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AN OCCLUDED GAS ION SOURCE

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ABSTRACT

An occluded gas pulsed ion source has been developed in which the hydrogen to be ionized has been previously absorbed by the arc electrode material. The principal advantages of the source are high output and economy in gas flow. The plasma is formed when a discharge is made to occur between alternate elements in a sandwich of insulators and hydrogenated titanium. The source has been adapted to both axial and radial extraction. Large beam currents, i.e. 1.5 amp per square inch of emitting area at 10 kv, are obtained in either case. The output has been magnetically analyzed and the ratio of atomic to molecular ions has been found to be high, but the major fraction of the total current consists of ions heavier than hydrogen. Reasons are given for believing that neutral hydrogen is released from the discharge points of the metal and is subsequently ionized. Steps in hydrogenating the metal are described.

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INTRODUCTION

This report covers work to date on development of a pulsed source of ions derived from hydrogen previously absorbed by an arc electrode material.[1] Several metals in the transition region of the periodic table have the ability to absorb exothermically, i.e., occlude, large quantities of hydrogen[2] (see Table I). This absorption is accompanied by large volume expansions, 10 to 15 percent, and by embrittlement. Titanium, at room temperature, has the largest known occlusive capacity. In fact, the saturation concentration of hydrogen in Ti is greater than in any other form in which hydrogen is known to exist. In addition, Ti metal is available commercially in many forms, and thus it was the logical choice for the occluding material.

DEVELOPMENT OF SOURCE GEOMETRY

One of the earliest attempts to obtain a pulsed release of gas involved discharging a low-impedance pulse line through a filament of hydrogenated titanium. With currents up to 2000 amp from the pulse line, no positive ion current was observed on a biased collector surrounding the filament. However, after only a few high-current pulses, a startling effect appeared: each pulse was accompanied by a bright blue flash in the vacuum system and positive ion current to the collector. Examination of the filament showed that the 0.002-in. Ti had ruptured and formed a joint of high resistance. The high pulse-line potential produced a spark discharge across this joint. Further experimentation showed that the collected ion current increased with the number of such joints and that the presence of a thin insulator in each gap lowered the spark threshold voltage and ensured a predictable electrical resistance. After considerable evolution the source appeared as shown in Fig. 1, a stack of alternately spaced Ti and mica washers, strung on an insulator for support and alignment. To measure the ion output, the source was surrounded with a collector biased with respect to the side of the source at fixed potential. Operating the source with high collector potentials would often initiate a spurious discharge so that the apparent collected current would become hundreds or thousands of amperes, depending only on the ability of the voltage-stabilizing capacitor to supply the current. Less ambiguous data obtained with a low-voltage biased collector are shown in Fig. 2. The circuitry consisted of a series combination of pulse line, termination, thyratron switch, and ion source.

Further experimentation showed that such an unconfined plasma is unsuited to the extraction of a concentrated beam of ions. Consequently, a rearrangement of the geometry was undertaken, which was to confine the discharge to the inside of the washer column. Limiting the volume resulted in a very dense plasma from which ions could be extracted at one end of the column. The initial mode of extraction, acceleration, and magnetic analysis of the beam was copied from procedures evolved in the PIG ion source

Hydrogen Solubility in cc STP per 100 g of metal.[2]

Temp°C		Ce	La	Nb	Pd	Ta	Th	Ti	V	Zr
20		------	------	10,400	------	------	12,500	40,300	15,000	24,000
150		------	------	------	------	------	-------	------	8,200	24,000
300		18,400	19,200	8,800	164	3,400	10,700	40,000	6,000	23,700
400		17,600	18,200	7,680	126	2,400	9,700	38,400	3,800	23,400
500		16,800	17,200	4,740	103	1,300	9,100	35,400	1,840	23,000
600		16,000	16,300	2,500	92.7	630	8,800	32,000	1,000	22,000
700		15,200	15,300	1,200	87.0	450	8,450	22,000	640	21,000
800		14,500	14,300	610	84.0	320	8,100	14,000	450	19,600
900		13,800	13,400	400	81.0	260	7,700	9,000	320	13,000
1000		13,000	12,300	------	78.5	230	2,600	6,500	240	------
1100		11,300	11,100	------	76.5	210	1,900	4,000	200	------
1200		5,300	4,100	------	------	------	1,750	------	------	------

Table I

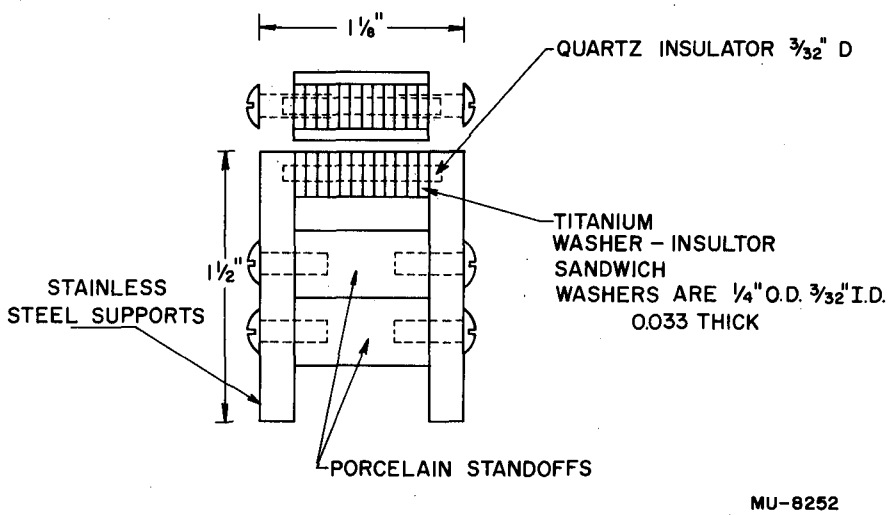

Fig. 1. Filament-type source.

