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Interphase fluctuation propagation and superposition model for boiling nucleation

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Abstract

Although boiling has been the subject of numerous investigations, there are still a number of fundamental nucleation phenomena needed to be explained consistently. A new proposed physical model termed the 'interphase fluctuation propagation and superposition model', was used to successfully explain the physical nature of heterogeneous nucleation processes. the new concepts of 'evaporating space' and 'fictitious boiling' were quoted here to explain nucleate boiling at the microscale level and resulted in a significantly enhanced understanding and a much clearer representation of the physical significance of the fundamental parameters that govern nucleate boiling. © 1998 Elsevier Science Ltd. All rights reserved.

Nomenclature

- G Gibbs function
- g specific Gibbs function
- m mass
- P pressure
- r bubble radius
- T temperature
- V volume
- v specific volume
- ρ density
- σ surface tension.

Subscripts

- e equilibrium
- g noncondensable gas or vapor
- i interface
- 1 liquid phase
- T temperature
- v vapor phase.

1. Introduction

Boiling nucleation and the associated bubble dynamics have been the subject of numerous previous investigations, and continue to be one of the basic research topics in the field of heat transfer. However, because of the complexity of the phenomena, no unified theory has been able to fully explain nucleate boiling phenomena [1]. The idea that nucleation occurs from small cavities with entrapped gases or vapor has been the most popular nucleation theory until recently, and a concave re-entrant cavity was thought to be the 'best' cavity geometry for entrapping gas or vapor [2, 3]. Several researchers from Russia developed a somewhat different nucleate theory which employed the idea of a convex 'nose' that serves as a nucleation site, to explain nucleate boiling phenomena in a pure working fluid where there are no noncondensable gases or impurities [4].

While these explanations are helpful, there are still a number of questions that occur for nucleate boiling phenomena. For example, when the smooth bottom surface of a beaker is touched by a smooth glass rod, bubbles will be generated at the point of contact for conditions at which boiling in an open pool would not previously occur, as shown in Fig. 1. Removal of the rod, however, eliminates the boiling and allows the liquid to return to a stable single phase state [2]. The formation of the bubbles cannot be explained by the well known 'entrapped gas or vapor model', or by the convex nose explanation.

In some cases, the onset of bubble nucleation is very difficult to achieve. For example, Lin et al. [5, 6] exper-

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Fig. 1. Bubble formation under a glass rod.

imentally observed bubble growth and the associated boiling characteristics on micro-wire and microchip heaters in microchannels. It was extremely difficult to generate bubbles in very small microchannels. The liquid superheat temperature required to initiate boiling or form bubbles in these microchannels was approximately the same order of magnitude as that for homogeneous nucleation in an unconstrained liquid. Peng et al. [7–11] have completed a series of experimental investigations on flow boiling through microchannels with rectangular cross-sections. The experimentally measured boiling curves indicated that boiling seemed to be initiated at wall superheats of only 2-8°C and immediately shifted to the fully developed nucleate boiling regime without any evidence of partial nucleate boiling. This was true even for highly subcooled liquid flows. However, at sufficiently high wall heat fluxes or wall superheats, which according to the previously observed boiling curves should produce nucleate boiling, no bubbles were observed in the microchannels even with the assistance of a high-power magnifying glass [1]. It would appear that the length scale of the liquid flow geometry may strongly influence the phase-change transition and corresponding flow boiling model. More recently, a nondimensional parameter, $N_{\rm mb}$ and the related criteria, $N_{\rm mb} \leq 1$, were derived by thermodynamic phase stability theory to determine the phase transition in microchannels [12]. The length scale was shown to have significant impact for sufficiently small hydraulic diameters, resulting in dramatically higher heat fluxes required for nucleation.

The hypothetical concept referred to as the 'evap-

orating space' [1], suggests that there exists a minimum amount of space necessary to generate a vapor bubble in a confined microchannel. This hypothesis is confirmed by the above nondimensional parameter and related criteria. Based on the analysis of bubble formation during nucleate boiling, the present work proposes a new model referred to as the 'interphase fluctuation propagation and superposition model' to explain nucleate boiling phenomena, and discusses qualitatively the concepts of 'evaporating space' and 'fictitious boiling' occurring in microscale boiling.

