

Forced Convection Subcooled Boiling of Binary Mixtures

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A model based on an additive mechanism of heat transfer is proposed for forced convection subcooled boiling of binary mixtures. The contributing modes of heat transfer are: (i) the heat transferred as latent heat by the rising bubbles, (ii) the heat transferred as the heat contained in the superheated thermal layer that is removed from the surface in the wake of the rising bubbles and (iii) the single phase forced convection heat transfer from the heating surface not influenced by the bubbles. Experimental data from the literature on binary systems show good agreement with the model, validating the postulated mechanism.

On propose un modèle basé sur un mécanisme de transfert de chaleur additif pour une ébullition sous-refroidie par convection forcée de mélanges binaires. Les modes de contribution de transfert de chaleur sont les suivants: 1° la chaleur transférée comme chaleur latente par les bulles montantes; 2° la chaleur transférée comme chaleur contenue dans la couche thermique surchauffée qui est retirée de la surface dans le sillage des bulles montantes; 3° le transfert de chaleur par convection forcée de la phase unique à partir de la surface de chauffe ne subissant pas l'influence des bulles. Les données expérimentales issues de la littérature sur les systèmes binaires montrent un bon accord avec le modèle, ce qui valide le mécanisme supposé.

Keywords: boiling, subcooled binary mixtures, forced convection, heat flux.

Subcooled boiling is a phenomenon where boiling occurs at the heating surface, followed by condensation in the colder bulk of the fluid without net generation of vapour. The liquid bulk is maintained below its saturation temperature, while the heating surface is above the liquid saturation temperature. In this study attention is focussed on forced convection subcooled boiling of binary mixtures as distinguished from pool boiling. Though published information on forced convection boiling of binary mixtures is available, very little is known with regard to the heat flux and heat transfer coefficient under subcooled conditions. Combining subcooled boiling of liquids with forced convection, extremely high heat flux densities can be attained. This makes it useful for such varied applications as cooling of rocket motors, nuclear accelerator targets, high temperature pressure transducers, high power electromagnets and other high temperature probes. While earlier studies were limited to single component systems, mainly water, this study concentrates on binary mixtures such as acetone-water, isopropanol-water and *n*-butanol-water. In subcooled boiling of binary mixtures maximum heat fluxes appear to coincide with low vapour production rate at the heating surface using systems of suitable compositions. This aspect raises the possibility of using binary mixtures as cooling media in heat exchangers and nuclear reactors.

Previous work

There are a number of empirical correlations for subcooled boiling, especially of single component liquids, and these have been reviewed by Sivagnanam (1984) and Sivagnanam and Varma (1990). Sivagnanam and Varma (1990) have given an empirical correlation, based on their own experiments, for estimating the heat transfer coefficient of subcooled boiling of binary mixtures under forced convection. They have reported that for a given driving force, ΔT_w the liquid velocity has a positive influence on the surface heat flux in the partial boiling region and ceases to influence in the fully developed region. The authors further noted that the higher the subcooling, the larger is the effect of liquid velocity on heat flux. Furthermore, as the concentration of

the more volatile component increases in the binary mixture, a higher wall super-heat is required to attain the same level of heat flux. This is particularly true for those compositions of binary mixtures whose absolute values of mass fraction difference of vapour and liquid increase with increase in concentration.

In this study attention is confined to the development of a mechanistic model to describe the subcooled forced convection boiling of binary mixtures. A brief survey of the mechanisms proposed by earlier investigators to explain the high rate of heat removal associated with bubble generation is presented below. Though the models pertain to single component boiling or pool boiling of binary mixtures, they are reviewed in order to provide continuity in the development of the proposed model for subcooled forced convective boiling of binary mixtures.

