Nucleation inception temperature in boiling due to rapid heating - A universal correlation

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Abstract

The boiling temperature, referred to as the saturation temperature is well known for an equilibrium process. It is defined, in a similar way to any thermodynamic state, by two independent thermodynamic properties, e.g. pressure and specific vapor. The collection of these points is referred to as the binodal.

Van-Der-Waals (VDW), however, "disagrees". Nucleation, according to his cubic equation of state, will occur at the spinodal, which is the collection of the thermodynamic states that are the limit of thermodynamic stability. The region that is bounded between the binodal and the spinodal is referred to as the metastable region.

We know that nucleation can occur at any point between the binodal and the spinodal. Furthermore, it is known that for a real liquid (not lacking of some impurities) the penetration depth into the metastable region depends on the heating rate. Moreover, the kinetic theory points out that for an infinite penetration rate, the thermodynamic limit of stability (the spinodal) will be reached.

What, if then, is the nucleation temperature as a function of the heating rate?

We propose a new simple universal correlation that predicts the onset of nucleation temperature following a rapid isobaric heating process, for intermediate to high temperature gradients ($10^5..10^9$ K/s). The prediction process does not require a-priori knowledge of the surface morphology. The correlation predicts the nucleation temperature, as a function of the heating rate, with knowledge of only the saturation and homogeneous nucleation conditions for various liquid types. The predictions are validated for different fluids: water, methanol, ethanol (polar) and heptane and toluene (non-polar). The unique approach presented here allows the prediction of the nucleation temperature without the need to characterize the morphology of the surface on which nucleation occurs.

Keywords

Onset of nucleate boiling ; Rapid heating ; Metastable liquid; Flux peaks

Introduction

Rapid heating occurs in many processes that span from inkjet printers [2], electronic devices [3], chill down of pipes by cryogenic propellant before space engines re-ignition [1] and accidents in the nuclear industry [4]. Rapid heating is a crucial element in the operation of the inkjet printers as they serve as the driving force for the creation of the bubble pump that leads to the well-controlled ink-jet. Per contra, flux peaks that lead to momentarily hot-spots is a phenomenon that should be understood in order to avoid damage. Nowadays, when micro-electronics world is dominating the market, hot-spots probably cannot be avoided, but if properly modeled, can be controlled so that normal operation of the device can be obtained. Rapid heating hold a potentially harmful effect especially for reactivity insertion accident (RIA), that again, if properly modeled can be appropriately handled.

The complete heat removal process requires a unified model that incorporates sub-models for the entire ebullition process, such as bubble growth, detachment, coalescence etc. The current study focus solely on the physical phenomenon of onset of nucleation in pool boiling.

Still, it is challenging to predict where within the metastable region (see Fig. 1), will the nucleation process begin. Bar-Kohany and Amsalem [5] suggested a simple universal model to predict the onset of nucleation temperature for different real liquids due to rapid heating processes following a rapid isobaric heating processes, for intermediate to high temperature gradients ($10^5..10^9$ K/s).

Thermodynamic model

It is well known that rapid processes, either heating [9, 10, 11] or depressurization [6, 7, 8], lead to the onset of nucleation to occur within the meta-stable zone. These studies and others often aimed to find out the maximal heating or depressurization rates to reach the spinodal, since it is established that the faster the processes, the deeper the penetration into the metastable zone.

Fig. 1 presents the various thermodynamic states and iso-curves (isobars & isotherms) pertaining to a phase change. For an isobaric process at P_1 the temperature can be rapidly increased from its initial value (T_1) to the nucleation temperature within the metastable zone, that depends on the temperature escalation rate $T_n(dT/dt)$. At that temperature, the equilibrium vapor pressure is lower than the appropriate saturation pressure ($(P_{ve} < P_s(T_1))$. It will coincide with the spinodal pressure ($P_{SP}(T_1)$) only at the theoretically infinite rates, that in practice are at the order of 10^9 K/s.

