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LIST OF ACRONYMS AND ABBREVIATIONS

ALTO	Accélérateur Linéaire et Tandem d'Orsay
ECR	Electron Cyclotron Resonance
FEBIAD	Forced Electron Beam Ion Arc Discharge
FEMM	Finite Element Method Magnetics
GANIL	Grand Accélérateur National d'Ions Lourds
HIE-ISOLDE	High Intensity Energy ISOLDE
IRENA	Ionization by Radial Electron Neat Adaptation
ISOL	Isotope Separation On Line
LPC Caen	Laboratoire de Physique Corpusculaire de Caen
ORNL	Oak Ridge National Laboratory
SIMION	ION and electron optics SIMulation program
SIRa	Radioactive Ion seperator
SPES	Selective Production of Exotic Species
SPIRAL	Système de Production d'Ions Radioactifs Accélérés en Ligne
TISS	Target Ion Source System
VADIS	Versatile Arc Discharge Ion Source

Executive Summary

The techniques for the production of radioactive ion beams - subject of the Beamlab Task - have been developed for more than fifty years, with the aim to improve the beam quality and intensity and therefore the access to the European facilities exploiting the Isotope Mass Separation OnLine (ISOL) method. Such a development goes hand in hand with research and development on target and ion source systems (TIS) which are at the heart of the ISOL technique. In particular, efforts have been concentrated on the improvement of the performances of the FEBIAD ion source types and the optimization of their operating parameters for the production of elements difficult to access by ISOL method. More particularly, the production of beams of refractory elements can be improved using chemical methods, such as volatile molecular formation with injection of reactive gas in the target and ion source production unit. Contemporary research in nuclear structure and nuclear physics aims at elucidating the forces and structures at play in the ever more exotic nuclei that can be produced at eg, ALTO, GANIL and HIE-ISOLDE.
A new delivery method for ^{34}S chemical was developed at ISOLDE in the context of BeamLab, and the production of $^{132}Sn^{34}S^{+}$ ions, separated at a mass 166 was tested. In particular, beams of germanium and tin sulfide ions were produced to provide exotic isotopes for different studies at HIE-ISOLDE.

As a hot plasma source, the FEBIAD ion source still presents many intricate parameters which control the performance of the ionization, among these, the thermal behavior and design shape of the ionization chamber geometry. In this framework, many optimization studies have been achieved by ALTO, GANIL and SPES for developing new reliable and efficient prototypes of FEBIAD ion sources.

Introduction

The first exotic neutron-rich krypton isotope beams were produced by the ISOL method at the Niels Bohr Institute more than sixty years ago from an irradiated thick UO_2 target. Along the years, different primary beam drivers were developed and different solutions for ionizing the intense RIBs in plasma sources have been tested. Among these, the use of Forced Electron Beam Ion Arc Discharge (FEBIAD) has been found to be one of the most universal solutions for performing the first ionization of atoms. As a general principle, these sources must be characterized by high ionization efficiency values because the production rate of some specific nuclei can be extremely low. The ionization efficiency is deeply influenced by the electron current impinging the anode and thus by the thermal behavior of the ion source. To achieve these goals new prototypes of FEBIAD type ion sources were developed at SPES and ALTO. At GANIL, the VADIS FEBIAD type ion source was successfully coupled to the SPIRAL1 target. The SPIRAL 1 upgrade makes use of an ECR charge breeder which permits the multi-ionization of the radioactive 1^{+} beams for their reacceleration by a compact cyclotron. This coupling scheme will make possible the application of the charge breeding technique for the production of ^{11}C for hadrontherapy.

A particular challenge for the ISOL facilities is the release and separation with appropriate efficiencies of refractory and chemically reactive beams, such as transition metals, post transition metals and metalloids. The problems come from their generally high melting points, low volatility at operating temperatures up to 2200°C commonly used for the ISOL target operation, associated with eventual chemical reactivity with the structural material components. These difficulties can be addressed using appropriate target and structural materials, and exploiting of the formation of more volatile and stable compounds with chemical reactants made available from impurities or from injection of traces. While the production and formation has been previously achieved at ORNL and ISOLDE in different conditions previously, here the formation and stabilization of these so-called molecular side-bands was monitored for the production of SnS^{+} molecular ions using the recently developed uranium carbide targets and VADIS ion sources at HIE-ISOLDE. The design of a new oven for the provision of elemental ^{34}S was also developed for the production of neutron-rich Sn beams at ISOLDE.

Section 1 – Thermal and Mechanical Developments for the new SPES FEBIAD ion source

The general working principle of plasma ion sources is the electron impact ionization mechanism, according to which free electrons are accelerated by means of an applied electric field, reaching an energy level that is sufficient to produce ionization phenomena when they collide with neutrals. The two main components of a plasma ion source are the cathode and the anode.
For the specific case of ISOL facilities, the former is usually made of tantalum, and is heated at very high temperature (close to 2200 ÷ 2300 °C) by Joule effect. In this way the cathode is capable of generating an intense thermionic emission of electrons on the surface facing the anode. At this point, the electrons can be accelerated by the positive anode potential, impinging the neutrals inside the anode chamber and generating the plasma from which the beam is extracted.