A 'microconvection' hypothesis assumes that the growing and departing vapour bubbles are followed by a back flow of cold liquid to the wall, agitating the liquid near the wall and thereby reducing the thermal resistance of the thin boundary layer adjacent to the heating surface. Hence, very high heat transfer rates are encountered [Gunther and Kreith (1950); Ellison (1953); Forster and Zuber (1954)]. Forster and Grief (1959) extended the microconvection model to the boiling of subcooled liquids, and proposed the 'vapour-liquid exchange model'. The model, developed primarily for subcooled nucleate boiling, assumes that when a bubble detaches from the heating surface, the hot liquid from the thermal layer is pushed into the bulk liquid and cold liquid replaces the bubble at the surface. Similarly, when a bubble collapses cold liquid is brought into contact with the heating surface. Thus, the individual vapour bubbles act as tiny, efficient pumps, pushing hot liquid from the thermal layer into the bulk liquid and drawing colder liquid from the bulk to the heating surface. The experimental results of Hsu (1962) showed evidence favouring the vapour-liquid exchange model.

A further extension of the above model is to postulate that the rising bubble entrains in its wake a portion of the thermal layer and transports it to the bulk liquid, while the cold liquid flows back to the heating surface to be superheated by transient conduction. Based on this 'transient conduction' hypothesis, Han and Griffith (1965) postulated the 'bulk convection' mechanism for saturated boiling. Characterising

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the total heat transfer area into (i) 'influenced' and (ii) 'not influenced' by the departing bubbles, the authors assumed that the area of influence for the thermal layer, drawn into the bulk liquid by a departing bubble, is equal to a circular area having a radius twice that of the departing bubble. Beer and Durst (1968) modified the above model and postulated a drift current behind the bubble, based on their calculations of the particle paths behind rising bubbles with the help of potential theory and interferometric investigations. They assumed that the total boiling heat transfer was the sum of three components: natural convection in the area not influenced by bubbles, the heat of vapourisation of bubble vapour content and the heat that was removed from the surface by the drift current.

Van Stralen (1970) proposed a 'relaxation microlayer hypothesis' which postulates that the bubble dome is surrounded by a superheated 'relaxation microlayer' to form an initially uniform thermal boundary layer at the wall which is pushed away by the rapidly growing bubbles. The excess enthalpy stored in this relaxation microlayer is entirely used for latent heat of vapourisation. Before bubble initiation occurs, that is in the waiting period, heat is presumed to be transferred to the liquid by transient conduction, as envisaged by Han and Griffith (1965). Snyder and Robin (1969) postulated the existence of an 'evaporation microlayer' of liquid beneath the bubble after its initial formation. Heat is transmitted through this microlayer, the thickness of which decreases with evaporation.

The model

A model is developed below for the forced convection subcooled boiling heat transfer of binary mixtures, taking into cognisance the models described in the earlier section. It is assumed that heat is removed from the heating surface during boiling by the following mechanisms:

- (a) the latent heat contained in the bubbles (Q_{b1})
- (b) the heat contained in the superheated thermal boundary layer that is removed periodically from the heating surface by the bubble in its wake (Q_{tb})
- (c) the heat transferred by single phase forced convection heat transfer (Q_{fc}).

Expressing these rates as heat fluxes, the total flux is

$$q_{tot} = q_{b1} + q_{tb} + q_{fc} \dots \dots \dots (1)$$

It is further assumed that the total area of heat transfer is divided into: i) the surface affected by bubbles (area of influence) and ii) the surface affected by forced convection heat transfer. Each bubble is assumed to be associated with an area of influence equivalent to twice its diameter [Han and Griffith (1965)]. Within this area a superheated thermal boundary layer develops which rises upwards with the bubble during bubble growth and is eventually pushed into the colder core in the wake of the bubble. Cold liquid flows into the space behind the ascending bubble and the entrained hot liquid, and comes into contact with the heating surface. This liquid is mainly heated through transient conduction whereby a new superheated boundary layer originates and the process repeats. The superheated layer is displaced at the rate of bubble frequency. The heat flux due to the heat content of the departing bubbles is obtained from an energy balance as

$$q_{b1} = (\pi d_m^3 / 6) \cdot \rho_v \cdot \lambda (N/A) \cdot f \dots \dots \dots (2)$$

