

High-Efficiency Target-Ion Sources for RIB Generation*

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Abstract

A brief review is given of high-efficiency ion sources which have been developed or are under development at ISOL facilities which show particular promise for use at existing, future, or radioactive ion beam (RIB) facilities now under construction. Emphasis will be placed on those sources which have demonstrated high ionization efficiency, species versatility, and operational reliability and which have been carefully designed for safe handling in the high level radioactivity radiation fields incumbent at such facilities. Particular attention will be given to the sources which have been selected for initial or future use at the Holifield Radioactive Ion Beam Facility now under construction at the Oak Ridge National Laboratory.

1.0 INTRODUCTION

During the past few years, world-wide interest has developed in the use of radioactive ion beams to address questions concerning the structure of the nucleus and on the nucleosynthesis burn cycles which power stellar processes and which are responsible for heavy element formation. Because many of the nuclear reactions important in nuclear, nuclear structure, and astrophysics are inaccessible to experimental study using stable/stable beam/target combinations, they can only be studied with accelerated radioactive ion beams (RIBs). Such beams, therefore, offer unique opportunities to further our knowledge of the nucleus and the energetics of our solar system. As a consequence, world-wide interest has led to the development and proposed development of RIB facilities in Asia, Europe, and North America [see, e.g., the facilities listed in Ref. 1]. Of the several facilities listed in Ref. 1, only the RIB facility at Louvain-la-Neuve is presently operational [2], while the Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory has been funded and is now under construction [3].

Our present knowledge of the properties of nuclei far from the region of beta stability can be, in part, attributed to the successful development of reliable, long-lived, and efficient ISOL ion sources with fast release properties. Many ion source developments

have been made over the past few decades for ISOL applications, many of which have been recently included in reviews by Ravn [4] and Van Duppen, et al. [5]. For information on the latest developments in on-line mass separators and target-ion sources associated with their use, readers are encouraged to consult the proceedings of the most recent electromagnetic isotope separator (EMIS) conferences [6,7]. In the present paper, the requirement of brevity places limitations on the number and types of sources which can be included; therefore, the review will be restricted to examples of sources which represent state-of-the-art developments of a particular source type which have demonstrated high ionization efficiencies and reliable performances at ISOL facilities.

2.0 CANDIDATE ION SOURCES FOR RIB GENERATION

The ISOL technique is complicated by high-temperature physics, chemistry, metallurgy, diffusion, and surface adsorption processes which take place in the target-ion source; all of these processes add to the delay times which result in losses of the short-lived radioactive species of interest. For RIB generation, the source should ideally exhibit the following properties: high efficiency; high temperature operation in order to minimize the diffusion times from the target and residence times on the surface; low energy spreads; chemical selectivity; flexibility for adaptation to different temperature ranges and modes of operation; target temperature control; long lifetime; and stable electrical and mechanical properties. The source should, as well, be designed for safe and expedient insertion/removal from the ISOL facility to permit changing of the target material and source repairs as required in high radioactivity radiation level fields.

2.1 Electron Beam Plasma Ion Sources

The FEBIAD ion source. The forced electron beam induced arc discharge (FEBIAD) source, developed at GSI by Kirchner [8-10], differs from conventional plasma discharge sources in that it does not require a minimum pressure for stable operation

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(commonly referred to as the Langmuir criterion for stable discharge). The source operates at pressures of more than one order of magnitude lower than the Nielsen plasma discharge source [11] as clearly demonstrated by Kirchner and Roeckl [12]. The source is well suited for ISOL applications which involve the use of heavy ions to produce the radioactive species of interest in that it operates stably and efficiently over a pressure range of $\sim 1 \times 10^{-5}$ to $\sim 2 \times 10^{-4}$ Torr at elevated temperatures. Several versions of the source have been developed at GSI (see, for example, Refs. 8-10 for specific details on the respective sources); these sources differ in their materials of construction, ionization chamber volumes and achievable target temperatures, depending on the particular source geometry. The efficiency of the FEBIAD ion source is quite high for slow moving heavy ions; for low mass, fast moving atoms with high ionization potentials, the source is not as impressive. For example, the measured ionization efficiencies for the noble gas elements, as reported in Ref. 13, are, respectively, Ne: 1.5%; Ar: 18%; Kr: 36%; and Xe: 54%. The following equation is found to be useful in approximating the ionization efficiencies η for the noble gases:

$$\eta_{\text{calc}} = \frac{4\langle\ell\rangle D_0 N_e}{A_0} \left(\frac{\pi M_i}{8kT_i} \right)^{1/2} \exp \{-I_p / kT_e\}$$

$$/[1 + \frac{4\langle\ell\rangle D_0 N_e}{A_0} \left(\frac{\pi M_i}{8kT_i} \right)^{1/2} \exp \{-I_p / kT_e\}] \quad (1)$$

