

On the effect of the dynamic contact angle of a vapor embryo interface trapped in a nucleation site

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Abstract The effect of boiling and cavitation phenomena on nucleation was first experimentally studied. Results highlight the fact that the "classical" theory of nucleation cannot describe such a configuration. New theoretical approaches were proposed in order to describe the dynamic effects which occur when the liquid pressure oscillates over time and when a heat flux imposed to the system. It then appears that the dynamic and the hysteresis of the contact angle may play a significant role in nucleation by simultaneous boiling and cavitation effects.

Keywords: Micro Flow, Nucleation, Boiling, Cavitation

1. Introduction

Numerous heat transfer enhancement techniques have been developed to meet the growing needs in terms of heat transfer efficiency and of compactness. Those implementing the liquid-vapor phase-change are promising. However, one of the constraints of these techniques is the high temperature to be achieved for the onset of the boiling process. A way for controlling the nucleation incipience temperature was proposed by Léal et al. [1, 2, 3]. It consists in simultaneously involving boiling and cavitation, using the dynamic deformation of a confinement wall. Indeed, nucleation can be obtained in two ways: by increasing the liquid temperature at constant pressure (boiling) or by decreasing the liquid pressure at constant temperature (cavitation). Thus, a decrease in pressure would then cause the decrease of the temperature necessary to the nucleation incipience. Furthermore, if the mechanisms governing nucleation were studied from the point of view of "boiling" and from the point of view of "cavitation", the simultaneous action of these two effects on the nucleation has not been analyzed yet.

An experimental device was designed to study the effect of the dynamic deformation of the confinement wall on nucleation conditions. A comparison between experimental nucleation incipience temperatures and those one evaluated using the existing theory [4] of nucleation is performed. It shows that the existing static theory cannot predict nucleation in such a dynamic configuration: certain dynamic effects are not taken into account in the existing theory. Thus, new theoretical approaches taking into account some dynamic effects on nucleation are proposed in order to identify the main mechanisms involved in such a configuration.

Firstly, a dynamic model of flow and of heat transfer in the vicinity of a vapor embryo trapped in a heated wall when an oscillation of pressure is imposed in the liquid was performed. It assumed that the contact line is attached to the singularity (aperture of the cavity) because this assumption provides satisfactory results in static case. The nucleation incipience superheats obtained by this

model are almost equal to the ones evaluated by the static theory. Knowing that this hypothesis does not allow explaining the results, a second hypothesis, frequently used for highly wetting fluid, is considered. The contact line is assumed to be located inside the cavity; in this configuration the dynamic of the contact angle as well as the contact angle hysteresis may change the nucleation conditions. Theoretical approaches are developed to study these dynamic effects of the contact line of the vapor embryo trapped into the wall cavity on nucleation. In a first time, the main results of the experimental study are reported. Then, the theoretical approach, studying the effect of the dynamic of the contact angle is realized.

2. Experimental study

An experimental device was developed to study nucleation incipience induced by the dynamic deformation of a confinement wall. As report by figure 1, in the test section, a fluid (n-pentane) is confined between a heated wall and a confinement wall. The confinement wall is dynamically deformed at its center by a piezoelectric actuator whereas its periphery is maintained fixed. The dynamic deformation yields to successive acceleration and deceleration of the confined liquid: the liquid pressure is temporally decreased. Thus, boiling and cavitation processes are simultaneously involved. Furthermore, the heated wall is instrumented by ten thermocouples and is polished to obtain ruggedness in order of magnitude of $0.2 \mu\text{m}$. The experimental protocol consists in applying successive steps of heat flux on the heated wall and waiting between each step the system reaches the stationary state. The nucleation incipience superheat is defined as the difference between the maximum temperature of the heated wall and the saturation temperature at ambient pressure: $\Delta T_{\text{ONB}} = T_{w,\text{max,ONB}} - T_{\text{sat}}(p_{\text{atm}})$. It is determined by the sudden decrease of the heated wall temperatures and by the appearance of bubbles at the periphery of the confined space. More details about the experimental device and protocol can be obtained in previous works [1, 3].

