

A COMBINED THERMAL DISSOCIATION AND ELECTRON IMPACT IONIZATION SOURCE FOR RIB GENERATION

G. D. Alton and C. Williams¹
Oak Ridge National Laboratory
P. O. Box 2008
Oak Ridge, Tennessee 37831-6368

Abstract

The probability for simultaneously dissociating and efficiently ionizing the individual atomic constituents of molecular feed materials with conventional, hot-cathode, electron-impact ion sources is low and consequently, the ion beams from these sources often appear as mixtures of several molecular sideband beams. This fragmentation process leads to dilution of the intensity of the species of interest for RIB applications where beam intensity is at a premium. We have conceived an ion source that combines the excellent molecular dissociation properties of a thermal dissociator and the high ionization efficiency characteristics of an electron impact ionization source that will, in principle, overcome this handicap. The source concept will be evaluated as a potential candidate for use for RIB generation at the Holifield Radioactive Ion Beam Facility (HRIBF), now under construction at the Oak Ridge National Laboratory. The design features and principles of operation of the source are described in this article.

1.0 Introduction

During on-line production of radioactive species, chemically active elements, which evolve through diffusion from high temperature target materials, are, more often than not, transported in a variety of molecular forms to the ionization chamber of the source. Because of the low probability of simultaneously dissociating the molecule and efficiently ionizing the atomic constituents in these sources, the species of interest may be present in a variety of molecular ion beam fragments; this fragmentation process leads to a dilution of the intensity of the species of interest. For high sensitivity mass spectrometry, on-line isotope separation and radioactive ion beam (RIB) applications, intensity is at a premium and therefore, it is extremely important to concentrate as much of the species of interest as possible into a single mass channel. The source concept described in this article was conceived for potential use at the Holifield Radioactive Ion Beam Facility [1], now under construction at the Oak Ridge National Laboratory, in an effort to increase the intensity of RIBs by reducing loss channels due

¹ Gilbert/Commonwealth Engineers & Consultants, Oak Ridge, TN.

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to molecular ion beam fragmentation. If the ion source concept proves to be a viable option, it will be used as a complement to the electron beam plasma ion sources already in use at the HRIBF [2,3]. The design features and principles of operation of the source are presented in this article.

2.0 Theory of Thermal Ionization

Collisions between gas particles at high temperatures may produce ionization, provided that their relative energies exceed the first ionization of the atoms or molecules that make up the gas. When the species is a monotonic gas, a fraction of the total number of 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 [4]. 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 pressures, respectively, of $p = 1 \times 10^{-3}$ and 1×10^{-2} Torr. The first reported applications of this technique are described in Refs. [5] and [6]. Such sources are relatively efficient for elements with ionization potentials $I_p \leq \sim 7$ eV. While the theory defined by Eq. 2 was derived for the case of thermal ionization of atomic species, it holds analogously for the case of molecular dissociation.

Table I compiles ionization efficiencies for a number of elements, including Ca, Sr, Nb, and La, all of the lanthanides, and U, Pu, and Cm as measured by the thermal ionization source described in Ref. [6]. Because of its chemical selectivity and efficiency for ionizing elements with relatively low ionization potentials ($I_p < 7.0$ eV), the source has been utilized for ionization of short-lived nuclei created on-line by

neutron activation [7] or by light ion transmutation of target materials [8]. While the source is used exclusively as a thermal ionization source, Table 1 clearly illustrates how effective the thermal process is for dissociation of strongly bound molecules.

3.0 Estimates of Electron Impact Ionization

The efficiencies of the electron impact ionization type ion sources, such as described in Refs. 2 and 3, 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. [9], 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 for the noble gases:

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

Where $\langle e \rangle$ 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 the FEBIAD ion source: $\langle kT_e \rangle = 3 \text{ eV}$; $T_i = 2273^\circ\text{K}$; and $4\langle\ell\rangle D_o A_o = 5.39 \times 10^3 \text{ cm/s}$.

Assuming the appropriateness of Eq. 3 for estimating other species, comparisons are made in Table II of measured ionization efficiencies with those calculated by use of Eq. 3. As noted the agreement for most cases is acceptably good.

