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A COMBINATION THERMAL DISSOCIATION/ELECTRON IMPACT IONIZATION SOURCE FOR RIB GENERATION

G. D. Alton¹, B. Cui², R. F. Welton³, C. Williams⁴,
Oak Ridge National Laboratory, * P. O. Box 2008, Oak Ridge, Tennessee 37831-6368

Abstract

The flourishing interest in radioactive ion beams (RIBs) with intensities adequate for astrophysics and nuclear physics research place a premium on targets that will swiftly release trace amounts of short lived radio-nuclei in the presence of bulk quantities of target material and ion sources that have the capability of efficiently ionizing the release products. Because of the low probability of simultaneously dissociating and efficiently ionizing the individual atomic constituents of molecules containing the element of interest with conventional, hot-cathode, electron-impact ion sources, the species of interest is often distributed in several mass channels in the form of molecular sideband beams and, consequently, the intensity 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 to address these problems. 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.

PACS:

Keywords: Thermal dissociation, ion source, thermal ionization, electron impact ionization

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¹ Corresponding author. Address: Oak Ridge National Laboratory, P. O. Box 2008, Oak Ridge, TN 37831-6368 USA. Phone: 423-574-4751, FAX: 423-574-1268, E-mail: gda@mail.phy.ornl.gov.

² China Institute of Atomic Energy, Beijing, China.

³ ORISE, Oak Ridge, TN.

⁴ Advanced Design Services, Oak Ridge, TN.

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A COMBINATION THERMAL DISSOCIATION/ELECTRON IMPACT IONIZATION SOURCE FOR RIB GENERATION

G. D. Alton⁵, B. Cui⁶, R. F. Welton⁷, C. Williams⁸,
Oak Ridge National Laboratory, P. O. Box 2008, Oak Ridge, Tennessee 37831-6368

1. Introduction

For ISOL and radioactive ion beam (RIB) research, radioactive nuclei, formed within the bulk of a selectively chosen target material by charged particle or neutron irradiation, must be diffused from the target material and transported to the ionization chamber of an ion source in a time span short with-respect-to the lifetime of the species. Chemically active species diffused from the target material often arrive at the ionization chamber of the source in a variety of molecular forms. Because of the low probability of simultaneously dissociating molecules and efficiently ionizing the atomic constituents in conventional hot cathode electron impact ion sources, the species of interest will be present in a variety of molecular fragment ion beams, and, as a consequence, the intensity of the species of interest will be diluted. For high sensitivity mass spectrometry, ISOL studies and RIB applications, where intensity of the species of interest is of paramount importance, it is highly desirable to eliminate these sidebands and concentrate as much of the species of interest as possible into a single mass channel. The combined thermal dissociation/electron impact ionization source, described in this article, offers the potential of simultaneously dissociating molecules and ionizing their atomic constituents efficiently. The source was specifically designed for potential use in generating RIBs at the Holifield Radioactive Ion Beam Facility [1] and if successful will be complementary to the modified CERN-ISOLDE type electron beam plasma ion sources (EBPISs) described in Refs. 2 and 3.

2. The Combined Thermal Dissociator/Electron Impact Ionization Source

Source Design. A schematic drawing of the prototype source, which will be evaluated for potential use at the HRIBF [1], is shown in Fig. 1. For RIB generation, collimated ^1H , ^2D , ^3He , and ^4He ion beams from the Oak Ridge Isochronous Cyclotron (ORIC) will pass through a thin window in the target reservoir where they will interact with 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 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 W hollow-cathode/vapor transport tube maintained at thermionic emission temperatures ($\sim 2200^\circ\text{C}$) and into the positively biased W anode which is heated to $\sim 2500^\circ\text{C}$ by acceleration of electrons emitted from the inner surface of the hollow-cathode. The anode structure of the source is inserted into the vapor transport tube from the ion extraction end of the source. The anode tube is solid for ~ 17.6 mm followed by a section with slots in the sides of the tube, also ~ 17.6 mm in length; the slotted portion of the anode is near the ion extraction end of the tube. The first part of the anode serves as the molecular dissociator while the second part of the anode allows electrons to bombard atomic constituents and form ions which are then extracted from the source.

⁵ Corresponding author. Address: Oak Ridge National Laboratory, P. O. Box 2008, Oak Ridge, TN 37831-6368 USA. Phone: 423-574-4751, FAX: 423-574-1268, E-mail: gda@mail.phy.ornl.gov.

⁶ China Institute of Atomic Energy, Beijing, China.

⁷ ORISE, Oak Ridge, TN.

⁸ Advanced Design Services, Oak Ridge, TN.

Anodic heating. The anode cylinder will be raised to thermal dissociation temperatures ($\sim 2500^\circ\text{C}$) by accelerating space charge limited electron beams, emitted by the hollow cathode, to the anode structure where they will dissipate their energies in the form of heat. The radially directed electron beam that passes through the slotted end of the anode structure, in combination with the axially directed electron beam, will be used to ionize the atomic constituents of dissociated molecules. Because of the importance of the temperature in dissociating molecules and the fact that the dissociation temperatures are molecule specific, the relationship between electron beam power and anode structure temperature is of primary importance.

