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Effect of capillarity at liquid–vapor interface on phase change without surfactant

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Abstract

Thermodynamic analysis has been employed in this paper to study the effect of capillarity on the interfacial phase change of liquid without surfactant, and a modified expression of phase change rate of liquid without surfactant has been developed. The new expression demonstrates that the capillarity of the concave liquid surface will promote the interfacial vaporization if sufficient heat is exerted on liquid, and the capillarity of the convex liquid surface will be helpful for increasing the condensation rate from vapor to liquid. The analyses of this paper showed that the newly developed expression is in accordance with practical observations reported in the literature. © 2002 Elsevier Science Ltd. All rights reserved.

1. Introduction

Phase change is of great importance in the thermal engineering field because of the huge transfer of latent heat during the phase change processes. For instance, boiling and condensation are two common and important heat removal approaches by phase change. For the formation of a vapor bubble in liquid boiling, the interfacial vaporization occurring at the liquid–vapor interface near the heated wall sustains the growth of the bubble. Heat is transferred from the heated wall to the vapor within bubble via the adsorption of latent heat during this interfacial vaporization. Meanwhile, vapor condensation into liquid will result in latent heat release and the corresponding heat transfer [1,2]. Due to the continued miniaturization of semiconductor devices, power electronics, biosensors and aerospace equipment, problems associated with overheating of these components have increased significantly. Accordingly, the innovative cooling techniques are required definitely to meet the demands of heat load removal from highly integrated electronic circuits and the electronic components of spacecraft designed for advanced long-term spacecraft missions. Those demands result in the rapid development of improved heat rejection techniques [3,4] such as the interfacial vaporization and condensation heat transfer of thin liquid film. Consequently, the study of the heat transfer of interfacial phase change is of fundamental importance to industrial applications.

The mechanism of phase change at liquid–vapor/gas interface has been extensively studied in decades based on the molecular kinetic theory. The fundamental expression of interfacial phase change rate was developed via those investigations as [5,6]

$$J = \left(\frac{M}{2\pi R T_{\mathrm{iv}}}\right)^{1/2} (C_1 P_{\mathrm{iv}}(T_{\mathrm{iv}}) - C_2 P_{\mathrm{v}}), \tag{1}$$

where J is the phase change rate, positive for the interfacial evaporation rate, and negative for the interfacial condensation rate; C_1 and C_2 are the accommodation coefficients of vaporization and condensation, respectively; M is the molecular weight; R is the general gas constant; $P_{\mathrm{iv}}(T_{\mathrm{iv}})$ is the saturated vapor pressure with respect to the interfacial temperature T_{iv}; P_{v} is the bulk vapor pressure.

Eq. (1) shows that the difference of interfacial and bulk vapor pressure is the basic driving force for interfacial

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Nomenclature

A	surface area of liquid–vapor interface
C	accommodation coefficient
F	free energy
f	thermodynamic fugacity of liquid
ΔH	ideal molar heat of vaporization
h_{fg}	latent heat
J	phase change rate
K	curvature of liquid surface
k	thermal conductivity
M	molecular weight
n	mole number of vapor at interface
P	pressure
q	heat flux
R	gas constant
r	radius of spheric surface
T	temperature
V	volume
Greek symbols	
δ	thickness of liquid film
γ	surface tension
ρ	density
Subscripts	
iv	liquid and vapor interface
l	liquid
v	vapor
w	wall

phase change. To express this pressure difference with common thermodynamic parameters is of great importance for the employment of this relation. For thermodynamic equilibrium or quasi-equilibrium, the interfacial saturation pressure, $P_{\mathrm{iv}}(T_{\mathrm{iv}})$ is equal to the practical interfacial pressure. For non-equilibrium, the saturated vapor pressure $P_{\mathrm{iv}}(T_{\mathrm{iv}})$ is actually the ideal vapor pressure, i.e., the fugacity of the corresponding liquid [7], which is related to interfacial free energy, and so, the superheat, practical interfacial pressure, and factors with interfacial capillarity and dispersion interaction [8]. The effects of superheat and practical interfacial pressure have been studied extensively, and right understanding has been got. However, the effect of interface capillarity in phase change is still unclear. This paper is devoted to exhibiting this effect on the base of thermodynamic analysis.

2. Review on the previous researches

Currently, three typical expressions have been developed from the fundamental relation, Eq. (1), in the thermal engineering field as given below. Among them, the expression developed by Wayner and co-workers [9–12] is most widely used.

