

A NEW CONCEPT TANDEM THERMAL DISSOCIATOR/ELECTRON IMPACT ION
SOURCE FOR RIB GENERATION

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Abstract

An innovative thermal dissociation/electron impact ionization positive ion source is presently under design at the Oak Ridge National Laboratory for potential use for generating RIBs at the Holifield Radioactive Ion Beam Facility (HRIBF). Because of the low probability of simultaneously dissociating and efficiently ionizing the individual atomic constituents with conventional, hot-cathode, electron-impact ion sources, the ion beams extracted from these sources often appear as a mixture of several molecular sideband beams. In this way, the intensity of the species of interest is diluted. We have conceived an ion source that combines the excellent molecular dissociation properties of a thermal dissociator and the high efficiency characteristics of an electron impact ionization source. If the concept proves to be a viable option, the source will be used as a complement to the electron beam plasma ion sources already in use at the HRIBF. The design features and principles of operation of the source are described in this article.

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1.0 Introduction

Chemical compounds are principally used as feed materials for more universal electron impact ion sources. These compounds may be vaporized directly in an oven or chemically synthesized insitu by passing a reactive vapor such as CCl_4 over an oxide containing the element of interest. The compounds which enter the ionization chamber must be dissociated and the element of interest ionized. For on-line isotope separation and radioactive ion beam generation applications, radioactive nuclei, formed within the bulk of a selectively chosen target material, must be diffused from the target material and transported to the ionization chamber of the source. For both applications, chemically active species may arrive at the ionization chamber of the source in a variety of molecular forms. Because of the low probability of simultaneously dissociating the molecule and efficiently ionizing the atomic constituents in conventional hot cathode electron impact ion sources, the species of interest may be present in a variety of molecular fragment ion beams, and, as a consequence, the intensity of the species of interest is diluted. For high sensitivity mass spectrometry and RIB applications, intensity of the species of interest is at a premium. Therefore, it is highly desirable to eliminate these side bands and concentrate as much of the species of interest as possible in a single mass channel. The combined thermal dissociator/electron impact ionization source offers the potential of simultaneously dissociating molecules and ionizing their atomic constituents relatively efficiently. The source described in this article was specifically designed to overcome molecular side-band problems associated with the electron impact ionization sources presently in use for future generation of RIBs at the Holifield Radioactive Ion Beam Facility, now under construction at the Oak Ridge National Laboratory [1].

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2.0 Theory of Thermal Ionization

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. Ionization equilibrium is a particular case of chemical equilibrium corresponding to a series of ionization reactions symbolically written as



where A_0 denotes a neutral atom and A_1, A_2 denote, respectively, singly and doubly ionized atoms and e is the electron removed in the collision. 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 [2]. The ionization efficiency for an atom with first ionization potential I_p can be expressed in the following form:

$$\eta = \left[1 + p \frac{g_0}{2g_1} \left(\frac{2\pi\hbar^2}{m} \right)^{3/2} \frac{\exp(I_p / kT)}{(kT)^{5/2}} \right]^{1/2}, \quad (2)$$

where m is the mass of the electron, T is the temperature, I_p is the first ionization potential of the atom, g_0, g_1 are, respectively, the statistical weights of the atom, and ion, and $\hbar = h/2\pi$, where h is Planck's constant. For atoms or ions $g = (2L + 1)(2S + 1)$, where L and S are the orbital and spin angular momentum quantum numbers of the atom or ion. This expression determines the degree of ionization as a function of pressure and temperature. The temperature need not be exceedingly high for a reasonable degree of ionization for easily ionized elements. Figure 1 illustrates the degree to which atomic hydrogen, mercury, calcium, and cesium are ionized as a function of temperature at $p = 1 \times 10^{-3}$ and 1×10^{-2} Torr. The first reported applications of this

technique are described in Refs. [3] and [4]. Such sources are relatively efficient for elements with ionization potentials $I_p \leq \sim 7$ eV.

