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SUPERHEATING IN NUCLEATE BOILING CALCULATED BY THE HETEROGENEOUS NUCLEATION THEORY†

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Abstract—With the heterogeneous nucleation theory the superheating of the liquid boundary layer in nucleate boiling is described not only for the onset of nuclear boiling but also for the boiling crisis. The rate of superheat depends on the thermodynamic stability of the metastable liquid, which is influenced by the statistical fluctuations in the liquid and the nucleation at the solid surface. Because of the fact that the cavities acting as nuclei are too small for microscopic observation, the size and distribution function of the nuclei on the surface necessary for the determination of the probability of bubble formation cannot be detected by measuring techniques. The work of bubble formation reduced by the nuclei can be represented by a simple empirical function, whose coefficients are determined from boiling experiments. With that the heterogeneous nucleation theory describes the superheating of the liquid, which was checked on several fluids like refrigerants, liquid gases, organic liquids and water.

NOMENCLATURE

A_c,	activation energy;
A_{c0},	activation energy for a spherical bubble;
b,	pressure ratio;
C,	constant;
J,	probability density;
J^*,	probability;
k,	Boltzmann constant;
$k_0 = A_c/A_{c0}$,	ratio of activation energies;
K,	coefficient of the k_0-function;
M,	mass per molecule;
p,	pressure;
p_s,	saturation pressure;
r_c,	critical bubble radius;
r_L,	mouth radius of cavity;
T,	temperature;
T_c,	critical temperature;
T_s,	saturation temperature;
V,	volume;
Z,	number of molecules per unit volume.

Greek symbols

β,	aperture angle;
γ,	exponent of the k_0-function;
Δ,	difference;
$\varepsilon = \dfrac{T_c - T}{T_c}$,	reduced temperature difference;
θ_0,	contact angle;
μ,	exponent of surface tension;
σ,	surface tension;
σ_0,	surface tension coefficient.

1. INTRODUCTION

BOILING can only occur in the superheated liquid layer of a surface. The amount of superheating ranges from a few hundredth of a Kelvin at normal atmospheric pressure up to several hundred Kelvin for liquid metal boiling at high heat flux.

From the thermodynamic point of view the superheated liquid layer is in a metastable state. This implies that the liquid may exist in single phase but a certain amount of disturbances may cause a change in phase and the stable two phase state will be achieved. Figure 1 demonstrates a pressure vs density plot of CO_2. The metastable isotherms in the two phase region are calculated by extrapolation of an equation of state [1]. The unstable region between the maxima and minima of the isotherms is bordered by the so called Spinodale line. The region between the coexistence line and the Spinodale line is the metastable region, in which phenomenological thermodynamics predict the existence of the metastable state up to the Spinodale line. With increasing distance from the coexistence line the probability that infinitesimal disturbances are able to cause a change of phase increases. Such disturbances are caused by statistical molecular fluctuations of local density, pressure and entropy and determines the homogeneous nucleation limit. In a real case the breakdown of the metastable state is anywhere between the homogeneous nucleation limit and the coexistence line depending on the solid surface or other effects acting as a nuclei.

2. HOMOGENEOUS NUCLEATION LIMIT

In the case of homogeneous nucleation limit an ideal pure liquid is assumed without effects of a solid surface, impurities or high energetic radiation. Disturbances are produced only by spontaneous