Where $\langle\ell\rangle$ = average path length for a particle in the plasma; D_0 = constant (cm^2/s); A_0 = emission area of the source; k = Boltzmann's constant; T_i = ion temperature; T_e = electron temperature; I_p = ionization potential; N_e = number of electrons in the valence shell of the atom with a given I_p ; and M_i = mass of species. The following values are used for terms in Eq. 1 when estimating ionization efficiencies for the FEBIAD ion source: $\langle kT_e \rangle = 3$ eV; $T_i = 2273^\circ\text{K}$; and $4\langle\ell\rangle D_0 / A_0 = 5.39 \times 10^3$ cm/s .

The CERN-ISOLDE ion source. The CERN-ISOLDE on-line source is similar in principle to the FEBIAD source. The design features of several versions of the source have recently been described by Sundell and Ravn [14]. The CERN ISOLDE target/ion source has been utilized extensively for the production of short-lived radioactive species and has been cleverly engineered to enable remote installation and removal from the facility as required for safe handling in high-radiation-level fields. Electron impact ionization sources of the FEBIAD-CERN-ISOLDE type are quite efficient for low-ionization-potential elements and elements which are heavy and,

therefore, move slowly through the ionization volume of the source. The ionization efficiencies for these atoms are close to those measured for the FEBIAD source. For example, the maximum efficiency recorded for Xe is 56% [15]. However, those sources do not appear to ionize low-mass elements or molecular materials efficiently, particularly those with high ionization potentials.

The high-temperature version of the CERN ISOLDE source [16] has been selected as the first source to be used for the generation of radioactive ion beams at the HRIBF because of its low emittance, $(\sim 2 \pi \text{ mm.mrad}) (\text{MeV})^{1/2}$ relatively high ionization efficiency, and capability for producing a broad range of radioactive species. Of equal importance, the source has been cleverly engineered for remote installation, removal and servicing as required in safe handling of highly radioactive contaminated sources, source components, and ancillary equipment. The source design also permits easy modification to lower temperature versions and conversion from electron impact ionization to either thermal or positive and negative surface ionization sources.

The HRIBF version of the CERN-ISOLDE source is described in Ref. 16. The high-temperature target and ionization chamber of the source are shown schematically in Figs. 1 and 2. A collimated ion beam from the ORIC will pass through a thin Re window where it interacts with the refractory target material chosen for the production of the desired radioactive beam. The Ta target material reservoir is lined with Ir or Re metal as is the beam transport tube and internal surfaces of the source.

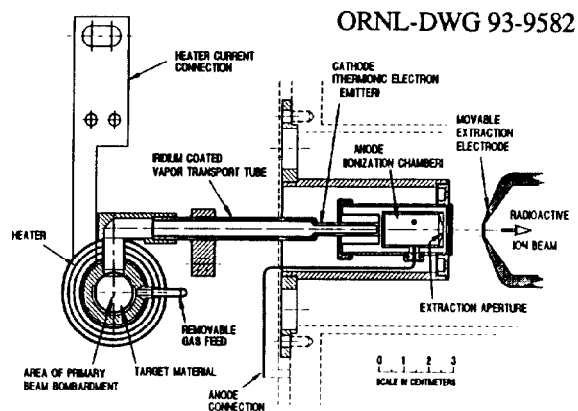


Figure 1. Cross-sectional side view of the HRIBF high-temperature target/ion source showing the target, vapor transport tube, and ionization chamber of the source [16].

The thickness of the target is chosen so that the projectile has an energy spread within the target medium which approximates that required for optimum radioactive species production. The

unreacted beam exits the target through a second Re window, then strikes a cooled C beam stop. This technique reduces the power deposited in the target and thereby simplifies temperature control problems.

