

# EFFECT OF DISSOLVED GAS ON BUBBLE NUCLEATION

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**Abstract**—The aim of this research is to make an experimental study of an effect of a dissolved gas on the liquid–gas homogeneous phase transition, and to compare the result with a bubble nucleation prediction.

The experiment was performed under the condition of no existence of gas bubble in a liquid. The superheat limit of the liquid was experimentally studied using a technique in which a droplet containing a dissolved gas was suspended in an immiscible fluid medium. The result shows that with an increase of the content of a dissolved gas homogeneous nucleation occurs at lower temperatures and in the presence of a sufficient dissolved gas at pressures above the saturation pressure of the pure solvent.

## NOMENCLATURE

$J$ ,	velocity of nucleation [ $1/m^3 \cdot s$ ];
$k$ ,	Boltzmann constant [ $J/K$ ];
$K$ ,	solubility coefficient [ $1/Pa$ ];
$m$ ,	mass per one molecule [ $g$ ];
$N$ ,	molecule number per unit volume in liquid;
$p$ ,	pressure [ $Pa$ or $atm$ ];
$q$ ,	molar latent heat [ $J$ ];
$r$ ,	radius of bubble [ $m$ ];
$T$ ,	temperature [ $^{\circ}C$ , $K$ ];
$x$ ,	mole fraction;
$\delta$ ,	surface tension [ $N/m$ ].

## Subscripts and superscripts

1,	liquid phase of droplet;
2,	noncondensable gas phase;
'	gas phase;
"	liquid phase;
c,	critical point.

## 1. INTRODUCTION

GENERATION of gas phase in the liquid phase caused by boiling, cavitation or such is one of the most important phenomena in studying thermal engineering and has been studied extensively from various standpoints. The study on the formation of the initial bubble is particularly important in clarifying the boiling cycle as a fundamental study on boiling or in making clear the minimum heat flux obtained from boiling, and is also essential in clarifying thermal explosion phenomena of liquid natural gas.

Studies on the formation of the initial bubble have been made mostly on the phenomena in the structure of two limit cases of boiling [1, 2] at the cavity or on the solid surface and of boiling [3–5] caused by homogeneous nucleation.

However, as to the formation of the initial bubble which is the most fundamental one in the boiling phenomena, it has been reported [6, 7] that the remaining bubble, dissolved gas, small floating solids, ions, cosmic rays etc. are possible to become the cause for originating bubble nucleations and that especially the effect due to the dissolved gas on the phase change

from liquid to gas is remarkable, on which many discussions have been made so far. However, it has not been made clear how the noncondensable gas gives effects on the phase change of boiling or such. For instance, in the study of cavitation, the effect by a dissolved gas is only taken up with the air content as parameter.

In order to clarify the effects extended by the remaining bubble and the dissolved gas on the liquid–gas phase change, it is required to obtain conditions of bubble formation from the liquid from experiments with accurate measurements of the volume of the dissolved gas and the existence of remaining bubbles. For instance, in experimental studies on formation of initial bubbles made so far, it is very difficult to make an accurate experimental study on the existence and size of the gas bubble kept in the small cavity artificially prepared on the wall surface of the container. Therefore, in order to examine behaviors of small bubbles in the liquid, it is necessary to make an experiment under the condition that a theoretical analysis can be made and, moreover, that the behavior of bubbles can be clearly observed.

Authors have reported in the other papers [8, 9] on the results of theoretical and experimental studies on the thermodynamical stability condition of the system consisting of a small insoluble droplet floating in a sufficient volume of liquid and a small bubble containing a noncondensable gas kept in this droplet. As a result, it is made clear that a gas bubble of a diameter smaller than a certain critical value does not exist in the liquid under thermal equilibrium condition, and that when the pressure exceeds a critical value, the gas phase of the bubble is dissolved and the bubble disappears. In the droplet system containing this dissolved gas, the volume of the dissolved gas in the droplet can be given accurately when the gas existed in the bubble, and as mentioned before, if a homogeneous nucleation is made from this condition, a quantitative experiment to clarify the effect by the dissolved gas extended on nucleation can be made. As mentioned before, there has been no experimental study on this problem, or on how the dissolved gas gives effects on the liquid–gas phase change. Moreover, although there are theoretical

or predictive studies [10–12] on the effect by a non-condensable gas on the superheat limit, that is, the homogeneous nucleation, no experimental study has been seen.

