A fast cryopump system for ultra-high vacuum

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Now that pumps which have a high pumping speed at low pressure have been developed it is easier to pump down to ultra-high vacuum, i.e. pressures of the order of $10^{-9}$ torr. However, the times in which such pressures can be attained — which do not only depend upon pumping speed, but on other factors as well — still leave something to be desired. In the article below a system is described in which, by the appropriate use of low temperature, the pumping time is considerably shortened.

Introduction

In the last ten years the art of achieving ultra-high vacuum, that is to say, pressures of less than $10^{-9}$ torr, has evolved from a complicated and time-consuming procedure to an established technique. The statement still holds even if the vacuum vessel is extremely large. A typical example of such a case is to be found in experiments designed to simulate the conditions prevailing in outer space. Apart from the classical method of pumping, using a diffusion pump, still widely employed, other methods have gained ground in which use is made of a getter ion pump, or of a cryopump [1].

The chief advantage of these pumps is that they cannot contaminate the space being evacuated, as they do not use a pump fluid. This advance has not however been associated with an appreciable shortening of the time in which the required vacuum is reached. The traditional method of reducing the pumping time is to bake out the system, but this results in a pumping time no shorter than a few hours. For experiments or processes of short duration which have to take place in a vacuum, a much greater reduction of the pumping time would therefore be very welcome. We have found that the pumping time can in fact be substantially reduced by the appropriate use of low temperature.

Generally speaking, lowering the temperature of part of the wall of a vacuum vessel has three effects, each of which causes a drop in pressure. The first is trivial: as the wall becomes colder the temperature of the gas drops and so too does the pressure (Charles's Law). The second effect is connected with the desorption, during evacuation, of gas which is adsorbed on the wall of the vacuum vessel and absorbed in the wall itself. Reduction of the temperature causes a considerable decrease in the rate of desorption of this gas. The pressure in the last phase of the pumping process, called the end pressure, is determined to a great extent by the competition between this desorption of gas and the performance of the pump. Lowering of the desorption rate and of the diffusion rate therefore leads to a lowering of the end pressure. The third and last effect is found only in gases whose partial pressure is higher than the saturation pressure corresponding to the temperature of the cooled part of the wall. A gas of this kind partly condenses on the cold wall, and its partial pressure then approaches the saturation pressure. The latter process, removal of the gas by condensation, is known as cryopumping.

The operation of the system described in this article [2], which can be used to achieve an ultra-high vacuum in a short time, is based on a combination of cryopumping and reduction of the rate of desorption. The Charles's Law temperature effect is also present, of course, but, as in other systems, it plays a relatively minor part. The required refrigeration is produced by a Philips two-stage gas-refrigerating machine [3], both

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stages of which are used — the first for reducing the desorption rate and the second for cryopumping. The walls to be cooled are in direct mechanical contact with the machine, so that no gas or fluid is necessary for the cold transport. Before describing the system we shall briefly discuss the two temperature effects underlying its operation.

Since low-temperature pumping calls for relatively expensive machines, it will not be the most economic method in all cases. It is undoubtedly an economic proposition for large systems, but for small systems it is only economic in certain cases. For example, a system of the type described in this article is attractive for short term experiments and processes, i.e. cases in which the pumping time figures largely in the costs.

Finally, it should be noted that circumstances may arise where an ultra-high vacuum can be attained quickly only by pumping simultaneously with pumps of different types. We shall not discuss situations of this kind here. Our purpose in the following is simply to show the contribution which the carefully considered use of low temperature can make to shortening the pumping time.

The two fundamental processes

Adsorption and desorption of residual gas

After pumping for some time, a point is reached where the pressure in a vacuum system begins to drop very slowly. The evacuated space then contains a gas mixture, the "residual gas", whose composition generally differs very considerably from that at the beginning of the pumping process, and whose pressure depends on a variety of processes. Chief among these processes is the release of gas adsorbed on the wall, but, as we indicated above, gas in solution inside the walls is also desorbed. Since different gases are adsorbed on the wall, whose desorption depends in different ways on temperature and pressure, the composition of the gas gradually changes during evacuation. If the system is baked out in a furnace, or a filament is heated to incandescence in the evacuated space, the composition may in addition be changed by interactions between the gas and the hot walls or the hot filament, resulting in chemical reactions.