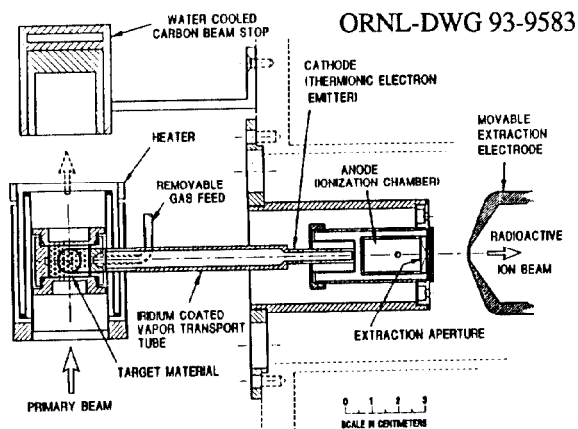


Figure 2. Cross-sectional top view of the HRIBF high-temperature target/ion source showing the target, vapor transport tube, and ionization chamber of the source [16].

The target reservoir is positioned within the inner diameter of a series-connected, resistively heated, three-cylinder, Ta tube. The reservoir can be heated to temperatures exceeding 2100°C by passing a current through the tubular structure. The power required to heat the assembly to 2100°C is estimated to be 5.5 kW (11 V at 500 A). Temperature control will be maintained within $\pm 2^\circ\text{C}$ by use of feedback circuitry driven by a two-color pyrometer to adjust the current through the heater.

The electron emitter cathode is also made of Ta and is resistively heated to thermionic emission temperatures, $\sim 2125^\circ\text{C}$. The electron beam, typically ≥ 250 mA, is accelerated through a potential difference of 200-300 V to the perforated anode plate where it passes into the cylindrical cavity of the anode structure and ionizes the gaseous material. Collimation of the electron beam is effected by adjusting the coaxially directed solenoidal magnetic field so as to optimize the ionization efficiency of the species of interest. The cathode power required to achieve thermionic emission temperature will be ~ 2 kW (400 A at 5 V).

The electron beam generated plasma (EBPG) ion source. The electron beam generated plasma (EBGP) ion source, developed for use at the OASIS facility by Nitschke [17] has demonstrated high efficiencies and very high temperature operation ($\sim 2700^\circ\text{C}$). Table 1 compares the efficiencies for a number of elements, as calculated from Eq. 1, with those measured from the EBGP, FEBIAD, and CERN-ISOLDE ion sources.

Table 1. Comparisons of calculated and experimentally measured ionization efficiencies η for electron beam plasma ion sources. Estimated ionization efficiencies were calculated by using Eq. 1.

Z	Element	$I_p(\text{eV})$	N_e	$\eta_{\text{calc}}(\%)$	$\eta_{\text{exp}}(\%)$	Ref.
10	^{20}Ne	21.56	8	2.0	1.6	10
18	^{40}Ar	15.76	8	16.6	19	10
24	^{54}Cr	6.77	1	37.0	>20	10
26	^{57}Fe	7.90	2	45.2	30	10
32	^{76}Ge	7.90	4	65.6	41	10
36	^{84}Kr	14.00	8	34.1	35	10
36	^{84}Kr	14.00	8	34.1	36	17
46	^{100}Pd	8.33	18	89.5	>25	10
47	^{107}Ag	7.58	1	38.7	47	10
47	^{109}Ag	7.58	1	38.9	50	10
50	^{116}Sn	7.34	4	74.0	53	10
50	^{124}Sn	7.34	4	74.6	54	10
54	^{129}Xe	12.13	8	54.6	52	10
54	^{132}Xe	12.13	8	54.8	53	10
54	^{132}Xe	12.13	8	54.8	56	15
79	^{197}Au	9.23	1	32.9	50	10
82	^{208}Pb	7.42	4	78.8	52.8	10
83	^{209}Bi	7.29	5	82.9	68.3	10

2.2 Thermal Ion Sources

At high temperatures, collisions between gas particles may produce ionization provided that their relative energies exceed the first ionization of the atoms or molecules that make up the gas. For the case of a monoatomic gas, a fraction of the total number of gas particles will be in various stages of ionization at thermal equilibrium. For such reactions, the law of mass action can be used to derive an expression for the ionization efficiency as a function of temperature and pressure [18]. The first reported applications of this technique are described in Refs. [19] and [20]. Such sources are relatively efficient for elements with ionization potentials $I_p \leq \sim 7$ eV.