In our present report, as a fundamental study to investigate the effect made by the dissolved gas on to the liquid–gas phase change such as boiling and cavitation etc., the gas bubble in the droplet is made to dissolve and extinct, and then an experimental investigation is made on the possibility of bubble formation in the droplet containing the dissolved gas, and on the effect made by the noncondensable gas (dissolved gas) on the superheat limit (supersaturation limit) of liquid.

## 2. EXPERIMENTAL APPARATUS AND METHODS

Figure 1 is the whole diagram of the experimental apparatus. The experimental apparatus consists of mixing section and bubble formation section, droplet formation section (bubble extinction section), and test

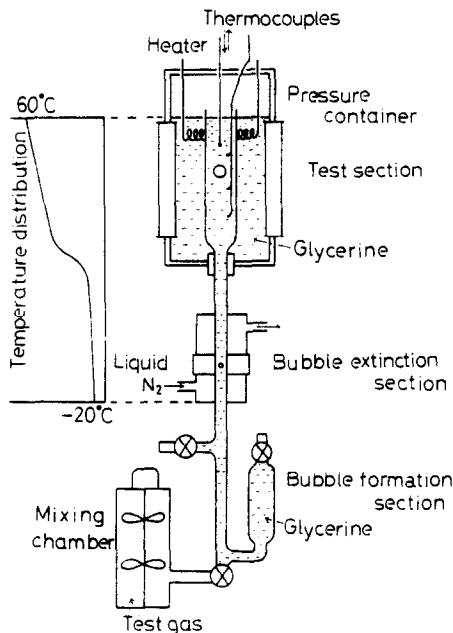


FIG. 1. Experimental apparatus.

section (nucleation section). The test section is 20 mm in inner diameter and 200 mm in length, and the droplet formation section and the nucleation section are incorporated with a pyrex-glass pipe of 4 mm I.D. and 400 mm length. The test section is placed in a pressure container for increasing pressure. At the mixing section,  $\text{CO}_2$  was added as the noncondensable gas to the vapor of test materials (freon 22, propane, isobutane) to make a mixture gas which was sent as a bubble from the lower part of the droplet formation section. The droplet formation section was cooled down to between  $-20$  and  $-30^\circ\text{C}$  with liquid nitrogen, making a droplet containing a noncondensable gas bubble. The glass pipe and the pressure container of the test section were filled with glycerine. Heaters and thermocouples were installed in glycerine in the pressure container to obtain a given temperature distribution

in the test section. A movable thermocouple was inserted from the upper part of the test section in order to obtain temperature distributions in the test section. The test section in the pressure container was heated at the upper part and was provided with a constant and linear temperature gradient to the vertical direction.

The droplet containing a gas bubble in the droplet formation section was pressurized above the bubble extinction pressure and then a droplet in which the gas was completely dissolved was made. This droplet after bubble extinction was sent by buoyancy from the bubble extinction section to the test section. The droplet rising with a slow speed was heated by the surrounding liquid and the temperature of the droplet rose gradually, and bubble formation by homogeneous nucleation occurred at a point in the test section. Temperature of this bubble formation position was obtained from the temperature distribution in the test section. This temperature is considered to be the temperature of supersaturation limit of the droplet. The accuracy of temperature of droplets and the gas volume in the droplet is discussed below. A rough illustration as to behaviors of the droplet system in the experimental apparatus mentioned above is shown in Fig. 2.

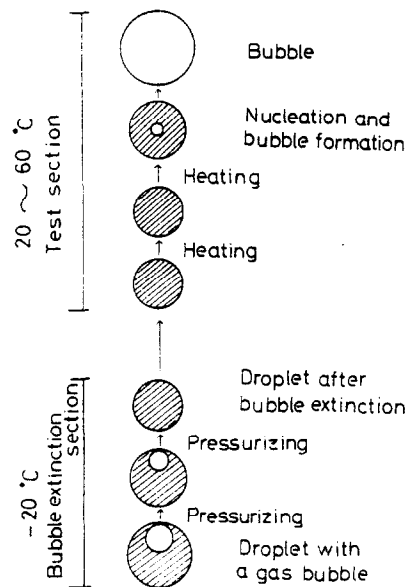


FIG. 2. Schematic pattern of experimental process.

