Conceptual design of $^4\text{He}$ film suppressor in Still of Dilution Refrigerator

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Abstract. The capacity of a dilution refrigerator in terms of its cooling power and base temperature is primarily governed by the flow rate of the isotopic mixture of helium. In the process of circulation of gas mixture, geometrical configuration of $^3\text{He}$ distillation chamber (Still) maintained at a temperature of around 0.7 K plays a significant role in controlling the flow rate. We have already employed a still in our existing dilution refrigerator, however, a new conceptual design is presented here and it is envisaged that this novel design will improve the gas circulation efficiency. Even though, thermodynamic conditions are major issues in eliminating unwanted $^4\text{He}$ film as compared to $^3\text{He}$ molecules, this design based on a new geometry is likely to enhance the pressure and the temperature at the still that helps in burning out the super-fluid $^4\text{He}$ film. The conceptual design of film suppressor and its relevance in the recently commissioned indigenous dilution refrigerator at Variable Energy Cyclotron Centre is presented in this paper.

1. Introduction

The dilution refrigerator (DR) requires a $^3\text{He}$ distillation chamber known as Still, maintained at a temperature of around 0.7 K that plays a significant role in controlling the flow rate of the isotopic mixture of $^3\text{He}/^4\text{He}$ especially for flow rate of $^3\text{He}$. The dilution refrigeration consists of five distinct units which are interconnected to make a complete and closed system. They are the condenser (1K-pot), the still, the heat exchanger (HeX), the MC and the gas circulating system. The cooling power of a dilution refrigerator with certain base temperature is primarily governed by the flow rate. In a close loop refrigerator, the presence of $^4\text{He}$ in the $^3\text{He}$ gas distilled stream downstream to the still and subsequent close loop circulation affect the cooling power, base temperature etc. at the Mixing chamber (MC) where the phase separation of isotropic liquid at the concentrated to dilute solution boundary takes place, thereby produces the cold. $^4\text{He}$ is superfluid below Lamda temperature (< 2.17K) and is co-existed within the mixture of liquid $^3\text{He}$ (as it becomes superfluid at 2.13 mK with no magnetic field) & superfluid $^4\text{He}$ at Still temperature. Creeping of this superfluid along the side wall is the cause of concern to the entire dynamic stability and performance of the refrigerator. To ensure the purity (x~1) of $^3\text{He}$, $^4\text{He}$ is restricted in the circulation of isotropic mixture and several effort are demonstrated in other laboratory in terms of a) Porous plug, b) Orifice or smaller diameter tube (imposing small flow perimeter), c) Heat exchanger for evaporation (large thermal conduction), d) knife edge device to stop the remaining flow by using atomically sharp edge.

We have already employed a Still [1] in our existing DR. We have demonstrated around 50 mK in DR refrigerator, recently in our laboratory. The Still with which the DR was commissioned, was
designed with the 2.6 mm diameter orifice. A typical circulation rate of 15 μmol/s of $^3$He was considered for the fridge. However, a new conceptual design for $^4$He superfluid film suppression aiming to improve the gas circulation efficiency is presented here.

2. Design and Development

2.1. Description of Still Performance

The room-temperature circulating pump extracts the mixture from the still and returns it to the dilution unit through successive cooling stages at 77K and 4K temperatures before it enters the condenser.

As time progresses, the $^3$He content of the circulating gas increases and the $^4$He largely settles down inside the dilute channel starting from MC to still. The circulating flow rate $m_3$ and the $^3$He concentration $x$ is controlled by the still heater for a given still design. In consequence the condensation of the circulating gas is affected and its condensing temperature reduces gradually.

The still [Fig.1 & 2] made of oxygen free high conductivity (OFHC) copper is a 40 cm$^3$, cylindrical, distillation chamber filled with $^3$He/$^4$He solution in dynamic equilibrium with its vapor phase. The vapor phase consists of mainly $^3$He with a small amount of $^4$He depending on the temperature and level of superfluid film creep. The primary drive of the dilution refrigeration process is the selective pumping of $^3$He vapor away from the still. This gas circulation produces cooling in the mixing chamber as it facilitates $^3$He flow across the phase boundary to maintain the equilibrium concentration of $^3$He in the dilute phase. The presence of excess $^4$He in the circulating gas mixture causes phase separation at relatively higher temperature and may lead to instabilities in the system. In order to maintain the amount of circulated $^4$He within the desired limit (1–10%), the still is provided with a heater as well as a plated orifice constriction and a critical heater power will ensure to sustain steady-state circulation by ensuring phase separation at a reasonably low temperature. This, in turn, produces the requisite osmotic pressure difference between the mixing chamber and the still. The still power for a given flow rate yielding the minimum fraction of $^4$He circulated can be arrived through simulation code SIDFO by J Pradhan et al [2, 3 & 4].