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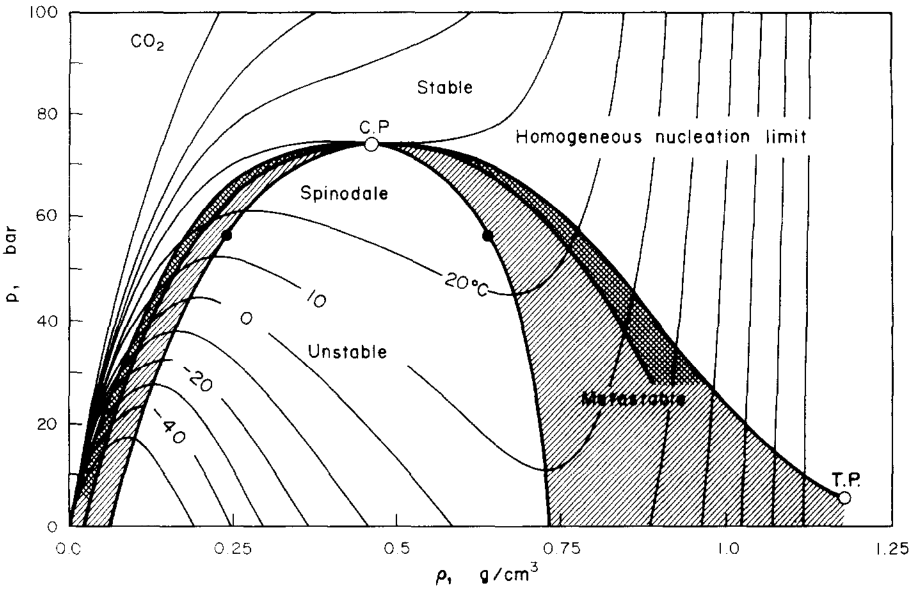

FIG. 1. p,T-plot of CO_2. The isotherms are calculated with the equation of state by Meyer-Pittroff [1]. In the two phase region the isotherms are extrapolated.

molecular fluctuations. From thermodynamic stability criteria it follows that a spherical bubble with critical radius r_c is stable in a metastable liquid, if equation (1) is satisfied:

$$r_c = \frac{2\sigma}{\Delta p}. \tag{1}$$

In this equation σ is the surface tension and $\Delta p = p_s - p$, the isothermal pressure difference between the saturation pressure p_s and the liquid pressure in the metastable state. If a bubble with a radius smaller than r_c is produced by a spontaneous fluctuation the bubble collapses, if a larger bubble is produced it is able to grow to macroscopic size. To produce an initial large bubble with r_c in a certain metastable state in a reversible isothermal way the critical activation energy A_{c0} is necessary, which results from a volume and a surface work term:

$$A_{c0} = \frac{16 \cdot \pi \cdot \sigma^3}{3 \cdot \Delta p^2}. \tag{2}$$

The amount of A_{c0} necessary depends on the surface tension σ, which is a function of temperature, and of the distance between the equilibrium state at saturation and the existing metastable state defined by Δp. The homogeneous nucleation theory assumes that the critical activation energy can be produced in a small volume only by an appropriate molecular fluctuation, which forms a void defect in the liquid lattice structure large enough for a spherical bubble with radius r_c. The probability that such a process takes place in a metastable liquid is given by the Boltzmann law:

$$J = C \cdot \exp(-A_{c0}/kT). \tag{3}$$

Here J is the probability per volume V and time t, the probability density, k is the Boltzmann constant and T the absolute temperature. Volmer [2] calculated the coefficient C in equation (3), which depends on the state of the liquid, using statistical mechanics as:

$$J^* = J \cdot V \cdot t = V \cdot t \cdot Z \cdot \left[\frac{6\sigma}{(3-b)\pi M}\right]^{1/2} \times \exp\left(-\frac{16\pi\sigma^3}{3kT\Delta p^2}\right). \tag{4}$$

J^* is now the probability of the existence of a critical bubble in the volume V and during the time interval t. The metastable liquid is defined by $\Delta p = p_s - p$. Z is the number of vapour molecules per unit volume, b is the pressure difference over saturation pressure $\Delta p/p_s$ and M is the mass per molecule.

If the probability J^* is equal to unity an initial critical bubble may be produced anywhere inside the metastable liquid. In this case equation (4) describes the homogeneous nucleation limit and the isothermal pressure difference Δp can be determined. With the vapour pressure curve the maximum superheating temperature at constant pressure p of a metastable liquid can be calculated as:

$$\Delta T_{\max} = T_s - T = \int_p^{p_s} \left(\frac{dT}{dp}\right)_s dp. \tag{5}$$

If J^* is less than unity no bubble of critical size is formed, the liquid still remains in the metastable state, but if J^* is larger than unity more bubbles can be produced in a certain time t.

