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MASTER

CRYOCONDENSATION PUMPING

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ABSTRACT

The work on cryocondensation of hydrogen and helium isotopes on smooth surfaces held at 4.2 K has been completed and published,¹⁻⁴ and the problems introduced by the presence of tritium in the pumping systems have been identified. In this report we want to summarize our findings and point out their implications on the design of large pumping systems. Presently, we plan to conduct a similar study on cryosorption in the temperature range of 10 to 22 K in order to compare the two methods of pumping.

1. H.J. Halama and J.A. Bamberger, Proc. Sixth Symp. Engineering Problems of Fusion Research, San Diego (IEEE, N.Y., 1975) p.202.
2. T.S. Chou and H.J. Halama, Proc., 7th International Vacuum Congress, Vienna, September 1977 p.65.
3. H.J. Halama, C.K. Lam and J.A. Bamberger, J. Vac. Sci. Technol. 14, 1201 (1977).
4. T.S. Chou and H.J. Halama, Proc., 7th Symposium on Engineering Problems of Fusion Research, Knoxville, October 1977, p.1790.

I. PURE GASES

1. Within a 10% accuracy the pumping speeds of H_2 isotopes are independent of surface coverage and gas fluxes to be pumped. The highest coverages and gas fluxes tested were 1.5×10^{21} molecules/cm² and 3×10^{-3} Torr liter/sec cm² respectively. Since the sticking probability of hydrogen isotopes is close to unity their pumping speed, S , in a typical pump is limited by the conductance of the chevron baffle, necessary to shield the cryopanel from thermal radiation. S is therefore proportional to the velocity of the molecules being pumped. For economical operation, typical pumping speeds for hydrogen, deuterium and tritium are 11, 8 and 6.5 liter/sec cm² respectively. In the case of tritium, He^3 has to be removed by other means.

2. Equilibrium pressure is also independent of surface coverage and is governed mainly by the residual helium pressure. If the residual helium gas is negligible, a pressure of approximately 1×10^{-6} and 5×10^{-8} Torr (N_2 equivalent) are typical in systems pumping H_2 and D_2 respectively. Tritium vapor pressure is $< 10^{-11}$ Torr, but He^3 has to be continuously pumped by auxiliary pumps.

3. Thermal radiation absorption of a 0.5 mm thick layer of H_2 or D_2 at 4.2 K is $< 5 \times 10^{-6}$ W/cm², which indicate that frozen films will not adversely effect the total emissivity of cryopanel. They will not raise liquid helium consumption.

II. MIXTURES - TRAPPING RATIOS

It is well known that on a 4.2 K smooth surface, only H_2 and He cannot be pumped down below 10^{-6} Torr beyond small coverages. Yet in fusion reactors these two gases may be quite abundant. We have, therefore, measured the pumping behavior of mixtures of D_2 and H_2 , D_2 and He, and H_2 and He. The results listed in Table I clearly indicate that one cannot use hydrogen isotopes to trap He. For comparison with other

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references, we list our trapping ratios of Ar which was used to trap H_2 and He.

Table 1. Trapping Ratio Q_x/Q_y

y \ x	H_2	D_2	T_2	Ar
H_2	--	9	--	1
He	5×10^4	5×10^4	--	30

In Q_x/Q_y , Q_x is the minimum flux of the trapping gas x, for which no trapped gas, y, diffuses from the composite film after the gas mixture flow has been closed. Q_y is the flux of the gas to be trapped. Tritium produces He^3 due to its β -decay and cannot therefore be used for trapping. It is evident that if helium has to be pumped, an auxiliary He pump will be required.

III. HEAT TO BE REMOVED BY CRYOGENS

In a cryopump most of the heat input due to the thermal radiation and the heat content of the gases present in the system are removed by a 77 K chevron baffle having a high emissivity and high thermal conductivity. High emissivity can be achieved by painting the chevron with a black paint or in the case of Al cheverons by anodizing. In a practical pump where $P \leq 3 \times 10^{-4}$ Torr power dissipation of the chevron (W_B) in watts is governed by thermal radiation and consequently

$$W_B = 5.7 \times 10^{-8} \epsilon_B T_B^4 A_B \text{ accuracy } \pm 15\%$$

where ϵ_B , T_B and A_B are emissivity temperature (K) and area (m^2) of the baffle.

ϵ_B of the anodized baffle was measured to be $0.75 \pm 15\%$ at 77 K. Values up to 0.9 are obtained for black paint.

It is about 20 times more expensive to remove heat at 4.2 K than at 80 K. Much more attention has to be paid to the design of system components operating at liquid helium temperature. The dissipation at ~ 4.2 K is governed not only by the properties of the material used in the construction of the pump, but also by the pressure in the system and the gas pumped. The total heat to be removed at this temperature, W_T , (watts) can be calculated by adding up the contributions from various sources.

