The He³/He⁴ Dilution Refrigerator

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Abstract. The principle of the He³/He⁴ dilution refrigerator is briefly discussed. Thermodynamic expressions are derived for the cooling capacity and for the ultimate temperature to be reached in a device of this novel type.

1. Introduction

Apart from gas liquefaction, the most common procedure for cooling is evaporation of a liquid by pumping off its vapor. Continuous and efficient refrigeration is obtained by this method. At low temperatures the vapor pressure is proportional to $\exp[-L(0)/RT]$, where $L(0)$ is the latent heat of evaporation per mole at $T = 0$ and R is the gas constant. The vapor pressure thus decreases exponentially with temperature, fewer and fewer molecules can be removed by the pump, and eventually a stable situation is reached in which cooling due to evaporation just balances the external heat leak. The lowest temperatures produced by this method, using He³ for which $L(0)/R = 2.5\,°K$, are about $0.25\,°K$.

For cooling still further, the only practical method available until quite recently was adiabatic demagnetization of a suitable paramagnetic salt. Temperatures in the millidegree region can be reached with this technique. Adiabatic demagnetization, however, is basically a 'one shot' method of cooling; after demagnetization the system begins to warm up due to the presence of heat leaks. For this reason, the method is unsuitable for experiments in which large continuous sources of heat cannot be avoided.

Recently, the first prototypes of the He³/He⁴ dilution refrigerator were made operational. A cryostat of this new type is a continuously working refrigerator, capable of absorbing large heat loads even with relatively modest pumping arrangements. In fact, as soon as the necessary 'know how' is available, a dilution machine is simpler in construction and easier to operate than a demagnetization cryostat. A further advantage is that magnetic fields, needed in many types of experiments, can be tolerated. It may thus safely be predicted that refrigerators of this new type will be very important in the immediate future.

The original suggestion for the principle of the dilution refrigerator was made by LONDON, CLARKE and MENDOZA [1] in 1962, and the first successful machines of this type were built by NEGANOV, BORISOV and LIBURG [2] in late 1965 and by HALL, FORD and THOMPSON [3] in early 1966. Further development work has been done, in particular, by WHEATLEY and his coworkers [4–7]. A dilution cryostat for Möss-

bauer effect research has been built by EHNHOLM, KATILA, LOUNASMAA and REIVARI
[8]. The low temperature record using a dilution refrigerator is held, at present, by
WHEATLEY, VILCHES and ABEL [7]: 0.010 °K in continuous operation and 0.0045 °K
for a short time. Several groups have quite recently reached 0.025 °K in continuous
operation.

2. Liquid Mixtures of He³ and He⁴

In order to describe the principle of the dilution refrigerator we must first review
the properties of liquid mixtures of He³ and He⁴. Figure 1 shows the phase diagram
in the (T, x)-plane where $x = n_3/(n_3 + n_4)$ is the concentration of He³. Above the
phase separation curve the liquid is superfluid or normal depending on whether the
point (T, x) is on the left or right side of the λ-curve, respectively. Below the phase
separation curve the liquid spontaneously separates into two components, one of the
phases being rich in He³ and the other rich in He⁴. Because of its lower density the
He³-rich phase floats on top of the He⁴-rich phase.
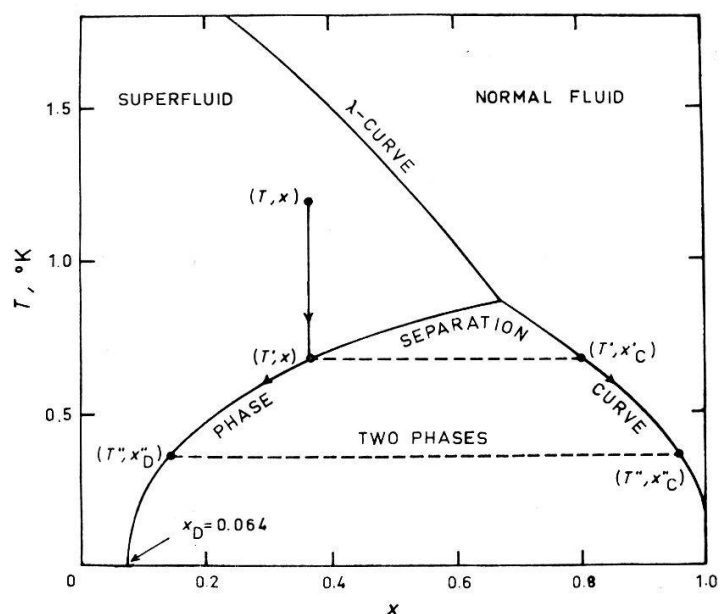

Figure 1

Phase diagram of He³/He⁴ mixtures; x is the He³ concentration.