Comparing results of boiling experiments with the homogeneous nucleation theory the measured superheating temperatures are much lower than those calculated from this theory; as this theory is based on idealistic assumptions, such as an ideal pure fluid which is not effected by nucleations from the solid surface, impurities or high energetic radiation. In reality, however, the liquid is surrounded by walls and is in direct contact with the heating surface on which small cavities act as nucleation sites and reduce the energy required for the formation of bubbles. It will be shown later that only near the critical point can bubble creation be described by the homogeneous nucleation theory.

3. HETEROGENEOUS NUCLEATION THEORY

During normal boiling processes the bubbles are formed always in cavities at the heating surface. Even in this case the thermodynamic stability criteria for the bubbles in equilibrium with the surrounding metastable liquid must be met and for the creation of the initial bubble, it must still have the critical radius r_c. If the bubble is created in a cavity, less volume and surface work is necessary than in the case of a spherical bubble with the same radius.

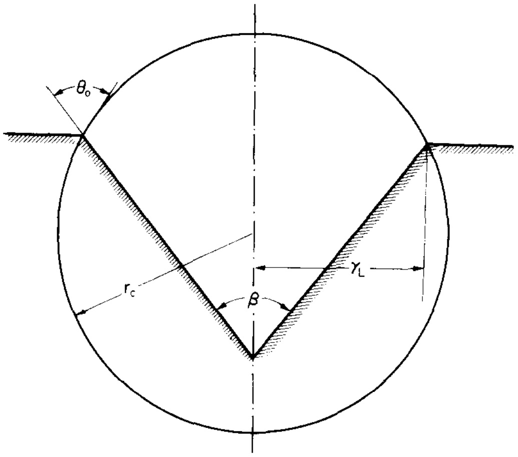

FIG. 2. Idealized conical cavity with a bubble. The cone has aperture angle β and mouth radius r_L. The bubble has radius r_c. The contact angle between liquid and wall is θ_0.

This is demonstrated with Fig. 2. The cavity has the mouth radius r_L and the aperture angle β, and the wetting angle θ_0 exists between liquid and solid surface. If it is assumed that the cavity is an ideal cone, Kottowsky [3] has determined a factor k_0 by geometrical considerations only. It is the ratio of surface and volume work A_c of a bubble created in the cavity to the work required for a spherical bubble A_{c0} of the same radius:

$$k_0 = A_c/A_{c0} = 1/4\cdot[2-3\sin(\theta_0-\beta/2) + \sin^3(\theta_0-\beta/2)]. \quad (6).$$

For the case of the heterogeneous nucleation the probability of creation of a critical bubble can be obtained by introducing the factor k_0 in equation (4):

$$J^* = V\cdot t\cdot Z\cdot\left[\frac{6\sigma}{(3-b)\pi M}\right]^{1/2} \times \exp\left(-k_0\frac{16\pi\sigma^3}{3kT\Delta p^2}\right). \quad (7)$$

Since the value of k_0 can only be between zero and unity, the activation energy for heterogeneous nucleation is always smaller than for the homogeneous case and the probability J^* increases. Therefore the influence of the solid surface or of any other nuclei which determines the value of k_0, reduces the range of the metastable region and shifts its limit nearer to the coexistence line. In special cases, $k_0 = 0$, this limit can coincide with the coexistence line, i.e. no superheating is possible.

In equation (7) the volume V is not that of the bulk liquid but can be calculated from the area of the heating surface times the thickness of the thin superheated liquid layer, which can be assumed to be of order of the critical radius. Δp is determined as in equation (5) with the saturation temperature and the temperature of the superheated liquid, which is assumed to be the wall temperature. J^* can be regarded as the quantity of bubbles produced in the time interval t and can be determined by the bubble frequency.

For the calculation of the ratio k_0 from equation (6) the exact knowledge of the statistical distribution function of the radii and the aperture angles would be necessary. It can be concluded from experimental investigations of boiling that the radii of the cavities must be between 10^{-9} and 5×10^{-8} m. This small size cannot be observed with any microscope. In the case that the cavity is an ideal cone there exists between the mouth radius r_L of the cavity the radius r_c, the wetting angle θ_0 and aperture angle β the geometrical relation:

$$r_L = r_c\cdot\cos(\theta_0-\beta/2). \quad (8)$$

Also the actual wetting angle θ_0 in equations (6) and (8) is unknown, because the formation process of a critical bubble is very fast. Therefore not the static but the dynamic wetting angle must be taken into account. The dynamic wetting angle may vary between 0 and 150° following the consideration of Class [4]. Furthermore it has to be considered that there is no ideal contact between pure liquid and pure metal, because at a surface there can exist some oxides and impurities, which influence the wetting angle in an unknown manner. Therefore it seems to be impossible to calculate the ratio k_0 from equation (6) for real boiling problems with normal liquids.

