ON $^3$He–$^4$He DILUTION REFRIGERATORS

PART I

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Abstract
An analysis is presented of the dilution refrigeration process in those cases in which $^3$He is circulated. Though the treatment is fundamentally equivalent to that of Wheatley and co-workers, our analysis leads via the exact solution of the differential equation involved, to a direct determination of cooling powers, lowest temperatures and temperature distributions along tubes. The special merits of a recently tested quasi-continuous dilution refrigeration method are elucidated within the framework of the presented thermodynamic description.

1. Introduction
In 1962 London, Clarke and Mendoza 1) proposed a method for refrigeration in the millikelvin region using $^3$He–$^4$He mixtures. The idea is based on the observation that at temperatures below 0.87 K a phase separation occurs in the liquid mixture into a $^3$He-rich and a $^3$He-poor phase and that there is a difference in entropy per $^3$He atom in the two phases. Due to differences in specific weight the separation in phases is such that the $^3$He-rich part floats on top of the $^3$He-poor part.

When $T$ is lower than about 0.15 K the $^3$He concentration in the $^3$He-poor phase, denoted by $X$, being the number of $^3$He atoms divided by the total number of atoms in this phase, satisfies the experimentally determined relationship 2) $X(T) = X(0) (1 + \beta T^2)$, where $\beta = 10.8 \text{K}^{-2}$ and where $X(0) = 0.064 \pm 0.0005$. Below 0.03 K the concentration at phase equilibrium can be put equal to $X(0)$, the error being at most one percent. The $^3$He-rich phase in this temperature range is 100% pure. In figure 1 the phase diagram of $^3$He–$^4$He mixtures at saturated vapour pressure $p$ is displayed below 2.17 K, where pure $^4$He becomes superfluid. Note that $^3$He–$^4$He mixtures are superfluid in the $(X, T)$ region marked II, contrary to the region I where a mixture is normal. A mixture of $^3$He and $^4$He at temperatures below 0.03 K, containing more than 6.4% $^3$He is thus composed of a 100% pure $^3$He liquid floating on top of a 6.4% solution of $^3$He in $^4$He.

Refrigeration is obtained, if $^3$He can be selectively exhausted from the $^3$He-poor phase. This will then in turn give rise to a supply of $^3$He out of the $^3$He-rich phase as equilibrium has to be maintained. As the entropy of $^3$He atoms appears to be much lower in the $^3$He-rich phase than in the $^3$He-poor phase,
Fig. 1. Phase diagram of $^3\text{He}^4\text{He}$ mixtures where $X = n_3/(n_3 + n_4)$ is the $^3\text{He}$ concentration, and $n_3$ ($n_4$) the number of moles $^3\text{He}$ ($^4\text{He}$) per unit volume. Below 0.87 K phase separation occurs. The $\lambda$-curve separates region II (superfluid region) and region I (normal region).

Fig. 2. Schematic drawing of a single-cycle dilution refrigerator. Essential elements are the mixing chamber $M$, the tube $P$ and the evaporation chamber $D$. 
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the result is cooling at the surface separating the two phases (interface). Selective exhaustion of $^3$He from the $^3$He-poor phase can be achieved by adding to the vessel in which the above mentioned mixture is contained (see for instance the chamber M in fig. 2 called the mixing chamber) a tube through which the 6.4% mixture is directly connected with a chamber that has a temperature of, say, 1 K. The $^3$He–$^4$He ratio in the vapour above the liquid mixture at 1 K is much larger than 1 due to the fact that the binding energy of $^3$He in the liquid mixture is lower than that of $^4$He. By pumping away this vapour the desired exhaustion of $^3$He can be obtained. This set-up is schematically indicated in fig. 2. There are two major disadvantages in this method of $^3$He exhaustion.

(i) Due to the much higher temperature in the evaporation chamber D, which is necessary for obtaining a sufficiently high vapour pressure there will be heat transport to the mixing chamber M.

(ii) The transport of $^3$He through the tube causes viscous loss, again limiting the cooling performance in M.

These two disadvantages have to be accepted, however, as unavoidable. Since heat conduction is favoured by large tube diameters, and viscous loss by small ones, it is possible at least with a given tube length to choose an optimum diameter. Any “dilution refrigerator” has to cope with this intrinsic problem of heat conduction and viscous loss. The purpose of the present article is to derive the temperature distribution in the close vicinity of the chamber M, not only in the case of fig. 2 but also in the case where during the cooling process $^3$He is continuously supplied to chamber M (see fig. 3). In section 2 we analyse in some detail the set-up given in fig. 2, and arrive at lowest attainable temperatures, as well as the cooling power of the device.

A drawback to this method of cooling is its finite duration. The cooling process stops as soon as all the $^3$He has been exhausted from chamber M. Figure 3 shows schematically how continuous cooling can be achieved. In this set-up, to be analysed in some detail in sec. 3, the $^3$He-gas that is pumped away is condensed again and led back to the chamber M via tube $P_2$. Though this $^3$He can be precooled somewhat by means of heat exchange with the cold tube $P_1$ that connects the mixing chamber M with the evaporation chamber D, an extra heat load on M is introduced in this way, of a nature similar to that described above. Most dilution refrigerators are built in this way. It is shown that, unless high quality heat exchangers are used, the lowest temperatures that can be reached are considerably higher than in the single-cycle set-up of fig. 2. A method is dealt with for determing the cooling powers. The first device produc- ing cooling on the above principle was built by Das, de Bruyn Ouboter and Taconis 3) in 1964. The lowest temperature they obtained was 0.22 K. Since then a number of improved versions have been designed, bringing down
the lowest attained temperature to values of the order of 2–5 mK. For a detailed analysis of a number of essential features in dilution refrigeration the reader is referred to the article 4) by J. C. Wheatley, O. E. Vilches and W. R. Abel on “Principles and methods of dilution refrigeration” and references quoted in that paper. Our aim is to reconsider the situation near the mixing chamber. It is shown that the resulting differential equations are exactly solvable.

