

Cryogen Free Refrigeration to 270mK by Adiabatic Expansion of He³ Gas

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Using the well-known phenomena of adiabatic expansion of gas and adsorption pumping, two new types of He³ refrigerator have been built at OIS. The first type operates in a liquid helium environment at 4.2K (so called “wet”). The second design is cryogen free and uses a Pulse Tube Cryocooler (“dry”). This ULT system is embodied in a commercial He³ Cryofree refrigerator Heliox AC-V, developed by Oxford Instruments. This system uses single-phase electrical power, does not require a water cooler and can be turned key in operation. Cooling performance and other features and instrumentation services will be discussed.

A lot of fascinating and intriguing phenomena in Physics are unravelled at ultra low temperatures (ULT). That is why ULT refrigerators are still in high demand. The first stage of ULT is temperatures below 1K, which can be achieved by the so-called He³ refrigerator. For many decades typical He³ refrigerators have comprised two refrigerators in one system. The first is a He⁴ refrigerator (so called 1K pot) for condensing He³ gas into liquid. The second is the He³ refrigerator itself, which produces cooling to a temperature below 300mK by pumping vapour above liquid He³ typically using a cryogenic adsorption pump [1].

An essential attribute for any adsorption He³ system is the 1K pot. This device produces the low temperature required for condensation of He³ gas. Usually this refrigerator reaches ~1.5K and the He³ liquefaction fraction is greater than 90% of the initial gas content. Starting He³ vapour pumping at such a low temperature means that very little He³ liquid needs to be evaporated to achieve base temperature. This leads to 2-3 days hold time with a residual heat leak of order 10 microWatt to the He³ pot – the coldest part of refrigerator. This type of the He³ refrigerator is reasonably simple to construct and operate, provides good temperature stability and is easy to control. Commercial He³ systems are usually automated [1]. The only complication is the necessity for a second refrigerator with liquid He⁴ with its needle valve or impedance and all pumping accessories, including a rotary pump, pumping line, valves etc. This complication makes the system expensive, where the main cost contribution is the additional He⁴ refrigerator.

However it has been shown that the He⁴ refrigerator is not required and can be avoided by using the procedure of adiabatic expansion of He³ gas. A suitable “expander” can be made using an adsorption pump at variable temperature [2]. This new method of He³ gas liquefaction reduces the cost of the whole system.

ADIABATIC (ISENTROPIC) EXPANSION OF HE³ GAS AS A SOLUTION FOR THE REPLACEMENT OF THE 1K POT

The adiabatic expansion of gases is a well known phenomenon and method of cooling gases. The majority of industrial liquefiers utilise adiabatic expansion for cooling and liquefaction of air, hydrogen and noble

gases including He⁴. Current commonly used cryogenic expansion machines are reciprocating piston or turbine expansion devices.

Only He³ has been excluded from this process. This has happened for obvious reasons. Expansion machines are expensive and complicated mechanical devices with a significant volume of operating gas. The 1K pot is simpler, smaller and cheaper than standard expansion devices and the quantity of He³ gas is very limited so the 1K pot arrangement is commonly used.

From the end of 1995 we were developing novel He³ systems at Oxford Instruments and an adiabatic method was under consideration from the beginning. To avoid a complicated expansion machine an attempt was made to cool He³ by expanding it into a reservoir. The cooling effect was seen straight away and some liquefaction of He³ was observed. Later we found an article by Troitskii and Fradkov, who observed this effect and used it for operating a He³ refrigerator [2]. Their result was very modest when compared with commercial He³ systems. The lowest temperature obtained was ~0.36K and could be maintained for only 9 hours. This may explain why the method of adiabatic expansion of He³ gas was not recognised until now.

The challenge was to improve the adiabatic expansion method to achieve the specification required for a commercial product. The results of this work are reported below.

Let us to remind ourselves briefly of the main, simple thermodynamic equations for gas adiabatic expansion and its implementation for He³.