4.0 The Combined Thermal Dissociator/Electron Impact Ionization Source

Schematic drawings of two prototype source concepts which will be evaluated for potential use at the HRIBF [1] are shown, respectively, in Figs. 2 and 3. The design details and principles of operation of these sources are given below.

For RIB generation, collimated ^1H , ^2D , ^3He , and ^4He ion beams from the Oak Ridge Isochronous Cyclotron (ORIC) will pass through a thin entrance window in the target material reservoir 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 ($\sim 2150^\circ\text{C}$). For efficient molecular

dissociation, the molecules must be heated to $\sim 2500^{\circ}\text{C}$; two source concepts have been proposed which will be evaluated in terms of their effectiveness for dissociating and ionizing the atomic constituents of molecules.

For the source shown in Fig. 1, the molecules enter a tubular, W anode which is positively biased with respect to the outer tube which serves as a hollow-cathode. The anode is inserted within the vapor transport tube from the extraction end of the source; the anode tube is heated to $\sim 2500^{\circ}\text{C}$ by acceleration of electrons emitted from the inner surface of the hollow-cathode. The anode tube is solid for ~ 20 mm which serves as the molecular dissociator. Following dissociation, the atomic constituents then enter a slotted part of the anode, also ~ 20 mm in length, where they are subjected to electron bombardment prior to extraction. Ion beams are formed by extraction through a 2 mm aperture in the extraction end of the anode tube. Because of the radial direction of electron bombardment, a magnetic field will not be used in this source version.

In the second version of the source, shown in Fig. 3, molecules are dissociated during passage through a capillary bored through a ~ 20 mm length of the solid portion of the vapor transport tube which is heated to $\sim 2500^{\circ}\text{C}$ by electron bombardment from a negatively biased filament positioned around the W tube. The dissociated atomic constituents then pass into a tubular anode where they are subjected to bombardment by electrons emitted by the high temperature end of the vapor transport tube to form the ion beam. The ability to independently control the electron energies used to heat the dissociator and those used to ionize the dissociation products is viewed as an advantage of this design over the source represented schematically in Fig. 2. For this version of the source, a weak magnetic field may be used to collimate the electron beam accelerated into the hollow anode.

5.0 Conclusions

Both high dissociation fractions and high ionization efficiencies are simultaneously possible by use of either of the source concepts displayed, respectively, in Figs. 2 and 3. If successfully developed, molecular-fragment ion beams will be avoided, and as a consequence, the ion beam intensity of the species of interest will be increased. These source concepts offer the prospect of overcoming the serious handicap of dilution of the intensity of chemically active species by concentrating the species of interest into a single mass channel. 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. Following successful development, the source will be a complementary replacement for the electron impact ionization of sources now in use at the HRIBF [2,3].

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Figure Captions

Fig 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.

Fig. 2. ORNL-DWG. 95-10131. Schematic drawing of the combined thermal dissociator/electron impact ionization source concept which utilizes a hollow cathode electron emitter for both dissociating and ionizing the atomic constituents. The source will be complementary to the electron impact ionization sources described in Refs. 2 and 3 now in use at the HRIBF [1].

Fig 3. ORNL-DWG. ???. Schematic drawing of the combined thermal dissociator/electron impact ionization source concept which utilizes a negatively biased electron emitter for heating a capillary molecular dissociator followed by a hot cathode/hollow anode structure for ionizing the atomic constituents of the dissociated molecules. The source will be complementary to the electron impact ionization sources described in Refs. 2 and 3 now in use at the HRIBF [1].