In section 4 a few remarks are devoted to the two compartment mixing chamber proposal 5) due to F. A. Staas and A. P. Severijns. The device is schematically given in fig. 4. The two compartments on top of each other are connected both by an ordinary tube and by a superleak (by definition a superleak passes only the superfluid component of a superfluid). It will be shown that such an arrangement has several advantages. First, when operating the device in the continuous way (the interface lies in M1 in this case, and 3He is continuously supplied to M1), chamber M2 will rather quickly assume the temperature $T_{M1}$, but it will not show any temperature fluctuation as occurs in M1. Secondly, when operating the device in a single-cycle experiment (the 3He supply to M1 has been stopped and the interface lies in M2 after some time) stable operation
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Fig. 4. Schematic drawing of a quasi-continuously operating dilution refrigerator. There are two mixing chambers $M_1$ and $M_2$, connected by a tube $P$ and a superleak $S$. The other elements are as in fig. 3.

can be obtained for a few hours, say. The method is therefore called quasi-continuous dilution refrigeration method. It is well understood that in both cases $M_2$ is the chamber where any experiment should be performed.

In the analysis of the above methods of dilution refrigeration it is often justified to use a number of simplifications in the temperature dependence of the following properties: solubility of $^3$He in $^4$He, viscosity, thermal conductivity and specific heat of both the dilute mixture ($^3$He-poor phase) and concentrated $^3$He ($^3$He-rich phase). Concerning the equilibrium concentration of $^3$He in $^4$He, this quantity, $X(T)$, can be put equal to the constant $X = X(0)$, the error being 1% at 30 mK and much less below. The viscosity coefficients $\eta_d$ (d for dilute) and $\eta_c$ (c for concentrated) show 4) low-temperature behaviour such that $n_d = \eta_d T^2$ approaches a value of about $5 \times 10^{-8}$ Pa s K$^2$, while $n_c = \eta_c T^2$ approaches the value of about $2 \times 10^{-7}$ Pa s K$^2$. Similarly, the thermal conductivities $\kappa_d$ and $\kappa_c$ are such that $k_d = \kappa_d T$ and $k_c = \kappa_c T$ approach the constant values $k_d = 3 \times 10^{-4}$ J s$^{-1}$ m$^{-1}$ and $k_c = 3.3 \times 10^{-4}$ J s$^{-1}$ m$^{-1}$, respectively, for $T \to 0$. Finally the specific heat at constant volume of a dilute mixture as well as that of concentrated $^3$He, both per unit mass
of $^3$He, approach the limiting forms $C_{3d}(T) \approx \alpha_2 T$ and $C_{3e}(T) \approx \alpha_1 T$, respectively. The quantities $\alpha_1$ and $\alpha_2$ can be written $\alpha_1 = a_1/m_3$ and $\alpha_2 = a_2/m_3$, where $m_3$ is the molar weight of $^3$He ($m_3 = 0.003$ kg mol$^{-1}$) and $a_1$ and $a_2$ are the more familiar specific heat quantities per mole $^3$He, being equal to $^6) a_1 \approx 24$ J K$^{-2}$ mol$^{-1}$ and $a_2 \approx 108$ J K$^{-2}$ mol$^{-1}$. The above limiting forms can be given an approximate validity below different temperatures. Accepting a few percent deviation, the simplified dependencies for $\eta_d, \eta_e$, $\kappa_d, \kappa_e, C_{3d}, C_{3e}$ are valid below about 0.1 K, 0.5 K, 0.01 K, 0.03 K, 0.1 K, 0.1 K, respectively. Therefore, especially the use of the limiting form for $\kappa_d$ seems to be prohibited in an analysis where temperatures are considered up to 0.03 K, say, though it can be argued that the relative importance of $\kappa_d$ will be small. The analysis presented deals only with those regions in the cooling system where temperatures are low enough to use the simplified expressions given above.

The advantage of the described quasi-continuous method of cooling lies in the fact that it is not necessary to insist on the application of high-quality heat exchangers, while the lowest attainable temperatures are lower than can be produced with any other continuous method known to us, including superfluid injection (see refs 7, 8, 9, 10) and multi-step mixing chamber systems $^11)$. However, if exceedingly long cooling times are needed, the quasi-continuous method described seems to be less interesting. In our opinion superfluid injection methods are to be preferred in such cases as heat exchange problems have been largely circumvented. These methods will be described in a separate paper. An abridged version of the presented material including a superfluid injection method can be found in ref. 12.

