#### CHAPTER VI

# **RIB SOURCES**

#### 6.1 General criteria for target and ion-sources

The ion-sources dedicated to the production of Radioactive Ion Beams (RIB) have to be highly efficient, selective (to reduce the isobar contamination) and fast (to limit the decay losses of short-lived isotopes). For radioactive beam generation the source must operate steadily for extended periods of time at elevated temperatures (up to 2000°C). The selection of the most appropriate choice for the target/ion source is of paramount importance since its performance determines the intensity, the beam quality, and the number of radioactive beams that can be provided for experimental use.

The world wide spread RIB facilities came up with a large variety of solutions to meet part or all of these requirements such as: surface, plasma, electron cyclotron resonance and laser ionsources [1]. A figure of merit of the 1+-ion sources for RIB is presented in Figure 6.1 as a function of the ionization potentials.



*Figure 6.1:* 1+ Ionization efficiencies measured with surface (black squares), plasma-FEBIAD (circles), laser (triangles) and ECR ion sources from ref [1]..

The choice for SPES project to develop a Target-Ion Source Chamber unit based on the ISOLDE one, implies the possibility of using a great part of sources developed at CERN, with the ability to choose and then to plug one of those into the transfer tube of the multi-foil SPES target. In an ISOL facility the volatile nuclear reaction products are released from the target material and diffuse via a transfer line into the ion source, so that the target and ion source system form a self-contained unit specifically optimized for each element or group of elements. The importance of choosing the ion source is due to primarily been dictated by efficiency and secondarily by its capability of selective ionization. All ions produced are accelerated towards the ion extraction electrode by a potential up to 60 kV.



Figure 6.2: The SPES fission isotopes produced by the three main ionization schemes considered

The fission isotopes produced in the UCx target, as shown in red colour in Fig 6.2, have different masses (between 60 < A < 160) and different chemical proprieties. A part of those, the refractory elements (Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd), do not escape from the target material because they have an very high vapour pressure. So we can classify the remaining of elements produced in the fission target according three sets in terms of the magnitude of first ionization potential I, thus ionization method, as reported in the Figure 6.3,

In the first group there are the Alkaline and the Rare Earth as Rb, Cs, Sr, Ba. They have a small +1 ionization potential, less than 6 eV, and they may be well ionized using a technique named 'Surface Ionization Method'. This technique is quite selective and we can reach high values of ionization efficiency, up to 60%.

In the second set we find the metal elements, like Ni, Cu, Zn, Ga, Ge, Ag, Cd, In, Sn, Sb and Te. They have an intermediate value of +1 ionization potential, approximately between 6 and 10 eV. They can be ionized successfully using a technique named 'Laser Ionization Method', using up to three laser tuneable beams. This technique is the most selective method in RIB applications.

Finally we have the noble gas (as Kr, Xe), the halogens (as Br, I), the As and Se elements. They have a high I value, even more than 10 eV. The powerful method in order to ionize this set of elements is named 'Ionization by Electron Impact' or 'Plasma Ionization Method'. This last technique is not selective, but we can reach quite high value ionization efficiency; for instance, for noble gas elements like Xe some experimental measurements reported efficiency up to 40 %. In Figure 6.3 we have reported briefly the main characteristics of the three ionization techniques discussed above.



*Figure 6.3:* The three different schemes for the +1 isotope ionization.

At the LNL SPES test bench laboratory, since May 2010, the off-line front end device has been working: this apparatus is focused in order to measure the ion source performances in terms of ionization efficiency and transversal emittance. It is important to point out that the SPES front end, see Figure 6.4 and 6.5, is an evolution of Isolde front end FE6.

In the off-line front end the production of the isotope beams originates in a dedicated oven, connect to the ionizer. In this oven, following a special method called 'Mass Marker Capillary Method', a fixed amount of stable atoms is inserting in.



