target development will first consist in testing different configurations of foils stacking that will be heated to 2000°C (see Figure 14). The resistance of the stackings to sintering will be studied over time in a heating test stand in 2018. In such metal foils targets, the sintering is a major cause of losses due to long diffusion of the reaction products in the bulk material.

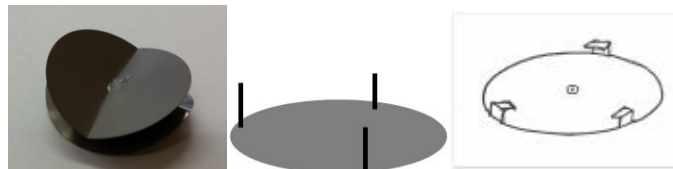


Figure 23: different stacking configurations for the Nb foils.

GANIL is additionally designing a new target-ion source for the production of radioactive beams through fusion evaporation reactions. In such target ion source (see Figure 15), the reaction of fusion – evaporation occurs in a thin target and the reaction products are stopped in a graphite catcher. Such a configuration (thin target and implantation of reaction products close to the surface of the catcher) is expected to favour the short diffusion of the reaction products, thus allowing the production of difficult ISOL beams (short half-lives and/or refractory isotopes). Some of the properties of the thin targets will be measured (thermal emissivity and electrical conductivity) on a dedicated test bench (see report for subtask 2.2). The off-line tests of the target – ion source should start in 2018.

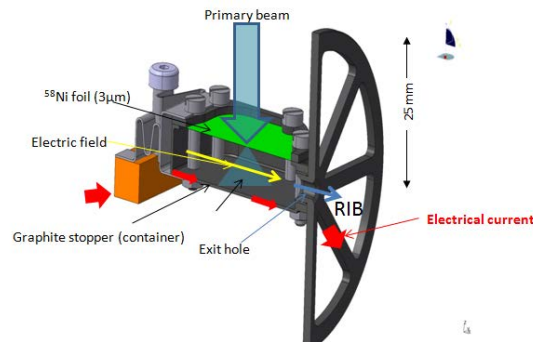


Figure 24: schematic view of the fusion – evaporation target assembly. From the PhD of V. Kuchi.

The proton to neutron converter at ISOLDE

The proton to neutron converter (p2n-converter) has been used at ISOLDE for many years to produce pure neutron rich fissions fragments when comparing with proton beam on the UCx target. This proton to neutron converter is a tungsten bar situated below the target, which acts as a spallation source when bombarded with the proton beam. The current p2n-converter design can be optimized to produce less neutron deficient isobars coming from the scattered protons hitting the target. It can also use a more efficient geometry to use the full solid angle of the isotropically emitted neutrons (e.g. use a converter-surrounding target). These optimizations would bring enhanced isotope production and beam purity, for standard UCx targets at ISOLDE and other facilities. Furthermore, the improved geometry p2n-converter is essential for the refractory carbonyl isotope production from a cold target (as detailed before).

A collaboration was started between TRIUMF, SCK-CEN and CERN-ISOLDE to develop an improved p2n-converter for TRIUMF and ISOLDE. While TRIUMF has a 500 MeV-100 μ A c.w. beam of 50 kW, ISOLDE has a 1.4 GeV – 2 μ A pulsed beam of 2.8 kW (1.2 GW of instantaneous power deposition). Due to the intrinsic difference between the driver beams, this leads to different concepts being developed for the two facilities. FLUKA was used to simulate the concept neutronics and ANSYS was used to simulate thermo-mechanical and thermo-electrical aspects of the target-converter-cooling block assembly.

In the last 6 months mainly the TRIUMF p2n-converter concept was addressed, where each institute involved in the collaboration has proposed a different concept and studied different issues (neutronics, cooling, target heating). An example of such concepts, the one developed by CERN, can be seen on Figure 16.

