The Electron-Beam-Plasma Ion Source as Source of Negative Fluorine

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Radioactive ion beams (RIBs) of short-lived isotopes of fluorine are in demand for investigating astrophysical phenomena related to the hot CNO cycle and rp processes responsible for stellar nucleosynthesis. Since negative ion beams are required for injection into tandem electrostatic accelerators, such as the 25 MV tandem accelerator used for post acceleration of RIBs for the Holifield Radioactive Ion Beam Facility (HRIBF) research program at the Oak Ridge National Laboratory (ORNL), efficient, direct-formation F⁻ ion sources are highly desirable for RIB applications involving this type of post-accelerator. We have conceived and evaluated a direct extraction F⁻ source for potential RIB applications which is predicated on the reverse polarity operation of a positive electron-beam-plasma target/ion source (EBPTIS) while simultaneously feeding fluorine rich compounds and Cs vapor into the source. The source is found to operate in two separately distinct temperature regimes for the generation of F⁻: (1) a high cathode temperature regime or plasma mode and (2) a lower cathode temperature regime or surface ionization mode. For the latter mode of operation, net efficiencies of $\eta = 0.2\%$ were attained for the EBPTIS; delay-times, τ , attributable to the transport of F and fluoride compounds from the target to the ionization chamber of the source, typically, were found to be $\tau \sim 50$ ns. Brief

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descriptions of the EBPTIS and experimental techniques used in the studies, as well as net efficiency and effusive flow data for the negative EBPTIS, are presented in this report.

1. Introduction

Radioactive ion beam (RIB) facilities have been constructed or proposed for construction around the world because of the flourishing interest in accelerated RIBs for studying nuclear reactions of fundamental importance in nuclear structure physics and nuclear astrophysics. These facilities utilize the well-established ISOL (Isotope Separation On-Line) technique [1,2] for the production and generation of RIBs by bombarding thick target materials, close-coupled to a high temperature target/ion source (TIS), with low-Z ions from a primary accelerator. Short-lived radioactive species, formed within the target must diffuse to the surface, desorb from the surface and then be transported to the ionization region of the TIS, ionized, extracted and post accelerated to research energies in a time span commensurate with the lifetime of the species of interest. Since intensity is of primary importance for RIB applications, a premium is placed on ion sources with these attributes.

Negative ion sources for RIB applications have, in general, received less development attention than their positive ion counterparts because of the universal character of the positive ion formation process and the consequent plurality of positive ion accelerators in spite of the fact that they offer decided advantages over their positive ion counterparts for processing certain high electron affinity elements such as the halogens (F, Cl, Br and I). Among these advantages are the following: beams formed through negative surface ionization exhibit low energy spreads; their use lowers the intensity and numbers of isobaric impurities due to the high degree of chemical selectivity of the negative surface ionization process; and, their use avoids having to provide for charge exchange for injection into tandem electrostatic accelerators. In principle, negative ion beams of these elements can be efficiently formed through surface ionization, by kinetic ejection of adsorbed halogens or halide compounds from a low work function surface or through electron dissociative attachment of a halide molecules. Although negative sources have been

developed for the heavier, less chemically reactive halogens and halogen compounds for ISOL experiments as well as other applications [3-9], efficient and reliable sources for F^- generation have proven to be more difficult. Presently there is considerable interest in RIBs for studying astrophysical processes such as the "hot" CNO cycle and the rp processes which occur in nuclear synthesis cycles of stellar systems; ^{17}F and ^{18}F are of particular interest for the study of the rp process [10]. Although, negative ion sources capable of efficiently producing negative ions of the isotopes of F have not reached final efficiency goals, steady progress is being made at the Holifield Radioactive Ion beam facility (HRIBF) of the Oak Ridge National Laboratory (ORNL) toward a solution to the F^- generation problem [11,12].

Although electron-beam-plasma type target/ion sources (EBPTISs) have been used extensively for generating positive RIBs [13-16] from many elements, they have never been evaluated in terms of their negative ion formation capabilities. The dissociative attachment and negative surface ionization mechanisms are among the most efficient processes for negative ion formation. The operational principles of the EBPTIS suggest that either or both of these mechanisms may be in play during reverse polarity operation, provided that a method for promoting either surface or volume negative ion formation is incorporated in the design of the source. In order to realize efficient negative ion formation, it has been found that the addition of highly electropositive elements such as Cs can greatly enhance H^- intensities extracted from "volume"-type H^- ion sources [17,18]. Therefore, the EBPTIS, developed for use in conjunction with the research program at the HRIBF [14,15], was evaluated for F^- generation by reversing the polarity of the source while feeding neutral Cs vapor and fluorine and fluoride compounds into the source. The operational parameters, ionization efficiencies for forming F^- and effusive-flow delay-times for fluorine and fluorides in the reverse polarity EBPTIS are presented in this report.