The heat flux from the heating surface over an area of influence, a , due to removal of superheated layer by bubble entrainment is also obtained from an energy balance as

$$q_{tb} = a \delta \rho_L C_p (T_w - T_L) (N/A) f \dots \dots \dots (3)$$

Van Stralen (1966) observed that the thickness of the conduction layer can be estimated from the conservation equations or from extrapolation of the convection region of the boiling curve to higher superheating in nucleate boiling. The thickness of the thermal boundary layer for single components, valid for both superheating and subcooling, is given by Van Stralen and Cole (1979) as

$$\delta = k / h_{fc} \dots \dots \dots (4)$$

This relation is assumed to be applicable to binary mixtures and inserted into Equation (4) to yield

$$q_{tb} = a \rho_L C_p (T_w - T_L) (k / h_{fc}) (N/A) f \dots \dots \dots (5)$$

The heat flux by single phase forced convection transfer over an area $\{1 - (N/A)a\}$, uninfluenced by bubbles is given by

$$q_{fc} = h_{fc} \{1 - (N/A)a\} (T_w - T_L) \dots \dots \dots (6)$$

The forced convection heat transfer coefficient, h_{fc} , is obtained from appropriate correlations depending upon flow geometry and flow condition.

The nucleation site density is evaluated from the experimental data (Figures 13 and 14 of Van Stralen and Sluyter, 1969) reported for boiling of water using vertical platinum wire as

$$(N/A) = 3.62 \times 10^4 (\Delta T_s)^{1.1} \dots \dots \dots (7)$$

The above dimensional equation is applicable for the specified material — platinum wire only.

The heat transfer from the heated surface to the liquid occurs in periodically repeating steps, namely, the growth time period, t_g and the waiting period, t_w . The waiting time between succeeding bubbles is assumed to be three times the growth time (Van Stralen et al., 1975). The total mean heat flux density at the area of influence during the entire cycle associated with bubble formation and growth may be written as

$$q_m = (q_{b1} t_g + q_{tb} t_w) / (t_g + t_w) \dots \dots \dots (8)$$

Equation (1) is now written as

$$q_{tot} = q_m + q_{fc} \dots \dots \dots (9)$$

To determine q_{b1} and q_m using Equations (2) and (8), it is necessary to estimate the maximum bubble diameter (d_m) and bubble growth time t_g during subcooled nucleate boiling. Based on a heat transfer controlled bubble model for subcooled nucleate flow boiling of water, Unal (1976) presented semi-empirical correlations for prediction of maximum bubble-departure diameter and maximum bubble-growth time (see appendix for details). These equations are now extended to binary mixtures.

Van Stralen et al. (1975) observed that the bubble growth in pure liquids depends on heat flow towards the bubble boundary to meet the heat requirement for evaporation. In mixtures, thermal diffusion is associated with an analogous slower mass diffusion of the more volatile component. Since $D \ll \alpha$, the bubble growth rate is retarded with binary

mixtures as compared for single component fluids at the same level of superheat. Moreover, the superheating available for bubble growth for binary mixtures is reduced by an amount $\Delta\theta$ due to simultaneous mass diffusion. The following assumptions are made in extending the equations due to Unal (1976) to binary mixtures:—

1. The liquid film at the base of the growing bubble is partially stripped of the more volatile component since its vapour mole fraction is greater than its liquid mole fraction. Consequently, the local liquid mole fraction at the base of the bubble becomes richer in the less volatile component. As a result, the superheating available for the bubble growth is reduced by an amount $\Delta\theta$ requiring the necessary correction in Equation (A-5).
2. The thickness of the thermal boundary layer is the same for boiling of single component liquids and for binary mixtures when $x \ll 1$.
3. The method used for estimating the latent heat and other transport and thermodynamic properties for single component liquids are applicable for binary mixtures as well.