2.2. A subsection

The gas circulation is generated by the evaporation in the still and in turn produces cooling by maintaining the equilibrium concentration of $^3$He in dilute phase within the mixing chamber. The selection of the appropriate still power is to ensure a given flow rate with the minimum amount of $^4$He.

The presence of $^4$He in the circulating gas tends to cause phase separation at higher temperatures in the concentrated side, hence reducing the cooling power. The least amount of $^4$He in the circulating gas occurs when the still heater power is just enough to suppress the film creep and is defined as the critical heater power, $Q_{hc}$, below which the $^4$He flow rate remains almost constant and depends only on the creep rate. The pressure and $^3$He concentration of the vapour within the still is determined from the energy balance within the still for a given still power. For a pump of a given throughput, the vapour pressure thus obtained determines the amount of helium circulating through the pump.

The cooling power per unit molar flow rate is calculated and compared with Frossati and Takano models [5]. $Q_m \text{ max at } MC \text{ temperature } T_m \text{ and is given by following eq. with } T_i = T_m$

$$Q_m \approx m^2 T_m^2 \text{ watt} \quad (1)$$
The generalized cooling power $Q_m$ in the MC with a molar flow rate $m$ and $^3$He concentration $x$ is given by the energy balance in the MC as put forward by Radebaugh [6]

$$Q_m = m[xH_1[T_m] - \{\frac{(x-x_f)H_{30}[T_f]}{1-x_f} + x_f(1-x_f)H_2[T_f]\}]$$  \hspace{1cm} (2)

**Fig.1:** 1K Pot, Still Bottom & Top flange; Still during soldering.

It is assumed that the phase boundary between the dilute and the concentrated phase is isothermal and that its temperature $T_m$ is equal to that of the dilute solution in the mixing chamber. Due to the presence of $^4$He in the circulating mixture the amount of pure $^3$He available for refrigeration is reduced to $m(x-x_f)(1-x_f)$ and the dilute solution already separated out at $T_f$ will contain $m(x)(1-x)(1-x_f)$ moles of $^4$He and thus the expression for cooling power in the MC is obtained assuming a negligible quantity of $^4$He. If we consider, $x = 1$, the above equation can be reduced to

$$Q_m = m[H_1[T_m] - H_{30}[T_f]]$$ \hspace{1cm} (3)

**Fig.2:** Schematic of DR Cryostat (Dilution kit assembled)

Thereby, the presence of $^4$He in the circulating gas reduces the achievable cooling power. Primary goal is to design a superfluid film suppressor so as to restrict the $^4$He within 1-10% in the circulating gas mixture i.e. $^3$He-$^4$He ratio of about 10 to 90 or even more than 300 as achieved by Kirk [7].

The $^3$He concentration gradient decreases gradually along the dilute channel towards the still. The applied heater power of still, $Q_{hs} > Q_{ht}$ decides the ultimate temperature and finally, cooling power by controlling the circulation flow rate and the $^4$He concentration. With reduction of the heater power, i.e. $Q_{ht} < Q_{hc}$, the total flow decreases, although, the steady superfluid film creep will result in an overall...
increase of the $^4$He concentration. The vapour pressure above the liquid surface is estimated from the still temperature $T_s$ and the $^3$He concentration, $x_s$, in the liquid phase using the following relation [8]:

$$P_v = 8.313 T_s x_s \exp \left( \frac{27.85 M_{he} x_s}{8.313 T_s} \right) \cdot 10^{-5}$$  \hspace{1cm} (4)

Where, $P_v$, $x_v$, $M_{he}$ are Vapour pressure in the still, $^3$He concentration in vapour phase, Effective mass $^3$He -dilute solution respectively and $x_s = m_3/( m_3 + m_4)$. The change in flow rate may be computed until the pressure at the pump inlet becomes sufficient to maintain the desired volume flow for a given pump capacity.