2.0 The Direct Extraction Negative EBPTIS

The ORNL EBPTIS, developed for use in the HRIBF research program [14,15], is schematically illustrated in Fig. 1. The only modifications required to evaluate the source in the negative ion mode, other than reversal of the high voltage power supply, were the additions of a Cs oven and a magnetic electron suppresser. Prior to operation in the negative ion mode, the source was operated in the positive ion mode and found to perform with the usual stability and ionization efficiency ($\eta \approx 15\%$ for Xe). During the measurements, Cs vapor was fed into the TIS through a $l = 32.9$ cm long Ta tube with average radius of $a = 1$ mm at typical operating Cs oven temperature ranging between 150° and 205°C , corresponding to a range of Cs vapor flow-rates between 1.5×10^{14} and 1.5×10^{15} atoms/s. Electron suppression was effected by the addition of a permanent magnet which produced a 250 G transverse magnetic field on axis in the extraction region of the source. The target material reservoir and vapor transport tube were heated resistively by passing independent currents through each component. The ionization chamber (anode) was heated by radiation from the end of the vapor transport tube and by electron bombardment when the cathode reached emission temperatures. Fig. 2 displays temperatures at various points along the transport tube (T1...T5) and ionization chamber of the EBPTIS as a function of transport tube heating current, as calculated using the finite element analysis code ANSYS [19] and verified by W-Re thermocouple measurements.

Thin, highly-permeable oxide target materials are preferred for proton production of RIBs of F because of their efficient release characteristics and refractory properties. Fibrous targets of Al_2O_3 ($\phi \sim 3 \mu\text{m}$) have been selected for use in generating F RIBs for the HRIBF research program [20] and shown to release $> \sim 90\%$ of the fluorine isotopes produced

during on-line experiments when operated at temperatures up to $\sim 1600^{\circ}\text{C}$ [21]. Therefore, the present experiments were conducted with fibrous Al_2O_3 in the target material reservoir of the reverse polarity EBPTIS to simulate the actual RIB generation process.

3.0 Ionization Efficiency and Effusive-Flow Delay-Time Measurements

3.1 Experimental equipment and procedures

The ionization efficiency and effusive-flow delay-time measurement techniques and associated equipment used in these studies were developed earlier for evaluating prototype RIB sources for the generation of ion beams from chemically active electronegative elements such as the halogens [22]. The efficiencies for generating beams of F were measured by injecting SF_6 through a cooled nozzle into the target material reservoir of the TIS (see Fig. 1) at flow-rates commensurate with on-line RIB generation. In order to reduce the temperature of the nozzle to values below the thermal dissociation temperature of the SF_6 molecule, it was decided to displace the nozzle 4.2 mm from the entrance to the target material reservoir. Since SF_6 is gaseous at the operating temperatures of the feed system (20° - 300°C), flow-rates can be controlled and estimated reasonably accurately. Since the target material reservoir of the TIS is maintained at a much higher temperature ($T \sim 1400^{\circ}\text{C}$) than the injection nozzle ($< 300^{\circ}\text{C}$) gaseous molecules completely thermally dissociate [23] upon entering the target material reservoir, thereby releasing atomic F into the TIS at a known flow-rate. The fraction of material lost through the gap was determined by measuring the positive ionization efficiency for Xe gas, introduced into the source through a fixed leak at a known flow rate, with and without the gap between the TIS and cooled nozzle [22]. Particle losses were also calculated by using well known formulas

taken from the kinetic theory of gases [24]; for these experiments, the fraction of gas lost between the gap and the target material reservoir was estimated to be 11.6%.

