The boiling suppression of liquid nitrogen

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Abstract

When He gas is injected from room temperature into boiling liquid N\textsubscript{2}, boiling is suppressed, leaving liquid surface flat like a mirror. Although the qualitative explanation for this phenomenon is known [Minkoff G J et al. Nature 1957;180(4599):1413-4.], it has not been studied quantitatively and comprehensively yet. In this report, we made careful simultaneous measurements of temperature and weight variation of the liquid. The results clearly indicate that the boiling suppression is caused by cooling of the liquid with “internal evaporation” of N\textsubscript{2} into the He bubbles.

Key words: Nitrogen(B)

1 Introduction

Boiling liquid N\textsubscript{2} is a common and useful cryogen. However, the boiling does various harms to experiments. For example, it makes the liquid a non-uniform medium for optical experiments and generates vibrational disturbances to the

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system. The boiling can be stopped by injecting He gas from room temperature into the liquid. It was reported that the cause of this phenomenon is the “internal evaporation”, i.e., liquid N$_2$ is forced to evaporate into injected He bubbles until the evaporation pressure reaches the equilibrium [1,2]. Because the latent heat is deprived by the evaporation, the liquid is cooled down. He bubbles mixed with N$_2$ gas rise up to the air, and new bubbles are supplied one after another, lowering the liquid temperature. Boiling is suppressed by this temperature dropdown, not by the pressure dropdown, as shown by the arrow in the Fig. [1]. However, this scenario has not been examined quantitatively enough. We studied this phenomenon quantitatively and comprehensively, which confirmed the above-mentioned scenario unambiguously [3].

2 Methods

Let us describe our experimental setup and procedure. We used a double-walled glass dewar (inner diameter 150 mm, depth 240 mm) without silver-plating. When liquid N$_2$ is poured into the dewar, violent boiling happens first. After a while, the inner wall of the dewar is cooled down and liquid N$_2$ boils steadily as shown in the photograph (Fig. 2(a)). The bubbles are formed one after another at nucleation centers, such as scratches and dust on the inner wall surface. Then He gas was injected into the liquid N$_2$ through a stainless steel pipe (5 mm in diameter), whose end was placed near the bottom of the dewar. The flow rate of injected gas was measured by a flow meter with a 5 % precision. After the injection, the boiling is suppressed and the liquid surface becomes flat like a mirror as seen in the photograph (Fig. 2(b)). The suppression continues for a while and finally the boiling starts...
again. This phenomenon is reproducible, and the suppression time depends on the amount of injected He gas.

We used a carbon resistance thermometer (CRT) \[4\] to measure the change of liquid temperature. CRT is calibrated at the boiling (\(T_b\)) and triple (\(T_t\)) points of liquid N\(_2\). Between the two points, the resistance-temperature relation is interpolated by a linear function, giving the maximum calibration error of ± 0.2 K. The thermometer is located away from the pipe end in order to measure the mean temperature of liquid.

In order to confirm that the cooling of liquid is caused by the internal evaporation, we monitored the evaporation rate by measuring the liquid weight with an electronic scale as shown in the photograph (Fig. 2(c)). The precision of the electronic scale is ±0.1 g.

### 3 Results and Discussions

Figure 3 shows the measured temperature changes by the gas injection at flow rates of 1.0, 1.5 and 2.0 ℓ/min for 60 s. The liquid temperature drops down while He gas is blown. After stopping the injection, it rises back to \(T_b\) at a constant speed. With increasing the flow rate, the temperature drop becomes larger, resulting in longer boiling suppression. It was observed that the boiling restarts just when the temperature returns to \(T_b\) which is unchanged from that before the injection.

The measured time evolutions of the liquid weight are shown in Fig. 4. The data shown in Figs. 3 and 4 were taken simultaneously. Without the gas injection, liquid N\(_2\) evaporates steadily at a constant rate under a constant heat
flow into the dewar from the environment. The evaporation rate increases by a factor of 3-5 after the gas injection starts at $t = 30$ s. After the injection is stopped, the evaporation rate becomes smaller than that in the steady state because the ambient heat leak is absorbed by the cooled liquid $N_2$. It returns to the steady-state speed and the boiling restarts when the temperature comes back to $T_b$ at $t = 270$, 320 and 360 s for 1.0, 1.5 and 2.0 ℓ/min, respectively. This weight measurement demonstrates unambiguously that liquid $N_2$ is cooled by the forced evaporation.

Let us calculate the temperature drop ($\Delta T$) due to this internal evaporation using the data at the flow rate of 2.0 ℓ/min in Fig. 4. At $t = 90$ s, the excess weight loss of liquid $N_2$ beyond the natural evaporation is $\Delta W = 3 \times 10^{-2}$ kg. Since the latent heat per unit mass of $N_2$ is $L = 200$ kJ/kg, the heat taken away by the forced evaporation is $\Delta Q = L\Delta W = 6$ kJ. Specific heat of liquid $N_2$ ($c_{N_2}$) and glass ($c_G$) is 2 and 0.7 kJ/kg·K, and the mass of liquid $N_2$ in the dewar ($M_{N_2}$) and the inner wall of glass dewar ($M_G$) is 1.2 and 1 kg, respectively. Thus, the total heat capacity is $C = c_{N_2}M_{N_2} + c_GM_G = 3.1$ kJ/K. A part of $\Delta Q$ should be used to cool the injected He gas from 300 K to approximately 77 K. Such heat is calculated as $\Delta Q_{He} = c_{He}M_{He} \times (300 - 77) = 0.42$ kJ, where $c_{He}(= 5.2$ kJ/kg·K) is T-independent specific heat of He and $M_{He}(= 3.6 \times 10^{-4}$ kg) is the weight of the injected He gas. Therefore, the temperature drop is calculated as $\Delta T = (\Delta Q - \Delta Q_{He})/C = 1.8$ K. This is roughly in accordance with the measured temperature drop (=1.4 K) at $t = 90$ s, which demonstrates the relevance of the forced evaporation mechanism quantitatively.