2. Single-cycle dilution

In deriving the temperature distribution near the mixing chamber M (see fig. 2) in the steady state, it is useful to consider first in detail the flow of $^3$He through the tube P from M to D. Let the cross-section of the tube be circular; let $l$ denote the length coordinate ($l = 0$ at M) and let the diameter $d$ be independent of $l$. If $\dot{n}_3$ is the flow rate of $^3$He (i.e. the number of moles $^3$He per second passing through a given cross-section) there is, necessarily, a compensating $^4$He backflow of, say, $\dot{n}_4$ moles per second. One mass unit of $^3$He possesses an amount of enthalpy equal to $\mu_3 + TS_3$, where $\mu_3$ is the chemical potential (free enthalpy) of $^3$He and $S_3$ the entropy, both per unit mass, and $T$ the temperature. Similarly, one mass unit of $^4$He possesses an amount of enthalpy equal to $\mu_4 + TS_4$. However, in treating the transport of $^3$He and $^4$He it should be remembered that entropy in a superfluid is transported only by the "normal" component. In the case of a superfluid $^3$He-$^4$He mixture this
normal component consists of both the $^3\text{He}$ and the normal part of the $^4\text{He}$. If the $^3\text{He}$ flows, the normal $^4\text{He}$, being rigidly attached to the $^3\text{He}$, moves along with the $^3\text{He}$. Therefore it does not carry an entropy $s_3$ but $s_3 + (\varrho_{3d}/\varrho_{3d}) s_4 = s_4$ per unit $^3\text{He}$ mass, where $\varrho_{3d} = 6.93 \text{ kg m}^{-3}$ and $\varrho_{4d} = 135.1 \text{ kg m}^{-3}$ are the densities of $^3\text{He}$ and $^4\text{He}$ with which they are present in the 6.4% mixture, respectively. If an energy balance is taken over a tube slice between $l$ and $l + dl$ we have to add the proper gravitational terms. If $z$ is the vertical projection of $l$ and $g$ the acceleration due to gravity the energy balance reads

$$\dot{n}_{3}m_{3}(\mu_{3} + gz + T\varsigma_{3}) - \dot{n}_{4}m_{4}(\mu_{4} + gz) - \frac{\pi d^{2}}{4} d\left(\kappa_{0}(T)\frac{dT}{dl}\right) - \dot{Q}dl = 0, \quad (1)$$

where $m_{4}$ is the molar weight of $^4\text{He}$ ($m_{4} = 0.004 \text{ kg mol}^{-1}$). The third term in eq. (1) is the amount of heat originating from heat conduction in the liquid mixture added to the tube slice per second, while $\dot{Q}dl$ is the heat that may possibly be added from outside. We neglect any heat conduction through the tube material, as it is much less than that through the liquid. Furthermore, we will put $\dot{Q} = 0$. In order to proceed with eq. (1) it is necessary to make use of the Gibbs–Duhem equation. As we need it in a rather unusual form we will recall its derivation. Consider a vessel containing a $^3\text{He}$-poor superfluid mixture with $n_{3}$ moles of $^3\text{He}$ and $n_{4}$ moles of $^4\text{He}$. The free enthalpy, the volume and the entropy of the superfluid $^3\text{He}–^4\text{He}$ mixture are given by

$$G = n_{3}m_{3}\mu_{3} + n_{4}m_{4}\mu_{4}, \quad (2)$$

$$V = n_{3}m_{3}v_{3} + n_{4}m_{4}v_{4}, \quad (3)$$

$$S = n_{3}m_{3}s_{3} + n_{4}m_{4}s_{4}, \quad (4)$$

where $v_{3}$ and $v_{4}$ are $^3\text{He}$ and $^4\text{He}$ partial volumes; note that $\varrho_{3d} = n_{3}m_{3}/V$ and $\varrho_{4d} = n_{4}m_{4}/V$. On the one hand we have

$$dG = n_{3}m_{3}d\mu_{3} + n_{4}m_{4}d\mu_{4} + \mu_{3}m_{3}dn_{3} + \mu_{4}m_{4}dn_{4} \quad (5)$$

while on the other hand

$$dG = -SdT + Vdp + \mu_{3}m_{3}dn_{3} + \mu_{4}m_{4}dn_{4}. \quad (6)$$

It is useful to introduce a reduced pressure

$$p' = p + (\varrho_{3d} + \varrho_{4d})gz, \quad (7)$$
such that differences in $p'$ have to do with hydrodynamic pressures only. In discussing liquid flow through a tube hydrostatic pressure differences are generally unimportant. Subtracting (6) from (5) and applying (7) one easily obtains
\[ d(\mu_3 + gz) = -\frac{\theta_{4a}}{\theta_{3d}} d(\mu_4 + gz) - s_d \, dT + \frac{dp'}{\theta_{3d}}, \]
which is the desired Gibbs–Duhem equation.

When rewriting (1) with the help of (8) it will be assumed that mutual friction can be neglected. This entails that
\[ d(\mu_4 + gz) = 0. \] (9)

In the general case, where $dT$ and $dp'$ differ from zero, expression (9) implies a difference in $^3$He concentration. In what follows these differences are assumed to be small enough to justify the use of a constant value for the quantity $\theta_{3d}$. Using both (8) and (9), eq. (1) can be transformed into
\[ \dot{n}_3 \, m_3 \left[ d(T_{3d}) - s_d \, dT + \frac{dp'}{\theta_{3d}} \right] - \frac{\pi d^2}{4} d \left[ \kappa_d(T) \frac{dT}{dl} \right] = 0. \] (10)

If we assume a viscous Poiseuille flow, we have according to Poiseuille's relation, valid for circularly cylindrical tubes,
\[ dp' = -\frac{\dot{n}_3 \, m_3}{\theta_{3d}} \frac{128}{\pi d^4} \eta_d \, dl, \] (11)
where $\dot{n}_3 \, m_3/\theta_{3d}$ is the volume rate of flow of the $^3$He, $128/(\pi d^4)$ is the well-known geometrical factor for circularly cylindrical tubes, and $\eta_d$ is the viscosity coefficient. Using $n_d = \eta_d \, T^2$; $k_d = \kappa_d \, T$ and
\[ s_d(T) = \int_0^T \frac{C_{3d}(T')}{T'} \, dT' \approx \frac{a_2}{m_3} \, T, \] (12)
eq (10) assumes the form
\[ \frac{\dot{n}_3 a_2}{2} \frac{dT}{dl}(T^2) - \left( \frac{\dot{n}_3 \, m_3}{\theta_{3d}} \right)^2 \frac{128}{\pi d^4} \eta_d \frac{1}{T^2} - \frac{\pi d^2}{4} k_d \frac{d}{dl} \left( \frac{1}{T} \frac{dT}{dl} \right) = 0. \] (13)

