^{3}He-^{4}He DILUTION REFRIGERATORS

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For over thirty years, the only way to obtain very low temperatures has been to perform adiabatic demagnetization experiments.

An entirely new refrigeration cycle was suggested, in 1951, by Dr. H. London 1) ; more definite proposals were formulated, in 1962, by himself, G.R. Clarke and E. Mendoza 2). Successful experiments were carried out almost simultaneously by H.E. Hall et al 3) at the University of Manchester and by B. Neganov and his coworkers 4) from the Institute for Nuclear Problems in Dubna.

This new method opens the way for new experiments in the very low temperature regions because it permits one to cope with fairly large amounts of heat and operates without magnetic fields.

The working principle of this refrigeration cycle rests on the properties of ^{3}He-^{4}He solutions and, more specifically, on the phase separation these solutions undergo below a certain critical temperature. The phase separation phenomenon is due to the large mass difference between ^{3}He and ^{4}He, which gives rise to large differences in zero point energies and anharmonic terms in the interaction potential. The phase separation curve, shown in figure 1, indicates that a given solution with a molar concentration in ^{3}He, x, larger than about 6 %, will separate below a certain temperature into two phases, the upper one richer than the other in ^{3}He.

Thus, at temperatures below a few tenths of a degree, there exist only very concentrated solutions and fairly dilute ones.

The thermodynamical properties of these solutions are, for our purposes, best described by means of the enthalpy H. The enthalpy diagram, shown in figure 2, is constructed from specific heat measurements for the rich phase and from a simple ideal gas model for the dilute phase. More details will be found in the appendix.

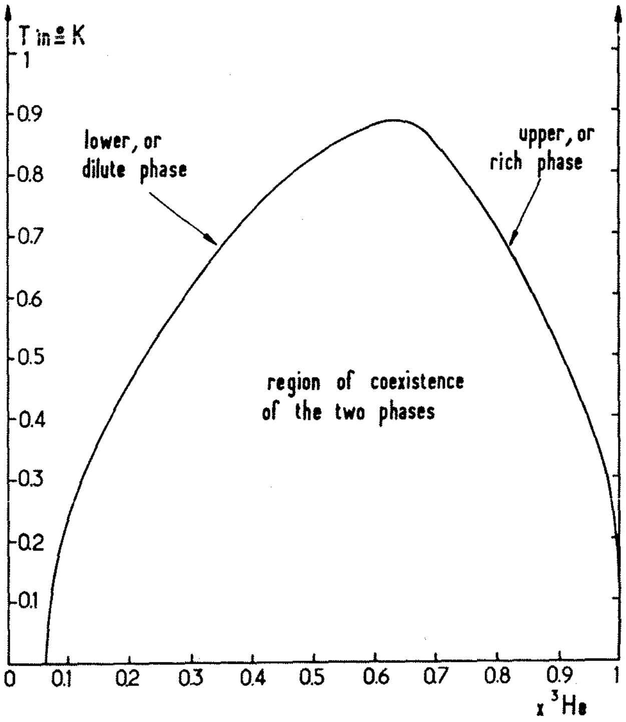

Fig. 1 Phase separation curve of helium solutions at satured vapour pressure.