Analytical approximations can be used to derive a correlation between electron beam power and anode temperature using the Langmuir-Blodgett relation for rectilinear flow between two closely spaced coaxial cylinders [4]. The space charge limited electron current I_c emitted by a length ℓ of the hollow cathode at potential V can be expressed by the relation

$$I_c = \frac{8\pi}{9} (2\eta)^{1/2} \epsilon_0 \frac{V^{3/2} \ell}{\beta^2 r} = P_c V^{3/2} \quad (1)$$

where P_c is the perveance of the electrode system, $\eta = e/m$ and ϵ_0 is the permittivity of free space. In Eq. 1, the function $\beta(r/r_c)$ is given by the series

$$\beta = \mu - 0.4\mu^2 + 0.092\mu^3 - 0.014\mu^4 + \dots \quad (2)$$

with $\mu = \ln(r/r_c)$ where r_c is the radius of the cathode.

Values for β^2 have been tabulated by several authors including those of Ref. 5. For the cathode/anode electrode structure, $\beta^2 \cong 0.071$. The perveance for the hollow-cathode/cylindrical anode structure is obtained by substituting β^2 into Eq. 1. The value for P_c is found to be

$$P_c = 2.5 \times 10^{-3} [A/V^{3/2}] \quad (3)$$

This value agrees identically with the results obtained from computational analyses of the electrode system using the program described in Ref. 6. Figure 2 displays equipotentials and electron trajectories derived by use of the program for simulation of anodic heating by electron impact.

The anode structure is heated both by radiation from the surrounding hollow-cathode which operates at $\sim 2200^\circ\text{C}$ and by the electron beam accelerated from the hollow-cathode tube to the anode surface. Cooling of the anode structure occurs by radiation to the hollow-cathode tube and to the surrounding vacuum enclosure walls. A rough estimation can be made of the power required to heat the anode to 2500°C from the Stefan-Boltzmann law of radiation if we assume that heating occurs solely by electron impact and cooling is solely through radiation losses to the surrounding walls. The Stefan-Boltzmann relation is given by

$$P = A_c \sigma \epsilon (T_1^4 - T_2^4) \quad (4)$$

where A_c is the radiating area of the W anode with emissivity ϵ at temperature T_1 , T_2 is the temperature of the surrounding body and σ is Stefann's constant with value

$$\sigma = 5.66 \times 10^{-8} [Wm^{-2} deg^{-4}] . \quad (5)$$

The power P required to heat the anode to $2500^{\circ}C$ is estimated by assuming that the surrounding surface to which the cylindrical anode radiates is at $2200^{\circ}C$; that to which the front side (ion emission side) of the anode mounting flange radiates is at $50^{\circ}C$ and those to which the back side of the anode mounting flange radiates are at $1350^{\circ}C$. Then the power P required to heat the anode to $2500^{\circ}C$ is $P_{max} = 1550$ W.

By equating the power-loss P through radiation to the power imparted to the anode structure by the accelerated electron beam, the voltage V that must be applied to the anode to heat it to $2500^{\circ}C$ can be obtained from the product of voltage V and Eq. 1 or

$$P = P_c V^{5/2} \quad (6)$$

From this expression, we find the maximum voltage required to heat the anode to $2500^{\circ}C$ to be

$$V = \{P/P_c\}^{2/5} = 213 \text{ V} . \quad (7)$$

The corresponding space-charge-limited current calculated from Eq. 1 is found to be $I_c = 7.7$ A. According to the Richardson-Dushman equation, the temperature limited electron current from the W hollow-cathode, when operated at $2200^{\circ}C$, is ~ 14 A.

The electron beam must simultaneously heat the anode to $2500^{\circ}C$ while efficiently ionizing the element in question. Since the optimum electron energy for ionization is the order of $5x$ the first ionization potential of the element of interest, the electron energy is reasonably compatible with this requirement, as well. The resulting RIB will be principally in atomic form, thereby increasing the beam intensity of the species of interest.

Ion Optics. The beam intensities from the present source when used for RIB generation will be of the order of a few μA and since the initial angular divergence of the beam is important for beam transport and for locating the first beam transport lens, a low value is desirable. We have used the code described in Ref. 6 to iteratively design the extraction electrode boundary. Figure 3 displays ion trajectories and equipotentials as well as an emittance diagram for the source. The angular divergence of the extracted beam is found to be reduced by a factor of ~ 2 over that calculated for the conventional electron beam plasma ion sources (EBPISs), described in Refs. 2 and 3, by extracting from a hemispherical focus electrode boundary rather than a slightly tapered surface.