4. DETERMINATION OF THE RATIO OF ACTIVATION ENERGY k_0

Kottowsky [3] showed that the heterogeneous nucleation theory can be applied for liquid metals. Their high surface tension implies larger critical radii and larger radii for the cavities than other liquids. Therefore the active cavities can be observed and their necessary probability function is obtainable.

Considering that all the bubbles during boiling are formed in cavities of the heated surface, which are filled with superheated liquid, there is no reason that the heterogeneous nucleation theory should not be valid for normal liquids. Although the parameters β and θ_0 for boiling experiments are not directly available by measuring techniques; the ratio k_0 can be indirectly determined from experiments by rearranging equation (7):

$$k_0 = \frac{\Delta p^2 \cdot 3 \cdot k \cdot T}{16 \cdot \pi \cdot \sigma^3} \cdot \ln\left\{\frac{V \cdot t}{J^*} \cdot Z \cdot \left[\frac{6\sigma}{(3-b) \cdot \pi \cdot M}\right]^{1/2}\right\}. \tag{9}$$

If one assumes that J^* is the number of bubbles produced in the volume V during the time t corresponding to the bubble frequency all terms of the equation (9) are known from the experimental conditions. Therefore k_0, the ratio of the activation energies required for a bubble to be formed within a cavity to the energy necessary for the formation of a spherical bubble, can be determined by boiling experiments, and with equations (1), (6) and (8) the "surface character" can be calculated.

It can be supposed that at constant temperature k_0 tends to a constant value for a certain combination of "surface character" and "liquid quality". But k_0 is not independent from the saturation temperature of the liquid, because the wetting angle β in equation (6) must be regarded as an unknown temperature function. In equation (9) the leading term for the temperature dependence is the surface tension, which can be described as a function of temperature from the triple-point up to the critical point for many liquids by a simple power law [5, 6]:

$$\sigma = \sigma_0 \cdot \left(\frac{T_c - T}{T_c}\right)^{\mu} = \sigma_0 \cdot \varepsilon^{\mu}. \tag{10}$$

In this equation T_c is the critical temperature of the fluid and ε the reduced temperature difference. The exponent μ can be regarded as a "universal" one for many liquids with a numerical value of 1.28 ± 0.05.

In the simplest case it will be assumed that the temperature dependence of k_0 follows a similar power law to the surface tension:

$$k_0 = K \cdot \left(\frac{T_c - T}{T_c}\right)^{\gamma} = K \cdot \varepsilon^{\gamma}. \tag{11}$$

It is obvious that the heterogeneous nucleation theory is valid for the first formation of an initial bubble at the onset of nuclear boiling. With the assumption that each bubble is formed within the cavity in the metastable liquid this theory should be valid for the whole bubble boiling region up to the maximum superheating temperature at boiling crisis at maximum heat flux. The ratio k_0 must not necessarily be constant for certain saturation temperatures, because it depends on the dynamic wetting angle and dynamic surface tension, which are dependent on the velocity of bubble formation. The nucleation probability J^* changes from unity at boiling onset up to 10^9 at maximum heat flux. The metastable volume of the liquid is, as mentioned, determined by the area of the heating surface times the critical radius, which changes with the superheating temperature ΔT as:

$$r_c = \frac{2\sigma}{\Delta p} \quad \text{where} \quad \Delta p = \int_{T_s}^{T_s + \Delta T} \left(\frac{\mathrm{d}p}{\mathrm{d}T}\right) \mathrm{d}T. \tag{12}$$

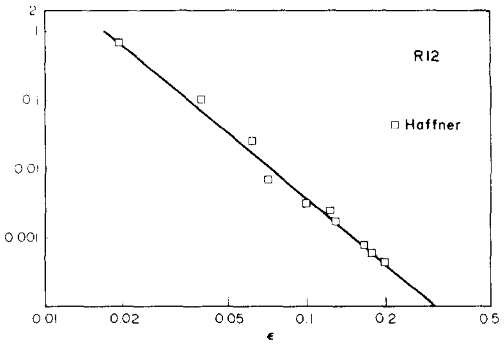

FIG. 3. The ratio of the reduced activation energy k_0 vs reduced temperature ε for boiling crisis of R12 by Haffner [7].