### 2.1. Selection of liquid

In order for the dissolved gas in the droplet not to diffuse through the droplet surface while the droplet is rising in the test section and for the temperature of the droplet to approach the temperature of the surrounding liquid, the rising velocity of the droplet must be well controlled. Therefore, in the present study, when measurement is made for the supersaturation limit of tested liquids by the use of the floating droplet method [13], the following conditions are required to select the surrounding liquid which has the temperature distribution in the vertical direction:

1. The dissolved gas in the droplet should not be

diffused in the surrounding liquid through the droplet surface.

2. Boiling should be avoided on the liquid-liquid interface of the tested and the surrounding liquids.

3. The temperature of the droplet should rise slowly enough as to be in thermal equilibrium with the surrounding liquid.

In order to meet the conditions mentioned above, a liquid of high viscosity and of low diffusive coefficient should be adopted as the surrounding liquid. Also, a combination of liquids of which liquid-liquid surface tension is large enough comparing with the surface tension of the test liquid (droplet) should be selected. As the surrounding liquid to satisfy these conditions, glycerine below 60°C was selected as the most appropriate. As tested liquid (droplet), considering the easiness of experiment and the convenience of experimental apparatus, hydro-carbons and halogen compounds, of which supersaturation limit will be below 60°C, are investigated as candidates. Considering that carbon dioxide is soluble into droplet and the physical property is fairly well known, and the physical property of liquid in which carbon dioxide is dissolved is known to a certain extent within the above mentioned temperature region, and also considering the relation between saturation pressure and temperature, freon 22, propane of 99.7% purity, isobutane of 99.9% purity were used. The major physical properties of liquids tested in the experiment are shown in Table 1.

Table 1

Liquid	Freon 22	Propane	Isobutane
Critical pressure (MPa)	4.91	4.25	3.65
Critical temperature (°C)	96.4	96.8	134.9
Boiling point (°C)	-40.8	-42.1	-11.7
Superheat limit (°C at 1 atm)	57	56	86

### 2.2. Size of droplet and temperature distribution in the test section

In order to raise the temperature accuracy of the droplet and also the accuracy of the gas volume in the droplet, the size of the droplet and the temperature gradient of the test section are determined by satisfying the following conditions.

1. *Accuracy of the gas volume in the droplet.* As this is influenced by the ratio of the time for the gas in the droplet to take to leak into the surrounding liquid to the residence time of the droplet in the test section, this value is to be made sufficiently large.

2. *Temperature accuracy of droplet.* As this is given by the ratio of the temperature relaxation time to the reciprocal of the product of the temperature gradient in the test section and the rising velocity of the droplet, this ratio should be made sufficiently small.

The volume of noncondensable gas diffusing into the surrounding liquid through the liquid-liquid interface

of the droplet is estimated to be about  $10^{-11}$  mol after 20 min, and the whole mole number of the noncondensable gas contained in the droplet system is  $10^{-6}$ – $10^{-7}$  mol. Therefore, the noncondensable gas in the droplet system actually does not leak into the surrounding glycerine. In the present experiment, the size of the droplet was 0.4–0.8 mm in diameter, and the temperature gradient was 1–2°C/cm. In this case, the measurable temperature accuracy is about  $\pm 1.5^\circ\text{C}$ , and the accuracy of the gas volume in the droplet is about 1%.

### 2.3. Possibility of bubble formation on the liquid-liquid surface

Possibility of bubble formation on the liquid-liquid surface is checked using Apfel's equation [14] with respect to nucleation at the liquid-liquid interface. According to Apfel, the nucleation on the liquid-liquid interface may be given by the following equation:

$$\left\{ \frac{16\pi Z(T)^3}{3kT(p'-p'')^2} \right\} \sim 30-60$$

where

$$Z(T) = \left\{ \frac{\sigma_1^3 + \sigma_2^3}{2} + \frac{\sigma_{12}^3}{16} + \frac{3\sigma_1^2\sigma_2}{8\sigma_{12}} - \frac{3\sigma_{12}(\sigma_1^2 + \sigma_2^2)}{8} - \frac{3(\sigma_1^4 + \sigma_2^4)}{16\sigma_{12}} \right\}^{\dagger}$$

where  $\sigma_{12}$  expresses the liquid-liquid surface tension.