3. Theory of Molecular Dissociation

Molecular dissociation of a homonuclear diatomic molecule at high temperatures is a form of chemical equilibrium that can be expressed symbolically through the relation

$$\sum_i v_i A_i = 0 \quad , \quad (8)$$

where A_i are the chemical symbols and v_i are positive or negative integers. For example, the equation for a diatomic molecule in chemical equilibrium at constant temperature and pressure can be written as

$$AB - A - B = 0 \quad (9)$$

The chemical potential for the gas mixture can be expressed as

$$\mu_i = kT \log p_i + \chi_i(T) \quad (10)$$

where p_i is the partial pressure and χ_i the temperature dependent part of the chemical potential of the i^{th} component of the mixture; $p_i = \frac{N_i}{N} p$ where N_i is the number of gas molecules of a specific gas and $N = \sum N_i$ is the total number of gas molecules in the mixture, k is Boltzmann's constant, T is the temperature. Correspondingly, the condition for chemical equilibrium of a mixture of gases at temperature T requires that the sum of the chemical potentials μ_i for the individual components which make up the mixture obey the following relation

$$\sum_i v_i \mu_i = kT \sum_i v_i \log p_{0i} + \sum_i v_i \chi_i = 0 \quad (11)$$

where the p_{0i} are the partial pressures of gases in a state of chemical equilibrium. The rate coefficient $K_p(T)$ can be expressed as

$$K_p(T) = e^{-\sum v_i \chi_i / kT} \quad (12)$$

so that

$$\prod_i p_{0i}^{v_i} = K_p(T) \quad (13)$$

When the gases used are in chemical equilibrium, we can express Eq. 13 in the form

$$\prod_i c_{0i}^{v_i} = p_{0i}^{-\sum v_i} K_p(T) = K_c(p, T) \quad (14)$$

where $c_{0i} = \frac{N_i}{N}$ is the concentration of the molecule of interest. The quantity $K_c(p, T)$ is called the chemical equilibrium constant and Eq. 13 or 14 is referred to as the law of mass action. For gases with constant specific heats, the function χ_i has the form

$$\chi_i(T) = \varepsilon_{0i} - c_{pi} kT \log kT - kT \xi_i \quad (15)$$

where ε_{0i} are the energy bands of the i^{th} molecule with specific heat c_{pi} and ξ_i is the chemical constant of the gas. By substituting Eq. 15 into Eq. 12, the rate coefficient $K_p(T)$ can be written as

$$K_p(T) = e^{\sum v_i \xi_i} (kT)^{\sum c_{pi} v_i} e^{-\sum v_i \varepsilon_{0i}/kT} \quad (16)$$

The law of mass action can be used to derive the degree of dissociation for a diatomic molecule, assumed to be in its ground vibrational and rotational states. The degree of dissociation η is defined as the ratio of the number of dissociated molecules to the total number of molecules which would exist in a non-dissociating gas. Accordingly, the degree of dissociation can be written as

$$\eta = N_A/2N_{AB}^0 = N_B/2N_{AB}^0 \quad (17)$$

where $N_{AB}^0 = N_{AB} + \frac{1}{2}N_A = N_{AB} + \frac{1}{2}N_B$. When applied to the dissociation of a diatomic molecule of identical atoms, Eq. 17 becomes

$$N_{AA}^0 = N_{AA} + \frac{1}{2}N_A \quad (18)$$

The rate constant $K_p(T)$ for the dissociation of a homonuclear diatomic molecule becomes

$$K_p(T) = p_{AA}/p_A^2 = \frac{N_{AA}(N_A + N_{AA})}{pN_A^2} = \frac{1-\eta^2}{4\eta^2 p} \quad (19)$$

and, therefore,

$$\eta = [1 + 4pK_p(T)]^{-1/2} \quad (20)$$

Substituting the specific heat for the dissociated atoms $c_{pA} = 5/2$ and the undissociated molecule $c_{pAA} = 9/2$ into Eq. 16 along with their respective chemical constants, $\xi_A = \log[g_A(M/2\pi\hbar^2)^{3/2}]$, and $\xi_{AA} = \log[(I/\hbar^6\omega)(M/\pi)^{3/2}]$. The rate constant now can be written

$$K_p(T) = \frac{8I\pi^{3/2}}{g_A^2\omega M^{3/2}(kT)^{1/2}} e^{\varepsilon_0/kT} \quad (21)$$

where ε_0 is the dissociation energy of the molecule, ω is the vibration frequency and I is the moment of inertia of the molecule M is the mass of each atom, g_A is the statistical weight for the

ground state of the atom A , given by $g_A = (2S + 1)(2L + 1)$ where S is the total spin and L the total angular momentum quantum numbers. Substituting Eq. 21 into Eq. 20, we arrive at the following equation for the dissociation of a homonuclear diatomic molecule

$$\eta = \left[1 + \frac{32pI\pi^{3/2}e^{\epsilon_0/kT}}{g_A^2\omega M^{3/2}(kT)^{1/2}} \right]^{-1/2} \quad (22)$$

For more details of thermal chemical equilibrium processes, reference is made to a number of statistical physics text books where these derivations can be found, including Ref. 7.

Al_2O_3 target material has been selected as the prime candidate for the production of $^{17,18}\text{F}$ through the respective fusion reactions $^{16}\text{O}(d,n)^{17}\text{F}$ and $^{18}\text{O}(p,n)^{18}\text{F}$ at the HRIBF [1]. The product species will most likely be released from the target material in the form of AlF . Because of the low probability of simultaneously dissociating the molecules and efficiently ionizing the atomic constituents (Al^+ and F^+) in conventional hot cathode electron impact ion sources, the species of interest (F) may be present in a variety of molecular fragment ion beams, and, as a consequence, the intensity of the species of interest will be diluted. To illustrate that this is indeed the case, the mass spectral data of beams extracted from a modified CERN-ISOLDE electron impact ionization source (EBPIS) [2, 3] that will be used on-line for generating RIBs at the HRIBF [1], are shown in Fig. 4. These data were obtained by passing very low flow rates of F , in the form of SF_6 , through hot Al_2O_3 located in the target chamber of an EBPIS.