In the case of plasma ion sources, the ionization efficiency is strongly influenced by the electron current impinging the anode and in general, the more intense the electron flux, the higher the ionization efficiency of the source. The parameters that directly control the electron flux are the cathode temperature and the cathode-anode gap by means of the well-known Richardson formula and Child-Langmuir relation [1]. In particular, the aforementioned thermionic emission of electrons can be maximized by increasing as much as possible the average temperature ($T_{cathode\text{-}anode}$) of the cathode surface facing the anode ($S_{cathode\text{-}anode}$); see dotted line in figure 1).

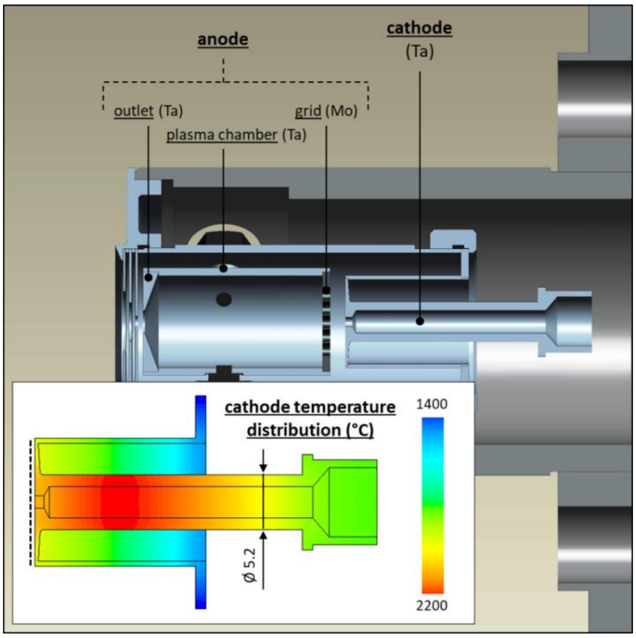

Figure 1. General representation of the first plasma ion source adopted for the SPES facility and focus on the cathode temperature field.

The plasma ion source initially adopted for the SPES facility (see figure 1) was based on the ISOLDE MK5 design [2] and the related cathode temperature distribution (obtained by means of coupled field electrical-thermal finite element simulations [1]) can be observed in figure 1. The correspondent heating current was accurately defined in order to fix the maximum temperature of the hot-spot at 2200 °C, that is the maximum temperature allowed in the case of Ta for reliable operation at vacuum levels between 10^{-5} and 10^{-6} mbar. Unfortunately, with this design the hot-spot at the center of the main tube limits $T_{cathode\text{-}anode}$ and consequently the thermionic emission of electrons.

With the aim to maximize $T_{cathode\text{-}anode}$ the cathode design was properly optimized in order to move the hot-spot as close as possible to $S_{cathode\text{-}anode}$. The resulting design, together with the optimization parameters adopted and the related temperature field is represented in figure 2.

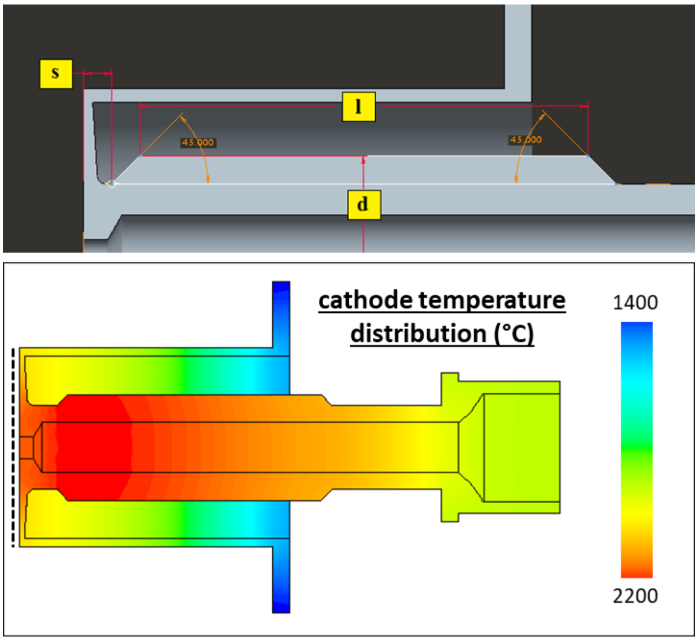

Figure 2. Optimized cathode design and focus on the related temperature field.

As properly discussed in [1], it is extremely important to control the cathode-anode gap and to prevent the contact of the cathode surface $S_{cathode\text{-}anode}$ with the anode grid. With this in mind, the 4-point alignment system represented in figure 3 was implemented in proximity of the cathode interface with the long, tubular transfer line connecting the plasma source with the production target. The aforementioned constraining system limits the axial force caused by the thermal expansion of the transfer line. In this way it is possible to limit the deformation of $S_{cathode\text{-}anode}$ and to prevent cathode-anode contacts, which make the ion source irreversibly inoperable.