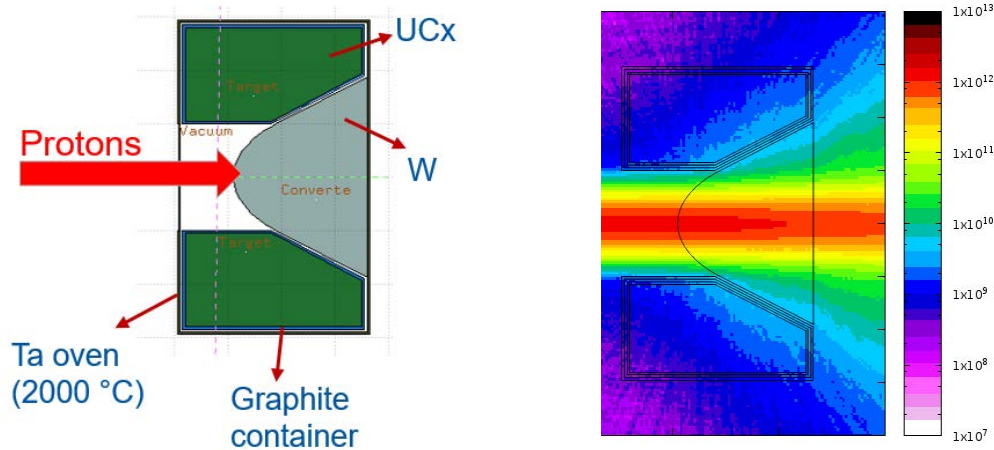


Figure 25: On the left, one of the p2nconverter concepts developed by CERN, where the converter and the target are coupled together. Each square is 1 cm. On the right, proton fluences in this concept (unit is in $/\mu\text{C}$).

The main features/requirements of the new concepts are:

- (i) having a circular target, surrounding the W converter, which is as close as possible to the W converter which has a central hole to avoid proton induced nuclear reactions;
- (ii) reduce the aspect ratio (standard 20:2) of the target to catch the zones of high neutron fluences;
- (iii) the converter has to be shifted relatively to the target downstream to the beam so scattered protons hitting the target are minimized;
- (iv) a very efficiency cooling solution since about 8 kW of power are deposited in the 2 cm converter and W is an inefficient conductor at high temperatures.

A simplified design of the converter, shown in Figure 16, has been selected to facilitate the experimental tests to come. This simplified design has the converter uncoupled from the target oven and shifted upstream to the beam. This brings a lower neutron induced fission rate but allows to uncouple the target heating from the converter heating – simplifying the cooling solution. The current neutron induced fissions are in the range of $1\text{E}11$ to $1\text{E}12$ fissions/s with about 7 to 15% of proton induced fissions from the total fissions. This range depends on the target aspect ratio and proximity of the target to the converter. There is no fixed geometry for the target yet, since it has still to be tested if 2000°C can be achieved in a large diameter target with the power supplied currently present at TRIUMF.

III CRIBE – Task 3 of JRA Eurisol (WP14)

Scientific Coordination: Manssour Fadil (GANIL)

A. General overview

The EURISOL JRA Task CRIBE is dedicated to the development of a tool (called in the following CRIBE) aiming for a presentation of the main characteristics of Radioactive Ion Beams (RIB) produced

in major European ISOL facilities. These characteristics will be mainly the nature of the produced isotope, its acceleration and/or pre-acceleration energy, the RIB intensity as well as purity when it is available.

Until now, three facilities have clearly shown their will to collaborate and to contribute to this work and their intention to publish their data on CRIBE: GANIL (SPIRAL1 and S3), ISOLDE and SPES.

The work on CRIBE is divided in two main endeavours:

1. the first deals with the technical development of the chart of beams. In this part, the mode of the visualization of the data, of their presentation, of the way of the uploading and downloading the data will be defined. It consists in an important amount of programming work.
2. The second deals with the data itself. In this part, the choice of the parameters characterizing RIBs, the format of the presentation and a compilation of the data have to be specified.

During the reporting period, the work on both chapters has started. A special meeting with some partners during EURISOL DF 2016 conference in Leuven (October, 2016) allowed to discuss about the data of CRIBE. Each facility defined its representative who will be in charge to provide the data to the developing team and to validate the chart before publication. An organizational structure of the task was submitted to the collaboration in April, 2016 (see figure 1).

Two students were hired to work in 2017 on CRIBE in GANIL. The first, Paul Jourdan from Normandy University, worked on technical specification of the chart of beams. The second, Mateusz Celary, in collaboration with IFJ-PAN Krakow, worked on the RIB data. Pierre-Marie Briéda, a student from Normandy University, contributed also to the summary of the specifications.

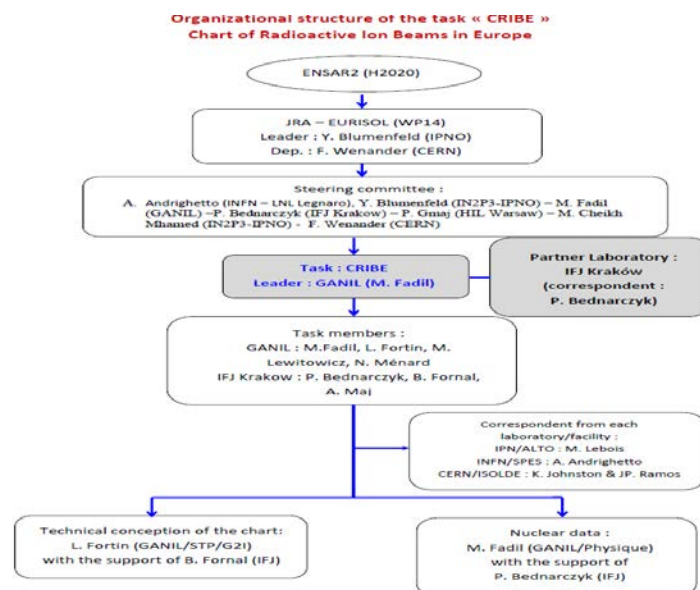


Figure 1: Organizational structure of CRIBE task submitted to and validated by collaboration