Calus and Leonidopoulos (1974) estimated the reduction in apparent temperature driving force for pool boiling of binary mixtures in terms of the vapour liquid equilibrium relationship, and their transport and thermodynamic properties as:

$$\Delta\theta = \Delta T_S \cdot F / (1 + F) \dots\dots\dots (10)$$

where

$$F = |y - x| (\alpha / D)^{0.5} (C_p / \lambda) dt / dx$$

Therefore, incorporating the correction for the available superheat, Equation (A-5) is modified to

$$a_1 = (\Delta T_S - \Delta\theta) k_L v / 2 \rho_v \lambda (\pi \beta_1)^{1/2} \dots\dots\dots (11)$$

Comparison with experimental data

The model is compared with the experimental data (Sivagnanam, 1984) of the following binary mixtures: (1) Acetone–water with $x = 0.05, 0.1$ and 0.15 and subcooling $\Delta T_{SUB} = 40, 30$ and 20°C ; (2) Isopropanol–water with $x = 0.05$ and subcooling of 40 and 30°C and (3) *n*-Butanol–water with $x = 0.02$ and $\Delta T_{SUB} = 40$ and 20°C .

The experimental set-up consisted of a vertical glass tube 47 mm I.D. and 590 mm long with a heating element consisting of a 0.3 mm diameter and 495 mm long platinum wire stretched at the centre line of the glass tube through which the subcooled binary liquid was allowed to flow upwards (Sivagnanam and Varma, 1990). The experiments were conducted at atmospheric pressure. The flux was measured through the voltage across and current through the wire. The feed to the test section was from a pre-heating tank with thermostatic control.

Results and discussion

Figures 1 to 3 show the boiling heat flux plotted against the temperature driving force $(T_w - T_L)$. The line represents the model prediction while the experimental data are shown as individual points. Since the present situation corresponds to longitudinal flow of liquid over a cylindrical heating element confined in a glass tube, the correlation by Stasiulevichius (1970) is used to estimate the forced convection nonboiling heat transfer coefficient, h_{fc} , for use in Equation (6):

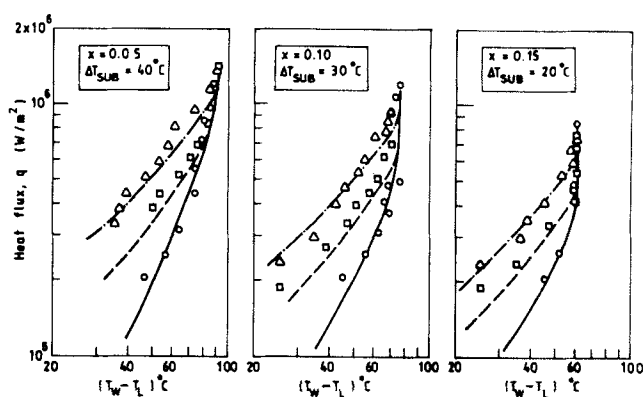


Figure 1 — Comparison of model with experimental data Acetone — Water mixtures; Experimental: \circ $v = 0.16$ m/s, \square $v = 0.55$ m/s, Δ $v = 1.0$ m/s; Theoretical: — $v = 0.16$ m/s, — — $v = 0.55$ m/s, - · - · $v = 1.0$ m/s.

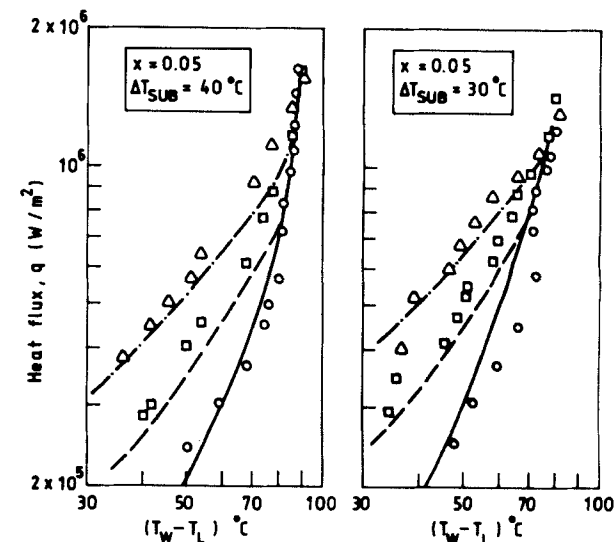


Figure 2 — Comparison of model with experimental data isopropanol — water mixtures; Experimental: \circ $v = 0.16$ m/s, \square $v = 0.55$ m/s, Δ $v = 1.0$ m/s; Theoretical: — $v = 0.16$ m/s, — — $v = 0.55$ m/s, - · - · $v = 1.0$ m/s.

