Thermodynamic Process and Analysis of Dilution Refrigerator

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Abstract. Thermodynamic analysis is essential for not only guiding the design of refrigerator, but also studying its operation mechanism. As the core component, the dilution unit plays an important role in deciding the performance of dilution refrigerator. In this study, a coupled thermodynamic model is established by analyzing each component separately for improving the operation of dilution refrigerator. And an optimization calculation based on that model is carried out to obtain some meaningful results on the dilution unit design. The optimal flow rate of the dilution refrigerator under different working conditions are pointed out. By considering the effect of viscous heat on the performance of the heat exchanger, the applicable conditions of the continuous heat exchanger and the methods to overcome the viscous heat are given.

1. Introduction
Dilution refrigerator, as a cryogenic technology which can obtain temperature below 100mK, has been applied in various fields such as aerospace and energy [1–4]. In order to fathom the mechanism of dilution process and optimize the design of dilution refrigerator, many analyses and verifications on the properties of ³He-⁴He mixtures and thermodynamic process of dilution unit were studied [5–10].

The complication of dilution unit lies not only in the varying properties of the circulating fluid but also in Kapitza boundary resistance due to extremely low temperature. In order to quantitatively analyze the cyclic migration of ³He atoms in the dilution refrigerator and its thermodynamic process, separates model were established for the three core components of the dilution unit and coupled analysis was performed. Based on analysis of the thermodynamic process in dilution unit, the parameters in the dilution unit were calculated quantitatively in the coupled model.
2. Models

2.1. Cycle

The operation process of dilution refrigerator considered for our model is shown in figure 1. Generally, dilution refrigerator is composed of five parts which are the precooling part, the still, the counterflow heat-exchanger, the mixing chamber and the gas circulating system. For a dry dilution refrigerator, precooling 1 and precooling 2 refer to the two stages of a precooling cryocooler respectively. The cycled working medium is first pre-cooled by a two-stage mechanical cryocooler to be below 4K. The purpose of the flow resistance is to make $^3$He obtain sufficient cohesion pressure and prevent gas from entering the heat exchanger. The $^3$He liquid coming from the flow resistance is further cooled by the still and then enters the counterflow heat-exchanger. The mixing chamber contains a concentrated phase (almost pure $^3$He) and a diluted phase ($^3$He-$^4$He mixtures). After passing through the counter-flow heat exchanger, $^3$He undergoes dilution process in the mixing chamber to produce a cooling power and then returns in the opposite direction to complete a cycle. The mixing chamber is the lowest temperature part of the system.

![Figure 1. The operation process of dilution refrigerator](image)

As the critical part of dilution refrigerator, the still, the counterflow heat-exchanger and the mixing chamber are called the dilution unit which is marked in figure 1. The temperature of this unit is always below 1 K during the operation and these components interact with each other. We first analyzed the migration process of helium from the dilution of the mixing chamber to the evaporation in the still, and developed thermodynamic analysis models for each component separately and synthesized these models to obtain the law of mutual influence between various parameters by MATLAB. In this model, the temperature of the concentrated side and the diluted side of the counterflow heat-exchanger are $T_c$ and $T_D$. The flow rate of circulating working medium is $\dot{n}$. The cooling power is $Q_{mc}$ at the temperature of $T_{mc}$ in the mixing chamber. The temperature of $^3$He fluid entering the still heat-exchanger is $T_{in}$. The temperature of the still and $^3$He fluid after the still is $T_s$.

2.2. Still model

The still is a $^3$He-$^4$He mixture container where $^3$He fluid entering the dilution unit is further cooled and $^3$He in $^3$He-$^4$He mixture evaporates to the next cycle. The gas evaporated from the still generally contains a small amount of $^4$He. figure 2 shows the thermodynamic model of the still. We assume that the molar concentration of $^3$He in the evaporated gas is $x$. The entering concentrated fluid is cooled from $T_{in}$ to $T_s$ by the still, so the enthalpy change of $^3$He fluid is $\Delta H_c = H_c(T_{in}) - H_c(t_s)$. The heating power of the still is
The energy conservation equation is given by

\[
\dot{Q}_{\text{ht}} = n \Delta h_c + \dot{Q}_n = nx h_{i0} + n(1-x) h_s + n \Delta h_D \tag{1}
\]

