IMPROVEMENT OF HYDROGEN PUMPING CHARACTERISTICS OF PENNING DISCHARGE GETTER-ION PUMPS*

C. M. Turner, J. G. Marinuzzi, and G. W. Bennett
Brookhaven National Laboratory
Upton, New York

Abstract

Pumping hydrogen with Penning discharge getter-ion pumps at the flow rates normally encountered with accelerators employing hydrogen ion sources presents problems that do not appear when these pumps are employed on systems where the only gas load is that arising from surface degassing and residual leakage. These problems are,-1) thermal instability resulting from reemission of absorbed hydrogen, and 2) deformation of the titanium cathodes with hydrogen absorption. This report outlines the pertinent considerations involved in modification of the design of the pumping elements of a standard 500 l/s pump (air rating), 1) to increase by a factor of about 10 the hydrogen gas load the pump can handle without danger of thermal instability, and 2) to virtually eliminate titanium deformation. Hydrogen gas loads of up to 5 atm cm^3/min can now be accomodated with a 500 l/s pump. While service life expectancy was previously limited to 2000-3000 hours by anode-cathode short circuit, it is presently limited by the ultimate hydrogen absorption capacity of the titanium. It is anticipated that life expectancies of up to 25000 hours can be realized with the 500 l/s pump at a hydrogen flow rate of 1 atm cm^3/min.

Background

The work summarized herewith was motivated by the desire to provide a fail safe vacuum system for the Van de Graaff injector for the BNL Cosmotron to eliminate hazards of oil vapor contamination of the acceleration tube. The Penning discharge getter-ion pump[1,2] with no moving parts or filaments appeared to be an ideal candidate for this application, and the 500 l/s pump was recommended by the manufacturer as being capable of meeting our requirements. However experience soon showed that this pump was incapable of reliable performance with hydrogen gas loads much greater than 0.5 atm cm^3/min. A simple solution would have been that of employing a substantially higher speed pump or several 500 l/s pumps in parallel. However space limitations made it highly desirable to meet the requirements with a small pump, and the unknown factors responsible for the erratic performance presented a challenge which led to the ensuing investigation which began in 1960 and has continued intermittently to the present.

For reference, Fig. 1 shows a complete 500 l/s pump assembly. Fig. 2 shows an assembled and Fig. 3 a disassembled pumping element.

Mechanism of Hydrogen Pumping

The mechanism of hydrogen pumping by a Penning discharge getter-ion pump employing titanium cathodes is that of diffusion of the hydrogen into the titanium to form a titanium-hydrogen association in the proportions represented by the formula $TiH_{1.76}$[3]. This association is believed to be that of a solid state solution of hydrogen in titanium[4] rather than a true chemical combination, although the detailed mechanism of binding is not involved in our present considerations. A factor of dominant importance however is the increase of vapor pressure of hydrogen from the solid state solution with increasing temperature, with it reaching the 10^{-5} torr range at 250°C. This establishes a critical temperature boundary condition which if exceeded by any portion of the titanium results in reemission of absorbed hydrogen, which can trigger thermal instability.

Equilibrium Temperature

The 500 l/s pump operates at a pressure of about $2x10^{-5}$ torr and with a power input of about 400 W when pumping hydrogen at the rate of 1 atm cm^3/min, or 100 W for each of the four pumping elements. The equilibrium operating temperature of the element is clearly sensitive to the thermal impedance between it and the pump housing. No provision has been made in the manufacture of the pump for good thermal contact since it is not required for high vacuum applications of the pump. If negligible thermal contact exists, element temperature will rise to a value sufficient to radiate the input power to the pump housing. Assuming a black body radiator the temperature will rise to about 280°C, and if the emissivity is 0.2 the temperature will rise to about 300°C. Randon thermal contact will reduce the temperature substantially, but it is clear from these numbers that a very marginal situation exists for the hydrogen gas load in question unless corrective measures are taken.

Improved Thermal Contact

In view of the above considerations it was decided to experiment with special clamps designed to improve by a large factor the thermal contact between titanium cathodes and pump housing. A very marked improvement of performance was realized and subsequently this approach was extended to include copper bars to reduce thermal gradients between clamps. Fig. 4 shows clamps (spreader jacks) and copper bars installed in a pump.