Figure 6.4: The devices of SPES front end

The target system, the ion source complex and the transfer tube are located under vacuum inside a water-cooled chamber are maintained at the average temperature of 2000°C: vacuum and high temperatures are two essential components to enhance the radioactive isotopes mobility, extraction and ionization.

The neutral atoms diffuse into the ionizer via a transfer line, where, once charged +1, are accelerated up to 30 kV voltage by the extraction electrode complex. After this stage the beam will interact with the front end Low Energy Beam Transport (LEBT) complex. The first device of the LEBT is the deflector system which consists of four electrostatic steerers. By tuning the steerer electric field, it is possible to modify the position of the beam centroid in the transverse plane.

The next stage of the front end LEBT system consists of a triplet of electrostatic quadruples responsible for bringing, by changing the values of the electric field, a focus in a desired downstream point.

Immediately after this beam focus system is inserted a diagnostic box which consists on a beam profiler and a Faraday cup. These devices are able to measure respectively, the beam spot and the total electric beam current.

Finally an emittance meter device, see Figure 6.6, is connected to the front end in order to measure the beam transversal emittance in the X and Y axis.



Figure 6.5: A picture of the SPES off-line front end working at LNL



Figure 6.6: The SPES Emittance Meter

We consider here all the three kind of ion sources for SPES: the Surface Ion Source (SIS), the Plasma Ion Source (PIS) based on the Ionization by electron Impact Method, and finally the Laser Ion Source (LIS). All of these three sources are used at ISOLDE and they constitute a good reference point for further SPES goals in the ion-source development.

## 6.2 The Surface Ion Source

The concept of surface ionisation has proven to be particularly successful for production of singly charged positive and negative radioactive ion-beams thanks to its simplicity, high efficiency and selectivity [2]. As shown by the Langmuir equation, ionisation efficiencies for positive ions of 50-100% may be obtained for elements with ionisation potential <5 eV and of negative ions for elements with electron affinity > 2 eV. Saha and Langmuir described the ionizing properties of a hot surface. Positive ions are produced when the minimal energy needed to remove an electron from a surface (its work function) is larger than the ionization potential, and negative ions are produced when the work function is smaller than the electron affinity of the atom impacting on the surface at thermal energies.

Surface ionization remains the most efficient ionization scheme for low ionization potential radioisotopes (alkalis and some lanthanides) that are currently produced with W,  $WO_3$  and Re surfaces. In Figure 6.7 is reported a picture of the SPES Positive surface Ion Source (SIS), which has been successfully tested to produce the first ionization test of Alkaline at LNL (Rb and Cs).



*Figure 6.7: The SPES Positive ion source set-up (with the Ta transfer line) connected to the target-ion source chamber frame.* 

The surface ion source adopted for the SPES facility is similar to the standard MK1 surface ion source developed and used at the CERN-ISOLDE facility. It is composed of a tungsten tubular ionizing cavity connected on one side to a tantalum support and on the other one to a Ta transfer line by means of connectors. The surface ion source and the transfer line are kept at high temperature thanks to a dedicated and independent 10 kW power supply, able to deliver a maximum electrical current and a maximum potential difference of 1000A and 10V, respectively. The electrical current flows through the W tubular ionizing cavity, called also hot-cavity, which dissipates, by Joule effect, the power needed to reach the desired temperature level. The same electrical current passes also through the Ta transfer line, thus keeping it at high temperature. In figure 6.8 it is reported the layout of the target-ion source chamber, where the surface ion source is connected to the oven system.



Figure 6.8: The SPES target chamber with the SIS ion source connects to the oven system.

A coupled thermal–electric Finite Element model was defined, in order to study in detail the thermal–electric behaviour of the surface ion source and of the transfer line at present adopted and to predict in a virtual environment the effect of some modifications that could be adopted in the future. Then, aims of simulations are as follows: 1) to calculate the heat dissipation by Joule effect in the SPES surface ion source and transfer line; 2) to calculate the correspondent temperature distributions; 3) to estimate the effects of some material and geometric variations on the temperature distributions .