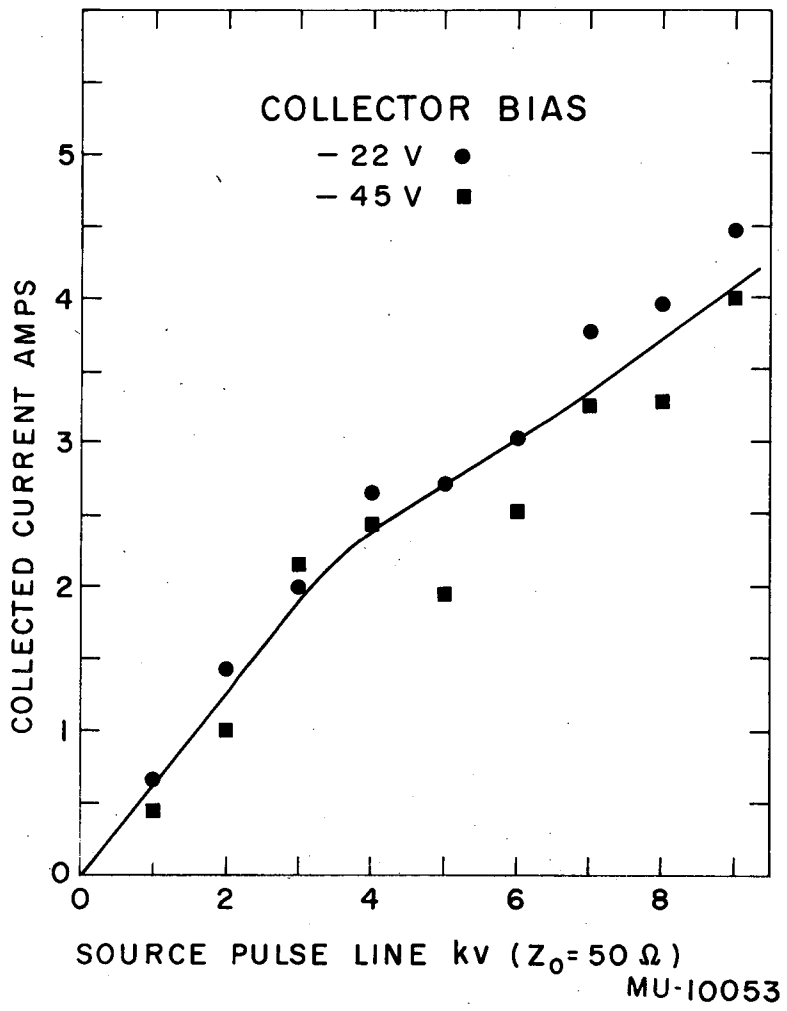

Fig. 2. Ion current versus charging voltage for filament-type source.

program.[3]

The new geometry consisted of a washer stack, minus the quartz insulator, clamped between two electrodes. One electrode was solid and the other contained a tapered hole, initially the diameter of the inside of the washers, but flaring out at 45^{o} to form a modified Pierce electrode. The extractor consisted of a 1-inch-diameter stainless steel cylinder tapered at one end to form the other Pierce electrode. The minimum opening in the extractor was about 0.010 inch larger than the inner diameter of the washers, and the usual spacing between the source exit aperture and the extractor was about 1/8 inch. An additional electrode was used to accelerate the ions to energies up to 60 kev to achieve constriction of the beam profile for magnetic analysis.

One problem that arose immediately was that of confining the discharge to the inside of the washer stack. Making the outer diameter of the insulators larger than that of the washers did not eliminate the tendency of the discharge frequently to prefer the outside of the stack. The additional feature that was found necessary was to trigger the discharge in the central region. To do this an insulated wire was inserted through the solid electrode so that the tip was concentric with the first washer. A 1-μsec high-voltage pulse was discharged through these elements. This produced ionization which both initiated the discharge of the pulse line and ensured the proper path. This trigger method made it possible to operate at lower pulse-line voltage than in the former scheme. The polarity of the source pulse line and the relative polarity of the source and the trigger pulses have been found to be of no consequence as far as performance is concerned. The complete source assembly is shown in Fig. 3.

As a circuit element, the source acts like a 60- to 80-volt hydrogen thyratron, and presents negligible impedance when in series with the pulse-line termination Z_o. For Z_o in the range 50 to 125 ohms, it has been found possible to deliver sufficient current through the source to provide space-charge-limited ion beams at reasonable pulse-line charging voltages, i.e. 3 to 10 kv. One method of providing the trigger pulse is to discharge a capacitor through a pulse transformer whose secondary is across the trigger gap. If the trigger pulse does not supply sufficient current, the pulse line is delayed -- i.e. it jitters -- or does not discharge at all. Satisfactory circuit components have been found to be a 0.5-μf capacitor and a 6:1 step-up transformer whose secondary insulation is adequate for the pulse-line voltage. In addition, it is desirable to finish the output end of the pulse line with an inductance instead of a capacitor in order to prevent any premature extinction of the discharge. If insulators are used whose dielectric strength is low enough so that appreciable leakage current is drawn by the source, a triggered ball gap is inserted in series with the pulse line.

In Figs. 4 and 5 are shown some data on beam currents for two methods of extraction. It will be seen that for the grid extractor, since a larger area of the plasma is effective, the beam currents are increased and show evidence of becoming emission-limited. In both these figures the currents shown should be multiplied by 0.5 to correct for secondary electrons that escape from the collector cup. This factor was determined subsequently with the use of a variable magnetic field in the cup.

The source is readily adapted for radial extraction as shown in Fig. 6.

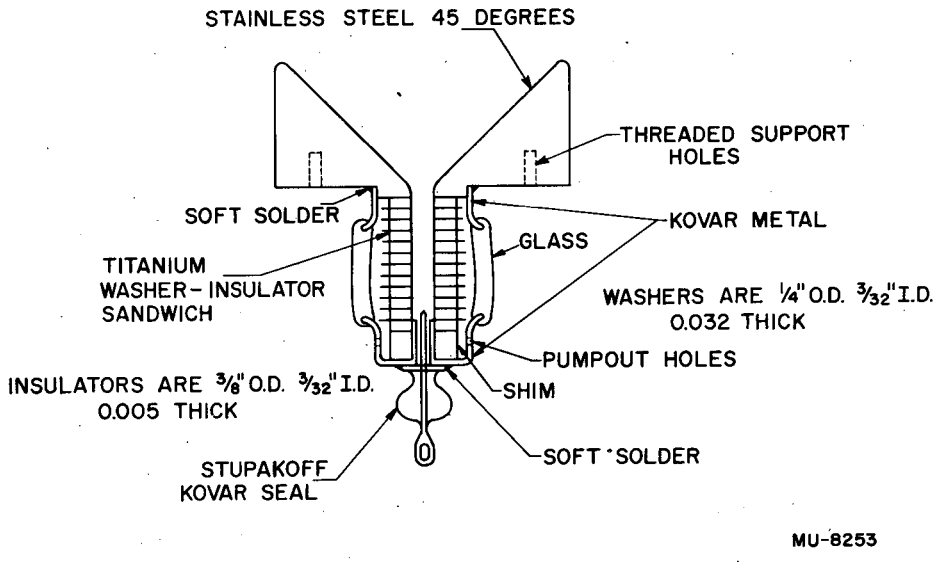

Fig. 3. Confined-volume source with exit geometry for axial extraction.