This minimum temperature may not be achieved in practice since the flow required is unlikely to realize due to high heat load on the still. For the given surface area, a higher circulation flow rate arising from increased heater power, may result in the positive effect of more atoms crossing the phase boundary which is likely to offset by the negative effect of an increasing heat load from the incoming concentrated stream on the mixing chamber. Ultimately the residual heat load decides the minimum temperature achieved during operation of the dilution refrigerator. The degradation of cooling power beyond a certain flow rate is attributed to viscous heating and the increase of $^4$He in the circulating gas.

Similarly, relatively higher still power is required for higher MC temperature to obtain maximum cooling power. The temperature of the still provides the primary information about molar flow rate under steady state situation and their dependence. The flow rate of $^3$He i.e. $^4$He concentration which tends to increase with the still temperature becomes minimum at 0.6 K, corresponding to a critical power of about 0.4 mW.

The variation with flow rate of the osmotic pressure difference between the mixing chamber and still for different mixing chamber temperatures to maintain a continuous flow of $^3$He through the dilute channel, the osmotic pressure difference ($\Delta P$) between the mixing chamber and the still should be maintained higher than the pressure drop across the dilute side of the refrigerator which primarily depends on the dilute-side impedance of the heat exchanger. The osmotic pressure remains almost constant for flow rates varying between 40 µmoles/s and 120 µmoles/s (50–80mK). For temperature of more than 70 mK, the osmotic pressure difference decreases sharply at lower flow rates which necessitates operation at a higher flow rate for a higher mixing chamber temperature and vice versa (higher flow rate from higher still power giving higher osmotic pressure). For lower temperatures (10 mK), the still power is comparatively less, with the lower flow rate being sufficient to maintain adequate osmotic pressure.

2.3. Design Aspects

Still is made of copper materials for its body and silver plated flange surfaces for lower resistance while mounting various sensors and heaters. Electrical resistance (manganin wire) of the heater is $\sim$ 400 Ω. A heat exchanger is wrapped over still wall to sub-cool $^3$He stream.

The film burner rather than Film suppressor is a heated copper piece located inside the $^3$He pump line close to the still. It is equipped with Cu tube which protrudes into the still pot to maintain the temperature distribution. The film burner is heated above $\lambda$- point which burns away the “super fluid” film. The $^4$He vapour re-condenses in the still but does not enter the pump line through which we selectively pump out $^3$He.

For “single cycle” operation, the tube size would be small whereas for higher throughput, the tube diameter would be large enough to give good characteristics.

Film suppressing or burner operation principle is essentially like this: the mobile $^4$He film is partially evaporated and made immobile before it can enter the exit tube of the Still. The two prominent criteria must be met to suppress $^4$He film flow – first, $^4$He mobile film must be evaporated at a rate of equals to critical flow rate of the film through the orifice. The evolved vapors must be deflected through baffles to condense immediately in the still. Secondly, the $^4$He film on the Cu pipe must not be in the super-fluid state. The temperature of the tube must be higher than the temperature of the liquid in the Still ($T_{\text{still}}$) such that the unsaturated $^4$He film on these surfaces is not a super fluid. So,
the overall design tends to minimize convective heat loss from the Cu tube which is directly connected to exit tube.

2.4. Orifice flow limitation

The maximum $^3$He flow occurs when the flow reaches the velocity of sound in the reducing orifice. The first law of thermodynamics shows that the change in enthalpy of the gas between the still and in the orifice, assuming adiabatic expansion,

$$
\gamma = C_p/C_v\text{ (Specific heat ratio) } = 5/3; \text{ for He (mono-atomic) adiabatic expansion } \tau_s = T_s \left( \frac{p_s}{p_s} \right)^{\gamma / (\gamma + 1)}
$$

$$
D, \text{ diameter of orifice, } T_s, \text{ temperature in Still, } M, \text{ molar mass, } R, \text{ the gas constant, } \psi, \text{ is function } \psi = \frac{n^2}{8MRT_s}
$$

$$
\psi^2 = \frac{\gamma - 1}{\gamma - 1} \left( \frac{p_s}{p_s} \right)^{\gamma - 1} \left( \frac{p_s}{p_s} \right)^{\gamma + 1} \left( \frac{p_s}{p_s} \right)^{\gamma + 1}
$$

For $^4$He $p_s/p_0 = 0.51$, gas velocity is maximum and equal to sound velocity at $T_0$.

