Proton Production in the Low Voltage Arc

E. S. LAMAR AND OVERTON LUHR, *Eastman Laboratories, Massachusetts Institute of Technology*

(Received May 15, 1934)

Further investigations with the proton source previously described have yielded considerable information about the processes involved in its operation. The tube has been redesigned with the object of obtaining large proton currents in a beam. Ions are drawn out of the arc through a hemispherical grid inserted in the cylindrical electrode. Part of the resulting converging beam passes through a hole in a second electrode which separates the arc chamber from a region maintained at a lower pressure by differential pumping. Total currents of over 0.5 milliampere at current densities of about 8.0 milliamperes per square cm were obtained on the low pressure side. The ratio of proton current to total current was as high as 98 percent with pressures in the discharge tube of 0.3 to 0.4 mm. In order to attain this percentage it was necessary to maintain certain minimum potentials on the cylinder and second electrode. At the same time the metal parts must be prevented from exceeding red heat since an adsorbed layer of hydrogen on the walls is necessary to prevent recombination of atomic hydrogen there. The proton currents obtainable are limited by the speed of the differential pumping and the degree of vacuum desired. The observed pressure ratios were improved by a factor of two when the arc was in operation owing to the increased temperature of the gas streaming through the hole.

INTRODUCTION

THE present widespread interest in the atomic nucleus has made desirable the development of a convenient proton source as one of the necessary tools for nuclear investigations. Large ion currents can be obtained from various types of electrical discharges in hydrogen. Mass-spectrograph studies show,[1, 2] however, that under most circumstances only about ten percent of the ions are protons. The remainder are either diatomic or triatomic ions depending upon the number of collisions with neutral molecules made by the primary diatomic ions before entering the mass-spectrograph. A very effective proton source was reported recently by Oliphant and Rutherford.[3] However, since the discharge tube described by them is large and requires high voltage and considerable power for its operation, it seemed desirable to see what could be done with other types of electrical discharges. As a most efficient source of ions, a low voltage arc of the type described by Langmuir and Jones[4] was employed. A preliminary report of this work has been published.[5]

[1] W. F. Bleakney, Phys. Rev. **35**, 1180 (1930).
[2] O. Luhr, Phys. Rev. **44**, 459 (1933).
[3] Oliphant and Rutherford, Proc. Roy. Soc. **A141**, 259 (1933).
[4] Langmuir and Jones, Science **52**, 380 (1924); Phys. Rev. **31**, 357 (1928).
[5] Lamar and Luhr, Phys. Rev. **44**, 947 (1933).

IDEALLY SIMPLE THEORY

Consider a hot filament cathode and a circular plate anode between which a low voltage arc is maintained in hydrogen gas. One might expect between five and ten percent of the ions in the arc, formed by direct impact of electrons with hydrogen molecules, to be protons and the remainder to be molecular ions. Imagine now this region of ionized gas enclosed by a third cylindrical electrode which is maintained negative with respect to the anode. Positive ions which drift into the space charge sheath enclosed by this electrode will be accelerated and will strike the cylinder with kinetic energy gained in the space charge sheath. Since the accommodation coefficient for hydrogen ions on the electrode is small, they will rebound as neutrals retaining a large fraction of the kinetic energy gained in the sheath. Because of collisions, these neutralized positive ions quickly reach equilibrium with the gas enclosed by the cylinder and cause an increase in the temperature of the gas above that of its surroundings. A thermal reaction should then take place in which molecular hydrogen dissociates with a reaction rate which depends upon the temperature of the gas. If this rate of production of atomic hydrogen greatly exceeds the rate of disappearance by diffusion to the walls, the enclosed gas will be largely atomic hydrogen and thus the ions produced by direct electron impact largely protons.

Apparatus and Experimental Procedure

A diagram of the discharge tube is shown in Fig. 1. The filament was a composite made by wrapping twelve mil tungsten with five mil nickel. The resulting (piano wire) was wound in a helix on a one-sixteenth inch mandrel. The filament was coated with a mixture of barium and strontium oxides, the nickel serving to increase the emitting area, and acting as a binder for the coating. The filament together with the flanged plate anode shown in the diagram comprised the two electrodes between which an arc was maintained in hydrogen. The