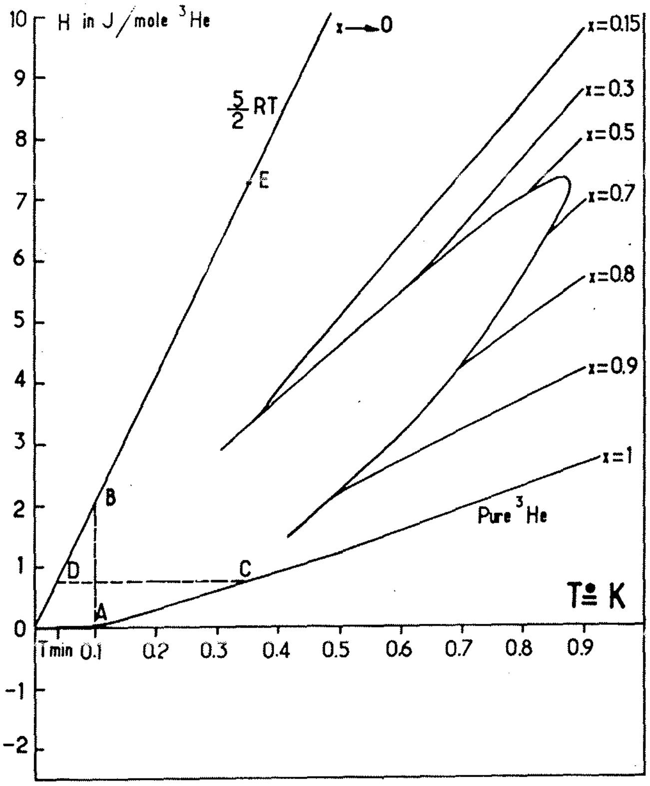

Fig. 2 Enthalpy diagram for ^{3}He pure and in solution. The straight line represents the enthalpy of an ideal classical gas with the same binding energy as pure ^{3}He.

In the rich phase, which is nearly pure ^{3}He, as can be seen in figure 1, the molar enthalpy is given by :

$$H_r(T) \simeq H_3(T) = - L_{03} + \int_0^T C_3 dT \tag{1}$$

where H_3 is the enthalpy of pure ^{3}He, L_{03} its latent heat of vaporization at absolute zero, and C_3 its specific heat under constant pressure.

In the dilute phase, ^{4}He, being superfluid, plays no significant thermodynamical or hydrodynamical rôle at very low temperatures since it has zero entropy and zero viscosity ; it can be said to act merely as a filler to increase the volume occupied by ^{3}He. The dilute component of the solution behaves like an ideal gas, the molar enthalpy of which is given by :

$$H_d(x,T) = - E_{03}(x) + \frac{5}{2} RT \tag{2}$$

where $E_{03}(x)$ is the binding energy of ^{3}He in the solution at absolute zero.

Therefore, upon mixing a number of moles n of ^{3}He in ^{4}He down to zero concentration, we can absorb a quantity of heat Q at the temperature T which is :

$$Q = n \left[L_{03} - E_{03} + \frac{5}{2} RT - \int_0^T C_3 dT\right] \quad . \tag{3}$$

This isothermal process is represented on the enthalpy diagram (fig. 2) by the line AB. This line shows the maximum cooling power available at temperature T. However, heat exchangers cannot be made perfect and it is in practice not possible to achieve the ideal cycle so that a more realistic path is to cool ^{3}He down to point C for example and to perform the dilution adiabatically to point D, and then to absorb a certain quantity of heat, back to B.

D represents the point at which, with given heat exchangers, no cooling power is available anymore and sets the minimum temperature one can reach. One also sees on the H-diagram that a considerable cooling power is available above the temperature T, when going from B to E for instance ; this will help quenching the heat leaks in an actual apparatus.

From formula 3, at a temperature T = 0.1° K and with a circulation rate $n = 10^{-4}$ mole per second, setting $E_{03}(x)$ nearly equal to L_{03}, we find a refrigerating capacity of the order of 2000 ergs per second.

The feasibility of a refrigerator working on the principle described rests on three essential facts : a) the very large cooling power of the dilution process ; b) the fact that it is possible to distil ^{3}He out of the solution in a very efficient way at a temperature of 0.6° K ; c) the fairly low heat conductivity of the mixture that enables one to perform the dilution in the mixing chamber ① (fig. 3) and to extract ^{3}He in a distillation chamber ② at a much higher temperature through a duct ③.