Table I provides thermal ionization efficiencies for a number of elements, including Ca, Sr, Nb, and La, all of the lanthanides, and U, Pu, and Cm as measured with the source described in Ref. [4]. All of these elements have ionization potentials ≤ 6.3 eV. While the source has been used exclusively as a thermal ionization source, Table I clearly illustrates how effective the thermal process is for dissociation of strongly bound molecules. Because of its chemical selectivity and efficiency for ionizing elements with relatively low ionization potentials, the source has been utilized for ionization of short-lived nuclei created on-line by neutron activation [5] and by light ion transmutation of target materials [6].

3.0 Estimates of Electron Impact Ionization

The efficiencies of FEBIAD-type electron impact ionization ion sources, such as described in Refs. 7-11, are 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. [10], 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:

$$\eta_{calc} = \frac{4\langle\ell\rangle D_o N_e \left(\frac{\pi M_i}{8kT_i}\right)^{1/2} \exp\{-I_p / \langle kT_e \rangle\}}{A_o \left[1 + \frac{4\langle\ell\rangle D_o N_e \left(\frac{\pi M_i}{8kT_i}\right)^{1/2} \exp\{-I_p / \langle kT_e \rangle\}}{A_o} \right]} \quad (3)$$

Where $\langle\ell\rangle$ is the average path length for a particle in the plasma; D_o is a constant (cm^2/s); A_o is the emission area of the source; k is Boltzmann's constant; T_i is the ion temperature; T_e is the electron temperature; I_p is the ionization potential; N_e is the number of electrons in the valence

shell of the atom with a given I_p ; and M_i is the mass of species. The following values are used for terms in Eq. 3 when estimating ionization efficiencies for FEBIAD-type ion sources: $\langle kT_e \rangle = 3$ eV; $T_i = 2273^\circ\text{K}$; and $4\langle \ell \rangle D_0 / A_0 = 5.39 \times 10^3$ cm/s. Assuming the appropriateness of Eq. 3 for estimating other species, comparisons are made in Table II of measured ionization efficiencies with those calculated by its use. As noted the agreement for most cases is acceptably good.

4.0 The Combined Thermal Dissociator/Electron Impact Ionization Source

A schematic drawing of the prototype source which will be evaluated for potential use at the HRIBF [1] is shown in Fig. 2. For RIB generation, collimated ^1H , ^2D , ^3He , and ^4He ion beams from the Oak Ridge Isochronous Cyclotron (ORIC) will pass through a thin window where they will interact with the refractory target material chosen for the production of the desired radioactive beam. 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. Radioactive species diffused from the high temperature target will be transported through the resistively heated vapor transport tube maintained at thermionic emission temperatures ($\sim 2150^\circ\text{C}$) and into the solid region of the positively biased W tube which is heated to $\sim 2500^\circ\text{C}$ by acceleration of electrons emitted from the inner surface of the cathode/vapor transport tube. The anode structure of the source is tubular and made of W and is inserted within the vapor transport tube from the extraction end of the source which also serves as the cathode. The anode tube is solid for ~ 20 mm followed by a section with slots in the sides of the tube, also ~ 20 mm in length; the slotted portion of the anode is near the ion extraction end of the tube. The first part of the tube serves as the thermal dissociator while the second part of the tube allows electrons to bombard the dissociated molecular atomic constituents to generate atomic ions which then are extracted from the tube to form the RIB beam. The resulting RIB will be principally in atomic form thereby

increasing the beam intensity of the species of interest. When successfully developed, the source will be a complementary replacement for the electron impact ionization of sources now in use at the HRIBF [7, 8]; the source, therefore, is designed to fit into the same vacuum housing.

5.0 Conclusions

The source shown in Fig. 1 is a prototype design and therefore, it is anticipated that further developments may be needed before arriving at the final design; however, the principles involved in the final iteration will remain unchanged. If successfully developed, this source offers the prospect of overcoming the serious handicap of dilution of the intensities of chemically active species through molecular fragment ionization that is characteristic of hot cathode, electron impact ion sources. Because of the importance of intensity for RIB applications, the source will find many applications in this field, as well as other fields where detection of very low abundance atomic species is crucially important such as in low abundance mass spectrometry. The concept may also be incorporated in more traditional ion sources which rely on the use of compound feed materials.