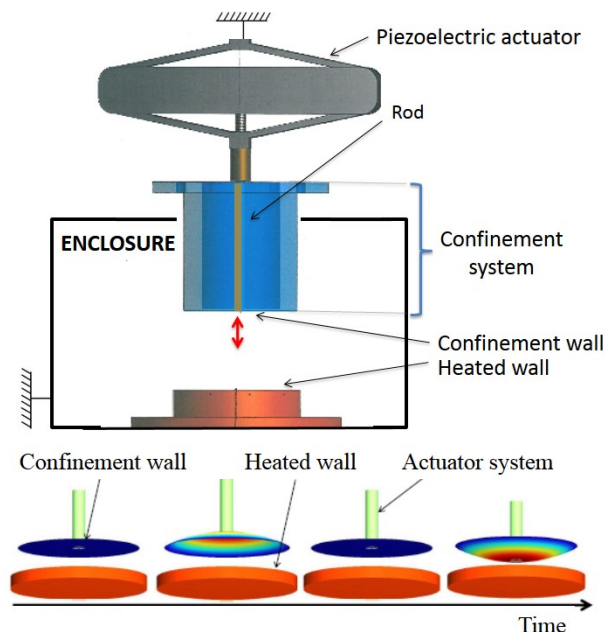


Figure 1: Schemes of the experimental device and of the dynamic deformation of the confinement wall over time

A parametric study was realized to get the effects of the amplitude and of the frequency of the dynamic deformation as well as the effect of the level of confinement on nucleation incipience superheats. Only the main conclusions are reported here. Thus, an increase in frequency or in amplitude leads to the decrease of the nucleation incipience superheats. The effect of the level of confinement was studied: two different distances between the heated wall and the confinement wall

are tested ($e=250 \mu\text{m}$ and $e=500 \mu\text{m}$). When no dynamic deformation is imposed to the confinement wall, the nucleation incipience superheat appears to be independent of the level of confinement. However, when the confinement wall is dynamically deformed, the superheat at nucleation greatly depends on the level of confinement. The effect of the dynamic deformation on nucleation process is even more pronounced that the level of confinement is high. Furthermore, this technique is very efficient. The maximum decrease of nucleation incipience superheat reaches 86% for the range of parameters studied. Indeed, the nucleation incipience superheat is almost equal to 23 K when the confinement wall is not deformed whereas the superheat at the onset of nucleation is reduced to set the value of 3.2 K when the dynamic deformation of the confinement wall is imposed ($a_0=210 \mu\text{m}$, $e=250 \mu\text{m}$, $f=100 \text{ Hz}$).

A hydrodynamic model [1, 3] was realized to obtain the minimum value of pressure reached over time in function of the dynamic deformation parameters as well as of the level of confinement. Using this model, it is possible to determine the effect of the minimum value of pressure reached over time on nucleation incipience superheat. Thus, as expected, the nucleation superheat decreases with decreasing the minimum value of liquid pressure. Furthermore, the comparison between the experimental and theoretical incipience superheats are compared (figure 2). The situation considered by the existing theory of nucleation consists in a vapor embryo attached at the aperture of the cavity of the heated wall ($r_c=0.2 \mu\text{m}$). The liquid pressure considered is the minimum pressure reached over time (static theory). The theoretical superheats at the onset of nucleation are determined using the following equation [4]:

$$r_c = \frac{2\sigma}{-p_l + p_{sat}(T_l) \exp\left(\frac{v_l(p_l - p_{sat}(T_l))}{RT_l}\right)} \quad (1)$$

In equation 1, r_c is the aperture radius of the wall cavity, σ is the the liquid/vapour surface tension, p_l is the liquid pressure and $p_{sat}(T_l)$ is the saturation pressure at the liquid temperature. When no dynamic deformation is imposed, theoretical and experimental nucleation incipience superheats are in good agreement. However, when the confinement wall is dynamically deformed, high discrepancies appear between the experimental and theoretical results (up to 20K). Thus, dynamic effects seem to not be taken into account in the existing theory.

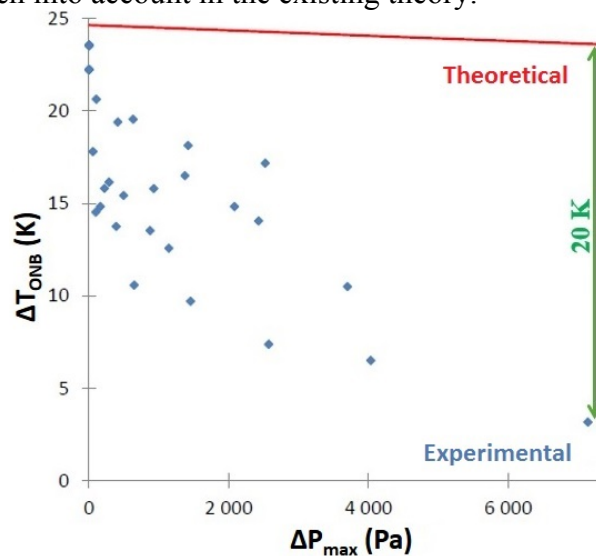


Figure 2: Comparison between experimental and theoretical nucleation incipience superheat

