A CRYOSTAT FOR THE EXPERIMENTAL OPTIMIZATION OF
VERY LOW PRESSURE CONDENSATION CRYOPUMPING

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Abstract

The observed deviation of the hydrogen saturated vapour pressure (S.V.P.) from the Clausius-Clapeyron law below about 30K, resulting in a temperature independent pressure limitation, might greatly reduce the usefulness of condensation cryopumping. This deviation was found to depend on the characteristics of the experimental apparatus, namely the nature of the condensing surface and of the thermal radiation reaching it. We present here the project and the performance of a cryostat permitting an easy change of the condensing surface and a variation of the quantity and of the spectrum of the radiation load; hence a complete investigation on the physical features of this deviation is made possible.
1. Introduction

In a vacuum system pumped by a cryosurface at a temperature \( T_0 \), the situation can be described by the following equation

\[
P = P^* + \frac{Q}{S} \quad \text{with} \quad 0 < P^* \leq P_{\text{sat}}(T_0)
\]  

(1)

in which \( Q \) is the load of a given gas, pumped with a pumping speed \( S \), and \( P^* \) represents the contribution to the pressure due to desorption from the cryosurface. This contribution, which ideally equals zero for zero coverage, increases with the time and is expected to reach a final value \( P_{\text{sat}} \) corresponding to the saturated vapour pressure of the considered gas at the temperature \( T_0 \). From this moment the surface should condense a practically infinite quantity of this gas without any further pressure increase.

The characteristic parameters of a condensation cryopump would seem therefore to be the temperature of the pumping surface and the S.V.P. curves of the gases to be pumped. In this respect we can observe\(^1\) that in an all-metal ultrahigh vacuum system hydrogen is generally the main component of the residual gas pressure, and it is also the only gas (apart from He, generally absent) presenting an appreciable vapour pressure at 4.2\(^0\)K (boiling temperature of Helium at atmospheric pressure). Nevertheless, extrapolating the available \( \text{H}_2 \) vapour pressure data\(^2,3,4\) (Fig. 1), an equilibrium pressure well below \( 10^{-12} \) torr is expected at a temperature \( (2.5\,\text{K}) \) which can easily be obtained by using liquid helium refrigeration. For this reason cryopumping by condensation has initially been chosen\(^5\) to produce pressures of \( 10^{-11} \) torr or lower in the intersection regions of the Intersecting Storage Rings at CERN.

However, experimental investigations on the pressure of \( \text{H}_2 \) done by Chubb\(^6\) and at CERN\(^7\) have shown (Fig. 2) a systematic departure, at low temperatures, from the Clausius-Clapeyron equation

\[
\log P = A - \frac{B}{T}
\]

\( P = \text{H}_2 \) saturated vapour pressure
\( T = \text{Temperature of the condensed H}_2 \)
\( A, B = \text{Constants.} \)

This deviation, resulting in an unexpected pressure limitation, was found to be strongly dependent on the characteristics of the experimental apparatus, namely:

- precondensing\(^7\) on the stainless steel surface of our "model B" pump (Fig. 3) a layer of a different gas (Ne, Ar or \( \text{O}_2 \)), we have observed a reduction of the measured \( \text{H}_2 \) saturated vapour pressure at 2.5\(^0\)K from \( 2 \times 10^{-11} \) torr on bare
Fig. 1  Saturated vapour pressure data for hydrogen
Fig. 2 Saturated vapour pressure data for hydrogen ($\sim 99\% H_2, \sim 1\% N_2$) from model A and model B cryopumps.
stainless steel) down to the measuring limit of $2 \times 10^{-13}$ torr (by extrapolating from high pressure values a pressure of $5 \times 10^{-15}$ torr is expected at this temperature);

- precondensed layers of HD and D$_2$ gave smaller reduction down to $9 \times 10^{-12}$ and $2.8 \times 10^{-12}$ torr, respectively;

- as was previously suggested$^{6,7}$, it was recently found$^8$ that the thermal radiation coming from the liquid N$_2$ cooled screens or from room temperature regions and loading the condensing surface is effective in desorbing H$_2$ molecules.