Thermal dissociation provides a means for reducing the molecular sideband problem provided the ion source can be operated at sufficiently high temperatures so that the molecules are dissociated. Figure 5 displays thermal dissociation fractions versus temperature of the most likely release products during production of $^{17,18}\text{F}$, AlF and AlOF , as calculated by use of the thermodynamic code Thermo-Calc [8]. As indicated by Eq. 22, thermal dissociation strongly depends on the binding energy of the molecule ϵ_0 .

3. Theory of Thermal Ionization

At high temperatures, collisions between gas particles may produce ionization, provided that their relative energies exceed the first ionization potential of the atoms or molecules that make up the gas. For the case of a monatomic 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 (Eq. 14) can be used to derive an expression for the ionization efficiency as a function of temperature and pressure [9]. The ionization efficiency for an atom with first ionization potential I_p can be expressed in the following form:

$$\eta = \left[1 + p \frac{g_0}{2g_1} \left(\frac{2\pi\hbar^2}{m} \right)^{3/2} \frac{\exp(I_p/kT)}{(kT)^{5/2}} \right]^{-1/2} \quad (24)$$

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 particular species. 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 elements with low to intermediate ionization potentials. Figure 6 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. 10 and 11. 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. 11. All of these elements have ionization potentials ≤ 6.3 eV. While the source has been used exclusively as a thermal ionization source, Table 1 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 [12] and by light-ion transmutation of target materials [13].

4. Estimates of Electron Impact Ionization

The efficiencies of EBPISSs, such as described in Refs. 14-16, 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. 15, 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:

$$\eta_{calc} = \frac{4\langle\ell\rangle D_0 N_e}{A_0} \left(\frac{\pi M_i}{8kT_i} \right)^{1/2} \exp\{-I_p/\langle kT_e \rangle\} \left/ \left[1 + \frac{4\langle\ell\rangle D_0 N_e}{A_0} \left(\frac{\pi M_i}{8kT_i} \right)^{1/2} \exp\{-I_p/\langle kT_e \rangle\} \right] \right. \quad (25)$$

where $\langle\ell\rangle$ is the average path length for a particle in the plasma; D_0 is a constant (cm^2/s); A_0 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 the species. The following values are used for terms in Eq. 25 when estimating ionization efficiencies for the EBPISS: $\langle kT_e \rangle = 3$ eV; $T_i = 2273$ K; and $4\langle\ell\rangle D_0/A_0 = 5.39 \times 10^3$ cm/s. Assuming the appropriateness of Eq. 25 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.

5. 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. If successfully developed, the source will find many applications for RIB generation where intensity is crucially important as well as other fields where the detection of very low abundance atomic species is required, 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.

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Figure Captions

1. ORNL DWG 96-7982. Schematic drawing of the combined 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. 2 and 3 now in use at the HRIBF [1].
2. ORNL-DWG 96M-7977(R2). Electron trajectories and equipotentials of the cathode-anode region of the combination thermal dissociation/electron impact ionization source shown schematically in Fig. 1. Anode-cathode voltage: $V = 213$ V; anode-cathode electron current: 7.7A; Perveance, P_c : $P_c = 2.5 \times 10^{-3}$ [A/V^{3/2}].
3. ORNL DWG 96M-6675B. Ion trajectories and equipotentials as well as an emittance diagram for the thermal dissociation/electron impact ionization source displayed in Fig. 1. Ion beam energy : 20 keV. The simulation code described in Ref. 6 was used to iteratively design the extraction system.
4. ORNL-DWG 96M-7314. Mass spectra of beams extracted from a modified CERN-ISOLDE electron impact ionization source (EBPIS) with Al₂O₃ as the target material and SF₆ as the fluorinating agent. (See Refs. 2 and 3 for more details on this source type).
5. ORNL-DWG 96-7315. Thermal dissociation of AlF, HF, and AlOF as calculated by use of Thermo-Calc [8].
6. ORNL-DWG 95M-8536B. Theoretical thermal ionization efficiency versus temperature at two pressures for Cs, Ca, Hg, and H as calculated by use of Eq. 25.

Table 1. Thermal Ionization efficiencies for various elements (From Ref. 11)

Sample			Yield data (% of initial charge)		
Element No.	Form	Ion beam	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 measured ionization efficiencies η for electron beam plasma ion sources (EBPISs). Estimated ionization efficiencies were calculated by using Eq. 25.