Let us start from the point (T, x) in the single-phase superfluid region (cf. Fig. 1)
and then lower the temperature, keeping x constant. When the coexistence curve is
reached at (T', x) the two phases begin to form. On further cooling from T' to T'' the
concentration of He³ increases in the upper phase while the lower phase becomes
richer in He⁴. The point representing the lower phase moves along the left branch of
the phase separation curve to (T'', x_D''). The (T, x)-relationship in the upper phase is
given by the right branch of the coexistence curve; upon cooling the representative
point moves from (T', x_C') to (T'', x_C''). Since the operating temperature of a dilution
refrigerator is below 0.1 °K we observe from Figure 1 that in the upper or concentrated
He³ phase $x_C \simeq 1$ (actually $x_C = 0.99997$ at 0.1 °K) and that in the lower or dilute
phase x_D varies slightly with temperature, from 0.064 at 0 °K to 0.070 at 0.1 °K.

For the operation of a dilution refrigerator it is of utmost importance that the equilibrium concentration of He³ in the dilute phase, even at the absolute zero, is finite and has the rather high value $x_D = 0.064$. This will become evident shortly. Basically, the 6.4% solubility of He³ into He⁴ at $T = 0$ is due to the fact that a single He³ atom is more strongly bound into pure liquid He⁴ than into pure He³. One might argue against the finite solubility of He³ into He⁴ at the absolute zero on the basis of the third law of thermodynamics as applied to the entropy of mixing. According to classical statistical mechanics the entropy of mixing is $- R [x_D \ln x_D + (1 - x_D) \ln (1 - x_D)]$, which, for $x_D = 0.064$, has the value $0.24\,R$. The third law requires, however, that the entropy is zero at $T = 0$. The dilemma is resolved by quantum statistics. In fact, both in the pure He³ phase and in the mixture the system will be at the ground state at $T = 0$, and thus the entropy vanishes. In the case of the mixture, the He⁴ particles, obeying Bose-Einstein statistics, have all condensed to their ground level, and the He³ particles, obeying Fermi-Dirac statistics, are filling all available energy levels within a precisely defined Fermi sphere.

3. Principle of the Dilution Refrigerator

The principal parts of a continuously operating dilution refrigerator are shown in Figure 2. The phase separation line occurs in the mixing chamber, and cooling is produced there by causing He³ atoms from the upper, almost pure He³ phase to move across the boundary to the lower, dilute phase. This process is, in many respects, analogous to ordinary evaporation, but here the upper phase corresponds to liquid and the lower phase to vapor. The continuous dissolving of He³ atoms from the concentrated to the dilute phase is obtained by circulating He³ in the system by
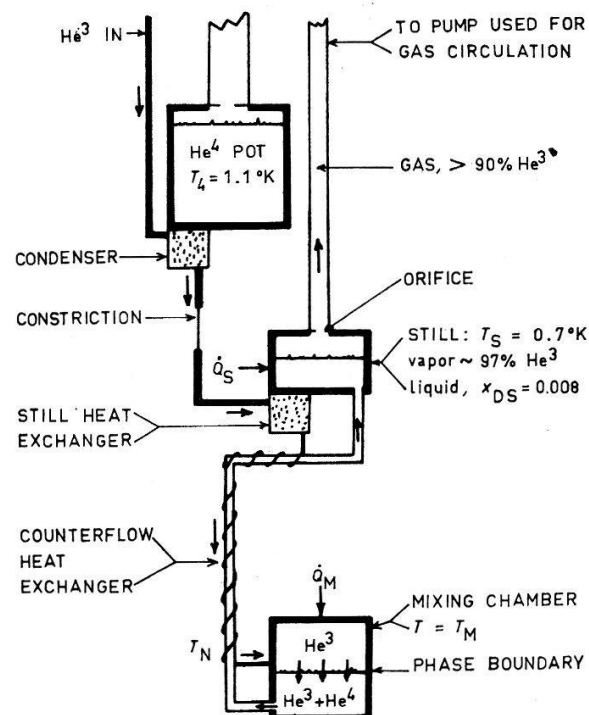

Figure 2

The principal parts of a continuously operating dilution refrigerator.