All theoretical results (both analytical and numerical) have been finally compared with thermal and electrical experimental measurements. The comparison of the theoretical results with the experimental data has been in a good agreement, as reported in Fig 6.9



Figure 6.9: SIS Finite Element Model's temperature plot and the comparison between FEM and experimental data.

The emittance of the SPES SIS ioniser has been measured recently at LNL, mainly for alkaline, for a number of different operating conditions in the SPES front end. The emittance is strongly dependent on the ionizer temperature and the extraction electrode position. Nevertheless

the average value of the transverse 90% emittance measured at 30 kV is less than 10  $\pi$  mm. mrad. and the energy spread is less than 10 eV.



Figure 6.10: Transversal (X axis) emittance of SPES SIS ion source.

### 6.3 The Plasma Ion Source:

The plasma ion-source PIS currently used in several RIB facilities is based on the Forced Electron Beam Induced Arc Discharge (FEBIAD) concept originally developed by Kirchner [4] at GSI. The principle is based on the capability of electrons, coming from an indirectly heated disc-shaped cathode and accelerated into the anode chamber by means of a grid, to ionize any atoms, floating in the anode chamber, with ionization potential smaller than the energy of incident electrons. As a consequence, the source is well suited for ISOL applications and it operates steadily and efficiently in conjunction with high temperature thick target materials over a pressure range of  $10^{-5}$  to  $10^{-4}$  Torr.

The PIS is particularly useful for the ionization of highly reactive or condensable elements for which wall sticking would limit their release from a surface ion source cavity. With electron impact energies between 100 and 200 eV, elements with very high ionization potentials (e.g. Xe and Kr) can be efficiently ionized. In fact the efficiency of the plasma ion source is quite high for slow moving heavy ions; for low mass, a fast moving atom with high ionization potentials, the source is not so impressive. For example, the measured ionization efficiencies for the noble gas elements are, respectively: Ne: 1.5%; Ar: 18%; Kr: 36%; and Xe: 54%.

The plasma ion source is also capable of producing multi-charged ions, but the limited selectivity, offered by this kind of ion source, can be improved by exploiting the chemical or physical properties of the atoms when as soon as they are released from the target.



Figure 6.11: The SPES high temperature plasma ion source inside the target container.

The SPES plasma ion source is an evolution of ISOLDE MK5 [5]. The cathode is made of tantalum, and it consists of three parts welded together an electron beam welding and press fitted into the transfer tube. The cathode temperature, and thus thermionic electron emission, is controlled by a DC current (arc discharge). The collimation of the electron beam optimizes the ionization of the species of interest and it is obtained by adjusting the magnetic field generated by a coaxial solenoid.

The discharge chamber and the anode assembly are made of molybdenum and screwed into a graphite cylinder rigidly fixed to the main target base as shown in Figure 6.11. The anode is insulated by three  $AlO_3$  insulators. The anode grid consists of a graphite disc with holes drilled through in order to let electrons being accelerated into the discharge chamber. The source is surrounded by three heat shields made of molybdenum.

A current flows through the transfer line via the cathode, the anode cylinder, the external graphite tube and back through the main target flange. The advantage of this design is that only one power supply is needed for heating the line, cathode and ion source. The same power supply can also be used for heating the tubular surface ionizers.

In the last months several off line tests have been performed at LNL SPES laboratory, as shown in Figure 6.12. In particularly, also for this ion source system, a dedicate coupled thermal–electric Finite Element (FE) model was defined, in order to study in detail the thermal–electric behaviour in order to predict, in a virtual environment, the effect of some modifications that could be adopted in the future (see Fig 6.13).



Figure 6.12: The SPES PIS during a temperature test performed in the LNL SPES HT laboratory.