Also, a homogeneous nucleation inside of the liquid is given by the following equation [14, 15]

$$\left\{ \frac{16\pi\sigma_1^3}{3kT(p'-p'')^2} \right\} \sim 45-70.$$

The energy required for heterogeneous nucleation at the liquid-liquid interface the propane-glycerine system used in this experiment is calculated to be about four times as much as that for a homogeneous nucleation inside the liquid. Therefore, as in the present experiment, if a combination of the surrounding liquid and the tested liquid is so selected as the surface tension of the surrounding liquid is sufficiently larger than that of the tested liquid, homogeneous nucleation in the droplet would be realized with much higher probability than heterogeneous nucleation, and a homogeneous nucleation inside the droplet would occur before the generation of nucleation at the interface. Therefore, the supersaturation limit obtained from the bubble nucleation of the droplet can be understood as homogeneous nucleation.

### 3. EXPERIMENTAL RESULTS

In this chapter, according to the experimental methods mentioned above, the results of measurements made on the superheat limit (supersaturation limit) of the droplet in which a noncondensable gas is completely dissolved are shown.

In Fig. 3, the pressure-temperature relations of the two component mixture system are shown for the propane-CO<sub>2</sub> and the superheating limit line of propane are also shown. The dotted chain line in the figure

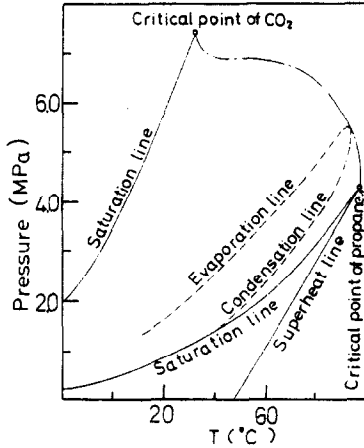


FIG. 3. Pressure-temperature relations of propane-CO<sub>2</sub>.

expresses the critical line of the mixture system, and the evaporation and condensation curves of constant composition of CO<sub>2</sub> are expressed with oblique broken lines.

The measured superheat limit of the pure liquid should be compared with the theory, and the experiments [3, 4] on superheat limit of hydrocarbons in the range of 100–200°C have so far been known. As shown later, it was confirmed in the present study that the superheat limit of the pure liquid agreed very well with the theoretical prediction of a homogeneous nucleation by Volmer-Döring [15]. However, no experiment on homogeneous nucleation when a dissolved gas is contained in the liquid has been known.

In order to provide an understanding of the procedure and scheme of the present experiment, the bubble extinction region and the homogeneous nucleation region are shown, together with the superheat limit line of the pure component in Fig. 4. The nucleation of this droplet in which a noncondensable gas is completely dissolved is on the left side of the superheat limit of the pure liquid.

The experimental result of the supersaturation limit of freon 22 liquid in which CO<sub>2</sub> gas is dissolved is shown in Fig. 5. The vertical axis in the figure shows the pressure of the system, while the horizontal axis

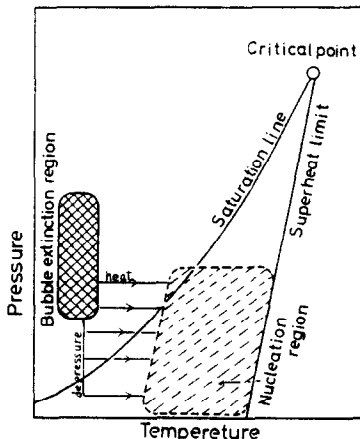


FIG. 4. Experimented regions.