In recent years investigations using mass spectrometers have yielded a great deal of information on the composition of the residual gas, but it is still not possible to give a quantitative prediction of how the total pressure in a particular pumping process will vary with time, particularly when dealing with high or ultra-high vacuum. This is easily understood if one considers that in a sphere of 1 dm³ in which the gas pressure is 10⁻³ torr, there are just as many molecules on the wall as inside the sphere when the wall is covered with only a monomolecular layer. If the pressure in the sphere is reduced to 10⁻¹⁰ torr the ratio between the number of adsorbed and the number of free molecules is no less than 10⁶ to 1. Obviously, therefore, the gas desorption from the wall has a decisive bearing on the course of the pumping process.

The rate of desorption, i.e. the number of molecules leaving the surface per second and per cm², is given by the expression

\[ dN/dt = N/\tau, \quad \ldots \ldots (1) \]

where \( N \) is the number of molecules per cm² of surface and \( \tau \) is the average time a molecule remains on the surface. This time is given by:

\[ \tau = \tau_0 \exp (E_d/RT). \quad \ldots \ldots (2) \]

In equation (2) \( \tau_0 \) is a constant, \( E_d \) is the desorption energy, \( R \) the gas constant and \( T \) the absolute temperature.

It is clear from (1) and (2) that a change in the temperature \( T \) of the wall from which the gas is desorbed has a marked effect on the rate of desorption. Cooling reduces the desorption rate considerably and thus results in a much lower end pressure.

The speed at which the gases dissolved in the wall reach the surface also decreases considerably as the temperature falls; the reciprocal of the diffusion constant varies with \( T \) in much the same way as \( \tau \).

By cooling the walls of a vacuum vessel and everything contained inside it to the temperature of liquid nitrogen, it is therefore possible in principle to produce an ultra-high vacuum without the need for any baking out [4].

Of course, in many vacuum systems it is not possible to make all the surfaces cold. The difficulties due to this differ from one case to another, and the right answer has to be found for each individual case. If, for example, a layer has to be deposited on a substrate by vacuum evaporation, the source has to be heated and a great deal of gas is released. The obvious answer here is a drastic increase in the pumping speed, i.e. the ratio of the quantity of gas pumped out per second and the pressure. Here too, as we shall now see, the use of low temperature can be very useful.

Condensation of gas; cryopumping

The saturation pressure of any substance depends to a great extent upon temperature. Fig. 1 shows the variation with temperature of the saturation pressure of various gases frequently found in vacuum systems. It can be seen that cooling to the temperature of liquid nitrogen (77 °K) is more than sufficient to reduce the vapour pressure of water to a negligible value, but that
at this temperature pressures in the ultra-high vacuum range cannot be reached if gases such as carbon monoxide, carbon dioxide or methane are present. Broadly speaking, such pressures can be reached when cooling to the temperature of liquid hydrogen (20 °K); at this temperature only hydrogen, helium and neon still have too high a pressure. For hydrogen the temperature of liquid helium (4 °K) is still too high; at this temperature hydrogen has a vapour pressure of about $10^{-7}$ torr. (If these gases are present in a quantity sufficient to upset the experiments, cryopumping must be combined with some other method of evacuation.)

If the pressure of a gas at a certain temperature is equal to the saturation pressure at that temperature, then the number of evaporating molecules is equal to the number of condensing molecules. Since we are concerned here with very low pressures, and the mean free path of the molecules is much larger than the dimension of the vacuum system, the temperature of the gas is entirely determined by the temperature of the walls and of the objects contained in the vacuum system. If now the temperature of part of the surface is lowered, so that the number of molecules evaporating from it is reduced, gas will condense there continuously; the part of the wall whose temperature has been reduced then operates as a cryopump.

