

A Negative Surface Ionization Source for RIB Generation

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Abstract. An efficient negative surface ionization source has been designed, fabricated, and initial tests begun for potential on-line use in generating radioactive ion beams of members of the group VIIA elements (F, Cl, Br, I, and At) for the Holifield Radioactive Ion Beam Facility research program. The source utilizes direct-surface ionization to form negative-ion beams resulting from interactions between highly electronegative atoms or molecules and a spherical-sector surface LaB₆ surface ionizer maintained at ~1722 °C. Despite its widely publicized propensity for being easily poisoned, no evidences of this effect were experienced during testing of the source. The source has been extensively evaluated off-line in terms of ionization efficiency for generating beams of Br⁻ by feeding AlBr₃ vapor at low feed rates into the source. The results of initial testing indicate that the source is reliable, stable and easy to operate, with nominal efficiencies of 15% for Br⁻ beam generation when account is taken of the fractional thermal dissociation of the AlBr₃ carrier molecule. The design features and principles of operation of the source are described and initial performance, operational parameter and beam quality (emittance) data are presented in this article.

INTRODUCTION

Ion sources based on the surface ionization principle are generally characterized by a high degree of ion beam purity (chemical selectivity), thermal energy spreads (~2 kT << 1 eV), and limited range of species capability. The ionization efficiency can be high or low, depending on the electrochemical character of the species in relation to the work function of the ionizing surface. Because of the fact that the surface-ionization process is highly chemically selective, it can be used to great advantage for radioactive ion beam (RIB) applications to eliminate isobaric contaminants that may compromise experimental results with these beams. Experimental methods and techniques for negative ion production by surface ionization have been reviewed by Kawano et al. [1-3].

Surface ionization has not been utilized frequently for generation of negative ion beams – principally due to the lack of chemically stable low-work-function materials for use as ionizers. Unfortunately, few chemically stable materials are available for this purpose in contradistinction to its positive ionization complement where several high-work-function metals may be chosen. LaB₆ is usually used for negative surface ionization because of its relatively low work function ϕ : 2.3 to 3.2 eV [4-8]) and ready availability, despite its widely publicized propensity for poisoning [9,10]. The poisoning mechanism appears when LaB₆ interacts with residual gases in the vacuum system,

usually under high flow rate conditions or higher than optimum pressure conditions. The effect raises the work function of the LaB₆ surface, thereby reducing the probability of ionizing electronegative atoms as they evaporate from the surface. Under high flow-rate conditions, the poisoning process also affects the reliability of operation of sources equipped with this material through time varying fluctuations of ion-beam intensity caused by variations in work function [10]. A raising of the work function causes an exponential diminution of the probability for negative ion formation and, consequently, a reduction in intensity of extracted negative ion beams.

Despite this problem, sources based on the use of LaB₆ ionizers have been described in the literature [10-12], including their use at ISOL facilities for negative ion generation of high-electron-affinity radioactive species [12]. A LaB₆ surface ionizer was also chosen for use with the source described in this article because the poisoning effects were not expected to be pronounced at existing flow-rates that characterize on-line source operation at ISOL based RIB facilities.

THEORY OF NEGATIVE ION FORMATION

The process of direct-surface ionization is statistical in nature, and therefore statistical and thermodynamic arguments can be used to determine the degrees of positive or negative ion formation. For thermodynamic equilibrium processes, the ratio of ions

to neutrals that leave an ideal surface can be predicted from Langmuir-Saha surface ionization theory appropriate for either positive or negative ion formation. The form of the Langmuir-Saha equation for the probability of negative-ion formation of neutral particles of electron affinity E_A interacting with a hot metal surface at temperature T and low work function, P_i , is given by

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

where r_- and r_0 are the reflection coefficients of the particle at the surface and ω_- and ω_0 are statistical weighting factors for the negative ion and neutral atom, respectively. ω_- and ω_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. From Eq. 1, it is evident that negative ion yields can be enhanced by lowering the work function, ϕ , or increasing the surface temperature T for elements where $E_A \gg \phi$.