The dynamic deformation of the confinement wall is an efficient way to control the superheat at nucleation. A strong decrease (up to 86%) in the superheat was obtained compared to the reference

case (i.e without dynamic deformation) at the maximum values of amplitude and frequency and at the maximum level of confinement (at $f=100\text{Hz}$, $e=250\mu\text{m}$, $a_0=210\mu\text{m}$). Furthermore, high discrepancies are obtained between theoretical and experimental results reports that dynamic effects, reflecting the non-inclusion of at least one dynamic phenomenon promoting nucleation in the theory. Thus, new theoretical approaches taking into account some dynamic effects on nucleation are proposed.

3. Theoretical study

Experimentally, an embryo is trapped into a wall cavity which the radius of aperture is about $0.2\mu\text{m}$. Furthermore, the pressure of the liquid surrounding the embryo oscillates over time and a heat flux is imposed. In order to determine which mechanisms prevail in such a configuration, a first model of flow and heat transfer in the vicinity of the embryo was performed. It consists in modeling the embryo trapped into the heated wall in taking into account the heat inertia, transport and diffusion as well as the liquid inertia. The main assumption of this model is that the contact line is attached to the aperture of the cavity. Results shown that only the mechanical equilibrium of the interface in a quasi-isothermal environment is needed to predict the embryo stability. The thermal equilibrium of the embryo with the wall is not affect (in the range experimentally explored) by the frequency of pressure oscillation (quasi-static oscillation). As the embryo can be described by as a succession of stationary state, the nucleation incipience superheats determined by this model are almost equal to those one evaluated by the static existing one. Nevertheless, in these two models, the contact line is assumed to be attached to the aperture of the cavity of the heated wall. However, for a highly wetting liquid (which is the case in the experimental configuration), the onset of nucleation takes place while the nucleus is trapped inside a cavity. The contact line is not attached to the geometrical singularity and can therefore move inside the cavity. The dynamic (including the hysteresis) of the contact angle can affect the conditions of nucleation.

Another theoretical approach is then developed to determine the effects of the dynamic of the contact line on the nucleation conditions. This study proposes a method to analyse the stability of the embryo and thus to predict the onset of nucleation. Before describing the model, the definitions of the dynamic of the contact line and of the hysteresis of the contact angle are briefly reported in the following part.

3.1. Hysteresis and dynamic of the contact angle

Figures 3 and 4 respectively report the definitions of the geometrical parameters defining the embryo and the definitions of the dynamic of the contact line and of the hysteresis of the contact angle. As shown by figure 3, the embryo is trapped into a cavity which the angle of aperture is 2β . The contact line, which the position is defined by h , is within the cavity and can move.

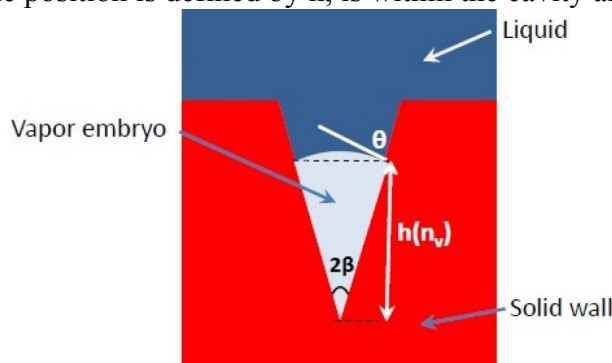


Figure 3: Description of vapor embryo in conical wall cavity

Figure 4 defines how the contact line can move. In static situation, the contact angle is equal to the static contact angle. In imposing an external action, the volume of the embryo can change. For

example, let us consider the case of an increasing volume of the embryo. Firstly, the contact line remains unmoving while the value of the contact angle decreases down to be equal to the receding contact angle (boundary of the hysteresis contact angle range). In the case where the volume continues to increase, the contact line begins to move within the cavity: the liquid frontline goes back. Thus, the contact line moves at a certain velocity which leads to modify the value of the contact angle. More explanations can be find in [4].