It turns out to be convenient to transform this second-order differential equation for $T(l)$ into two first-order differential equations. It is easily shown that eq. (13)
is equivalent to the two coupled first-order differential equations
\[
\frac{df_d(t)}{dt} = \frac{2 [1 - f_d(t)]}{t} - \frac{1}{t^7 f_d(t)}, \quad (14)
\]
\[
\frac{d\lambda}{dt} = \frac{1}{t^3 f_d(t)}, \quad (15)
\]
where we introduced a dimensionless temperature \( t = T/T_a \), and a dimensionless length parameter \( \lambda = l/l_d \), with
\[
T_a = \left( \frac{128 n_d k_d m_2^2}{a_2^2 \theta_3 d^2} \right)^{1/6} \frac{1}{d^{1/3}} \quad (16)
\]
and
\[
l_d = \left( \frac{\pi^3 k_d^2 \theta_3 d^2}{1024 a_2 n_d m_3^2} \right)^{1/3} \frac{d^{2/3}}{\hat{n}_3}. \quad (17)
\]
The function \( f_d(t) \) is defined by
\[
\frac{\pi d^2}{4} k_d \frac{1}{T} \frac{dT}{dl} \equiv f_d(t) \frac{\hat{n}_3 a_2}{2} T^2. \quad (18)
\]
It can be given the following physical interpretation: \( f_d(t) \) expresses the heat flow rate due to heat conduction in terms of the temperature-dependent part of the \(^3\)He enthalpy flow rate. Introduction of ratios such as \( f_d \) enables us to present energy balances in a transparent way, especially in cases in which more than one tube is involved. Equations (14) and (15), being equivalent to the differential equations established by Wheatley et al.\(^4\)), can both be integrated explicitly. The solutions are given by
\[
\lambda = 2^{-1/3} x + \gamma_1 \quad (19)
\]
and
\[
Ai'(x) + \gamma_2 Bi'(x) = y [Ai(x) + \gamma_2 Bi(x)] \quad (20)
\]
with
\[
x = 2^{-2/3} \left\{ t^4 [f_d(t) - 1]^2 - t^2 \right\} \quad (20a)
\]
and
\[
y = 2^{-1/3} t^2 [f_d(t) - 1]. \quad (20b)
\]
Here \( \gamma_1 \) and \( \gamma_2 \) are two integration constants, while \( Ai(x) \) and \( Bi(x) \) are two linearly independent solutions \(^{13}\) of the differential equation \( W'' - x W = 0 \); the functions \( Ai'(x) \) and \( Bi'(x) \) are derivatives with respect to \( x \). It is easily verified that differentiation of (19) with respect to \( t \) yields eq. (15). The ex-
pression (20) is arrived at after the consecutive substitutions $f_1 = t^2 (f_d - 1)$; $t_0 = -1/(2t^2)$; $t_1 = t_0 + f_1^2/2$, transforming eq. (14) into $df_1/dt_1 = -f_1^2 + 2t_1$. Introduction of $u = \exp(\int f_1 \, dt_1)$ yields the equation $u'' = 2t_1 \, u$. A final substitution $x = 2^{-1/3} \, t_1$ leads to the Airy differential equation in its usual form. A set of solutions for $f_d(t)$ is given in fig. 5. Though an explicit expression for $t(\lambda)$ follows immediately from (20) if one uses $y^2 = x + 2^{-2/3} \, t^{-2}$, we proceed with the above parametric representation for physical reasons.

Fig. 5. Graphical representations of solutions of the differential equation

$$\frac{df}{dt} = \frac{2 \, [1 - f(t)]}{t} - \frac{1}{t^2 \, f(t)}$$

for positive $t$. Positive $f$ values apply if the heat conductive flow in a tube has a direction opposite to that of the enthalpy flow. Negative $f$ values apply if these flows have equal directions. The first case applies to tubes through which $^3$He is exhausted from the mixing chamber, in which case instead of $f$ the symbol $f_d$ is used in the text. In the second case, the symbol $f_d$ is used with reference to the $^3$He supply tube. The asymptotic curve is indicated with the symbol $A$. The intersection of $A$ with the line $f_d = 1.70$ is seen to give $t = 0.691$. Intersection with the line $f_d = 1.55$ gives $t = 0.707$. 
With respect to the determination of the integration constants $\gamma_1$ and $\gamma_2$ we first derive a relation at $l = 0$, that is at the very entrance of the mixing chamber. Let chamber $M$ in the steady state, at a given time, contain $N_{3c}$ moles of pure $^3$He and $N_{3d}$ moles of dilute $^3$He. The volume of $M$ can then be put equal to $V_M = N_{3c} m_3/\rho_{3c} + N_{3d} m_3/\rho_{3d}$, where $\rho_{3c}$ is the density of pure $^3$He ($\rho_{3c} = 81.5$ kg m$^{-3}$). It follows that $N_{3c} m_3/\rho_{3c} + N_{3d} m_3/\rho_{3d} = 0$. As $\dot{n}_3 = -(\dot{N}_{3c} + \dot{N}_{3d})$ one obtains $\dot{N}_{3d} = \rho_{3d} \dot{n}_3/(\rho_{3c} - \rho_{3d})$ and $\dot{N}_{3c} = -\rho_{3c} \dot{n}_3 / (\rho_{3c} - \rho_{3d})$.