Z	Element	I_p (eV)	N_e	$\eta_{\text{calc}}(\%)$	$\eta_{\text{exp}}(\%)$	Ref.
10	^{20}Ne	21.56	8	2.0	1.6	15
18	^{40}Ar	15.76	8	16.6	19	15
24	^{54}Cr	6.77	1	37.0	>20	15
26	^{57}Fe	7.90	2	45.2	30	15
32	^{76}Ge	7.90	4	65.6	41	15
36	^{84}Kr	14.00	8	34.1	35	15
36	^{84}Kr	14.00	8	34.1	36	16
46	^{100}Pd	8.33	18	89.5	>25	15
47	^{107}Ag	7.58	1	38.7	47	15
47	^{109}Ag	7.58	1	38.9	50	15
50	^{116}Sn	7.34	4	74.0	53	15
50	^{124}Sn	7.34	4	74.6	54	15
54	^{129}Xe	12.13	8	54.6	52	15
54	^{132}Xe	12.13	8	54.8	53	15
54	^{132}Xe	12.13	8	54.8	56	14
79	^{197}Au	9.23	1	32.9	50	15
83	^{209}Bi	7.29	5	82.9	68.3	15

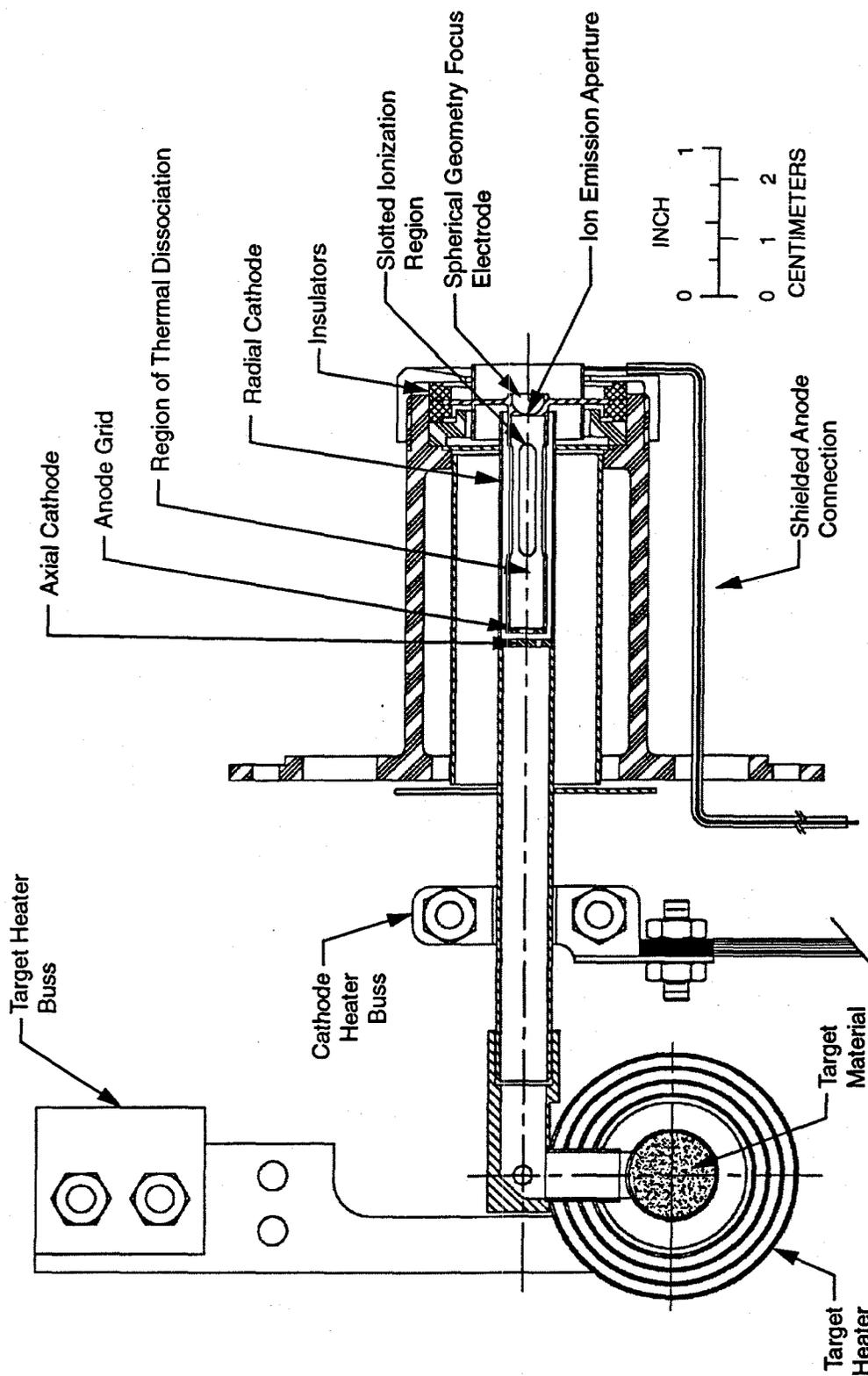


Fig.1

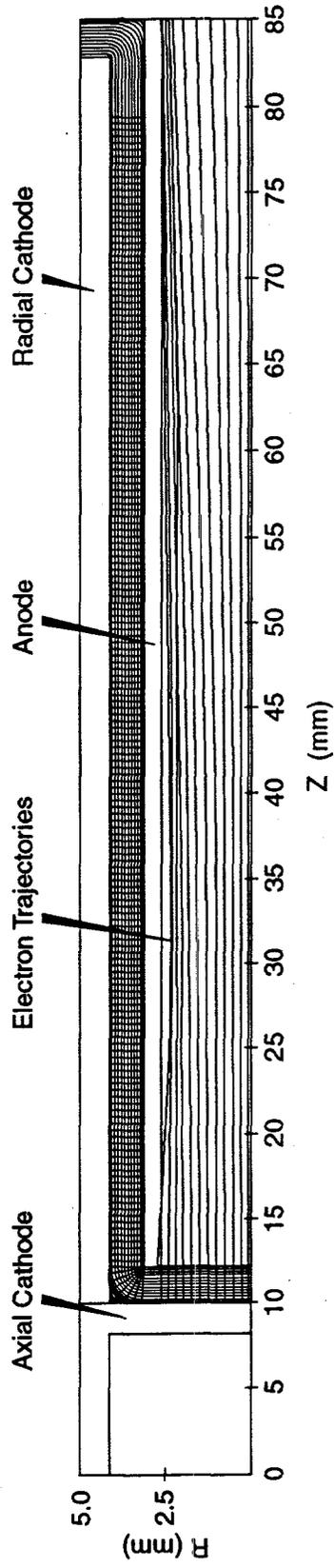


Fig. 2

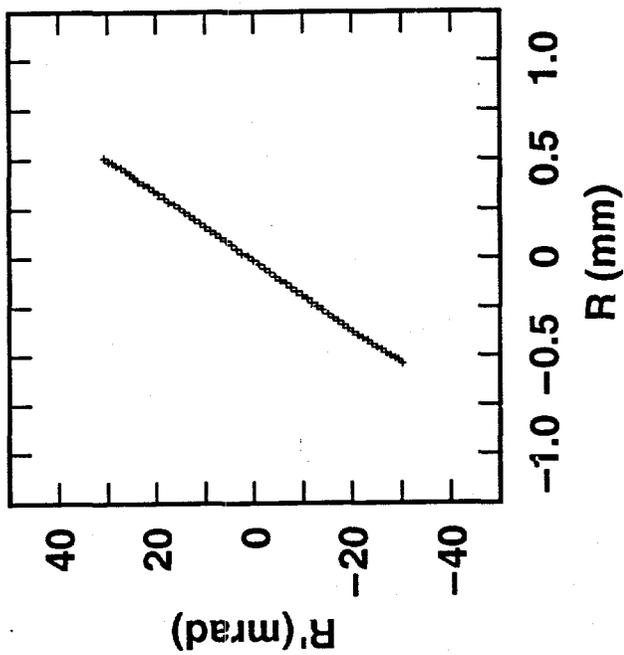
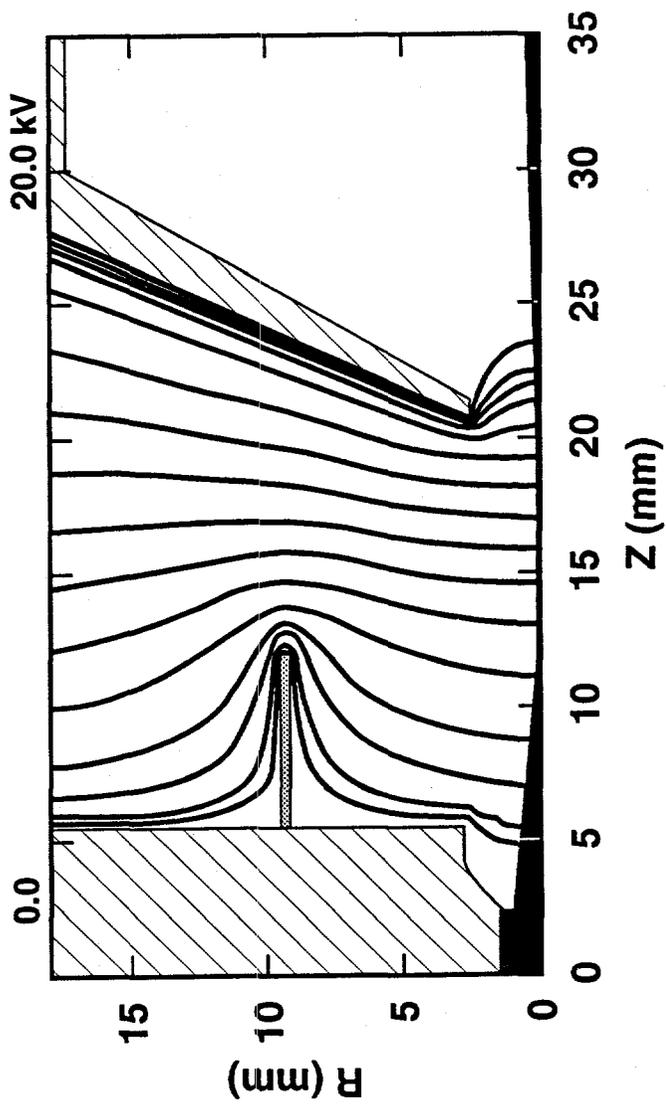