means of a pump at room temperature. Incoming gas is first precooled in the condenser attached to a He^4 pot at 1.0–1.2°K. The pressure of He^3 is kept sufficiently high for condensation to occur by means of a flow-limiting constriction. The liquid then enters the still heat exchanger at 0.6–0.8°K, the counterflow heat exchanger, and finally, the mixing chamber. After crossing the phase boundary He^3 atoms, driven by an osmotic pressure gradient, proceed through the counterflow heat exchanger, along an unbroken column of the dilute superfluid phase, to the still. Vapor is removed from the still by pumping. Over 90% of the outcoming gas is He^3 because He^4 atoms are, even at the still temperature, relatively inert. The small amount of He^4 in the circulating gas can be ignored in a simplified discussion of the dilution refrigerator. External heating of the still is required to keep its temperature high enough to achieve a sufficient gas circulation rate.

It is now clear that the main reason for the success of the dilution refrigerator is due to the 6.4% solubility of He^3 into He^4 even at $T = 0$. When the temperature of the mixing chamber is lowered, during the operation of the refrigerator, we do not 'run out' of He^3 molecules in the dilute or 'gas' phase. The circulation rate of He^3 is thus independent of the mixing chamber temperature.

Besides the continuously operating model discussed above, there are two other kinds of dilution refrigerators on which development work has been done: the single-cycle type and the superleak-operated type.

In the single-cycle dilution refrigerator no inlet is provided for the concentrated He^3; circulation of He^3 is thus not possible. During operation the concentrated phase in the mixing chamber is continuously depleted by pumping away He^3, and the refrigeration capacity is rather soon exhausted, when all He^3 has been removed. On the other hand, because there is no heat leak due to cooling of the incoming He^3 to the temperature of the mixing chamber, it is possible to reach very low temperatures in a single-cycle refrigerator. WHEATLEY and his collaborators [6–7] were able to obtain 0.0045°K in a device of this type, precooled by a continuously operating dilution refrigerator.

In the superleak-operated dilution refrigerator the mixing chamber is, at the beginning, partially filled with He^3. The mixing chamber is connected by means of a superleak, a tube filled with fine powder and allowing only superfluid He^4 to pass through it, to another chamber containing pure He^4. Cooling is achieved by forcing gradually, using external pressure, He^4 into the mixing chamber whereby the dilute phase continuously grows at the expense of the concentrated phase. So far, however, a superleak-operated refrigerator has only produced 0.15°K [9].

4. Entropy and Enthalpy

Using thermodynamics, we shall next derive expressions for entropy and enthalpy which are important for an understanding of the dilution process.

Because of its zero nuclear spin and superfluid properties, He^4 is thermally and hydrodynamically inert at temperatures below 0.5°K and acts just as a 'supporting' medium for He^3 in the dilute phase. Experimental measurements show that the specific heat of this phase is given by the specific heat of an ideal Fermi-Dirac gas, provided that a slight variation is allowed in the effective mass m^* as a function of

He³ concentration. The density of the Fermi gas must be taken to be the same as the number density of He³ in the dilute solution. Therefore, at $T \ll T_F$, we find for the specific heat per mole of liquid, $C_D = x_D R \pi^2 T/2 T_F$, or per mole of He³ in the solution, $C_{3D} = R \pi^2 T/2 T_F$. (Cf. the expression for the electronic specific heat of a metal.) The Fermi temperature $T_F = 0.38°K$ at $x_D = 0.064$. The entropy of the dilute phase can be calculated from the specific heat by integration. At sufficiently low temperatures, below 0.04 °K, one finds

$$S_{3D} = \int_0^T (C_{3D}/T) \, dT = R \pi^2 T/2 T_F = 107 \; T \; \text{J/(mole of He}^3) \times °K^2. \qquad (1)$$

The thermodynamic functions of the concentrated phase can be calculated from experimental measurements of the specific heat of pure liquid He³. Below 0.04 °K, $C_3 = 24 \; T$ J/mole × °K² whereby we find

$$H_3 = H_3(0) + \int_0^T C_3 \, dT = H_3(0) + 12 \; T^2 \; \text{J/mole} \times °K^2. \qquad (2)$$

$H_3(0)$ is the value of H_3 at $T = 0$. The entropy becomes

$$S_3 = \int_0^T (C_3/T) \, dT = 24 \; T \; \text{J/mole} \times °K^2. \qquad (3)$$

In the mixing chamber, under equilibrium conditions, the partial chemical potentials $\mu_{3C} = \mu_3$ and μ_{3D} of He³ in the concentrated and dilute phases, respectively, are equal, i.e.