Here \( h_c, h_{i0}, h_s, h_D \) respectively represent the specific enthalpy of concentrated phase, pure \(^3\text{He}\), pure \(^4\text{He}\) and diluted phase. On an ideal condition, the gas evaporated from the still is pure \(^3\text{He}\), \( x=1 \). The energy conservation can be given by

\[
n \Delta h_c + \dot{Q}_{\text{ht}} = nh_{i0} + n \Delta h_D \tag{2}
\]

2.3. Counterflow heat-exchanger

The counterflow heat-exchanger, located between the still and the mixing chamber, is the most pivotal component in the dilution unit where the incoming concentrated liquid from the still exchanges heat with the diluted mixtures from the mixing chamber. The temperature of incoming concentrated \(^3\text{He}\) drops from \( h_{i0} \) to \( h_{i} \).

At extremely low temperatures especially below 100 mK, Kapitza boundary resistance between solids and liquids increases sharply as the temperature decreases. Kapitza boundary resistance is defined by \[ R = \frac{\Delta T}{Q} \tag{3} \]

The boundary between liquid helium and solid is \( R \propto T^{-3} \) and Kapitza boundary conductivity is \( \alpha = 1/(4 R_{KM}) \) for extremely low temperatures especially below 100 mK \( (R_{KM} = A R T^3) \). But in some other literature, \( R_s \) does not conform to the above-mentioned law\[14,15\]. We use it as a set value. The energy conservation equations of two sides of the counterflow heat-exchanger are respectively given by

\[
s_D \frac{d}{dz} \left( k_d \frac{dT_D}{dz} \right) + \mu v_D \frac{d^2T_D}{dz^2} + \frac{dA}{dz} \left( T_D^4 - T_D^4 \right) \alpha = \dot{n}C_p \frac{dT_D}{dz} \tag{4}
\]

\[
s_C \frac{d}{dz} \left( k_e \frac{dT_C}{dz} \right) + \mu v_C \frac{d^2T_C}{dz^2} + \frac{dA}{dz} \left( T_C^4 - T_D^4 \right) \alpha = \dot{n}C_p \frac{dT_C}{dz} \tag{5}
\]

Where \( s, k, \mu, A, C \) respectively represent cross-sectional area, thermal conductivity, viscosity, heat exchange area, specific heat and the following tables C and D indicate the concentrated and diluted phase. Ignoring the thermal conductance and viscous heat, we can simplify the above two equations to

\[
\frac{dA}{dz} \left( T_C^4 - T_D^4 \right) \frac{4R_{KM}}{4R_{KM}} = \dot{n}C_p \frac{dT_C}{dz} = \dot{n}C_p \frac{dT_C}{dz} \tag{6}
\]

After a series of transformations and integration, the equation can be simplified to \( A/(50 R_{KM} \dot{n}) = T_C^{-1}(0) - T_C^{-1}(z) \). We have assumed previously that \( T_s(z) = T_i = 0.7K \), but commonly \( T_C(0) \) is below 0.1K in the mixing chamber. According to \( T_C^{-1}(0) >> T_C^{-1}(z) \), a simplified equation on the relationship between the performance and the parameters of the counterflow heat-exchanger is given by

\[
T_C^{-1}(0) = 50 R_{KM} \dot{n} \frac{1}{A} \tag{7}
\]
2.4. Dilution process in the mixing chamber

As is shown in Figure 4, phase separation will occur in the mixing chamber when the temperature is below phase separation temperature.