DESCRIPTION OF THE SOURCE

The salient design features of the negative surface ionization source are schematically illustrated in Fig. 1 which shows a cross sectional side view of the target reservoir, the vapor transport tube, ionization region and extraction electrode system of the source. The target material reservoir is positioned within the inner diameter of a series-connected, resistively-heated, Ta tube designed to reach temperatures exceeding 2000 °C. The vapor transport tube is typically heated resistively to ~1400-2000 °C by passing a current through the tubular structure. In order to provide halogen atoms for evaluating the efficiency of the source, AlB₃ vapor was fed into the target material reservoir from the vapor feed system described below.

According to Eq. 1, electronegative species that strike and are subsequently evaporated from the spherical-geometry LaB₆ ionizer surface have a finite probability of being negatively ionized, these species are then extracted by applying a positive voltage to the extraction electrode.

Ionizer Design

The spherical geometry ionizer (spherical radius: 2.5 mm; diameter: 4.3 mm) is machined from a solid

LaB₆ rod and pressed into a 6 mm diameter Ta holder with 0.63 mm deep slots machined in the outer periphery through which vapor flows from the transport tube into the ionization region of the source. The temperature distribution along the vapor transport system and at the LaB₆ surface ionizer were computed as function of the heating current by use of the thermal transport code ANSYS [13]. These results were used to design the transport tube so that the tube can be operated at temperatures up to ~2000 °C while achieving a maximum temperature at the surface of the ionizer of ≤ 1740 °C, the thermal dissociation temperature of LaB₆ (calculated by use of the chemical reaction code described in Ref. 14).

This effect was achieved by adjusting the thickness of the transport tube and the interface position of the return current bus relative to the position of the LaB₆ ionizer holder so that an optimum temperature of ~1722 °C could be maintained during normal operation of the source.

Ion Optics

Figure 2 displays the ion optics for the ion extraction system of the source. As noted, the spherical geometry electrode system is designed to focus the beam through a small aperture (ϕ : 0.41mm). The perveance, P_c , for space-charge-limited flow of Br⁻ through the electrode system is $P_c = 1.0 \times 10^{-9}$ [A/{V_{ex}}^{3/2}] as calculated by use of the computer code described in Ref. 15. Of course, this figure of merit has no practical meaning at the very low flow rates used in these experiments or that will be present during on-line operation of the source where space charge effects are negligible.

The Vapor Feed System and Vapor Transport

The system (see, e.g., Fig. 3) consists of a reservoir, located external to the main vacuum system of the source, for holding a relatively volatile halogen feed material (e.g., AlBr₃ or AlCl₃). The material is fed through a small diameter transfer tube of effective radius, a , and length, l that is designed to conductance limit the flow rate of vapor from the reservoir to the source. The transfer tube is isolated from the heat-sink effects of the vacuum chamber wall by means of a thermally isolated vacuum feed-through. Feed material, placed in the reservoir, is raised to temperature, T_R , where it is vaporized, creating a pressure gradient, $(p_R - p_{IS})/l = \Delta p/l$ along the tube due to differences in pressure in the reservoir, p_R , and in the ionization region of the source, p_{IS} . The reservoir is heated by thermal conduction from the independently heated transfer tube with temperature control maintained by adjusting the transfer tube temperature.