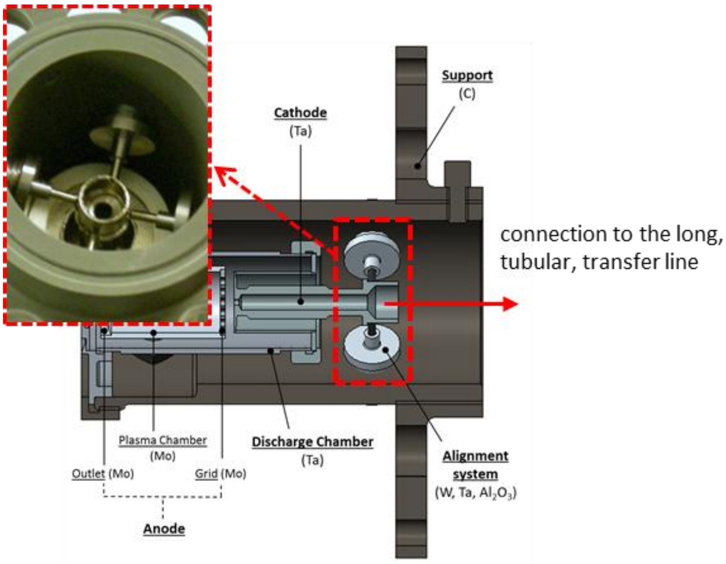

Figure 3. The cathode alignment system.

The two modifications described above led to the definition of the new SPES plasma ion source. It was tested with stable beams for long periods at Legnaro National Laboratories, and proved to have a stable behavior for both intermediate (150 mA) and high (250 mA) anode current (electron current) values. In particular, after ≈ 40 heating - cooling cycles and ≈ 300 working hours at 2200 °C the cathode proved to retain good general properties.

Figure 4. Optimized cathode after ≈ 40 heating - cooling cycles and ≈ 300 working hours at 2200 °C.

The new SPES plasma ion source was tested off-line with noble gases and metallic elements. The ionization efficiency measurements performed are reported in table1.

beam	ion. eff. (%)	injection mode	cathode temp. (°C)
Ar	6	gas tube	2200
Br	8	oven	2200
Kr	9	gas tube	2200
Y	< 0.1	oven	2300
Sn	10	oven	2200
I	19	oven	2200
Xe	11	gas tube	2200
Cu	10	oven	2200
Ag	15	oven	2200

Table 1. Ionization efficiency values measured at Legnaro National Laboratories for the new SPES plasma ion source.

The same ion source was adopted also for the production of SnS molecular beams. Dedicated experimental campaigns are ongoing and the related results will be soon available.

SECTION 2- NEW BEAMS WITH FEBIAD ION SOURCE AT GANIL-SPIRAL2 AND ISOLDE

2.1. *LATEST DEVELOPMENT AND TESTS AT GANIL-SPIRAL2*

At GANIL, the FEBIAD Target Ion Source System (TISS) couples standard SPIRAL1 1200W carbon targets to the VADIS [3] via an ohmic heated tantalum transfer tube (figure 5). The TISS was developed in a staged approach. The early versions were first tested online in 2011 using the SIRa test bench at modest primary beam power. They demonstrated the efficient ionization of radioactive isotopes of 8 new elements: the alkali Na, K, metallic Mg, Al, Fe, Cu, Mn and halogen Cl elements. A more reliable prototype has been tested at nominal power (1200W of ^{36}Ar at 95 AMeV) in the SPIRAL 1 beam lines in December 2013, leading to the first scientific results obtained at SPIRAL 1 with a FEBIAD source [4] [5]. These on-line tests at SPIRAL1 gave generally even better secondary beam intensities than expected from the measurements at SIRa, showing that the yields do not scale linearly with the primary beam power. The intensities shown in figure 6 were published in [6]. In order to increase the reliability of the target ion source, a program of test was rigorously followed on the SPIRAL1 off-line test bench with the aim to select the best insulator, which could stand operation under high temperature. The results of these tests showed that on the test bench boron nitride (BN) insulators permitted a stable operation over a period of about 3 weeks. 2016 and 2017 were used to provision the FEBIAD target ion sources for the startup of the upgrade and diverse characterizations detailed below, which were part of the Beamlab activities.