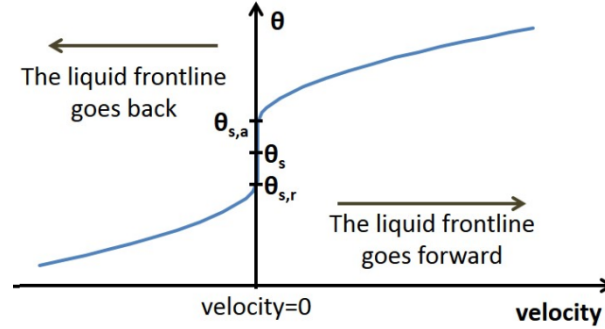


Figure 4: Evolution of the contact angle in function of the velocity of the contact line

To describe the dynamic of the contact angle in a simplified manner, the study is limited to the configuration in which the contact line moves and the contact angle is a function of its velocity. The methodology developed hereafter does not replace the need for much detailed modeling phenomena that occur at these scales but simply provides an attractive alternative for capturing different phenomena and to guide future studies for understanding the experimental results.

In the next part, the equilibrium of an embryo trapped into a cavity is defined. Furthermore, a new method is developed to analyze the stability of the embryo.

3.2. Equilibrium and stability of an embryo trapped into a cavity

To be at the equilibrium, a vapour embryo must be at the mechanic, thermal and thermodynamic equilibria which are expressed by the following equations:

$$p_t = p_{nc} + p_v = p_l + \frac{2\sigma}{r} \quad (2)$$

$$T_v = T_l = T_w = T_{l/v} \quad (3)$$

$$p_v = p_{sat}(T_v) \quad (4)$$

$$p_i = \frac{n_i RT}{V_{embryon}} \quad (5)$$

$$p_v = \frac{n_v RT}{V_{embryon}} \quad (6)$$

$$V_{embryon} = f(h, \theta, \beta) \quad (7)$$

In these equations, p_t , p_{nc} , p_v and p_l are respectively the total pressure of the embryo, the partial pressure of non-condensable gas, the partial pressure of vapour and the pressure of liquid. T_v , T_l , T_w and $T_{l/v}$ are respectively the temperature of vapor, of liquid, of solid wall and the temperature of the liquid/vapour interface. r is the curvature radius of the interface. In this model, non-condensable gas and vapour are assumed to be ideal gases.

A method to determine the stability of each situation of equilibrium is developed. It consists in imposing a fluctuation of the number of vapour mole n_v to the embryo which was at equilibrium state. Thus, a certain quantity of vapour moles crosses through the liquid/vapour interface. The embryo is then temporally in a non-equilibrium state. Nevertheless, the interface may be assumed to

be at equilibrium state (from mechanical and thermodynamical point of view) because of the low time required to reach the interface equilibrium compared to heat diffusion time in the surrounding environment. Furthermore, as the characteristic time of the diffusion inside the vapor embryo is very low (nanosecond), the vapor temperature is supposed to equal to the interface temperature.

Increasing the number of the vapour mole inside the embryo can lead to increase or decrease of the temperature of the embryo. The two situations are studied to determine if they are stable or not.

The first situation considered is the case where the increase in the number of vapour moles inside the embryo leads to the increase of the vapour pressure (and consequently of the temperature of the liquid/vapour interface). Thus, the temperature of the interface becomes higher than those one of the surrounding fluid which means that a heat flux appears from the interface to the surrounding fluid. The embryo is then condensing: the number of vapour moles decreases. As it is initially assumed that the decrease in the number of vapour moles leads to the decrease in the interface temperature, the number of vapor moles decreases until the thermal equilibrium is reached again. As the reasoning is perfectly symmetric, imposing the decrease of the number of vapour moles leads to the vaporisation until the equilibrium is reached. Thus, this configuration is stable.