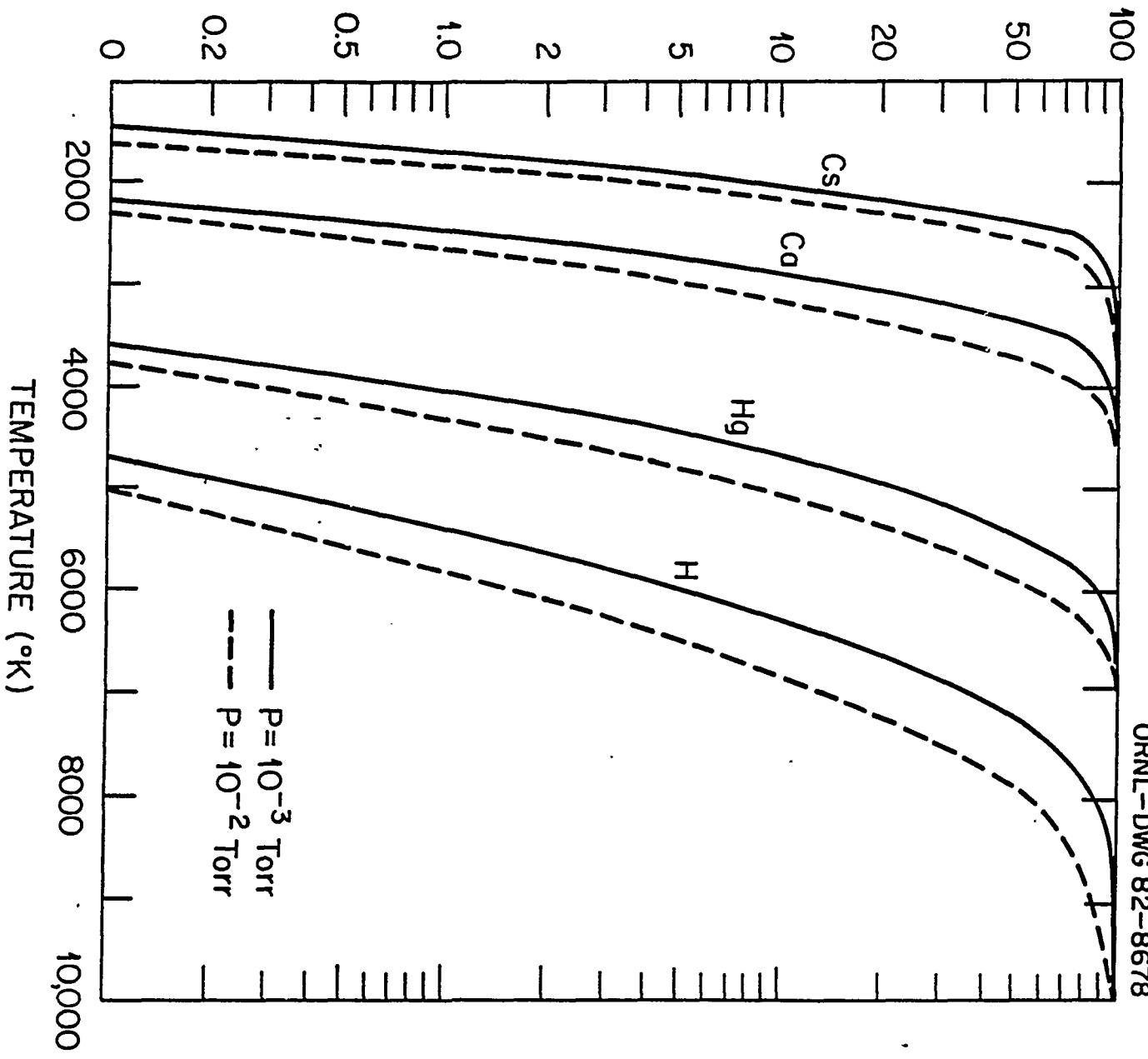
References

1. A Proposal for Physics with Exotic Beams at the Holifield Heavy Ion Research Facility, eds. J. D. Garrett and D. K. Olsen, Physics Division, ORNL, March 1991, unpublished.
2. M. N. Saha, *Philos. Mag.* **40**, 472 (1920).
3. G. J. Beyer, E. Herrmann, A. Peotrowski, V. I. Raiko, and H. Tyroff, *Nucl. Instr. and Meth.* **96**, 347 (1971).
4. P. G. Johnson, A. Bolson, and C. M. Henderson, *Nucl. Instr. and Meth.* **106**, 83 (1973).
5. A. Peotrowski, R. L. Gill, and D. McDonald, *Nucl. Instr. and Meth.* **224**, 1 (1984).
6. R. Kirchner, *Nucl. Instr. and Meth.* **A292**, 203 (1990).
7. G. D. Alton, D. C. Haynes, G. D. Mills, and D. K. Olsen, *Nucl. Instr. and Meth.* **A328**, 325 (1993).
8. G. D. Alton, *Particle Accelerators*, **47** 133 (1994).
9. G. D. Alton and S. Sundell, unpublished.
10. R. Kirchner, *Nucl. Instr. and Meth.* **B70**, 186 (1992).
11. S. Sundell and H. L. Ravn, *Nucl. Instr. and Meth.*, **B70** (1992) 160.
12. J. M. Nitschke, *Nucl. Instr. and Meth.*, **A236**, 1 (1985).

Figure Captions

1. ORNL-DWG 82-8678. Theoretical thermal ionization efficiency versus temperature ($^{\circ}\text{K}$) at two pressures for Cs, Ca, Hg, and H as calculated from Eq. 2.
2. ORNL-DWG 95-10131. Schematic drawing of the tandem thermal dissociator/electron impact ionization source concept for use in sequential dissociation of molecular and electron transport ionization of their atomic constituents. The source will be complementary to the electron impact ionization sources described in Refs. 7 and 8 now in use at the HRIBF [1].

THERMAL IONIZATION EFFICIENCY, η (%)