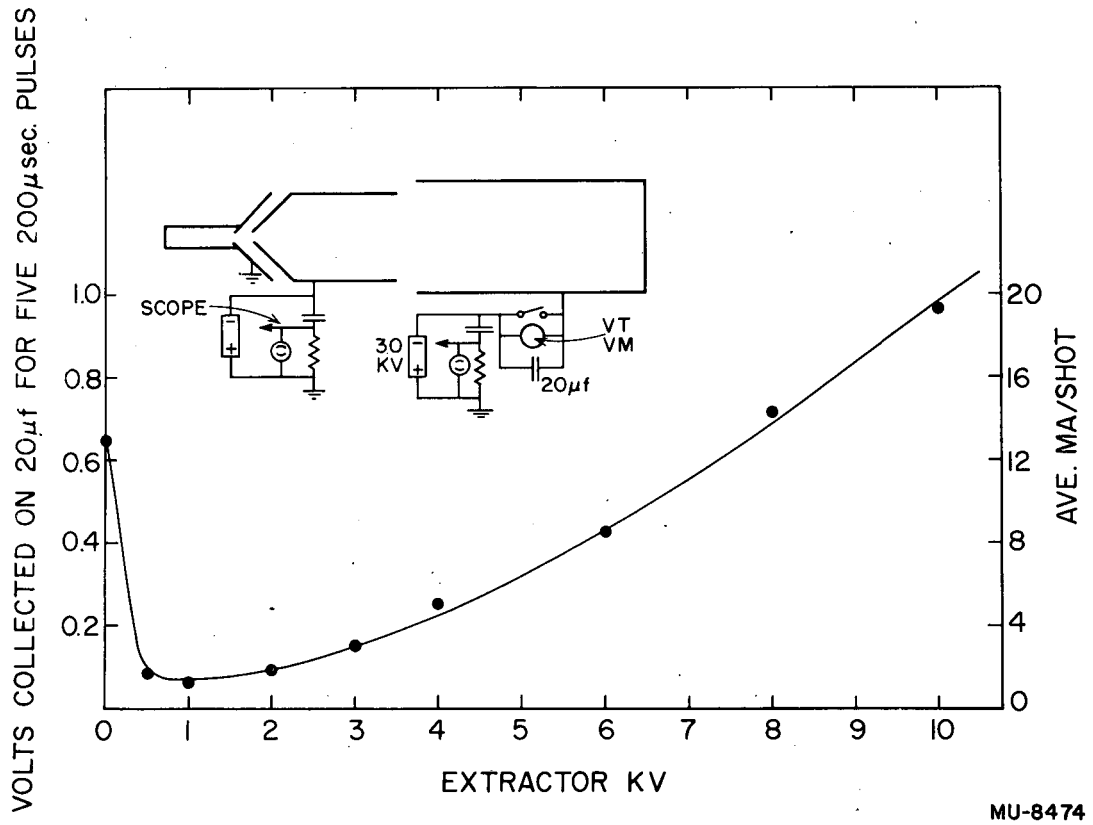

Fig. 4. Ion current versus extractor voltage for aperture-type axial extraction.

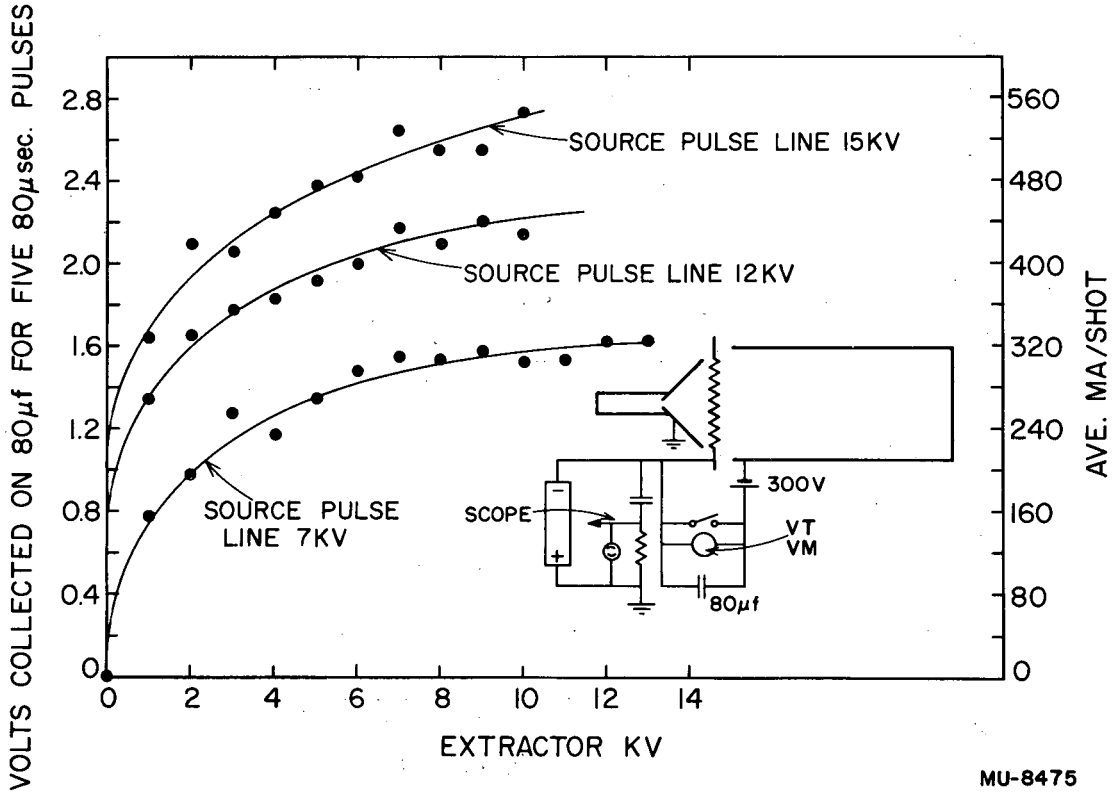

Fig. 5. Ion current versus extractor voltage for grid-type axial extraction.

