# ION SOURCES FOR INITIAL USE AT THE HOLIFIELD RADIOACTIVE ION BEAM FACILITY

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The Holifield Radioactive Ion Beam Facility (HRIBF) now under construction at the Oak Ridge National Laboratory will use the 25-MV tandem accelerator for the acceleration of radioactive ion beams to energies appropriate for research in nuclear physics; negative ion beams are, therefore, required for injection into the tandem accelerator. Because charge exchange is an efficient means for converting initially positive ion beams to negative ion beams, both positive and negative ion sources are viable options for use at the facility; the choice of the type of ion source will depend on the overall efficiency for generating the radioactive species of interest. A high-temperature version of the CERN-ISOLDE positive ion source has been selected and a modified version of the source designed and fabricated for initial use at the HRIBF because of its low emittance, relatively high ionization efficiencies and species versatility, and because it has been engineered for remote installation, removal and servicing as required for safe handling in a high-radiation-level ISOL facility. Prototype plasma-sputter negative ion sources and negative surface-ionization sources are also under design consideration for generating negative radioactive ion beams from high-electron-affinity elements. The design features of these sources and expected efficiencies and beam qualities (emittances) will be described in this report.

KEY WORDS: Ion sources, radioactive-ion-beam, radioactivity, ISOL

## **1 INTRODUCTION**

Many of the nuclear reactions important for the understanding of nuclear structure and astrophysical phenomena are inaccessible to study using stable projectile/stable target combinations; they, therefore, can only be studied with accelerated radioactive ion beams (RIBs). As a consequence of these unique research opportunities, a number of RIB facilities have been proposed throughout the world,<sup>1</sup> including the Holifield Radioactive Ion Beam Facility (HRIBF)<sup>2,3</sup> now under construction at the Oak Ridge National Laboratory.

The HRIBF has as its objective the production and acceleration of short-lived, proton-rich nuclei for the study of nuclear structure physics and astrophysics. In the HRIBF concept, intense light ion beams from the Oak Ridge Isochronous Cyclotron (ORIC) will be used to produce radioactive atoms in a thick-target, ISOL-type ion source mounted on a high-voltage platform which will serve as a second injector for the 25-MV tandem accelerator.

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Since the tandem accelerator requires negative ion beams, positive ion beams must be converted to negative ion beams through charge exchange prior to their injection into the tandem accelerator. Among the proposed initial species are radioactive beams of C, O, F, Na, Si, P, S, Cl, K, Cu, Ga, Ge, As, Se, Br, and Rb. An important capability of the HRIBF will be the ability to accelerate all of these beams to energies of at least 5 MeV/amu.

The success of this project will be critically dependent on the choice, design, and performance of the target/ion sources. For radioactive ion beam generation, these sources must operate stably for extended periods of time at elevated temperatures (>2000°C) and be designed to facilitate expedient and remote (if necessary) installation, removal, and servicing, as required for safe handling of high-level, radioactively contaminated sources, source components, and ancillary equipment. The selection of a particular target/ion source or sources is critically important since their respective performances determine the intensity, beam quality, and the number of radioactive beams that can be provided for experimental use. High-ionization-efficiency sources are clearly desirable because of the low production rates of the radioactive species of interest, release losses which may be incurred through trapping within the target, and surface chemical reactions during transport from the target and through the source. 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 surfaces; 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. While there are no sources which simultaneously meet all of these criteria, a number of sources have been developed at ISOL facilities which meet some of the requirements noted above. We have selected and designed an electron-beam plasma-type source patterned after the CERN-ISOLDE source<sup>4</sup> for initial use at the HRIBF. (The HRIBF source is also described in Reference 5.) This source was chosen because of a number of reasons, including the following: 1) it is competitively efficient; 2) it has a wide range of species capabilities; 3) it has demonstrated reliability over many years of operation at the CERN-ISOLDE facility; 4) the target temperature can be controlled independently of the discharge parameters of the source; 5) the basic geometry can be readily adapted to other types of sources; 6) it has been engineered for safe removal and installation in the high-radiation-level fields present at ISOL facilities; 7) the source has a low emittance ( $\sim 2\pi$  mm.mrad MeV<sup>1/2</sup>).