The second configuration considered is the situation where the increase of the number of vapour moles inside the embryo leads to the decrease in the vapour pressure (and the interface temperature). Thus, the temperature of the interface becomes lower than those one of the surrounding fluid. The embryo is then condensing: the number of vapour moles decreases leading to the further decrease of the interface temperature. The equilibrium state cannot be reached: this configuration is not stable. As this kind of phenomena is extremely fast (compared to the oscillation of pressure over time), it is assumed that this configuration leads to the onset of nucleation. Thus, it is possible to define stability criterion as:

$$\frac{\partial T_{l/v}}{\partial n_v} > 0 \text{ assuming a constant value of liquid pressure and non-condensable gas}$$

Furthermore, as the characteristic time of mechanical effects is assumed to be negligible compared to those of thermal diffusivity, the evolution of the embryo can be analysed as two successive steps. The first step is a quasi-instantaneous one during which thermal transfer does not occur. The embryo can be considered as being at equilibrium state assuming that the system is thermally isolated. The second step is slower. During this step, the system goes back to thermal equilibrium. Thus, in these conditions, the system changes between two equilibrium states.

The limit of stability can be expressed as:

$$\frac{\partial T_{v, equilibrium}}{\partial n_v} > 0 \text{ assuming a constant value of liquid pressure and non-condensable gas} \quad (8)$$

The stability of each equilibrium state is, thus, easy to graphically determine. In the following, an example of the method used is given.

The description of the motion of the contact line used for the example consists in assuming that the value of the contact angle is constant ($\theta = \theta_s$) and that the contact line can move ($h = h(n_v)$). Furthermore, the liquid pressure and the number of moles of non-condensable gas are imposed constant. To be consistent with experimental data, the value of the contact angle is the one of n-pentane on the AU4G heated wall ($\theta = 10$ degrees) and the number of non-condensable moles is chosen in order to obtained the onset of boiling at 23 K (superheat experimentally obtained at 10^5 Pa). The number of non-condensable gas is equal to $4.3825 \cdot 10^{-19}$ mol. First, the equilibrium states for an embryo trapped into a cavity ($2\beta = 20$ degrees) are evaluated using equations 2 to 7. Then, for each equilibrium state reached over time, the stability of the embryo is checked (equation 8).

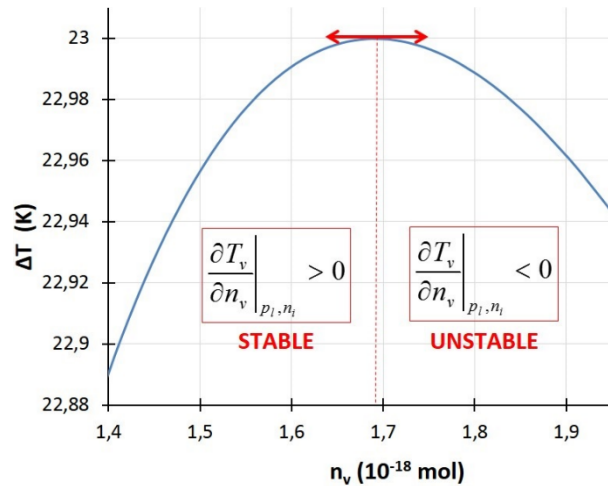


Figure 5: Effect of the increase in the number of moles of vapour on the superheat ($\beta=20$ degrees, $\theta=10$ degrees, $n_{nc}=4.3825 \cdot 10^{-19}$ mol, $p_l=10^5$ Pa)

Figure 5 reports the effect of the increase of n_v on the wall superheat at equilibrium state. At lower n_v , wall superheat increases with increasing n_v whereas at higher n_v , wall superheat decreases with increasing n_v . Thus, the curve representing the wall superheat in function of n_v has a maximum which is, according to the stability criterion, the limit of stability. The stability study just consists in determining the sign of $\frac{\partial T_v}{\partial n_v}$ at equilibrium states. Thus, only the equilibria at the left can endure: a fluctuation from an equilibrium state at the right part of the curve leads to the onset of nucleation assuming that the growing of nucleus is explosive.

3.3. Results

In this paper only situations where hysteresis phenomena do not exist ($\theta_{s,a} = \theta_{s,r}$, figure 4) are considered however the contact angle can change due to the velocity of the triple line.

- In a first time, the equilibrium states (and their stability) of the embryo are studied in the case where the temperature is gradually increased. It is supposed that when a non-stable equilibrium is reached, the nucleation begins. The situation considered here consists in an embryo trapped into a cavity (its contact line is no longer attached to the aperture of the cavity). Furthermore, we assume firstly that the value of its contact angle remains constant over time, which means the modifications of the contact angle induced by the velocity of the triple line are neglected (the velocity of the triple line is negligible when the temperature of the wall is gradually increased).