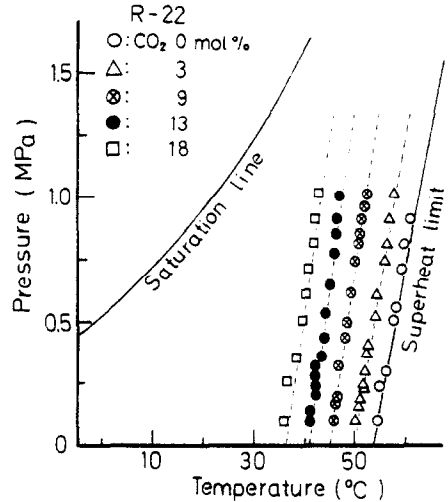


FIG. 5. R-22 with CO<sub>2</sub> dissolved.

indicates the temperature when the nucleation occurred, and the measured points are shown with the CO<sub>2</sub> concentration in mol% as parameter. The white circle in the figure is the experimental value of the superheat limit of pure freon, and the solid line crossing the white circles is the theoretical curve of the superheat limit of pure freon 22 by Volmer-Döring's [15] theory. In the figure, four typical experimental conditions are chosen as shown on the left top of the figure. Also, in the figure, the saturation pressure curve of pure freon 22 is shown in a solid line. By adding CO<sub>2</sub>, the supersaturation temperature of liquid falls down by about 10°C, and rises about 15 atm in pressure. With increase of CO<sub>2</sub> concentration, it is supposed that there exists a possibility of bubble formation also on the saturation line of the pure substance. Therefore, in order to investigate whether the supersaturation limit crosses the saturation line of the pure substance by increasing the CO<sub>2</sub> concentration or by changing the combination of tested liquid and CO<sub>2</sub>, an experiment was made on isobutane.

The experimental result on the supersaturation limit of isobutane liquid in which CO<sub>2</sub> is completely dissolved is shown in Fig. 6. The solid line on the right

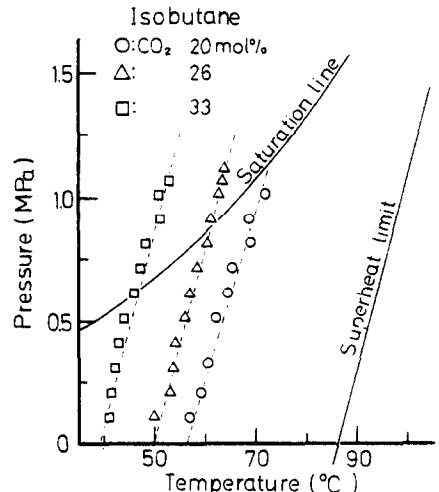
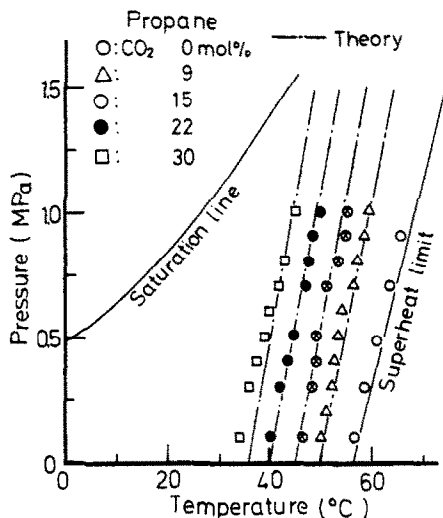


FIG. 6. Isobutane with CO<sub>2</sub> dissolved.

side of the figure is the superheat limit of pure isobutane theoretically calculated by Volmer-Döring theory. The solid line on the left side in the figure shows the saturation pressure curve of pure isobutane. As shown in Fig. 6, it is found from the experiment that the dissolved CO<sub>2</sub> gives a great effect on the supersaturation limit of isobutane. Especially the supersaturation limit of the liquid in which CO<sub>2</sub> is dissolved crosses over the saturation pressure curve of the pure substance. This interesting and important result suggests a possibility of bubble formation by homogeneous nucleation in the liquid containing a dissolved gas, even at the saturation condition.