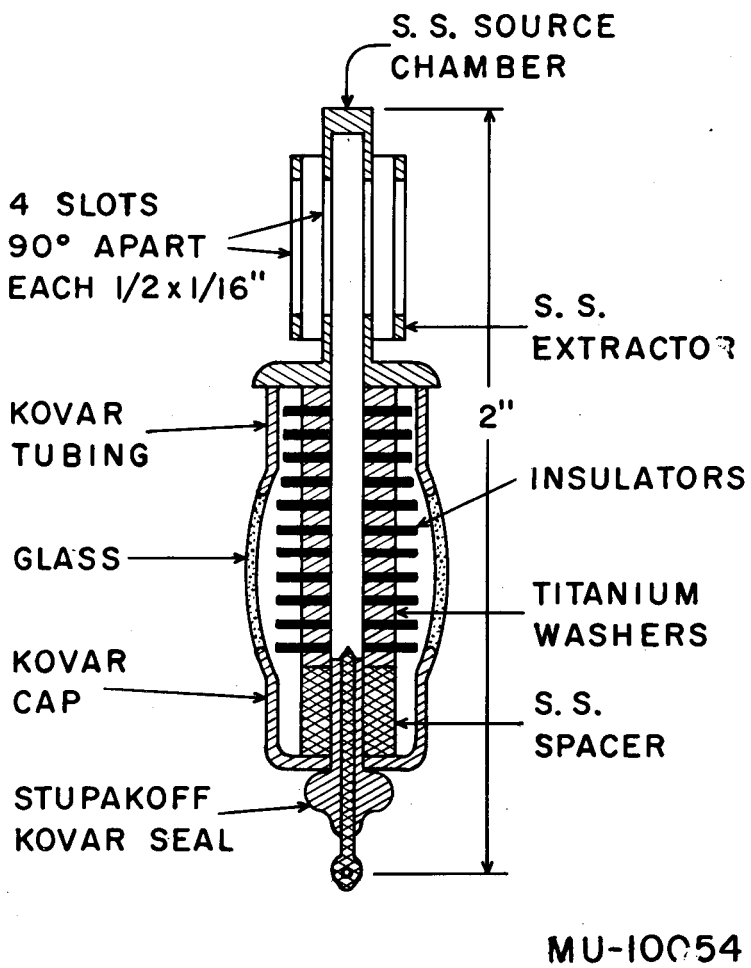

Fig. 6. Confined-volume source with exit geometry for radial extraction.

In contrast to the axial extraction geometry, where an axial magnetic field has no effect on beam output, the radially extracted beam is increased in the presence of such a field. Also, the beam is enhanced if the potential of the rear of the column, near the trigger, is negative with respect to the front. This is probably due to the repulsion of electrons into the extractor region, whereupon the potential from the separation of charge causes the ions to follow. In Fig. 7 are shown data for beam current collected through four slots, each 1/2 by 1/16 in. The extractor, which has four similar slots, was set at a spacing of 1/16 in. Since the data were taken in a field of 1300 gauss, the correction for secondary electrons is believed to be small.

MAGNETIC ANALYSIS

The magnetic analysis system and the associated electronics are shown in Fig. 8. Mass analyses of the ions from sources made with several different insulator materials are shown in Figs. 9, 10, 11, and 12. The curves have not been corrected for an instrumental effect which increases the widths of all peaks by an amount proportional to the magnetic field at which they are measured. In the most favorable case, muscovite mica, the fraction of all ions that are D^+ is 0.41. For a deuterium source a possible contribution to the H^+ peak may result from the absorption of hydrogen by the titanium owing to operation in an oil-pumped system. For an ordinary hydrogen source, the situation would be particularly ambiguous. The striking difference when Teflon insulators are used can be explained with the assumption that what was assumed to be Ti^+ is mostly CF_2^+ and Ti^{++} mostly CF_2^{++}, and that the discharge is principally supported on the volatile flourocarbons.

For analyses at different ion energies, a variation in the focusing properties of the system as a function of mass was noted. That is, at any one energy, the focus is optimum for only one mass. This effect was accentuated by the fact that the collimating system transmitted only a very small sample of the total beam. In the future, the analyses will be repeated with the source and detector in a large magnet so that the entire output is utilizing 180° focusing. Thus selective mass defocusing can be minimized.

DEGASSING AND LOADING

Achieving a high concentration of ordinary hydrogen or deuterium in titanium, i.e., approaching 400 cc at STP per gram of metal, involves no difficulty. Indeed, it is necessary to limit the concentration to less than this amount in order that the loaded metal does not become too brittle to permit subsequent assembly. When loading with deuterium it is desirable to use a mercury pumping system and to pay particular attention to adequate degassing, in order that the contamination from ordinary hydrogen be small. In addition, when using commercial grade hydrogen, the gas should be first dried in some fashion or passed through a palladium valve.

In degassing, the metal may be heated (in vacuum) in any one of several ways. Induction heating proves one of the most convenient. The degassing time depends upon the temperature and the dimensions of the specimen.[4] Because of this and the embrittlement effect, the titanium washers are fabricated before loading. The optimum temperature for degassing is 815°C.[4] For 1/4-inch Ti rod at this temperature, the percentage of initially absorbed

