

BUBBLE GROWTH RATES IN PURE AND BINARY SYSTEMS: COMBINED EFFECT OF RELAXATION AND EVAPORATION MICROLAYERS

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Abstract—Pohlhausen's equation has been used to determine the initial thickness of the evaporating microlayer beneath a hemispherical vapour bubble on a superheated horizontal wall. Microlayer thickness is proportional to the square root of the distance to the nucleation site during early bubble growth, while a linear relationship exists during advanced growth.

A (heat and mass) diffusion-type solution is derived for advanced bubble growth, which accounts for the interaction of the mutually dependent contributions due to the relaxation microlayer (around the bubble dome) and the evaporation microlayer. The entire bubble behaviour during adherence is determined by a combination of this asymptotic solution and the Rayleigh solution, which governs early growth. Also, expressions are derived for both the radius of the dry area and the radius of the maximum contact area between bubble and wall.

At low concentrations of the more volatile component in binary systems, the dominating influence of mass diffusion is demonstrated by the following effects: (i) asymptotic bubble growth is slowed down substantially; (ii) the formation of dry areas beneath bubbles is prevented, even at subatmospheric pressures; (iii) the lower part of the bubble is contracted; (iv) the evaporation microlayer contribution to bubble growth is negligible at atmospheric and at elevated pressures.

NOMENCLATURE

a , = $k/\rho_1 c$, liquid thermal diffusivity [m^2/s];
 A_1, A_2, A_3 , bubble growth factor in pure liquid [$\text{m}/\text{s}^{1/2}\text{K}$];
 b, b^* , dimensionless bubble growth parameter during adherence;
 B , = $(2\rho_2 l\theta_0/3\rho_1 T)^{1/2} = R/t$, factor in Rayleigh solution [m/s];
 c , liquid specific heat at constant pressure [J/kgK];
 C_1 , = $C_{1,p} = R/\theta_0 t^{1/2}$, bubble growth constant in pure liquid [$\text{m}/\text{s}^{1/2}\text{K}$];
 $C_{1,m}$, bubble growth constant in binary mixture [$\text{m}/\text{s}^{1/2}\text{K}$];
 C_2 , = $R/t^{1/2}$, bubble growth factor [$\text{m}/\text{s}^{1/2}$];
 $d(r, t)$, thermal boundary-layer thickness of evaporation microlayer beneath hemispherical bubble [m];
 $d_0(r)$, initial thermal boundary-layer thickness of evaporation microlayer [m];
 d_0^* , initial thickness of relaxation microlayer around curved bubble surface [m];

$d_{w,co}$, thickness of equivalent conduction boundary layer at heating surface [m];
 D , mass diffusivity of more volatile component in less volatile component [m^2/s];
 e , = 2.718, ..., base of natural logarithms;
 F , = $(k\rho_1 c)^{1/2}$ [$\text{J}/\text{s}^{1/2}\text{mK}$];
 g , gravitational acceleration [m/s^2];
 G , vaporized mass fraction;
 H_1 , = $2b^*R(t_1)$, maximal height of spherical bubble segment covered with relaxation microlayer [m];
 Ja , = $(\rho_1 c/\rho_2 l)\theta_0$, Jakob number for superheated pure liquid;
 Ja_m , = $(C_{1,m}/C_{1,p})Ja$, modified Jakob number for superheated binary mixture;
 k , liquid thermal conductivity [W/mK];
 K , equilibrium constant of more volatile component in binary mixture (ratio of mass fractions of vapour and liquid);
 l , latent heat of vaporization [J/kg];
 m , exponent in $R^* \sim t^m$;
 n , exponent in $R_m^* = (C_{1,m}/C_{1,p})^n R^*$;
 Nu , = $q_w r/k\theta_0$, local Nusselt number;
 p , ambient pressure [$Pa = \text{N}/\text{m}^2 = \text{kg}/\text{ms}^2$];
 Δp , excess pressure on vapour side of bubble boundary [kg/ms^2];
 Pr , = ν/a , liquid Prandtl number;

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- q , heat flux density [W/m^2];
 r , radial distance from bubble centre [m];
 R , $= R^*/2^{1/3}$, equivalent spherical bubble radius [m];
 R_0 , $= 2\sigma T/\rho_2 l\theta_0$, equilibrium bubble radius [m];
 $R(t_1)$, equivalent spherical bubble departure radius [m];
 R_1 , equivalent bubble radius according to modified Rayleigh solution [m];
 R_2 , $= A_3 t^{1/2}$, equivalent bubble radius according to total diffusion (combined evaporation and relaxation microlayer) solution [m];
 $R_{2,1}$, $= A_1 t^{1/2}$, equivalent bubble radius accounting for evaporation microlayer [m];
 $R_{2,2}$, $= A_2 t^{1/2}$, equivalent bubble radius accounting for relaxation microlayer [m];
 R^* , radius of hemispherical bubble [m];
 R_c^* , radius of contact area between bubble and heating surface [m];
 R_d^* , radius of dry area beneath hemispherical bubble [m];
 R_f^* , hemispherical bubble radius according to Rayleigh solution [m];
 R_g^* , hemispherical bubble radius according to Cooper solution [m];
 R^+ , $= BR/C_2^2$, dimensionless bubble radius;
 Re , $= r\dot{R}^*/\nu$, local Reynolds number;
 t , bubble growth time during adherence or during delay [s];
 t_1 , bubble departure time [s];
 t_2 , waiting time between succeeding bubbles [s];
 t_c , instant of maximum contact radius [s];
 t_d , instant of complete evaporation of microlayer [s];
 t_r , instant at which $R^*(t_r) = r$ [s];
 t^+ , $= (B/C_2)t$, dimensionless bubble growth time;
 t_2^+ , $= (B/C_2)^2 t_2$, dimensionless bubble waiting time;
 T , absolute boiling temperature [K];
 $T(x)$, absolute boiling temperature of liquid at bubble boundary in binary mixture [K];
 $T(x_0)$, absolute boiling temperature of original liquid in binary mixture [K];
 ΔT , $= T(x) - T(x_0)$, increase in temperature of liquid at bubble boundary with respect to original liquid [K];
 x , $= x_0/\{1 + (K-1)G\}$, mass fraction of more volatile component in liquid at bubble boundary in binary mixture;
 x_0 , mass fraction of more volatile component in original binary mixture.

Greek symbols

- $\delta(r, t)$, hydrodynamic boundary-layer thickness of evaporation microlayer [m];
 $\delta_0(r)$, initial hydrodynamic boundary-layer thickness of evaporation microlayer [m];
 η , liquid dynamic viscosity [kg/sm];
 θ , uniform superheating of relaxation microlayer around curved bubble surface [K];
 θ_0 , superheating of heating surface [K];
 θ_0^* , asymptotic superheating of contact surface between two semi-infinite bodies [K];
 θ_R^* , superheating of bubble vapour-liquid interface [K];
 θ_w , wall superheating [K];
 $\Delta\theta_0$, superheating of bulk liquid [K];
 ν , $= \eta/\rho_1$, liquid kinematic viscosity [m^2/s];
 ρ_1 , liquid density [kg/m^3];
 ρ_2 , saturated vapour density [kg/m^3];
 σ , surface tension constant [kg/s^2].

Subscripts

- co , corresponding convective contribution to total heat flux density;
 m , value in binary mixture;
 0 , initial value;
 p , value in pure liquid;
 w , value for heating surface.

1. SCOPE OF THE PRESENT INVESTIGATION

THE PRIMARY object of the present work is an experimental verification and reexamination of the physical background of various bubble growth theories including those based upon microlayer evaporation, cf. [41].

Both bubbles in water at a range of high Jakob numbers (occurring by varying the ambient pressure) and in aqueous binary mixtures with a more volatile component will be investigated experimentally; the mixtures being used to demonstrate the expected predominant slowing-down effect of mass diffusion on the asymptotic stage of bubble growth.

It is of substantial importance to vary the pressure in a certain sub-atmospheric range, as initially hydrodynamic inertia may be expected to govern bubble growth according to: $R \sim (\Delta p)^{1/2} t \sim (p\theta_0)^{1/2} t$ (the asymptotic Rayleigh solution), in contradistinction to the succeeding diffusion-controlled stage, during which $R \sim \theta_0 t^{1/2}/p$.

Bubbles are generated on an artificial nucleus with a radius $R_0 = 2\sigma T/(\rho_2 l\theta_0) \sim 1/(p\theta_0)$, i.e. for a given R_0 the required wall superheating for "equilibrium" bubble formation $\theta_0 \sim 1/p$, as according to the Clausius-Clapeyron equation for an ideal gas

$$dp/dT = \rho_2 l/T \sim p.$$

The present paper is concerned with theory. Forthcoming articles will give experimental results for water and aqueous binary mixtures with a more volatile component. Also, theoretical predictions will be compared to the new experimental data and to the results obtained previously by Stewart and Cole [1].