For the purpose of comparing our results with a theory on homogeneous nucleation in the dilute solution stated later, the supersaturation limit of the liquid of the propane-CO<sub>2</sub> system was measured. This is because there exists an experimental value on solubility of CO<sub>2</sub> in liquid propane and a comparison with the theory is possible. The experimental result of the effect of CO<sub>2</sub> on the supersaturation limit of liquid propane is shown in Fig. 7. The white circles in the figure are



the experimental values of the pure propane, and the solid line on the right side of the figure is the calculation value obtained by Volmer-Döring's theory. In the figure, four theoretical lines for the respective experimental conditions are shown. The solid line on the left side of the figure is the saturation pressure curve of pure propane. When CO<sub>2</sub> is added, the supersaturation limit moves to the lower temperature side almost in parallel with the supersaturation limit of pure liquid propane.

#### 4. DISCUSSIONS

No theoretical study on homogeneous nucleation in liquid containing dissolved gas has been seen. Only Ward [10] uses the equation of homogeneous nucleation in the single component liquid suggested by Volmer-Döring [15], and performs a theoretical analysis, assuming that the effect of the dissolved gas is extended only to the pressure in the critical bubble

nucleus. That is, the pressure in the critical bubble nucleus is given as the sum of vapor pressure of pure liquid and partial pressure of dissolved gas in liquid, but the effect of the surface tension by the dissolved gas and the change of rate speed of nucleation by the dissolved gas are not taken into consideration. According to Ward's result, the speed of homogeneous nucleation in which a gas is dissolved is given by the following equation:

$$J = N \exp\left(\frac{-q}{kT}\right) \left\{ \frac{6\sigma_1}{(3-b)\pi m} \right\}^{\frac{1}{2}} \exp\left\{ \frac{-16\pi\sigma_1^3}{3kT(p'_1 + p'_2 - p'')^2} \right\}$$

where  $b = (p'_1 - p'')/p'_1$  and  $p'_1$  is the vapor pressure in the critical bubble nucleus,  $p'_2$  the partial pressure of noncondensable gas in the critical bubble nucleus. According to Henry's law,  $p'_2$  is given by the gas volume dissolved in the liquid. In order to calculate the supersaturation limit of the liquid in which gas is dissolved according to the above equation, the solubility of gas in the liquid should be given. There are no data for CO<sub>2</sub> gas solubility in liquid freon 22 and isobutane, but the solubility of CO<sub>2</sub> in liquid propane has been reported by Poettmann, Reamer and Schneider [16-18]. Therefore, here a comparison of our results with the theoretical analysis only for propane was made.

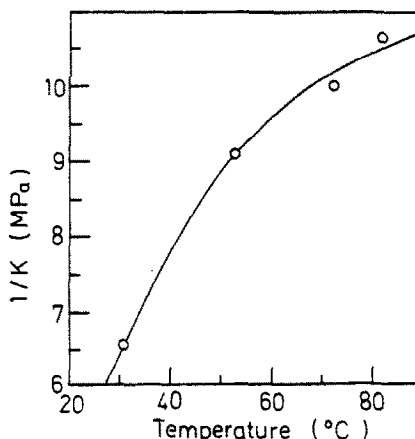


FIG. 8. Solubility coefficient.

In Fig. 8, the temperature dependency of CO<sub>2</sub> gas solubility coefficient in liquid propane is shown. The white circles are calculated from Poettmann's experimental values. Poettmann's experimental results show that CO<sub>2</sub> solubility is almost proportional to the pressure up to about 30% of CO<sub>2</sub> concentration. Therefore, the solubility coefficient can be considered constant up to about 30% of CO<sub>2</sub> concentration, and Henry's law is found applicable. In the calculation of theoretical curves, the following equation was used for the surface tension:

$$\sigma = 55 \times 10^3 \times (1 - T/T_c)^{1.22}, \text{ N/m.}$$

This equation was obtained by the extrapolation of the measured values reported in API table [19] (-84 to -40°C) up to the critical temperature.

The result of calculation regarding the effect of CO<sub>2</sub> on the supersaturation limit of liquid propane accord-

ing to Ward's theory is shown in Fig. 7 in one dotted chain lines. With increase of the dissolved gas concentration, the superheat limit moves to the low temperature side, and a good agreement is seen between the theoretical prediction and our experimental results. With the further increase of dissolved gas, it can be considered that the supersaturation limit line should cross over the saturation vapor pressure of the pure propane as in the case of isobutane, but due to the safety of pressure vessel, no experiment has been done for this purpose in regard to propane. This predicts that with the decrease of pressure for a given temperature or with the increase of temperature for a given pressure by an appropriate combination of a liquid and a dissolved noncondensable gas, the bubble formation by homogeneous nucleation begins before reaching the saturation pressure line of a pure substance. In the present experiment, though the experimental result agrees with the theoretical prediction stated above, there might leave some doubts as to whether the theory on homogeneous nucleation of pure liquid could be applicable to the case when a noncondensable gas is dissolved. A new development of a theory in this field is greatly expected.