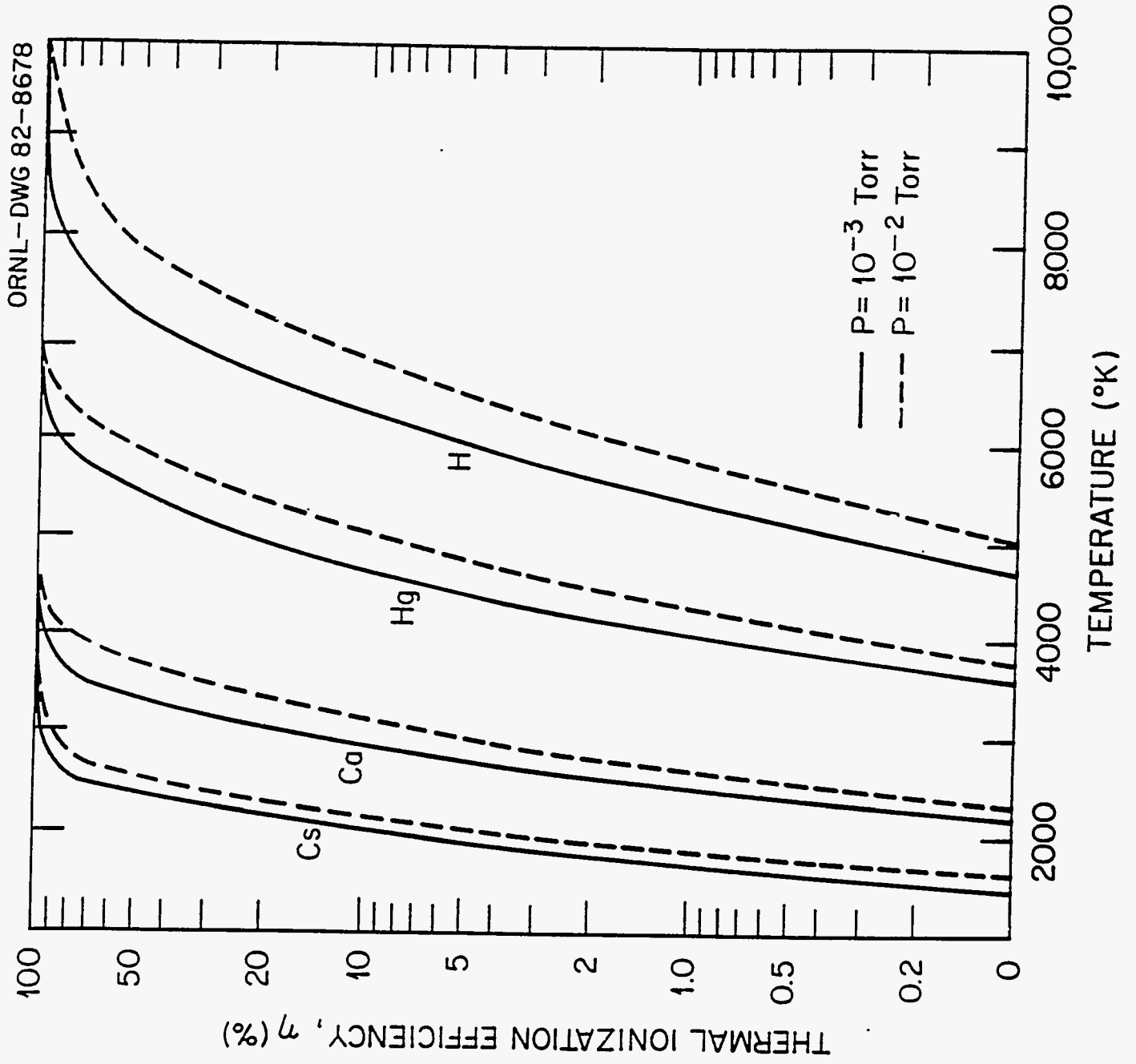


Fig. 1