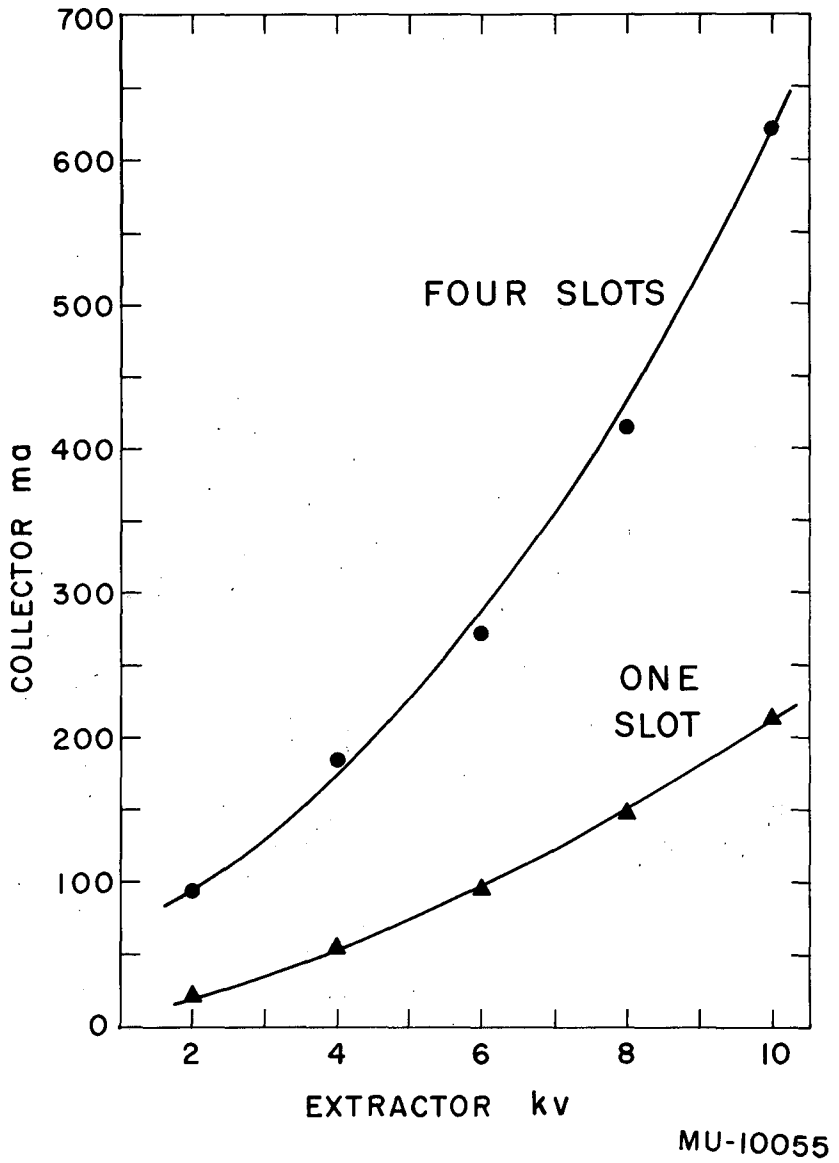

Fig. 7. Ion current versus extractor voltage for radial extraction.

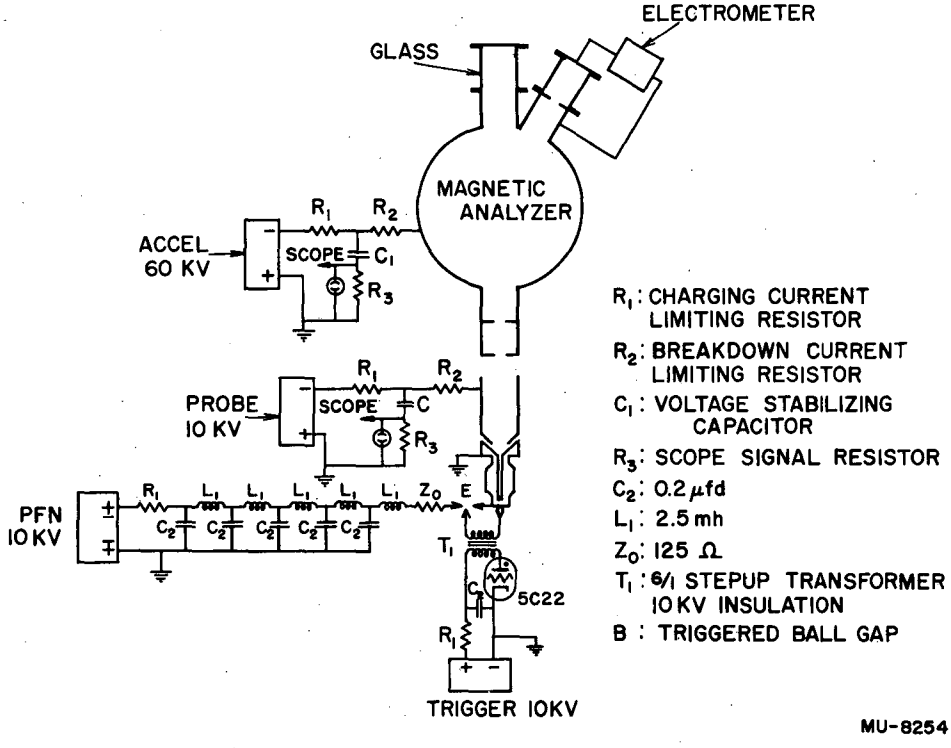

Fig. 8. Magnetic mass-analysis system and associated electronics.

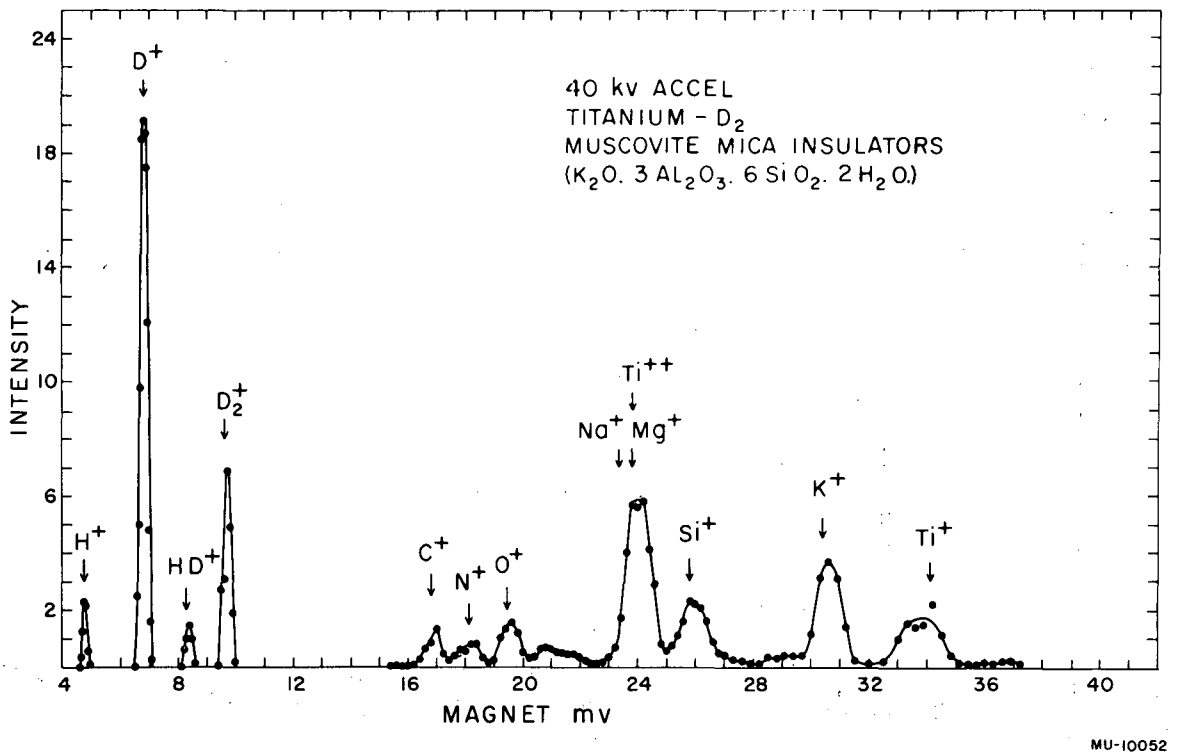

Fig. 9 Mass spectrum for Ti-D with mica insulators.