The original reduction of the Wayner et al. expression [9] was given as

$$\mathrm{d}\ln f_{\mathrm{l}} = \frac{V_{\mathrm{l}}}{RT_{\mathrm{l}}}\mathrm{d}P_{\mathrm{l}} + \frac{\Delta H^{\mathrm{i}}}{RT_{\mathrm{l}}^2}\mathrm{d}T_{\mathrm{l}}, \tag{2}$$

where f_{l} is the fugacity of liquid; and ΔH^{i} is the ideal molar heat of vaporization for a liquid expanding to a vacuum, which can be approximated by the ordinary latent heat Mh_{fg}. The question arose from the integration path chosen by Wayner et al. [9]. They selected the integration path: $(P_{\mathrm{v}}, T_{\mathrm{v}}) \to (P_{\mathrm{v}}, T_{\mathrm{iv}})$ for the first term on the right-hand side of Eq. (2); $(P_{\mathrm{v}}, T_{\mathrm{iv}}) \to (P_{\mathrm{l}}, T_{\mathrm{iv}})$ for the second term on the right-hand side of Eq. (2); and $(P_{\mathrm{v}}, T_{\mathrm{v}}) \to (P_{\mathrm{iv}}, T_{\mathrm{iv}})$ for the term on the left-hand side of Eq. (2), that is,

$$\int_{(P_{\mathrm{v}},T_{\mathrm{v}})}^{(P_{\mathrm{iv}},T_{\mathrm{iv}})} \mathrm{d}\ln f_{\mathrm{l}} = \int_{(P_{\mathrm{v}},T_{\mathrm{v}})}^{(P_{\mathrm{v}},T_{\mathrm{iv}})} \frac{Mh_{\mathrm{fg}}}{RT_{\mathrm{i}}^2}\mathrm{d}T_{\mathrm{i}} + \int_{(P_{\mathrm{v}},T_{\mathrm{iv}})}^{(P_{\mathrm{l}},T_{\mathrm{iv}})} \frac{V_{\mathrm{l}}}{RT_{\mathrm{l}}}\mathrm{d}P_{\mathrm{l}}. \tag{3}$$

The integration path of the left- and right-hand sides of Eq. (3) is not consistent, so that the integration derived by Wayner et al. as

$$\ln\frac{P_{\mathrm{iv}}(T_{\mathrm{iv}})}{P_{\mathrm{v}}} = \frac{Mh_{\mathrm{fg}}}{RT_{\mathrm{v}}T_{\mathrm{iv}}}(T_{\mathrm{iv}} - T_{\mathrm{v}}) + \frac{V_{\mathrm{l}}(P_{\mathrm{l}} - P_{\mathrm{v}})}{RT_{\mathrm{iv}}} \tag{4}$$

may be invalid, the interfacial saturation pressure, as well as the expression of phase change rate, proposed by Wayner et al. from Eq. (4) would be questionable [8].

Faghri and his co-workers [4,13] proposed an expression of interfacial phase change rate as

$$J = -\left(\frac{2\alpha}{2-\alpha}\right)\left(\frac{M}{2\pi R}\right)^{1/2}\left[\frac{P_{\mathrm{v}}}{T_{\mathrm{v}}^{1/2}} - \frac{P_{\mathrm{iv}}}{T_{\mathrm{iv}}^{1/2}}\right] \tag{5}$$

and

$$P_{\mathrm{iv}} = P_{\mathrm{iv}}(T_{\mathrm{iv}})\exp\left[\frac{P_{\mathrm{iv}} - P_{\mathrm{iv}}(T_{\mathrm{iv}}) + P_{\mathrm{d}} - \gamma K}{\rho_{\mathrm{l}}RT_{\mathrm{iv}}}\right]. \tag{6}$$

They declared that Eq. (6) was the so-called Kelvin equation. However, comparing Eq. (6) with the original version of the Kelvin equation quoted in [7,14–16], they misarranged the pressure terms in the Kelvin equation by misunderstanding the physical meaning of vapor and saturated vapor pressure, and hence, the expression of Faghri et al. [4,13] would still be questionable.

The expression of interfacial phase change rate by Carey [17] is

$$J = -\left(\frac{2\alpha}{2-\alpha}\right)\left(\frac{M}{2\pi R}\right)^{1/2}\left[\frac{P_v}{T_v^{1/2}} - \frac{P_l}{T_l^{1/2}}\right]. \tag{7}$$

Confusion arises from comparing Eq. (7) with the fundamental expression of the interfacial phase change rate, Eq. (1). The saturated vapor pressure in Eq. (1), $P_{iv}(T_{iv})$, was replaced with P_l in Eq. (7) by Carey. However, the relation, $P_l = P_{iv}(T_{iv})$, is merely valid for flat liquid–vapor interface in thermodynamic equilibrium, whereas no net phase change occurs at the interface. For non-thermodynamic equilibrium, this replacement is prohibited, and so, Eq. (7) may be also questionable.