As it is well known for an ideal gas one can write

$$PV = mRT \quad (1)$$

Where P is the pressure, V is the volume, R is the gas constant and m is the mass of the gas at the temperature T . The reversible adiabatic (isentropic) process means that entropy of the system is constant, hence

$$\Delta S = c_v \ln\left(\frac{T_2}{T_1}\right) + R \ln\left(\frac{V_2}{V_1}\right) = 0 \quad (2)$$

The relationships between thermodynamic parameters of the system are defined as

$$\left(\frac{T_2}{T_1}\right) = \left(\frac{P_2}{P_1}\right)^{(\gamma-1)/\gamma}; \quad \left(\frac{T_2}{T_1}\right) \left(\frac{V_2}{V_1}\right)^{R/c_v} = 1 \quad (3)$$

He³ is a monoatomic gas, hence $\gamma=5/3$

When the pressure of He³ gas drops from P_1 to P_2 the final temperature T_2 is defined by the expression

$$T_2 = T_1 \left(\frac{P_2}{P_1}\right)^{0.4} \quad (4)$$

For quantitative evaluation of (4) let's consider a very modest set of conditions:

- initial temperature of He^3 gas $T_1 = 4.2\text{K}$
- the pressure of the gas drops adiabatically from 3 Bar to 1 Bar.

According to (4) T_2 should decrease from 4.2 K to 2.7K, which means it has dropped below the boiling point of 3.2K for 1 bar. This means that liquefaction should take place under these conditions. This effect is easy to demonstrate on the Entropy -Temperature diagram, obtained by Daunt [3] and shown on Fig.1

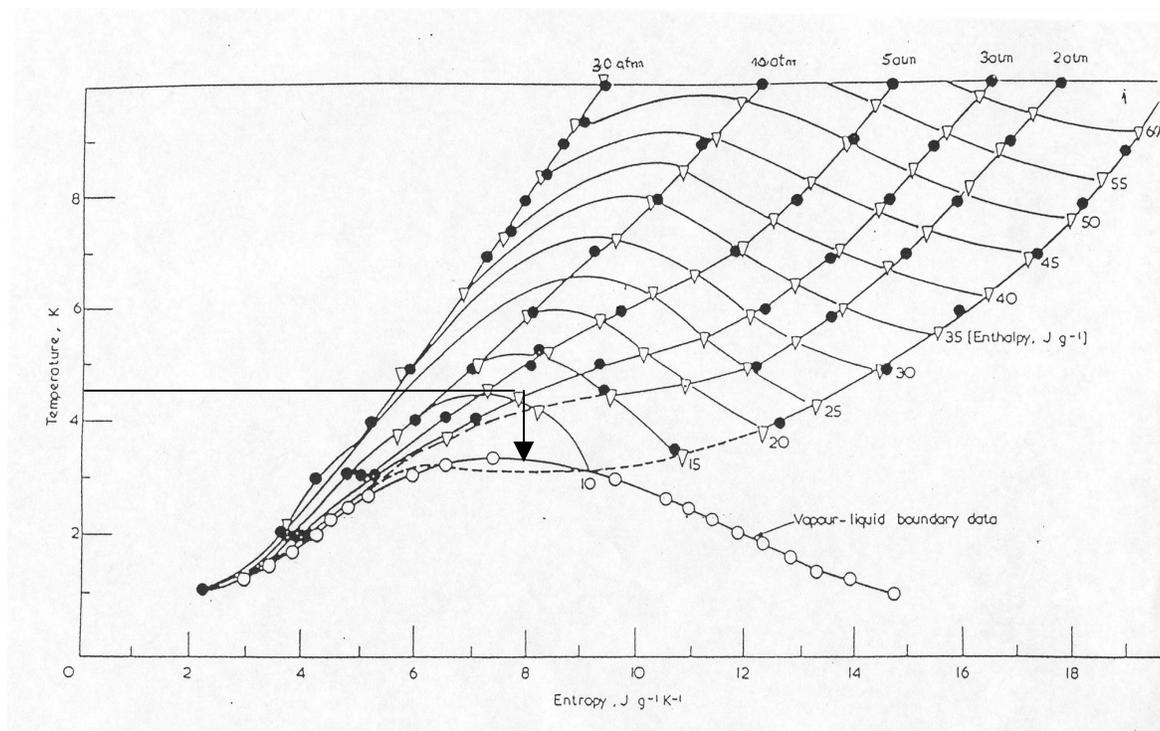


Fig 1. Isobars and isenthalps on the temperature, T, versus entropy, S, chart for He^3

The intersection of the horizontal line at the temperature 4.2K with the 3 bar Isobar (3 atm on the picture) corresponds to the initial state of the He^3 gas. The vertical line down from 3 bar isobar to 1 isobar describes the adiabatic expansion process and the intersection of this line with the vapour-liquid curve corresponds to He^3 liquefaction. In fact this process will start slightly earlier at the critical point for He^3 which is 3.3K. The liquefaction process continues until the pressure of the He^3 gas is above the saturated vapour pressure of the liquid.