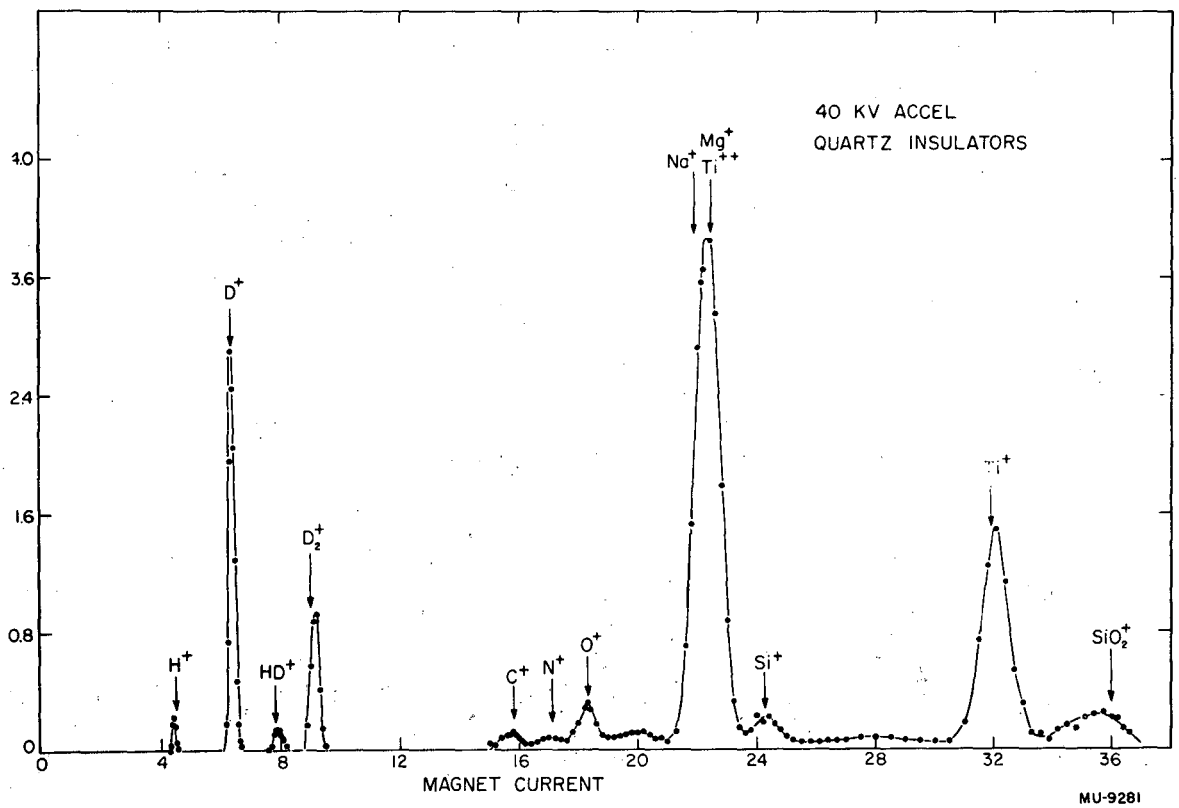

Fig. 10, Mass spectrum for Ti-D with quartz insulators.

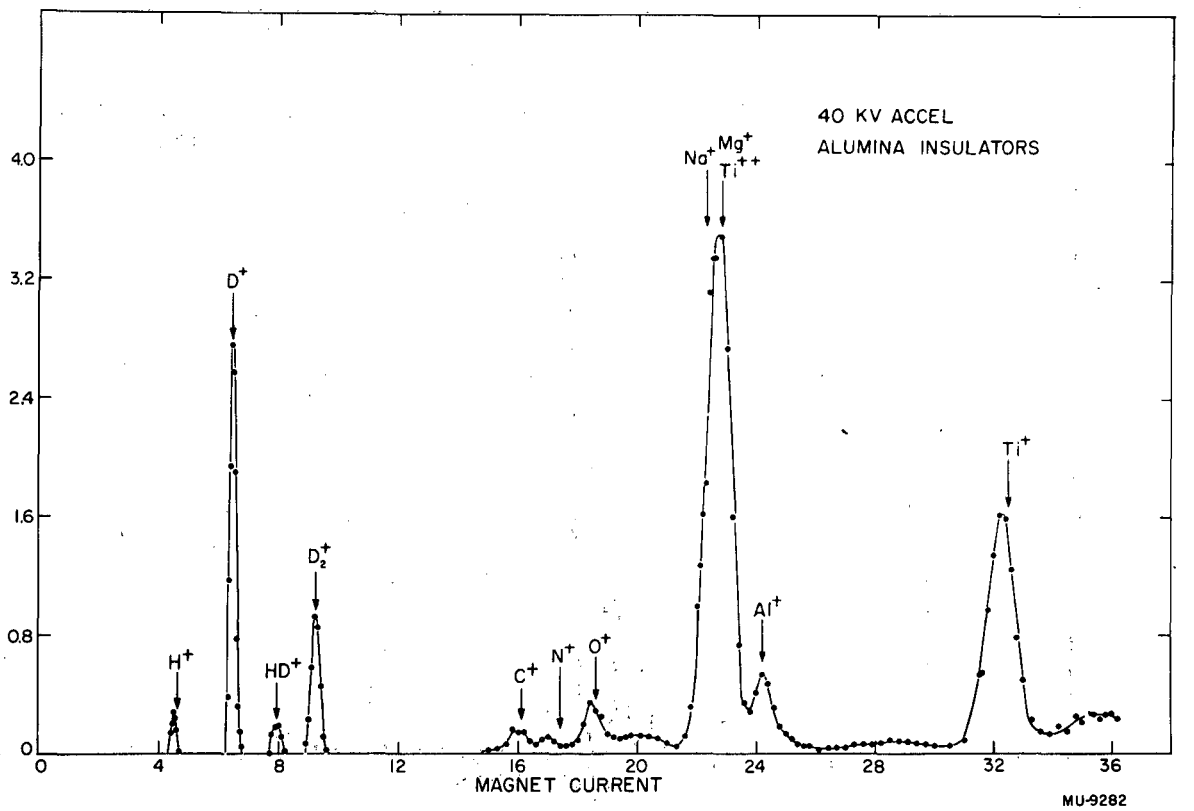

Fig. 11. Mass spectrum for Ti-D with alumina insulators.