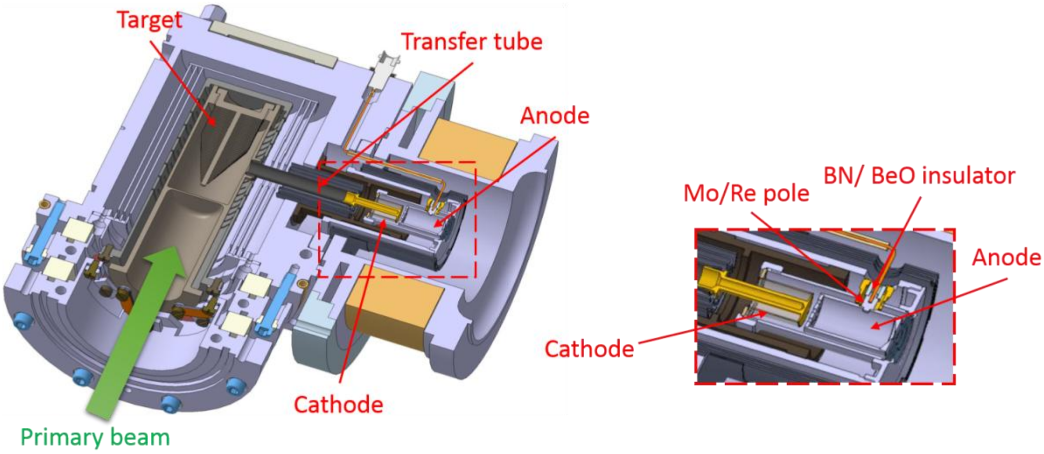

Figure 5: FEBIAD TISS. The right inset is a zoom on the ion source critical parts. See text for details.

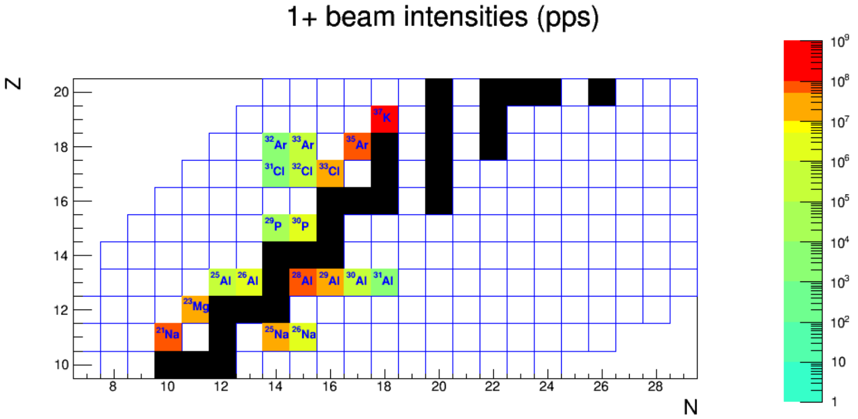

Figure 6: Secondary intensities measured with the SPIRAL 1 FEBIAD TISS with 36Ar at nominal power (~1200W) in 2013.

In April and May 2018, the SPIRAL 1 upgraded facility was commissioned for radioactive ion beam production. A beam of 95A MeV of ^{20}Ne first impinged on the SPIRAL 1 target for the production of a ^{17}F beam for the E750 experiment. Unexpectedly, the anode entered again into a short with the cathode very rapidly (6-8h) after the beam was tuned on target. Again, the anode insulators were identified as the probable cause for the FEBIAD breakdown, contrasting with the observations done on the test bench without beam on target. During discussions with a panel of experts from ISOLDE and TRIUMF, the possible explanations mentioned for the different behavior of the source on-line and off-line were the different heating /thermal conditioning of the target ion source as well as the possible deposition of direct C vapors on different parts of the source, including the insulators. In order to fix these issues, a new FEBIAD ion source was conditioned with a different configuration of heat shields permitting to cool down the insulators. A helical chicane was inserted in the transfer tube to stop the direct C vapor from the irradiated target (figure 7). With these modifications, the FEBIAD target ion source could be run with first a low power (100-200W) ^{40}Ca beam and finally a high power (>800W) ^{36}Ar beam, both at 95A MeV without failure. The secondary beam intensities measured for the latest test are shown in figure 8. As it clearly appeared later, the diagnostic used to center the primary beam on target was malfunctioning during the commissioning tests of 2018. The wrong steering caused lower yields than expected for the most reactive elements: Cl and Al. As it was observed later with a Nanogan TISS delivering ^{14}O to the E744 experiment, a factor of 50 to 100 improvement of the production yield could be obtained at the SPIRAL identification station [7] by tuning the primary beam steerers to correct for the faulty diagnostic.

Figure 7: photograph of the transfer tube and associated helical chicane.

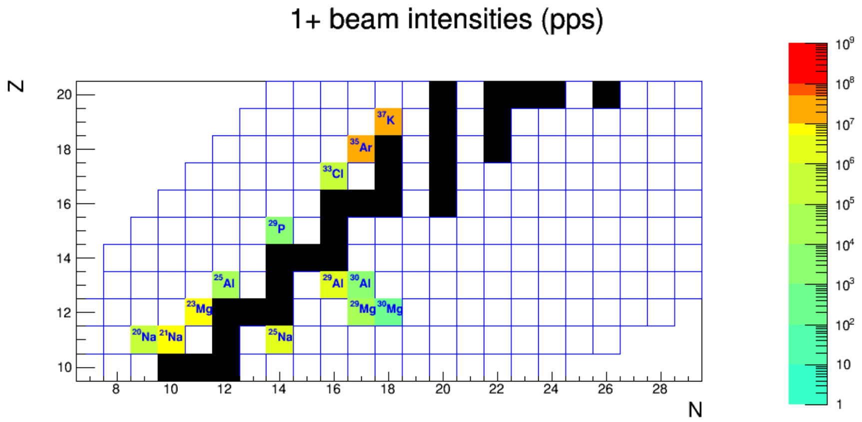

Figure 8: Secondary intensities measured with the SPIRAL 1 FEBIAD TISS with ^{36}Ar at nominal power (800 - 1200W) in 2018. A wrong steering of the primary beam is invoked as the reason for the lower yields as compared to 2013.