The liquid fraction achieved is a key parameter and strongly depends on the initial gas conditions of pressure and temperature. It is important to note that to have a significant high pressure of He^3 at 4.2K in the sealed volume one would need a very high gas pressure at room temperature.

There are advantages to expanding into an additional reservoir that is being cooled. A cooled reservoir with a given mass of gas will have a reduced pressure according to (1) as $T_{\text{hot}}/T_{\text{cold}} = P_{\text{hot}}/P_{\text{cold}}$. If this reservoir is connected to our He^3 pot at high pressure it will cause expansion of He^3 from the He^3 pot to the cooled vessel. Neglecting friction and any heat losses due to thermal conductivity, the expansion process is adiabatic and the He^3 gas produces work during its flow from the He^3 pot to the additional

reservoir. This process is essentially single shot, but in the case of He³ gas even single shot expansion is very effective because the operating temperature range is wide. For example cooling of the reservoir from 80K down to 4K corresponds to a pressure drop of 20 times. This effect could be described in different language as an increase of its effective volume by factor 20 according to $T_{hot}/T_{cold} = V_{cold}/V_{hot}$ at constant pressure (analogy with the piston expander).

At low temperatures the effect of pressure drop will be increased because this reservoir is filled with an adsorbent and becomes an adsorption pump, which at a temperature below 20K starts to adsorb He³ gas and decrease the pressure still further. After liquefaction of part of the He³ gas the process automatically goes to the standard operation mode of pumping vapour above the He³ liquid.

Analysis shows that to make such a single expansion refrigerator effective for operation from 4.2K one needs to create a very high initial pressure and optimise the volumes in the whole system and its temperature profile. That is because this system has as its essential element an adsorption pump with limited sorption capacity for He³ gas, at a pressure below 10⁻³ mbar, which corresponds T~300mK. The adsorption pump should operate without saturation the whole time until all liquid He³ in the He³ pot has been evaporated .

The simple calculations of the expansion conditions are based on the simplified picture Fig2. The maximum pressure for this system will be defined by the quantity of He³ gas, adsorbed at T=4.2K and P= 10⁻³ mbar. For simplicity we neglect the volume of the connecting tube. Then

$$P_{max} = C_0 \rho V_0 / [V_0(T_0) + V_3(T_3)] \quad (5)$$

where ρ is density of adsorbent, and C_0 –sorption capacity at 10⁻³ mbar , V_3 -volume of He³ Pot, T_0, T_3 -initial temperatures of adsorption pump and He³ pot before expansion respectively.

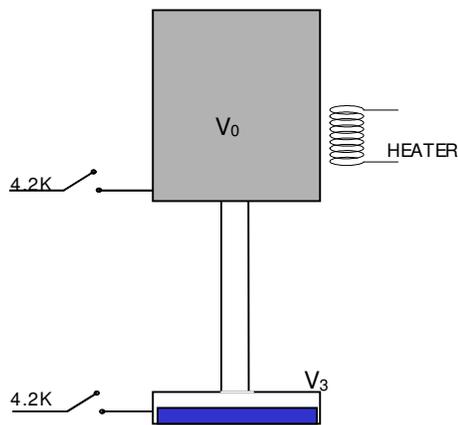


Fig.2 Schematic of a single expansion He³ refrigerator vessel with a reservoir V₀ with variable temperature. The heater and heat switches provide for heating and cooling of components to 4.2K.

The maximum pre-expansion pressure will be defined by the amount of adsorbed He³ gas, the volume of the He³ pot and the initial temperature of the adsorption pump. The limited amount of He³ in the system will lead to a small liquefied fraction and restrict the refrigerator to a short hold time at the base temperature. In practice to get a reasonable liquid fraction after expansion the He³ gas initial pressure should be more then 100 bar at the room temperature. Naturally, such a high pressure in the He³ system impacts the system design and safety requirements.

In this work we describe another method of significantly increasing the liquid fraction without needing a very high initial pressure. We called it the multi-expansion process [4].

The main feature of this method is to deliberately saturate the adsorption pump with He³ at the initial stage with the aim of being able to increase the gas pressure. For this we utilised the pressure dependence of the sorption capacity. For example at a pressure of about 100 mbar the adsorption pump can accept a double quantity of He³ gas compared with 10⁻³ mbar. According to (5) P_{max} will rise by the same factor, which is good, but we should remember that after expansion with such a large amount of gas the adsorbent will be saturated. It will not then be able to operate as a pump at the pressure of 10⁻³ mbar to obtain the base temperature.