From this equation it follows that at constant saturation temperature the critical radius decreases with increasing superheating, this means that the activation energy is reduced and that the nucleation probability increased.

To check equations (9) and (11) a number of boiling experiments on different liquids are evaluated in the whole temperature region from triple point up to critical point and are discussed as follows. In most cases of this evaluation the superheating experiments at boiling crisis are used, because at maximum heat flux more reliable experiments are available as on boiling onset.

Good values of the maximum superheating temperature at boiling crisis on the refrigerant R12 have been measured inside a vertical tube at forced convection by Haffner [7] over a wide range of saturation temperature starting near the triple-point up to near the critical point. In Fig. 3 the calculated ratio k_0 from these experiments is plotted vs the reduced temperature ε in a log–log plot. It is demonstrated that the temperature dependence of k_0 can be described by equation (11) from the triple-point up to $\varepsilon = 0.017$. The coefficient K and the exponent of this equation are determined by a least square fit method with an average deviation of about 37%, resulting mainly from scattering of experimental values. The numerical values of K and γ are listed in Table 1 with the results of other fluids.

Pool boiling results on a platinum wire by Feurstein [8] for the refrigerant R13 near the critical point are shown in Fig. 4. The circles result from boiling crisis and the crosses from boiling onset experiments. For ε larger than 0.05, k_0 can be described by equation (11), for ε less than 0.05, k_0 has a value of about unity. The deviations from unity may be explained by experimental uncertainty. One has to consider that the superheating is very small near the critical point, and additionally the relatively large influence of a small error in the surface tension. Therefore small errors in temperature measurements can lead to large deviations in k_0. A value of unity for k_0 is probable and means homogeneous nucleation. This is not in contradiction with experience, because near the critical point bubbles forming inside the liquid are observed.

Table 1. Coefficient and exponent for equation

Liquid	Author	K	γ	Valid for $\varepsilon >$
R22	Haffner [7]	5.30×10^{-6}	-2.81	0.01
R12	Haffner [7]	2.42×10^{-6}	-3.16	0.01
N_2	Kosky [10]	3.80×10^{-6}	-5.12	0.08
N_2	Roubeau [11]	1.95×10^{-6}	-4.35	0.06
O_2	Kosky [10]	8.51×10^{-6}	-6.07	0.1
C_2H_6O	Cichelli [12]	1.71×10^{-7}	-5.63	0.07

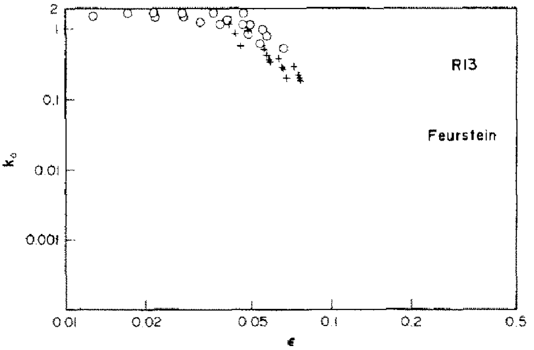

FIG. 4. k_0 vs reduced temperature ε for R12 by Feurstein [8]. The circles represent experiments of boiling crisis, the crosses experiments for the beginning of boiling.

This effect may be explained in Fig. 1. It can be seen that when approaching the critical point, the stable, metastable and unstable region as well as homogeneous and heterogeneous nucleation limits are converging. At the critical point the surface tension approaches zero. Regarding equations (2) the necessary activation energy approaches zero too. In addition it is known that by approaching the critical point the average size of molecular fluctuation called the correlation length increases rapidly and achieves dimensions in the order of the critical radii. Therefore near the critical point homogeneous nucleation is possible and only a very small superheating of the thin liquid layer on the heating surface is necessary without additional nucleation.