The FEBIAD target ion source is being presently further consolidated for final adjustment on the test bench, before being recommissioned online in the upcoming running period. In addition to the aforementioned modifications, a few adjustments are tested. In particular the material of the poles supporting the fragile insulators, originally in Mo, are replaced by Re (see right inset of figure 5), which has a lower thermal conductivity. These Re poles will permit to further protect the insulators from the hottest parts of the ion source. With these adjustments, it is believed that the FEBIAD target ion source will have most chances to behave very well online.

During the conditioning of the various versions of the TISS, the ionization efficiencies of the FEBIAD have been repeatedly monitored on the off-line test bench with stable rare gases. These ionization efficiencies are generally reproducing the efficiencies of the traditional MK5 of ISOLDE, which are a factor of ~4 lower than the VADIS ones quoted in [3]. ISOLDE equally reports that similarly lower efficiencies are regularly obtained on-line with the VADIS. Nevertheless, it is remarkable that the efficiencies quoted in [3] could be sometimes obtained during the conditioning of the TISS at the SPIRAL 1 test bench, over periods of a few hours. The conditions for stabilizing this enhanced ionization regime have not yet been found, and are being investigated. One of the latest manifestations of this particular regime happened during the injection of a tiny leak of Xe (figure 9), whose flow rate enabled the control of the ionization efficiency, switching gradually from the standard low ionization regime to the enhanced ionization regime and vice-versa, for a couple of hours.

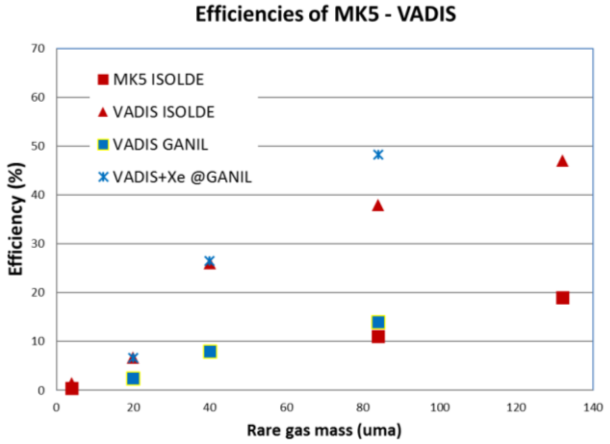

Figure 9: Ionization efficiencies measured at ISOLDE and GANIL with the different FEBIAD sources and in different ionization regimes. See text for more explanations.

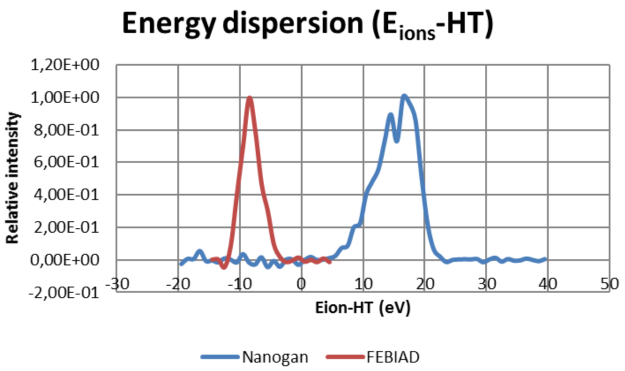

Figure 10: energy profiles of the Nanogan and FEBIAD ion sources.

The energy profile of the 1^+ beam from the FEBIAD source is of high interest for the charge breeding performances. The SPIRAL 1 upgrade makes use of an Electron Cyclotron Resonance (ECR) charge breeder which permits the multi-ionization of the radioactive 1+ beams for their reacceleration by a compact cyclotron. The energy acceptance of the ECR charge breeder is defined by the width of its "ΔV" curve; the n+ extracted ion beam current as a function of the difference of voltage between the 1^+ source and ECR charge breeder. The acceptance of ECR charge breeders is typically of 5 to 10 eV FWHM for condensable elements. The energy profiles of the FEBIAD and Nanogan III could recently be measured and compared using an analyzer manufactured by LPC Caen (figure 10). The energy dispersion of the FEBIAD ion source is of the order of σ_{Ey}=1.5eV, which is well within the acceptance range measured with the SPIRAL 1 charge breeder. It is also remarkable that the plasma potentials of the Nanogan and FEBIAD source differ considerably. The origin for the voltage shown in figure 10 is somewhat arbitrary, because of the lack of calibration of our test bench HV power supplies. It corresponds to the 1+ source HV voltage within 20-30 V error bar. Latest simulations show that the FEBIAD can have plasma potentials as low as -70V, with an electron beam current as high as 300 mA (figure 11). This result was experimentally corroborated by the first charge breeding of a radioactive ion performed at SPIRAL 1 [8].