ORNL-DWG 82-8678

Fig. 1

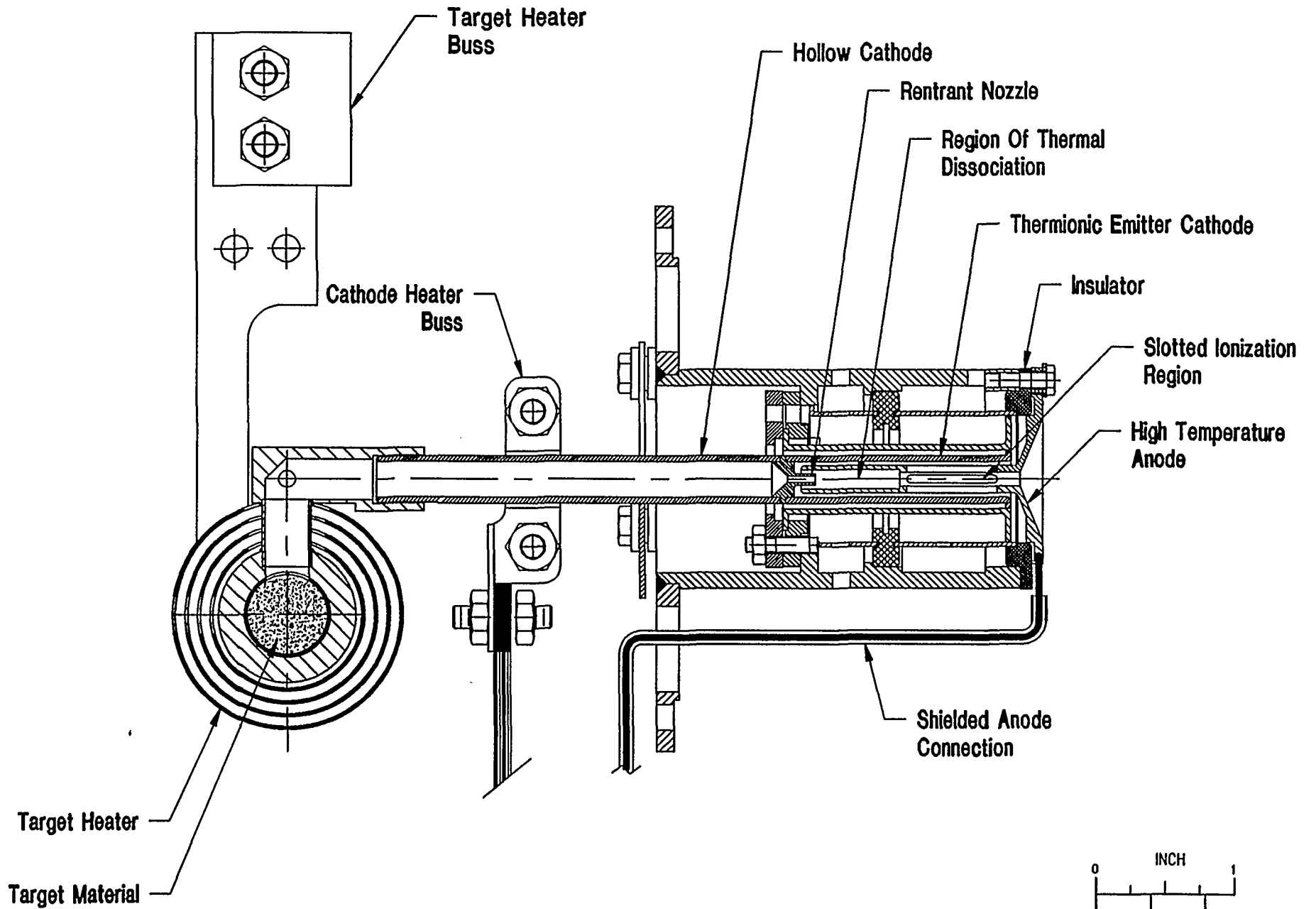


Fig. 2

Table I. Thermal Ionization efficiencies for various elements (From Ref. 4)

Element No.	Sample Form	Ion beam	Yield data (% of initial charge)		
			Low	High	Average
20	CaO	Ca ⁺		13.2	13.2
38	SrCO ₃	Sr ⁺	20.4	74.0	44.1
39	Y ₂ O ₃	Y ⁺	4.6	27.5	16.0
41	Nb ₂ O ₅	Nb ⁺	0.12	15.0	2.31
57	La ₂ O ₃	La ⁺	10.0	41.0	24.6
58	CeO ₂	Ce ⁺	-	36.0	36.0
59	Pr ₆ O ₁₁	Pr ⁺	21.4	40.3	33.0
60	Nd ₂ O ₃	Nd ⁺	19.9	61.0	42.9
61	Pm ₂ O ₃	Pm ⁺	50.5	93.5	75.3
62	Sm ₂ O ₃	Sm ⁺	57.0	71.5	65.9
63	Eu ₂ O ₃	Eu ⁺	13.1	73.0	45.8
64	Gd ₂ O ₃	Gd ⁺	18.0	28.0	23.0
65	Tb ₂ O ₃	Tb ⁺	17.4	51.8	28.8
66	Dy ₂ O ₃	Dy ⁺	33.3	51.8	43.8
67	Ho ₂ O ₃	Ho ⁺	26.6	44.4	32.5
68	Er ₂ O ₃	Er ⁺	28.1	38.1	32.6
69	Tm ₂ O ₃	Tm ⁺	20.6	37.6	32.4
70	Yb ₂ O ₃	Yb ⁺	8.4	23.0	14.9
71	Lu ₂ O ₃	Lu ⁺	14.5	39.2	19.5
92	U ₃ O ₈	U ⁺	10.8	20.2	15.1
94	PuO ₂	Pu ⁺	7.9	24.4	16.2
96	CmO ₂	Cm ⁺	1.0	14.8	8.8

Table II. Comparisons of Calculated and Experimentally Measured Ionization Efficiencies η for Electron Beam Plasma Ion Sources. Estimated ionization efficiencies were calculated by using Eq. 3.

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	12
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	9
54	^{132}Xe	12.13	8	54.8	56	10
79	^{197}Au	9.23	1	32.9	50	10
83	^{209}Bi	7.29	5	82.9	68.3	10