The pumping speed of such a pump and the lowest attainable pressure can easily be calculated as follows. The number of molecules $\nu_1$ colliding per cm$^2$ and per second with the wall — and hence with the part whose temperature has been reduced — is given from the kinetic theory of gases, by:

$$\nu_1 = 3.513 \times 10^{22} \frac{p_1(MT_1)^{1/2}}{T_1^{3/2}}.$$  \hspace{1cm} (3)

Here $p_1$ is the gas pressure in torr, $M$ the molecular weight and $T_1$ the temperature of the gas, i.e. the temperature of the walls apart from the area at reduced temperature. Not every molecule colliding with this part of the surface condenses, however, although there is a great probability of this happening if the molecule is not travelling exceptionally fast. Let $\alpha_{12}\nu_1$ be the number of condensing molecules per cm$^2$ and per second; the factor $\alpha_{12}$, the fraction that condenses, we shall call the condensation coefficient of a molecule of temperature $T_1$ which comes into contact with a surface of temperature $T_2$. The number of molecules evaporating per cm$^2$ and per second from the part reduced in temperature is equal to $\alpha_{22}\nu_2$. Here $\alpha_{22}$ is the condensation coefficient for molecules of temperature $T_2$ that collide with a surface having the same temperature. The number $\nu_2$ is found from (3) by substituting for $p_1$ and $T_1$ in that expression the values of $p_2$ and $T_2$; $p_2$ is the saturation pressure at the temperature $T_2$. For every cm$^2$ of the surface of the cold wall, therefore, there disappear per second from the evacuated space a number of molecules

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equal to \(a_{12}p_1 - a_{22}p_2\). If this number is converted into practical units, the pumping speed \(S\) per cm\(^2\) of cold surface is found to be:

\[
S = 3.64a_{12} \left( \frac{T_1}{M} \right)^{1/4} \left( 1 - \frac{a_{22}}{a_{12}} \frac{p_2}{p_1} \left( \frac{T_2}{T_1} \right)^{-1/12} \right) \quad (4a)
\]

The lowest pressure attainable is the pressure at which \(S\) has decreased to zero. This value of \(p_1\), usually denoted by \(p_{eq}\), is given by:

\[
p_{eq} = \frac{a_{22}}{a_{12}} p_2 \left( \frac{T_2}{T_1} \right)^{-1/12} \quad (5)
\]

As can be seen, \(p_{eq}\) is not equal to the saturation pressure \(p_2\) that corresponds to the temperature \(T_2\) of the pumping plate. This is because only part of the wall has the temperature \(T_2\).

With the aid of equation (5) the equation for the pumping speed can be reduced to:

\[
S = S_0 \left( 1 - \frac{p_{eq}}{p_1} \right) \quad (4b)
\]

where \(S_0 = 3.64a_{12}(T_1/M)^{-1/12}\). This expression (see fig. 2) shows rather more clearly than (4a) the way in which the pumping speed drops to zero during evacuation in a particular case (i.e. at a given \(S_0\)). The advantage offered by cryopumping in terms of gas kinetics also becomes clear: if the "temperature" of the molecules is not too far above that of the pumping surface, then \(a_{12}\) is roughly unity, which is 4 to 5 times greater than the constant, corresponding to \(a\), occurring in the expression for the pumping speed of a well-designed diffusion pump.

A vacuum system with a two-stage gas-refrigerating machine

The heart of our vacuum system for rapid production of ultra-high vacuum is the two-stage gas-refrigerating machine mentioned earlier. In the design of the system good use was made of the ability of this machine to supply cold at two temperatures, the cold production at the lowest temperature (about 12 °K) being only slightly influenced by the cold production at the other temperature (50 °K to 80 °K).