Figure 5 shows the results of boiling crisis experiments by Hesse [9] with the refrigerant R114. He used a smooth vertical Ni-tube of 14 mm diameter in pool boiling. From the experiments on oxygen by Kosky [10] in Fig. 6, it can be demonstrated that this theory is also valid for liquid gases. Kosky used a plain polished platinum disc of 1.9 mm dia.

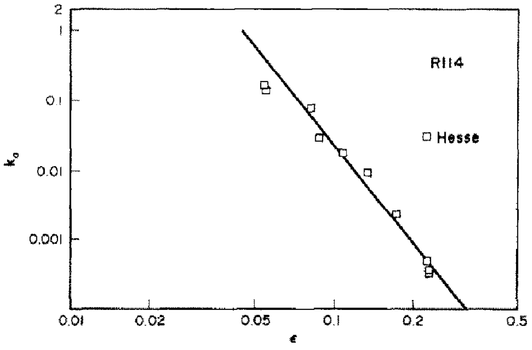

FIG. 5. k_0 vs reduced temperature ε for the boiling crisis of R114 by Hesse [9].

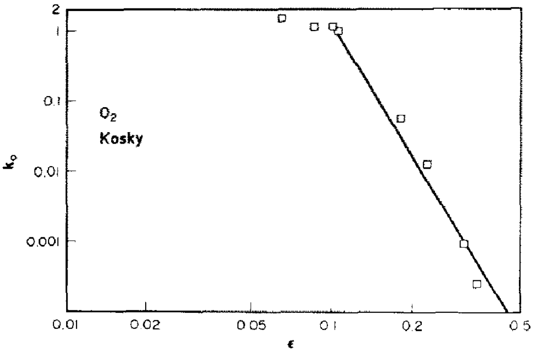

FIG. 6. k_0 vs reduced temperature ε for the boiling crisis of O_2 by Kosky [10].

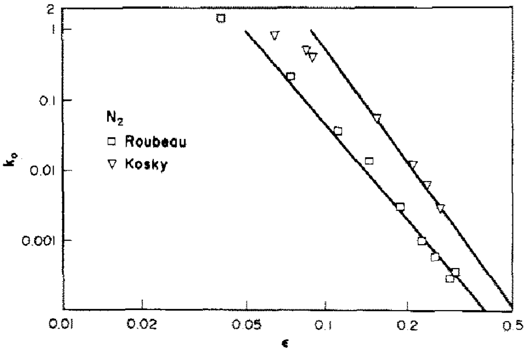

FIG. 7. k_0 vs reduced temperature ε for the boiling crisis of N_2 by Roubeau [11] (squares) and Kosky [10] (triangles).

In Fig. 7 boiling crisis experiments by Roubeau [11] and Kosky [10] on nitrogen are compared. Each set of data can be represented with equation (11), but k_0 from Kosky's results is generally larger. This could be caused on the influence of the polished platinum disc used by him with probably less nuclei on the surface than on that of Roubeau. Another influence on these results could be, that the maximum superheating is influenced by hydrodynamic effects and by subjective observation methods.

As an example for organic liquids, experiments by Cichelli [12] on ethylalcohol are evaluated in Fig. 8. The heating surface was a vertical copper plate coated with polished electro-plated chromium.

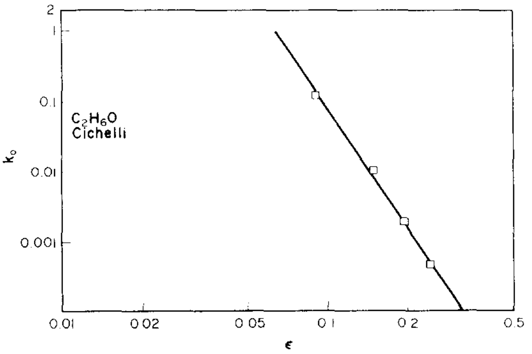

FIG. 8. k_0 vs reduced temperature ε for the boiling crisis of ethylether by Cichelli [12].