Cathode Hot Spots

Square and Round Anode Cells

Pumping element anodes were originally made with square cells, but are now made with round cells. Both produce a discharge pattern with a sharp intensity maximum on the axis as shown by the pitting of the titanium in Fig. 3. This concentration of the discharge onto a small fraction of the total surface area of the titanium results in temperature spikes or hot spots on the titanium.Since maximum titanium temperature occurs in these hot spots, we can expect thermal instability to be directly associated with them.

Experimental observations indicate that with a

*Work performed under the auspices of the United States Atomic Energy Commission

hydrogen gas load of 1 atm cm^3/min the temperature in at least some of the hot spots equals or exceeds the critical temperature of 250°C. Under this condition an increase of discharge power, resulting from an increase either of pressure or of anode voltage, will increase the volume of titanium heated to a temperature above the critical temperature. If the rate of release of hydrogen from this volume is greater than the rate of its reabsorption in cooler areas of the titanium, thermal instability will result. Conversely, a decrease of discharge power will allow a small volume of titanium to cool sufficiently to again retain hydrogen and thus give a transient increase of pumping speed. Fig. 5 illustrates these effects.

It can be predicted from the above observations that stability can be improved if discharge power can be made invariant with pressure and anode voltage.The inclusion of a suitable series resistor in the anode lead, which gives an approximation to a constant power characteristic, improves stability. A more sophisticated constant power circuit could readily be devised.

Rectangular Anode Cells

Observations have been made with experimental pumping elements employing anodes with rectangular cells to give a discharge with plane rather than axial symmetry to eliminate the hot spots that occur with square and round cells. Fig. 6 shows an experimental anode with rectangular cells together with a mating cathode after about 500 hours of operation. Preliminary results are sufficiently promising to justify further evaluation. Series stabilizing resistance has been found unnecessary with this geometry.

Pumping Speed vs. Temperature

When the Cosmotron operating schedule was reduced from 21 to 15 shifts per week requiring weekend shutdown, it was observed that the cold startup pumping speed of the pump was only about 25% of its equilibrium speed after warmup,- with a period of 4-5 hours being required to reach equilibrium,- suggesting a rapid variation of pumping speed with temperature. This led to an extended series of observations of equilibrium pumping speed vs. temperature over the range from -50°C, obtained by surrounding the pump with dry ice, to 250°C, obtained by wrapping the pump with fiberglass insulation and inserting auxiliary heaters beneath it. Fig. 7 shows the surprisingly small variation of pumping speed that occurs over this temperature range,- aside from the rapid decrease as the temperature approaches 250°C.

The loss of pumping speed on cold startup was not accounted for by the variation of pumping speed with temperature. Another explanation had to be found, and it became apparent when it was observed that if the pump was turned off for the weekend while operating at a temperature of 150-200°C no delay in recovery of full speed on cold startup occurred. The normal equilibrium operating temperature without thermal insulation is 110°C.
The distribution of hydrogen in the titanium in the immediate vicinity of the hot spots and its rate of change with time and temperature appears to provide a satisfactory explanation for the observations as illustrated by Fig. 8. The equilibrium distribution while hydrogen is being pumped has a minimun in the center due to the high temperature (A). When the pump is turned off at the normal operating temperature of 110°C diffusion of hydrogen in the titanium during cooling will give a distribution somewhat as shown by B. Due to the increased concentration of hydrogen on the axis, hydrogen will be reemitted from the center of the hot spots on cold startup until the equilibrium distribution is reestablished. The pumping speed will be lower during this period. With turnoff at 200°C, the increased mobility of hydrogen at the higher temperature and the longer time required for cooling permits the outward diffusion of hydrogen to reduce the concentration at the center to a value sufficiently small that reemission does not occur on startup (C). These observations, together with that on deformation noted below, provide evidence that the mobility of hydrogen in titanium is a rapidly varying function of temperature.

Titanium Deformation

Initial Short Circuit Life

Fig. 9 shows the nature of the titanium deformation that led to anode-cathode short circuit after 2000-3000 hours of operation before corrective steps were taken.