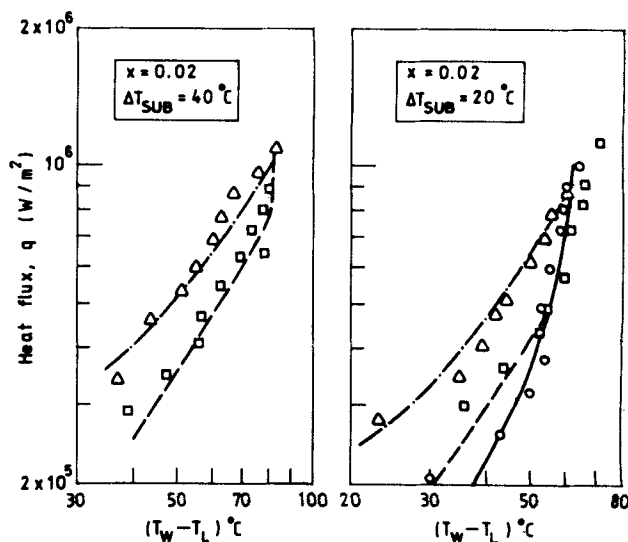


Figure 3 — Comparison of model with experimental data *n*-butanol — water; Experimental: \circ $v = 0.16$ m/s, \square $v = 0.55$ m/s, Δ $v = 1.0$ m/s; Theoretical: — $v = 0.16$ m/s, — — $v = 0.55$ m/s, - · - · $v = 1.0$ m/s.

TABLE I
Physical Properties of Binary Mixtures at Saturation Temperature, T_s

x (-)	y (-)	T_s K	ρ_L kg/m ³	ρ_v kg/m ³	$\lambda \times 10^6$ J/kg	C_p J/kg · C	$\mu \times 10^{-3}$ Pa · s	k W/m · C	Pr (-)	σ N/m	$\alpha \times 10^{-6}$ m ² /s	$D \times 10^{-9}$ m ² /s
<i>Acetone - Water Systems</i>												
0.05	0.645	362	552	1.11	2.19	4370	0.286	0.492	2.54	0.0396	0.132	0.676
0.10	0.790	354	545	1.40	2.12	4180	0.285	0.468	2.54	0.0408	0.133	0.603
0.15	0.804	348	583	1.45	2.05	4020	0.284	0.446	2.56	0.0418	0.133	0.553
<i>Isopropanol - Water Systems</i>												
0.05	0.530	365	840	0.971	2.20	4460	0.290	0.494	2.61	0.0386	0.132	2.00
0.10	0.695	360	823	1.210	2.13	4340	0.293	0.471	2.70	0.0390	0.132	1.63
<i>n-Butanol - Water Systems</i>												
0.02	0.370	369	855	0.839	2.24	4520	0.287	0.508	2.55	0.0384	0.132	1.56
0.04	0.450	368	850	0.921	2.21	4460	0.287	0.499	2.57	0.0386	0.132	1.27

$$St = 0.183 [\log(Re_z)]^{-2.45} (T_w/T_L)^{-0.25} \Omega \dots (12)$$

where $\Omega = 1 + 0.37(z/r)^{0.8} (Re_r)^{-0.2}$ and T_w and T_L are in °K.

The physical properties of the binary liquid mixtures are estimated using established methods as summarized by Holland et al. (1970). Table I shows the property values of the binary mixtures at their saturation temperatures. Similarly, property values at any other liquid temperature can also be determined. There is good agreement between the experimental data and the model predictions, proving the validity of the model.

The model predicts the partial boiling region at low temperature driving force and the fully developed boiling region at high driving force for the forced convection subcooled boiling of binary mixtures. The liquid velocity and the degree of subcooling do not have a significant effect on the boiling heat flux in the fully developed boiling region. This is in agreement with the observations of McAdams et al. (1949), Jens and Lottes (1951), Clark and Rohsenow (1954), Bergles and Rohsenow (1964) and Thom et al. (1965) made in respect of boiling of single component liquids.