The penetration depth can be characterized by the Gibbs or the Jakob numbers (Eq. (1) and Eq. (2)). These dimensionless numbers represent the ratio between the sensible to the latent heat and the ratio between the work
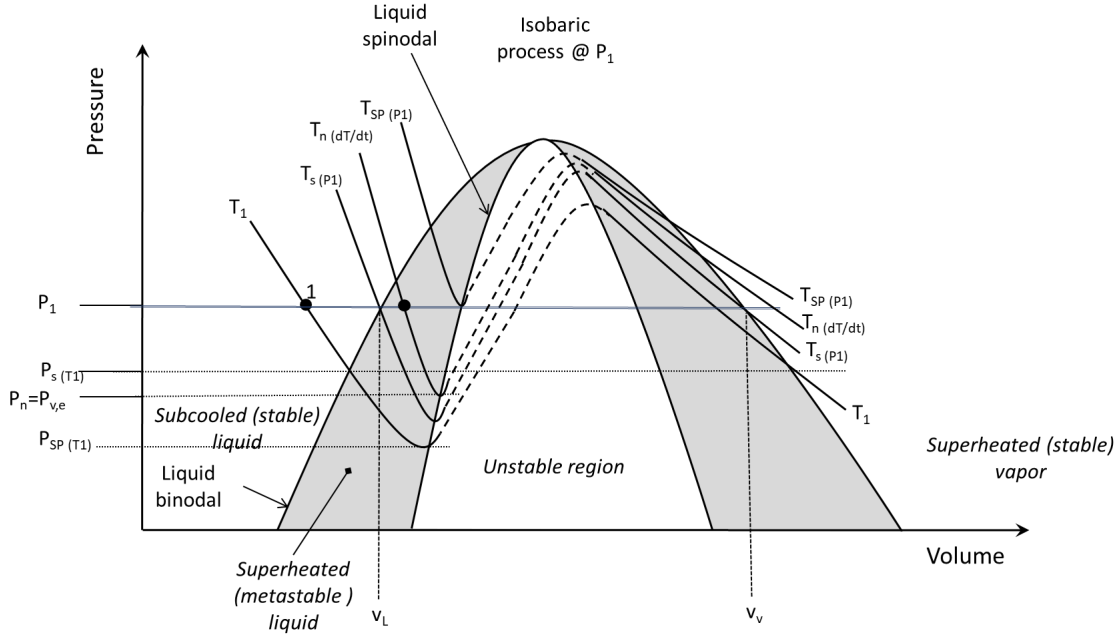
Figure 1. P-v diagram.

required to form a critical vapor embryo vs. the energy required to distance molecules form one another in order to change the phase of the fluid.

$$Gb \equiv \frac{W_c}{k_B T}\phi = \frac{16\pi\sigma^3}{3k_B T(P_L - P_{v,e})^2}\phi \qquad (1)$$

Where ρ_v and ρ_L are the vapor and liquid density values at the saturation conditions. P_L is the liquid pressure and $P_{v,e}$ is the pressure within the vapor bubble at equilibrium; namely, this is the pressure within the bubble that corresponds to the nucleation temperature (and can be considered as the nucleation pressure for an isothermal process).

$$Ja = \frac{\rho_L}{\rho_v}\frac{C_{p,L}(T - T_s)}{h_{fg}} \qquad (2)$$

Where $C_{P,L}$ is the heat capacity of the liquid and h_{fg} is the latent heat of vaporization.

These are prominent thermodynamic potentials, and as such, do not consider kinetic aspects of the process. Thus, correction functions are introduced to take into account the dynamics of the process. The effects of rapid heating ($\dot{T} \equiv dT/dt$) and depressurization ($\Sigma \equiv dP/dt$) are characterized separately by unique functions. In this paper, we will discuss an isothermal process, thus only two correction functions are considered. Interested readers are referred to [5] for the full development.

$$\frac{Gb}{\phi} = \sqrt{\frac{1}{f_1(P_1, T_1)f_2(\dot{T})}} \qquad (3)$$

$$f_1(P_L) \propto \frac{T_s}{\Delta T_s}$$
$$f_2(\dot{T}) \propto \dot{T}^{c/Ja_{HN}} \qquad (4)$$
$$\Delta T_s(P_1) = \frac{v_v}{h_{fg}}\sqrt{\frac{16\pi\sigma^3}{3k_B}T_s}$$

As we discuss here an isobaric process, the pressure difference that appears in the denominator of the Gibbs number should be transformed into a temperature difference. This is performed with the aid of the Clausius-Clapeyron equation. Although this is certainly an approximation, it is a common practice in phase change processes (see for example [12, 13]) and even more, it is concluded that its effect is not dramatic.

Combining Equations 1..4, yields the final, simple universal correlation for the nucleation temperature following a rapid isobaric process, as a function of the temperature escalation rate:

$$\Delta T_n = C \cdot T_s \dot{T}^{c/Ja_{HN}} \qquad (5)$$

Results and discussion
The following figure presents the normalized nucleation temperatures as a function of the temporal gradient of the temperature for various fluids: water, methanol, ethanol, heptane, toluene that were addressed in our previous work [5] and an additional fluid: butanol. The nucleation temperature values were normalized with respect to the critical temperature.