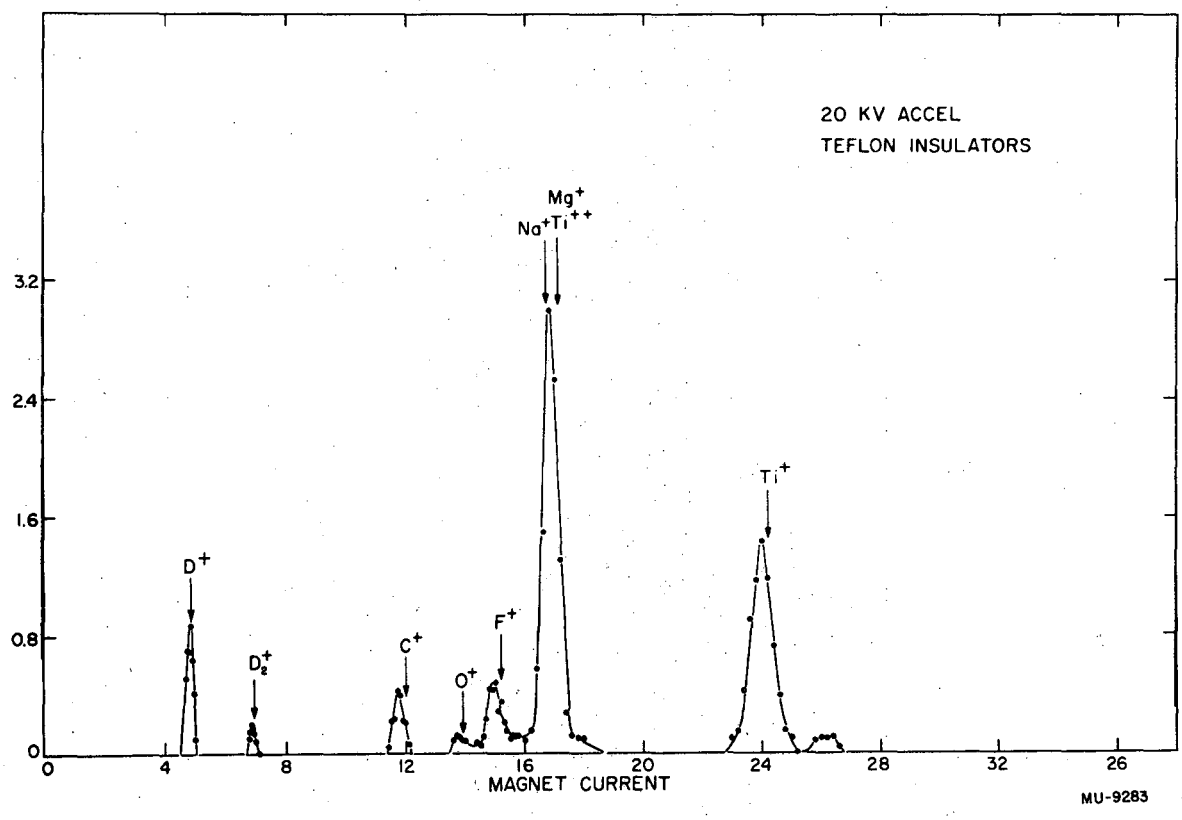

Fig. 12. Mass spectrum for Ti-D with teflon insulators.

hydrogen removed rises almost exponentially with time. Eighty-three percent has been removed after 2000 sec and ninety-seven percent after 6000 sec. The progress of degassing can be assayed by observing the change in pressure in the vacuum system as the titanium is alternately heated and cooled.

After degassing, the metal is allowed to cool and hydrogen is admitted to the system. The temperature is raised and at about 400°C a point is reached at which the occlusion process starts. As the reaction is exothermic, the temperature of the metal rises further as the reaction proceeds. The uptake can be accelerated by taking the temperature initially to about 1000°C, which opens the β phase. The metal is allowed to cool and the amount of hydrogen absorbed depends only on the pressure in the system and on the solubility limit. Any residual hydrogen in the vacuum system is then pumped away, the pressure is relieved, and the loaded specimen is removed.

MISCELLANEOUS

A study was made of the effect of scaling up the axial source. The diameters of all elements were approximately doubled. The resulting collected currents under comparable operating conditions roughly doubled. The dependence upon diameter rather than area is to be expected from the fact that only the periphery of the extractor aperture is effective. Inside the aperture the electric field is weakened by the geometrical effect as well as by space charge. To counteract this, the extractor voltage could be increased when larger apertures are desired. For a given geometry a minimum voltage is required to contain the plasma and prevent penetration of the aperture by electrons and low-energy ions.

For the work reported herein the rate of pulsing of the sources has been about 1/2 pps. For application to linear accelerators, a higher repetition rate is necessary. A period of operation was arranged at 7.5 pps consisting of over 10^5 shots of 8 μsec each. The beam level was observed to decrease in just a few shots to 2/3 of the low repetition rate level and to remain there for the duration of the run.

Other metals with d-shell deficiencies similar to Ti are known to occlude hydrogen to a large extent (see Table I). Excluding the radioactive elements, zirconium and hafnium should be the next strongest occluders. A source was made with Zr and gave the same total space-charge-limited current as those with Ti. Magnetic analysis of this source is shown in Fig. 13. The Zr^+ peak is not shown, but it is smaller than the Zr^{++} peak. The presence here of Zr^{++} indicates that the ambiguous peak in the Ti spectra is Ti^{++}.