Figure 6.13: The SPES PIS Finite Element Model with the 3D drawing.

### 6.4 The Laser Ion Source

The Laser Ion Source (LIS) method is nowadays the most powerful tool for radioactive ion beam production at on-line facilities, because it provides a selective ionization process with inherent suppression of unwanted isobaric contaminations at the ion source. It is worth to note that LIS operates on the same mechanical set-up designed for and used in the surface ionization technique (Figure 6.3, Figure 6.7).

The photoionization pathway (Figure 6.14) usually involves a photon absorption ladder within the electronic levels of the atom, each step being resonant with the optical transition of the desired atomic species. In this stepwise model, a valence electron is brought or directly to the continuum or to a highly excited Rydberg level in presence of an electric field or to an autoionizing state. These latter two techniques, when feasible, have better efficiency. Of course each optical transition requires a dedicated laser wavelength, i.e. colour. Usually two or three different colours are required for a given chemical element. Using tuneable lasers (solid state or dye or a combination of the two) it is possible to match the photon energy of the laser lights to the electronic transitions of the desired atomic species. For many elements, this ionization by stepwise resonance photon absorption can provide an unmatched level of selectivity because it is the unique electronic structure of different atomic species that gives this process its selectivity. In few selected cases, isotopes or even isomers of some elements can be isolated by operating the lasers in a narrower band mode, typically inserting an etalon into the tuneable laser cavity.



Figure 6.14: Schematic of laser photoionization for Ni (from P. Van Duppen) [6]

The ionization efficiency is heavily reliant on the saturation of the resonant photon absorption steps; therefore the spectral radiance requested to the laser system depends also from the optical atomic parameters (spectroscopy) of the atomic sample released from the target [7]. Furthermore the last step, for kinetic reasons and especially when involving a direct ionization, is demanding for a high irradiance. This goal can be accomplished using pulsed lasers, which must operate at high repetition rate in order to process all the fragments as they are coming out from the target. To this purpose, currently the above mentioned tuneable lasers are pumped by the second harmonic of Nd-Yag, in turn pumped by diode laser, instead of the old copper vapour lasers.

As far as the emittance of the beam, it is even better than that obtained with the surface ionization technique, by a factor 0.75 [8]. Of course, this comparison is meaningful because it is possible to use the same mechanical set-up, as above specified.

Typical laser systems are the well known RILIS at ISOLDE, which provide beams for more than 20 elements or that planned at Oak Ridge National Laboratory [9]. Incidentally, the ISOLDE system has been very recently updated for even better performances. The laser beams are directed through a window and focussed into the target located more than 10 meters away from the laser room, in agreement of health physics regulations.

The Laser Ionization Method has been investigated in the past at Pavia University [3], as a spin-off of the atomic vapour laser isotope separation. Accordingly, during the last two years collaboration started up between Legnaro LNL and Pavia in order to begin the preparation of the SPES laser ion source. As first step, dye laser will be used to generate resonant light source. These laser are in turn pumped by the second harmonic of Nd-Yag laser. All these units are already present in the Pavia laboratory. The main work done has been to bring back into use the Nd-Yag, a Quanta System model, and three dye lasers, manufactured by Lambda Physik. This has been successfully done and three step photoionization has been obtained for instance on Neodymium. The atomic source is a hollow cathode lamp and the signals are optogalvanic type. The final goal is the set-up of a system made of three tunable lasers, an atomic beam and a time of flight mass spectrometer. This system is intended for a full diagnostic of LIS applied to the chemical elements belonging to the fission fragments selected by SPES group.

In Pavia the tunable laser is and will be pumped by the Nd-Yag running at 10 pulse per second. At the moment this is sufficient for diagnostic purpose. However the frequency repetition rate of laser is of paramount importance in the effective ionization system.



Figure 6.15: The Pavia dye laser pumped by Nd:YAG-SHG.