2. Thermodynamic aspects of bubble formation and growth

Figure 2 illustrates a system held at a constant temperature, T_1 , and pressure P_1 , which contains a superheated pure liquid, in which a spherical embryo vapor bubble has been formed. If the Gibbs function, G, defined as

$$G = U - TS + PV \tag{1}$$

is evaluated for this system after the formation of the embryo bubble, the Gibbs function for the system is the sum of three components, or

$$G = G_v + G_1 + G_i \tag{2}$$



Fig. 2. Formation of an embryo vapor bubble in superheated liquid.

where $G_{\rm x}$, $G_{\rm l}$ and $G_{\rm i}$ are the free energy contributions due to the vapor bubble, and surrounding liquid, and the interface, which is due to interfacial tension, respectively.

Prior to the formation of the embryo, the entire system consists of superheated liquid and the Gibbs function is given by

$$G_0 = (m_1 + m_y)g_1(T_1, p_1)$$
(3)

where m_1 and m_y are the mass of liquid surrounding the bubble, and the mass of the bubble, respectively, and g_1 is the specific Gibbs function for the liquid.

Given this system, the change in the Gibbs function of the system associated with the formation of the embryo can be expressed as [3]

$$\Delta G = G - G_0 = G_v + G_1 + G_1 - G_0$$

= $m_v [g_v(T_1, P_v) - g_1(T_1, P_1) + (P_1 - P_v)v_v] + 4\pi r^2 \sigma.$ (4)

• 0

If the embryo is of the exact size so as to be in equilibrium with the surrounding superheated liquid, i.e. $r = r_{e}$, then $P_{\rm y}$ can be expressed in terms of the Young–Laplace equation as

$$P_{\rm v} = P_{\rm ve} = P_{\rm l} + \frac{2\sigma}{r_{\rm e}} \tag{5}$$

and the change in the Gibbs function will be equal to

$$\Delta G_{\rm e} = m_{\rm v} \left[g_{\rm v}(T_{\rm l}, P_{\rm ve}) - g_{\rm l}(T_{\rm l}, P_{\rm l}) - \frac{2\sigma v_{\rm v}}{r_{\rm e}} \right] + 4\pi r_{\rm e}^2 \sigma.$$
 (6)

For a pure substance, the chemical potential equals the specific Gibbs free energy, $\mu = g$. Because $\mu_1 = g_1$ must equal $\mu_{\rm v} = g_{\rm v}$ at equilibrium, the $g_{\rm l}$ and $g_{\rm v}$ terms in the above equation cancel. Recognizing that $m_v v_v$ is equal to the total volume of the embryo, the above relationship can be simplified to

$$\Delta G_{\rm e} = \left(\frac{4}{3}\right)\pi r_{\rm e}^2 \sigma. \tag{7}$$

Because T_1 and P_1 are fixed, and assuming that the surface tension, σ , is constant, which is true for small changes in temperature, equation (6) suggests that ΔG is a function of P_{v} , v_{v} and r. If the vapor is assumed to behave as an ideal gas, i.e. $P_v v_v = RT_v$ and the embryo is in mechanical equilibrium and satisfies the Young-Laplace equation regardless of the size of the bubble, then the dependency of ΔG on P_{y} and v_{y} can be removed and ΔG is only a function of r. With these idealizations, ΔG can be expanded using a Taylor series expansion technique about $r = r_e$ to yield [3]

$$\Delta G = \frac{4}{3}\pi r_{\rm e}^2 \,\sigma - \frac{4}{3}\pi \sigma \bigg[2 + \frac{1P_1}{P_{\rm ve}} \bigg] (r - r_{\rm e})^2 + \dots \tag{8}$$

The variation of ΔG with respect to the radius, r, is illustrated in Fig. 3. As shown, ΔG clearly has a maximum value and this value occurs at $r = r_e$. Because the Gibbs free energy, G, must be at a minimum for a stable equilibrium and G_0 is a constant, in order for the entire system to be in stable equilibrium, $\Delta G = G - G_0$ should be at a



Fig. 3. Variation of the Gibbs function with bubble radius for a vapor bubble spontaneously formed in a superheated liquid.

minimum value. Using this rationale, it is clear that an embryo of radius $r = r_{e}$, such as the one under consideration here, is in an unstable equilibrium state. Thus, if any density fluctuations in the metastable liquid lead to the loss of a single molecule from the embryo and produces an embryo bubble of radius $r < r_e$, the bubble will collapse. Conversely, if an embryo of radius $r = r_e$ gains one molecule from the density fluctuations in the metastable liquid and enters the range $r > r_e$, the embryo will grow spontaneously. This density fluctuation is termed the 'interphase fluctuation', and implies that a nucleation site or an embryo in a metastable liquid is the direct result of density fluctuations, which can be strengthened by superposition or weakened by dissipation. An embryo can grow only when density fluctuation causes an increase in the radius r making it larger than $r_{\rm e}$. Then and only then will nucleate boiling begin.