MECHANISM OF PLASMA PRODUCTION

Underlying the practical aspects in the production of accelerated ion beams is the fundamental question of how the plasma is generated. The evidence is quite conclusive[5] that hydrogen exists in those metals which are exothermic occluders not only in lattice rifts, as for other hydrogen-absorbing substances, but also distributed regularly within the lattice structure. This hydrogen is contained in the atomic form, at least partially ionized, and, in fact, probably in the metallic state.[6] This last conclusion is suggested by

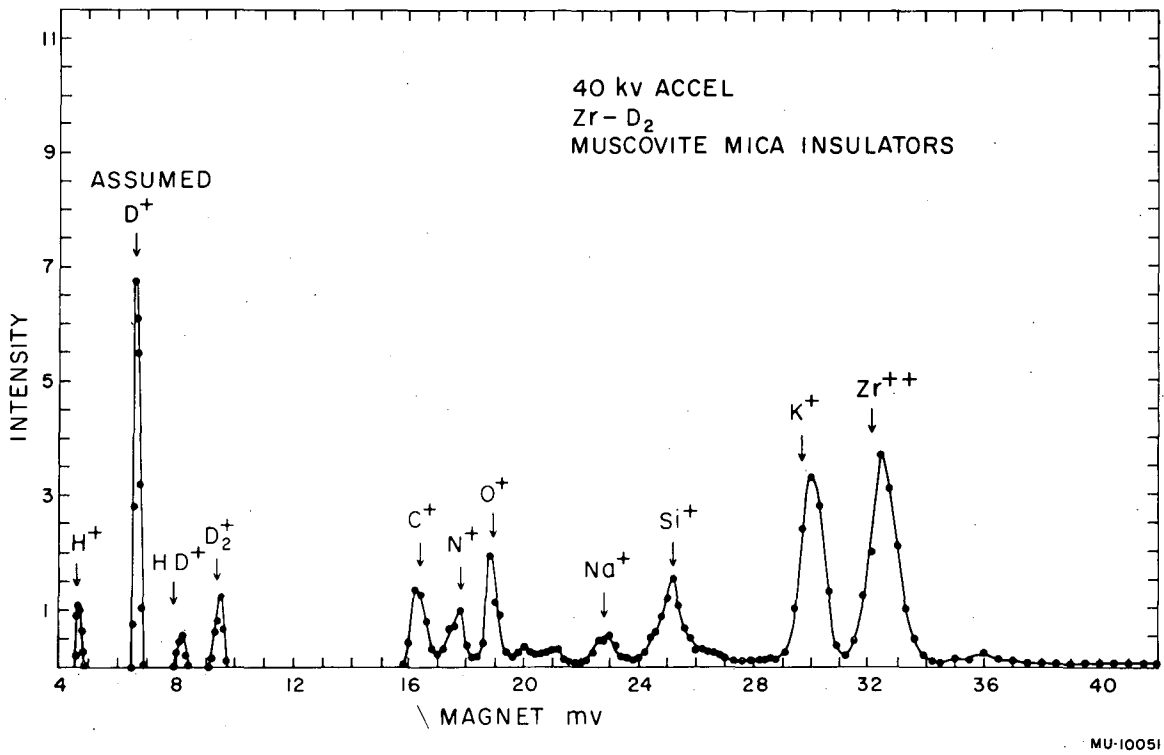

Fig. 13. Mass spectrum for Zr-D with mica insulators.

the fact that although the hydrogen atom is small enough to require no expansion of the Pd lattice to contain it, the observed expansion is 11 percent. It has been calculated that metallic hydrogen would be stable relative to covalent hydrogen at pressures not less than 2.5×10^5 atmospheres, and such an internal pressure in Pd would produce a volume increase of about 10 percent.

A controversy exists in the literature as to whether ions can be obtained from palladium, the most thoroughly studied exothermic occluder, solely by heating of the metal. The situation is reviewed by Stansfield.[7] He concludes, in accord with our own experience, that there is no spontaneous emission of protons from hydrogenated palladium. However, in a recent reinvestigation of the question at Syracuse University,[8] a proton current of 10^{-10} amp was observed[9] from heated Pd, and with a sufficiently low collector voltage -- 3 to 5 volts -- to ensure the absence of spurious discharges. The current thus obtained is sensitive to the degree of hydrogenation of the Pd. However, this current is far lower than those at issue; e.g., Franzini[10] observed over 0.3 ma. It is possible that the latter result and others[9] similar to it can be explained on the basis that unobserved low-current discharges produced the ionization found in the pressure of heated palladium. On the theoretical side, it has been calculated,[11] on the basis of energy conservation, that the binding energy of H^+ in Pd is 10.7 electron volts. This would preclude any thermionic emission of protons.

The evidence indicates that neutral hydrogen is released in the form of either H, H_2 or Ti-H, and subsequently ionized in the discharge. A characteristic of low-voltage ion sources is the presence of the ion H_3^+. While it is not clear what reactions are responsible for the high percentages of H^+ observed is such sources,[12] the reaction leading to H_3^+ is believed to be $H_2^+ + H_2 \rightarrow H_3^+ + H_1 + 1.7$ ev. The formation of H_3^+ thus depends on the presence of molecular hydrogen. In a careful search for the mass-three peak with a source loaded with ordinary hydrogen, the ratio $H^+ : H_2^+ : H_3^+ : D_2^+ = 4.0 : 0.6 : 0.015 : 0.0003$ was obtained. Had the $H_3^+ : H_2^+$ ratio been of order unity, as in many other low-voltage sources, this would have indicated the presence of considerable molecular hydrogen. The disparity in the observed ratio, however, does not indicate necessarily that atomic hydrogen is the primary material, as similar ratios have been obtained in well-collimated high-current discharges in molecular hydrogen.[13]

CONCLUSION

Perhaps the foremost advantage of the occluded-gas ion source, aside from high output, is economy in gas flow. This is a consequence of the high ionization efficiency and the absence of any gas emission between pulses. The compact size and the elimination (with axial extraction) of the need for an auxiliary magnet, are of considerable benefit when space is a problem. Also, the ratio of atomic to molecular ions compares favorably with sources of other types. It is unfortunate that the major fraction of the large beam currents available consist of ions heavier than hydrogen, but the problem of separating them out does not appear very formidable.

A program is now under way, designed to increase the output of the source and produce a suitably focused and accelerated beam of ions for application in accelerators having axial symmetry. Experimenters who plan to make use of this type of source in its present state of development should be prepared necessarily to undertake some research.

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