2. INTRODUCTION ON THE MECHANISM OF BUBBLE GROWTH

The present state of knowledge on vapour bubble growth on a heated horizontal wall at high Jakob numbers—occurring in general at subatmospheric pressures and at high superheatings as $Ja = \rho_1 c \theta_0 / (\rho_2 l) \sim \theta_0 / \rho_2 \sim \theta_0 / p$ —involves the predominant importance of the following governing mechanisms:

(i) The “evaporation microlayer” model

A thin liquid microlayer is present under the hemispherical bubble during a certain stage of growth, which occurs shortly after the initial formation of the equilibrium radius.

Heat is transmitted through the microlayer, the thickness of which is diminishing by evaporation. As a result, a dry surface area is formed around the nucleation site; the radius of this dry spot increases gradually with time. In general, the initial superheating enthalpy of the microlayer is not taken into account, and conductive heat transmission through the layer is assumed, i.e. the laminar liquid flow in the microlayer is neglected.

The concept of a microlayer under a vapour bubble during nucleate pool boiling of water on a flat plate was first proposed by Snyder [2] and later used by Moore and Mesler [3] to explain the occurrence of fluctuations in the local wall surface temperature. Similar observations in liquid metals have been made by Madsen and Bonilla [4].

Bonnet, Macke and Morin [5] showed, that the instant of the rapid temperature dip coincides with the high initial growth rate of the bubble. The local interface temperature then increases gradually to the original superheating shortly before bubble departure. Following departure, another rapid but smaller temperature dip may occur as a result of cold fluid from the bulk impinging upon the surface. The surface temperature again recovers, increasing continuously during the succeeding waiting time, until the next bubble is generated on the same nucleus.

More recently, presence of the microlayer has been shown by thermal effects using very thin temperature-sensitive solid state elements—Cooper and Lloyd [6–8]—and by optical effects using the principle of total reflection of light [9–11] or using interferometry [12, 13].

(ii) The “relaxation microlayer” model

Simultaneously during bubble growth, evaporation occurs at the entire or at the lower part of the liquid–vapour interface at the bubble dome. According to Van Stralen [14, 15], this additional contribution to bubble growth in a non-uniform temperature field is due to the excess enthalpy of a superheated “relaxation microlayer”, which surrounds the dome as the original thermal boundary layer at the wall is pushed away by the initially rapidly growing bubble.

Van Stralen used the transient conduction equation for semi-infinite bodies to determine the heat flux (density) at the heating surface, which is given by [16]:

$$q_w = \frac{F_w}{F_w + F} \frac{F}{(\pi t)^{1/2}} \theta_0 = \frac{F}{(\pi t)^{1/2}} \theta_0^*, \quad (1)$$

where

$$\theta_0^* = \frac{F_w}{F_w + F} \theta_0 \quad (2)$$

denotes the interface superheating in case of a step change in temperature at the contact surface between liquid and wall with initially uniform temperatures T and $T + \theta_0$, respectively. The coefficients $F = (k \rho_1 c)^{1/2}$ and $F_w = (k_w \rho_w c_w)^{1/2}$ refer to the liquid and the wall material.

It follows from equation (1), that the heat flux is determined mainly by the poorest thermal conductor, i.e. equation (1) is simplified to:

$$q_w = \frac{F}{(\pi t)^{1/2}} \theta_0 = \frac{k}{(\pi a t)^{1/2}} \theta_0 \quad (3)$$

in case of $F_w \gg F$, whence $\theta_0^* \rightarrow \theta_0$ for a wall of high conductivity; this case applies to commonly used boiling apparatus. In case of $F_w \ll F$:

$$q_w = \frac{F_w}{(\pi t)^{1/2}} \theta_0, \quad (4)$$

whence $\theta_0^* \rightarrow 0$ for a wall of low conductivity.

(iii) The combination of the evaporation and the relaxation microlayer

Several theoretical models have been presented in literature to describe the contributions of (i) and (ii) to the total bubble growth separately. Until now, the problem of combining both mechanisms has not been solved satisfactorily.

Actually, the situation is complicated by the occurrence of different (hydrodynamically or diffusion-controlled) stages in the bubble growth. These regimes occur already in the more simple case of a free spherical bubble in an initially uniformly superheated infinite, incompressible and inviscid liquid, where the influence of buoyancy pressure, which amounts to $\frac{1}{2} \rho_1 g R$, can be

neglected. The complexity of the problem is demonstrated by the nonlinearity of the second order Rayleigh differential equation of motion, which is derived from the energy equation in combination with continuity.

3. SURVEY OF THEORIES ON BUBBLE GROWTH IN PURE LIQUIDS BOILING AT LOW PRESSURES

3.1. Models involving asymptotic microlayer evaporation

3.1.1. Cooper [8] and Cooper and Lloyd [7] suggested that low system pressure is one of the factors promoting the formation of a liquid microlayer and leading to a hemispherical shape during initial bubble growth. The initial thickness of the hydrodynamic microlayer under a bubble is $\delta_0(r) = \frac{1}{2}(r/R^*)(\pi vt)^{1/2} = \frac{1}{2}(r/R^*)(\pi at)^{1/2} Pr^{1/2}$, obtained by solving a simpler form of the Navier–Stokes equation for the liquid velocity in the microlayer, and by assuming that $R^* \sim t^{1/2}$.

Actually, the value of the constant $\frac{1}{2}\pi^{1/2} = 0.89$ applies to the introduced time exponent of 0.50; a value of 0.80 is in better agreement with experimental results and with the expression

$$\delta_0(r) = \frac{1}{2}\{\pi^2/(\pi^2 + 1)\}(\pi vt_r)^{1/2},$$

which is also given in [7]. According to Cooper *et al.* [7, 8, 17], the neglect of the thermal capacity of the microlayer corresponds to ignoring a vapour volume equal to $Pr/6$ times the bubble volume resulting in discrepancies of 100 per cent at low pressures. The average thermal resistance of the microlayer is taken to be constant at $\delta_0/2k$, which is apparently connected to a reduction of the thickness due to evaporation.

The two resulting asymptotic relations for the extreme cases of a high and a low conductivity wall are based upon conduction relations for heat flow across the microlayer (hydrodynamic case) and through the wall (thermally controlled case), respectively. In general, in accordance with equation (1), the heat transmission is governed by the poorest heat conductor:

$$R^*(t) \cong \frac{4}{\pi^{1/2}} Pr^{-1/2} Ja(at)^{1/2} = 2.26 Pr^{-1/2} Ja(at)^{1/2} \quad (5)$$

for a wall of high conductivity (hydrodynamically controlled case), and

$$R^*(t) \cong \frac{2}{\pi^{1/2}} \frac{F_w}{F} Ja(at)^{1/2} = 1.13 \frac{F_w}{F} Ja(at)^{1/2} \quad (6)$$

for a wall of low conductivity (thermally controlled case).

Equation (5) can be derived easily from the heat flux balance taking hemispherical bubble shape and the mean thermal resistance of the microlayer into account:

$$q_w = 2\rho_2 l \dot{R}^* = 2 \frac{k\theta_0}{\delta_0} = \frac{4k\theta_0}{(\pi at)^{1/2} Pr^{1/2}} \quad (7)$$

In principle, equation (5) can also be derived from equation (3) by replacing the thermal penetration depth, $(\pi at)^{1/2}$, by the corresponding hydrodynamic boundary-layer thickness, $(\pi at)^{1/2} Pr^{1/2}$, of the evaporating microlayer.

Similarly, equation (6) follows directly from equation (4) as:

$$q_w = 2\rho_2 l \dot{R}^* = 2 \frac{F_w}{F} \frac{F}{(\pi t)^{1/2}} \theta_0 = -\rho_1 l \dot{\delta}. \quad (8)$$

Apparently, a factor of 2 has been introduced by Cooper [8] in the second expression of equation (8).

Cooper *et al.* [7, 8, 17] added also a term to the r.h.s. of equations (5) and (6) accounting for evaporation at the curved surface of the bubble dome. This term has been taken from the asymptotic growth equation derived by Forster and Zuber [18] and Plesset and Zwick [19] for a free spherical bubble in an initially uniform superheated infinite liquid:

$$R(t) \cong \left(\frac{12}{\pi}\right)^{1/2} Ja(at)^{1/2} = 1.954 Ja(at)^{1/2}, \quad (9)$$

in which the Jakob number applies to the liquid superheating.

If the bulk liquid is not at uniform temperature initially, the effect of assumed profile of temperature variation is incorporated by an analysis based upon the work of Skinner and Bankoff [20, 21]. For this case, it has been suggested to apply the Jakob numbers in equations (5) and (9) during advanced bubble growth to the bulk liquid superheating or to the remote liquid superheating.

3.1.2. For $F_w \gg F$, Kotake [22] showed theoretically the existence of a hydrodynamic microlayer with $\delta_0(R^*)/R^* \sim (Re)^{-1/2}$, where the bubble Reynolds number is defined as $Re = R^* \dot{R}^*/\nu$. In the thermally controlled case ($F_w \ll F$), $\delta_0(R^*)/R^* \sim \rho_2/\rho_1$, in agreement with equation (8).