But in the actual operation of the dilution refrigerator, the vaporized gas in the still cannot be pure $^3$He. Suppose the mole fraction of $^3$He in the vaporized gas is $x = n_3/n$ and in the diluted phase is $x_D = n_3/(n_3 + n_4)$, and then the mole fraction of $^4$He in the vaporized gas is $1 - x$ and in the diluted phase is $1 - x_D$. According to the conservation of energy, we get

$$\dot{Q}_m + \frac{x - x_D}{1 - x_D} n [h_c(T_c) - h_m(T_m)] + \frac{1 - x}{1 - x_D} (x_D n) [h_D(T_c) - h_m(T_m)] = \frac{x - x_D}{1 - x_D} n [h_D(T_c) - h_c(T_c)]$$

Under ideal circumstances, $x = 1$, Equation 7 can be simplified as

$$\dot{Q}_m + n [h_c(T_c) - h_m(T_m)] = n [h_D(T_m) - h_c(T_m)]$$

The temperature of the mixing chamber is usually below 0.1K when the dilution refrigerator is running stably. In this temperature range, $h_c(T_c) = 12 T_c^2$ and $h_D(T_m) = 96 T_m^2$ [12]. It can be obtained that the minimum temperature of the dilution refrigerator without heat load is $T_{mc} = T_c/2.8$ which depends on the performance of the heat exchanger in a sense.

3. Coupled analysis and results

The above analysis shows that the performance of the dilution refrigerator is closely related to the still, the counterflow heat-exchanger and the mixing chamber which have an inseparable influence on each other. Without a doubt, heat leakage from heat conduction, viscous heat and radiant heat also affect the dilution unit, but it can be minimized through rigorous design. The integrated relationship can be given by coupling the above model that

$$T_m^2 = 6.25 R_M \frac{n}{A} + 0.01 \frac{\dot{Q}}{n}$$

or
From equation 10, the relationship formula of the minimum temperature of the dilution refrigerator can be obtained. When there is no heat load, \( T_{mc}^2 = 6.25R_{KM} n/A \). It’s worth mentioning that heat leakage is ignored in this relationship. If a sleeve-type counterflow heat exchanger is used, where the heat exchange area is too large, it causes excessive viscous heat. However, the design of the sintered heat-exchangers can ensure sufficient heat exchange area while meeting the minimum temperature requirement.

Equation 11 converted from Equation 10, can directly show the relationship between the performance of the dilution refrigerator and the heat exchanger, evaporator and mixing chamber. For a certain dilution refrigerator design, when the thermal resistance and heat exchange area are constant, the cooling capacity is related to the flow rate and the minimum temperature. The optimal flow value (or saturated flow rate) at the maximum cooling capacity at different temperatures can be obtained by deriving the flow rate by the cooling capacity,

\[
\frac{d\dot{Q}}{dn} = -1250 \frac{R_{KM}}{A} n + 96T_{mc}^2 = 0
\]

Then the optimal flow value at different temperatures can be expressed as

\[
n = 0.0768T_{mc}^2 \frac{A}{R_{KM}}
\]

When the flow rate is higher than the optimal flow value, which leads the low gas purity, insufficient heat exchange, and large viscous heat leakage, the cooling capacity could decrease instead of increasing.

**Figure 5.** Ideal cooling capacity-temperature curve

**Figure 6.** Influence of \(^3\)He purity on cooling capacity

The ideal cooling capacity-temperature curve (\( n = 1 \times 10^{-4} \text{ mol/s} \)) is shown in Figure 5, with complete heat exchange and no heat leakage. Now for a specific dilution refrigerator, assuming that the material of the tube heat exchanger is CuNi alloy \((R_{KM} = 12 \times 10^{-3} \text{ K}^4 \text{m}^2/\text{W})\), the following analysis is based on this premise) [13], the influence of \(^3\)He purity on cooling capacity (\( n = 1 \times 10^{-4} \text{ mol/s} \) and the heat exchange area is 100 cm\(^2\)) is shown in Figure 6. The flow rate increases with the temperature rising. But the purity of \(^3\)He decreases subsequently, affecting performance. Therefore, the still temperature usually does not exceed 0.8 K.
Figure 7 shows the relationship between heat exchange area and the minimum temperature \((n = 1 \times 10^{-4} \text{ mol/s})\). It can be seen that the heat exchange area increases exponentially with the decrease of the minimum temperature. When the minimum temperature is lower than 20mK, the heat exchange area sharply becomes huge.