The figures indicate that an increase in temperature driving force increases the heat flux rapidly in the fully developed boiling region. This can be attributed to the fact that the contribution of the heat flux due to forced convection becomes less significant than the contribution of the boiling component in this region. This is in agreement with the observations of Rohsenow (1964) for single component liquids.

The model takes into account the mass transfer resistance, caused by the presence of a second liquid in the mixture. This results in the reduction of the apparent temperature driving force. To establish the applicability of the model for the limiting case of single component boiling (i.e., $x = 0$), experimental data using water is compared in Figure 4. While the model matches well the experimental data obtained using platinum wire as the heating surface, the deviation of the data using Inconel alloy heating element (Lung et al., 1977) indicates the specificity of Equation (7).

Summary

A model to predict the heat flux in forced convection subcooled boiling of binary mixtures has been proposed assuming an additive mechanism for heat transfer. A bubble cycle consisting of a growth period and a waiting period

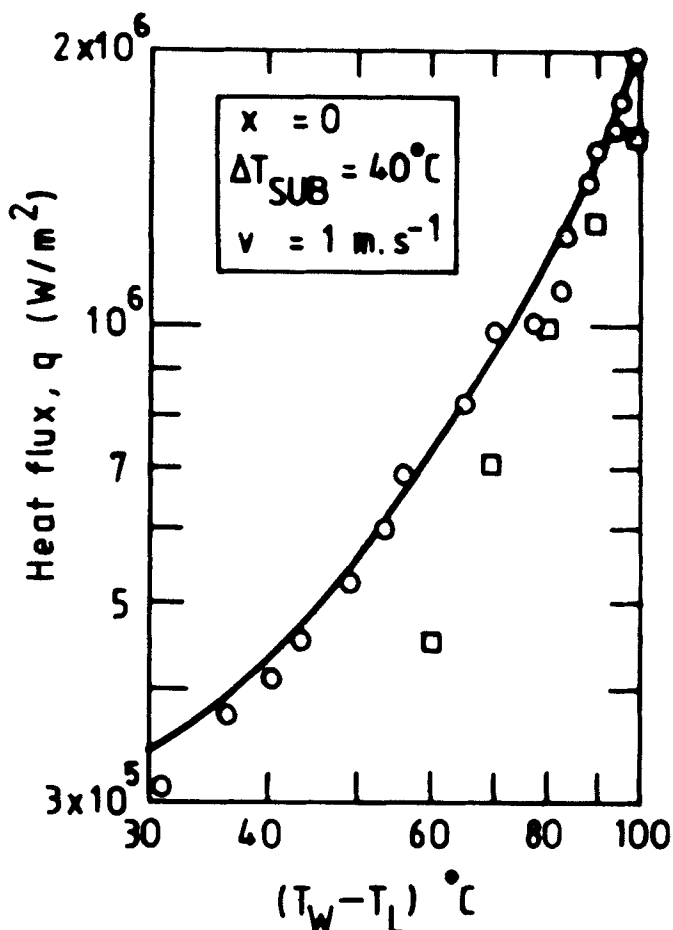


Figure 4 — Comparison of model with experimental data. Pure water; \circ Sivagnanam (1984); \square Lung et al. (1977); — Model prediction.

is visualised. Heat transfer from the heating surface to the liquid occurs by the following mechanisms: (i) the departing bubble carries an amount of heat equivalent to the latent heat of the mass of the bubble, (ii) the bulk liquid rushes towards the nucleation site and gets superheated and this represents the waiting period, and (iii) the total heat transfer area is characterised into an 'area of influence' and an area uninfluenced by bubbles; it is assumed that heat is transferred in the latter region by single phase forced convection mechanism.

The model predictions agree well with the observations based on experimental data obtained in forced convection

subcooled boiling of single component liquids as well as binary liquid mixtures. The additional diffusional resistance, caused in binary mixtures, to heat transfer in forced convective boiling is similar to that reported for pool boiling.