Kotake extended equation (2) for the superheating of the wall for the case of $\delta_0(R^*) \gg (at)^{1/2}$ to:

$$\theta_0^*(t) = \frac{F_w}{(\pi at)^{1/2}} \theta_0 \rightarrow \frac{\delta_0(R^*)}{F_w + \frac{F}{\delta_0(R^*)}} \frac{F_w}{F} \theta_0. \quad (10)$$

3.1.3. Van Ouwkerk [10, 11] assumed the hemispherical self-similar bubble growth equation as $R^* \sim t^{1/2}$ (for which the bubble Reynolds number is independent of time) and carried out an elaborate analysis of the Navier–Stokes equation for the hydrodynamically formed laminar microlayer. The initial microlayer thickness is given by the self-similar solution $\delta_0(r) = (r/R^*)\delta_0(R^*) = 1.27 (r/R^*)(vt)^{1/2}$. The final bubble growth relation is connected to the radius of the dry area and to evaporation on the curved surface for the case of an initially uniformly superheated system with $\Delta\theta_0 = \theta_0$, cf. equation (9) for the uniform bulk.

For $F_w = F$:

$$R^*(t) \cong \frac{2}{\pi^{1/2}} (1 + 3^{1/2}) Ja(at)^{1/2} \\ = 1.128(1 + 1.73) Ja(at)^{1/2}, \quad (11)$$

in which case microlayer evaporation accounts, independent of pressure, for 37 per cent of bubble radius, i.e. only for 5 per cent of total vapour volume. Obviously, in the practically more common case of $\Delta\theta_0 \ll \theta_0$, the relative contribution of the microlayer will be substantially larger. The microlayer contribution in equation (11) is independent of liquid viscosity.

For different thermal properties of wall and liquid, Van Ouwkerk [10, 11] presented numerical solutions for the microlayer contribution to bubble growth using the dimensionless quantities F_w/F , $c\theta_0/l$ and Pr as parameters. An increase of the F_w/F -ratio from 1 to 10 and a simultaneous decrease of Pr from 6 to 0.6 enlarges the bubble growth constant corresponding to microlayer evaporation by a factor of approximately 1.6 at $c\theta_0/l = 0.10$. In principle these effects are obvious, as they are caused by larger heat fluxes at the wall according to equation (1), or according to a diminishing initial thickness of the microlayer as $Pr = \nu/a$ and $\delta_0 \sim \nu^{1/2}$.

A serious objection against both Coopers and Van Ouwkerk's procedure to derive the total bubble radius by superposition, i.e. addition of two separate contributions, cf. e.g. equation (11), is that the evaporation at the bubble dome increases the bubble Reynolds number, whence δ_0 is diminishing, and the microlayer evaporation is thus increased; cf. Section 5.1.

3.1.4. Van Beek and Vennekens [23] started from the heat flux balance:

$$q_w(t) = -\rho_l l \dot{d} = k \frac{\theta_w(t)}{d}. \quad (12)$$

Integration over the time interval required for complete evaporation of the microlayer yields an expression for the initial thickness:

$$d_0^2 = 2 \frac{k}{\rho_l l} \int_{t_r}^{t_d} \theta_w(t) dt. \quad (13)$$

The fluctuating wall superheating under the bubble was measured locally using very thin nickel resistance thermometers during bubble growth and inserted into equation (13). Actually, the superheat enthalpy of the microlayer was also taken into account in the r.h.s. of equation (12), and d_0 was taken from computer calculations as the best-fit value to the experimental $\theta_w(t)$. The Van Beek and Vennekens thermal model is based upon the common assumptions that the liquid in the microlayer is at rest, hence a linear temperature profile for $c\theta_0 \ll 1$, and that the vapour temperature remains

constant throughout at saturation, i.e. the model is asymptotic.

The final results in *n*-heptane boiling on a horizontal quartz plate at a subatmospheric pressure of 6.7 kPa and at an initial $\theta_w(0) = \theta_0 = 14$ K are:

$$d_0(r) = 3.5 \times 10^{-4} r^{1/2}, \quad (14)$$

in contrast to other theoretical models predicting: $d_0(r) \sim r$, cf. Sections 3.1.1–3.1.3, and

$$\delta_0(r) = 0.48(\nu t_r)^{1/2}, \quad (15)$$

which means a reduction to only 38 per cent of Van Ouwkerk's value. The radius of the dry spot has been calculated as $R_d^* = 5.6 \times 10^{-2} t$. Possibly, discrepancies in the value of the constant in equation (15) are also caused by inserting k (of heptane) instead of k_w (of quartz) into equations (12) and (13). In the latter case, one calculates $\delta_0(r) = 1.7(\nu t_r)^{1/2}$. Actually however, the ratio F_w/F has to be introduced into equations (12) and (13), cf. equation (1). The neglect of the superheat enthalpy of the microlayer diminishes the microlayer thickness by less than 10 per cent; this effect is substantially lower than Cooper's prediction, cf. Section 3.1.1.

3.1.5. *Application of the Pohlhausen equation.* In principle, Pohlhausen's solution [24, 25] will be used here as an approximation assuming that the liquid microlayer beneath a growing hemispherical vapour bubble is initially formed as a laminar boundary layer in parallel flow along a horizontal throughout uniformly superheated plate. Similarly to all other theoretical models, the eventual occurrence of slip phenomena at the vapour-liquid interface of the microlayer is neglected. This seems to be allowed as the bubble displaces liquid both in front ("relaxation microlayer", cf. Section 3.2.2) and along the plate ("evaporation microlayer"). The exact Pohlhausen solution (for one-dimensional flow, constant wall temperature and constant free vapour velocity) is based on a simple Navier-Stokes equation without pressure gradient in combination with continuity and heat conduction.

The local Nusselt number depends here on a local Reynolds number $Re(r) = r\dot{R}^*/\nu = (r/R^*)(R^*\dot{R}^*/\nu)$, where $R^*\dot{R}^*/\nu$ denotes the bubble Reynolds number:

$$Nu(r) = \frac{q_w(r)r}{k\theta_0} = \frac{r}{d_0(r)} = 0.332 Re^{1/2}(r) Pr^{1/3} \\ = 0.332 \left(\frac{r}{R^*} \right)^{1/2} \left(\frac{R^*\dot{R}^*}{\nu} \right)^{1/2} Pr^{1/3}. \quad (16)$$

The meaning of this equation is restricted here to the initial formation of the evaporation microlayer at the instant, t_r , at which the bubble boundary crosses the radius under consideration: $R^*(t_r) = r$. Assuming

$R^* \sim t^m$, this yields an expression for the initial local thickness of the thermal boundary layer, which must be independent of time:

$$\begin{aligned} d_0(r) &= \frac{k\theta_0}{q_w} = 3.012 \left(\frac{r}{R^*}\right)^{1/2m} \left(\frac{\nu R^*}{\dot{R}^*}\right)^{1/2} Pr^{-1/3} \\ &= 3.012 \left(\frac{r}{R^*}\right)^{1/2m} \left(\frac{\nu t}{m}\right)^{1/2} Pr^{-1/3}. \end{aligned} \quad (17)$$

The thickness of the corresponding hydrodynamic boundary layer amounts to:

$$\delta_0(r) = 3.012 \left(\frac{r}{R^*}\right)^{1/2m} \left(\frac{\nu R^*}{\dot{R}^*}\right)^{1/2}. \quad (18)$$

Two cases are of special interest:

(i) Asymptotic bubble growth ($R^* \sim t^{1/2}$), the only case in which the bubble Reynolds number is independent of time. Equation (17) reduces then to:

$$\begin{aligned} d_0(r) &= 4.260 Pr^{-1/3} \frac{r}{R^*} (\nu t)^{1/2} = 4.260 Pr^{-1/3} (\nu t_r)^{1/2} \\ &= 4.260 Pr^{1/6} (at_r)^{1/2}. \end{aligned} \quad (19)$$

(ii) Initial bubble growth ($R^* \sim t$), the only case in which \dot{R}^* is independent of time, cf. the original derivation of the Pohlhausen equation, where:

$$\begin{aligned} d_0(r) &= 3.012 Pr^{-1/3} \left(\frac{r}{R^*}\right)^{1/2} (\nu t)^{1/2} \\ &= 3.012 Pr^{-1/3} (\nu t_r)^{1/2}. \end{aligned} \quad (20)$$

A considerable advantage of equation (20) over other theoretical models is that $d_0(r) \sim r^{1/2}$ during initial bubble growth in agreement with Van Beek and Vennekens' experimental relation, equation (14). Also, this prediction is in good agreement with the data obtained by Cooper and Lloyd [7] on toluene boiling at pressures of 7 and 14 kPa on a ceramic plate, and on *iso*-propanol at 7 kPa on a glass plate. From these data, one calculates: $d_0(r) \sim r^{0.53}$ and $d_0(r) \sim r^{0.59}$, respectively.