If we assume the mixing chamber to be at uniform pressure $p$ and uniform temperature $T_M = T(0)$ we can easily equate the apparent decrease of enthalpy content in $M$ with incoming and outgoing energy rates at $l = 0$, that is

$$\dot{N}_{3c} m_3 (\mu_{3c} + T_M s_{3c}) + \dot{N}_{3d} m_3 (\mu_3 + T_M s_d) = -\dot{n}_3 m_3 (\mu_3 + T_M s_d) + \frac{\pi d^2}{4} k_d \frac{1}{T_M} \left( \frac{dT}{dl} \right)_M + \dot{Q}_M. \quad (21)$$

Here $\dot{Q}_M$ is the amount of heat added to $M$ per second, which will be called cooling power. The concentrated $^3$He in $M$ has free enthalpy $\mu_{3c}$ and entropy $s_{3c}$ per unit mass. Similar to (13) one has

$$s_{3c}(T) = \int_0^T \frac{C_{3c}(T')} {T'} \, dT' \leq \frac{a_1} {m_3} T. \quad (22)$$

Thermal equilibrium in $M$ yields $\mu_3 = \mu_{3c}$. Equation (21) can thus be reduced to

$$f_a(t_M) = \frac{2\rho_{3c} (a_2 - a_1)} {a_2 (\rho_{3c} - \rho_{3d})} - \frac{2\dot{Q}_M} {\dot{n}_3 a_2 T_d^2 t_M^2}, \quad (23)$$

where $t_M = T_M/T_d$. Equation (23) is only one of the starting conditions that are needed to determine $\gamma_1$ and $\gamma_2$. The natural way to find the other starting condition would be to give the temperature in the evaporation chamber $D$; if the distance along the tube from $M$ to $D$ equalled $L_D$ this condition would read $T(L_D) = T_D$. However, as stated in the introduction, the temperature of $D$ is of the order of 1 K, implying that our analysis of the flow in the pipe is certainly not valid in the neighbourhood of $D$. This would have been the case only if none of the simplified assumptions about the temperature dependence of the relevant physical quantities had been made. On the other hand, a complete treatment without these simplifications is in principle possible, although explicit solutions of the differential equations can then no longer be found. We "solve" this starting condition problem by taking quite arbitrarily, at a
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certain distance $L_d$ along the tube the value $T(L_d)$. In an actual case of a conventional dilution refrigerator we can for instance choose $L_d$ to indicate the position on the tube where it enters the lowest heat exchanger. It will be clear, however, that any change in $n_3$ will give rise to a change in $T(L_d)$. The above problem concerning the second starting condition is in fact fundamental in all types of dilution refrigerators. Physically, the reason is that the tube is connected to the outside world in a way that is not always easily specified. The adopted "solution" implies in fact that the tube at $l = L_d$ ends in a heat bath at the temperature $T(L_d)$. It will be shown that conclusions concerning lowest temperatures and cooling powers can nevertheless be drawn. So the second starting condition from now on reads

$$t\left(\frac{L_d}{l_d}\right) = \frac{T_h}{T_d} = t_h,$$

where $T_h$ is a given (bath) temperature, and $t_h$ is the related dimensionless temperature. The integration constant $\gamma_1$ does not play a physical role and is eliminated by subtracting the R.H.S. of (19) taken at $t = t_h$ from its value at $t = t_M$, yielding

$$\frac{L_d}{l_d} = -\frac{1}{2t_h^2} + \frac{t_h^4}{2} \left[f_d(t_h) - 1\right]^2 + \frac{1}{2t_M^2} - \frac{t_M^4}{2} \left[f_d(t_M) - 1\right]^2. \quad (25)$$

The integration constant $\gamma_2$ can in principle be determined by applying eq. (20) for $t = t_M$ as well as for $t = t_h$. This, together with (23) and (25), gives four equations for the four unknowns $\gamma_2$, $t_M$, $f_d(t_M)$ and $f_d(t_h)$ where the values for $T_h$, $L_d$, $d$, $n_3$ and $Q_M$ are supposed to be given.

In actual practice it is convenient to use the following procedure. At given $T_h$, $n_3$, $L_d$ and $d$ we choose some starting value $f_d(t_h) > 0$. It is then possible to calculate $x_H = x(t_h, f_d(t_h))$ and $y_H = y(t_h, f_d(t_h))$. Application of (20) with these $x$ and $y$ values gives a $\gamma_2$ value; then we take the $x$ value $x_M = x_H - (L_d/l_d) (2)^{1/3}$; applying (20) using $x_M$ and $\gamma_2$, we obtain the value $y_M$; we then calculate $t_M$ and $f_d(t_M)$ from $x_M$ and $y_M$. Application of (23) gives $Q_M$, which is the cooling power at a temperature $T_M = t_M T_d$. By varying the starting value $f_d(t_h)$ the cooling power $Q_M$ can be obtained as a function of the temperature in the lowest mixing chamber. The minimum attainable temperature is the value of $T_M$ for which $Q_M = 0$. Inserting the values for $\omega_3$, $\omega_{3d}$, $a_2$ and $a_1$ in (23) and taking $Q_M = 0$ we find $f_d(t_M) = 1.70$ which is therefore the maximum $f_d$ value possible in the mixing chamber $M$.