Stiffening Posts

Short circuit life expectancy was markedly increased by the insertion of stainless steel stiffening posts between cathodes on the axis of selected anode cells. This led to a lesser problem resulting from the sputtering of post material onto the anode with subsequent flaking and short circuit. These shorts are easily cleared however with a condenser discharge. The sputtering is caused by a cold cathode magnetron discharge occurring in the volume between post and anode cell and is discussed further below.

Titanium Segmentation

Segmentation of the titanium into relatively small pieces is an obvious approach to the elimination of the cumulative effect of distortion over large areas. The cathode shown in Fig. 6 has 1 inch square titanium pieces attached to a stainless steel support plate. This technique provides a completely adequate solution to the deformation problem. However as noted below high temperature operation provides a much simpler solution.

High Temperature Operation

The high temperature operation that was found to eliminate cold startup problems has also been found to virtually eliminate distortion of the titanium with hydrogen absorption, presumably due to the greater mobility of hydrogen in the titanium with corresponding reduction of concentration gradients.

Operation of the titanium at the highest temperature permitted by the 250°C critical temperature boundary condition appears to be of major importance in the realization of optimum performance for hydrogen pumping. Operation of the titanium at the highest allowable average temperature requires, 1) the use of either low or carefully controlled thermal impedance between titanium cathodes and pump housing, 2) minimization of temperature gradients over the titanium, and 3) avoidance of hot spots due to discharge concentrations.

The increased mobility of hydrogen in titanium at high temperature will permit the use of thicker titanium cathodes. This will give the advantages of smaller temperature gradients and greater ultimate storage capacity for hydrogen.

Pumping Speed

The large change of hydrogen mobility and the small change of pumping speed observed with changes of temperature suggest that pumping speed is determined primarily by physical processes occurring at the surface. The observed increase of pumping speed by a factor of 2-4 after about 100 hours of operation has been shown to be associated with surface cracking and corresponding increase of surface area. For this reason in order to maximize pumping speed it appears desirable to maximize surface to volume ratio. Possible approaches are slotting the titanium or fabricating porous cathodes by sintering titanium granules or powder.

Magnetron Pumping

Following the observations noted above that a magnetron discharge occurs in cells with a center post, an experimental pumping element was fabricated with a titanium rod on the axis of each cell to determine the effectiveness of the magnetron discharge compared to the Penning discharge. Only very preliminary results are available but they indicate, 1) that a stable magnetron discharge occurs in all cells, 2) that the discharge has a strongly negative current-voltage characteristic, and 3) that pumping action starts at a substantially higher pressure than with the Penning discharge. The magnetron discharge has the advantage of avoiding the hot spot problem. Further work is required to fully evaluate the potentialities of this type of discharge.

Startup

Startup of a pump which has been thoroughly degassed and exposed only to dry gases is relatively easy, and is simply a matter of reducing the pressure to about 1 micron where the Penning discharge starts to pump. However startup of a new pump, one which has been exposed to wet air, or one which has been apart for rework is more difficult due to the evolution of gas from the surfaces. The degassing process can be greatly accelerated by heat which can be supplied either with bakeout heaters or by the glow discharge which occurs during the startup phase. However the circuits supplied with the pumps are poorly suited to the delivery of appreciable power into the glow discharge due to the low voltage at which it occurs (about 400 volts). It appears desirable to employ a special startup power supply capable of delivering 2-5 KW into the glow discharge for rapid heating and degassing. While the pump can be forced to ingest the products of degassing, it is preferable to remove them from the pump with a sorption, sublimation, or turbomolecular pump during the startup phase.

Further Development

Further work is indicated to fully evaluate, 1) rectangular anode cells, 2) slotted or porous cathodes, 3) magnetron discharge pumping, and 4) accelerated startup. On the basis of presently available information it appears possible to design pumping elements for a 500 l/s pump capable of pumping hydrogen continuously for periods up to 3-5 years at a rate of 1 atm cm^3/min, or at rates of up to at least 5 atm cm^3/min for correspondingly shorted periods. Application of these design principles to larger pumps should give proportionately higher ratings.

Acknowledgements

The assistance in this work of the following Brookhaven National Laboratory personnel is gratefully acknowledged,- W. R. French, E. W. Hoyle, W. J. Jordan, and E. A. Lange.