Asymptotic bubble growth. It follows by inserting equation (19) into the heat balance equation:†

$$2\pi(R^*)^2 \dot{R}^* = \frac{k\theta_0}{\rho_2 l} \int_0^{R^*} \frac{2\pi r}{d_0(r)} dr, \quad (21)$$

†This is allowed as the heat flux in the relatively small dry area, cf. equation (23), increases during the period of decreasing microlayer thickness, but drops to zero after complete evaporation. At larger distances from the nucleation site, the relative decrease in $d_0(r)$ is small. This conclusion is confirmed by the mass balance, which yields a bubble vapour mass of only 0.6 per cent of the total liquid mass in the microlayer.

that the radius of a hemisphere bubble is:

$$R^*(t) \cong 0.470 Pr^{-1/6} Ja(at)^{1/2}. \quad (22)$$

Equation (22) is more or less similar to the asymptotic heat-diffusion growth equation for a free bubble in an initially uniformly superheated infinite liquid, but the growth constant has been reduced substantially, cf. equation (9). The similarity makes it difficult to separate from experimental bubble growth curves the contributions due to microlayer and at the bubble dome.

For water boiling at a subatmospheric pressure of 4 kPa ($T = 303$ K, $Pr = 5.43$), the numerical value of $0.470 Pr^{-1/6} = 0.35$; Cooper's equation (5) and Van Ouwkerk's equation (11), corrected to $F_w/F = 10$, are giving 0.97 and 1.24, respectively, i.e. approximately a factor of three higher values. At atmospheric pressure ($Pr = 1.75$) the values are 0.40, 1.71 and 1.41, respectively however, cf. Section 5.1.

Radius of dry area. The radius R_d^* of the dry area around the nucleation site is characterized by complete local evaporation of the microlayer, whence

$$q_w = k\theta_0/d = -\rho_1 l \dot{d} \quad \text{with} \quad \int_{t_r}^{t_d} \dot{d} dt = -d_0.$$

Integration of the local heat flux balance over the time interval required for complete evaporation yields:

$$\frac{k\theta_0}{\rho_1 l} (t_d - t_r) = - \int_{t_r}^{t_d} \dot{d} dt = \frac{1}{2} d_0^2(r) = 9.07 Pr^{-2/3} \nu t_r$$

according to equation (19), whence:

$$\frac{t_d}{t_r} = 1 + 9.07 \frac{\rho_1 l}{k\theta_0} Pr^{-2/3} \nu \approx 9.07 \frac{\eta l}{k\theta_0} Pr^{-2/3}. \quad (23)$$

In general $t_d \gg t_r$, i.e. formation of the microlayer occurs rapidly in comparison with evaporation.

In the asymptotic case, the ratio of the radius of the dry area to the bubble radius is given by:

$$\frac{R_d^*}{R^*} = \frac{R^*(t_r)}{R^*(t_d)} = \left(\frac{t_r}{t_d}\right)^{1/2} \approx 0.332 \left(\frac{k\theta_0}{\eta l}\right)^{1/2} Pr^{1/3}, \quad (24)$$

so that $R_d^* \sim \theta_0^{3/2}$, a prediction being an important feature for the onset of film boiling or the occurrence of burnout. In general, the maximum radius of the dry area is proportional to the bubble departure radius. In systems with prescribed heat flux, high local wall temperatures may occur at dry spots.

The combination of equations (22) and (24) yields an asymptotic approximation for the radius of the dry area:

$$\begin{aligned} R_d^*(t) &\cong 0.156 \left(\frac{k\theta_0}{\eta l}\right)^{1/2} Pr^{1/6} Ja(at)^{1/2} \\ &= 0.156 \left(\frac{\rho_2}{\rho_1}\right)^{1/2} Pr^{-1/3} Ja^{3/2}(at)^{1/2}, \end{aligned} \quad (25)$$

whence $R_d^* \sim \nu^{-1/3}$.

Initially, equation (20) has to be applied instead of equation (19). One has in this case:

$$\begin{aligned} \frac{R_d^*}{R^*} &= \frac{t_r}{t_d} = 0.220 \frac{k\theta_0}{\eta l} Pr^{2/3} = 0.220 \frac{c\theta_0}{l} Pr^{-1/3} \\ &= 0.220 \frac{\rho_2}{\rho_1} Ja Pr^{-1/3}, \end{aligned} \quad (26)$$

whence $R_d^* \sim t$, which is in qualitative agreement with Van Beek and Vennekens' results [23], cf. Section 3.1.4 and the Appendix.

Conclusions. The reader is referred to the Appendix, where numerical values for water show that hardly any dry spot will be present beneath hemispherical bubbles in pure liquids boiling at atmospheric pressure.

3.2. Models for asymptotic bubble growth due to evaporation at the curved surface

3.2.1. Mikic and Rohsenow [26] presented a one-dimensional model of heat diffusion for bubble growth in a non-uniform temperature field. A thermal boundary layer is formed by superposition of two heat fluxes, one due to continuous liquid heating at the wall during a complete bubble cycle, and the other due to the bubble acting as a spherical heat sink starting after a waiting time t_2 ; hence:

$$\rho_2 l \dot{R} = 3^{1/2} k \left[\frac{\theta_0}{(\pi a t)^{1/2}} - \frac{\theta_0 - \Delta\theta_0}{\{\pi a(t + t_2)\}^{1/2}} \right]. \quad (27)$$

The equivalent bubble radius follows by integration:

$$\begin{aligned} R(t) &\cong \left(\frac{12}{\pi}\right)^{1/2} Ja(at)^{1/2} \\ &\times \left[1 - \frac{\theta_0 - \Delta\theta_0}{\theta_0} \left\{ \left(1 + \frac{t_2}{t}\right)^{1/2} - \left(\frac{t_2}{t}\right)^{1/2} \right\} \right]. \end{aligned} \quad (28)$$

Equation (28) simplifies to equation (9), the uniform superheat case, for $t_2 = \infty$.

3.2.2. The "relaxation microlayer" model of Van Stralen [14, 15] is originally based on a different interpretation of the local fluctuations in wall temperature during nucleate boiling. Bubble generation is considered as a relaxation phenomenon of the superheating of the convective thermal boundary layer at the heating surface. The thin liquid thermal layer ("relaxation microlayer") is pushed away locally (due to the radial motion of the bubble boundary) from the heating surface during rapid initial growth, and is surrounding a part of the curved bubble boundary until bubble departure from the surface.

In pure liquids, the relaxation microlayer supplies entirely the required latent heat of vaporization for the bubble from its excess enthalpy above the saturation temperature, i.e. the layer is assumed to be thermally insulated on the bulk liquid side. During the waiting

time when colder liquid of the bulk replaces the original thermal boundary at the heating surface, the convective heat transfer is supplied at the wall side and simultaneously removed at the opposite side. The heat of the relaxation microlayer was then extended to a complete bubble period [14, 15]. Ultimately, the action of the bubbles increases the corresponding convective heat transfer.

The variation in the temperature around the bubble was demonstrated using thin thermocouples by Van Stralen and Sluyter [27] and can be seen in the interferometer cinematographic results of Beer [28] and Matekunas and Winter [29].

Van Stralen's final expression for the equivalent radius during asymptotic bubble growth at a superheated wall is related to equation (9):

$$R(t) \cong \left(\frac{12}{\pi}\right)^{1/2} b Ja(at)^{1/2} = 1.954b Ja(at)^{1/2}, \quad (29)$$

where the Jakob number depends on time according to:

$$Ja = \frac{\rho_1 c}{\rho_2 l} \theta(t) = \frac{\rho_1 c}{\rho_2 l} \theta_0 \exp - \left(\frac{t}{t_1}\right)^{1/2}, \quad (30)$$

t_1 = departure time. The time-independent bubble growth parameter b is proportional to the relative height of that part of the curved surface of the bubble, which is covered with superheated liquid; a maximal value of b occurs if the surface is completely covered, i.e. $b \leq 2^{-1/3} = 0.794$ for a hemispherical bubble, and $b \leq 1$ for a sphere. Equation (29) is simplified to equation (9) for the case of uniform liquid superheating by taking $b = 1$ and $t_1 = \infty$.

The bubble departure radius follows from equation (29):

$$R(t_1) = \frac{b}{e} \left(\frac{12}{\pi}\right)^{1/2} \frac{\rho_1 c}{\rho_2 l} \theta_0 (at_1)^{1/2} = 0.719b Ja(at_1)^{1/2}. \quad (31)$$

The initial thickness of the relaxation microlayer d_0^* in pure liquids is derived from the corresponding convective heat flux according to:

$$d_0^* = \left(\frac{12}{\pi}\right)^{1/2} at_1 = 0.791 d_{w,co} = 0.791 \frac{k\theta_0}{q_{w,co}}, \quad (32)$$

or

$$d_0^* = 1.582 \frac{k}{q_{w,co}} \frac{\sigma T}{\rho_2 l R_0} \quad (33)$$

for a nucleus with radius R_0 . As $\rho_2 \sim p$, it follows from equation (33): $t_1^{1/2} \sim p^{-1}$. It follows from equation (33), that the departure time t_1 , and hence also the departure radius R_1 are derived by extrapolation of the convective part of the boiling curve or from an appropriate convection correlation.