3.2 Ionization efficiency measurements

During evaluation of the reverse polarity EBPTIS, two distinctly different modes were found for optimum F^- generation: a low cathode temperature mode (low electron density mode) corresponding to a low ionization chamber temperature ($\sim 625^\circ - 650^\circ\text{C}$) where the cathode was operated below emission temperatures for plasma ignition (e.g., $< 2100^\circ\text{C}$) and a high cathode temperature mode (high electron density mode) corresponding to ionization chamber temperatures between 700° and 900°C where the cathode was operated up to plasma ignition temperatures (e.g., $\sim 2100^\circ\text{C}$). F^- ion beam intensity versus ionization chamber temperature and vapor transport tube heating current data for fixed Cs vapor and F flow-rates are displayed in Fig. 3; also shown are emission current versus vapor transport tube heating current data for the same Cs and F flow-rate conditions. The F^- beam intensity for the low ionization chamber temperature mode (low electron emission mode) is ~ 6 times greater than that for the high ionization chamber temperature mode (high electron emission mode). As noted, the F^- current reaches a maximum value at an ionization chamber temperature of $\sim 625^\circ - 650^\circ\text{C}$ at which point it decreases steadily with increasing temperature. The low cathode temperature regime (surface ionization mode) is commensurate with conditions in which the dominant negative ionization mechanism is most likely that of surface ionization while the high cathode temperature regime (volume ionization mode) is more compatible with a dissociative attachment negative ion formation mechanism. Since both modes require the presence of Cs vapor, surface effects most likely contribute to negative ion formation in both modes of operation. Also, when the

source is operated in the high cathode temperature regime, conditions are very favorable for electron detachment of negative ions formed either through dissociative attachment or surface processes because of the high electron densities accompanying this mode of operation. Thus, it is decidedly more advantageous to operate the source at cathode temperatures which ensure that the ionization chamber temperature reach values between 625° and 650°C.

The probability of negative surface ionization depends exponentially on the magnitude of the difference between the electron affinity E_a of the species and the work function of the surface from which the atom or molecule is evaporated or ejected [25]. Thus, a low work function surface is necessary in order to realize optimum ionization efficiencies of a particular species. Less than one monolayer of a highly electropositive adsorbate has the effect of lowering the surface work function of an adsorbent. (A minimum work function of ~ 1.8 eV can be realized by the adsorption of ~ 0.5 monolayer of Cs on a Ta surface [25].) Therefore, Cs was introduced into the EBPTIS to effect a lowering of the work function of the Ta ionization chamber surface. According to calculations, $\sim$ a monolayer of Cs is adsorbed on the walls of the Ta ionization chamber at temperatures below 700°C which corresponds to a Cs flow-rate of 1.5×10^{15} atoms/s [26]. As the ionization chamber temperature is increased above 700°C, the coverage abruptly decreases until an insignificant fraction of Cs is present on the surface at a temperature of $\sim 1000^\circ\text{C}$ which corresponds approximately to the onset temperature for Cs^+ evaporation [9].

F^- beam intensity versus Cs flow-rate and Cs oven temperature data, for the F^- feed-rates and vapor transport tube heating currents required for optimum ionization efficiencies for both the volume and surface ionization modes, are shown in Fig. 4. Plots of F^- beam

intensity versus anode potential for an optimum Cs flow-rate and a fixed F flow-rate of 2.6×10^{13} atoms/s for each mode of operation are displayed in Fig. 5. The F^- ion current was found to vary linearly over the F flow-rate regime of 1×10^{12} - 5×10^{13} atoms/s, suggesting constant F^- formation probabilities within this range of feed-rate parameters.

As indicated in Fig. 3, a maximum in the $^{19}F^-$ efficiency of $\eta = 2 \times 10^{-3}$ for the surface ionization mode was reached at an ionization chamber temperature of 625°C , corresponding to a vapor transport tube temperature (T5) of 1660°C . Earlier investigations in which the EBPTIS was operated in the positive ion extraction mode with Al_2O_3 fibrous target material in the reservoir showed that F atoms are released from the target material and transported through the TIS in the molecular forms of AlF and BeF_2 [22]. Calculations show that the thermal dissociation fractions of AlF and BeF_2 at 1660°C are, respectively, 2.64×10^{-3} and 6.3×10^{-3} [23]. Since the maximum ionization probability $\eta = 2 \times 10^{-3}$ is in close agreement with the calculated dissociation efficiencies of these compounds, the probability for ionizing the dissociated atoms of F species appears to be quite high. Thus, for achieving high efficiencies and consequently high ion beam intensities, it will be necessary to incorporate means for efficient dissociation of compounds in future RIB sources, as first pointed out in Refs. 12, 27 and 28.