High-efficiency negative-ion sources are obviously preferred for applications involving the postacceleration of RIBs with tandem accelerators such as at the HRIBF. We have designed a plasma-sputter negative-ion source specifically for generation of ion beams from high-electron-affinity elements, e.g., the group IV-A, V-A, VI-A, and VII-A elements. The source will be housed in the vacuum envelope of the HRIBF source. The design aspects and expected performance of this source type for a selected number of elements will also be presented. A complementary negative-surface-ionization source has also been designed for alternate use with the plasma-sputter negative ion source in generating RIBs from certain members of the group VI-A and VII-A elements; this source will also be briefly described.

## 2 THE HRIBF POSITIVE ION SOURCE

The CERN-ISOLDE on-line source<sup>4</sup> is in the class of electron beam plasma ion sources and, therefore, similar, in principle, to the forced-electron-beam-induced-arc-discharge (FEBIAD) source developed at GSI.<sup>6–8</sup> Sources of this type differ from conventional plasma discharge sources in that they do not require a minimum pressure for stable operation (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 plasmadischarge source<sup>9</sup> as reported in Reference 10. The CERN-ISOLDE source is well suited for ISOL applications because, unlike the FEBIAD ion source, the target temperature can be independently controlled. Both types of sources operate stably and efficiently over a pressure range of  $\sim 1 \times 10^{-5}$  to  $2 \times 10^{-4}$  Torr at elevated temperatures.<sup>10</sup>

#### 2.1 Ionization Efficiencies

The ionization efficiencies of the CERN-ISOLDE and FEBIAD ion sources are quite high for slow moving heavy ions; for low mass, fast moving atoms with high ionization potentials, the efficiencies are not as impressive. This effect can be readily observed by comparing the ionization efficiencies of the noble gas elements using the FEBIAD ion source [11]; the efficiencies for these elements were measured to be: Ne: 1.5%; Ar: 18%; Kr: 36%; and Xe: 54%. The ionization efficiencies for the noble gas elements, using the CERN-ISOLDE source, are close to those measured for the FEBIAD source. For example, the maximum efficiency recorded for the CERN-ISOLDE source with Xe is 56%.<sup>12</sup>

The following empirical equation has been found to be useful in approximating ionization efficiencies  $\eta$  for noble gases in sources of this type:

$$\eta_{\text{calc}} = \frac{4 < \ell > D_o N_e}{A_o} \left(\frac{\pi M_i}{8kT_i}\right)^{1/2} \exp\left\{-I_p / < kT_e > \right\} / \left[1 + \frac{4 < \ell > D_o N_e}{A_o} \left(\frac{\pi M_i}{8kT_i}\right)^{1/2} \exp\left\{-I_p / < kT_e > \right\}\right].$$
(1)

In Equation 1, < l > = average path length for a particle in the plasma;  $D_o$  = constant (cm<sup>2</sup>/s);  $A_o$  = 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 Equation 1 when estimating ionization efficiencies for the FEBIAD ion source:  $< kT_e > = 3.029$  eV;  $T_i = 2273$ K; and  $4 < l > D_o/A_o = 5.39 \times 10^5$  cm/s. Table 1 compares the efficiencies for a number of elements as calculated from Equation 1, with those measured by using the FEBIAD, CERN-ISOLDE, and the electron-beam-generated plasma (EBGP) source.<sup>13</sup>

## 2.2 Source Design Features

In designing the HRIBF target/ion source, particular emphasis was placed on the materials of construction, the thermal transport properties, the size and geometry of the target, and the number of species that the source can be used to process. Equal attention was given to design features which facilitate assembly and disassembly of contaminated source components to avoid potential radiation hazards to personnel during maintenance periods.