Next we made temperature measurements with $H_2$, Ne and $N_2$ gases at a constant flow rate of 1.0 ℓ/min. It is difficult to conduct this experiment with
other kinds of gas with higher boiling points than N\textsubscript{2}, since they are liquefied into liquid N\textsubscript{2}. The results are shown in Fig. 5. The cooling rates during the gas injection of Ne and H\textsubscript{2} are almost the same as that of He. On the other hand, the temperature drop is not observed with N\textsubscript{2} injection. This shows that the role of injected gas is to increase the liquid surface area and to displace the evaporated gas to the atmosphere efficiently regardless of kind of injected gas.

For H\textsubscript{2} gas in Fig. 5, the temperature rises above \(T_b\) and suddenly returns to \(T_b\) at \(t = 340\) s, at which boiling restarts. This is overheating, which is sometimes observed in the case of less nucleation centers, i.e., when liquid N\textsubscript{2} is not contaminated by ice grains condensed from the atmosphere and the inner surface of the dewar is clean enough.

We found that the cooling power does not depend on the size of bubbles by changing the pipe diameter \((d = 5, 10, 15\) mm) with a fixed flow rate as shown in Fig. 6. This indicates that N\textsubscript{2} vapor pressure in the bubbles is saturated before arriving at the liquid surface. The N\textsubscript{2} boiling suppression was also observed by replacing the air above the liquid N\textsubscript{2} surface with He gas as was reported in Ref. [1], although the temperature drop is smaller than that caused by the injection.

Finally, we tried to lower the temperature of liquid N\textsubscript{2} by the forced evaporation method as much as possible. As shown in Fig. 1, the vapor pressure of N\textsubscript{2} at the triple point is as high as 1.25 \times 10^4\) Pa, and the temperature difference between \(T_t = 63.2\) K and \(T_b = 77.4\) K is only 14.2 K. Hence, in principle, it is possible to solidify N\textsubscript{2} by this method, which was demonstrated by the previous workers [2]. We used a metallic container rather than the transparent
dewar in order to decrease the ambient heat leak, and He gas was precooled before injection. Figure 7 is the result of this cooling experiment. Here, the flow rate is increased stepwise at \( t = 2240, 2890 \) and \( 3310 \) s indicated by the arrows in Fig. 7 up to \( 5.0 \ell/\text{min} \) at the end. We reached the lowest temperature of \( 63.7 \pm 0.2 \, \text{K} \), which is very close to \( T_t \) as shown in Fig. 7. The boiling was suppressed for more than 150 min after stopping the injection. Considering the fact that CRT was placed near the pipe end and the temperature drop was saturated, it is plausible that we reached the triple point actually. However, solid \( \text{N}_2 \) was not identified at least by eyes. This is presumably because the liquid was heavily contaminated with the ice dust.

4 Conclusion

We confirmed that the \( \text{N}_2 \) boiling suppression with He gas injection is caused by the liquid temperature drop due to the forced evaporation. This was made by measuring the liquid temperature and weight simultaneously. This phenomenon is practically useful for experiments where one has to minimize vibrational and optical disturbances.

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Fig. 1. The phase diagram of N$_2$. The liquid is cooled along the arrow.
Fig. 2. (a) Boiling liquid $\text{N}_2$. Bubbles are formed on the inner wall of the glass dewar. (b) After the injection of He gas. Boiling is suppressed and the surface is quiet like a mirror. (c) The experimental setup of the weight measurement of liquid $\text{N}_2$ by an electronic scale.
Fig. 3. The temperature changes of liquid $\text{N}_2$ when He gas is injected at a constant flow rate denoted. The gas injection starts at $t = 30$ s and ends at $t = 90$ s.
Fig. 4. The weight changes of liquid N\textsubscript{2} when He gas is injected at a constant flow rate denoted. The solid line is the weight change for the steady-state evaporation under the ambient heat leak.
Fig. 5. The temperature changes of liquid N\textsubscript{2} when various kinds of gas are injected at a constant flow rate of 1.0 ℓ/min.
Fig. 6. The weight changes of liquid \( \text{N}_2 \) when He gas is injected through pipes of different diameters \( (d) \). The flow rate is fixed at 1.5 \( \ell/min \).
Fig. 7. The attempt to cool liquid \( \text{N}_2 \) as much as possible, using a metal dewar. The flow rate is increased stepwise at the times indicated by the arrows. The highest flow rate is 5.0 ℓ/min.