References

1. L. D. Hall, Electronic Ultra-High Vacuum Pump, R. S. I. 29, 367 (1958).
2. R. L. Jepsen, Important Characteristics of a New Type Getter-Ion Pump, Le Vide 80, 80 (1959).
3. Varian Associates Vacuum Products Division Instruction Manual #87-400 079, Sept. 1964, p10.
4. S. L. Rutherford and R. L. Jepsen, Enhanced Hydrogen Pumping with Sputter-Ion Pumps, R. S. I. 32, 1144 (1961).

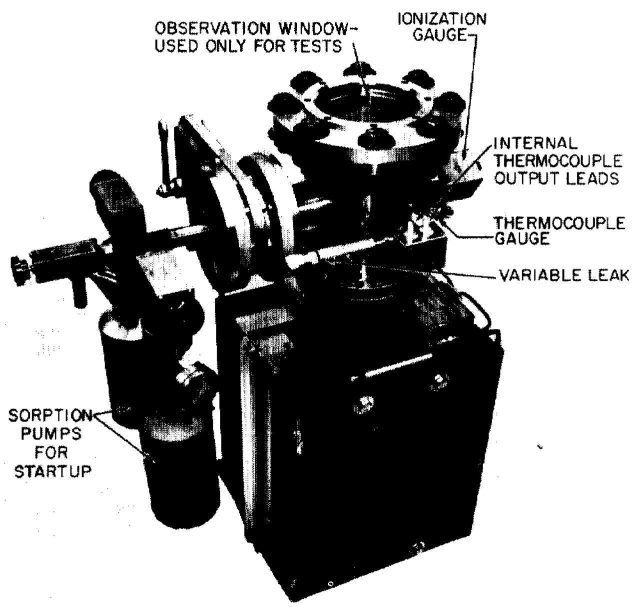

Fig. 1. Complete 500 l/s (air rating) VacIon pump assembly. Pump manufactured by Varian Associates, Palo Alto, Calif. Hydrogen pumping speed of new elements is approximately 500 l/s, increasing after about 100 hours of hydrogen pumping to approximately 1200 l/s. A fine mesh tungsten screen across the pump throat confines the startup plasma to the volume of the pump. The internal thermocouple is provided to monitor titanium temperature which is a critical parameter for reliable hydrogen pumping.

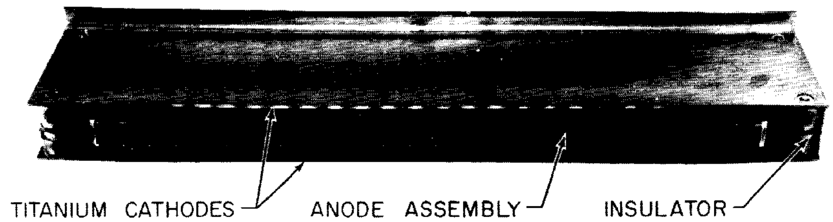

Fig. 2. Assembled pumping element with round cell anode. Four elements are used in the 500 l/s pump. Length = 18 1/2 in, width = 3 7/8 in, depth = 1 5/8 in.

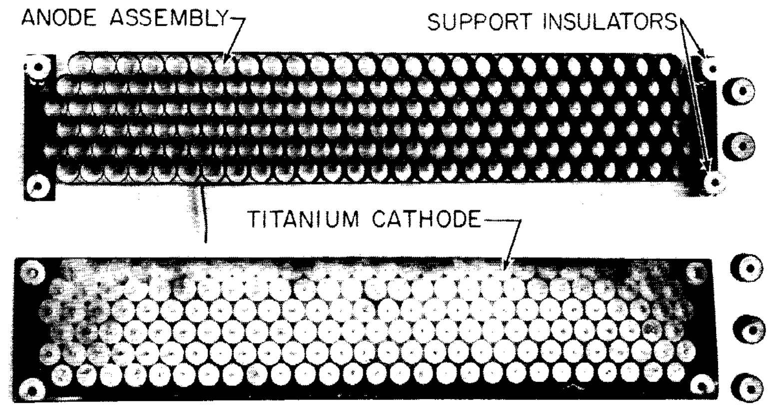

Fig. 3. Disassembled pumping element, showing one cathode only. This element was operated for 2000 hours at a hydrogen gas load of 1 atm cm^3/min when it shorted due to titanium deformation. Pitting of the titanium by the concentrated discharge in each anode cell is to be noted.