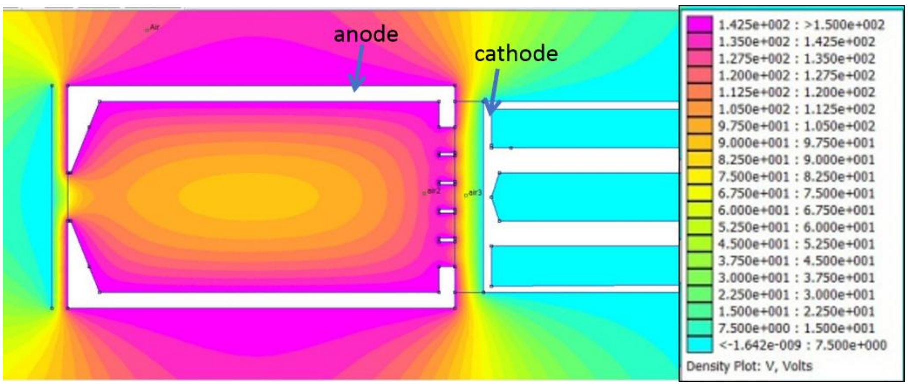

Figure 11: first result of the combined FEMM4.2 – SIMION simulations, showing the potential calculated in the anode of the FEBIAD by a 300 mA electron current. The voltage of the anode body is 150V with respect to the cathode.

In order to identify the most important mechanisms that govern the ionization performances of the FEBIAD, simulations combining the SIMION software for calculating the trajectory of charged particles (electrons and ions) in fields, and FEMM 4.2 for accounting for space charge effects, are being undertaken, for which figure 11 is an example of a preliminary result. At present, these simulations reproduce the plasma potential reasonably well, as the latter defines the energy profile of the 1^+ beam (mean energy and dispersion) of the extracted beam. Possible ionization regimes by pure electron impact or in plasma will have then to be investigated to attempt to reproduce the order of magnitudes of the measured efficiencies.

2.2. IMPROVED DELIVERY SYSTEM FOR SULFUR REACTANT WITH FEBIAD ION SOURCE AT ISOLDE

The formation of molecular beams can be promoted by introducing reactive compounds to the target and ion source volumes. To ensure a constant delivered ion beam quality, it is required to be able to adjust the reaction rates by controlling the quantity of the injected reactive component.

There are two main concepts for introducing additional compounds to the target and ion source units. Gaseous compounds can be injected through the buffer gas delivery system for the plasma ion source. Here the concentration of the reactive compounds can be easily adjusted by regulating the pressure applied to a specially tailored vacuum leak. This method has been very successful for the production of Fluoride beams where the reactive gas can be for example CF_4.

Elements with high melting point on the other hand can be introduced using the mass-marker technique. Here the compound containing the element is placed inside a tantalum capillary which is electrical and thermally connected to the target and ion source volume. The evaporation rate can be adjusted through adjustment of the electrical heating power applied to the capillary. This method works well for metals.

For the case of sulfide beams, it is desired to introduce isotopically pure ^{34}S in elemental form. It has been shown that when using the mass marker technique, all sulfur is released already during the initial conditioning and calibration of the target. The reason for this is the good thermal contact between the mass marker capillary and the hot target itself. Measurements with thermocouples have shown that the coldest point in the mass marker capillary still exceeds the boiling point of sulfur of 444 °C.

In order to be able to control the release of sulfur, a gradual improvement of the mass marker concept was performed. First the length of the capillary was increased, and the heat screening was improved. In a second step, the capillary was separated electrically and thermally though an adapter made from boron nitride. The latest stage of the development is shown in figure 12. Here the sulfur is placed in a heater assembly that is made from boron nitride. A tantalum wire is used to provide heat to the assembly. The cartridge is placed in a special aluminum case which is machined to match the vacuum feedthroughs of the target base. This allows placing the sulfur reservoir in a naturally cold area of the target unit assembly. Once vaporized, the sulfur diffuses through a standard mass-marker tantalum capillary to the target and ion source volume.

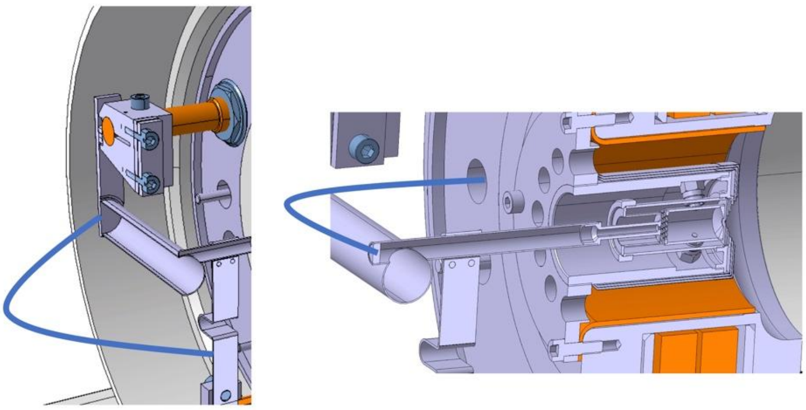

Figure 12: Top: In blue, Mass Marker or gas leak capillaries used for chemical traces injection in the target container or in the transfer line. Bottom: Heater assembly for reactants with low boiling point. From left to right: Aluminum case, heating cartridge: boron nitride insulating the tantalum heating wires and contains the sulfur powder, plug.