In short, the confusions existed in the expressions of interfacial saturation pressure, so the expression of interfacial phase change rate in heat transfer field. As a result, it is necessary to develop a new expression of interfacial phase change rate to clarify those confusions.

3. A new expression proposed for the interfacial phase change rate

Fig. 1 is the physical model for the study herein. Phase change will occur once the unbalance of free energy, i.e., thermodynamic fugacity, exerted at liquid–vapor interface. Fugacity, f, is the measure of the escaping tendency of vapor molecules in phase change of thermodynamic non-equilibrium, as [7,18]

$$\mathrm{d}\ln f = \frac{\mathrm{d}F}{RT} = \frac{1}{\rho RT}\mathrm{d}P + \frac{Mh_{fg}}{RT^2}\mathrm{d}T + \frac{\gamma \mathrm{d}A_{iv}}{n_{iv}RT} + \frac{\mathrm{d}P_d}{\rho RT}, \tag{8}$$

where A_{iv} is the surface area of liquid–vapor interface, F is the specific free energy, n_{iv} is the mole number of vapor at interface, P_d is the disjoining pressure [19], γ is the surface tension of liquid–vapor interface, and ρ is the molar density. The surface area of liquid–vapor interface, A_{iv}, is related to the surface curvature. In general, the differential area of curved surface can be considered to be a part of sphere surface. For this consideration, the differential area, $\mathrm{d}A_{iv}$, relates to the differential volume of the sphere, $\mathrm{d}V_{iv}$ and the radius of the surface curvature by [7]

$$\mathrm{d}A_{iv} = \frac{2\mathrm{d}V_{iv}}{r}n_{iv}, \tag{9}$$

where r is the radius of the spheric surface, positive for concave liquid surface and negative for convex liquid surface; V_{iv} is the molar specific volume.

Substituting Eq. (9) into Eq. (8) and then integrating with the integration path from (P_v, T_v) to (P_{iv}, T_{iv}), yield

$$\ln\left(\frac{f_{iv}}{f_v}\right) = \frac{1}{\rho_{iv}RT_{iv}}\Delta P_{iv} + \frac{Mh_{fg}}{RT_{iv}}\left(\frac{T_{iv}-T_v}{T_v}\right) + \frac{2\gamma}{r\rho_{iv}RT_{iv}} + \frac{P_d}{\rho_{iv}RT_{iv}}, \tag{10}$$

where ΔP_{iv} is the practical pressure difference between the interface and the bulk vapor, i.e., $\Delta P_{iv} = P_{iv} - P_v$, whereas P_{iv} is the practical vapor pressure at the liquid–vapor interface, and P_v is the bulk vapor pressure adjacent to the interface; ρ_{iv} is $1/V_{iv}$. Accordingly, the bulk vapor pressure is equal to its fugacity [7], or $P_v = f_v$; and the ideal interfacial vapor pressure, $P_{iv}(T_{iv})$, is actually the interfacial fugacity, f_{iv}. As a result, Eq. (10) can be expressed by vapor pressure as

$$\ln\left(\frac{P_{iv}(T_{iv})}{P_v}\right) = \frac{1}{\rho_{iv}RT_{iv}}\Delta P_{iv} + \frac{Mh_{fg}}{RT_{iv}}\left(\frac{T_{iv}-T_v}{T_v}\right) + \frac{2\gamma/r + P_d}{\rho_{iv}RT_{iv}}. \tag{11}$$

If the liquid and vapor adjacent to the interface are not far from thermodynamic equilibrium, the saturated vapor pressure, $P_{iv}(T_{iv})$, will be nearly equal to the bulk vapor pressure, P_v, so that, by remaining the first order term, the logarithmic term of Eq. (11) can be expanded approximately to

$$P_{iv}(T_{iv}) - P_v = \frac{P_v}{\rho_{iv}RT_{iv}}\Delta P_{iv} + \frac{P_vMh_{fg}}{RT_{iv}}\left(\frac{T_{iv}-T_v}{T_v}\right) + \frac{P_v}{\rho_{iv}RT_{iv}}\left(\frac{2\gamma}{r} + P_d\right). \tag{12}$$