The picture emerging is, therefore, one of a complex interaction between the thermal radiation, the condensing surface and the condensed H$_2$.

2. The fundamental features of the new cryostat

To get more information about the physical mechanism of this interaction a new cryostat (model C) was built up offering the following advantages:

a) the possibility of investigating the effect of the thermal radiation by varying the temperature and the efficiency of the cold screens;

b) the possibility of extending the investigation on the influence of the nature of the condensing surface to any stable and bakeable substrate: the condensing surface is easily changeable;

c) "cleanliness" of the experimental information, that is to say:

i) the condensing surface is well defined; the area of the regions at intermediate temperatures is small and the regions where temperature variations during the measurements might occur (this is the case, in particular$^7$), of the walls of the liquid helium container are completely screened off;

ii) the area of the condensing surface is big enough to ensure that the two main disturbing phenomena in the adsorption isotherms measurements (the continuous H$_2$ injection due to the outgassing from the walls and the pumping of gauges) are negligible;

iii) the H$_2$ outgassing from the measuring dome is small, therefore, the Q/S term in Eq. 1) is also small and the P$^*$ term can be measured down to very low pressures.

3. Description of the "Model C" cryostat

The new cryostat is shown in Fig. 4. It consists of two distinct parts connected by a flange (1): thus it is demountable and modifications to the inner part can be made easily.

a) The outside

It is practically a liquid nitrogen container (2) insulated by a rough vacuum (3) independent of the experimental vacuum in the cryostat. The main contri-
Fig. 3  Model B cryopump

Fig. 4  Model C cryostat and cold He gas transfer line
b) The inside and the transfer line

A liquid He container (10) of 6 litres capacity is connected by a tube 10 mm in diameter (11) to a smaller lenticular vessel (12) giving a condensing surface of \(-700 \text{ cm}^2\). The container (10) is enclosed at the top by a liquid \(N_2\) container (17), at the bottom by a bellow (18) welded on the tube (11) and all around by the cylinder (19).

This solution presents the following advantages:

i) the condensing surface is well defined (see 2c, point 1)) and can be easily changed (see 2a)) by cutting the tube (11) just above the condensing surface (12);

ii) the thermal conduction to the liquid He is reduced by the bellow (18), which also supplies the elasticity needed to compensate differential contractions of the two connected parts;

iii) any trouble connected with changes of temperature on the He container walls (see 2c, point 1)) is removed;

iv) the danger represented by leakages inside the volume (13) is reduced.

The spiral (20) welded on the lower part of the cylinder (19) permits one to regulate the temperature (see 2b) of the inner screen (9), above \(77^\circ\text{K}\) by circulating warm \(N_2\) gas and below \(77^\circ\text{K}\) (down to \(10^\circ\text{K}\) if necessary) by circulating cold He gas. The temperature of the enclosure surrounding the surface (12) can be measured by means of the thermocouples (in 15 and 21) and of a vapour pressure thermometer (in 16). The transfer of cold He gas from a normal liquid He storage dewar is done by means of the transfer line also shown in Fig. 4. The cold gas is transmitted through the central tube (22) and, circulating in the spiral (20), cools down the screen (9). Coming back to the transfer line (where it cools a radiation screen (23) before being pumped through the flange (24)) it reduces the heat leaks into the liquid He container by reducing the temperature of the tube (25) and of the
screen (26). The insulation vacuum in the transfer line is obtained by pumping through the flange (28). The connection between the transfer line and the cryostat is made by the double-joint flange (27), shown in details in Fig. 5. The inner gasket (1) which has a working temperature lower than 77\(^0\)K is indium; the outer one (2), always at room temperature, is viton.