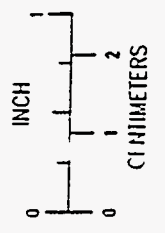
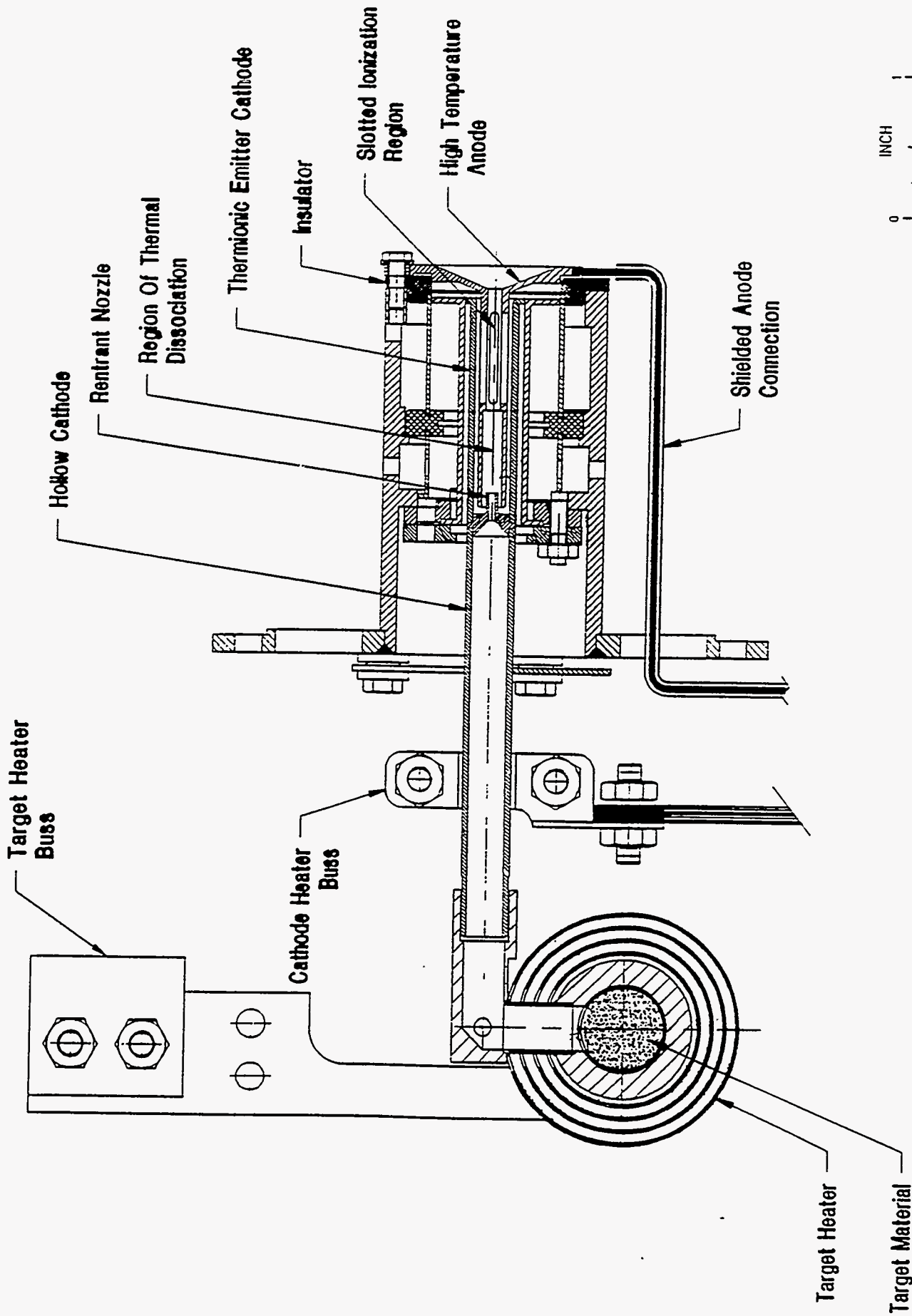