According to Plesset and Prosperetti [5], the two accommodation coefficients, C_1 and C_2, in Eq. (1) are approximately equal if phase change is near thermodynamic equilibrium. With this consideration, incorporating Eq. (12) into Eq. (1) yields

$$J = C\left(\frac{M}{2\pi RT_{iv}}\right)^{1/2}\left[\frac{P_v}{\rho_{iv}RT_{iv}}\Delta P_{iv} + \frac{P_vMh_{fg}}{RT_{iv}T_v}(T_{iv}-T_v) + \frac{P_v}{\rho_{iv}RT_{iv}}\left(\frac{2\gamma}{r} + P_d\right)\right], \tag{13}$$

where C is the accommodation coefficient. The first term of the right-hand side of Eq. (13) represents the effect of

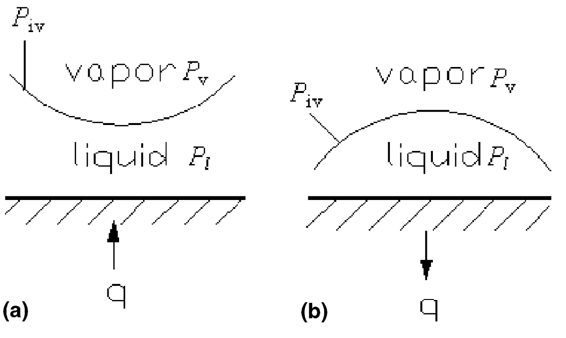

Fig. 1. Physical model, where P_l and P_v represent liquid and vapor pressures, respectively.

practical interfacial vapor pressure and bulk vapor pressure on phase change. Obviously, the positive pressure difference is to promote interfacial vaporization, while the negative pressure difference is to increase interfacial condensation. The second term represents the effect of superheat or subcooling on interfacial phase change. The first part of the third term stands for the effect of capillarity, it is positive for interfacial vaporization on concave liquid surface or for interfacial condensation on convex liquid surface. The second part in the third term represents the effect of disjoining pressure for extreme thin film.

For convenience to apply Eq. (13) in practice, the original superheat in Eq. (13) was related to the wall superheat in the following reduction. Provided that heat transfer is one-dimensional heat conduction through thin liquid film, we have

$$q_{\mathrm{w}} = \frac{k}{\delta}(T_{\mathrm{w}} - T_{\mathrm{iv}}) \tag{14}$$

and

$$T_{\mathrm{iv}} = T_{\mathrm{w}} - \frac{\delta}{k}q_{\mathrm{w}}, \tag{15}$$

where k is the thermal conductivity of liquid film, q_{w} is the wall heat flux, T_{w} is the wall temperature, and δ is the thickness of the thin liquid film.

Incorporating Eq. (15) into Eq. (13) yields the expression of interfacial phase change rate as

$$J = aC\left(\frac{M}{2\pi RT_{\mathrm{iv}}}\right)^{1/2}\left[\frac{P_{\mathrm{v}}}{\rho_{\mathrm{iv}}RT_{\mathrm{iv}}}\Delta P_{\mathrm{iv}} + \frac{P_{\mathrm{v}}Mh_{\mathrm{fg}}}{RT_{\mathrm{iv}}T_{\mathrm{v}}}(T_{\mathrm{w}} - T_{\mathrm{v}}) + \frac{P_{\mathrm{v}}}{\rho_{\mathrm{iv}}RT_{\mathrm{iv}}}\left(\frac{2\gamma}{r} + P_{\mathrm{d}}\right)\right], \tag{16}$$

where

$$a = \left[1 + h_{\mathrm{fg}}C\left(\frac{M}{2\pi RT_{\mathrm{iv}}}\right)^{1/2}\frac{P_{\mathrm{v}}Mh_{\mathrm{fg}}}{RT_{\mathrm{iv}}T_{\mathrm{v}}}\frac{\delta}{k}\right]^{-1}. \tag{17}$$

It would be better to define $(T_{\mathrm{w}} - T_{\mathrm{v}})$ as the wall-superheat to distinguish from the superheat $(T_{\mathrm{iv}} - T_{\mathrm{v}})$ defined in Eq. (13).