The above procedure is correct only if
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$$\int_0^{t_H} \frac{dt}{t^3 f_d(t)} > \frac{L_d}{l_d}, \quad (26)$$

as otherwise no proper $t_M$ and $f_d(t_M)$ values can be found. Furthermore, it is noted that one can also obtain negative $Q_M$ values, implying that the corresponding (lower) $T_M$ values are realistic only if an amount of heat $-Q_M$ is withdrawn from $M$ by some other cooling device.

When realistic values are inserted for $d$ (a few mm) and $n_3$ (of the order of $10^{-4}$ mol s$^{-1}$) in eqs (16) and (17) it appears that $T_d$ is of the order of 5 mK, while $l_d$ is about 5 mm. A reasonable value for $L_d$ is from 10 to 20 cm. A practical problem now to be solved is to determine which temperature differences $T_H - T_M$ can be bridged in the given set-up. It is useful in this respect to consider the $f(t)$ versus $t$ curves in fig. 5 in some detail. Suppose we assume that $T_H$ has approximately the value of $T_d$ or is even smaller. We then observe from fig. 5 that no large reduction in temperature is to be expected, because the $f(t)$ curves for $t < 1$ are nearly vertical. Moreover, the tube length compatible with such a temperature difference $T_H - T_M$ would turn out to be extremely small. On the other hand, if we take $T_H$ to be equal to a few times $T_d$, the starting value $t_H$ is such that a sizeable reduction in $T$ can be obtained. It can then be deduced from fig. 5, at least in a qualitative way, that choosing an $f(t_H)$ value not too close to 1 makes it possible, at least for $Q_M = 0$, to reach a very well defined minimum value of $t_M$. This value is obtained from the intersection point of the line $f_d = 1.70$ with the asymptotic curve into which each $f(t)$ curve is seen to merge. This $t_M$ value is found to be $t_{min} = 0.691$. For this reason one can state that in practice the minimum temperature to be obtained is

$$T_{min} = 0.691 T_d. \quad (27)$$

This expression is in agreement with that given by Wheatley et al.\textsuperscript{4}). These authors arrived at the minimum temperature expression by means of numerical integration of a differential equation closely related to eq. (14). It is emphasized that the derivation of the above minimum temperature is largely independent of the starting value $t_H$. The same observation holds if we compute $t_M$ values at finite $Q_M$. This shows clearly that the cooling possibilities of the set-up under consideration can indeed be investigated without specifying the starting conditions at the high temperature side in great detail. It makes sense therefore to compute the $Q_M$ versus $t_M$ curve in “practical” cases, as defined above. In figure 6 cooling powers are given as a function of the mixing chamber temperature for some tube diameters $d$ and flow rates $n_3$. The calculations were
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done with the aid of a computer. Temperature distributions along the tube between M and D were also calculated, and some results are presented in fig. 7.

It follows from (27) and (16) that \( d \) is the only parameter influencing \( T_{\text{min}} \). There appears to be no fundamental lower limit, as \( d \) can be increased at will. However, it should be realized that a large \( d \), according to (17) leads to a large \( I_d \), and therefore to a large \( L_d \). This can be compensated only by increasing \( \dot{n}_3 \) accordingly. These considerations can easily be seen to lead to a "practical" lower temperature limit of the order of 1 to 2 mK.

3. Continuous dilution refrigeration, \(^3\)He circulation only

If \(^3\)He is circulated in the way shown in fig. 3, the \(^3\)He supplied via tube \( P_2 \) leads to an extra heat load on mixing chamber M, if one compares the configuration with that of fig. 2. Part of this extra load is precisely known and equals the enthalpy flow rate of the incoming pure \(^3\)He at the position where it enters M. In addition, however, there is an unknown heat conduction contribution. In order to be able to find the magnitude of this latter term it is necessary to consider the fluid flow in tube \( P_2 \) in detail. Fortunately, the treatment is quite similar to that of tube \( P_1 \) already dealt with in sec. 2. For if one supposes that the temperatures are low enough to guarantee that the \(^3\)He supplied to M is 100\% pure, the energy balance taken over a tube slice between \( l \) and \( l + dl \) (again \( l = 0 \) at the entrance in M) reads

\[
\dot{n}_3 m_3 d(\mu_{3e} + g z + T s_{3e}) - \frac{\pi d_e^2}{2} d \left[ \kappa_e(T) \frac{dT}{dl} \right] = 0,
\]

where \( d_e \) is the diameter of the (circular) supply tube. If the positive \( l \) direction and that of \( \dot{n}_3 \) are taken parallel, application of relations analogous to eqs (8), (10), (11) and (12) leads in a straightforward way to an equation which can be obtained from eq. (13) by replacing \( a_2, \varphi_{3d}, d \) and \( k_d \) by \( a_1, \varphi_{3e}, d_e \) and \( k_e \). From the analysis in sec. 2 it follows immediately that eq. (28) is equivalent to the two coupled first-order differential equations

\[
\frac{df_e(t)}{dt} = \frac{2 \left[ 1 - f_e(t) \right]}{t} - \frac{1}{t^7 f_e(t)^3},
\]

\[
\frac{d\lambda}{dt} = \frac{1}{t^3 f_e(t)},
\]

where \( t = T/T_e \); \( \lambda = l/l_e \) with

\[
T_e = \left( \frac{128 n_e k_e m_3^2}{a_1^2 \varphi_{3e}^2} \right)^{1/6} \left( \frac{1}{d_e^{1/3}} \right)
\]

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Fig. 6. Cooling power as a function of mixing chamber temperature for the single-cycle case described in sec. 2 (see fig. 2). Results are shown for some parameter sets $T_H$, $L_d$, $d$ and $n_3$. The temperature $T_{\text{min}}$ is obtained for $Q_M = 0$.