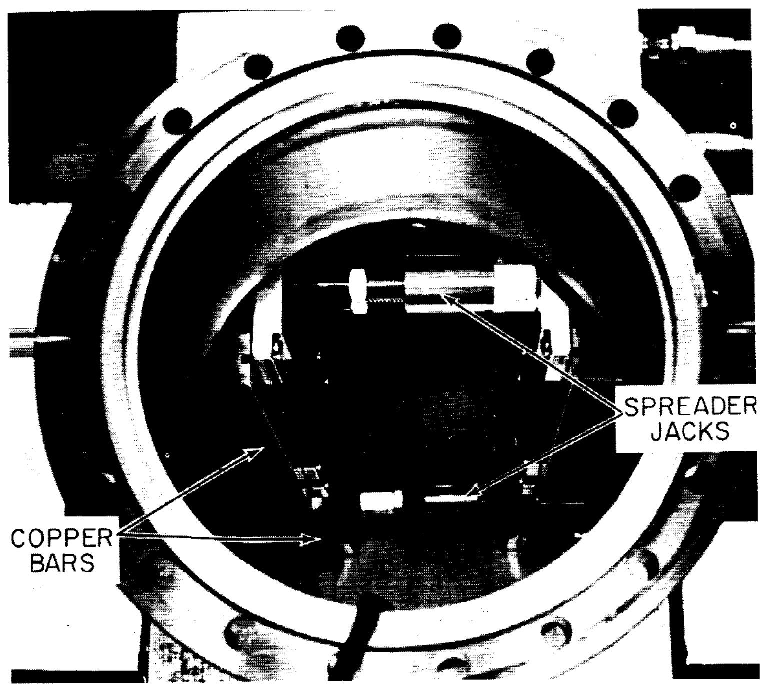

Fig. 4. Pair of pumping elements with copper heat equalizing bars installed in a pump. The spreader jacks establish intimate thermal contact between titanium cathodes and pump housing.

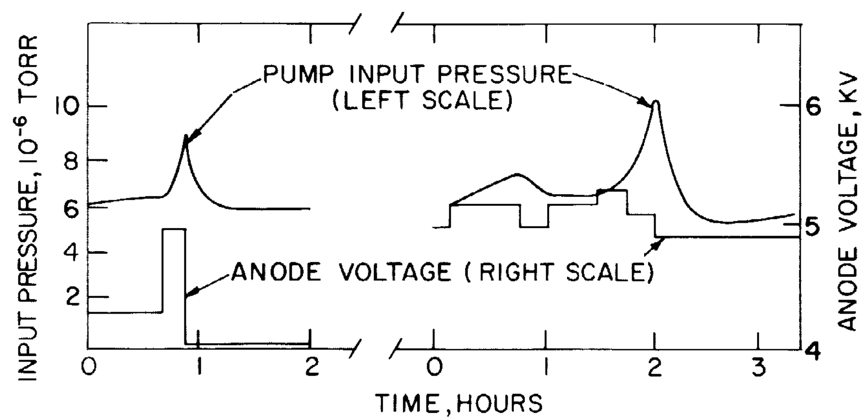

Fig. 5. Pressure vs. time following changes of anode voltage at constant hydrogen feed rate. Time lag due to the time required for the distribution of hydrogen in the vicinity of the hot spots to reach a new equilibrium.

