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A Windowless Frozen Hydrogen Target System

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Abstract

A cryogenic target system has been constructed in which gaseous mixtures of all three hydrogen isotopes have been frozen onto a thin, 65 mm diameter gold foil. The foil is cooled to 3 K while inside a 70 K radiation shield, all of which is mounted in a vacuum system maintained at 10^{-9} torr. Stable multi-layer hydrogen targets of known uniformity and thickness have been maintained for required measurement times of up to several days. To date, hundreds of targets have been successfully used in muon-catalyzed fusion experiments at TRIUMF.

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1. Motivation

Experiments involving negative muons and hydrogen isotopes have been carried out for many years. In particular, a negative muon stopped in hydrogen will form muonic atoms and molecules and, due to the small size of muonic hydrogen molecules, the nuclei often undergo fusion. The reactions leading to muon-catalyzed fusion are complex, and the interested reader is referred to the recent reviews [1,2].

It was shown at TRIUMF that beams of muonic deuterium atoms were emitted from a thin solid layer of natural hydrogen ($^1H_2$ with 140 ppm $^2H_2$) maintained in a high vacuum environment [3]. Emitted beams of muonic tritium have also been observed from targets of tritium-doped protium [4]. Time-of-flight measurements with muonic tritium atoms permitted the direct exploration of the resonance formation of muonic molecules, dμt, in the region of epithermal interactions [5]. Use of the target has allowed other physics processes in muonic atoms to be studied [6-8].

2. Design and Construction

Problems in cryogenics, vacuum design, gas mixing, and tritium safety were addressed in the design of this target system for experimental studies of muon-catalyzed fusion in hydrogen isotopes.

Fortunately the most demanding design restriction, the compatibility with tritium, was not often in conflict with the other requirements. The design had to eliminate, or reduce to a very low level, the possibility of leaks, and ensure that any release of tritium would occur in an area (or at a rate) which would not endanger personnel. Internal contamination had to be minimized to permit target disassembly, so materials such as elastomers were avoided to reduce contamination through replacement reactions of the tritium with any compound containing hydrogen.

The observed lifetime of a muonic hydrogen atom is very sensitive to contamination from heavier nuclei due to transfer reactions which compete strongly with the reactions being investigated. The all-metal vacuum system helped to achieve both cleanliness and a high vacuum, and in addition reduced the possibility of tritium contamination of internal surfaces.

2.1. The Cryostat

The cryogenic system was designed by a company specializing in custom refrigerators[†]. The upper portion of the target system is represented in Fig. 1, the core of which is a modified continuous-flow cryostat (detail A). A standard unit[‡] was extended to a length of 38 cm to reduce the thermal conductivity between the mounting flange and the cold stage. At two points along the body of the cryostat, cylindrical copper blocks (detail B) were attached to provide thermal contact points for the external target attachments (details G,I). The blocks provide conduction paths to the 3 K and the 70 K stages of the cryostat.

The cryostat functions by the continuous transfer of cold helium gas from a storage dewar through the cryostat and out through a pump. A needle valve located in the section of the transfer line inserted in the dewar restricts the flow, while the pump provides low pressure allowing the helium to vapourise and cool. The flow of cold helium gas through baffles in the cryostat provides the refrigeration. Additional steel wool installed in the helium gas flow path increased the cooling power of the 70 K thermal contact stage to 10 W.

[†]Quantum Technology Corporation, 1370, Alpha Lake Road, Unit 15, Whistler, B.C. Canada

[‡]SuperTran, Janis Research Company, Inc. Wilmington MA, 01887-0696

The cryostat unit was inserted into a long closed cylindrical stainless steel tube of 500 μm wall thickness (detail C) whose upper end was welded to a 200–mm diameter knife-edge vacuum flange. The tube acted as a second barrier between the helium flow in the cryostat and the region of the vacuum system exposed to tritium. Thus the continuous flow of helium was separated from possible tritium contamination by two all-welded vacuum barriers; the tube, and the cryostat itself. Doubling the containment of the tritium strongly reduced the chance of introducing tritium contamination into the helium circuit, thereby allowing the helium to be safely recycled. The tritium barrier space, or TBS (detail D), between the outside of the cryostat and the inside of the tube was separately evacuated and monitored.