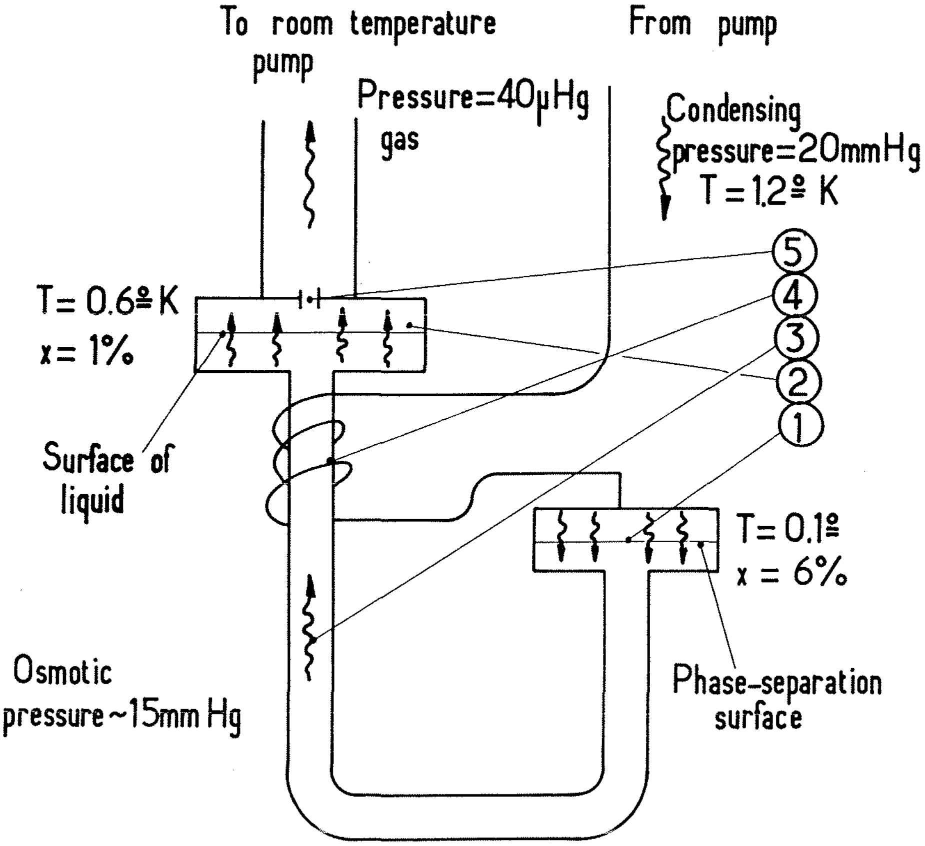

Fig. 3 Schematic description of a dilution refrigerator : ① mixing chamber, ② distillation chamber, ③ duct, ④ heat exchanger, ⑤ constriction on the pumping line. The direction of circulation of ^{3}He is indicated by the wiggly arrows to which are attached the relevant physical parameters of a typical working situation.

The problems met with in the construction of such a refrigerator are : a) the conception of efficient heat exchangers ④ to precool the incoming ^{3}He, to take advantage of fact a. b) the superfluid helium film creep along the wall of the distillation chamber, which has to be stopped by a constriction ⑤, to take advantage of fact b. c) the design of the duct ③ which must introduce adequate thermal isolation and a negligible osmotic pressure drop, by taking advantage of fact c.

Five such machines have been reported working at the 10th International Conference on Low Temperature Physics in September. Their performances are summarized in figure 4.

	Neganov Dubna	Hall Manchester	Wheatley Illinois	Zinov'eva Moscow	Leyden group
$\dot{n}$ circulation rate in mole/s	1.8×10^{-4}	3×10^{-5}	3×10^{-5}	1×10^{-5}	10^{-5} to 10^{-6}
Minimum temperature T_{min}	25 m° K	55 m° K	30 m° K	$\lesssim$ 0.1° K	88 m° K
Cooling power at 0.1° K $\dot{Q}$ (erg/s)	1800	40	200	0	-
Factor of merit $\dot{Q}/\dot{n}$ (erg/mole)	10^7	1.33×10^6	6.6×10^6	0	-

Fig. 4 Comparative table of performances.

APPENDIX

In a more sophisticated attempt to derive the H-diagram in the low temperature limit, we shall use the Landau-Pomeranchuk 5,6) model for dilute ^{3}He-^{4}He solutions which is confirmed by experiments 7,8) and according to which the Fermi excitations in the solution have the energy-momentum relation :

$$\mathcal{E} = - \frac{E_{03}(x)}{N_A} + \frac{p^2}{2m_3^*}$$

and the same number density as ^{3}He atoms in the solution. The effective mass m_3^* is independent of the concentration and equal to 2.4 times the real mass of ^{3}He 7). The binding energy $E_{03}(x)$ is temperature independent and varies linearly with x 9). The rich-phase is taken to be pure ^{3}He 10,11).

The condition for equilibrium at absolute zero is found by equating the chemical potentials of ^{3}He in the two phases :

$$- E_{03}(x_\ell) + RT_F(x_\ell) = - L_{03} \qquad (4)$$

where $RT_F(x_\ell)$ is 3/5 times the kinetic energy U of ^{3}He excitation gas and x_ℓ is the maximum stable concentration.