A comparison of boiling crisis experiments on R22 by Haffner with our own experiments [13] is shown in Fig. 9. In our experiments the bulk liquid in the pressure vessel was quasi-steadily brought into the metastable region by isochoric cooling with slow cooling rates. The temperature, pressure and density of the metastable liquid were measured exactly at the breakdown of the metastable state which was registrated by an abrupt increase of the pressure and temperature when the initial bubble is created anywhere on the wall of the pressure vessel. As described above this bubble creation is a statistical process, where in a nucleus a bubble with a critical radius is formed. These experiments may be regarded as boiling onset and it is not surprising that there is a larger statistical scattering observed than in the boiling experiments. In this case the large number of bubbles created during the time of observation cause a good average value of k_0. Regarding these aspects the agreement between Haffner's and our own experimental results and the theory of heterogeneous nucleation is satisfactory.

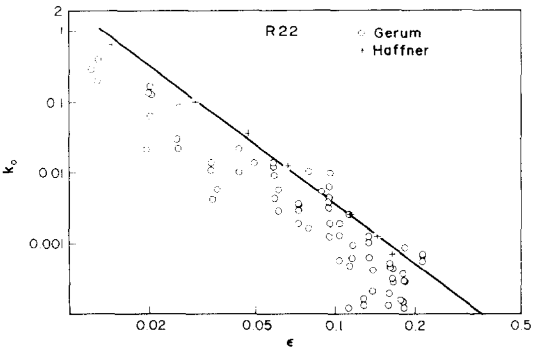

FIG. 9. k_0 vs reduced temperature ε of R22 for the boiling crisis by Haffner [7] (crosses) and for the beginning of boiling (circles)—own measurements.

Pool boiling experiments on water from Turton [14] and Rohsenow cited by Kreith [15] are compared in Fig. 10 with our own experiments on water, in which the limit of the metastable region is determined in a similar quasistationary manner, as mentioned above. Other experiments of boiling crisis at low temperatures (ε between 0.4 and 0.5) on thin platinum wires by Pitschmann [16] agree so well with our own measurement [13] that they are not plotted in Fig. 10. All results for water can be fitted with an exponential function as:

$$k_0 = 67.44 \cdot \exp(-45.5\,\varepsilon). \tag{13}$$

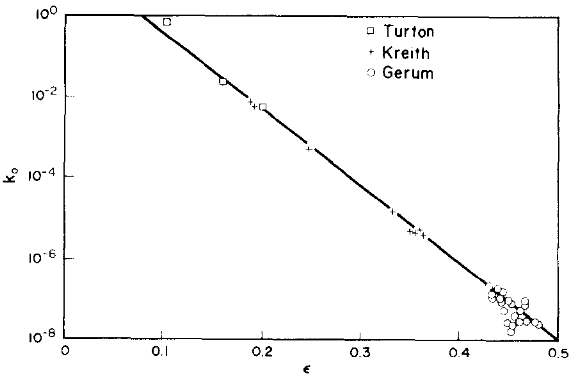

FIG. 10. k_0 vs reduced temperature ε of water for the boiling crisis by Turton [14] (squares) and by Kreith [15] (crosses) and the beginning of boiling from our own experiments.

In the case of water, k_0 is much less than on other liquids. Partly this could be traced back to the higher surface tension of water [17], which according to equation (9) influences strongly the reduction of the activation energy.

If the temperature dependence of k_0 would only be influenced by the surface tension, then it follows from equation (10) that the exponent γ in equation (11) should be -3.84. In Table 1 the values of γ from this evaluation are between -2.8 and -6.1 and different from liquid to liquid.

From this result it seems to be confirmed that k_0 depends not only on the properties of the liquid, but additionally on the characterica of the surface, which can only be defined indirectly by boiling experiments. By systematic investigations on different liquids on the same and different surface characterica the influence of the surface on the reduction of the activation energy for superheating the liquids in boiling could be checked.

5. CONCLUSIONS

Introducing a ratio k_0 for the reduced activation energy the heterogeneous nucleation theory is used to describe the metastable state of a liquid which means the temperature of superheating at a boiling surface. The active nuclei on the surface are so small that they cannot be observed with microscopes, therefore the ratio k_0 is determined indirectly from boiling experiments. This ratio depends on the saturation temperature of the liquid and this dependence could be described by a simple power law.