4. Discussions

Eq. (13) or (16) demonstrates that phase change is dominated by difference of practical vapor pressure, superheat and pressure difference of capillarity and disjoining pressure. In order to extrude the effect of capillarity on phase change, it was provided that the system is in isothermal state, the practical pressure difference, $(P_{\mathrm{iv}} - P_{\mathrm{v}})$, of Eq. (12), is zero, and the disjoining pressure which is only significant for extreme thin film is ignored. With this consideration, the only factor acting on phase change is the capillarity at liquid–vapor interfacial surface. Eq. (12) obviously indicates that for convex liquid surface, whereas the radius of curvature is negative, the capillarity promotes the bulk vapor pressure, P_{v}, to make it larger than the saturated vapor pressure, $P_{\mathrm{iv}}(T_{\mathrm{iv}})$, so that the vapor over the convex liquid surface is in supersaturated state [7,8,14–16]. On this line, Eq. (13) or (16) predicts that the capillarity at convex liquid surface is to promote interfacial condensation. This conclusion is in accordance with the theoretical deduction of the Kelvin equation [7,14–16]. As exhibited by the Kelvin equation [7] and Eq. (13), the bulk vapor pressure over a small droplet will be larger than that of a plane surface of the same liquid. As a result, the supersaturated vapor over the convex liquid surface will spontaneously condense into liquid as stated out by the experimental observations [7]. Those experimental observations demonstrated that uniform-sized droplets of a nonvolatile solvent produced and then equilibrated with the vapor over a solution of the same solvent and a volatile solute rapidly grew to a larger size spontaneously.

Eq. (12) has indicated that the capillarity at the concave liquid surface produces a positive vapor pressure difference. Consequently, the capillarity at concave liquid surface is to promote evaporation once heat is applied to liquid. As pointed out [7], if the liquid surface is concave towards the vapor, the fugacity is decreased instead of being increased, whereas the radius of curvature in Eq. (12) will be positive. Therefore, the chemical potential of the vapor over the interface is lower than the saturated value of the same vapor temperature, and the vapor is superheated as pointed out by Keenan [15]. For this situation, the tendency of thermodynamic process is that, molecules of higher chemical potential at liquid surface are to evaporate into vapor of lower chemical potential if heat is applied to liquid. The difference from that of convex liquid surface is that the vaporization cannot work spontaneously. Heat must be brought into liquid to help liquid molecule to escape into vapor by getting over the energy barrier at interface. As a result, vaporization due to the effect of capillarity cannot maintain unless sequent heat is applied.

In order to highlight the effect of capillarity, the interfacial evaporation rate as a function of the capillarity at concave liquid surface of different liquids was estimated under isothermal limit. Those estimations implied that the required heat has been applied to liquid to maintain the isothermal state of liquid during evaporation. The accommodation coefficient in estimation was chosen as $C = 1$ [5] and the bulk vapor pressure was $P_{\mathrm{v}} = 1.01325 \times 10^{5}$ Pa. The results are illustrated in Fig. 2 that the promotion of interfacial evaporation by the capillarity at the concave liquid surface is enhanced with decreasing the radius of curvature. At the same time, the estimations with the questionable model of Wayner et al. [9–12] are also presented in Fig. 2 for comparison. The