For vapour pressure, in the temperature range, $0.6 < T < 1K$

$$
p_s = p_{atm} \approx 1.22 \times 10^6 e^{T_s / 9}
$$

Limiting value for $^4$He or $^3$He flow,

$$
n_s = 4.2D_s p_s T_s^{-\gamma / 2}
$$

$$
n_s = 3.6D_s p_s T_s^{-\gamma / 2}
$$

As per Radebaugh\cite{6} (temp: $0.6 < T_s < 0.8$ K)

$$
p_s = p_{3He} \approx 0.705 e^{T_s / 3}
$$

2.5. Compressible flow effect

In pipes of constant cross-section, the Mach no $M_a = 1$ characterizes the highest achievable velocity as subsonic gas only be accelerated to $M_a < 1$ in converging/diverging nozzles. As a consequence, the diameter of a pump line is chosen such that the velocity of the fluid at the desired flow rate does not exceed the speed of sound ‘$a$’, which depends solely on the temperature $T$ and can be calculated by $a = \sqrt{\gamma RT}$ as per CERN thesis 2007\cite{9}; $\gamma = 5/3$ for ideal mono-atomic gas, $M$ is the molar mass of the gas. Close to the still, for instance, where the temperature is in the range of 700 mK and $M = 3$ g/mol, the speed of sound, $a$, is 56.86 m/s.

2.6. Superfluid Film Flow

As the pump line is used to reduce the vapour pressure above the bath below the Lambda point, a superfluid film creeps up the pump wall to warmer areas where it evaporates. The volume, $V_{\text{film}}$ of liquid $^4$He escaping the still can be calculated by multiplying the cross-section of the film by its flow speed.

$$
V_{\text{film}} = 2\pi rf W_{\text{crit}}
$$

where, $r$ is the radius of the tube/orifice, and $W_{\text{crit}}$ is the velocity of the superfluid film (approximately 20 cm/s). The factor $f$ accounts for the difference between geometric and effective perimeter of the
tube; it amounts to approximately 3 if no special measure (such as electro-polishing) are taken. The thickness, \( t \) of the superfluid film can be estimated as \( t \approx 30h^{1/3} \), where \( h \) is the height above the free surface of the bulk liquid in cm. The film thickness \( t \) is yielded in nm. The amount of liquid lost due to superfluid film flow can easily be converted into an equivalent heat load (calculated as \(~0.36 \text{ mW}\) through multiplying by the latent heat of evaporation for \(^4\text{He}\).

The mass flow rate of the film, \( m_{\text{film}} \) as per Shirron P et. al [10] is

\[
m_{\text{film}} = \rho_s C \nu_c (d - d_0)
\]

where, \( \rho_s \) is the superfluid density (which at the temperature of interest is equal to bulk density, \( \rho \)), \( d \) is the film thickness, \( d_0 \) is the inert layer thickness (almost 1 layer, 0.36nm), and \( C \) is the smallest perimeter of the vent line or flow channel. The critical velocity of superfluid, \( \nu_c \) is a function of only \( d \) and temperature \( T \).

2.7. **Extended tube with heater**

We are planning to upgrade the superfluid film suppression with a thin wall 3 mm diameter tubing connected to orifice as per Fig.3 [option-I] with heater element inside the still.

![Fig.3: Proposed superfluid film suppressor for Dilution refrigerator.](image)

**2.7.1. Principle** The heater will evaporate the film of superfluid before entering into the suction tube however, the upstream side of the tube will expose to helium vapor whereas the downstream side towards orifice will expose to superfluid rich helium vapor. To avoid the pressure the length and the heater power are to be controlled for smooth operation. The thin wall will enhance the lesser gradient of temperature across the wall; whereas, the stainless steel, the tubing material, will maintain a longitudinal temperature gradient.