In pure liquids, the waiting time between succeeding bubbles is related to the departure time:

$$t_2 = 3t_1. \quad (34)$$

In binary mixtures, one has [14, 15]:

$$d_{\delta,m}^* = 1.582 \frac{C_{1,m}}{C_{1,p}} \left(\frac{t_{1,m}}{t_{1,p}} \right)^{1/2} \frac{k}{q_{w,co}} \frac{\sigma_m T}{\rho_2 l R_0}, \quad (35)$$

whence it follows from equations (35) and (33) that:

$$\frac{t_{1,m}}{t_{1,p}} = \left(\frac{\sigma_m}{\sigma_p} \right)^2 \quad (36)$$

i.e. in "positive" mixtures, $t_{1,m} < t_{1,p}$.

It may be worth noticing, that, depending on experimental conditions, $q_{w,co}$ in equations (33) and (35) has sometimes to be replaced by a conductive (instead of a convective) heat flux, which follows from heat penetration from the superheated wall into the liquid, cf. also [26].

3.3. Initial bubble growth

The Rayleigh equation of motion for a spherical bubble can be derived from the energy equation in combination with continuity [30, 18, 19, 31, 32]:

$$\begin{aligned} R\dot{R} + \frac{3}{2}\dot{R}^2 &= \frac{1}{2R^2\dot{R}} \frac{d}{dt} (R^3\dot{R}^2) \\ &= \frac{2\sigma}{\rho_1 R_0} \frac{\theta_R(t)}{\theta_0} - \frac{2\sigma}{\rho_1 R} - 4\nu \frac{\dot{R}}{R}, \end{aligned} \quad (37)$$

where the equilibrium radius R_0 follows from the Clausius-Clapeyron equation $\Delta p(0) = (\rho_2 l/T)\theta_0$:

$$R_0 = \frac{2\sigma}{\Delta p(0)} = \frac{2\sigma T}{\rho_2 l \theta_0}. \quad (38)$$

In general, the viscous dissipation term $4\nu\dot{R}/R$ is neglected due to the small value of kinematic viscosity for most liquids.

The asymptotic (i.e. $R \gg R_0$) Rayleigh solution for the isothermal case is obtained by taking the instantaneous superheating at the vapour-liquid interface $\theta_R(t) \equiv \theta_0 = \text{constant}$, i.e. by taking

$$\Delta p(t) = \{\theta_R(t)/\theta_0\} \Delta p(0) \equiv \Delta p(0) = \Delta p = \text{constant:}$$

$$R(t) \cong \left(\frac{2\Delta p}{3\rho_1} \right)^{1/2} t = \left(\frac{4\sigma}{3\rho_1 R_0} \right)^{1/2} t = \left(\frac{2\rho_2 l \theta_0}{3\rho_1 T} \right)^{1/2} t. \quad (39)$$

In contradistinction to the mode of advanced bubble growth, which is determined by heat diffusion, initial bubble growth is seen to be determined by the hydrodynamic inertia terms $R\dot{R}$ and $\frac{3}{2}\dot{R}^2$. Diffusion-controlled growth can, according to Forster and Zuber [18] and to Plesset and Zwick [19], also be described by equation (37), but only if a negative evaporation term (accounting for bubble growth as a moving heat sink) is added to the r.h.s. of equation (37):

$$R\dot{R} + \frac{3}{2}\dot{R}^2 = \frac{2\sigma}{\rho_1 R_0 \theta_0} \left[\theta_0 - \frac{\rho_2 l}{\left(\frac{12}{\pi} a \right)^{1/2} \rho_1 c} C_2 \right] - \frac{2\sigma}{R}, \quad (40)$$

where $R = C_2 t^{1/2} = C_1 \theta_0 t^{1/2}$ yields the asymptotic equation (9) for the isobaric case as only the term between brackets remains as $t \rightarrow \infty$.

Actually, during an intermediate time interval, a gradual transition range will be present during which bubble growth can be described by an appropriate combination of equations (39) and (9), as $\theta_R(0) = \theta_0$ and $\theta_R(t) \rightarrow (R_0/R)\theta_0 \rightarrow 0$ as $t \rightarrow \infty$ (i.e. both the vapour and interface temperatures approximate the saturation value).

3.4. Transition between isothermal and isobaric bubble growth

3.4.1. Cooper and Vijuk [17] apply equation (37) for a hemispherical bubble at a wall with constant superheating θ_0 , i.e. at a wall of high conductivity. The instantaneous superheating of the vapour-liquid interface is given by:

$$\theta_{R^*}(t) = \frac{\rho_1 T}{\rho_2 l} \left[R^* \dot{R}^* + \frac{3}{2} (\dot{R}^*)^2 + \frac{2\sigma}{\rho_1 R^*} \right]. \quad (41)$$

The heat flux balance, equation (7), is used to obtain the driving instantaneous temperature difference across the microlayer:

$$\theta_0 - \theta_{R^*}(t) = \frac{(\pi\nu)^{1/2} \rho_2 l (R^*)^2 \dot{R}^*}{2k} \int_0^{R^*} \frac{r dr}{l_r^{1/2}}. \quad (42)$$

If the Cooper *et al.* initial microlayer thickness $\delta_0(r) = \frac{1}{2}(r/R^*)(\pi\nu t)^{1/2}$ and the asymptotic $R^*(t)$ of equation (5) are inserted, equation (42) reduces to $\theta_0 - \theta_{R^*}(\infty) = \theta_0$, i.e. $\theta_{R^*}(\infty) = 0$. This result is also obtained by inserting equations (19) and (21) into equation (42).

With the purpose of eliminating the unknown function $\theta_{R^*}(t)$, Cooper and Vijuk add equations (41) and (42), omitting the surface tension term in equation (41) being unimportant for the transition from the linear growth phase to the parabolic growth phase. An analytical solution does not exist for the transition period. The following approximation to a numerical solution has been suggested [17]:

$$R^*(t) = \frac{1}{\left(\frac{3\rho_1 T}{2\rho_2 l \theta_0 t^2} \right)^{1/2} + \frac{(\pi Pr)^{1/2}}{4Ja(at)^{1/2}}}. \quad (43)$$

Equation (43) has the form

$$R^*(t) = 1/[1/R_1^*(t) + 1/R_2^*(t)],$$

where $R_1^*(t)$ denotes the Rayleigh solution for linear growth, equation (39), and $R_2^*(t)$ the Cooper *et al.* solution for asymptotic microlayer growth, equation (5). Obviously, equation (43) has been derived in analogy to two thermal resistances connected in series, with temperature drops $\theta_{R^*}(t)$ and $\theta_0 - \theta_{R^*}(t)$, respectively, i.e.

with a constant total temperature drop of θ_0 . Equation (43) is independent of the bulk liquid temperature and the properties of the solid wall material; the latter is evident as the wall is supposed to be of high thermal conductivity.

3.4.2. Mikic, Rohsenow and Griffith [33] combined the Rayleigh solution accounting for inertia, equation (39), with the heat diffusion solution, equation (9), in case of a uniformly superheated liquid, or with equation (28) in case of non-uniformity. Similarly to Cooper and Vijuk [17], Mikic *et al.* eliminate the unknown instantaneous vapour superheating $\theta_R(t)$ during the transition stage. This elimination has been done only seemingly, as actually $\theta_R(t)$ is determined empirically by applying equation (39) to $\theta_R(t)$ and equation (9) to $\theta_0 - \theta_R(t)$ in case of uniform superheat. The objection against this procedure is that one could eliminate R similarly whence an equation for $\theta_R(t)$ remains.

Obviously, the Mikic *et al.* model does not incorporate microlayer evaporation. The final equation is given in the following dimensionless form:

$$\frac{dR^+}{dt^+} = \left[t^+ + 1 - \frac{\theta_0 - \Delta\theta_0}{\theta_0} \left(\frac{t^+}{t^+ + t_2^+} \right)^{1/2} \right]^{1/2} - (t^+)^{1/2}, \dagger \quad (44)$$

where:

$$R^+ = \frac{B}{C_2} R \quad \text{and} \quad t^+ = \left(\frac{B}{C_2} \right)^2 t, \quad (45)$$

B follows by writing equation (39) as $R = Bt$ and C_2 by writing equation (9) as $R = C_2 t^{1/2}$. In case of a bubble at a wall, the factor $2/3$ in equation (34) is replaced by $\pi/7$. According to equation (44), bubble growth rates increase by extending the preceding waiting time during which a thermal "boundary layer" is formed.

For a uniformly superheated liquid ($\Delta\theta_0 = \theta_0$) the dimensionless waiting time $t_2^+ = \infty$, whence equation (44) simplifies to:

$$\frac{dR^+}{dt^+} = (t^+ + 1)^{1/2} - (t^+)^{1/2}. \quad (46)$$

A common feature of equations (43) and (44) is, that both reduce to equation (39), the Rayleigh solution accounting for liquid inertia, for $t \rightarrow 0$. Also, both reduce to an asymptotic heat diffusion solution for $t \rightarrow \infty$, i.e. to equation (5) accounting for microlayer evaporation, or to equation (9) accounting for evaporation on the curved surface, respectively.