B. Chart developing

The technical specifications of the chart were worked out during the spring 2017. These specifications define the guidelines of three facets of the chart:

1. **Functionality:** the chart has to be interactive and dynamic. This interactivity will allow distinguishing the already available beams from those that are expected in the future. It will also allow visualizing the RIB data by clicking on any isotope. Thank to this interactivity one can search for any beams of interest considering one or more criteria: production facility, energy, intensity and availability.
2. **Administration:** we define two levels of the chart administrators: a principal and a secondary one. The principal administrator will have rights to modify the chart (adding or removing isotopes, elements, creation of administrator account...). The secondary administrator, appointed at each involved facility, will have rights to add, to remove or to modify the local RIB data.
3. **Presentation:** two ideas of the chart presentation were proposed: Z versus N presentation (Karlsruhe nuclides chart like) or Mendeleev table (table of elements).

C. RIB data format

In order to begin the process of collecting RIB data from the European facilities, analysis of RIB nuclear data for each facility was carried out. It considered two main contexts: data in the view of different production techniques at different facilities and data seen from the users point of view.

It resulted in two proposals of formats of presenting RIB data to users of CRIBE, among which the best one was chosen and is presented below. It will be discussed with representatives of facilities in the CRIBE task.

1. Data analysis summary

We considered the data published by the following facilities:

Facility:	Public RIB database (later called "table"):
GANIL, France	http://u.ganil-spiral2.eu/chartbeams/
ISOLDE, Germany	http://test-isolde-yields.web.cern.ch/test-isolde-yields/query_tgt.htm
SPES, Italy	https://web.infn.it/spes/index.php/characteristics/spes-beams-7037/spesbeamstable
ALTO, France	http://ipnwww.in2p3.fr/lons-disponibles,687

Also additional information, from e-mail exchanges about new planned ISOLDE tables, as well as different discussions with physicists and engineers of GANIL were considered.

1.1. Manner of presenting the data in tables from different facilities

Facilities show RIB data in tables with additional comments. GANIL/SPIRAL1 and ISOLDE data are shown as different lines or "records" for each primary beam – target material – ion source

configuration. For SPIRAL1 also data for diverse primary beam energies, primary and secondary charge states are given. Beam data in SPES tables are presented in a similar manner, but not distinguishing intensity values obtained for different ion sources and targets.

It is proposed for each isotope to minimize number of beam records given by each facility in the CRIBE database. This minimization could follow these priorities:

1. Prefer measured data over calculations,
2. For calculations prefer soonest available beams (related to the target, ion-source and primary beam developments),
3. For many available records for a given isotope take the beam with the highest intensity.

It is proposed, in some cases (e.g. measurements done for not the most abundant charge states and not optimal primary beam energy and power) to give among measured value also calculated expectation.

1.2. Beam parameters shown in tables

Radioactive beams and their production can be described by a set of parameters. Table 1 presents information about every beam parameter for each facility.

1.3. Origin of data in tables and beam availability

Yields for GANIL/S3 and SPES are results of calculations or extrapolation based on measured cross sections. ISOLDE provides pre-acceleration data from two sources: for some isotopes from measurements done on currently working (1.4 GeV) accelerator or from previous (0.6 GeV) one, the latest treated only as an estimate. ISOLDE post-acceleration yields can be estimated by user, with a recipe available on the REX-ISOLDE website. For GANIL/SPIRAL1 for most of the elements data come from calculations, for others from experiments and extrapolations.

Due to constant development of RIBs facilities, a year of availability of beams after different facility upgrade stages was considered.