1. Pressure independent dissipation W_o (watts). Since the heat conducted along supporting structure of the LHe vessel is generally small, W_o is dominated by the emissivity ϵ of the material used in the pump and the temperature of the baffle

$$W_o \approx 0.1 [A_T - A_B + A_B (T_B/77)^4] \epsilon / 0.03 \text{ (accuracy } \pm 15\%)$$

A_T is the total area of the cryopanel in m^2 .

2. Residual helium pressure dependent dissipation, W_{He}

$$W_{He} = 450 \Delta T A_T P_{He} \text{ (accuracy } \pm 20\%)$$

where P_{He} (Torr) is the residual helium pressure in the pump. ΔT is the temperature difference between the cryopanel and the chevron. From thermodynamical calculations this relation implies an average accommodation coefficient of He gas between 4.2 and 80 K of 0.7.

3. Dissipation with D_2 input, W_{D_2} . For gas fluxes Q_{D_2} up to 50 Torr liter/sec m^2 and $77 < T_B < 90$ K.

$$W_{D_2} = (0.18 \pm 0.02) Q_{D_2}$$

4. Dissipation with H_2 input, W_{H_2} ,

$$W_{H_2} = 0.1 A_T + (0.25 \pm 0.03) Q_{H_2}$$

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where Q_{H_2} is hydrogen gas being pumped in Torr liter/sec.

5. Dissipation with T_2 input, W_{T_2} ,

$$W_{T_2} = (0.29 \pm 0.03) Q_{T_2}$$

Q_{T_2} is T_2 gas flux in Torr liter/sec.

Dissipations from 3, 4 and 5, which were derived experimentally, W , are compared with the thermodynamical calculations of total heat content between 4 and 80 K, H_{tot} , in Table II.

Table II.

Isotopes	H_2	D_2	T_2
H_{tot} (J/mole)	3308.11	3482.68	4074.5
H_{tot} (W/Tl/s)	0.19	0.20	0.24
W (W/Tl/s)	0.25 ± 0.03	0.18 ± 0.02	0.29 ± 0.03

IV. TRITIUM

In this section we summarize the differences in behavior between tritium and the other hydrogen isotopes, which were investigated during the course of our experiments. Our findings, suggest that water should be removed from surface of pumping systems pumping tritium. Compared with D_2 and H_2 , tritium behaves very differently chemically in our stainless steel system. Its reactivity is much higher than one would expect from known noncatalyzed experimental and theoretical data. In order to explain the fast disappearance of T_2 gas in our system at room temperature, we think its surface behaves as a catalyst. Two things have been done to investigate this possibility.

1. The product of surface reaction is detected from its outgassing by quadrupole mass spectrometer. Identification of mass 20 as HTO,

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agrees with Beavis'⁵ finding in his study on D_2 reaction with H_2O on stainless steel walls. The mass 20 of the residual gas in the system will decrease if one flushes the system with H_2 instead of T_2 . This strongly indicates that the isotope exchange is taking place at the surface. The high activity of the surface and its volatility were found after examining the surface during the dismantling of the system. This finding further indicates the high reactivity of our surface.

2. We looked into the rate and the equilibrium constants of hydrogen isotope exchange reaction. The equilibrium constant, K , at room temperature agrees well with Dorfman.⁶ The equilibrium constants are determined from mass spectrometric measurements assuming equal ionization efficiency of various hydrogen isotopes. There is no available K value in low temperature region. All measured K values are listed in Table III.

Table III. Equilibrium Constants

	$H_2 + T_2 \rightarrow 2 HT$	$D_2 + T_2 \rightarrow 2 DT$
Temperature		
	$K = \frac{(HT)^2}{(H_2)(T_2)}$	$K = \frac{(DT)^2}{(D_2)(T_2)}$
Room Temp.	2.5 ± 1	--
$4.2 K < t < 77 K$	$(2.5 \pm 0.5) \times 10^{-2}$	$(1.5 \pm 0.2) \times 10^{-2}$

It should be pointed out that although our K value agrees with Dorfman's,⁶ our reaction rate is much faster. Again, this can be explained by the surface catalytic action.

5. L.C. Beavis, J. Vac. Sci. Technol., 10 (1973), p.386.

6. L.M. Dorfman and H.C. Matraw, J. Phys. Chem., 57 (1953), p.723.

V. TRITIUM CONTAMINATION

Tritium gas, as we find from our study, reacts readily with hydrogen-containing compounds such as water (H_2O) pumping oil, grease (carbon or silicone base hydride) and other compounds present on surfaces. Our Sergeant-Welch 240 l/s turbomolecular pump is lubricated by low vapor pressure oil which is the major source of tritium contamination. Backing pump for turbomolecular pump is also grossly contaminated with tritides.

Physisorbed water on the metallic surfaces of the pump, as well as the surfaces themselves, introduce an additional source of contamination. After tritium is isotopically exchanged with hydrogen in water and/or oil, it has a tendency to diffuse into the bulk. This diffusion mechanism explains the reappearance of tritium contamination after the surfaces have been thoroughly cleaned. This phenomenon is found repeatedly in our experiments, but it is not fully understood. All the findings discussed above will complicate the safety aspects of fusion reactors which will use tritium as fuel. A detail study is needed.

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