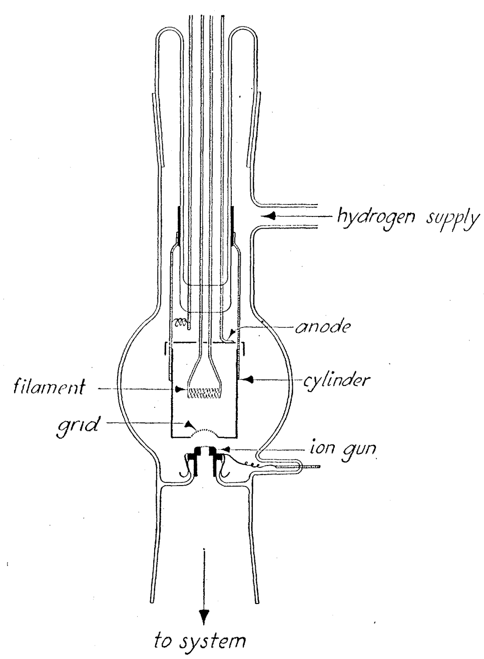

Fig. 1. Experimental tube.

resulting region of partly ionized gas was completely enclosed by a nickel cylinder 4.0 cm long and 3.0 cm in diameter. This cylindrical electrode was always maintained sufficiently negative with respect to the anode to prevent primary electrons from the filament from reaching it. At its lower end, the cylinder was fitted with a hemispherical grid about one cm in diameter, made of forty mesh nickel gauze. Of the ions which drifted into the space charge sheath covering the grid, about half were collected by the grid and the other half drawn from the arc. The grid tended to focus the ions drawn from the arc onto the surface of a fourth electrode or ion gun shown in the diagram. In an earlier tube the ion gun was placed directly in an opening in the cylinder, omitting the hemispherical grid, but this type of construction never yielded such high percentages of protons. The results indicate the desirability of reducing to a minimum all openings in the metal walls enclosing the discharge. The available evidence seems to show that the loss of atomic hydrogen from this region is largely by diffusion through such openings.

The ion gun was made of aluminum and the shape was as shown in the diagram. Several different ion guns having different sized openings in their upper faces were used in the course of the experiments. The ion gun fitted loosely in a glass tube whose upper end had been ground flat. A molybdenum spring held the ion gun against this flat grind, thus sealing the arc chamber from the region of high vacuum. Since the ion gun fitted loosely in the glass tube, any expansion of the metal due to heating introduced no mechanical strains in the glass.

A conical grind at the lower end of the tube made possible its attachment either to the mass-spectrograph or to any desired system of pumps and collecting electrodes. The upper end of the tube was also fitted with a conical grind to facilitate repairs or alterations of the metal parts.

When the tube was fastened to the mass-spectrograph the ion gun had in its upper face a hole 0.3 mm in diameter, countersunk from the back to reduce its thickness to a negligible value. Since the pumping system for the mass-spectrograph was not designed to take care of large quantities of gas, a larger hole in the ion gun would not permit the necessary low pressures to be maintained in the mass-spectrograph.

The filament was heated by alternating current, twenty-five watts being more than sufficient to maintain an arc current of one ampere measured at the cathode. The relative numbers of the different types of ions drawn from the arc were studied by means of the mass-spectrograph as functions of the negative voltages on cylinder and ion gun, pressure in the arc chamber, and previous history of the arc.

When the arc was first started, most of the ions measured were those of water vapor, CO and CO_2. After operating the arc for about an hour at low pressure and high arc drop so that the metal parts became bright red, these impurities were cleaned up.

After studying the nature of the ions with the mass-spectrograph, experiments were undertaken to determine the magnitude of the ion current which could be obtained from the arc with any degree of reliability. For these tests the tube was taken off the mass-spectrograph and put onto a system having a net pumping speed for air of about ten liters per second. A collecting electrode was placed several centimeters behind the ion gun. Several different types of ion guns were tried but the most satisfactory design was that shown in the figure. The hole in the face of the ion gun was 4.0 mm in diameter and 4.0 mm long. The hole was covered by a forty mesh grid to prevent distortion of the electric field at the face and thus to avoid undue spreading of the emerging ion beam. It was found advisable to round off the edges of the ion gun to prevent the concentration of electric field and thus the concentration of ions onto the edges of the gun.

Discussion of Results

The range of pressures in the discharge tube over which satisfactory proton yields were obtained was from 0.20 to 0.60 mm. In this range, the arc drop varied from about 25 to 50 volts. It was found possible to increase the arc drop by decreasing the heating current to the filament without undue damage to the filament. The higher arc drop gave greater ion currents to the cylinder. Results, in general, were found therefore to be more satisfactory with an arc drop between 40 and 50 volts than at lower voltages. All of the tests here reported were with an arc current of one ampere measured at the cathode. The current to the cylinder ranged from 0.23 to 0.32 ampere.

Fig. 2 shows two typical mass-spectrograph curves. The lower curve was taken under fairly ideal conditions and shows about 98 percent protons and the rest triatomic ions. The upper curve was taken under conditions not quite ideal in order to show the presence of diatomic ions. The peaks do not occur at the exact positions to be expected from the applied potentials because of collisions made by the ions before leaving the discharge chamber. The shift in the position of the maximum is greatest for the protons, and least for the triatomic ions.