At finite temperature, this equation reads :

$$- E_3(x) + \mu_F(x,T) = - L_{03} - \int_0^T S_3 dT \qquad (5)$$

where μ_F is the kinetic part of the chemical potential and S_3 the entropy of pure ^{3}He. Equation 5 has been used by D.O. Edwards et al 9) to determine $E_{03}(x) - L_{03}$ knowing the phase separation curve $x_S(T)$.

The molar enthalpies in the two phases and the heat of dilution are given, in this quasi-particle theory, by :

$$H_d(x,T) = - E_{03}(x) + \frac{5}{3} U(x,T) \qquad (6)$$

$$H_r(T) = - L_{03} + \int_0^T C_3 dT \qquad (7)$$

$$\Delta H(T) = L_{03} - E_{03}(x_S) + \frac{5}{3} U(x_S,T) - \int_0^T C_3 dT \qquad (8)$$

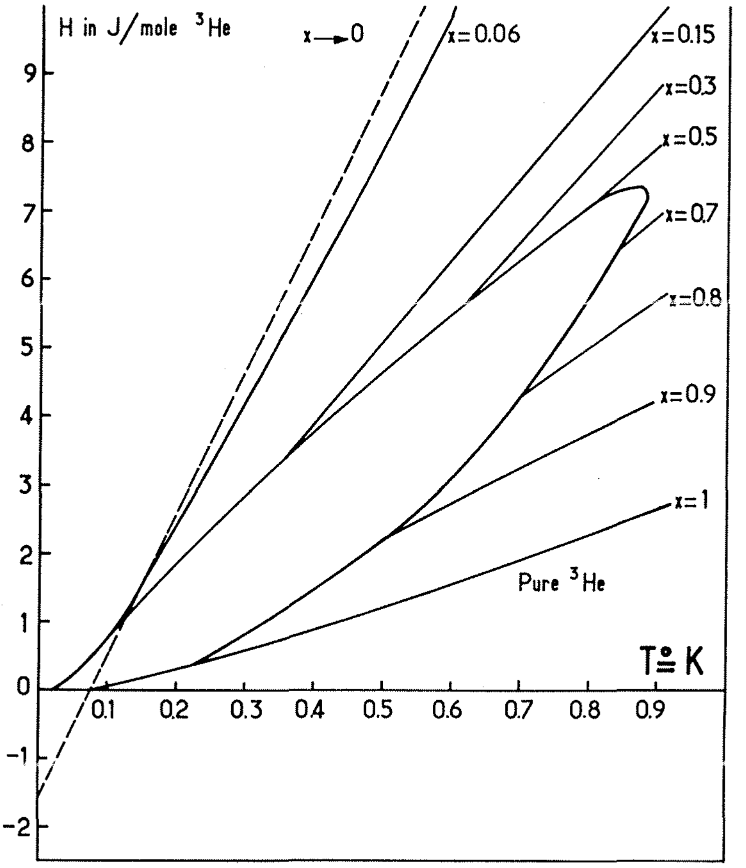

Fig. 5 Enthalpy diagram for ^{3}He pure and in solution in the quasi-particle model for an osmotic pressure equal to the zero-point pressure of the equivalent Fermi gas. The straight line represents the enthalpy of an infinitely dilute gas $\frac{5}{2}$ RT + L_{03} - $E_{03}(0)$.

C_3, the specific heat of pure ^{3}He at constant pressure, is known experimentally to a fair accuracy. The numerical evaluation of equation (8) can be easily performed with the help of the Fermi integral tables [12] and the results are plotted on figure 5.

In the case of a reversible dilution, the heat of mixing tends to zero as T^2, as already predicted by Peshkov [13] on other grounds. Finally, we wish to point out that the value we have derived for ΔH(0.1° K) in the frame of the Landau-Pomeranchuk model differs from the one observed by Neganov [4], the predicted one being too small by 30 %.

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Also, O.E. Vilches and J.C. Wheatley (to be published) who give a precise experimental confirmation of equation (8) and have obtained an ultimate temperature of 20 m° K (13 m° K in transient regime) ; we are grateful to O.E. Vilches and J.C. Wheatley for sending a preprint to us.