![Figure 7. Relationship between heat exchange area and the minimum temperature](image1)

Figure 8 shows Q-n-T Coupling influence relationship for a specific dilution refrigerator. The curve on the bottom plane represents the optimal flow value at the maximum cooling capacity at different temperatures. In order to facilitate observation, we choose \(T_{mc}=50 \text{ mK}\) and \(T_{mc}=75 \text{ mK}\) for analysis, as shown in Figure 9. Comparing the two figures, it can be seen that the cooling capacity depends on the flow and temperature, and the optimal flow value at the maximum cooling capacity increases with the increase of temperature. This is of great significance for the best performance under different working conditions.

![Figure 8. Q-n-T Coupling influence relationship for a specific dilution refrigerator](image2)

Figure 9. Saturated flow rate in the Q-n curve at \(T_{mc}=50\text{mK}\) and \(T_{mc}=75\text{mK}\)

![Figure 9. Saturated flow rate in the Q-n curve at \(T_{mc}=50\text{mK}\) and \(T_{mc}=75\text{mK}\)](image3)

When the flow rate is greater than the optimal flow rate, the cooling capacity does not increase but decreases, limited by the heat exchange area. As shown in Figure 10, this limitation continues to attenuate as the heat exchange area increases. That is, as long as the heat exchange area is large enough, the cooling capacity will always increase as the flow rate increases. The above results are based on the ideal condition \(x=1\) and our future plans will include analyses with \(x < 1\).

![Figure 10. Influence of heat exchange area on Q-n curve](image4)

4. Discussion
Certainly, too large heat exchange area is unnecessary, because it will bring other losses, such as viscous heat, a factor that cannot be ignored in extremely low temperature pipelines, given by
\[
\dot{Q}_{vis} = \dot{V} \Delta p = Z \eta \dot{v}^2 \dot{n}^2
\]  
(13)

Where \( Z = 128L / \pi D^4 \) represents flow resistance, \( \dot{V} \) the molar volume, \( \eta \) viscosity. The temperature rise caused by viscous heat is
\[
dT = \frac{d\dot{Q}_{vis}}{nc} = \frac{d(\dot{h}Z\eta \dot{v}^2)}{c}
\]  
(13)

Larger heat exchange area means longer pipeline, greater flow resistance and temperature rise. Assuming the \(^3\)He fluid channel diameter is 0.6 mm, Table 1 compares the outlet temperature rise and the outlet temperature of the heat-exchanger with viscous heat considered under different length of heat-exchanger.

Table 1 The outlet temperature rise caused by viscous heat

| Diameter(mm) | Length(m) | HE area(m²) | Z(cm⁻³) | n(mol/s) | \( T_{vis}(mK) \) | \( T_c(mK) \) |
|-------------|-----------|-------------|---------|---------|----------------|-------------|
| 0.6         | 6         | 0.011       | 1.86×10¹⁰ | 0.001   | 13             | 86          |
| 0.6         | 5         | 0.0094      | 1.55×10¹⁰ | 0.0001  | 11             | 91          |
| 0.6         | 4         | 0.0075      | 1.24×10¹⁰ | 0.0001  | 8.5            | 97.5        |
| 0.6         | 3         | 0.0057      | 9.3×10⁹   | 0.0001  | 6.3            | 109.3       |
| 0.6         | 2         | 0.0038      | 6.21×10⁹  | 0.0001  | 4.2            | 130         |
| 0.6         | 1         | 0.0012      | 3.1×10⁹   | 0.0001  | 2              | 180         |

Figure 11 Effect of viscous temperature rise on the performance of heat-exchanger

It can be seen from Figure 11 that as the pipeline lengthens, the outlet temperature decline trend becomes gentler, while the viscosity temperature rise continues to increase. For the 0.6 mm flow channel, the effect of the heat exchanger is longer than 6 meters. In this case, increasing the heat exchange area by increasing the length of the double-pipe heat exchanger cannot improve the performance of the dilution refrigerator.