3.3 Delay time measurements

It is imperative to minimize delay times τ in a given TIS in order to reduce losses due to decay of short lived RIB species during their vapor transport from the target to the ionization chamber of the TIS. Several factors affect the effusive-flow delay times,

including: the operating temperature of the transport tube system and the enthalpies of adsorption of the species on the transport surfaces of the TIS. The enthalpy of adsorption for most species can be minimized by proper choice of the materials of construction for the transport system of the TIS. Re and Ir have been suggested for this purpose because of their chemical inertness and refractory characteristics [29]. The rate of arrival of radioactive particles with half-life $\tau_{1/2}$ at the ionization chamber of the TIS depends sensitively on the delay time during transit τ as seen from the following relations

$$I = e\eta_i\eta_{ex} I_0 \exp(-\lambda\tau) = e\eta I_0 \exp(-\lambda\tau) \quad (1)$$

where I_0 is the initial flow-rate of particles leaving the target and entering the transport tube; $\lambda = 0.693/\tau_{1/2}$ where $\tau_{1/2}$ is the half-life of the species; τ is the characteristic delay time for transit through the transport tube to the ionization chamber of the source system; e is the charge on the electron; I and I_0 are, respectively, the equivalent electrical currents of the radioactive species, η_i and η_{ex} are, respectively, the probabilities for ionization and extraction from the ion source and $\eta = \eta_i \eta_{ex}$ is the overall efficiency of the source, assuming no further losses in the beam transport system. As noted, the effusive-flow delay-times *must* be less than or approximately equal to the half-life of the species in question if debilitating losses due to decay are avoided.

As is obvious from Eq. 1, delay times affect the achievable beam intensity for a given species/TIS combination, and therefore, their magnitudes are crucially important. The effusive-flow delay-times of F and fluoride compounds were measured by use of the off-line apparatus and techniques described in Ref. 22 by injecting low flow-rates of SF_6 into an isolated Ta chamber and into the EBPTIS through a Ta needle valve, both of which were maintained at source operating temperatures. The mean delay-times were extracted from the

time dependence of the mass analyzed F^- current by opening/closing the valve for a length of time sufficiently long to achieve a steady state F^- signal. Effusive-flow delay times were then extricated from these data by fitting a single exponential to the rise or fall side of the F^- signal versus time curve for each mode of operation with the operational parameters of the source adjusted for optimum efficiency [30]; these data are displayed in Table I. As noted, delay times of 65 s and 42 s are characteristic of the source when operated, respectively, in the surface ionization and plasma ionization modes. These values are less than or approximately equal to the halflives of ^{17}F ($\tau_{1/2} = 64.5$ s) and ^{18}F ($\tau_{1/2} = 1.83$ h), both of which are of paramount interest for astrophysics research. Research at the Oak Ridge National Laboratory is supported by the U.S. Department of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp.

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Table I. Measured effusive-flow delay-times τ for source operating conditions which correspond to the maximum ionization efficiencies in the volume and surface ionization modes.

Ionization mode	τ (s)	Ionization Chamber	F Flow rate into TIS	Cs Flow rate into TIS
		Temperature (C)	(atoms/s)	(atoms/s)
surface	65	625	3×10^{12}	1.5×10^{15}
plasma	42	1025	3×10^{12}	1.5×10^{15}

Figure Captions

Fig. 1. Schematic drawing of the EBPTIS (side view) used in these studies. The source was developed for use as a positive ion source for RIB generation at the HRIBF [14,15]. For negative ion generation, the polarity of the source is reversed and the source is equipped with an external oven for supplying Cs vapor and with provisions for electron suppression.

Fig. 2. Temperature versus transport tube heating current for the transport tube, cathode emission surface and ionization chamber of the EBPTIS as calculated with the finite element code ANSYS [19] and verified with W-Re thermocouple measurements. The points T1 through T5 are shown in Fig. 1.

Fig. 3. Mass analyzed F^- beam intensity versus ionization chamber temperature and transport tube current for fixed Cs and F flow-rates; also shown is the dependence of emission current on the same parameters.

Fig. 4. Mass analyzed F^- ion current versus Cs oven temperature and Cs flow-rate into the TIS for a fixed F flow-rate; the source was operated with the cathode heating currents which were found to maximum the ionization efficiencies for surface and plasma ionization modes. The Cs flow-rates were calculated under Knudsen flow-conditions [24].

Fig. 5. Mass analyzed F^- ion current versus anode potential at a fixed F feed rate and at the Cs flow-rate, and transport tube heating currents required to optimize F^- ionization efficiency for both surface and volume ionization modes.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5