The functioning of the new delivery system was verified at the heating test stand using a residual gas analyzer: The signal of Sulfur was recorded over time while the heating current was changed.

Finally the new design was applied in an on-line experiment where $^{132}Sn^{34}S^{+}$ beams were delivered. A former capillary system was already used at HIE-ISOLDE to deliver $^{132}Sn^{34}S^{+}$ [9].

SECTION 3- NEW DEVELOPMENTS ON IRENA FEBIAD ION SOURCE AT ALTO

A new FEBIAD-type ion source, named IRENA, has been developed at IPN Orsay to operate efficiently and steadily under strong radiation conditions [10]. It has been designed with a radial configuration of the anode–cathode set to allow both efficient ionization and the confinement of the positive ions for an efficient extraction. The operation without magnet is an important advantage since this particularly ensures the reliability and reduces substantially the ion source part of the radioactive waste. To optimize the anode–cathode system and to improve the mechanical and electrical reliability, the second prototype was completely modeled with 3D-Lorentz simulation code [11] [12]. Simulations for the third prototype of the IRENA ion source were achieved, in particular, for ionization, extraction process and thermal design.

3.1. NEW IONIZATION CHAMBER AND EXTRACTION DESIGN FOR IRENA

The ion source should have enough space charge effect for repelling the electrons. Not enough space charge implies not enough time for electrons to impact the atoms which will result in low ionization efficiency. For this kind of radial-type FEBIAD ion source, figuring out what kind of anode and cathode will provide enough space charge effect and ionization area for electrons is a key design point. Nevertheless, the lifetime of the electrons is also very important to improve ionization efficiency. In order to investigate the space charge effect as well as the electron mean lifetime, a seven step multi-electron simulation is performed. Finding the optimal operating parameters for the anode is the primary task for the ion source study. However, the anode radius is fixed at 8 mm because of the mechanical constraints, only the distance between anode and cathode can be optimized in the ion source design. As shown in figure 13, that the best distance between anode and cathode is around 1 mm, and that the electron motion is very sensitive to the applied voltage.

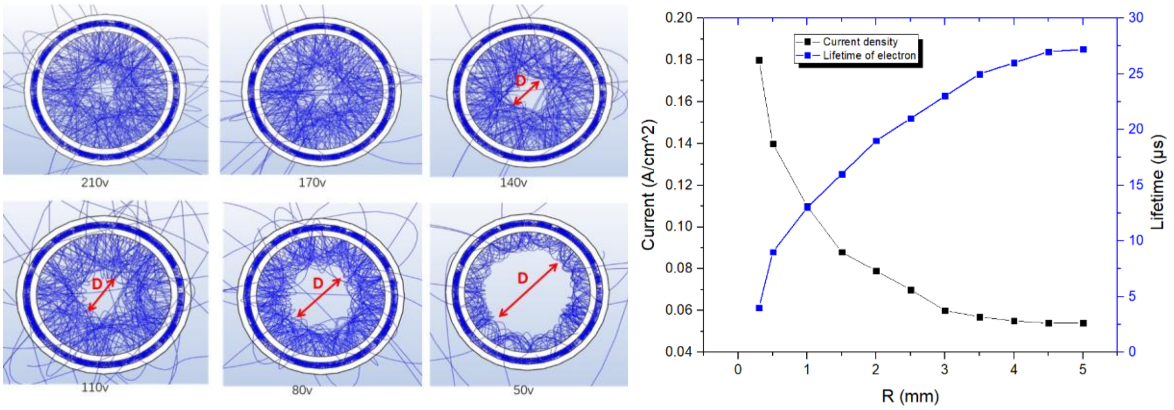

Figure 13: The electron trajectory simulation results for IRENA3. R represents the distance between anode and cathode, and D the diameter of the potential well created inside the ionization chamber.

According to the simulation results, the R has a little influence on the space charge effect. The space charge is manly dependent on the applied voltage between anode and cathode and thus on the velocity of the electrons. To get the best ionization efficiency a compromise between the lifetime and the electron current needs to be found. The blue curve in figure 13 represents the mean lifetime of the electrons. It's very hard to simulate the ionization in a plasma environment. We propose a simple simulation which deals with electron motion and collisions between electrons and gas. We deduce the relative weights for current and lifetime, *Ionization=current*40%+lifetime*60%*. The output of this study leads to an optimal R value of 1.5 mm.

To design an optimized extraction system, detailed multiparametric simulations have been carried out. There are five mechanical parameters which have to be optimized. Simulation results are shown in figure 14. L1 is the distance between anode and cathode and L2 is the distance between ground electrode and extraction electrode. For L1 and D1, D2, D3 the best distances are 2 mm. The extraction electrode will be on -30 kV. The simulation results are showed in figure 14 (right). The emittance value is strongly correlated with the L2 value. The best L2 value is 10 mm.