Fig. 3

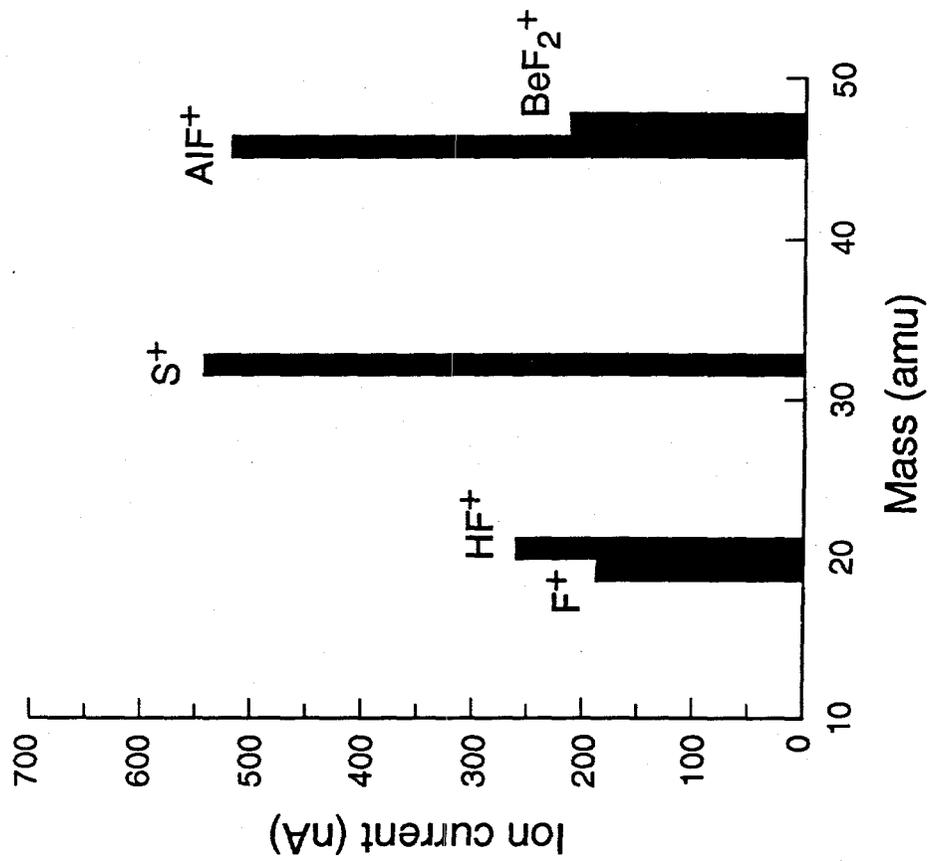


Fig. 4

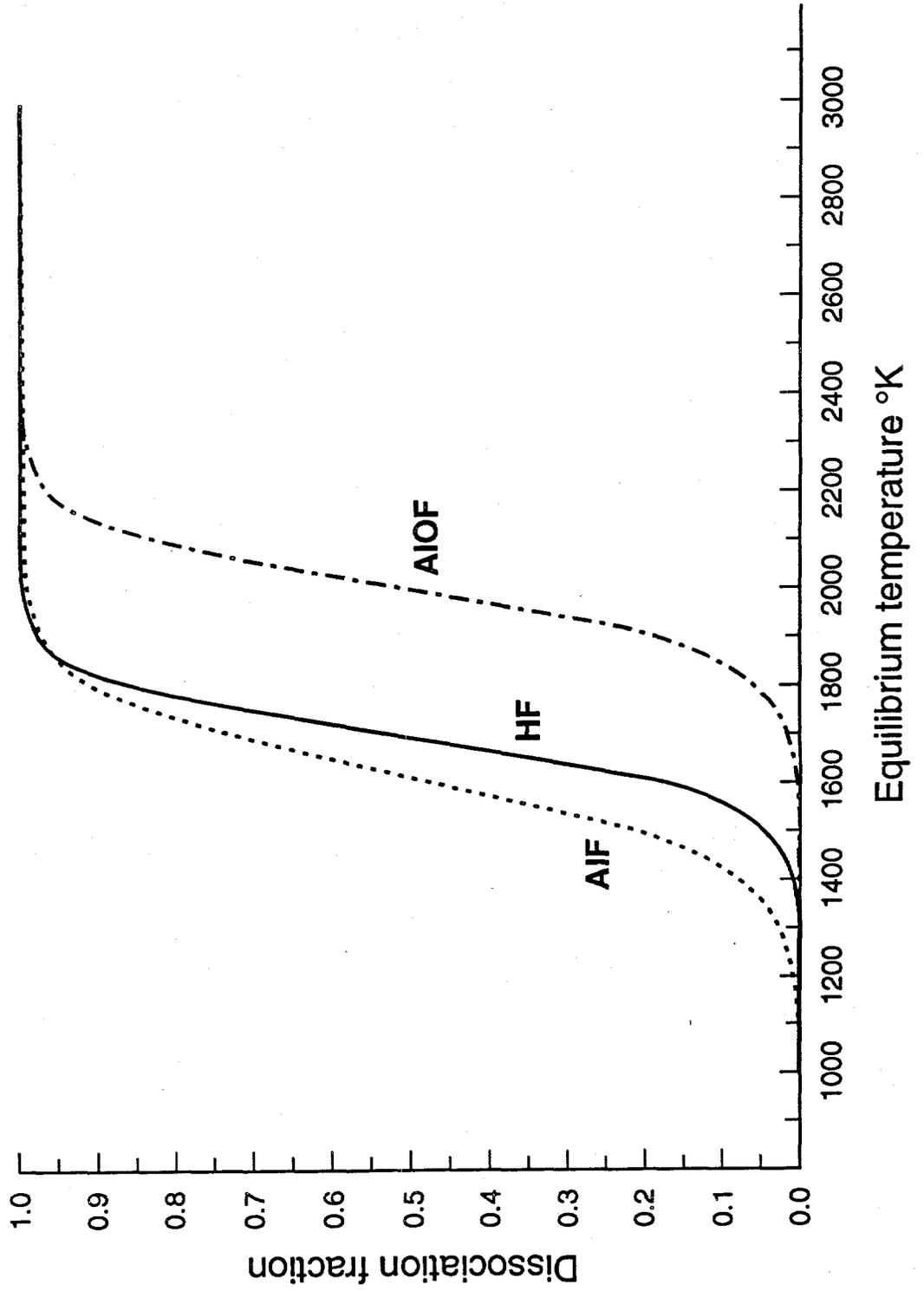