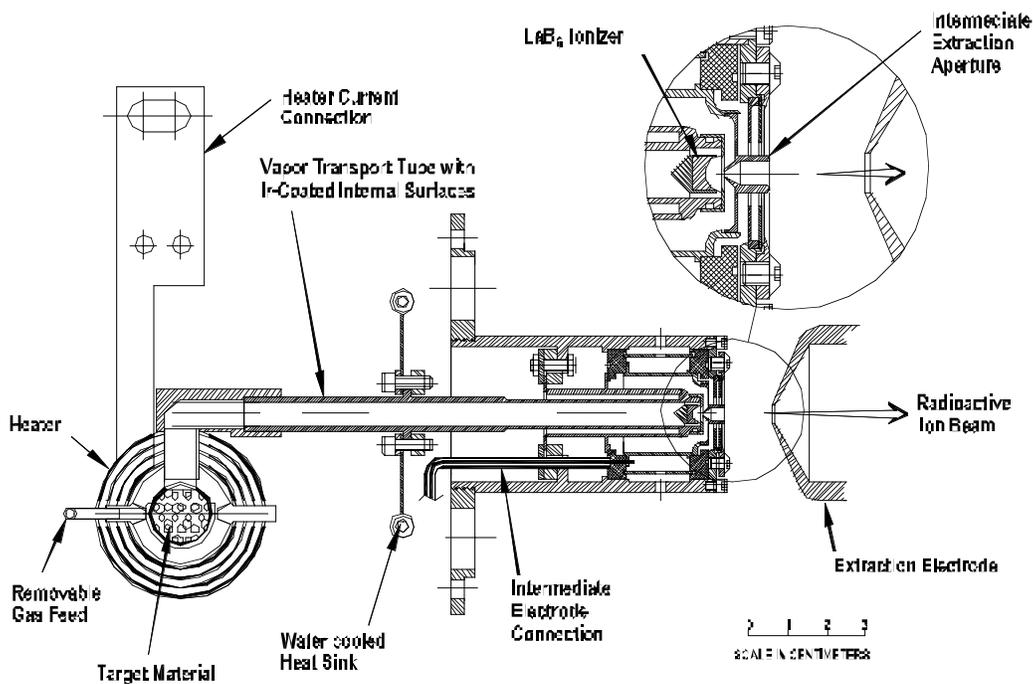


FIGURE 1. Schematic drawing of the negative ionization source equipped with a spherical geometry ionizer.

Vapor pressure

The vapor pressure of the AlBr_3 feed material, used to investigate the ionization efficiency of the source, was computed by use of the thermo-chemistry computer code, ThermoCalc [16]. During all measurements, the reservoir was operated between 20 and 22 °C, correlating to a range of vapor pressures between 3.5×10^{-1} Pa and 4.6×10^{-1} Pa.

Flow rate of Br atoms into the ion source

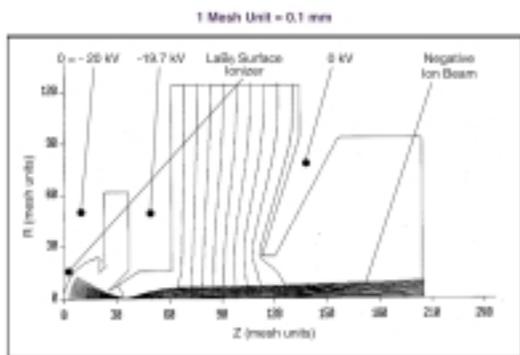


FIGURE 2. Ion optics of the spherical-geometry surface-ionization source for extraction of Br^- . 1 Mesh Unit = 0.1

The rate of flow of halogen molecules through the transfer tube into the source can be estimated from the familiar relation (see, e. g., Ref. 17), given by

$$dN_M/dt = \{2\pi a^3/3k_B T\} v_M \Delta p / l \quad (2)$$

In Eq. 2, Δp is the pressure drop across the transport tube of length, l , and effective radius, a ; T is the average temperature from the reservoir to the ionization volume of the source and; v_M is the average velocity of a AlBr_3 molecule in transit from the feed material reservoir to the ionizer volume of the source. At the operational temperature of the vapor transfer

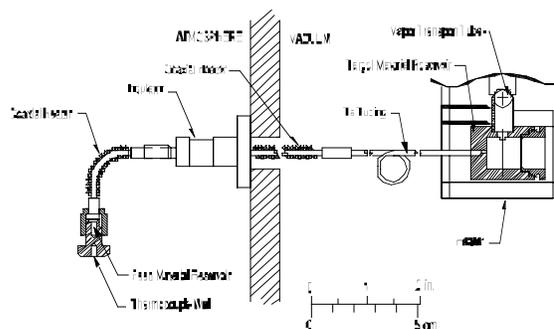


FIGURE 3. Vapor transport system used to feed AlBr_3 vapor into the source

tube, AlBr_3 thermally dissociates. Therefore, the number of halogen atoms entering the source per unit time, dN_A/dt , is equal to