4. BUBBLE GROWTH RATES IN BINARY SYSTEMS

4.1. The combined heat and mass diffusion controlled asymptotic stage

4.1.1. Van Stralen [14, 32] extended the asymptotic equation (29) for bubble growth due to evaporation on

the curved surface in case of bubble growth on a superheated wall in a binary mixture to:

$$\begin{aligned} R_m(t) &\cong bC_{1,m}\theta(t)t^{1/2} \\ &= \left(\frac{12}{\pi} \right)^{1/2} b \frac{(\theta_0 - \Delta T) \exp-(t/t_1)^{1/2}}{\rho_2 l / \rho_1 c} (at)^{1/2} \\ &= \left(\frac{12}{\pi} \right)^{1/2} b \frac{\theta_0 \exp-(t/t_1)^{1/2}}{\frac{\rho_2}{\rho_1} \left[\frac{l}{c} + \left(\frac{a}{D} \right)^{1/2} \frac{\Delta T}{G} \right]} \end{aligned} \quad (47)$$

$\Delta T(x_0)$ denotes the increase in dew point of the vapour depending on the mass (or mole) fraction of the more volatile component; G is the vaporized mass fraction. For mixtures with $x_0 \ll 1$, the numerical values of the properties may be considered to be independent on composition.

The ratio

$$\frac{\Delta T}{G}(x_0) = -x_0 \{ K(x_0) - 1 \} \left(\frac{dT}{dx} \right)_{x=x_0} \quad (48)$$

is independent of G and of θ_0 (as both $\Delta T = T(x) - T(x_0)$ and G are proportional to θ_0) and of x (the mass fraction of the more volatile component at the interface) for relatively small values of G .

This ratio can also be evaluated graphically (in dependence on composition) from an equilibrium diagram, a $T(x, y)$ diagram at constant pressure [34–36]. The minimum value of the bubble growth constant $C_{1,m}$ in binary systems can thus easily be derived by substituting the obtained $\Delta T/G$ in equation (47). For the system water–2-butanone boiling at atmospheric and subatmospheric pressures, the minimal growth rate occurs at $x_0 = 0.04$; the bubble growth constant $C_{1,m}$ has been reduced to 0.25 times the value in water boiling at the same pressure.

It follows from equation (48), that also for binary mixtures with $x_0 \ll 1$, $R_m \sim 1/\rho_2 \sim 1/p$. Equation (31) for the bubble departure radius still holds for binary mixtures, if θ_0 is replaced by $\theta_0 - \Delta T$, i.e. by $0.25 \theta_0$ for the mixture considered above.

4.1.2. Application of the Pohlhausen equation. As the dew point of vapour in mixtures has been increased by an amount ΔT , the effective temperature difference across the microlayer equals $\theta_0 - \Delta T$; $\theta_0 =$ wall superheating; the diffusion penetration depth $2(Dt)^{1/2} \ll d_{0,m}(r)$ already after a very short time due to the small numerical value of the mass diffusivity, which is of the order of $D = 10^{-9} \text{ m}^2/\text{s}$; the order of ν for water at room temperature is $10^{-6} \text{ m}^2/\text{s}$.

† Equation (17) of [33] has a misprint.

For binary mixtures, one can introduce a modified Jakob number Ja_m , which is based on $\theta_0 - \Delta T$ instead of θ_0 :

$$\begin{aligned} Ja_m &= \frac{\rho_1 c}{\rho_2 l} (\theta_0 - \Delta T) = \frac{\theta_0 - \Delta T}{\theta_0} Ja \\ &= \frac{C_{1,m}}{C_{1,p}} Ja = \frac{1}{1 + \frac{c}{l} \left(\frac{a}{D}\right)^{1/2} \frac{\Delta T}{G}} Ja, \end{aligned} \quad (49)$$

where $Ja = \rho_1 c \theta_0 / (\rho_2 l)$ denotes the Jakob number of the excess (less volatile) component. For convenience, however, we will use only Ja and characterize bubble microlayer behaviour in mixtures only by the ratio of the growth constants $C_{1,m}/C_{1,p}$.

As equation (16) has to be consistent with $q_w = 2\rho_2 l \dot{R}^*$, it follows by taking $R_m^*(t) = (C_{1,m}/C_{1,p})^n R^*(t)$, that also $q_{w,m} \sim (C_{1,m}/C_{1,p})^n$. Equation (16) is giving:

$$d_{0,m}(r) \sim (C_{1,m}/C_{1,p})^{-n/2}$$

and

$$q_{w,m} \sim (C_{1,m}/C_{1,p})^{(n/2)+1},$$

whence

$$(n/2) + 1 = n \quad \text{or} \quad n = 2,$$

i.e. $q_{w,m} \sim (C_{1,m}/C_{1,p})^2$. Hence a substantial reduction in heat flux in mixtures with a low bubble growth constant occurs; this is caused by a diminished driving temperature difference across the microlayer in combination with an increase in microlayer thickness, the latter being due to the decreased bubble growth rates.

It follows from equation (16), that the initial microlayer thickness:

$$d_{0,m}(r) = \left(\frac{C_{1,p}}{C_{1,m}}\right) d_0(r). \quad (50)$$

Equation (22) must be replaced by:

$$R_m^*(t) \cong 0.470 \left(\frac{C_{1,m}}{C_{1,p}}\right)^2 Pr^{-1/6} Ja(at)^{1/2}. \quad (51)$$

One has here instead of equation (24):

$$\frac{R_{d,m}^*}{R_m^*} \cong 0.332 \left(\frac{C_{1,m}}{C_{1,p}}\right)^{3/2} \left(\frac{k\theta_0}{\eta l}\right)^{1/2} Pr^{1/3} \quad (52)$$

and equation (25) is replaced by:

$$R_{d,m}^*(t) \cong 0.156 \left(\frac{C_{1,m}}{C_{1,p}}\right)^{7/2} \left(\frac{k\theta_0}{\eta l}\right)^{1/2} Pr^{1/6} Ja(at)^{1/2}. \quad (53)$$

Equations (50)–(53) are simplified to the original equations if $C_{1,m} = C_{1,p}$. The substantial reduction of the radius of the dry area beneath a growing bubble in case of $C_{1,m} < C_{1,p}$, equation (53), has a practical consequence for the onset of film boiling or for the occurrence of burnout, cf. Van Stralen's "boiling paradox" [14, 15] and Van Ouwkerk's discussion on boiling stability [37].

It follows from equations (9) and (47), that both $C_{1,p} \sim 1/p$ and $C_{1,m} \sim 1/p$, whence this effect is independent of pressure. The reader is referred to the Appendix for numerical data on the mixture $x_0 = 0.04$ in the binary system water–2-butanone, where $C_{1,m} = 0.25C_{1,p}$.

Conclusions. If the predictions of Section 3.1.5 for water are also taken into account, the conclusions are the following: (i) dry areas will be formed in pure liquids only at low subatmospheric pressures; and (ii) in binary mixtures with a small bubble growth constant, dry areas will even not occur at very low pressures.

Some of the present predictions are more or less similar to those obtained by Van Ouwkerk [38], who extended the self-similar solution for the microlayer, cf. Section 3.1.3, using previous bubble growth theories for mixtures by Scriven [39] and Van Stralen [31, 32].

4.1.3. *Combination of evaporation microlayer and relaxation microlayer.* According to equation (51), bubble growth rates due to the evaporation microlayer are reduced in mixtures with a factor of $(C_{1,m}/C_{1,p})^2$, i.e. they are even more substantially diminished than the growth due to evaporation on the curved bubble surface, as the latter is reduced with a factor of $C_{1,m}/C_{1,p}$, cf. equations (47) and (49). A combination of both separate contributions is only possible, cf. Section 3.1.3, if the dependence of the microlayer evaporation on the total bubble Reynolds number, cf. equation (16), is taken into account, i.e. the microlayer evaporation depends on the evaporation rate of the relaxation microlayer surrounding the curved bubble surface.

Conversely, the evaporation microlayer removes a part of the relaxation microlayer resulting in an interaction of both mechanisms, cf. Section 5.1.

4.2. Transition between isothermal and isobaric bubble growth

According to equation (39), the Rayleigh solution, initial bubble growth is hydrodynamically determined by liquid inertia, and thus independent of heat and mass diffusion towards the bubble boundary, which is only important at advanced time.

Similarly to a pure liquid, the equilibrium bubble in a mixture has an initial superheating θ_0 at the interface. This implies that no effect of composition is expected during initial bubble growth for mixtures with $x_0 \ll 1$, as liquid properties are then nearly the same.

Experimental data on bubble growth rates in binary mixtures are thus not only important for the diffusion-controlled stage and for the formation of dry spots, but also for checking the validity of the Rayleigh solution, i.e. the effect of liquid inertia during initial bubble growth.

4.3. Bubble shape in mixtures

Apparently, a nearly complete exhaustion of the more volatile component in the evaporating microlayer (if present) may be expected to occur soon after initial bubble formation. In "positive mixtures" (in which the more volatile component has the lowest surface tension constant), this will result in a contraction of the lower part of the originally hemispherical bubble due to the "Marangoni effect", cf. [40], i.e. the mixture $x_0 = 0.04$ in the binary system water-2-butanone, is predicted to show this behaviour.

5. COMBINATION OF RELAXATION AND EVAPORATION MICROLAYERS

5.1. Interaction of the microlayer contributions to bubble growth

To date, theoretical bubble growth models have not accounted for the interaction of the mutually dependent contributions due to the relaxation microlayer (around the bubble dome) and the evaporation microlayer (beneath the bubble). An explanation of the interaction effect, together with the procedure for incorporating it into the theoretical growth model, is detailed below.