With the cryostat installed in the tube, the cylindrical copper blocks ensured a good thermal contact by providing sufficient surface area to overcome the limited thermal conductivity of the steel tube (C). Indium was used in the joints to increase the thermal conductance. Fig. 2 shows the cross section of the upper contact region (Section S in Fig. 1) detailing how external thermal loads (detail I, and similarly for G) were attached to the outside of the tube at the positions of the copper blocks. Stainless steel clamp rings (detail E) held together with 5/16" steel bolts, provided a high pressure on the joint (limited by the sheer strength of the bolts) sufficient to conduct 10 W at 70 K.

The cross section of the external attachments changed from circular, at the contact points (detail E, Fig. 1), to square, which simplified the manufacture of the target attachments and maximized the usable internal volume.

The two gold target foils 50–μm thick and 65 mm in diameter were cold soldered, using indium, to gold-plated copper frames (detail F). The copper frames were attached to the coldest stage of the cryostat (detail G) by a clamp which allowed the spacing between the foils to be varied between 16 and 40 mm (detail H). Indium was used to ensure a good thermal contact between the removable target holding frames and the 3 K cold stage.

A radiation shield (detail I) fully surrounding the cold target stage, was attached to the cryostat at the 70 K cooling stage. The 10 W heat load due to thermal radiation from the room temperature vacuum system was thus removed by utilizing the cold exhaust gas from the 3 K cooling stage.

This radiation shield provided a convenient mount for silicon detectors used to observe reaction products originating at the targets. The shield also supported a guide rail system (detail J) for the precise insertion of the target gas deposition unit. The gas deposition unit, discussed below, was cooled via conductive copper braids (detail K).

The use of conventional super-insulation was precluded by the incompatibility between tritium and hydrogen-containing compounds such as plastics and elastomers. Instead, the low emissivity of gold-plate was used to reduce the radiative heat load from all internal surfaces. The inert nature of the plating made it resistant to oxidation, and the low permeability to tritium reduced the contamination absorbed by the internal components.

Stray muons reacting with the gold produce a neutron background due to muon capture, but the neutron emission time distribution from such nuclei is prompt and well understood. The dominance of gold on all exposed surfaces ensures that only this well understood background was present.

The temperatures of parts of the target were monitored to verify proper operation. Constant-current silicon diodes were used at several places to monitor temperatures to an accuracy of a few Kelvin, while sensors specific to low temperature, such as carbon glass resistors or a CERNOX©§ low temperature sensor, were used on the target frames themselves where better information on target temperature was required.

§All thermometers were acquired from LakeShore Cryotronics, Inc., Westerville, Ohio

Discrepancies of ≈ 0.5 K were observed in measured temperatures. Thermometers mounted on the target support foils indicated a temperature different from other similar devices, as well as from that determined from the relatively well known vapour pressure of hydrogen at low temperatures. This was attributed to difficulty in thermally anchoring the thermometer leads without the use of glues and materials incompatible with tritium. Although the lack of an absolute calibration was troublesome, the thermometers reliably indicated changes in the temperature. Experience has shown that the vapour pressure of hydrogen in the vacuum system was a more useful measure of the target operation temperature and its stability — the parameters of greatest value in monitoring target performance.

2.2. The Deposition System

Hydrogen gas was deposited on the target foils by releasing it through the device pictured in Fig. 3. A central gold-plated copper sheet (detail L) supported two gas diffusers, independently connected to the gas supply to permit the deposition of a solid 60-mm diameter layer of hydrogen onto either of the two target foils.

Each side of the deposition system consisted of a gas supply line (detail M) from the gas mixing manifold, and a specialized chamber (detail N) designed to deposit the gas evenly on the chosen cold target with less than 0.5% deposited on the opposite target. The chamber was built from a thin cylindrical hollow disk, the diffuser covered with a 500-μm thick layer of 2 μm porosity-sintered metal and the other by a gas tight backing of stainless foil 50 μm thick. A section view of a diffuser is shown in Fig. 4. The gas was supplied through 2 mm inside-diameter tubing connected to the deposition chamber with a short reducing junction. With the two diffusers mounted on the copper support the total thickness of the part inserted between the target foils was 9 mm. This limited the separation between the target foils to a minimum of about 16 mm when thermal distortion precautions and maximum hydrogen target thickness were included.

The copper sheet and diffusers were cooled to 100 K to suppress radiation loading of the target. Two flexible copper braids (detail K, Figs. 1,3) were connected to the copper sheet and the bottom of the radiation shield to provide a flexible thermal conduction path. The braids were long enough to allow the diffuser mount to move a total vertical distance of 160 mm.