The liquid pressure and the number of non-condensable gas moles are always imposed (constant). As the system has only one degree of freedom (the position of the contact line), it is possible to describe the increase of temperature by the increase of the number of vapour moles. As it was previously mentioned, while the embryo is stable, the increase of the number of vapour moles leads to the increase of the embryo temperature. The number of non-condensable gas moles is determined in using the experimental conditions when no dynamic deformation is imposed to the confinement wall: the number of non-condensable gas moles is imposed considering that the superheat required to reach the onset of the nucleation is equal to 23 K, in the case where the liquid pressure is constant ($p_l=10^5$ Pa) and the value of the contact angle is equal to 10 degree (experimental condition). The number of non-condensable gas moles is evaluated for the different angles of aperture of the cavity.

- Knowing the composition of embryos which may explode in response to a gradual increase of temperature without hysteresis phenomena we can now consider dynamic situations where both pressure and contact angle can be modified (dynamic deformation of the wall). Nevertheless the limits of stability of embryos reported hereafter are only determined by considering that the first

moments of destabilization occur with constant value (which may be different from the static contact angle value) of the contact angle. So situations where the destabilization occurs with contact angle value inside the hysteresis are not considered.

Choosing the values of the liquid pressure and of the contact angle, the superheat at the onset of nucleation is determined. Figure 6 reports the effect of the values of the contact angle on the nucleation superheat for different liquid pressures. It can be noticed that a decrease of the liquid pressure equal to 0.1 bar (static) only decreases the superheat at the nucleation incipience of about 2 K which means that the boiling incipience superheat is weakly sensible to the value of the static liquid pressure. However, the value of the contact angle has a very significative effect on the pool boiling nucleation incipience superheat: the superheat is decreased up to 20 K ($\theta=10$ to 90 degrees). The effect of the angle of aperture of the cavity (β) is also studied. The variation of the nucleation superheat in function of the contact angle depends of β . The decrease of the nucleation superheat remains important.

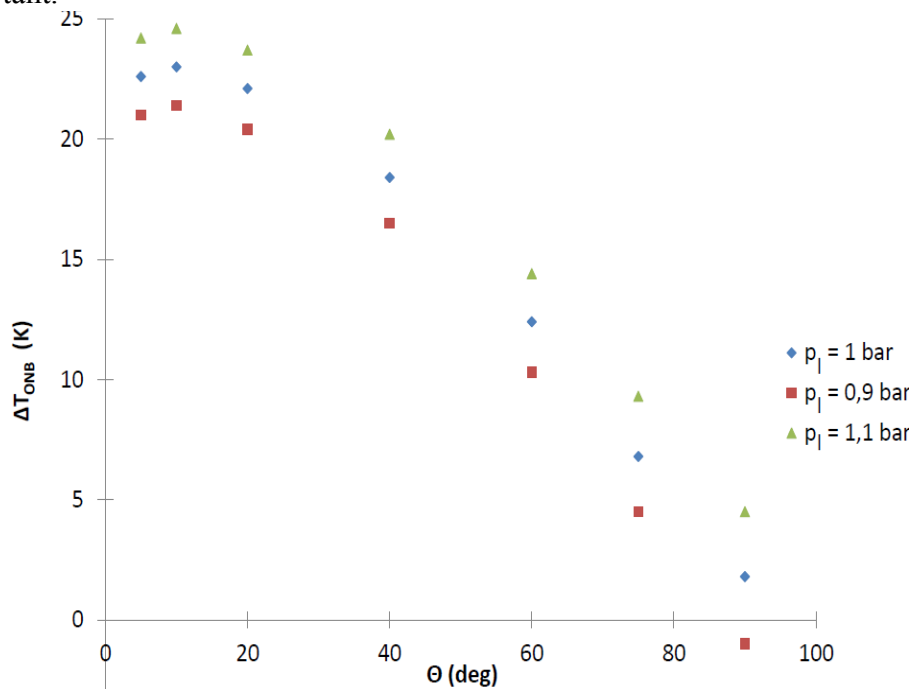


Figure 6: Evolution of the nucleation superheat in function of the contact angle for different liquid pressure ($\beta=10$ degrees)

This parametric study showed that the lower the liquid pressure (in the case where the confinement wall is not dynamically deformed), the lower the nucleation superheat is. Nevertheless, the decrease of the superheat required to the onset of boiling remains weak: only 2 K for the decrease of liquid pressure equal to 10^4 Pa. However the superheat at the boiling incipience strongly depends on the value of the contact angle: the nucleation superheat for $\theta=90$ degrees is 20 K lower than the nucleation superheat for $\theta=10$ degrees. Thus, if by dynamic effects, the value of the contact angle is significantly increased then the superheat required to the nucleation can be sharply decreased. Thus the next study consists in imposing the oscillation of the liquid pressure over time (the confinement wall is dynamically deformed). The change in liquid pressure leads to the moving of the contact line: the value of the contact angle changes (dynamic contact angle phenomenon).