A diagram of the whole system is shown in fig. 3. The space to be evacuated is contained inside the copper housing \(W_1\), mounted on the freezer \(F_1\) of the first stage. The head of the machine, with freezer \(F_1\), is inside this housing. Fitted to the top of \(F_1\) is a copper plate \(C\) with a surface area of about 80 cm\(^2\), which functions as the cryopump. To prevent the condensation of atmospheric vapours and gases on the outside wall of the vacuum chamber — this would give a loss of cold and could cause short-circuiting between electrical leads through the wall — the whole system is contained inside a much larger vacuum vessel (wall \(W_2\)) in which a pressure of \(10^{-8}\) to \(10^{-6}\) torr prevails. This pressure can easily be obtained by means of a rotary oil pump.

Since there are advantages in putting the cryopump into operation only when the pressure is already fairly low, the vacuum chamber can be connected by a pipe \(P\) to another type of pump, for example a rotary pump or a combination of a sorption pump using zeolite \(\text{[5]}\) and a diffusion pump, or a getter ion pump. In addition, the wall \(W_1\) is fitted with a number of flanges (not shown in the diagram) which can be used to connect an ionization gauge for measuring the total pressure, to connect instruments for measuring the partial pressures, for feed-through of electrical leads, etc.

Since, after completion of an experiment, a vacuum vessel of this kind can be opened only when the temperature has again become roughly equal to room temperature, the outside of the wall \(W_1\) is fitted with an electric heating element to raise the temperature quickly. This element can, if necessary, also be used for baking out the wall to some extent before or during the first phase of the pumping process. Another method of heating, which is undoubtedly attractive, is to let the gas-refrigerating machine itself produce the heat. In principle, this only requires the direction of rotation to be reversed. With the machines at present on the market, however, this would involve some slight modifications.

Experience gained with the system

The experience gained with the system has completely fulfilled our expectations. Starting from atmospheric pressure, a vacuum of less than \(10^{-6}\) torr can be reached within about an hour. This result will not, of course, be achieved easily in all circumstances. It is necessary to take account of the particular characteristics of the instruments arranged inside the vacuum space and of
the pressure gauges connected to it, and suitable measures must be taken to offset any adverse effects they may have. To some extent, these are precautions which have to be taken in any ultra-high vacuum system, but they are more important here owing to the shortening of the time scale. These precautions are also applied because the gas cannot be completely evacuated from a gauge. After out-gassing the pressure again dropped quickly, and in a good hour a pressure as low as $10^{-10}$ torr was reached.

Another feature of the system came to light in experiments designed to simulate the gas yield due to desorption from the wall; this was done by admitting a gentle stream of nitrogen into the vacuum space while

system like the one illustrated in fig. 3, some gas always remaining on the wall.

One of the most troublesome components of the system itself is the ionization gauge. Its hot filament gives off gas and moreover heats the wall, giving rise to some unwanted desorption. This makes it necessary to out-gas the gauge and everything near it. The importance of this can be clearly understood from fig. 4. In the experiment which gave these results the refrigerating machine was connected in after a pressure of $7 \times 10^{-5}$ torr had been reached with an auxiliary pump. The pressure then dropped within half an hour to about $10^{-8}$ torr, but from then on the pressure dropped very slowly because of the release of gas from the ion pumping. The end pressure established in these experiments was $10^{-6}$ to $10^{-7}$ torr. When the supply of nitrogen was stopped, the pressure dropped much more slowly than one would have expected. The reason proved to be that the admitted nitrogen was quickly adsorbed on the wall and was gradually desorbed when the supply was stopped; the temperature of the wall in our equipment (about 55 °K) is apparently not low enough

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Zeolites are Al-Mg silicates which, upon heating, lose their water of crystallization without any change taking place in their crystal structure, giving a molecular-porous material. This material has an extremely large internal surface area and can bind a great deal of gas. Since this takes place both by adsorption and absorption, the term sorption pumps is used, which also distinguishes these pumps from getters which trap gases chemically.
to bind nitrogen drastically and not high enough for rapid desorption of nitrogen. This is the very same effect which is found with water vapour in systems at room temperature. This difficulty can easily be overcome by increasing the temperature of the wall by 5 to 10 °K. The nitrogen is then released more quickly. This sends the pressure up for a little while, but after this it falls rapidly.