For convenience the ordinates for the curves of Fig. 3 were computed from the heights of the peaks in curves of the type of Fig. 2 rather than from the areas under the curves since it was found that the error thus introduced was less than the error inherent in the experiments. Curve number 1 of Fig. 3 is a plot of the fraction of the ion current carried by protons as measured by the mass-spectrograph as a function of the negative potential of the cylinder with respect to the anode. The arc current was 1.0 ampere and the potential of the ion gun with respect to the anode was 300 volts. As can be seen from the curve, the proton percentage rose rapidly with increase in voltage of the cylinder, reaching a maximum at about 60 volts. Further increase in voltage resulted in a decrease in the percentage of protons. As a possible explanation for this decrease it was assumed that under the best conditions the recombination of atomic hydrogen on the cylinder was hindered by the presence of an adsorbed layer of hydrogen on the surface. Since an increase in voltage of the cylinder means an increase in power input, and thus in temperature of the cylinder, a temperature should be reached at which the adsorbed layer can no longer exist. Thus the rate of disappearance of atomic hydrogen to the walls should increase and the proton percentage should decrease if the cylinder voltage is raised above an optimum point.

In order to test the above hypothesis, the tube was water-cooled by wrapping with three-sixteenths inch rubber tubing. Unfortunately, a test on the exact same tube which yielded curve number 1 of Fig. 3 was not possible since a minor accident made it necessary to replace the filament before any test could be made after the discharge tube had been wrapped for cooling. When re-assembled, the filament was closer to the grid than previously and, further, it was learned later that the grid and ion gun were

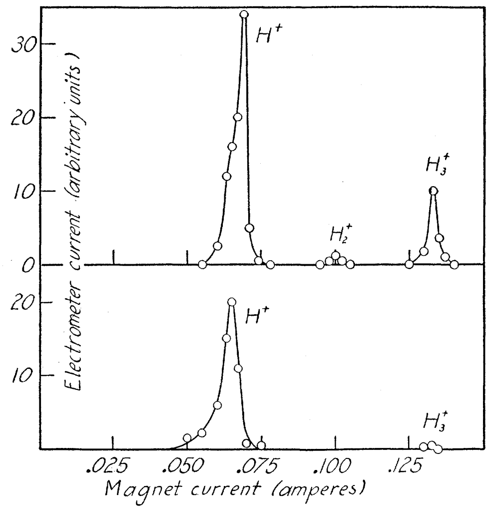

FIG. 2.

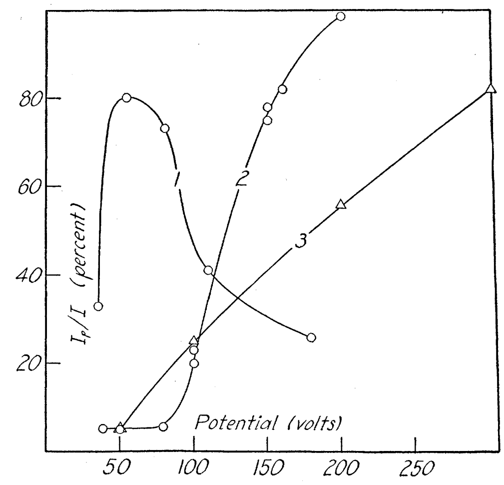

FIG. 3.

FIG. 2. Typical mass-spectrograph curves. (Tube water-cooled.) Upper curve: pressure = 0.230 mm; cylinder potential = 150 volts; ion gun potential = 300 volts; H^+ = 75 percent of total current. Lower curve: pressure = 0.380 mm; cylinder potential = 200 volts; ion gun potential = 300 volts; H^+ = 98 percent of total current.

FIG. 3. Ordinates give fraction in percent of measured current carried by protons. Abscissae, curves 1 and 2, negative voltage of cylinder with respect to anode. Abscissa, curve 3, ion gun voltage.

not in as perfect alignment as before. Curve number 2, of Fig. 3, is a plot of the proton percentage as a function of cylinder voltage for the water-cooled tube. It is to be noticed that although this curve starts up at a higher voltage than does curve number 1, possibly for the reason stated above, it continues on up to about 98 percent.

Curve number 3, Fig. 3, shows the proton percentage as a function of voltage on the ion gun for a cylinder potential of 162 volts for the water-cooled tube. For the tube whose grid was in good alignment with the ion gun, no change in proton percentage was observed when the potential of the ion gun was changed from 300 to 100 volts. Below 100 volts the proton percentage decreased with decreasing potential on the ion gun.