The target/ion source assembly, shown schematically in Figures 1-3, incorporates many of the engineering features utilized in the CERN ISOLDE design.<sup>4</sup> The source assembly has been engineered to enable remote installation and removal in high-radiation fields and the transport of contaminated sources and components to and from remote servicing and storage areas. To install the source assembly, for example, the assembly is lowered by means of a remotely controlled overhead hoist mechanism onto locator pins associated with a linear motion carriage. The carriage is then pushed forward and clamped against the vacuum interface flange by a remotely controlled pneumatic actuator mechanism. The source assembly plugs into the interface flange, during which all electrical, gas feed, coolant, vacuum, and temperature monitoring connections are mated together. A schematic illustration of the source in the operational position is shown in Figure 1. Following this operation, the vacuum seal between the beam entrance port on the source vacuum chamber and beam line from the ORIC is also made by remote actuation of a similar pneumatic clamping device. The vacuum chambers can then be pumped down or vented by remote control of the pumps and valves associated with the source and ORIC beam lines. To remove the source, the procedure is reversed. For servicing and storage, the source is simply lifted from the linear-motion carriage by means of a remotely controlled electro-mechanical handler and transported to a separated shielded work or storage area.



FIGURE 1: Side cross-sectional view of the HRIBF target/ion source assembly in the operational position.



FIGURE 2: Cross-sectional side view of the HRIBF high-temperature target/ion source showing the target, vapor-transport tube, and ionization chamber of the source.



FIGURE 3: Cross-sectional top view of the HRIBF high-temperature target/ion source showing the target, vaportransport tube, and ionization chamber of the source.

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The high-temperature target and ionization chamber of the source are shown schematically in Figures 2 and 3. Following vacuum pump down, the source is outgassed by incrementally adding power to both the target reservoir and to the cathode until the source achieves a steady-state vacuum level at the anticipated operational temperature. Collimated <sup>1</sup>H, <sup>2</sup>D, <sup>3</sup>He, and <sup>4</sup>He ion beams from the ORIC will pass through a thin Re window where they will interact 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 are beam transport tubing and internal surfaces of the source to reduce the residence times of chemically active elements during adsorption. 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.

The target reservoir is positioned within the inner diameter of a series-connected, resistively heated, triaxial Ta tube. The reservoir can be heated to temperatures exceeding 2000°C by passing a current through the tubular structure. The power required to heat the assembly to 2000°C is measured to be  $\sim 2.25 \text{ kW}$  (4.5 V at 500 A). Temperature control will be maintained within  $\pm 5^{\circ}$ 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,  $\geq 2125^{\circ}$ C. The electron beam, typically  $\geq 250$  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 fields so as to optimize the ionization efficiency of the species of interest. The cathode power required to achieve thermionic emission temperature will be  $\sim 1.2$  kW (400 A at 5 V). The total power required to heat the target and cathode to the temperatures listed above and to ionize the vaporous material transported from the target to the ionization chamber of the source will be of the order of 3.5 kW.

The ion extraction electrode system for the source was iteratively designed by use of the computer code described in Reference 14, which includes space charge and plasma optical effects. The extraction gap can be varied in order to optimally transport ion beams over a wide energy range as required for optimally converting positive ion beams to negative ion beams through charge exchange.

## 3 THE PLASMA-SPUTTER NEGATIVE ION SOURCE

The adsorption of less than a monolayer of a highly electropositive adsorbate material on the surface of a sample undergoing particle bombardment greatly enhances the probability for secondary negative ion formation.<sup>15</sup>

In the prescription of Nørskov and Lundqvist,<sup>16</sup> the probability for negative ion formation during sputtering can be represented by the following simple energy-dependent relation:

$$\eta_i = \frac{2}{\pi} \exp\left[-\beta \sqrt{M_2} \left\{\phi(\sigma) - E_A + V_i\right\} / \sqrt{2E_2} \cos\theta\right]$$
(2)

In Equation 2,  $\phi$  is the work function of the surface which is a function of the relative adsorbate coverage  $\sigma$ ,  $E_A$  is the electron affinity of the ejected particle of mass  $M_2$  and energy  $E_2$ ,  $V_i$  is the image potential induced in the surface by the escaping ion,  $\theta$  is the polar angle of the sputtered ion with respect to the surface normal and  $\beta$  is a constant. In Equation 2,  $\sqrt{2E_2/M_2} \cos \theta = v_{\perp}$  is the component of the velocity of the escaping particle perpendicular to the sample surface.