We may note in passing that the refrigerating capacity of the machine at, say, 20 °K, is generally much greater than the heat of condensation from the pump plate plus the energy of the incident radiation. We might, therefore, have made the pump plate and hence the pumping speed much larger. For our purpose however, this was not necessary. In fact, in systems for carrying out certain heat-evolving processes under vacuum a large pump plate can be undesirable. It is better then to keep a fair amount of refrigerating capacity "in reserve".

If required, it is possible, using a two-stage gas-refrigerating machine, to build an ultra-high vacuum system of an entirely different type, leaving the walls of the vacuum chamber at room temperature. A diagram of the cross-section of such a system is shown in fig. 6. The cryopumping surface is surrounded on all sides by a screen of the type illustrated in fig. 5, which is fitted to the first stage of the machine. In a system of this kind a substantially higher pumping speed can be reached than is possible with the pumping methods used hitherto. Very large vacuum systems using a cryopump are nearly always designed on the lines illustrated in fig. 6; the screen is usually cooled with liquid nitrogen, and the pumping surface with liquid hydrogen or helium.

When an evaporation process is carried out in the vacuum chamber a considerable amount of heat is generated which reaches the wall as radiation. How does this affect the temperature of the surface acting as the cryopump? The situation is relatively favourable, since the refrigerating capacity of the gas-refrigerating machine is already fairly high at temperatures only just above the minimum temperature (as much as 85 W at 20 °K). Even so, it is more advantageous to dissipate the entire radiant energy through the first stage of the machine. For if the pumping surface becomes a few degrees warmer, this can have a fairly considerable effect on the pressure if the residual gas has a component whose partial pressure is close to the saturation pressure. A slight increase of temperature in the first stage of the machine, on the other hand, has hardly any effect on the temperature of the pumping surface and cannot therefore seriously affect the pumping speed. The pumping surface can be very effectively screened by means of a grid connected to the wall of the vacuum chamber and directly connected to the first stage of the machine. A grid of this kind, with a cross-section like that shown in fig. 5, lets gas through readily but not the radiation. It does, of course, reduce the pumping speed slightly.

Arrangement for measuring condensation coefficients

To conclude, as an example of a special application of our vacuum system, we shall briefly describe an arrangement for measuring condensation coefficients (fig. 7). The arrangement consists basically of two vacuum chambers I and II, interconnected by means of a circular hole of known diameter (conductivity F). Immediately above the plate C, which operates as the cryopump, there is a diaphragm, and this means that the area of the pumping surface is also exactly known. In the equilibrium state the quantity of gas flowing per
second from II to I is equal to the quantity condensed per unit time on the pumping plate C. Expressed as an equation:

\[(p_{II} - p_I)F = p_I S; \quad \ldots \quad (6a)\]

hence:

\[S = F \{(p_{II}/p_I) - 1\}. \quad \ldots \quad (6b)\]

Summary. Now that pumps are available which have a high pumping speed at low pressure, it is no longer particularly difficult to produce an ultra-high vacuum \(\leq 10^{-9}\) torr. The pumping time can be drastically shortened by cooling the wall of the system to about 50 °K. This minimizes the rate of gas desorption. The Philips two-stage gas-refrigerating machine is particularly useful in such a system. The wall is then mounted on the first stage. The head of the machine (at about 12 °K) is inside the vacuum and on top of it there is a copper plate which acts as a cryopump, i.e. it lowers the partial pressure of the gas components to a value slightly above their saturation pressure at 12 °K. With an arrangement of this kind \(10^{-9}\) torr can be reached in one hour. If the system contains an internal heating source, as in a vacuum-evaporation equipment, a grid is arranged in front of the plate to shield it from radiation: the heat received by the grid is taken off through the first stage of the machine.

Using equation (6b) the pumping speed can be directly derived from the ratio of the pressures \(p_{II}\) and \(p_I\). From the value of \(S\) then found the condensation coefficient can be calculated directly. In this way we found a coefficient of 0.98 for the condensation of nitrogen at 60 °K on a surface at 18 °K.