APPENDIX

Considering a lumped formulation of subcooled flow boiling bubble model, Unal (1976) presented the following equations for maximum bubble diameter, d_m and for maximum bubble growth time t_g :

$$d_m = 1.21 a_1 \beta / (bc\phi)^{1/2} \dots\dots\dots (A-1)$$

$$t_g = 1/1.46(bc\phi) \dots\dots\dots (A-2)$$

where

$$\phi = 1 \text{ for } v \leq 0.61 \text{ m/s} \\ = (v/0.61)^{0.47} \text{ for } v > 0.61 \text{ m/s} \dots\dots\dots (A-3)$$

$$c = 65 - 5.69 \times 10^{-5} (P - 10^5) \dots\dots\dots (A-4)$$

$$a_1 = k_L v \Delta T_S / 2 \rho_v \lambda (\pi \beta_1)^{1/2} \dots\dots\dots (A-5)$$

$$b = \Delta T_{SUB} / 2 [1 - (\rho_v / \rho_L)] \dots\dots\dots (A-6)$$

$$\beta = 1 - (d_d^2 / d_b^2) \dots\dots\dots (A-7)$$

$$\Delta T_S = [(q - h \Delta T_{SUB}) / C_1]^3 \dots\dots\dots (A-8)$$

$$\beta_1 = k_S / \rho_S C_{pS} \dots\dots\dots (A-9)$$

$$v = k_W \rho_W C_{pW} / k_S \rho_S C_{pS} \dots\dots\dots (A-10)$$

$$C_1 = \lambda \mu_L \left[\frac{C_p}{0.013 \lambda P_n^{1.7}} \right]^3 / \frac{\sigma}{[(\rho_L - \rho_v)g]^{1/2}} \dots\dots\dots (A-11)$$

The range of application of the above equations is $0.1 < P < 17.7$; $0.47 < q < 10.64$; $0.08 < v < 9.15$; $3 < \Delta T_{SUB} < 86$ (Unal, 1976). Taking into account the additional mass transfer resistance due to the presence of the second liquid, the available superheat, ΔT_S is corrected to give Equation (11).

Nomenclature

- a = area of influence of bubble on heating surface, m
- a_1 = parameter defined in Equation (A-5)
- A = area of heating surface, m
- b = parameter defined in Equation (A-6)
- c = parameter defined in Equation (A-4), MN/m²
- C_1 = parameter defined in Equation (A-11), W/m² · C³
- C_p = specific heat of liquid, J/kg · C
- d_b = bubble diameter, m
- d_d = diameter of dry area under bubble, m
- d_m = maximum bubble diameter, m
- D = mass diffusivity, m²/s
- f = frequency of bubbles, s⁻¹
- G = mass velocity, kg/m² · s
- h = heat transfer coefficient, W/m² · C
- k = thermal conductivity, W/m · C
- N = number of active nucleation sites
- P = pressure, MN/m²
- Q = heat transfer rate, W
- q = heat flux (= Q/A), W/m²
- Re = Reynolds number (= $d \cdot v \cdot \rho / \mu$)
- St = Stanton number (= $h / C_p \cdot G$)

- t_g = bubble growth time, s
- t_w = waiting period, s
- T = temperature, K
- ΔT_S = (= $T_w - T_S$), degree of superheating, C
- ΔT_{SUB} = (= $T_S - T_L$), degree of subcooling, C
- ΔT_w = (= $T_w - T_L$), temperature driving force, C
- v = liquid velocity, m/s
- x = weight fraction of more volatile component in liquid mixture
- y = weight fraction of more volatile component in vapours
- z = length, m

Greek letters

- α = thermal diffusivity, m²/s
- β = parameter defined in Equation (A-7)
- β_λ = thermal diffusivity of liquid at saturation, m²/s
- δ = thickness of the thermal layer, m
- λ = latent heat of vapourisation, J/kg
- μ = viscosity, Pa · s
- ν = parameter defined by Equation (A-10)
- ρ = density, kg/m³
- σ = surface tension, N/m
- ϕ = parameter defined by Equation (A-3), m/s

Subscripts

- b = boiling
- fc = single phase forced convection heat transfer
- L = liquid
- r = based on radius
- S = at saturation temperature
- SUB = subcooling
- tb = thermal boundary layer
- tot = total
- v = vapour
- W = wall
- z = based on length

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