The above analysis for homogeneous nucleation within a metastable liquid phase can be extended to heterogeneous nucleation, including an embryo formed on a heated wall, but the term G_i will be much more complicated, since the interface is governed by the presence of the vapor, liquid and solid.

3. Interphase fluctuation propagation and superposition

Figure 4 illustrates the density distribution near a liquid-vapor interface. This type of interface will be formed when two different phases meet together. In this situation, the density in the interface region will always be lower than that occurring in the bulk phase. As a result, the density in this region is transitory in nature and is continually fluctuating in the interface region. This continually varying density distribution in the interface region, illustrated in Fig. 4, will be stable only when the



Fig. 4. Density variation in the interfacial region.

vapor and liquid are in a precise equilibrium state. When the temperature is changed, i.e. heated or cooled, or the system is heated without causing a temperature change, the density in the interface region will become unstable and will fluctuate. This density fluctuation will then propagate into the bulk phase. If the liquid is superheated, i.e. $(T_1 - T_g) > 0$, this density fluctuation will become an interphase fluctuation source. All of the fluctuations and changes of density in the superheated liquid can be stimulated as interphase fluctuations by varying the temperature or being heated without causing a temperature change. These interphase fluctuations will then propagate in all directions in the liquid, and may have an interdependency effect, such that any of the interphase fluctuations can be strengthened by superposition or weakened by dissipation.

If a vapor or noncondensable gas is present in the liquid or in the interface region of the solid and liquid, the location of the vapor or noncondensable gas may appear to be the nucleation site. When other interphase fluctuations are induced by temperature variations or other factors are superposed, an embryo which generates from the vapor or noncondensable gas will grow and when its radius is greater than $r_{\rm e}$, a bubble will be formed and will grow from the liquid or solid-liquid interface, resulting in the onset of nucleate boiling. This boiling process will occur only when the liquid molecules become unstable and transfer to the vapor phase, or when the nucleation site or embryo in the liquid is enhanced by other interphase fluctuations. The boiling process will not occur if the interphase fluctuations have a dissipative effect on the nucleate site or the embryo, causing the nucleation site or the embryo to be resolved or dissipated into a larger number of smaller interphase fluctuations.

The intensity of a given interphase fluctuation can be formulated by the Gibbs function as expressed previously

$$\delta G = \delta G_{\rm i} + \delta G_{\rm g} + \delta G_{\rm T} \tag{9}$$

where the subscripts i, g and T symbolize the density fluctuation of the phase interface, the noncondensable gas, and the density fluctuation of temperature, respectively.

4. Heterogeneous nucleation in a liquid

In homogeneous nucleation, only the density fluctuation caused by the temperature is considered. For this case, equation (9) simplifies to

$$\delta G = \delta G_{\rm T}.\tag{10}$$

It is well known and widely accepted that the superheat required for nucleate boiling in this case is very high. For example, the superheat needed for water to boil at atmospheric pressure is approximately 170°C. But, in fact, the superheat required for heterogeneous nucleation that occurs on a superheated wall is much lower. If there are no noncondensable gases present, the existing 'entrapped gas or vapor model' cannot adequately explain this lower temperature needed in this heterogeneous nucleation.

The new 'interphase fluctuation propagation and superposition model' can be introduced here and used to explain the early onset of heterogeneous nucleation. If there are no noncondensable gases, i.e. δG_g in equation (9) is equal to zero, then equation (9) can be rewritten as

$$\delta G = \delta G_{\rm i} + \delta G_{\rm T}.\tag{11}$$

This implies that the density fluctuations can be caused by interface and/or temperature variations, and that the temperature will in all probability induce interface density fluctuations.