The flash lamp Nd-Yag must be replaced by, for instance, pulsed diode pumped Nd-Yag. In this case a frequency repetition rate, as large as 10 KHz, can be achieved by current technology. Furthermore, the dye laser could be replaced by Ti-Sa units, this last option being not for sure convenience.

In the framework of target SPES activities, beside an important target development and characterization, is fundamental a deep understanding of the ionization, extraction and shaping capability of the ion beam by the front-end.

In order to evaluate all these properties, studies and experiments are under development in Legnaro. To evaluate the shape of the beam, and the capability of manipulate it, is possible to measure beam properties with Faraday cup, beam profiler and emittance meter, activities that are nowadays in progress.

Other key property is the capability to extract ions from the ionization tube, which is under test with mass marker technique. With the same setup used for surface ionization is possible to use laser ionization, and characterize the system also with laser ionization technique.

Among the materials useful for these experiments, Aluminum is a well candidate in order to evaluate the laser ionization efficiency: the choosing of this material is motivated by several reasons. In the initial phase of target prototyping, SPES has developed a SiC target beside UCx.

This target, already tested in HRIBF in Oak Ridge, will be the first target used in SPES Online Facility. Impinging SiC target with protons, Aluminum atoms comes from (Si,p) nuclear interaction. Is important to point out that the aluminum is, most probably, the first p-rich beam of the SPES facility.

In fact from the experimental data collected at Oak Ridge, it is possible to observe, using the SiC target, that the isotope production for Al spans from mass 26 to mass 29.



Figure 6.16: SiC target prototypes tested at HRIBF.

The ionization path used involves only one wavelength, 308 nm, which is resonant for the first light absorption step and brings electron towards continuum energies with a second photon of same energy.

Using a standard notation it is possible to write:

Al [308.215;308.215 e–] Al+



The first resonant transition implies a intrinsic selectivity of the process, because any other element, except Aluminum, present in the ionization chamber, theoretically, will not absorb photon at 308 nm.

This process is known as one color two step ionization. Thanks to single color, two step ionization, is possible to use a single laser to achieve ionization, and an excimer laser, which filled with XeCl emits photon at 308 nm, is already present in LNL.

Excited Dimers (Excimers) are molecules which are bound in excited states but are unstable in their electronic ground states. These excimers are ideal candidates to form the active medium of lasers, since inversion between the upper bound state and the dissociating lower state is automatically maintained.

These are the main characteristics of the Lambda Phisyk LPX200 excimer laser (the laser working at LNL) by the manufacturer:

Power supply	380 VAC, 50 Hz
Output wavelength	308 nm
Energy per pulse	Up to 400 mJ (1Hz)
Max repetition rate	Up to 200 Hz
Gas mixture	HCL/He/H2 + Ne + Xe
Total pressure	2900 mbar

Pumping sources are provided by high voltage, high current electron beam source, or by fast transverse discharges. Works in order to use the laser are under development in SPES Front End Laboratory at LNL, to propagate the laser beam leaving the room also to other measurements.





Figure 6.17: LNL Front End layout laboratory study and disposal.



Figure 6.18: Laser beam spot and path towards ionization chamber.

Among the salts used in mass marker capillarity technique, is possible to charge the furnace with an Aluminum salt, which once evaporated allows to the Aluminum atoms to reach ionization tube.

Nowadays placing a calibrated amount of salts in a oven directly connected to the ionizer it is possible to perform ionization test using a laser beam in the off-line SPES front end.

# 6.5 Characterization of the SPES surface and plasma ion sources

The surface ion source described in the previous paragraphs is directly connected to a tantalum transfer line. Both of them are heated by Joule effect using the same electrical circuit. A 250 mm long tantalum tube, with external and internal diameters of 2 mm and 1 mm, respectively, is used as an oven device to feed the atoms of the desired element to the ion source (see figure 6.19).



Figure 6.19: The SPES surface ion source.