To solve this problem we used a multi-expansion process, which needs an additional volume sufficient to accommodate all the extra He³ gas during the first stage of expansion. For simple expansion the extra gas can be transferred from the He³ pot to the room temperature reservoir [5]. To accommodate more He³ during the first stage of expansion we made a pre-expansion reservoir with small volume, but cooled by attachment to the first stage of the pulse tube refrigerator (PTR). As a result the room temperature pressure in our system does not exceed 10 Bar (good safety feature).

The multi-expansion layout is shown in Fig3. The procedure is as follows:

1. The adsorption pump is cold and has adsorbed the maximum amount of He³ gas from the cold dump (valve is open) until it saturated at approximately 100 mbar.
 2. The-adsorption pump is warmed up to 60-80K and the valve is closed to create maximum pressure in the He³ pot.
 3. Wait for about 20-30min until the He³ Pot has pre-cooled to ~4.2K.
 4. The first stage of expansion, with the adsorption pump warm, happens by opening the valve and discharging high pressure He³ gas into the cold dump
 5. The second expansion stage occurs by cooling the adsorption pump with the valve closed.
- After cooling of the adsorption pump below 20K it works like a standard He³ evaporating refrigerator down to the base temperature of <300mK. The typical fraction of He³ liquid obtained with this procedure is ~50%.

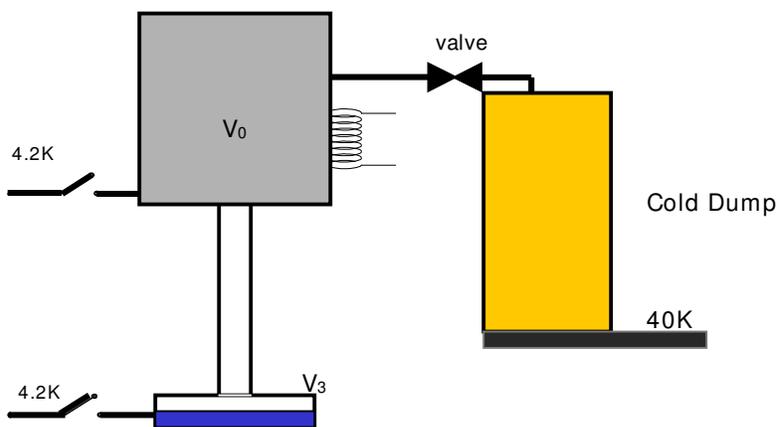


Fig 3. Schematic view of multi-expansion layout

NOTE: It is important to note that the valve operation does not have a significant effect on the refrigerator's vibration performance because the valve's operation takes place only during the regeneration mode and not at base temperature.

Utilising the multi-expansion method we have built two types of He³ refrigerator, i.e. “wet” and “dry”, at Oxford Instruments Superconductivity.



Fig.4 Experimental “wet” prototype of expansion He³ refrigerator for operating in liquid He⁴

The “wet” system means that the He³ refrigerator is operating in a dewar with liquid He⁴ at 4.2K. The refrigerator itself has a vacuum jacket with a capillary inside for liquid He⁴ flow through heat exchangers for pre-cooling the He³ gas and the adsorption pump. The development prototype was tested in January 2003 and an its external view shown in Fig.4.

The “wet” system shown in Fig.4 (vacuum jacket has been removed) has achieved the following specification:

Base temperature <270 mK, Hold time >70hours, Cooling Power 100 microWatt at T<350mK Total He³ amount ~7 L STP

This system is genuinely vibration less and could satisfy the most serious demands for vibration sensitive measurement.