Figure 5 illustrates three different kinds of surfaces. For a given liquid, the solid-liquid interface area of the ideal smooth surface shown in Fig. 5(A) is the smallest when compared with those of the convex or concave surface, so, the value of ΔG_i for the ideal smooth surface is also the smallest. For this ideal smooth surface, the interface fluctuation is nearly the same everywhere and will spread uniformly over the hemispherical space. As a result, nucleation will be determined entirely by the intensity of the interface fluctuations. Conversely, the Gibbs function of the interface ΔG_i , and also the gross value of ΔG increases with increases in the interface area of the convex or concave surfaces shown in Fig. 5(B) and (C), and hence, nucleate boiling occurs more easily on the convex or concave surfaces than on the ideal smooth surface. The 'nose' point of the convex surface, i.e. point 'a' shown in Fig. 5(B) will be more easily influenced by fluctuations from different directions and hence, the value of ΔG for point 'a' increases faster than for any other point on the surface that bubbles might be formed. This phenomena makes point 'a' appear to be a preferential



Fig. 5. Different kinds of surface.

nucleation site. Point 'a' on the concave surface as shown in Fig. 5(C) will more easily accumulate fluctuations due to the presence of the side surfaces and as a result, will induce nucleate boiling. In addition, when the surface is heated uniformly, the temperature of the region with convex or concave surfaces will increase more rapidly than for a smooth surface. Furthermore, the liquid inside the concave area has more specific surface area per unit volume than the other regions, so the liquid temperature of this region will clearly increase. If the liquid in this concave region is superheated, bubbles will be induced from this concave area if there are other fluctuations superposed on the partially superheated liquid before the bulk temperature reaches the saturation temperature. The reason for the formation of bubbles between the rod and the heated bottom of the glass, mentioned previously can be explained using a similar analysis.

To verify this hypothesis, a simple pool boiling experimental system was designed to observe bubble formation and generation in nucleate boiling. Figure 6 shows a schematic of the test apparatus used in this investigation. The test section (3) used was a microchanneled stainless steel plate, 15 mm wide, 60 mm long and 2 mm thick. There are three identically sized microchannels uniformly distributed on the plate. The cross-section of the channels was rectangular, 0.6 mm wide and 0.7 mm high. This microchanneled plate was used in previous research and introduced in detail in [7]. Six T-type thermocouples were embedded directly into the plate to measure the wall temperature. A heat flux control system directly heated the plate (3) through electrodes (6). Insulation spacer (9) and washer (10) were made of Teflon, to provide adequate insulation. The working fluids evaluated were pure water and methanol.

When the system was heated to an average temperature of approximately 95° C for pure water (about 60° C for methanol), bubbles were generated only at the ends of the microchannels around the glass plate (4) provided the glass plate was pressed tightly on the test plate and no gap existed.No bubbles were observed anywhere else along the microchannel. The wall temperature in the region under the glass plate (4) at this moment was greater than 105° C for water (for methanol, the temperature was at least 66° C). If the glass plate (4) was removed at this temperature, only a few bubbles could be observed.



1. Test stand 2. Glass lid 3. Test section 4. Glass plate 5. Nut 6. Electrode 7. Copper bar 8. Nut 9. Insulation spacer 10. Washer 11. Thermocouple

Fig. 6. Schematic of test apparatus. 1 Test stand; 2 glass lid; 3 test section; 4 glass plate; 5 nut; 6 electrode; 7 copper bar; 8 nut; 9 insulation spacer; 10 washer; 11 thermocouple.

Bubbles were generated in the microchannels when the system was heated to a temperature of 108° C for water (70°C for methanol) without the glass plate (4). If the test section was changed to a flat unpolished plate, bubbles were formed at a number of sites on the surface of the test section around the edges of the glass plate (4). These bubbles occurred when the system temperature was approximately 98°C for water and 63°C for methanol, while the partial wall temperature under the glass plate (4) was almost 106°C for water and 68°C for methanol.

These interesting phenomena can also be successfully explained by the interphase fluctuation propagation and superposition model. In this experimental system, the microchannels serve as very good concave cavities and the glass plate (4) used as a cover for the test plate, effectively increases the interface area allowing the liquid to become partially superheated. For this case, the interphase fluctuation is very strong. The larger outside spaces of the contacts between the microchannels and the glass plate, make the fluctuations superpose quite easily. As a result, the bubbles can be seen to generate initially at the two ends of the microchannels around the glass plate (4) provided the glass plate was pressed tightly on the test plate. When the unpolished plate was substituted for the microchanneled plate, the defects resulting from the rough surface served as the convex or concave surfaces. Some of these defects, located just under the edge of the glass plate (4) then served as the active nucleation sites. Bubbles formed at these sites first, once the temperature is high enough.