		GANIL/S3	ISOLDE	ALTO	SPES	GANIL/SPIRAL 1	
PRIMARY BEAM	isotop	28Si	proton	electron	proton	86Kr	
	current	2E+14 pps / 4,4 kW	2 uA	10 uA	5 uA	0,8 kW	
	energy	9,3 MeV/u	1,4 GeV	50 MeV	40 MeV	57,9 MeV/u	
PRODUCTION	target material	40Ca	U Carbide, UC2.201	UCx	UCx	Carbon	
	ion source	-	RILIS	FEBIAD/SIS/LIS	FEBIAD	-	
SECONDARY BEAM	element	65Se	124Cd	79Se	79Se	79Se	
		half-life	33ms	1.25 s	-	3.57E+13 s	295 ky
	PRE-ACC	intensity	MIN s: 1,3E-3 pps AVE s: 2,7 E-3 pps MAX s: 5,7E-3 pps	AVE for mat: 7.7 E+6 ions/C	CUM 1,3E+5 pps	4,1E+5 pps	1E+5 pps
		energy	(?)10 - 30 keV	30 - 60 keV	30 keV	20 - 40 keV	10 - 24 keV
		charge	always +1	always +1	always +1	always +1	possible +n / +1
	POST-ACC	purity	-	-	-	given as a commentary	given as a commentary
		intensity	no postacc	another data: intruction for calculating yield estimate given	no postacc	for q: 8,2E+3 pps	for q: 5,7 E+6 pps
		energy				12 MeV/u	MIN: 1,2 MeV/u MAX: 8,4 MeV/u
		charge				most abundant +15	most abundant +14
	Other parameters provided by facilities:		-	efficiency 10% temperature of target target thickness (50g/cm ²) temperature of source n-converter usage laser usage release info(rise, fall etc) transferline (hot,warm)	eff. of ionisation % temperature 2000° nr of fissions/s 1E+11 diffusion/desorption exit time	temperature 2000° number fissions/s 1E+13 spectrometer A/Q (transferline compatibility)	-

Table 1. Beam parameters comparison. Black color indicates the information taken from the facility tables, red color is information coming from the references [18], [19], [20], [21].

2. Proposed format of presenting the data

After analysis, values describing beams which seem to be the most important to users and facilities were chosen (described below). An additional parameter – “beam purity” was proposed to be added. It resulted in data format proposition shown in Table 2.

isotope	half-life	Pre-accelerated RIB		Post-accelerated RIB				Production		Beam Availability	Facility	
		intensity	purity min %	intensity	purity min %	charge state	energy MeV/A		target material			primary beam
							min	max				
72Kr	17s	2E+4 ions/uC	-	-	-	-	-	-	Y2O3	proton	now	ISOLDE
72Kr	17s	# 2E+2 pps	-	4E+1 pps	-	+11	1,8	6,3	Carbon	78Kr	2018	GANIL/SP1
72Kr	17s	1,7E+3 pps	-	2,8E+1 pps	-	+14	1,2	10,1	Carbon	78Kr	2018	GANIL/SP1

2E+2 pps measured value	#	Measurement was done for non-optimal primary beam energy and/or power.
2E+2 pps extrapolation from preacc mesaurement		Possible intensity optimisation. In case of ISOLDE data taken from previous SC accelerator, and available intensity value can vary.
2E+2 pps calculated value		
-		No data yet.
Energy, for a fixed charge state, can be adjusted within a given range without intensity lowering. Wider energy range is possible with intensity lowering.		
For more information about facilities, and beam production: see additional description.		
For questions, and submitting beam proposals: contact representatives.		

Table 2. Table of parameters. Example of 72Kr isotope. Each line shows one radioactive beam, for one production method (primary beam, target etc.) which can be extracted before or after acceleration.

PRE-ACCELERATED	
Intensity	RIB intensity after the first ion source. This value can be considered as final after losses in the transmission line are taken into account. Should be decided among facilities if it's measured in the same way.
Purity	Beam purity defined as a ratio of the intensity of the given RIB to the intensity of

	all transmitted simultaneously isotopes. We propose that facilities provide lower limit of purity.
POST-ACCELERATED	
Intensity	RIB intensity after post acceleration. To discuss if it is better to leave ISOLDE ions/ μC units, what would make comparison more difficult, or recalculate accordingly to effective primary beam current to pps. This operation could be later reversed by a user, and info about time structure of beams would be placed in the description of tables.
Charge state	It is a usual way to include charge state, to give a user knowledge, that intensity depends on it (one charge state can be more abundant than another), as well as energy.
Energy min, max	Range of possible post-acc energies is given, for fixed charge state energy can be adjusted in accelerator without changing beam intensity.
PRODUCTION	
Primary beam nature	At GANIL different beams are used, and sometimes they are very expensive.
Ion source	While in ISOLDE proposal form it is required to give information about ion source, we propose not to give this information in this table, since we prefer scientist to submit proposal after contacting representatives of chosen facility.
OTHER INFO	
Year of availability	Approximate year for future beams, for beams that are available "now" tag.
Reliability, and source of the data	We propose to add information if given intensity values come from measurement, extrapolation of experimental data, or were calculated, to give notification about reliability of data. It can be indicated for example by the color of the font.

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