4. Performances and results

The results obtained during about one year of experimental work have been partially published\(^9\) or will be published in the near future; they will be mentioned here only as far as the performances of the cryostat are concerned.

a) The change of the radiation load on the condensing surface

i) The radiation from 300\(^0\)K

Combining differently various screens in 6) and 9) the S.V.P. of H\(_2\) condensed on stainless steel was measured under 300\(^0\)K radiation loads in the range 2.7 to 1.5 x 10\(^{-3}\) mW cm\(^{-2}\). These figures correspond to radiation absorbed on the pumping surface, and are estimated by subtracting from the measured He boil-off rates the contributions coming from heat influx onto the upper container 10) and onto the pumping surface itself but due to 77\(^0\)K radiation. Both these two contributions to the consumption have been measured together by putting in 9) a completely closed screen at 77\(^0\)K. In order to increase the sensitivity when very small consumptions are involved, the efficiencies of the screens 6) and 9) are estimated from separate measurements by this method and the efficiency of the combination calculated as the product of the two.

ii) The change of the temperature of the screen

By circulating cold helium gas as previously described (see 3b)) the temperature of the inner screen 9) can be stabilized at any desired value between 13 and 77\(^0\)K. In practice, however, only temperatures higher than 50\(^0\)K are interesting, because below this temperature the H\(_2\) adsorption desorption phenomena on the screen are too important. Above about 50\(^0\)K the H\(_2\) S.V.P. measurements are reproducible, i.e. independent of the temperature of the screen during the H\(_2\) injection and of the direction from which the chosen screen temperature is approached.

Any change of cold helium gas flow in 20) produces a non-equilibrium situation concerning either the temperature distribution or the H\(_2\) coverage of the screen (and therefore the H\(_2\) pressure in the system). A new equilibrium is reached after a time which is longer for lower temperatures and bigger temperature variations. About 2 hours are needed, for example, to get 50 \(\pm 1\)\(^0\)K on the thermocouples 15) and 21) starting from 77\(^0\)K and
Fig. 5  The connection flange between the model C cryostat and the cold He gas transfer line
about 2 hours more for the corresponding pressure stabilization; but a complete equilibrium is obtained more quickly when stabilizing the temperature above 700 K: a variation from 77 to 200 ± 50 K is possible in 3-4 hours, but the pressure equilibrium follows immediately.

While in the case of 3000 K radiation the absorbed radiation is known by consumption measurements, this method does not apply to the present situation because the consumption from the upper container 10) is also changing continuously; for this reason (and also in order to reduce the consumption, i.e. to increase the life-time of an experiment) for measurements between 200 and 3000 K the inner screen 9) was removed and the temperature of the outer screen 2) was varied. A discrete temperature regulation of this screen is easily obtained filling the N₂ container with warmer refrigerant fluids.

As result of these measurements the correlation between the H₂ S.V.P. and the temperature of the screen surrounding the condensing surface (or between the H₂ S.V.P. and the radiation absorbed) was obtained⁹); a physical model for the desorption mechanism was also proposed⁹).

For the practical applications it is important to remark that the upper limit of the S.V.P. of H₂ condensed on a stainless steel (for example) surface surrounded by a screen at 770 K, permitting no access of 3000 K radiation, was of about 3 x 10⁻¹³ (N₂ equivalent, pressure in the room temperature working chamber) This means that, usually, the observed deviation from the Clausius-Clapeyron law is due to room temperature radiation. Very efficient 770 K screens are needed in order to keep this deviation small; for example, in order to get a H₂ S.V.P. lower than 1 x 10⁻¹² a screening efficiency better than 1/1000 is required. Obviously this introduces a decrease of pumping speed per cm² of pumping surface and consequently a corresponding increase of the size of the cryopumps. For this reason the optimization of the screening efficiency is extremely important, and an investigation programme in this direction is in progress.

b) The change of the condensing surfaces

As we had suspected on the basis of the important change introduced by a precondensed layer of a different gas⁷), the desorption efficiency of H₂ by thermal radiation was found to be strongly dependent on the nature of the condensing surface⁹).

Up to now, ten different stable and bakeable surfaces have been tested; the "model C", cryostat permits one to investigate a surface per week. In a standard test the H₂ S.V.P. on a given surface is measured in presence of two different 3000 K radiation loads. The inner screen 9) is removed and the outer screen 2) is either at 3000 K or at 770 K. In this second case the 3000 K radiation load is reduced to 1/25 of the value corresponding to full exposure of room temperature radiation.
Although a few containers were made in different materials (Cu, Be, Pyrex) in general stainless steel containers coated by the material to be tested are used. The coating has been made either electrolytically (Pb, Ag, Au, Chromium black, average thickness about 10 μ) or by vacuum deposition (CsI, MgF₂, average thickness about 1 μ). A 10² fold variation⁹ in the H₂ S.V.P. between various substrates under the same radiation load was observed.