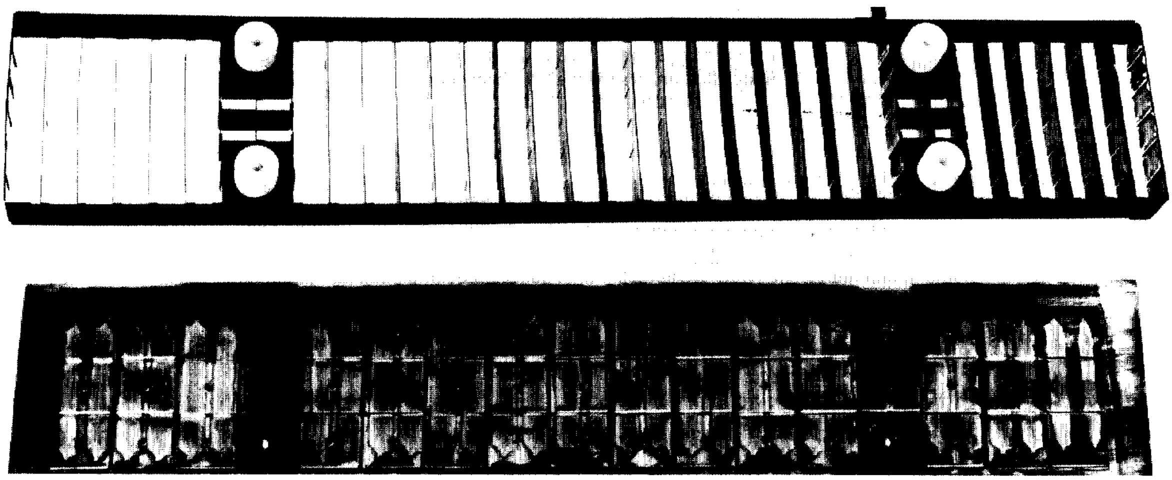

Fig. 6. Experimental rectangular cell anode with one mating cathode used with it for 500 hours. Cathode has titanium segmented into 1 inch squares attached to a stainless steel backing plate. Length = 18 1/2 in, width = 3 7/8 in.

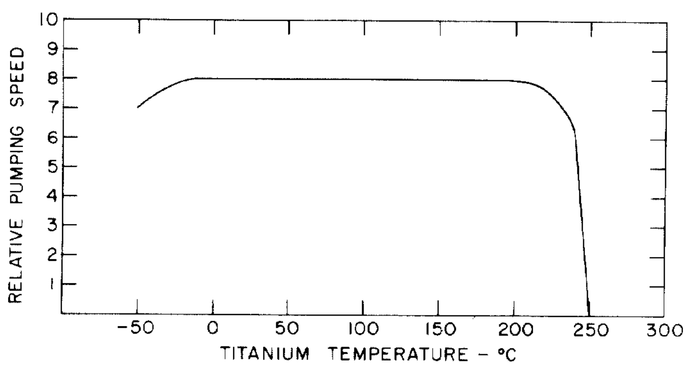

Fig. 7. Pumping speed for hydrogen vs. temperature of titanium.

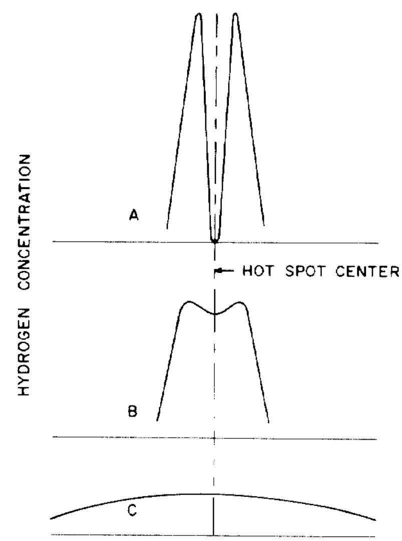

Fig. 8. Postulated distribution of hydrogen in the vicinity of a discharge hot spot.
A. Equilibrium distribution during operation.
B. Distribution 48 hours after 110°C turnoff.
C. Distribution 48 hours after 200°C turnoff.

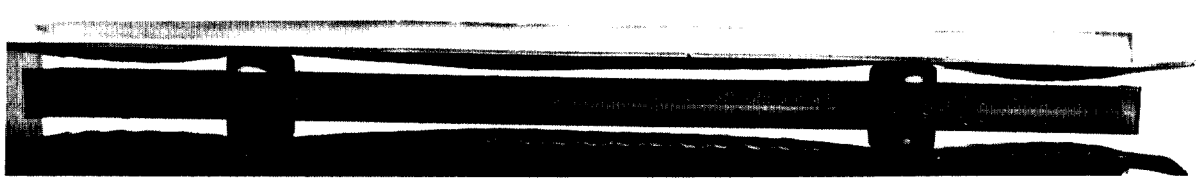

Fig. 9. Titanium distortion with hydrogen absorption. Short circuit occurred after 4000 hours of operation at a hydrogen input rate of 1 atm cm^3/min.