The contact angle imposed here is no longer the static contact angle because the change of the volume of the embryo implies the change of the velocity on the contact line as well as the variation of the contact angle. To evaluate the value of the contact angle in function of the velocity of the contact line, Seeberg's law is applied [5]. To simplify the model, the values of the receding and advancing contact angles are supposed equal to the static contact angle (10 degrees) of the

experimental case. The temperature and the angle of the aperture of the cavity are maintained constant as well as the number of non-condensable moles which is determined in using the previous method. Thus, the variable of the system is the liquid pressure: $p_l(t) = p_{l,avg} - \Delta p_l \sin(2\pi ft)$. The averaged pressure $p_{l,avg}$ as well as the frequency f and the amplitude Δp_l of the oscillation of the liquid pressure are the parameters of the model.

The parametric study consists in reporting the effects of the frequency and of the amplitude of the liquid pressure oscillation on the dynamic contact angle ($\theta(t)$) to determine the maximal variation of the contact angle. Indeed, as previously mentioned, a great variation of the contact angle induces the important decrease of the nucleation superheat.

Figure 7 reports the effect of the frequency of the oscillation of the liquid pressure on the maximum amplitude of the contact angle variation ($\Delta T = 20\text{K}$, $\Delta p_l = 10^4\text{ Pa}$). Even for a strong increase of the frequency (up to 1500 Hz), the amplitude of the variation of the contact angle remains weak (up to 5 K). Figure 8 reports the effect of the amplitude of the oscillation of the liquid pressure on the amplitude of the variation of the contact angle ($\Delta T = 15\text{K}$, $f = 1500\text{ Hz}$). The contact angle increases with the increase of the amplitude of the oscillation of liquid pressure. Nevertheless, the change in contact angle value is not very important.

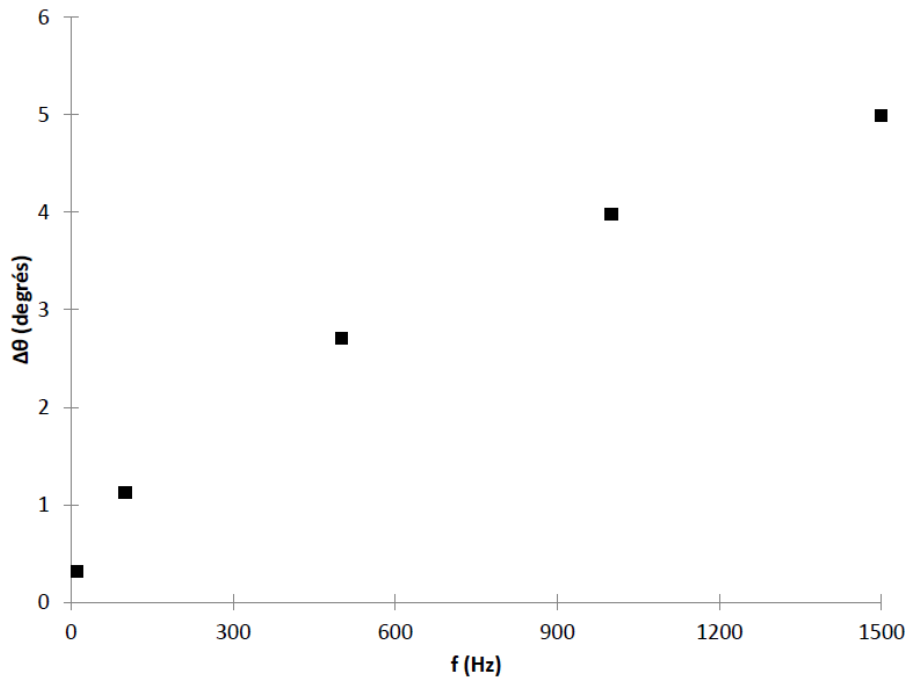


Figure 7: Effect of the amplitude of the oscillation of the liquid pressure on the amplitude of variation of the contact angle ($\beta = 10$ degrees, $n_i = 8.1829 \times 10^{-19}$ mol, $\Delta T = 20\text{ K}$, $\Delta p_l = 10^4\text{ Pa}$)