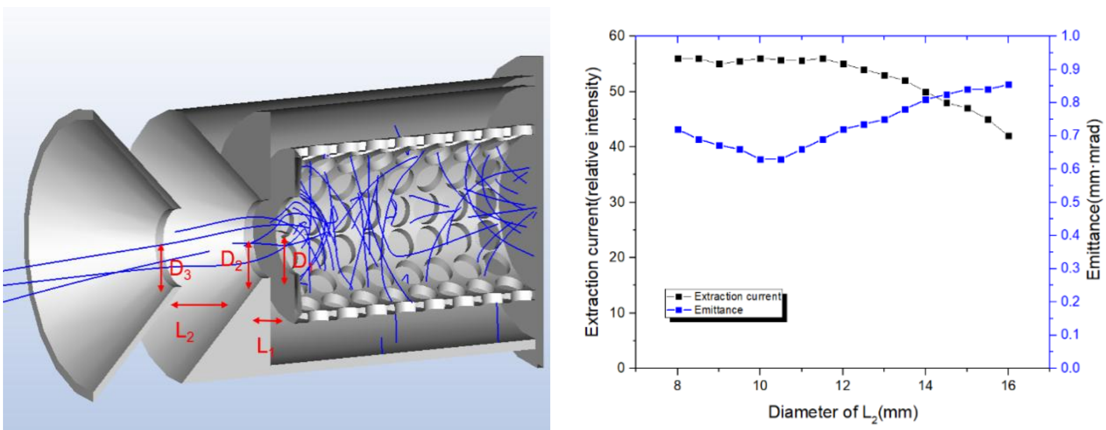

Figure 14: The extraction simulation for IRENA3

3.2. THERMAL DESIGN SIMULATION FOR IRENA

To produce enough electrons for ionization, a cathode temperature around 2200℃ is needed. This is extremely important for IRENA3 designs. Thermal simulations done by ANSYS 19.0 were carried out to optimize the thermal behavior of this prototype.

To withstand the high temperature, the round wings and anode are made of heat-resisting material tantalum. The new design for IRENA3 shown in figure 15 (left) is simpler and lighter.
In order to protect other assembly units from the thermal radiation, reflectors made by 0.1mm tantalum foils are used to reflect the thermal emissions. Figure 15 (right) shows the simulation result with 4 layers of thermal reflectors around the cathode. We found that 810 A is the current needed to reach 2270 ℃ on the cathode. We note that for this new design of IRENA3, only one third of tantalum mass is needed compared with the IRENA2 prototype. Furthermore the fixation system of the anode and cathode is also simpler and more reliable.

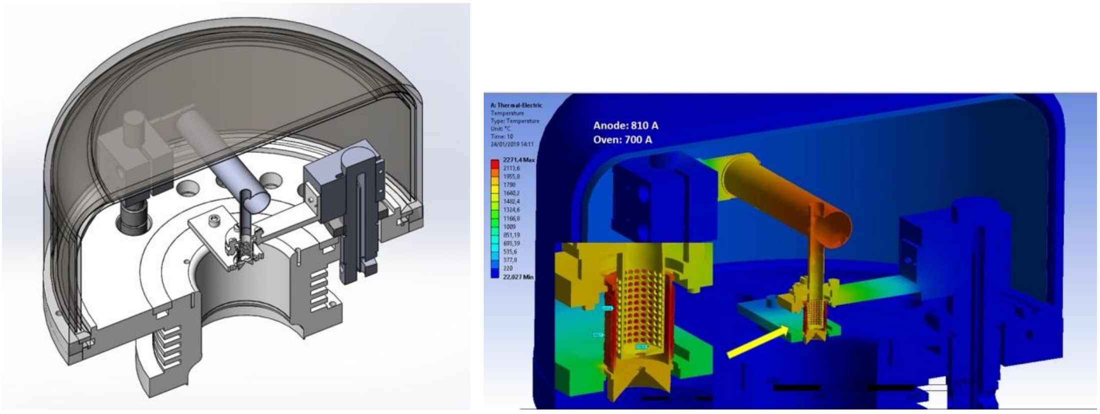

Figure 15: The new design and the thermal simulation result for IRENA3

CONCLUSION

The critical and most key parameters of radioactive plasma ion sources were studied by both numerical simulations and experimental tests. The numerical simulations led to the development of new prototypes optimized to be operational in the different existing ISOL facilities such as ISOLDE, GANIL and ALTO and for the SPES project. Due to this collaborative work, FEBIAD type ion sources become more and more reliable and give access to new beams which were difficult to access previously with this kind of ion source. The VADIS FEBIAD type ion source was used at ISOLDE for the development of new molecular beams around the doubly magic ^{132}Sn. The Sulfur bond chemistry was exploited by providing enriched ^{34}S in the target gas phase for the formation of SnS molecules before their ionization and formation as a beam.