Fig. 7. Temperature distribution along the tube between M and D for the single-cycle case described in sec. 2 (see fig. 2). Results are shown for some parameter sets $T_H$, $L_d$, $d$ and $n_3$. The cooling power is equal to zero at $T = T_M$. 
and

\[ l_c = \left( \frac{\pi^3 k_e^2 Q_{2c}^2}{1024 a_1 n_c m_3^2} \right)^{1/3} d_c^{8/3} \hat{n}_3. \]  

(32)

The function \( f_c(t) \) is defined by

\[ \frac{\pi d_c^2}{4} k_e \frac{1}{T} \frac{dT}{dl} = f_c(t) \frac{\hat{n}_3 a_1}{2} T^2. \]  

(33)

Here \( f_c(t) \) expresses the heat flow rate due to heat conduction in terms of the temperature-dependent part of the \( ^3\text{He} \) enthalpy flow rate. As the heat conductive flow has a direction parallel to that of the enthalpy flow, \( f_c(t) \) will assume negative values. Solutions of (29) and (30) are given by (19) and (20) where \( f_c(t) \) has to be substituted instead of \( f_d(t) \).

The supply tube causes obvious changes in additional conditions. Instead of two, there are now four integration constants \( \gamma_1, \gamma_2 \) and \( \gamma'_1, \gamma'_2 \). There is one condition on \( \mathcal{M} \), similar to but different from (21). Furthermore, giving the temperature \( T_{H'} \) at the position \( l = -L_c \) of the supply tube means that there is an additional condition equivalent to (25). Application of eq. (20) at \( T_M/T_d, T_M/T_c, \frac{T_H}{T_d} \) and \( \frac{T_H}{T_c} \) gives four equations. One can then determine the temperature distribution, at least at given \( L_d, L_c, d, d_c, \hat{n}_3 \) and \( \mathcal{Q}_M \). In some cases the analysis leads to non-monotonic functions \( T(l) \), implying the occurrence of negative as well as positive \( f_c(t) \) values.

For convenience it will be assumed that \( T_{H'} = T_H \). The physical situation we have in mind is that of an ideal heat exchanger, in which at the lower end the outgoing pure \( ^3\text{He} \) flow as well as the incoming dilute \( ^3\text{He} \) flow have equal temperature \( T_H \). The condition replacing (21) is easily derived to be

\[ \hat{n}_3 T_M^2 \left[ a_1 \left( 1 - \frac{f_c(T_M/T_c)}{2} \right) - a_2 \left( 1 - \frac{f_d(T_M/T_d)}{2} \right) \right] + \mathcal{Q}_M = 0, \]  

(34)

while the additional condition equivalent to (25) reads

\[ \frac{L_c}{l_c} = \frac{T_c^2}{2} \left( \frac{1}{T_M^2} - \frac{1}{T_{H'}^2} \right) + \frac{1}{2T_c^4} \left[ T_H^4 \left( f_c(T_H/T_c) - 1 \right)^2 - T_M^4 \left( f_c(T_M/T_c) - 1 \right)^2 \right]. \]  

(35)

In writing down eq. (35), the constant \( \gamma'_1 \) is eliminated. One may consider the problem of determining the temperature \( T_M \) as follows: there are seven unknowns \( f_c(T_H/T_c), f_d(T_H/T_d), f_c(T_M/T_c), f_d(T_M/T_d), T_M, \gamma_2 \) and \( \gamma'_2 \) to be calculated from a set of seven coupled equations, namely (34), (25), (35) and four equations to be obtained from eq. (20) by inserting \( t = T_H/T_c, T_H/T_d, T_M/T_c \)
and $T_M/T_d$, respectively. By varying $\dot{Q}_M$ the cooling power can be obtained as a function of $T_M$.

In figure 8 cooling powers have been given as a function of mixing chamber temperatures for some parameter sets $T_H, L_d, L_e, d, d_e$ and $\dot{n}_3$. For comparison corresponding results for single-cycle cooling with equal $T_H, L_d, d$ and $\dot{n}_3$ have been presented as well. It can be deduced from the inset in fig. 8 that for the higher $T_H$ values, continuous cooling leads to temperatures $T_M$ that are lower by a factor of 2.8 than $T_H$. This factor is smaller when the $T_H$ values are lower. This can also be directly understood from fig. 5, where the $f$ curves for negative values of $f$ can bridge a factor of 2.8 only if the $t$ values involved are large enough.

It should be realized in this connection that a comparison with the single-cycle results is misleading to a certain extent. As we arbitrarily fixed the value $T_H$, one might conclude at first sight that conventional cooling is by far inferior to single-cycle cooling. However, if nearly ideal heat exchangers are available, the cooling process in $M$ can bring the temperature $T_H$ down to very low values. On the other hand, the temperature $T_M$ at given values of $L_d, d, \dot{n}_3$ and $\dot{Q}_M (= 0)$ will not reach values as low as given in eq. (27). In the following argument an
explicit expression is derived, yielding a lower temperature limit for the (hypothetical) case in which the two tubes are in intimate contact, such that at equal \( T \), equal temperatures are found in the respective tubes. Though this limiting situation can never be reached in practice, it shows how little the derived minimum temperature differs from the single-cycle case. One has in this ideal case