For cases where noncondensable gases exist, the concave surface can easily entrap gases while the smooth and convex surfaces will not. In this case, there are gas-liquid interfaces and gas-solid interfaces in addition to the liquid-solid interfaces. For this situation, the term δG_i can be written as

$$\delta G_{\rm i} = \delta G_{\rm sl} + \delta G_{\rm sg} + \delta G_{\rm lg}. \tag{12}$$

Correspondingly, equation (9) can be rewritten as

$$\delta G = \delta G_{\rm i} + \delta G_{\rm g} = \delta G_{\rm sl} + \delta G_{\rm sg} + \delta G_{\rm lg} + \delta G_{\rm g}. \tag{13}$$

As a result, the concave surface with entrapped gas can more easily become the site of nucleation and induce nucleate boiling.

5. Boiling nucleation in microscale channels

Experimental observations and analysis imply that, besides the necessary conditions, such as superheat and nucleation sites, the length scale of the bulk liquid must be large enough for bubbles to exist and grow in the liquid [1]. The term 'evaporating space' was developed to imply that there is a minimum liquid bulk size that allows internal evaporation to occur and allows bubbles to grow within the liquid. Only when the bulk liquid size is larger than this required evaporating space will boiling occur and bubbles grow. This evaporation will be affected by the boundary conditions of the liquid and will vary with the thermophysical properties of the different liquids [12].

Experimental results and corresponding analyses have often confirmed that the heat transfer rates for systems without boiling phase change, such as vaporization of sessile droplets or convection evaporation of stagnant or flowing liquid films, are much higher than for fully developed boiling. These high heat transfer rates cannot be explained even by considering the combined latent heat of vaporization at the surface and the sensible heat absorbed by the liquid. Experiments conducted with flow boiling inside microchannels revealed that for conditions corresponding to the nucleate boiling regime, but for which no bubbles can be seen in the liquid [7, 11], the superheat of the heating wall was less than 10°C and the superheat of the liquid was even lower, so the actual heat transfer rates (as high as 10^6 W/m^2) could not be reached solely by the absorption of the sensible heat alone. For this reason, a new concept referred to as 'fictitious boiling' was proposed to explain these phenomena [1]. This fictitious boiling encompasses two facets :

- the liquid has reached conventional nucleate boiling conditions but the bubble growth heat transfer processes have not been realized
- (2) the heated liquid will evaporate and bubbles will generate and grow from the free surface as soon as it enters a space which is large enough for bubble formation to occur.

When liquid flows through the microscale channels, the interface area of the unit volume is very large; correspondingly, δG_i increases significantly. The interactions between the interface fluctuations are so frequent and violent that the fluctuations are dispersed into many smaller interphase fluctuations. These fluctuations scatter and propagate into a larger space, (here the outlet of the microchannels), where they can superpose together again to form bubbles. In this situation, the small microscale space will disperse the fluctuations into even smaller ones until there exists an evaporating space for bubble generation. The intensity of the scattering, however, will be different for different liquids and different boundary conditions, and hence, so too will the evaporating space required for different liquids. Also, because the fluctuations are scattered into smaller ones, bubble formation may be delayed. All of these smaller fluctuations, however, carry very high energy, so the heat transfer in the microchannels is intensified even though no bubbles are formed in the microchannels, hence the term 'fictitious boiling'. In addition, if the outlet of the microchannels cannot meet the conditions for the superposition of the interphase fluctuations, the fluctuations will continue spreading. The scattering among the interphase fluctuations in the microchannels are in fact somehow commensurate with the boundary effects in the microscale.

6. Conclusions

The interphase fluctuation propagation and superposition model proposed for boiling nucleation has been used to qualitatively explain the physical mechanisms of both macroscale nucleate boiling and microscale fictitious boiling. In this model, the interphase fluctuations that promote the unstable liquid are the source of the phase change. The liquid must be superheated or partially superheated before it becomes unstable. However, during the nucleation, fluctuations resulting from the change of temperature are not significant, and the fluctuations from the interface are the main factors causing the liquid to be

unstable. The effects of any entrapped vapors or noncondensable gases are that they provide a vapor-liquid interface. Additional experiments are still necessary to verify and understand the new model in more detail; and more quantitative data are required to determine the extent to which these factors affect the nucleate boiling behavior.

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