$$\begin{aligned} dN_A/dt &= 3F_D dN_M/dt \\ &= F_D \left\{ 2\pi a^3 / 3k_B T \right\} v_m \Delta p / l \end{aligned} \quad (3)$$

where F_D is the dissociation fraction for releasing atomic Br atoms into the source from the AlBr_3 molecule during transit through the hot vapor transport tube and the factor, 3, is the number of Br atoms in the AlBr_3 molecule. F_D can be estimated by computing the equilibrium composition of the molecule as a function of temperature, T . The equilibrium composition versus temperature of AlBr_3 in a Ta tube is displayed in Fig. 4 as computed with the chemical reaction and equilibrium code described in Ref. 14. As noted the equilibrium dissociation fraction for AlBr_3 is 0.666 at $T \geq 1500$ °C. For optimum efficiency, the LaB₆ ionizer operates at ~ 1722 °C.

The rate of neutral Br atoms striking the LaB₆ ionizer

The number of neutral Br atoms, dN_{IS}/dt , striking the ionizer per unit area per unit time can be expressed as

$$\begin{aligned} dN_{IS}/dt &= \left\{ dN_A/dAdt \right\} A_{IS} \\ &= \left\{ F_D \left\{ 2\pi a^3 / 3k_B T \right\} v_A \Delta p / l \right\} A_{IS} / A_T \quad (4) \\ &= n_A v_A \left\{ A_{IS} \right\} / 4 \end{aligned}$$

where v_A is the average velocity of a Br atom within the volume surrounding the ionizer surface at temperature T ; n_A is the number of particles per unit volume in the region of the ionizer, A_{IS} is the effective area of ion extraction from the ionizer surface, determined from ion optic data (see Fig. 2) and A_T is the total surface area of the ionization chamber.

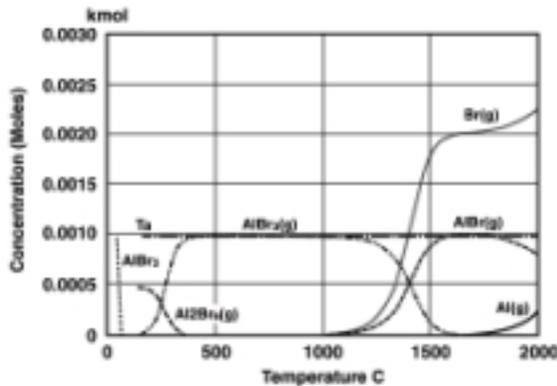


FIGURE 4. Equilibrium composition of AlBr_3 in a Ta tube.

The negative ion-beam intensity

Since the negative-ion-beam intensity is proportional to the number of neutral Br atoms striking the ionizer surface per unit time, the Br^- intensity can be estimated by multiplying Eq. 4 by the probability of negative ion formation given by Eq. 1.

Operational Parameters

In order to optimize the performance of the source, the dependence of Br^- beam intensity on the following parameters must be known: (1) Br^- beam intensity versus feed material reservoir temperature; (2) Br^- beam intensity versus vapor transport tube current (ionizer temperature); and (3) Br^- beam intensity versus extraction voltage.

Br^- beam intensity versus feed material reservoir temperature

The feed material reservoir temperature was held essentially constant during all measurements, at 22 °C, correlating to an equivalent flow rate of AlBr_3 into the source of ~ 3.33 μA at a vapor transport tube current of 368 A.

Br^- beam intensity versus vapor transport tube temperature

Onsets are observed in the $^{79}\text{Br}^-$ versus vapor transport tube current (ionizer temperature), shown in