Ionization efficiencies for a number of elements, including Ca, Sr, Nb, and La, all of the lanthanides, and U, Pu, and Cm of the actinides have been measured by use of the thermal ionization source described in Ref. 20. The efficiencies for ionizing Ti, Cr, Fe, Ni, Sr, Pb, and U have been measured by Kirchner [21]. The source can be very efficient as indicated. A thermal ion source has also been designed for use at the TRISTAN facility [22]. A thermal source is also now under design at the HRIBF. The vapor transport tube, made of W or Ta, will be resistively heated to $\sim 2100^\circ\text{C}$. In addition, the thermal ionization region of the vapor transport tube will be heated to $\sim 2600^\circ\text{C}$ by electron bombardment.

2.3 Surface Ionization Sources

Positive surface ionization. For thermodynamic equilibrium processes, the ratio of ions to neutrals that leave an ideal surface can be predicted from Langmuir-Saha surface ionization theory.

Positive surface ionization sources. Positive surface ionization sources are quite simple and easy to operate. The principle has been used at CERN-ISOLDE [23] and TSIOL facility [24] to generate ion beams from low-ionization-potential elements. Although they have limited applications in terms of species, the process is highly chemically selective. The CERN-ISOLDE source can be easily retrofitted to accommodate the positive surface ionization source mode of operation, as demonstrated at INS to ionize ^{38}K [25]. A positive surface ionization source will be used as complementary to the CERN-ISOLDE source at the HRIBF. The ionizer and vapor transport tube will be Ir coated tantalum and heated resistively to $\sim 1100^\circ\text{C}$.

Negative surface ionization. For thermodynamic equilibrium processes, the ratio of ions to neutrals which leave an ideal surface can be predicted from Langmuir-Saha surface ionization theory appropriate for negative ion formation.

The negative form of surface ionization is also highly chemically selective and, therefore, can be used for the generation of high-electron-affinity elements such as the group VIIA (halogens). Unfortunately, there is limited availability of a wide variety of stable, low work materials. LaB_6 is the most frequently used low-work-function surface ionizer, having a work function $\phi = \sim 2.7$ eV for polycrystalline and $\phi \approx 2.36$ eV for single crystalline material.

Negative surface ionization sources. An on-line form of the negative surface ionization source has been developed at CERN-ISOLDE, which is equipped with a LaB_6 surface ionizer [26]. The CERN-ISOLDE-type source can easily be retrofitted with a LaB_6 ionizer and used to efficiently ionize high-electron-affinity elements as clearly evidenced by the successful application described in Ref. 26. A spherical-geometry negative-surface source is planned for use at the HRIBF.

2.4 ECR Ion Sources

Electrons moving along the field lines of an external magnetic field of flux density B can be resonantly accelerated by the electric field associated with microwave radiation of the proper frequency which matches the electron-cyclotron resonance condition, $\omega_{\text{ECR}} = \omega_{\text{rf}} = Be/m$. The region where the ECR condition is met is referred to as the ECR zone. Several ion sources, based on the ECR

principle, have been developed for ISOL applications including those described in References 27-29.

The ECR ion source is superior in terms of ionization efficiencies for low mass, high ionization potential elements and in terms of their capabilities for producing multiply charged ion beams. Another principal advantage of the ECR ion source is that it does not rely on a negatively biased hot cathode for generating and sustaining the plasma which limits the lifetimes of conventional electron beam plasma sources due to physical sputtering of the cathode material. The principal disadvantage of the source is that, in its present state of development, the discharge chamber can only be operated at modestly high temperatures which severely limits the number of species that the source can be used to process. The ionization efficiencies for condensable materials will be less than those for more volatile elements. However, due to the high probability for ionization during transit through the ionization volume of the source, there is evidence that the efficiencies for condensable materials may be relatively high, as well. The ECR ion source, however, is particularly well suited for the processing of highly volatile or gaseous materials and usually out performs electron beam plasma sources for low atomic number species. The prototype on-line ECR ion source was first developed at Karlsruhe [27] for ISOL applications; the source has demonstrated efficiencies for C of 10%; for N up to 27%; for O up to 55%, for Ne up to 31% and Xe up to 65%. This source was duplicated at TRIUMF [28]. Ionization efficiencies of 18% for N, 37% for C and 14 % for Ne and 35% for Ar have been measured with the on-line ECR ion sources at Louvain-la-Neuve [29-31]. The ionization efficiencies for the ECR ion source are sensitively dependent on the pressure in the discharge chamber as evidenced by the measurements made at Louvain-la-Neuve. These sources are clearly more efficient for low mass, high ionization potential elements than conventional, hot cathode electron beam plasma type sources such as the FEBIAD and CERN-ISOLDE type ion sources. On the other hand, the ionization efficiencies for Xe in this source are close to those recorded at CERN-ISOLDE [15] and GSI [13] for Xe ($\sim 54\%$). These sources are particularly impressive for difficult to ionize elements such as He and Ne. A compact, single stage, permanent magnet ECR ion source is now being developed for on-line use at GANIL for potential RIB applications [32].