Fig. 5

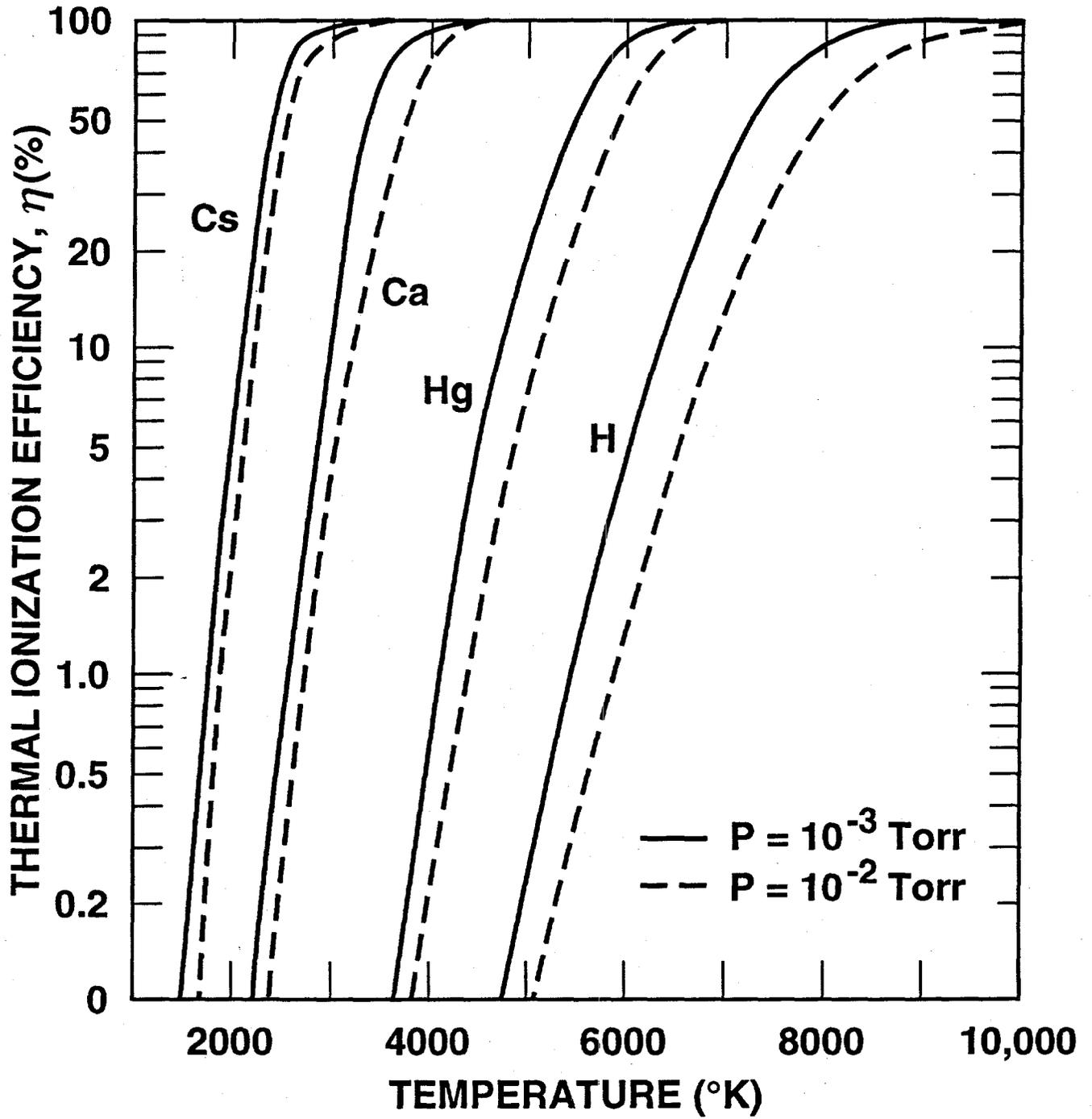


Fig. 6