In a few cases very low consumptions of liquid helium were obtained (for situations where the temperature of the screens was 77⁰K); for example, when a silver coated container was mounted (screening radiation efficiency 5/10⁴), the evaporation rate was of only 20cc of liquid helium per hour; the corresponding pumping speed for H₂ about 700 1. sec⁻¹.

c) Measurements of H₂ adsorption isotherms

i) Residual pressures

When liquid helium is poured into the cryostat, the H₂ pressure falls from about 2 x 10⁻¹⁰ to a few 10⁻¹³ torr, the latter value depending on the choice of the screening system 6) (and 9).

This measured H₂ pressure is due to a combination of P* and Q/S (see formula 1; in this case Q is the total H₂ flow degassing from the measuring dome) plus the limit pressure of the measuring instrument. Since P* depends, for a given coverage and temperature, only on the thermal radiation loading the pumping surface, Q/S only on the pumping speed to the measuring dome, and the limit pressure of the gauge only on the gauge parameters, then by changing independently the radiation load and S a complete evaluation of the different contributions was obtained¹⁰). Measurements in the 10⁻¹³ torr range were usually done by means of an Omegatron mass spectrometer calibrated for absolute pressures and linearity (for H₂) against the H₂ saturated vapour isostere⁷).

ii) Pressures after H₂ injections

In this case the range of pressures to be measured goes from 10⁻¹³ up to 10⁻⁶ torr or higher depending on the degree of coverage and on the temperature of the condensing surface; furthermore, H₂ quantities must be evaluated. The different possible sources of error to be considered, concerning therefore both the quantity and the pressure measurements, are:

- the continuous H₂ injection due to H₂ degassing;
  - about 5 x 10⁻⁹ torr 1 sec⁻¹ of H₂ degas from the walls of the vacuum system; since the monolayer quantity is in the present case of the order of 10⁻¹ torr 1, and usually quantities of at least the order of 10⁻² torr 1 are injected, it follows that even for stabilization times of about 10 h only quantities of the order of 1/100 of the injected quantities are involved.
- high pressure pumping of the gauges;

four pressure gauges are normally used during the experiment, having a total pumping speed for \( H_2 \), measured in the \( 10^{-7} \) torr pressure range, of \( -10^{-4} \) sec\(^{-1} \). This means that for pressures in the \( 10^{-6} \) torr range, and equilibrium times of a few hours, quantities of the order of 1/10 of the injected quantities might be pumped. Fortunately the higher the pressures, the shorter are the equilibrium times (of the order of \( 10' \) in \( 10^{-6} \) torr pressure range) and only one gauge need be used in this situation.

- contamination of the system during \( H_2 \) injections;

since coverages up to \(-100\) monolayers are investigated (corresponding to a few torr 1 of \( H_2 \) injected) the vacuum system is exposed to relatively high pressures during the injection. On the measuring dome (this is the lowest pressure point of the system during the injection; pressures 1000 times higher are measured on the injection dome) a \( H_2 \) pressure of \( 6 \times 10^{-8} \) torr over 2 hours may be maintained. After injection, pressures in the \( 10^{-13} \) torr range should often be measured, and the pressure due to the adsorption-desorption equilibrium on the pumping surface must be distinguished from memory effects of the system and of the measuring gauges. This is generally possible, in the model C cryostat, by profiting from the radiation dependence of the equilibrium \( H_2 \) pressure\(^10\).

Conclusions

By means of the model C cryostat the effect of the thermal radiation on the S.V.P. of \( H_2 \) condensed on different surfaces has been studied varying the efficiency and the temperature of the radiation screens.

A model of the physical mechanism of the description has been proposed.

In spite of the observed deviation from the Clausius Clapeyron law of the \( H_2 \) S.V.P. cryopumping by condensation has been proved to be an interesting means to get high pumping speeds at very low pressures. Consequently an intersection region of the ISR at CERN will be cryopumped in order to reach pressures down to \( 10^{-12} \) torr.

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