The copper sheet was mounted on a tall thin-walled cylinder of stainless steel (detail O) which served as a rigid mechanical, yet relatively poor thermal, contact to a movable flange. The movable flange was held between two bellows (detail P) to remove atmospheric pressure from the moving components, thus lowering the force required for motion and reducing the possibility of component failure. Two guide shafts and a lead screw (detail Q) provided well-controlled vertical motion. Guide rails on the inside of the radiation shield (detail J, Figs. 1,5) ensured that the copper sheet with its deposition units was accurately located between the target support frames.

The present deposition system design was based on an earlier version constructed using 0.8 mm inside-diameter tubing, and a chamber with a 70-mm diameter stainless steel foil pierced with approximately seven hundred 0.2 mm diameter holes spaced in a 1 mm rectangular array. Difficulties arose in controlling the deposition rate and total amount of gas released while using this system, so conductance measurements were made to optimize the current design [9]. In the previous design, the poor conductance of the long small-diameter supply lines resulted in a very long pump-down time to clean the lines, and a pressure of several Torr at the gas inlet even when the target region was at a very high vacuum (10^{-7} Torr). To reduce the time required for target deposition and to gain more precise control of the amount of gas deposited, larger diameter tubing was installed.

2.3. *The Target Vacuum System*

The gas deposition system and the cryostat were both attached to a stainless-steel cube using knife-edge seals. The cryostat was introduced through the top port of the cube which had been modified from a circular to a square aperture to accommodate the profile of the heat shield. The deposition and pumping systems were attached through the bottom port, leaving the four remaining sides of the cube for a muon entry port and windows through which detectors could view the target. Fig. 5 shows a view of the cube. The target support foils (detail F) are seen through the radiation shield (detail I) which is drawn with one of its side plates removed for clarity. The partially inserted gas deposition system is shown entering through the bottom of the radiation shield (detail L).

Muons enter the system through a custom flange which had a thin (25 μm) stainless-steel vacuum isolation window to protect the beam line vacuum from possible tritium contamination, and to protect the clean high-vacuum target from oil and other contaminants from the beam line vacuum system. Thin stainless-steel windows (nominally 50 μm thick) covered the remaining cube ports through which emitted radiations such as neutrons, gammas, and muon-decay electrons exit to the external detectors. The left and right side windows of the cube were used for neutron detectors, germanium x-ray and γ-ray counters, or wire chamber position tracking detectors, each of which was mounted externally to the vacuum system. The internally located silicon detectors, designed to see the charged products of muon-catalyzed fusion, were mounted on the thermal shield.

The entire vacuum system was constructed using all-metal, knife-edge fittings of a nominal 150 mm diameter, and all-metal valves[¶]. The vacuum system (without the cryostat installed) was cleaned after assembly by use of an oxygen plasma. A pressure of 300 mTorr of oxygen was required to start the cleaning. Once the plasma ignited, a pressure of 60 mTorr and a current of 1.3 A (which provided the brightest glow from the inside of the vacuum system) was maintained for 30 minutes. Pressures of $\sim 10^{-7}$ Torr were then routinely achieved after baking when pumped by a magnetic levitation turbo pump[‖], backed by an oil and elastomer free roughing pump system[**]. The exhaust of the pumps was directed to an evacuated 100 l volume to provide a closed-cycle pumping system for safe collection of residual tritium from the vacuum system.

2.4. *Gas Mixing and Handling System*

To permit work with mixtures of hydrogen isotopes, a comprehensive system for purifying and mixing hydrogen gas was constructed. Experience from two previous hydrogen mixing systems was incorporated in the design shown in block-schematic layout in Fig. 6.

Isotopically pure protium was generated by electrolysis of deuterium-depleted water using a small commercial unit[††]. Deuterium was obtained from bottled research-grade deuterium which was passed through a commercial palladium filter[‡‡] (detail R). Because small admixtures of protium in the deuterium would not disturb our measurements, ordinary research grade deuterium cleaned of non-hydrogens was sufficient. Other gasses, such as He or Ne, were supplied from bottles through an external port.