A possible explanation for the dependence of proton percentage on ion gun voltage is as follows: The rate of disappearance of atomic hydrogen from the inside of the cylinder by diffusion through the grid depends upon the atomic hydrogen concentration gradient at the grid. Collisions with gas molecules made by ions travelling toward the ion gun and by neutralized ions returning from the ion gun should be effective in producing atomic hydrogen in the region between grid and ion gun. As a result, the gradient at the grid, and thus the rate of loss of atomic hydrogen from inside the cylinder should be reduced. As soon as the voltage of the ion gun is so great that the diffusion of atomic hydrogen through the grid is small in comparison with the diffusion through other openings in the tube, the ion gun is no longer the controlling factor; thus further increase in its voltage does not increase the proton percentage.

The proton percentages, with the water-cooled tube seemed to be independent of pressure within the errors of measurement over the range of pressures in the arc chamber, from 0.23 to 0.4 mm. Above 0.4 mm, for the same voltages, the proton percentage was definitely higher than for the range given above, and for pressures below 0.23 mm, the proton percentage decreased rapidly until at 2×10^{-2} mm only about five percent protons were observed.

Two possible contributing causes for the decrease in proton percentage with decreasing pressure are as follows. When the mean free path for the neutralized positive ions rebounding from the cylinder becomes comparable with the dimensions of the cylinder, the neutralized positives lose their energy in collisions with the cylinder rather than in collisions with the gas. The gas is therefore not raised to a temperature high enough to cause thermal dissociation. Also the cooling of the cylinder may not have been sufficient to keep its temperature at a value sufficiently low to permit the existence of an adsorbed layer of hydrogen at the lower pressures. Thus the rate of recombination of atomic hydrogen on the walls may have been high at low pressures.

Although they have not been tried, cooling fins of carbonized nickel spot-welded to the outside of the cylinder are suggested to replace the less convenient water-cooling employed in these experiments.

The maximum ion current obtainable from the arc depends upon the current density at the face of the ion gun, the pressure in the arc chamber, the speed of differential pumping, and the degree of vacuum desired on the high vacuum side of the ion gun. In these experiments the current density at the surface of the ion gun was about 8 milliamperes per square cm. The ion gun used in these experiments had in its surface an opening 4 mm in diameter and was covered by a mesh grid which reduced the open area by a factor of two. The total current through the ion gun was therefore about 0.5 milliampere.

Earlier experiments with holes of the order of 1 mm in diameter in the ion gun indicated considerable distortion of the electric field at the surface of the ion gun and thus wide spreading of the ion beam after leaving the arc chamber. With the introduction of the grid, however, it was possible to realize saturation current with only 200 volts between the ion gun and the collector on the high vacuum side. In other words, the ions were projected through the grid straight enough to prevent measurable loss to the walls in travelling the 4 mm length of the hole in the ion gun.

The pumping system employed in the second set of experiments had a net pumping speed for air of about 10 liters per second. When the arc was not in operation and hydrogen was being pumped through the 4 mm opening in the ion gun, the pressure ratio maintained between the arc chamber and the high vacuum side was about 20. This ratio increased by a factor of two when the arc was in normal operation. This increase in pressure ratio probably resulted in part from the fact that the gas diffusing through the opening in the ion gun was partly atomic, and in part from the decrease in gas density in the arc chamber resulting from the increased temperature of the gas.

As can be seen from the diagram, the entire apparatus is small, the voltages required can be obtained from storage batteries, and since the power requirements are small, these batteries need not be heavy duty, and thus need not require much space. It was hoped that the apparatus would yield high proton percentages at much lower pressures so that the problem of differential pumping for any high voltage installation would be less difficult. If the mechanism outlined above for the dissociation of hydrogen is the correct one, however, the apparatus should yield protons only at pressures such that the mean free path, for the high speed neutralized positive ions returning from the cylinder, is small in comparison with the dimensions of the apparatus. If one increases the dimensions, one must also increase the power input to maintain the same ion concentration. The present apparatus may of course be limited by recombination of atomic hydrogen on clean walls, and if so, more adequate cooling would insure the existence of an adsorbed hydrogen layer at lower pressures, and thus less recombination. Granted these two limitations, the other alternative is to increase the current density at the surface of the ion gun so as to reduce the size of the opening required for any desired ion current. Eight milliamperes per square centimeter were realized in these experiments and it may be hoped that by careful alignment of ion gun and grid, this current density might be somewhat increased.

The authors wish to express their indebtedness to Dr. K. T. Compton for his continued interest, advice and many valuable suggestions during the progress of these experiments. They wish also to thank various members of the staffs in the departments of Physics and Chemistry for many helpful discussions.