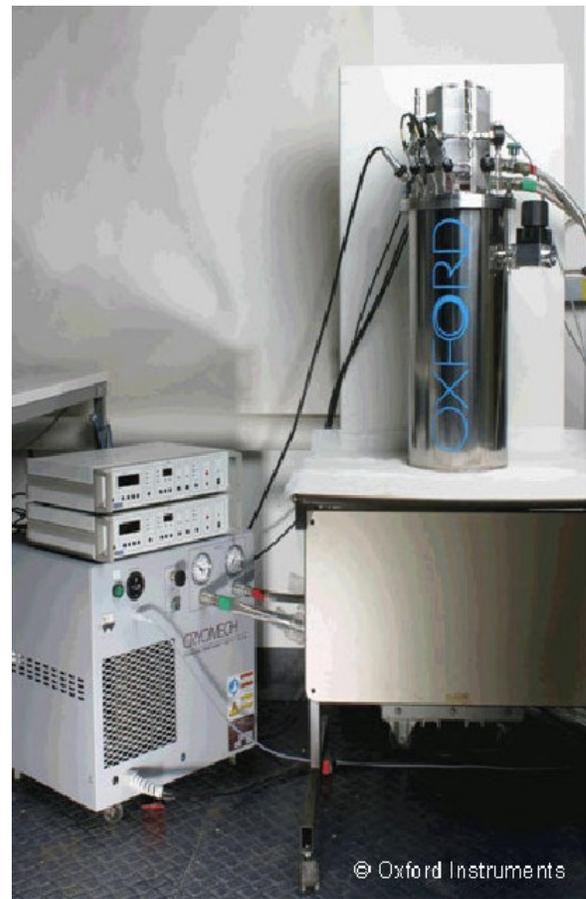
The second type of refrigerator is based on the “dry” version. It is a so-called cryogen free system based on a pulse tube cryocooler (Cryomech PT405).

The base temperature and hold time of this Cryofree system is very similar to the “wet” system. It was also established that the adiabatic He³ refrigerator is rather economical and does not require the high cooling power of the PT 405, which is 0.5 Watt at 4.2K. The disadvantage of this type PTR's is the existence of a water-cooled compressor, which makes the cryocooler very bulky and has high electrical Power consumption.

Recently Cryomech Ltd has developed a special pulse tube PT403 with an air-cooled 3KW compressor and the cooling power 300 mW at 4.2K.



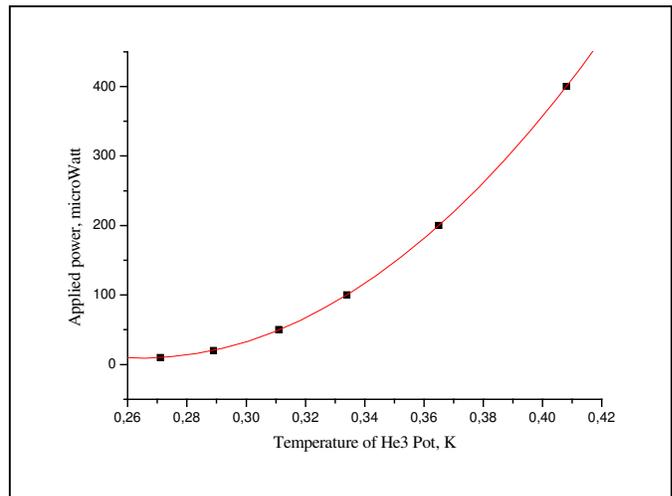
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Fig 5 The Cold Head of PT403 with adiabatic expansion He³ refrigerator and the general view of Cryogen free system Heliox AC-V

Fig 6. Cooling Power curve for Heliox AC-V



That is sufficient cooling power for operation of our adiabatic system and OIS has developed a product Heliox AC-V based on the air-cooled PT403.

Additional specification parameters:

- Temperature stability - better than ± 2 mK at < 5 K
- Regeneration time 1 hour typically
- Cooldown < 10 hours typically

CONCLUSIONS

Oxford Instruments Superconductivity has developed and built two types of He³ refrigerator with liquefaction of He³ gas by the adiabatic expansion process. Both systems have a similar specification. The “wet” system is a genuine vibrationless refrigerator for the most sensitive measurements. The “dry” system is operated from single-phase electrical power, does not need a water cooler and hence became the first ultra low temperature “kitchen” refrigerator. It is important to mention the reliability of the expansion refrigerator. Because current cryocoolers produce cooling down to <3K, this causes an extra liquid fraction by direct condensation into the He³ pot. However this temperature is close to the He³ critical point so any failure in the pulse tube and subsequent deterioration of the minimum temperature achieved by the PTR will completely stop the operation of a traditional He³ refrigerator. For our He³ system this limit is much higher at 4.2K or even 5K, where adiabatic expansion of He³ still produces liquefaction of He³ and hence ultra low temperatures. This fact was proven experimentally.

We believe that the described here method will progress in the future. This point is supported by a recent publication [6] by a group from Roma University of a “wet” refrigerator based on adiabatic expansion of He³.

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