[¶] VAT, Haag, CH-9469, Switzerland, and MDC, Hayward CA, 94545

[‖] TurboVac 340M, Leybold Vacuum Products Inc., D5000 Cologne 51, Germany

[**] A combination of a Normetex D7 spiral pump, and a PV-12 bellows pump, Normetex corporation, France

[††] Elhygen Mark V, Milton Roy, Ireland

[‡‡] Johnson-Mathey Inc., Wayne PA, 19087

The small quantity of tritium required for a deposition (of the order of 10 Ci per target) was removed from a small (5 g) uranium getter bed by heating the bed (detail T). The storage on and liberation of hydrogen from uranium is a well-understood technology [10,11]. In this way the hydrogen was contained in solid form below atmospheric pressure and the difficulties associated with the storage of large amounts of gaseous radioactive material were avoided.

The mixing system consisted of two accurate capacitive manometers [§§] (detail U) and one accurately measured volume (detail V). The volumes of all the other mixing components were obtained to better than 1% uncertainty by accurate pressure measurements of gasses as they were expanded into the volumes. Target gas mixtures were made by capturing a calculated amount of each constituent gas in a separate measured volume, and then opening the interconnections between the volumes.

Special attention was taken to reduce dead volume and to maximize conductance and pumping speed. To reduce the amount of wasted gas (especially tritiated gas mixtures) the total volume of interconnecting tubing between the standard volumes was kept small. To allow rapid mixing between the standard volumes there were short-length large-conductance connections between them. To reduce the possibility of leaks in the tritium-compatible system, all-welded construction was required. These requirements were achieved by the use of two all-welded "pucks" — each an octagonal block of stainless steel with nine connection ports, one per octagon face, plus one port in the back. The ports were fitted with all-metal face-seal connections[¶¶]. The resulting unit allowed the attachment of nine components (such as gas supply and exhaust, mixing volumes, and pressure sensors) with all components connected through low-volume high-conductance lines. Components were joined either by welding, or by all-metal face-seal connections. The valves [|||] in general use were fitted with with Vespal shut-off stem tips; the valves exposed to pure tritium were fitted with copper tips. A quadrupole gas analyzer was added to the system as a leak detector, and as a monitor on the purity of the target gas (detail Z).

With this system, complete mixing of initially separate target gasses required two hours. This time was measured with the gas analyzer by repeatedly sampling the target gas at one point in the system until the ratio of component gasses was stable. Once mixed, the gas was released through one of the two deposition lines (detail M) at a slow rate controlled by a metering valve (detail W). The gas passed through the diffuser system and onto the selected cold target foil.

The tritiated hydrogen isotopes remaining in the gas handling system following a deposition, as well as the frozen target at the conclusion of the experiment, were pumped onto a large (3.6 kg) uranium-hydride getter pump (detail X). Any residual gas not collected in this way was moved into the waste volume by the pumping system (detail Y).

To cope with a catastrophic failure of one of the system components when tritium was in use, the entire experimental system was placed in an enclosure which was vented at a rate of 50 m^3/min of air to a roof exhaust. Stoppered ventilation holes allowed air to enter the enclosure, but permitted the box to be closed when the vent was off. Tritium-in-air monitors were used to check the vent exhaust for tritium; no unexpected releases were observed. Controlled releases have been made while purging the system after use and while testing the safety system.

[§§] MKS Instruments Canada Ltd., Nepean, Ontario, Canada

[¶¶] VCR metal gasket, Cajon, Macedonia, Ohio, 44056

[|||] SS-4BK-V51, Nupro, Willoughby, Ohio, 44094

3. Operation

The continuous flow commercial cooling unit has provided good stability and reliability of operation. Cooling of the target system from room temperature can be accomplished in about four hours, with optimum running conditions achieved after twelve hours. When cold, the system consumes approximately 2.8 *l* of liquid helium per hour to maintain the 3 K target foil temperature. By use of a 500 *l* dewar of He, the target system was able to maintain a solid hydrogen layer for about a week, permitting studies at low event rates without alteration of the target conditions.

Of major concern in the design was the maximum achievable cooling power. High cooling power was essential not only to maintain the 3 K temperature when a target had been deposited, but also to deposit a target of roughly one standard litre of hydrogen in a reasonable time. The measured cooling power of the cryostat is ~100 mW/K at the nominal 3 K operating temperature. This is sufficient to freeze a 1 *l* target to a foil in about 30 minutes.

A nominal pressure of 10^{-7} Torr was achieved at room temperature using the turbo pump, with water the dominant residual gas. When cold, the cryostat, because of the large surface area of the thermal shield, pumped more rapidly than the turbo pump. Under these conditions, the turbo pump was isolated from the vacuum space, and the cryostat maintained the vacuum outside of the cold shield at 10^{-9} Torr.