2.5 Plasma Sputter Negative Ion Sources

The technique of sputtering a surface covered with a fractional layer of a highly electropositive adsorbate material such as cesium has proved to be a universal method for generating atomic and molecular negative ion beams from most chemically active

elements. Positive ion beams, usually formed by either direct surface ionization of a group IA element or in a heavy noble gas (Ar, Kr, or Xe) plasma discharge seeded with alkali metal vapor, are accelerated to energies between a few hundred eV and several keV where they sputter a sample containing the element of interest. A small fraction of the sputter ejected particles leave the adsorbate covered surface as negative ions and are accelerated through an extraction aperture in the source. Several sources predicated on this principle have been developed, some of which are described in Ref. 33. Sources based on this principle are particularly appealing for applications involving the postacceleration of RIBs with tandem accelerators such as at the HRIBF. In particular, sources which use a plasma to sputter the sample [34-37] are especially attractive because this technique assures uniform sputtering and, consequently, good overlap of the bombarding species and the material containing the radioactive ion beam.

Figure 3 displays a schematical representation of a plasma sputter source now under design for use at the HRIBF. The source will be housed in the source vacuum envelope as the CERN-ISOLDE source.

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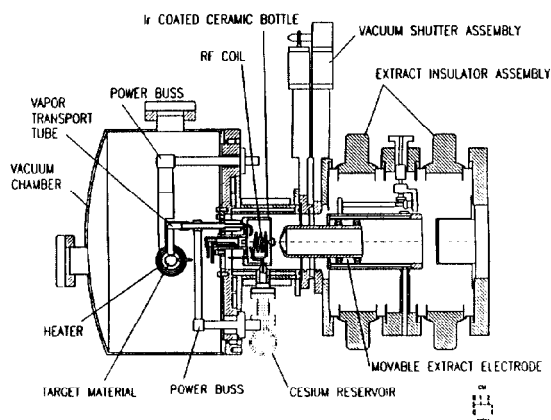


Figure 3. Schematic drawing of a plasma sputter negative ion source now under design for use in the HRIBF. The source will be complementary to the CERN-ISOLDE electron beam plasma source displayed in Figs. 1 and 2.

Radioactive species from the target will be transported at high temperatures through the vapor transport tube into the plasma discharge chamber where the vapor will be condensed on the cold cathode surface. A Xenon plasma, seeded with cesium from an external oven, will be ignited either by a filament or a rf coil. The radioactive ion beam will be formed by sputter ejection of atoms or molecules from the negative biased spherical geometry sputter probe covered with a partial layer of cesium adsorbate material. The

double sheath surrounding the negatively biased sputter probe (spherical radius 30 mm and diameter $\phi = 12.5$ mm), which is maintained at a variable voltage (0-1000 V) relative to housing, serves as the acceleration gap and lens for focusing the negative-ion beam through the exit aperture (diameter $\phi = 3$ mm). At this point, the ion beam is further accelerated to energies up to 50 keV prior to mass analysis. The efficiencies of several negative ion species have been estimated by Tsuji and Ishikawa [37]. Their results are shown in Table 2.

Table 2. Estimates of the probability for negative ion formation by xenon sputtering at optimum cesium coverage (Ref. 37)

Negative Ion	C-	Si-	Cu-	Ge-	Mo-	Ta-	W-
Probability (%)	18.3	15.6	12.1	13.6	(0.52)	(1.59)	8.07

The emittance of the source is quite good ($\sim 8\pi$ mm.mrad $[\text{MeV}]^{1/2}$) as measured by Mori for the compact plasma sputter described in Ref. 36 when operated with mA beams of Cu-. The emittance is much lower than the acceptance of the 25-MV tandem accelerator [38] and, therefore, should be easily transported to the terminal stripper in the machine.