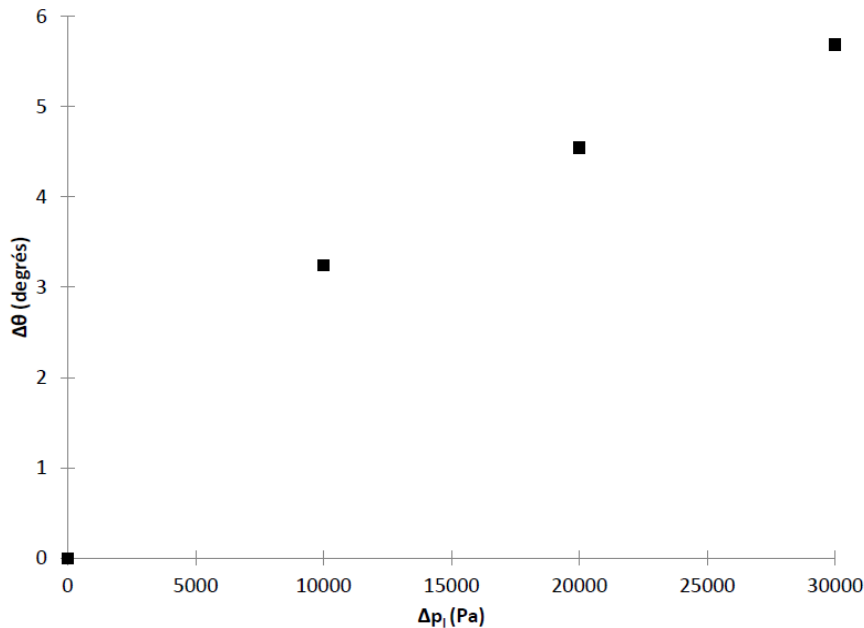


Figure 8: Effect of the amplitude of the oscillation of the liquid pressure on the amplitude of variation of the contact angle ($\beta=10$ degrees, $n_l=8.1829e-19$ mol, $\Delta T= 15$ K, $f=1500$ Hz)

The first conclusion of this study is that the decrease in the static liquid pressure has a weak effect on the superheat required to the onset of boiling (almost 2 K for a decrease in the liquid pressure of 0.1 bar). The boiling incipience appears to be more sensitive to the dynamic of the contact angle. A variation of this contact angle during an oscillation from 10 degrees to 90 degrees implies about 20K of reduction in the wall superheat at the onset of boiling. Thus, if dynamical effects lead to the change in the contact angle, it appears possible to obtain the onset of nucleation at very low superheat. As the liquid pressure is changed over time, the contact line moves with a certain velocity that changes the value of the contact angle. So, the effect of the dynamic (including hysteresis) of the contact angle appears as being the possible mechanism promoting nucleation in dynamic configuration. The evaluation of the variation of the contact angle is realized in using the Seeberg's law. The results obtained show that the variation of the contact angle is weak. Nevertheless, the Seeberg's law was developed to study the dynamic contact angle of a droplet of liquid which the characteristic dimension is 1 mm. In the case considered here, the characteristic dimension is about 10^{-7} m. The validity of the Seeberg's law is then questionable. Thus, further studies are required to quantify the effect of the velocity of the contact line on the value of the contact angle at nanoscale, and consequently to determine the amplitude of the variation of the contact angle in function of the oscillation of the liquid pressure.

4. Conclusion

It has been experimentally shown that imposing a periodic deformation of the wall (and as a consequence a pressure oscillation) confining a heated liquid may drastically reduce the wall superheat at the onset of the nucleate boiling. Theory considering the existence of a vapor embryo trapped within a cavity and attached to the aperture radius cannot reproduce such a decrease in the wall superheat at ONB. To explain these results, the effect the dynamic of the contact angle as well as its hysteresis within the nucleation site prior the onset of the nucleate boiling may be considered. To validate the role of these two parameters, further studies are required, in particular regarding the correlation between the dynamic contact angle and the velocity of the triple line in a cavity of very low order of magnitude in dimension.

5. Acknowledgement

Financial support from CNRS Energie CITAMPE PR09-3.1.3-2 and FNRAE SYRTIPE are gratefully acknowledged

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