The thickness and uniformity of the solid targets, studied by measuring the energy loss of alpha particles transmitted through the layer [12], is reproducible to 1% and uniform to 15%. This reliability extends to targets which are deposited in several different steps of different isotope concentrations even when many hours have passed between the application of each layer.

Safe operation with tritium required an interlock system. Several key valves, and many sensors, were connected to a programmable logic control (PLC) which automatically implemented basic protective measures in case of an emergency. The main use of the PLC was to monitor the levels of the tritium-in-air monitors and to warn the operators of changes. The PLC also sensed any failure in the roof vent airflow or in the cooling of the cryostat, and signaled an alarm.

The disassembly and modification of the target, to change the target separation for example, occurs in a glove box specifically designed to extract the cryostat system from the vacuum system and to securely hold the apparatus during the modification. For personnel protection the extraction and modification procedure occur entirely within the vented enclosure system.

Acknowledgements

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Figure captions

- Fig. 1 A schematic view of the cryostat showing the commercial cryostat (A), heat transfer copper cylinders (B), secondary containment (C), TBS vacuum (D), attachment clamps (E), target support foils (F), 3 K cold stage (G), variable clamp (H), heat shield (I), diffuser guide rails (J), and diffuser cooling braids (K). The section S, Fig. 2, shows a cross-section of the heat shield attachment. To permit a clear view of the relevant components, the drawing is not to scale.

- Fig. 2 A cross section of an attachment point, section S from Fig. 1. The cryostat (A), heat transfer copper cylinders (B), secondary containment (C), TBS vacuum (D), and attachment clamps (E) are shown in relation to the target attachments (I, and similarly for G).

- Fig. 3 A view of the gas deposition diffuser showing the central diffuser support (L), braids to the heat shield (K), gas supply lines (M), diffuser chamber (N), mechanical support (O), bellows and vertical motion guide shafts (P,Q). The projection shows only one side of the full diffuser system; the second diffuser is obscured by the support.

- Fig. 4 A vertical-section view of the gas diffusion assembly. The *horizontal* size is magnified by a factor of eight and the gas supply tubing has been removed for clarity. The top of the copper support, (L), is shown where the two diffusion chambers are mounted in recesses in the sheet. The gas flow path into the chamber and out through the sintered metal is illustrated.

- Fig. 5 A perspective view of the assembled target system viewed through an open side window with the thermal shield side plate removed. The orientation of the target foils (F), heat shield (I), and the diffuser (L) in its guide rails (J), is illustrated.

• Fig. 6 The vacuum system and gas mixing topology for the target system. Block (R) illustrates the protium/deuterium supply, (T) the small uranium getter bed supplying tritium. Other components include the capacitive manometers (U), mixing volume (V), metering valve (W) and deposition lines (M). The large uranium getter is block (X), while the pumping and RGA analysis are contained in blocks (Y) and (Z) respectively. The arrows show the flow scheme of gasses in the system.