$$H_3 - T S_3 = H_{3D} - T S_{3D}. \qquad (4)$$

We thus find, using Equations (1)–(3)

$$H_{3D} = H_3 + T (S_{3D} - S_3) = H_3(0) + 95 \; T^2 \; \text{J/(mole of He}^3) \times °K^2. \qquad (5)$$

This relation is valid below 0.04 °K at $x_D = 0.064$. Large differences, due to the binding energy and interactions between the He³ atoms, are found between the enthalpy of the dilute solution and of an ideal Fermi gas. This is why we cannot calculate H_{3D} by integration from the specific heat C_{3D} of an ideal Fermi gas.

A rather comprehensive treatment of the thermodynamics of the dilution refrigerator, with many useful tables and graphs, has been published by RADEBAUGH [10]. We also refer to a paper by PESHKOV [11].

5. Cooling Capacity and Ultimate Temperature

We next assume that the dilution refrigerator has reached a steady state, the temperatures of the mixing chamber and the still being constant at T_M and T_S, respectively. Further, let $\dot{Q}_M = dQ_M/dt$ and $\dot{Q}_S$ be the rates of heat input (cf. Fig. 2). We make the simplifying assumption that only He³ is circulated, and calculate the cooling capacity and ultimate temperature of a continuously operating dilution refrigerator under these idealized conditions.

External heating of the mixing chamber is caused by $\dot{Q}_M$ and by the incoming liquid He³ which must be cooled at constant pressure ($P = 0$) from the temperature T_N, at which it leaves the counterflow heat exchanger, to T_M. Cooling is simultaneously produced at the rate $\dot{N}_3 [H_{3D}(T_M) - H_3(T_M)]$ by dilution; here $\dot{N}_3$ is the molar circulation rate of He³. In equilibrium

$$\dot{Q}_M + \dot{N}_3 [H_3(T_N) - H_3(T_M)] = \dot{N}_3[H_{3D}(T_M) - H_3(T_M)] \tag{6}$$

giving, by applying (2) and (5),

$$\dot{Q}_M = \dot{N}_3 [H_{3D}(T_M) - H_3(T_N)] = \dot{N}_3 (95\ T_M^2 - 12\ T_N^2)\ \text{Watt/mole} \times {}^\circ\text{K}^2. \tag{7}$$

With an ideal heat exchanger $T_N = T_M$, provided that, as in the present case, the cooling capacity of the outgoing liquid is sufficient, and (7) yields $\dot{Q}_M = 83\ \dot{N}_3\ T_M^2$ Watt/mole $\times {}^\circ\text{K}^2$ (this also applies to the single-cycle dilution refrigerator); the cooling capacity is thus proportional to the square of the mixing chamber temperature. As was emphasized before, $\dot{N}_3$ is not a function of T_M because x_D is independent of temperature below 0.04 °K. We now clearly see the difference between the dilution process and an ordinary evaporation type refrigerator. In the former case the number of He³ atoms removed by the pump per unit time is independent of T but the cooling capacity per He³ atom decreases proportionally to T^2, whereas in the latter case the number of molecules removed from the vapor phase decreases exponentially with T while the cooling capacity per particle remains constant at $L(0)/N_0$ (N_0 is Avogadro's number).

By assuming $\dot{Q}_M = 3$ erg/s and $\dot{N}_3 = 3 \times 10^{-5}$ mole/s one obtains in the ideal case $T_M = 0.011$ °K. With $T_N/T_M = 2$ and with the same values of $\dot{Q}_M$ and $\dot{N}_3$ we find $T_M = 0.015$ °K. These calculations show the capabilities of a dilution refrigerator; even with a relatively modest gas circulation rate, easily handled by a rather small diffusion pump, and with a considerable heat leak to the mixing chamber, temperatures below 0.02 °K can be reached. Experimental curves for the cooling capacity of three different dilution refrigerators are shown in Figure 3.
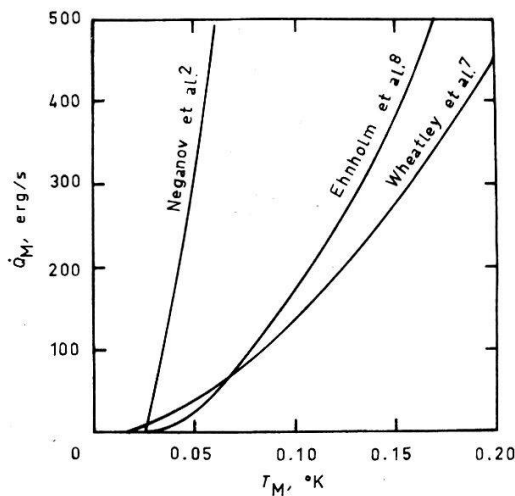
Figure 3

Cooling capacity of three dilution refrigerators as a function of the mixing chamber temperature.