\[
\frac{\pi d_c^2}{4} k_c \left( \frac{1}{T} \frac{dT}{dl} \right)_M = \frac{d_c^2 k_c}{d^2 k_d} f_d \left( \frac{T_M}{T_d} \right) \frac{n_3 a_2}{2} T_m^2,
\]

and the appropriate additional condition on mixing chamber \( M \) reads

\[
\frac{n_3 T_m^2}{2} (a_1 - a_2) + \frac{n_3 a_2}{2} T_m^2 f_d \left( \frac{T_M}{T_d} \right) \left( 1 + \frac{d_c^2 k_c}{d^2 k_d} \right) + \dot{Q}_M = 0,
\]

yielding

\[
f_d \left( \frac{T_M}{T_d} \right) = \frac{d^2 k_d}{d^2 k_d + d_c^2 k_c} \left[ \frac{2 (a_2 - a_1)}{a_2} - \frac{2 \dot{Q}_M}{n_3 a_2 T_m^2} \right]
\]

instead of (23). If \( \dot{Q}_M = 0 \) and \( d_c/d \ll 1 \) one obtains (note that \( k_c \approx k_d \)) the value 1.55 for \( f_d(T_M/T_d) \), instead of the value 1.70 obtained in the single-cycle case. It can be seen from (23) and (38) that the difference is caused by the factor \( \varrho_2(\varrho_3 - \varrho_3) \) which is rather close to 1. From fig. 5 one then finds \( t_{\text{min}} = 0.707 \) by intersecting the line \( f_d = 1.55 \) with the asymptotic curve. This represents a minimum temperature very close to the one obtained in single-cycle cooling.

The conclusion is therefore that the performance of continuous dilution refrigerators with \(^3\)He circulation depends to a large extent on the quality of heat exchangers. Frossati and co-workers have been very successful in finding high-quality heat exchangers \(^{14}\), while superfluid injection (see refs 7, 8, 9 and 10) and multi-step mixing chamber systems (see ref. 11) can be viewed as alternative attempts to reduce the heat exchange problem. Nevertheless, the quasi-continuous method, described in the introduction and in the next section, seems worth considering as it avoids heat exchange completely.

4. Quasi-continuous dilution refrigeration

From sections 2 and 3 it clearly follows (i) that the lowest temperatures in dilution refrigeration are to be obtained in single-cycle cooling, and (ii) that continuous cooling introduces heat exchange difficulties which lead to higher minimum temperatures, even in the hypothetical case of ideal heat exchange. In this section we return to the quasi-continuous set-up, indicated in fig. 4. It was stated in the introduction that when the refrigerator is operated in the
continuous mode (presence of an external $^3$He circulation) the temperature in chamber $M_2$ assumes the temperature of chamber $M_1$ rather quickly. This can be elucidated in the following way.

Compare the thermodynamical situation in the superfluid mixture near the lower end of superleak $S$ with that of a droplet of dilute mixture placed on top of $S$ in $M_2$. Consider more specifically differences in the quantities $\mu_3 + gz$, $\mu_4 + gz$, $T$ and $p'$. As far as $p'$ is concerned, there is no difference, the only pressure difference being hydrostatical. Suppose, however, that $T_{M_2} > T_{M_1}$. It will be clear that then also $(\mu_3 + gz)_{M_2} > (\mu_3 + gz)_{M_1}$. In view of the Gibbs–Duhem equation (8) it follows that $(\mu_4 + gz)_{M_2} < (\mu_4 + gz)_{M_1}$, implying that superfluid $^4$He will flow from $M_1$ to $M_2$. The result is that chamber $M_2$ will be cooled as $^3$He dissolves in the pure $^4$He injected into $M_2$. Due to gravity (there is a density difference between the “enriched” $^4$He and the surrounding $^3$He) the enriched $^4$He is transported to $M_1$. The adopted model is that the enriched $^4$He falls in the form of isolated droplets. This transport is counterbalanced by a flow of $^3$He going upwards from $M_1$ to $M_2$. The process will stop as soon as $T_{M_2} = T_{M_1}$.

An interesting consequence is that if $T_{M_2}$ at some instant should be lower than $T_{M_1}$, no backflow of $^4$He from $M_2$ to $M_1$ via the superleak can take place, there being no $^4$He in $M_2$. The superleak acts therefore as a rectifying device, stabilizing the temperature in $M_2$, as the other connection between $M_2$ and $M_1$, being the tube $P$, is assumed to be thin enough to avoid a serious heat load from below.

The proposal is to perform continuous cooling of $M_1$ and $M_2$ first, then to stop the external $^3$He supply and to go over to single-cycle cooling. After a certain time the phase separation level enters the tube $P$ and minimum temperatures and cooling powers can be obtained just as described in sec. 2, the advantage being that due to the superleak $S$, chamber $M_2$ is already at a very low initial temperature.

The above described quasi-continuous cooling method deserves attention for the following reasons:

(i) no heat exchange problem is encountered;
(ii) the cooling duration can be chosen in the order of a few hours, simply by giving chamber $M_2$ compatible dimensions;
(iii) the temperature in the mixing chamber tends to be much more stable than in other methods.

In our opinion the method is fundamentally superior to any continuous method known to us. A double mixing chamber based on this principle has been built by A. P. Severijns et al.15) and was shown indeed to provide the expected lowest temperatures.
5. Concluding remark

As a final remark we mention the fact that the heat exchange problem can be avoided to a large extent in superfluid injection dilution refrigerators. As the theoretical description of the movement of enriched \(^4\)He droplets in an environment of pure \(^3\)He is much more delicate than that of the \(^3\)He flow considered in this paper, we treat this matter in a separate paper.

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Eindhoven, October 1978

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