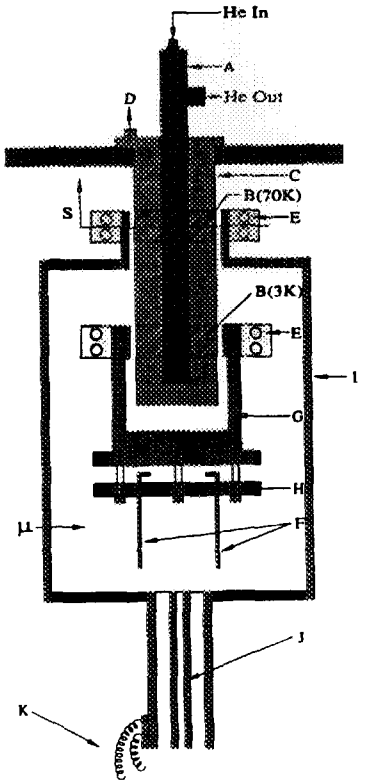

Fig. 1

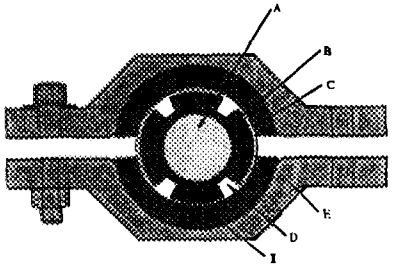

Fig. 2

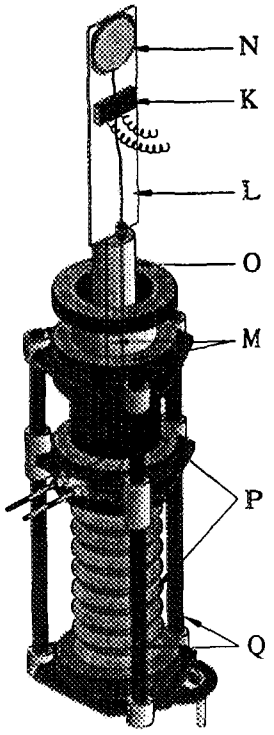

Fig. 3

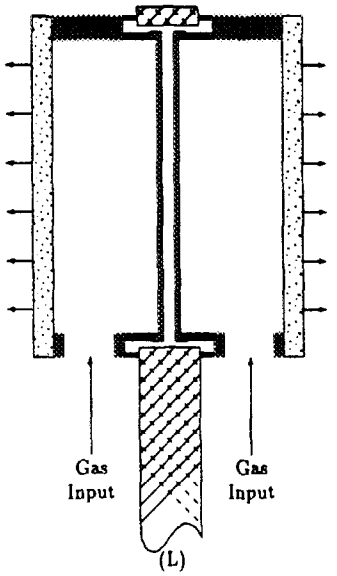

Fig. 4

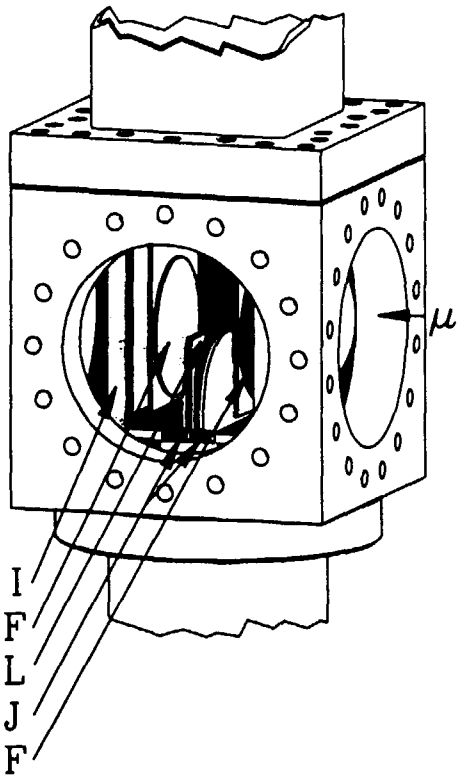
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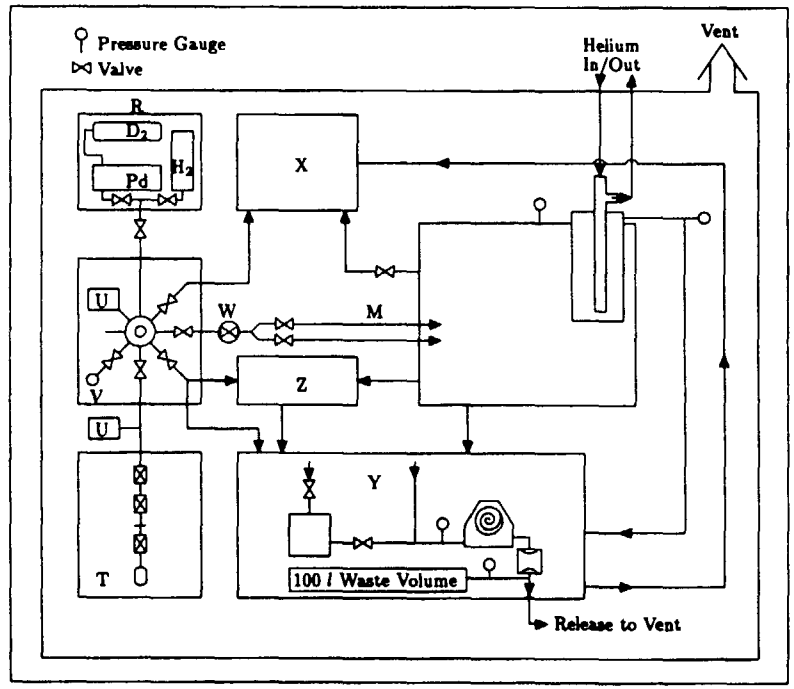
Fig. 5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H_2
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100 l Waste Volume
Release to Vent

Fig. 6