2.6 Multi-photon Resonance Ionization Sources

Multi-photon resonance ionization spectroscopy (RIS) has been utilized for a number of years to selectively ionize atoms [39]. The scheme is, in principle, very simple; the difficult challenge is to find the most efficient scheme for ionizing a particular atom. Two or more lasers are used to selectively ionize the species of interest. Because the ionization process requires the precise matching of the photon energies to each of the energy levels of the particular atom, the process is resonant and uniquely species selective; the RIS scheme is, therefore, highly discriminatory against potential contaminants. Thus, RIS offers a means of generating fully ionized, isotopically and isobarically pure RIBs. Approximately 80% of the elements in the periodic chart can be resonantly ionized with the RIS technique with existing lasers.

Several groups are actively developing RIS laser ion sources for future ISOL applications. The RIS scheme has been applied to resonantly ionize atoms released from an ISOL target and effused through tubular or insulated cavities which are operated at high temperatures to prevent condensation. Two or three tunable dye laser beams are collinearly focused through the cavity with their wave lengths chosen to resonantly ionize the species of interest. This source is handicapped by the fact that some of the atoms are in excited states or may be thermally or surface

ionized due to the high temperatures required to prevent condensation on the walls of the cavity. The otherwise chemical selectivity character of the RIS process is, therefore, compromised. Nevertheless, the hot cavity-RIS technique has been used off-line by Andreev, et al. to selectively ionize Sr to efficiencies of 17% [40] and by Ames, et al. to ionize Tc to efficiencies of 13% [41]. These groups both employed Cu vapor lasers which operate at high repetition rates to pump tunable dye lasers. This RIS scheme has been duplicated by Alkhazov, et al. to study the ionization of the rare earth elements Yb, Ho, Tl, and Sm [42]. Mishin, et al. used the RIS technique to determine the ionization efficiencies for Sn, Tm, Yb, and Li [43]. Efficiencies up to 15% were recorded for Yb. In these studies, it was found that the initial chemical selectivity factor for Tm, compromised by surface and thermal excitation/ionization processes, could be increased from 10 to 10000 by the proper choice of cavity material and by reducing the cavity temperature; the gain in selectivity was made by suppressing thermal and surface ionization processes. Furthermore, it was found that another factor of 10 could be gained by using gated detection techniques on the bunched beam. The laser desorption technique has been used to desorb Pt and Au which were ion implanted into samples and to study the Pt-Au isobar contamination problem by using the RIS technique. Ionization efficiencies for this study only reached 5×10^{-3} [44]. However, the desorption efficiencies for the process reached 60%.

3.0 Conclusions

ISOL ion source development continues to be driven by needs for sources with improved chemical selectivity, high duty factors, and more universal species capabilities. Despite the fact that electron beam plasma ion sources have poor chemical selectivity characteristics, they have a decided advantage in that they are closer to being universal than other ISOL sources that have been developed to date. Of the electron beam sources, the CERN-ISOLDE source is very appealing for RIB generation applications.

Although ECR ion source development has made considerable progress, this source type still suffers from the fact that no really satisfactory solution to the low wall temperature/condensation problem has been found; as a consequence, the source, in its present state of development, has very limited species capabilities for ISOL applications. However, there is ample incentive to solve this problem because of the obvious advantage of the ECR source over conventional source types in terms of ionization efficiency.

The results from the testing of RIS laser sources has been rather encouraging and it is expected that

this technology will rapidly advance in the future. The RIS technique offers the idealistic prospect of eliminating the need for expensive isotope and isobar electromagnetic separation devices.

Plasma or cesium sputter ion sources offer another possibility for the efficient formation of negative ion beams from high electron affinity elements. Sources, based on this well developed technology, do not suffer from poisoning effects as do direct negative surface ionization sources and are very appealing for use at tandem accelerator based RIB facilities such as the HRIBF.

Thermal and surface ionization sources have reached a certain degree of maturity in their development but still play important roles for the efficient generation of ion beams from specific elements. Surface ionization sources, in particular, offer a high degree of chemical selectivity and are simple and easy to operate.

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