Fig. 2

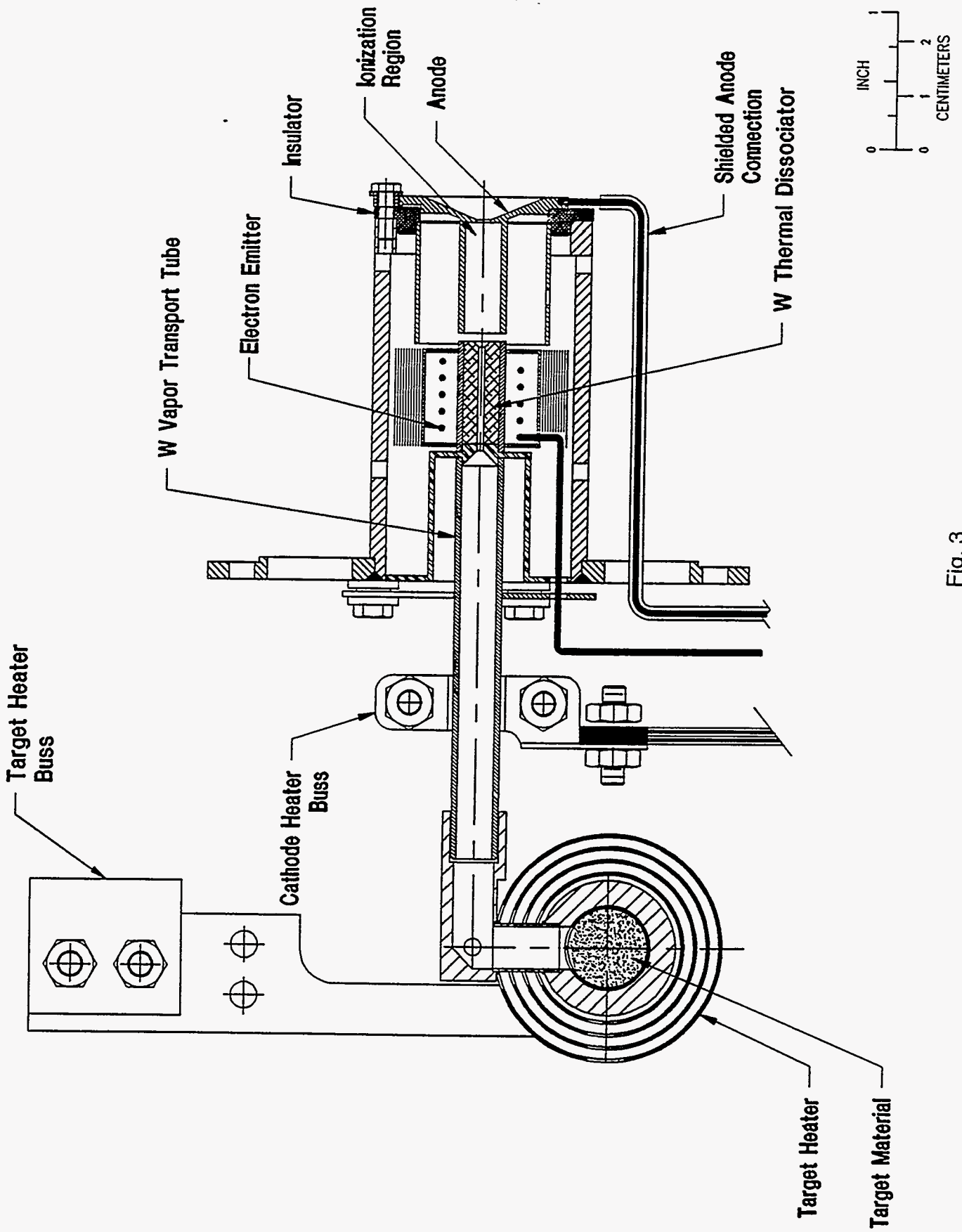


Fig. 3

Table 1. Thermal Ionization efficiencies for various elements (From Ref. 6)

Sample Element No.	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 2. Comparisons of Calculated and Experimentally Measures Ionization Efficiencies η for Electron Beam Plasma Ion Sources. Estimated ionization efficiencies were calculated by using Eq. 3.

Z	Element	I_p (eV)	N_e	$\eta_{\text{calc}}(\%)$	$\eta_{\text{exp}}(\%)$	Ref.
10	²⁰ Ne	21.56	8	2.0	1.6	9
18	⁴⁰ Ar	15.76	8	16.6	19	9
24	⁵⁴ Cr	6.77	1	37.0	>20	9
26	⁵⁷ Fe	7.90	2	45.2	30	9
32	⁷⁶ Ge	7.90	4	65.6	41	9
36	⁸⁴ Kr	14.00	8	34.1	35	9
36	⁸⁴ Kr	14.00	8	34.1	36	11
46	¹⁰⁰ Pd	8.33	18	89.5	>25	9
47	¹⁰⁷ Ag	7.58	1	38.7	47	9
47	¹⁰⁹ Ag	7.58	1	38.9	50	9
50	¹¹⁶ Sn	7.34	4	74.0	53	9
50	¹²⁴ Sn	7.34	4	74.6	54	9
54	¹²⁹ Xe	12.13	8	54.6	52	9
54	¹³² Xe	12.13	8	54.8	53	10
54	¹³² Xe	12.13	8	54.8	56	9
79	¹⁹⁷ Au	9.23	1	32.9	50	9
83	²⁰⁹ Bi	7.29	5	82.9	68.3	9