#### 5. CONCLUSION

With respect to homogeneous nucleation in connection with boiling and cavitation, the effect of noncondensable gas was experimentally made clear when a bubble nucleus does not exist in the liquid phase with the gas completely dissolved in. A special attention was paid to the accurate measurements of the amount of gas dissolved and the temperature.

The superheat limit of pure liquid not containing a dissolved gas agrees with Volmer-Döring's theory, and the supersaturation limit of liquid containing a dissolved gas does decrease with the content of the gas. With the increase of dissolved gas concentration in the liquid, the supersaturation limit line crosses over the saturation pressure curve of the pure liquid substance, and even at pressures larger than the saturation pressure, bubbles are generated by homogeneous nucleation. The experimental result agrees with the Ward's theory concerning the superheat limit.

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#### EFFET DES GAZ DISSOUS SUR LA NUCLEATION DES BULLES

**Résumé**—Le but de la recherche effectuée est une étude expérimentale de l'effet des gaz dissous sur le changement de phase homogène liquide-vapeur, et la comparaison des résultats obtenus aux prévisions de nucléation des bulles.

L'expérience a été réalisée en l'absence de bulles gazeuses dans le liquide. La limite de surchauffe du liquide a été étudiée expérimentalement à l'aide d'une technique qui permet de maintenir en suspension dans un milieu fluide non miscible une gouttelette contenant un gaz dissous. Les résultats montrent que si la quantité de gaz dissous est augmentée la nucléation homogène se produit à des températures plus basses et qu'en présence d'une quantité suffisante de gaz dissous elle se produit à des pressions supérieures à la pression de saturation du solvant pur.

## DER EINFLUSS GELÖSTER GASE AUF DIE BLASENENTSTEHUNG

**Zusammenfassung**—Das Ziel dieser Forschungsarbeit ist eine experimentelle Studie des Einflusses gelöster Gase auf den homogenen Phasenwechsel Flüssigkeit—Dampf sowie der Vergleich der Ergebnisse mit Vorhersagen mit Hilfe der Keimbildungstheorie. Die Versuchsbedingungen waren so, daß sich keine Gasblasen in der Flüssigkeit befanden. Die Überhitzungsgrenze der Flüssigkeit wurde experimentell untersucht; dabei wurde ein Verfahren benutzt, bei dem ein Tropfen, der gelöstes Gas enthält, in ein nicht mischbares Fluid getaucht wird. Die Ergebnisse zeigen, daß mit zunehmendem Gehalt an gelöstem Gas die Temperatur, bei der eine homogene Keimbildung eintritt, niedriger wird. Bei Vorhandensein von ausreichend gelöstem Gas tritt die Keimbildung schon bei Drücken oberhalb des Sättigungsdruckes des reinen Lösungsmittels auf.

ВЛИЯНИЕ РАСТВОРЕННОГО ГАЗА НА ОБРАЗОВАНИЕ ЗАРОДЫШЕЙ  
ПРИ ПУЗЫРЬКОВОМ КИПЕНИИ

**Аннотация** — Целью данной работы являлось экспериментальное исследование влияния растворенного газа на переход жидкость—пар в гомогенной фазе. Полученные результаты сравнивались с расчетными данными по образованию зародышей при пузырьковом кипении. Эксперимент проводился при отсутствии пузырьков газа в жидкости. Предел перегрева жидкости изучался экспериментально с помощью метода, при котором капля с содержащимся в ней растворенным газом помещалась в несмешивающуюся жидкую среду. Найдено, что с увеличением содержания растворенного газа образование зародышей в гомогенной фазе происходит при пониженных температурах и при наличии достаточного количества растворенного газа при давлении, превышающем давление насыщения чистого растворителя.