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. In addition to being versatile in terms of species, sources based on this concept are simple in design, easy to operate, and generally have long lifetimes. Because of these factors, such sources are used extensively in most tandem electrostatic accelerator heavy ion physics research laboratories, as well as for use in a growing number of other applications, including high-energy ion implantation and tandem accelerator mass spectrometry. 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. The presence of a fractional layer of a highly electropositive adsorbate material is critically important to the enhancement of negative ion formation during the sputtering process. A 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 Reference 17. 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<sup>18,19</sup> 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 4 displays a schematic representation of a plasma sputter source now under design for potential use at the HRIBF. The source will be housed in the same vacuum envelope as the positive HRIBF sources, described in Section 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 hot filament or an RF antenna. The final choice of the two means of igniting the plasma will be determined during off-line testing of the source. The radioactive ion beam will be formed by sputter ejection of atoms or molecules from the negatively biased sphericalgeometry 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 negativeion beam through the exit aperture (diameter  $\phi = 3$  mm). At this point, the ion beam



FIGURE 4: Schematic drawing of a plasma-sputter negative-ion source equipped with a RF plasma igniter which is now under design for potential use in the HRIBF. The source will be complementary to the HRIBF electronbeam-plasma source displayed in Figures 1–3.

is further accelerated to energies up to 50 keV prior to mass analysis. The efficiencies of several negative ion species, estimated by Tsuji and Ishikawa,<sup>19</sup> are shown in Table 2. The measured emittance of a similar geometry source is quite good ( $\sim 6\pi$  mm.mrad MeV<sup>1/2</sup>)<sup>20</sup> even at mA intensity levels of Cu<sup>-</sup>. This emittance is lower than the acceptance of the HRIBF 25-MV tandem accelerator.<sup>21</sup>

#### 4 NEGATIVE-SURFACE-IONIZATION SOURCE

For atoms or molecules leaving a heated surface, the probability of negative ion formation  $\eta_i$  arriving at a position far from the metal in a given state depends on the magnitude of the difference between the electron affinity  $E_A$  and the surface work function  $\phi$  of the atom or molecule, i.e.,  $(E_A - \phi)$ . 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 form of the Langmuir-Saha equation for the probability of negative ion formation for neutral particles of electron affinity  $E_A$  interacting with a hot metal surface at temperature T and constant work function  $\phi$  is given by

Z	Element	$I_p(eV)$	N <sub>e</sub>	$\eta_{\rm calc}(\%)$	$\eta_{\exp}(\%)$	Ref.
10	<sup>20</sup> Ne	21.56	8	2.2	1.6	8
18	<sup>40</sup> Ar	15.76	8	17.8	19	8
24	<sup>54</sup> Cr	6.77	1	38.0	>20	8
26	<sup>57</sup> Fe	7.90	2	46.4	30	8
32	<sup>76</sup> Ge	7.90	4	66.7	41	8
36	<sup>84</sup> Kr	14.00	8	36.0	35	8
36	<sup>84</sup> Kr	14.00	8	36.0	36	13
46	<sup>100</sup> Pd	8.33	18	90.0	>25	8
47	<sup>107</sup> Ag	7.58	1	39.8	47	8
47	<sup>109</sup> Ag	7.58	1	40.2	50	8
50	<sup>116</sup> Sn	7.34	4	74.8	53	8
50	<sup>124</sup> Sn	7.34	4	77.4	54	8
54	<sup>129</sup> Xe	12.13	8	56.0	52	8
54	<sup>132</sup> Xe	12.13	8	56.6	53	8
54	<sup>132</sup> Xe	12.13	8	56.6	56	12
79	<sup>197</sup> Au	9.23	1	34.2	50	8
82	<sup>208</sup> Pd	7.42	4	79.5	52.8	8
83	<sup>209</sup> Bi	7.29	5	83.5	68.3	8

TABLE 1: Comparisons of calculated  $\eta_{calc}$  and experimentally measured  $\eta_{exp}$  ionization efficiencies for electron beam plasma ion sources. Estimated ionization efficiencies were calculated by using Equation 1.