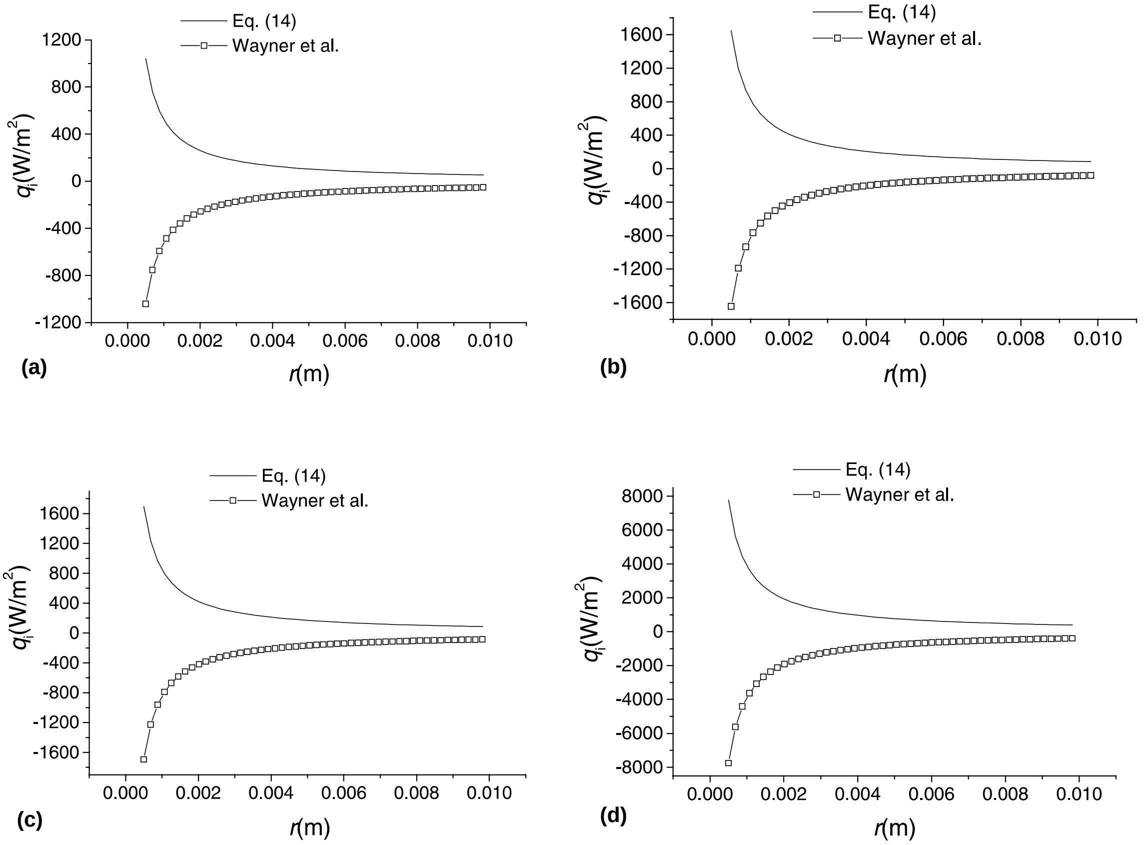

Fig. 2. Effect of the capillarity of concave liquid surface on the interfacial evaporation: (a) acetone; (b) ethanol; (c) methanol; (d) water.

model of Wayner et al. suggested that the effect of capillarity at concave liquid surface would cause interfacial condensation, which opposes the conventional understanding [7,15,16] about the phase-change tendency mentioned above. The comparisons in Fig. 2 indicate that the discrepancy between the new proposed model and that of Wayner et al.'s model is enlarged significantly as the latent heat of liquid and the curvature of the concave liquid surface increase.

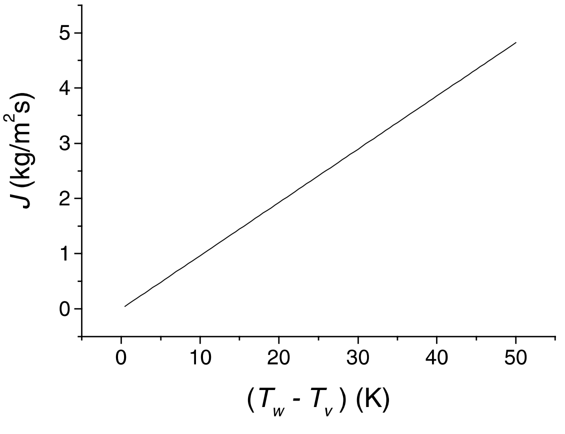

Fig. 3. Effect of superheat on the interfacial evaporation.

Fig. 3 illustrates an estimation of the interfacial evaporation rate as a function of the variation of the water film superheat, at $T_{iv} = 100$ °C and $P_v = 1.01325 \times 10^5$ Pa with the film thickness of 0.1 mm. For such thick film, the disjoining pressure can be neglected. The accommodation coefficient was the same as that for the calculations of Fig. 2. Fig. 3 illustrates that the effect of capillarity is significant when the wall superheat is not high.

5. Conclusion

The review conducted in this paper exhibited that confusions exist with the models of phase change in thermal engineering fields. The analyses demonstrated that those confusions arise from ignoring the difference between the saturated and practical vapor pressure in thermodynamic non-equilibrium. A new proposed model of phase change has been derived in this paper.

The new proposed model indicates that the capillarity of convex liquid surface promotes the vapor pressure over the interface to make the vapor supersaturated. As a result, the capillarity of convex liquid surface promotes the condensation tendency of the vapor over the surface. For the concave liquid surface, the effect of capillarity depresses the vapor pressure to make the vapor superheated over the interface so as to promote the tendency of evaporation. The analyses demonstrate that these conclusions are in accordance with the classical Kelvin equation and the experimental observations.

Estimations demonstrate that the effect of capillarity is significant when superheat is small, enhanced with increasing the latent heat of working fluid and curvature of liquid surface.

Acknowledgements

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