The formation of the evaporation microlayer has to be induced by an externally caused bubble growth rate—according to equation (16)—i.e. by an instantaneous superheating at the bubble boundary (resulting in the Rayleigh solution, Section 3.3) in combination with the effect of the relaxation microlayer.

It follows from equation (19) by inserting $R_{2,1} = A_1 t^{1/2}$, equation (22), that during advanced growth:

$$d_0(r) = \frac{4 \cdot 260 Pr^{1/6} a^{1/2}}{A_1} r. \quad (54)$$

The initial thickness of the evaporation microlayer at a fixed distance from the nucleation site is thus inversely proportional to the bubble growth factor A_1 , in agreement with equation (50). The simultaneous additional evaporation of liquid at the curved bubble boundary increases A_1 considerably to the value A_3 . As a consequence, $d_0(r)$ decreases as follows:

$$d_0(r) = \frac{4 \cdot 260 Pr^{1/6} a^{1/2}}{A_3} r, \quad (55)$$

which results in additional microlayer evaporation. In the reverse sense, the evaporation microlayer keeps a quantity of superheated liquid at the wall, which is not removed to support the relaxation microlayer. $R_2 = A_3 t^{1/2}$ accounts for the total combined diffusion-controlled growth.

The interaction between both microlayer contributions in pure systems is given by the heat balance during the mode of advanced bubble growth, which is

an extension of equation (21):

$$2\pi R_2^2 \dot{R}_2 = \frac{k\theta_0}{2^{1/3}\rho_2 l} \int_0^{R_2} \frac{2\pi r}{d_0(r)} dr + 2\pi R_2^2 \dot{R}_{2,2} \quad (56)$$

$R_{2,2} = A_2 t^{1/2}$ accounts for the relaxation microlayer and is given by equations (29) and (31) by replacing here $\theta(t)$ by θ_0 and b by b^* . The factor $2^{-1/3} = 0.794$ in the evaporation microlayer term in equation (56) is introduced by considering the equivalent bubble radius.

Equation (56) is independent of time and yields by inserting the corresponding expressions for R_2 , $R_{2,2}$ and $R_{2,1}$ of equation (22) using equation (55): $A_3^3 = A_3^2 A_1 + A_3^2 A_2$, or:

$$A_3 = A_1 + A_2. \quad (57)$$

It is emphasized, that the interaction of both microlayers is incorporated in this superposition-type equation. If the interaction should have been neglected by inserting equation (54) instead of equation (55) into equation (56), the result would have been:

$$A_3^3 = A_3 A_1^2 + A_3^2 A_2, \quad \text{or} \quad A_3^2 = A_1^2 + A_3 A_2.$$

It follows from (57), that both in pure and binary systems the diffusion solution is given by, cf. equation (51):

$$R_2(t) \cong \left\{ 0.3730 Pr^{-1/6} \frac{C_{1,m}}{C_{1,p}} + 1.9544 b^* \right\} \frac{C_{1,m}}{C_{1,p}} Ja(at)^{1/2}. \quad (58)$$

It follows that the ratio of the contributions to the bubble growth rate due to evaporation microlayer and relaxation microlayer is independent of both time and of initial wall superheating (cf. Section 5.3) and amounts to:

$$\frac{0.1908 Pr^{-1/6} C_{1,m}}{b^* C_{1,p}} : 1. \quad (59)$$

5.2. Height of the relaxation microlayer in dependence on pressure

On account of its physical meaning, one may expect, that $b^* \rightarrow 1$ with increasing pressure [14, 15, 31, 32], and that b^* is of the order of magnitude of 10^{-1} (cf. the forthcoming paper [41] on experimental results) for a saturated bulk liquid at very low subatmospheric pressures.

It follows now from equation (59), that the relative contribution of the evaporation microlayer decreases with increasing pressure. For thin wires used as a heating surface instead of a flat plate, the evaporation microlayer contribution will be small at any pressure.

The maximum height of the spherical segment (at the lower part of the bubble dome, consistent with [28, 29]), which is covered with superheated liquid is given by [14, 15]:

$$H_1 = 2b^* R(t_1). \quad (60)$$

As $t_1^{1/2} \sim \theta_0 \sim p^{-1}$, cf. equation (33), and approximately $b^* \sim p$, one may expect for a constant cavity radius R_0 acting at the minimum required θ_0 , that

$$H_1 \sim p^2 p^{-1} Ja \sim p^2 p^{-1} p^{-2} \sim p^{-1}$$

(as $Ja \sim \rho_2^{-2} \sim p^{-2}$), i.e. H_1 is predicted to be inversely proportional to the pressure. This means, that the distance, across which the original thermal boundary layer at the heating wall can be displaced by the moving bubble in the upward direction, will be dependent on pressure. Actually, however, the operational θ_0 may exceed the required minimum value substantially.

5.3. Final equation for bubble growth during adherence

Until now, the gradually diminishing excess enthalpy of the evaporation microlayer has been neglected. Actually, this effect can be incorporated into the final bubble growth equation by introducing a time-dependent microlayer (uniform) superheating $\theta(t)$ similarly to equation (30). Both microlayers show then the same temperature dependence, in good agreement with observed temperature fluctuations at the wall beneath growing bubbles [4-8, 23], which occur even in case of a heating material of high thermal conductivity. The introduction of $\theta(t) = \theta_0 \exp -(t/t_1)^{1/2}$ instead of a constant θ_0 has no influence on the derivation of equation (57) from the heat balance (56).

The proposed procedure is not in contradiction with equations (21), (22) and (13), cf. [14, 15]. During the succeeding waiting time after bubble departure, the superheating of the wall increases gradually to θ_0 according to equation (1), the validity of which has previously been extended to the entire bubble period, cf. [14, 15] and Section 3.1.1.

Due to its physical background, the Cooper and Vijk procedure [17], cf. Section 3.4.1, will be used here to combine the total asymptotic diffusion solution with a modified Rayleigh solution, cf. equation (39), which dominates during the early growth period.

The proposed final bubble growth equation, which is valid (both in pure liquids and in binary mixtures) during the entire adherence time is assumed to be of following form:

$$R(t) = \frac{1}{\{1/R_1(t) + 1/R_2(t)\}} = \frac{R_1(t)R_2(t)}{R_1(t) + R_2(t)} \quad (61)$$

The modified Rayleigh solution is given by:

$$R_1(t) = 0.8165 \left\{ \frac{\rho_2 l \theta_0 \exp -(t/t_1)^{1/2}}{\rho_1 T} \right\}^{1/2} t, \quad (62)$$

and the combined relaxation and evaporation microlayers (heat and mass) diffusion solution, cf. equations

(47), (51) and (58) is given by:

$$R_2(t) = 1.9544 \frac{C_{1,m}}{C_{1,p}} \times \left[\{b^* \exp -(t/t_1)^{1/2}\} + \frac{\Delta\theta_0}{\theta_0} \right] Ja(at)^{1/2} + 0.3730 \left(\frac{C_{1,m}}{C_{1,p}} \right)^2 Pr^{-1/6} \times \{ \exp -(t/t_1) \}^{1/2} Ja(at)^{1/2}. \quad (63)$$

The Jakob number $Ja = \rho_1 c \theta_0 / \rho_2 l$ has to be applied here to the pure less volatile component. In pure liquids $C_{1,m}/C_{1,p} = 1$.

Obviously, $R(t) \rightarrow R_1(t) \rightarrow$ equation (39) as $t \rightarrow 0$, i.e. bubble growth during the early stage is governed by liquid inertia, thus by hydrodynamics. $R(t) \rightarrow R_2(t_1)$ as $t \rightarrow t_1$, the departure time, at which $R_2(t_1) = 0$ for the case of $\Delta\theta_0 = 0$. The bubble departure radius follows from equation (61):

$$R(t_1) = \frac{R_1(t_1)R_2(t_1)}{R_1(t_1) + R_2(t_1)}, \quad (64)$$

where

$$R_1(t_1) = 0.4952 \left(\frac{\rho_2 l \theta_0}{\rho_1 T} \right)^{1/2} t_1, \quad (65)$$

and (in case $\Delta\theta_0 = 0$):

$$R_2(t_1) = 0.7190 \frac{C_{1,m}}{C_{1,p}} b^* Ja(at_1)^{1/2} + 0.1372 \left(\frac{C_{1,m}}{C_{1,p}} \right)^2 Pr^{-1/6} Ja(at_1)^{1/2}, \quad (66)$$

whence b^* is determined by the following expression:

$$b^* = 1.3908 \frac{C_{1,p}}{C_{1,m}} \frac{R_2(t_1)}{Ja(at_1)^{1/2}} - 0.1908 \frac{C_{1,m}}{C_{1,p}} Pr^{-1/6}. \quad (67)$$

This expression is an extension of equation (31), cf. also Section 5.1.