$$\eta_i = \frac{\omega_-}{\omega_0} \left( \frac{1 - r_-}{1 - r_0} \right) \exp\left( \frac{E_A - \phi}{kT} \right) \times \left[ + \frac{\omega_-}{\omega_0} \left( \frac{1 - r_-}{1 - r_-} \right) \exp\left( \frac{E_A - \phi}{kT} \right) \right]^{-1} , \quad (3)$$

where  $r_{-}/r_{0}$  are the reflection coefficients of the negative/neutral particles at the surface and  $\omega_{-}$  and  $\omega_{0}$  are statistical weighting factors for the negative ion and neutral atom, respectively.  $\omega_{-}$  and  $\omega_{0}$  are related to the total spin of the respective species given by  $\omega = 2\sum_{i} s_{i} + 1$ , where  $s_{i}$  is the spin of the electron. In principle, the halogen group of elements can be ionized with unit efficiency on a clean LaB<sub>6</sub> surface maintained at 1373°K. From Equation 3, it is evident that negative ion yields could be enhanced by lowering the work function  $\phi$  or increasing the surface temperature T for elements where  $E_{A} \leq \phi$ . The former can be effected by surface adsorption of minute amounts of low-work-function materials such as the group I-A and II-A elements. Analogously, the adsorption of minute amounts of highly electronegative atoms or molecules such as oxygen or the halogens can deleteriously effect the negative surface ionization efficiency by raising the work function.

 TABLE 2:
 Estimates of the probability for negative ion formation by xenon sputtering at optimum cesium coverage (Ref. 19)

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



FIGURE 5: A negative-surface-ionizaton source design now under consideration for use in generating RIBs at the HRIBF from elements with high electron affinities such as the group VI-A and VII-A elements.

The negative form of surface ionization is also highly chemically selective and, therefore, can be used to great advantage for the generation of high-electron-affinity elements such as the group VII-A (halogens). Unfortunately, there is limited availability of a wide variety of stable, low-work-function materials. LaB<sub>6</sub> is the most frequently used low-work-function surface ionizer, having a work function  $\phi = \sim 2.7$  eV for polycrystalline and  $\phi \simeq 2.36$  eV for single-crystalline material.

An on-line form of the negative-surface-ionization source has been developed at CERN-ISOLDE, which is equipped with a LaB<sub>6</sub> surface ionizer.<sup>22</sup> The positive/negative-surfaceionization source described in Reference 23 has also been used to generate beams of  $UF_6^-$ . The HRIBF source can easily be retrofitted with a LaB<sub>6</sub> ionizer and used to efficiently ionize high-electron-affinity elements as clearly evidenced by the successful application described in Reference 22. Figure 5 displays a negative-surface-ionization source which will be used as a complement to other HRIBF sources. The vapor transport tube will be Ir coated tantalum and heated resistively to  $\sim 1100^{\circ}$ C to again minimize the residence times on surfaces of chemically active materials. This source will serve as a backup source to the plasma-sputter-negative-ion source described in Section 3 for the generation of RIBs from the group VIIA elements.

## 5 DISCUSSION

The most difficult challenge related to the efficient generation of RIBs using the ISOL technique is associated with target and surface adsorption issues and not the ion source itself. The challenge is to select a target material which will withstand the high temperatures required to efficiently and promptly release the short-lived species of interest while preserving the physical integrity of the target material and the vacuum requirements of the ion source. The speed at which the element is released from the target is directly related to the diffusion properties of the species/target material combination.<sup>24</sup> As pointed out by Ravn,<sup>25</sup> in general, each element must be considered separately, often requiring dedicated efforts to solve specific problems in order to generate useful RIBs of the element. Much work remains to be done in the area of target material selection and the development of techniques to enhance the release of elements from a properly chosen target material.

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, a modified version of the CERN-ISOLDE source has been chosen for use at the HRIBF because of the following: 1) it is competitively efficient; 2) it has demonstrated reliability over many years of operation at CERN-ISOLDE; 3) the target temperature can be controlled independently of the discharge parameters of the source; 4) it can be readily adapted for a wide range of operational temperatures and for other modes of operation; 5) it is readily adaptable to other types of ion sources; and 6) it has been engineered for safe removal and installation in the high level radiation fields present at an ISOL facility.

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 welldeveloped technology are not as sensitive to poisoning effects as are direct-negative-surfaceionization sources and, therefore, are very appealing for use at tandem-accelerator-based RIB facilities such as the HRIBF.

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. They offer a high degree of chemical selectivity and are usually simple and easy to operate. However, the ionizers in negative-surface-ionization sources may be easily poisoned and, therefore, may pose challenges for use at tandem-accelerator-based RIB facilities such as the HRIBF.

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