To pursue a temperature of 30 mK or even lower, we must find another way. Sintered metal powder heat exchanger can avoid the above troubles. Sintered heat exchangers, usually made of copper powder or silver powder, can greatly reduce the effect of viscous heat. The specific surface area can reach more than 1, which can easily meet the demand for higher heat exchange area [12]. We will compare the two heat transfer methods through theory and experiment in the future.
5. Conclusion
Based on the conservation of energy and the Kapitza thermal resistance effect, the thermodynamic analysis of the dilution unit of the basic dilution refrigerator is carried out. The results reveal the thermodynamic laws inside the dilution refrigerator, especially the coupling influence relationship among refrigeration capacity, flow rate, heat exchange area, and minimum temperature, and the optimization direction in the design. The results show that corresponding to different heat exchange areas, a minimum temperature and an optimal flow rate can be obtained for each. Taking the optimal flow as the node, the cooling capacity first increases and then decreases as the flow increases.

In addition, we quantitatively analyzed the effect of viscous heat on the performance of the heat exchanger and its reasons, and gave the general application of the continuous heat exchanger. These results have an important guiding role for the design of dilution refrigerators. In the next step, we will compare the theoretical results through experimental research and analyze the main reasons why the cooling capacity decreases with the flow rate rising.

6. References
[1] S. Triqueneaux, L. Sentis, P. Camus et al. 2006 Design and performance of the dilution cooler system for the Planck mission. Cryogenics 46, 288–297
[2] P.A. Scholz, S. Kraft-Bermuth, V. 2016 Andrianov, Systematic vibration studies on a cryogen-free $^3$He/$^4$He dilution refrigerator for X-ray spectroscopy at storage rings. J. Low Temp. Phys. 184, 576–582
[3] F. Jessen, M. Knufinke, S.C. Bell et al. 2014 Trapping of ultracold atoms in a $^3$He/$^4$He dilution refrigerator. Appl. Phys. B 116, 665–671
[4] L. Duband, L. Hui, A. Lange. 1990 Space-borne $^3$He refrigerator. Cryogenics 30, 263–270
[5] B.W. Mueller, F.K. Miller. 2016 Development of a thermodynamic model for a cold cycle $^3$He–$^4$He dilution refrigerator. Cryogenics 79, 85–95
[6] J. Pradhan, N.K. Das, A. Chakraborty. 2013 Thermo-dynamical process simulation of dilution refrigerator. Cryogenics 57, 158–165
[7] R. Radebaugh, J. D. Siegwarth. 1971 Dilution refrigerator technology. Cryogenics
[8] J.G.M. Kuerten, C.A.M. Castelijns, T.A.M. de Waele and, H.M. Gijsman. 1985 Thermodynamic properties of liquid $^3$He–$^4$He mixtures at zero pressure for temperatures below 250 mK and $^3$He concentrations below 8%. Cryogenics 25:419-443
[9] Masaki NAKAMURA, Yoshiko FUJII, Toshinobu SHIGEMATSU. 1987 Solubility of $^4$He in Liquid $^3$He at Very Low Temperatures. Japanese Journal of Applied Physics
[10] Patrick Wikus, Tapio O, Niinikosko. 2010 Theoretical Models for the Cooling Power and Base Temperature of Dilution Refrigerators. J Low Tem Phys 158:901-91
[11] John C. Wheatley, R. E. Rapp, R. T. 1971 Johnson. Principles and methods of dilution refrigeration. II. Journal of Low Temperature Physics 4(1):1-39
[12] O. V. Lounasmaa. 1974 Experimental Principles and Methods Below 1 K. Academic Press.
[13] G.Frossati. 1992 Experimental Techniques: Methods for Cooling Below 300mK. J Low Tem Phys 87:595-633
[14] Ramiere, Aymeric, Sebastian Volz, and Jay Amrit. "Thermal resistance at a solid/superfluid helium interface." Nature materials 15.5 (2016): 512-516.
[15] Maris, Humphrey J., and T. E. Huber. "Kapitza resistance between liquid and solid helium. I. Theory." Journal of Low Temperature Physics 48.1 (1982): 99-109.

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