As expected, the nucleation temperature increases as the heating rate increases. Yet, its increase rate depends on each fluid. Mainly, as we have identified, the Jakob number is a key factor in determining the slope of the curves. Higher maximal Jakob number values (Ja_{HN}, see Table 1) are indication of higher penetration into the metastable zone; namely larger energy quantities are required in order to achieve the same level of superheating degree as compared to a fluid with a lower Jakob number. Hence, water, who's Jakob number is the largest, displays the most moderate slope of all of the fluids that were examined.
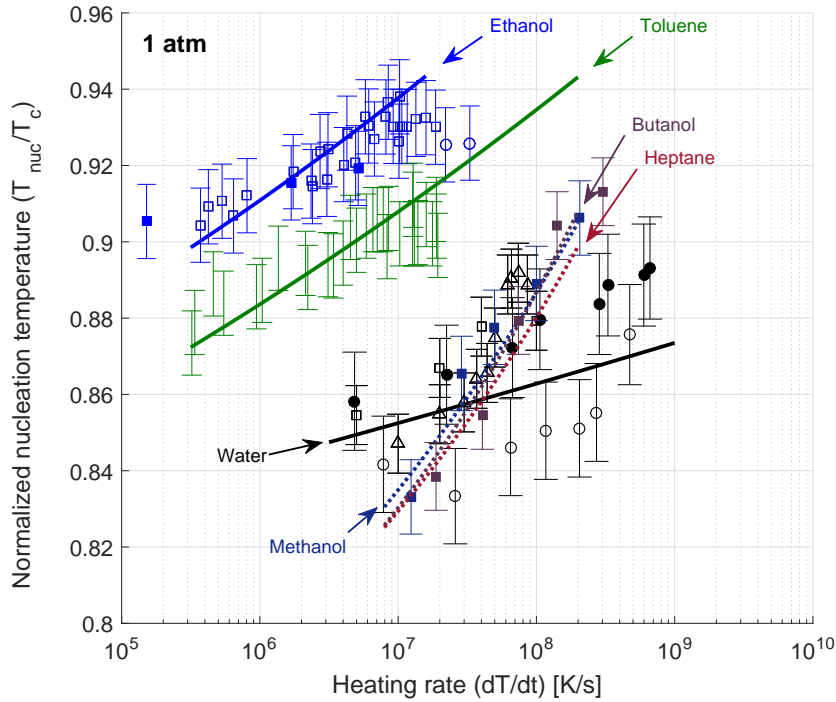
Figure 2. P-v diagram.

Table 1. Jakob numbers at homogeneous nucleation conditions.

Fluid	Water	Methanol	Ethanol	Toluene	Butanol	Heptane
Ja_{HN}	626	198	206	236	146	136

In order to simplify the correlation and to make it easy to use by any engineering evaluation, we suggested to use an easily known value of the Jakob number at the spinodal limit ($Ja(T_{HN})$), rather than the appropriate Jakob number for each heating rate ($Ja(\dot{T})$). This approach spared the need for an iterative method. Table 2 presents the appropriate values of the constants that were used for the various fluids. In our previous publication we set the numerator of the Jakob power to be $c = 10$ for all the fluids, originating from different studies of different researchers. The pre-factors (C) were all of the same order of magnitude, and varied between ~ 0.1 to ~ 0.4. We noticed, though, that all the results that originated from Ching et. al [2] had a steeper slope. Though we do not fully understand the reason, we found that for this group of experiments, a different power constant is more pertinent ($c = 22$), which leads to a reduction of the pre-factors (C) by approximately an order of magnitude, as seen in Table 2.

Table 2. Constant.

Fluid	Water	Methanol	Ethanol	Toluene	Butanol	Heptane
C	0.370	0.045	0.170	0.205	0.0174	0.0245
c	10	22	10	10	22	22

Conclusions

The universal correlation that was presented here enables the prediction of the nucleation temperature of various different fluids, due to a rapid isobaric process. The correlation can be easily incorporated to any numerical or engineering calculation, and is most appropriate for intermediate to high heating rates ($10^5..10^9$ K/s). One prominent advantage of this correlation, and approach, is the ability to predict the nucleation temperature without the need to characterize the morphology of the surface.

We show that using the Jakob number as the denominator of the Jakob power, the acceleration and deceleration nature of the nucleation process, with respect to the heating rate, is accounted. The results show good agreement for the results that were found in the literature.

The current correlation was developed to atmospheric pressure, with accordance to the available experimental results. It is expected, however, to be applicable for different pressure values, by somewhat modifying the numerator of the Jakob power. The authors are currently engaged with designing an experimental apparatus that will enable that modification. The pre-factor is expected to be a function of the surface morphology, especially for lower heating rates. The authors are planning to examine the extent of this approach on lower heating rates as well.

Acknowledgements

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