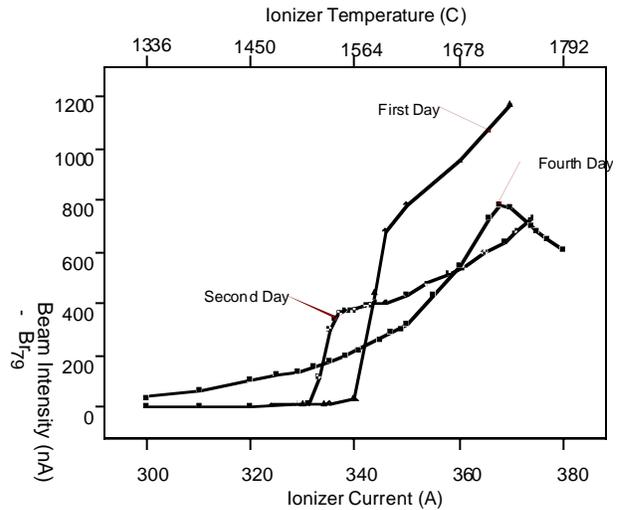


FIGURE 5. $^{79}\text{Br}^-$ ion beam current and ionizer temperature versus ionizer current.

Fig. 5, for data taken after the *First Day* and *Second Day* of source operation that we attribute to the dissociation of AlBr_3 ($\text{AlBr}_3 = \text{AlBr} + 2\text{Br}$). (The surface temperature of the LaB_6 ionizer, measured with an optical pyrometer, is also shown on days of operation, the $^{79}\text{Br}^-$ versus vapor transport tube the top abscissa scale.). However, after more than two current relation reverted to the curve designated as *Fourth Day*. As noted, the beam intensity increases monotonically with vapor transport tube current until it reaches an optimum value of ~ 368 A, beyond which the intensity drops. (This current correlates to a temperature of ~ 1722 °C at the surface of the LaB_6 ionizer.) The decrease in intensity beyond the optimum value for the *Fourth Day* of operation is presumably attributable to the onset of thermal dissociation of LaB_6 (~ 1740 °C).

Br⁻ beam intensity versus extraction voltage ΔV_{ex}

Figure 6 displays a typical Br^- ion-beam intensity versus extraction voltage curve. As noted, the extracted ion current increases with extraction voltage, ΔV_{ex} , until it reaches a constant value (saturates) at ~ 150 V, suggesting that the Br ions are extracted as fast as they are evaporated from the hot LaB_6 surface.

Source Performance: Experimental Results

The negative surface ionization source (Fig. 1) was characterized by feeding AlBr_3 vapor from a feed material reservoir located external to the main vacuum system, as displayed in Fig. 3. All efficiency measurements were made with the feed material reservoir temperature set at 22°C and the transport tube current fixed at 368 A, correlating to an ionizer temperature of ~ 1722 °C.

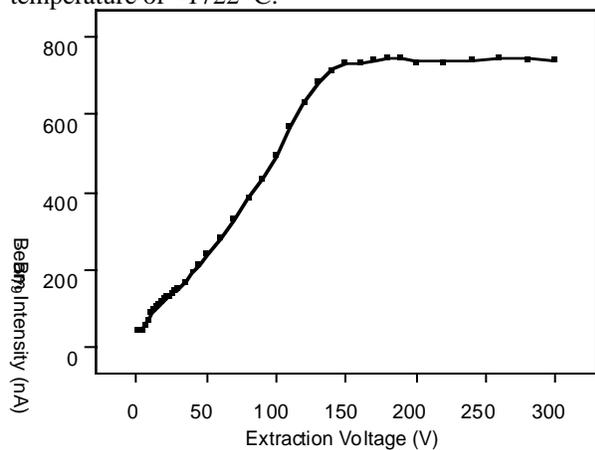


FIGURE 6. Br^- ion-beam intensity versus extraction voltage.

Mass spectrum

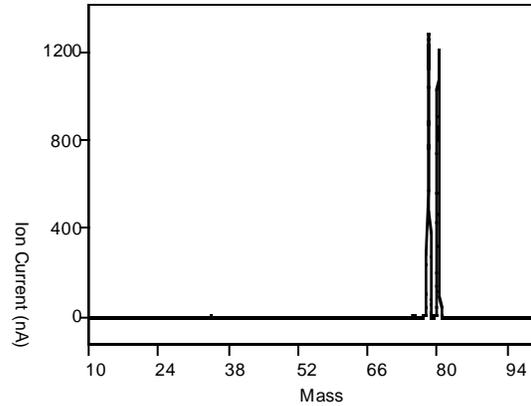


FIGURE 7. Br^- mass spectrum.