2.8. **Porous Plug**

We are also planning to upgrade the superfluid film suppression with a porous plug as per Fig. 3 [option-II].

**2.8.1. Principle** The porous plug is traditional device to segregate helium gas from the superfluid helium even in zero gravity by using thermo-mechanical effect. It is made of porous material such as sintered alumina, stainless steel etc. The size of the pore is typically in the order of only few micron. When the upstream side of the plug is connected to the superfluid based gas mixture, the superfluid flows through the pores to the downstream side and evaporates immediately lowering the local
temperature to a few tens of mK because of absorbed latent heat. The superfluid tends to move towards higher temperature and thereby the liquid helium is retreated into the pores hence the helium vapor is separated out from the liquid.

2.8.2. Layout of porous plug The mass flow rate of superfluid helium, $M$, through the plug as per H Nakai et al [11 & 12], Martin F et al [13] and Chaudhry et al [14] is ideally as

$$M = -\frac{ST}{k + ST} \frac{qA\lambda}{\eta_n} r_p$$

(13)

where, $q$, $S$, $T$ and $k$ are density, specific entropy, temperature and latent heat of the superfluid helium. $A$ and $\lambda$ are cross sectional area and permeability of the porous plug. $\eta$ is a normal component of viscosity and $r_p = (p_{He} - p_S)/t$, $t$ the plug thickness] is the pressure gradient across the porous plug. For a typical values of $q = 0.145 \text{ g/cm}^3$ below 1.3K, $S = 5.881e-3 \text{ J.mole}^{-1}\text{K}^{-1}$ at 600 mK, $k = 72.09 \text{ J.mole}^{-1}$, $r_p \sim 1e5 \text{ Pa.m}^{-1}$, $A = 2.1 \text{ cm}^2$, $\lambda = 1.96e-14 \text{ m}^2$, $\eta_n = 2.1 \mu\text{Pa.s}$, $M = 0.139 \mu\text{g/s}$.

The mass flow rate of helium vapour, $M_v$, through the plug as per H Nakai is ideally as

$$M_v = -\frac{q_v A\lambda}{\eta_n} \frac{\Delta p_v}{l_v}$$

(14)

where, $q_v$, $\Delta p_v (p_{sat} - p_0)$ and $l_v$ are density, pressure drop and thickness of vapour phase respectively. Flow rate increases as the downstream temperature drops further. This is normal because of higher evaporation rate of the superfluid helium inside the porous plug.

The sizing of the porous plug is mainly guided by the above equations and the $\Delta T$ of design at certain base temperature. The thickness of the plug depends on the temperature gradient, $r_T$, which is proportional to $r_p$, the pressure gradient too.

3. Results and Discussion

The performance of only plated orifice in the still, in terms of superfluid Helium suppression, needs further experiment, however, the proposed superfluid film suppressor scheme are also advantageous. The uncertainty in liquid level inside still will greatly disturb the functioning of porous plug. Their performance will be validated through experiments in the laboratory. In addition to the film burning function in the DR refrigerator at the laboratory, the scheme can be extended even in $^3$He purification application with suitable modification. However, this is beyond the scope of our DR.

4. Conclusion

Influence of $^4$He content in the circulating gas and its outcome in terms of cooling power and mixing chamber temperature is discussed in great details. It is seen that increasing flow rate does not necessarily increase the cooling power beyond a certain value. Osmotic pressure within the dilute channel is accordingly begins to reduce. However, cooling power increases linearly up to certain flow and then start to decrease. The still power plays a key role on the flow rate vis a vis $Q_m$ and $T_m$ for a given design of the system.

It achieves the desired performance of the machine considering intricate relationships among all the sub-systems and helps to diagnose the cause of malfunctioning. The present film suppressor with a combination of precise plated orifice and still heater is working to a certain extent. In spite of the technological challenges in developing proper porous plug, the conceptual design of the superfluid helium film suppression on partial modification of the Still through either or combined options will be useful in terms of

a) Limiting the superfluid film creep, thereby, restricting percentage of $^4$He<<10% reduces the thermo-dynamical instability of the DR performance.

b) Possibility lower base temperature in the MC is feasible.

c) The cooling power of the DR can be achieved near to the maximum as theoretically achievable.
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