According to Equation (7) the cooling capacity $\dot{Q}_M$ and the ultimate temperature in the mixing chamber both depend critically on the ratio T_N/T_M. An effective heat exchange between the incoming concentrated He³ and the outgoing dilute He³ is thus essential. At the operating temperatures of a dilution refrigerator the main thermal barrier is the Kapitza resistance, generally observed between liquid helium and a solid body. Experimental measurements show that this resistance is proportional to T^{-3}, i.e. it increases rapidly towards lower temperatures. It is very important to reduce the Kapitza resistance as much as possible by having large surface areas in the heat exchanger, especially at low temperatures.
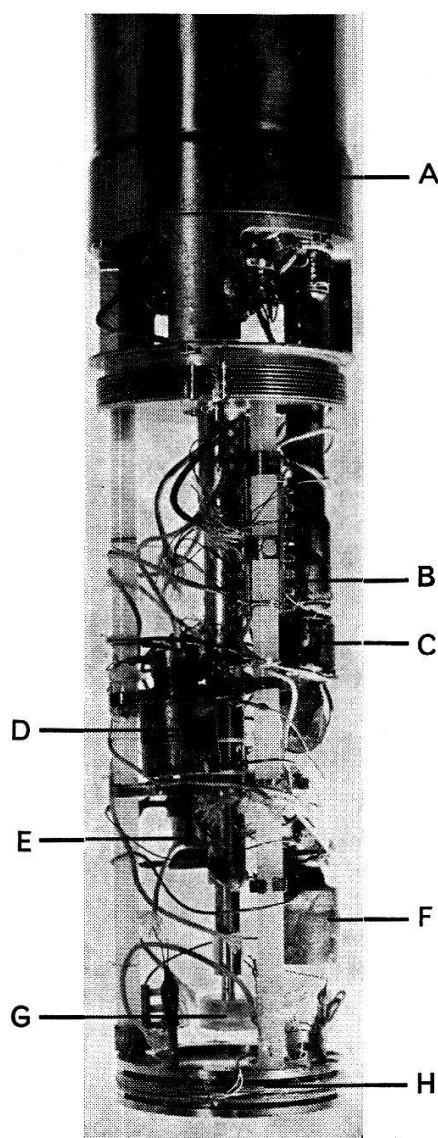

Figure 4

The dilution refrigerator of EHNHOLM et al. [8] A = He⁴ pot, B = still, C = still heat exchanger, D, E, F = heat exchangers, G = source holder, H = mixing chamber.

We do not intend to discuss here the constructional details of dilution cryostats. As an example, however, Figure 4 shows the dilution refrigerator built by EHNHOLM, KATILA, LOUNASMAA and REIVARI [8]. This apparatus is used for Mössbauer studies at very low temperatures.

Acknowledgments

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Section efficace totale de la réaction $^{16}O(d,n)^{17}F$

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I. Introduction

La détermination de la section efficace totale de la réaction $^{16}O(d, n)^{17}F$ présente avant tout un intérêt d'ordre pratique. Cette réaction permet d'une part d'effectuer un dosage rapide et précis de l'oxygène dans de nombreux échantillons dont l'activation par deutons ne produit pas d'activité comparable à celle de ^{17}F (β^+, 1,75 MeV, T 1/2 = 66 s). D'autre part, des activités appréciables de ^{17}F sont constatées dans l'eau lourde de réacteurs où elles sont induites par les deutons ayant subi une diffusion élastique de neutrons rapides. Une connaissance détaillée du processus pourrait être d'une certaine utilité.

BONDELID et al. [1] ont déterminé avec précision l'énergie du seuil de la réaction: 1829,2 ± 0,6 keV. La répartition angulaire des neutrons et des sections efficaces différentielles ont été données pour des énergies inférieures à 5,02 MeV par HODGSON [2] et par YARAMIS [3]. Seul le premier niveau excité 1/2+ de ^{17}F a pu être observé [4, 5]. L'analyse des distributions angulaires effectuée dans le cadre de la théorie de BUTLER [3] et par DWBA [2] montre qu'il s'agit principalement d'une réaction de stripping à