The mass spectrum obtained while feeding AlBr_3 in to the source is shown in Fig. 7. The selective nature of the surface ionization process is clearly demonstrated by the cleanliness of the spectrum. The only masses with significant intensity are the two isotopes of Br (i.e., ^{79}Br and ^{81}Br).

Emittance Data

Isobaric contamination problems can cause serious difficulties in interpretation and analysis of on-line experimental data and seriously compromise experimental results. In such cases, high quality beams (low energy spread, low-emittance) are very important, making possible mass resolution of such contaminants with existing magnetic isobar separation systems. Fig. 8 displays a typical emittance diagram for a 20 keV Br^- beam generated with the source. As noted the emittance is quite small.

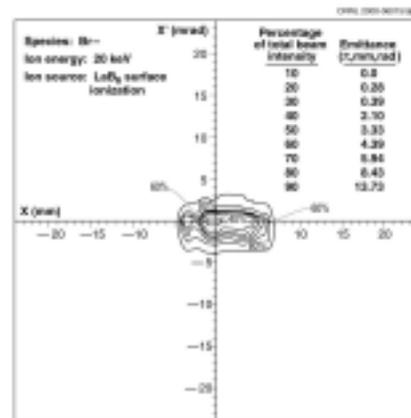


FIGURE 8. A typical emittance plot.

Br⁻ ionization efficiency estimates

According to the results of thermal equilibrium composition calculations, displayed in Fig. 4, the dissociation fraction of AlBr_3 is 0.66 at $T = 1722$ °C, the operational temperature of the ionizer. Thus, two of three of the Br atoms are available for ionization. The feed material reservoir was operated at 22 °C for all efficiency measurements, correlating to a vapor pressure of $p_R \cong 0.46$ Pa. The halogen atom flow rate into the ionization region of the source was calculated from Eq. 3, yielding an equivalent flow-rate of atomic Br into the source of ~ 10 μA . The overall efficiency for negative ion formation is then computed from the following relation:

$$\eta \cong I^-(\text{Br}) / \{dN_A/dt\} \quad (7)$$

Mass analyzed $^{79}\text{Br}^-$ intensities ranging between 0.8 and 1.5 μA were typical of the currents recorded during source evaluation, corresponding to $\sim 10\%$ to 17% total efficiency. Thus, the overall efficiency for generating beams of Br^- is quite high ($\sim 24.3\%$ for the *First Day* of operation), as required for on-line applications. Fig. 9 displays efficiency versus temperature of the LaB_6 ionizer.

CONCLUSIONS

The surface ionization source has proved to be a stable, reliable, versatile, and efficient means for generating beams of highly electronegative species. These characteristics and flexibility make it a viable candidate for use in several research and applied science applications, including RIB generation. Off-line tests, the source has demonstrated that it can be used to efficiently ionize feed materials such as AlBr_3 .

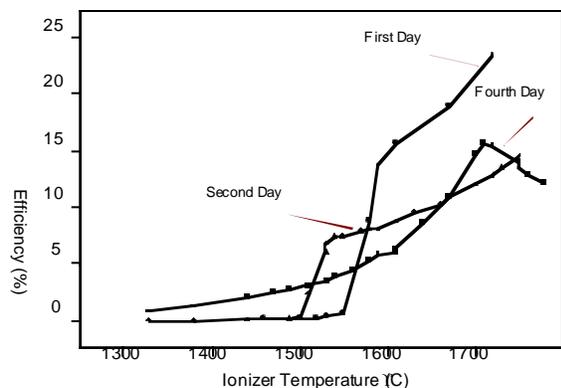


FIGURE 9. Efficiency versus surface ionizer temperature.

The source was found to be easy to operate with no evidence of poisoning effects which plague more traditional negative surface ionization sources equipped with LaB_6 ionizers that operate under higher flow rate conditions [10]. Preliminary estimates suggest that source can be used to ionize all halogens except F. Mass analyzed $^{79}\text{Br}^-$ intensities ranging between 0.5 and 0.75 μA were typical of the currents recorded during source evaluation, corresponding to $\sim 10\%$ to 15% total efficiency.

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