At one end of the oven a calibrated quantity of atoms is introduced and hermetically sealed in; the other end is accurately connected to the transfer line and thus to the ion source. During operation the flux of atoms that reach the ion source by effusion is regulated controlling the heating current (passing through the tubular oven device) by a dedicated power supply [10]. In this work ionization efficiency measurements for Cs and Rb were performed using calibrated Cs/Rb samples housed inside the aforementioned oven tube. We underline the importance of Cs and Rb for the SPES project, since they are the only alkali metal isotopes produced by fission in the UCx SPES target. At the beginning of each measurement the ion source temperature was rapidly increased up to 2000 °C in the ionizing cavity. Then the oven was gradually heated (reaching temperatures of approximately 2000 °C, with heating currents up to 70 A) in order to melt the dehydrated salts, dissociate the nitrate molecules, and vaporize the Cs and Rb atoms. The ion beam current, kept between 1 and 3  $\mu$ A for about 5 hours, was continuously recorded until the calibrated sample completely petered out. Both for Cs and Rb the ionization efficiency was calculated as the ratio of the integrated number of detected ions, to the total number of atoms in the calibrated sample. Experimental measurements are proposed in the following.



Figure 6.20: Temperature dependence of the Cs ionization efficiency (Ta hot-cavity).



Figure 6.21: Ionization efficiency measurements with a Ta hot-cavity.



Figure 6.22: Ionization efficiency measurements with a Re hot-cavity.

For the ionization efficiency testing of the plasma ion source, the cathode current was gradually increased up to 310 A (the lower heating current capable to produce an electron density sufficient to start the ionization process, and so to produce an ion beam), and the anode voltage was set at 150 V.



Figure 6.23: The test bench used to characterize the plasma ion source.



Figure 6.24: Example of mass scan (main region, cathode current at 370 A).

In the meantime the magnetic field in the anode region was stabilized at approximately 300 G, and the Ar gas pressure before the leak was slowly regulated at 1 bar, the leak reference value. In this way the flux of Ar atoms entering the ion source was univocally defined  $(1\cdot10^{14} \text{ atoms/s})$ .

At this point, the procedure for the ionization efficiency measurement started. The ion beam was opportunely focused and monitored in the Faraday cup of the diagnostic box 1 (FC1). Once stabilized the total beam current, its intensity value was opportunely stored. Then the ion beam was focalized in the Faraday cup placed in the diagnostic box 2 (FC2), and an accurate mass scan was performed in order to identify the elements composing the beam and their partial ion beam currents (see Figure 6.24). Once determined the current percentage associated to the  $Ar^{1+}$  ions, it was multiplied by the total beam current value previously stored by the FC1, obtaining in this way the total  $Ar^{1+}$  current. It is fundamental to refer to the FC1 when a precise absolute value of the beam current is needed (that is the case of ionization efficiency measurements), since the FC2 is placed after the system of slits of the Wien filter, where part of the beam is dissipated (Wien filter transmission efficiency lower than 100%). At this point the  $Ar^{1+}$  ionization efficiency was easily calculated as the ratio of the total  $Ar^{1+}$  current and the flux of Ar atoms entering the ion source. The efficiency measurement was repeated for different cathode current values (and consequently, for different electron density values), up to 370 A.



*Figure 6.25*: Ar ionization efficiency measurements for cathode currents ranging from 310 A ( $T_{cathode} \approx 1800 \text{ °C}$ ) up to 370 A ( $T_{cathode} \approx 2200 \text{ °C}$ ).

Figure 6.25 presents the three sets of measurements performed at Legnaro National Laboratories (LNL). The  $Ar^{1+}$  ionization efficiency increases gradually with the cathode current, reaching the saturation value of approximately 8%, when the cathode current is equal to 360 A and the cathode temperature level is approximately equal to 2200 °C. Efficiency measurements with other gases (mainly Kr and Xe) are currently ongoing at LNL.

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