Near the critical point homogeneous nucleation is achieved.

With this extended heterogeneous nucleation theory it seems to be possible to determine the maximum superheating at boiling crisis over the whole saturation line with a few experimental points only. At the moment it is not possible to calculate the maximum heat flux, because the heat-transfer coefficient is no more linear near boiling crisis.

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CALCUL DE LA SURCHAUFFE EN EBULLITION NUCLEE PAR LA THEORIE DE LA NUCLEATION HETEROGENE

Résumé—A l'aide de la théorie de la nucléation hétérogène, on décrit la surchauffe de la couche limite liquide dans l'ébullition nucléée non seulement en ce qui concerne l'apparition de l'ébullition nucléée mais aussi la crise d'ébullition. Le flux de surchauffe dépend de la stabilité thermodynamique du liquide métastable qui est influencée par les fluctuations statistiques dans le liquide et la nucléation sur la surface solide. Du fait que les cavités agissant sur la nucléation sont trop petites pour une observation microscopique, on ne peut déterminer expérimentalement la probabilité de formation des bulles. La formation des bulles peut être représentée par une fonction empirique dont les coefficients sont déterminés à partir des mesures. Ainsi la théorie de la nucléation homogène décrit la surchauffe du liquide, qui a été vérifiée pour différents fluides tels que les réfrigérants, des gaz liquéfiés, des liquides organiques et l'eau.

ÜBERHITZUNG BEIM BLASENSIEDEN, BERECHNET MIT DER HETEROGENEN KEIMBILDUNGSTHEORIE

Zusammenfassung—Mit der heterogenen Keimbildungstheorie kann die Überhitzung der Flüssigkeitsgrenzschicht beim Blasensieden sowohl für den Siedebeginn wie auch für die Siedekrisis beschrieben werden. Der Grad der Überhitzung hängt von der thermodynamischen Stabilität der metastabilen Flüssigkeit ab, die durch die statistischen Schwankungen der Flüssigkeit und durch die Keimwirkung der festen Oberfläche beeinflußt wird. Da die auf der Oberfläche als Keime wirkenden Kavernen zu klein für die mikroskopische Beobachtung sind, können deren Größe und Verteilungsfunktion nicht vermessen werden, was zur Bestimmung der Wahrscheinlichkeit des Blasenbildungsprozesses notwendig wäre. Die durch die Keime reduzierte Blasenbildungsarbeit läßt sich durch eine einfache empirische Funktion darstellen, deren Koeffizienten aus Siedeversuchen bestimmt werden. Damit kann die heterogene Keimbildungstheorie die Überhitzung der Flüssigkeit beschreiben, was an verschiedenen Fluiden, wie Kältemitteln, flüssigen Gasen, organischen Flüssigkeiten und Wasser geprüft wurde.

ИСПОЛЬЗОВАНИЕ ГЕТЕРОГЕННОЙ ТЕОРИИ ЗАРОЖДЕНИЯ ПУЗЫРЬКОВ ДЛЯ РАСЧЁТА ПЕРЕГРЕВА ПРИ ПУЗЫРЬКОВОМ КИПЕНИИ

Аннотация — С помощью гетерогенной теории зарождения пузырьков описывается перегрев пограничного слоя жидкости при пузырьковом кипении не только в начале кипения, но и в момент наступления кризиса. Степень перегрева зависит от термодинамической устойчивости метастабильной жидкости, на которую оказывают влияние статистические флуктуации в жидкости и зарождение пузырьков на твердой поверхности. Поскольку впадины, являющиеся центрами образования пузырьков, очень малы, то обычными измерительными методами невозможно определить их размеры и распределение по поверхности, т. е. найти вероятность образования пузырьков. Это можно сделать с помощью простого эмпирического выражения для работы образования пузырька, в которое входят экспериментально определяемые коэффициенты. С учётом этого выражения гетерогенная теория зарождения пузырьков позволяет описать перегрев жидкости. Справедливость формулы проверена на таких жидкостях, как хладагенты, жидкие газы, органические жидкости и вода.