According to equation (33), the bubble departure time increases with decreasing pressure. In general,

$$R_1(t_1) \sim p^{1/2} p^{-2} = p^{-3/2}, \\ R_2(t_1) \sim (p^{-1} p^{-1} + p^{1/2} p^{-1} p^{-1}) \approx p^{-2},$$

whence $R(t_1) \sim p^{-2}$ at pressures, at which heat and mass diffusion governs bubble growth (the more common case) and $R(t_1) \sim P^{-3/2}$ at very low sub-atmospheric pressures, at which liquid inertia dominates. Apparently, in any case, the bubble departure radius decreases substantially at increasing pressure. A gradual decrease of $R(t_1)$ at decreasing θ_0 is also obvious. Both the absolute and the relative instant (in relation to t_1), at which $R_1 = R_2$, increase at decreasing pressure, i.e. at very low subatmospheric pressures,

liquid inertia governs bubble growth during nearly the entire adherence time. Contrarily, at elevated pressures, where the diffusion solution dominates during the entire adherence time, equation (61) is simplified to equation (29), and equation (64) to (31).

The derived bubble growth and departure radius equations (61)–(66) can be generalized easily to the case of a wall of low thermal conductivity by replacing θ_0 by $\theta_0^* = F_w \theta_0 / (F_w + F)$ according to equation (2). I.e. the superheating enthalpy of the wall material is taken into account.

5.4. The maximum radius of the contact area between bubble and wall

The ratio of the contact area between bubble and heating wall to the bubble radius (R_c^*/R^*) equals one during the early hemispherical growth period. During the succeeding mode of advanced growth, the lower part of the bubble boundary contracts (due to surface tension gradient forces in combination with buoyancy), whence R_c^*/R decreases gradually and R_c^* shows a maximum value at t_c as $R_c^*(t_c) = 0$. For pure liquids, the assumption is made, that the maximum contact radius $R^*(t_c)$ occurs at the instant t_c , which is determined by the following condition, cf. equations (19) and (32):

$$\begin{aligned} d_0\{R^*(t_c)\} \\ = 4.260Pr^{-1/6}(at_c)^{1/2} = d_{w,co} = 2.471(at_1)^{1/2}, \end{aligned} \quad (68)$$

whence t_c is proportional to the departure time:

$$t_c = 0.336Pr^{-1/3}t_1. \quad (69)$$

The physical basis of equation (68) is, that the evaporation microlayer keeps only superheated liquid beneath the bubble. Afterwards, the contact area is diminishing due to the combined action of buoyancy and the occurrence of gradients in the surface tension along the bubble boundary, resulting in a contraction of the outer area.

At very low pressures (below 3 kPa), where the Rayleigh solution governs bubble growth during nearly the entire departure time, one has to use equation (20) for the initial thickness of the evaporation microlayer instead of equation (19). In this case, equation (69) is replaced by:

$$t_c = 0.673Pr^{-1/3}t_1. \quad (70)$$

5.5. Temperature fluctuations at the surface and inside the heating material

It follows from equation (63), that the temperature difference across the evaporation microlayer decreases during bubble growth. Consequently, a substantial temperature dip occurs simultaneously at the heating surface. During the waiting time between succeeding bubbles, the temperature difference across the microlayer increases again gradually.

These temperature fluctuations show only a slight damping of the amplitude in case of a highly conducting (metal) wall. Consequently, one may expect the occurrence of considerable temperature fluctuations inside the heating material even at relatively large distances from the upper surface. The fluctuations give rise to unwanted periodic thermal expansions resulting in material fatigue. Of course, this effect will even be of greater importance at the dry spots formed beneath bubbles at which much higher local temperatures occur in case of uniform heat production inside the heating material. This may also promote local corrosion of the heating surface.

5.6. Turbulent flow pattern inside and in the neighbourhood of rapidly growing bubbles

Especially in water boiling at low pressures, the equivalent bubble Reynolds number $R\dot{R}/\nu$ reaches high values of the order of magnitude of 10^4 – 10^5 , which results in the occurrence of turbulent eddies in the adjacent liquid at the bubble dome. Also, in the vapour inside the bubble space, turbulent eddies may be expected to occur, although the kinematic viscosity of vapour is nearly one order or magnitude larger in comparison to the kinematic viscosity of liquid. Violent turbulent flow pattern have actually been observed by the present authors on high-speed cinematographic recordings, cf. the forthcoming paper on experimental bubble growth data in water boiling at subatmospheric pressures [41]. Similar observations have been made by Beer [42].

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APPENDIX

The Evaporation Microlayer Contribution to Bubble Growth

1. Numerical values for water

(i) *At a low pressure of 4kPa.* For $\theta_0 = 35$ K ($Ja = 1970$) and $t = 6.25 \times 10^{-2}$ s (i.e. $t \approx \frac{1}{2}t_1$), one has, equation (25): $R_d^* = 4.14 \times 10^{-3}$ m; under these conditions, equation (26) predicts a ratio $R_d^*/R^* = 4.61 \times 10^{-2}$, equation (22) yields $R^* = 6.83 \times 10^{-2}$ m.

(ii) *At atmospheric pressure.* For $\theta_0 = 20$ K ($Ja = 60$) and $t = 4.9 \times 10^{-3}$ s (i.e. $t \approx \frac{1}{2}t_1$), $R_d^* = 4.25 \times 10^{-5}$ m, $R_d^*/R^* = 4.43 \times 10^{-2}$ and $R^* = 7.31 \times 10^{-4}$ m. Apparently, the theoretical asymptotic microlayer value of R^* at 4kPa exceeds the total experimental value (4.5×10^{-2} m); cf. Section 5.

At atmospheric pressure the experimental $R^* = 1.24 \times 10^{-3}$ m exceeds the asymptotic microlayer contribution with

70 per cent. Due to the small value of R_d^* , which is only thirty times the dimension of nucleation sites, hardly any dry spot will be present in water boiling at atmospheric pressure.

It may be worth noticing, that the value of the constant, 0.156 in equation (25) may be increased maximally by a factor of 5 due to the interaction with the relaxation microlayer, cf. Section 5, i.e. dry areas of the order of 10^{-2} m are predicted to occur in nucleate boiling of water at 4 kPa.

2. Numerical values for an aqueous binary mixture

Some numerical values are following for the mixture $x_0 = 0.04$ in the binary system water-2-butanone, where $C_{1,m} = 0.25C_{1,p}$, $C_{1,p}$ is the corresponding value for water boiling at the same pressure.

(i) At a pressure of 4 kPa, cf. Section 4.1.2, the predictions are giving for $\theta_0 = 35$ K and $t = 6.25 \times 10^{-2}$ s; $R_m^* = 4.27 \times 10^{-3}$ m, $R_{d,m}^*/R_m^* = 7.70 \times 10^{-3}$ and $R_{d,m}^* = 3.24 \times 10^{-5}$ m, a very small value, which equals the dimensions of nucleation sites (3×10^{-5} m), i.e. hardly any dry area will be formed in this mixture.

(ii) At atmospheric pressure at $\theta_0 = 20$ K and $t = 4.9 \times 10^{-3}$ s, the predictions are: $R_m^* = 4.55 \times 10^{-5}$ m, $R_{d,m}^*/R_m^* = 7.40 \times 10^{-3}$ and $R_{d,m}^* = 3.32 \times 10^{-7}$ m, a value even an order of magnitude smaller than the dimensions of nucleation sites (1.5×10^{-6} m).

3. Numerical values for heptane

(i) *Microlayer thickness.* With the purpose to compare theoretical predictions with Van Beek and Vennekens' experimental equation (14): $d_0(r) = 3.5 \times 10^{-4} r^{1/2}$ for the

initial thickness of the evaporation microlayer, the following distinction is made:

(a) Asymptotic bubble growth, cf. equation (19). The bubble radius in *n*-heptane boiling at a pressure of 6.7 kPa at $\theta_0 = 14$ K is estimated both from Van Ouwkerk's experimental data [10,11] at 16.6–24.6 kPa and from equation (9) to: $R^* = 0.10t^{1/2}$. Equation (19) yields: $d_0(r) = 1.10 \times 10^{-3} r$ at $t = 10^{-3}$ s, and $d_0(r) = 1.95 \times 10^{-3} r$ at $t = 10^{-2}$ s.

(b) Initial bubble growth, cf. equation (20); equation (39) gives: $R^* = 1.87t$, whence equation (20) yields: $d_0(r) = 10.1 \times 10^{-4} r^{1/2}$, which is in satisfactory agreement with equation (14), if one considers the neglect of microlayer interaction.

(ii) *Radius of dry area.* During initial bubble growth: $R_d^*/R^* = 0.22 (k\theta_0/\eta)Pr^{2/3}$, cf. Section 3.1.5, equation (26). For *n*-heptane at the above-mentioned boiling conditions, $R_d^* = 2.0 \times 10^{-2}t$. This prediction is also in satisfactory agreement with Van Beek and Vennekens' experimental results [23], cf. Section 3.1.4.

Van Ouwkerk's data [10,11] on *n*-heptane boiling at 16.6 kPa on a perspex plate show a ratio: $R_d^*/R^* = 8.2 \times 10^{-2}$, in quantitative agreement with equation (24), which yields a ratio of 7.2×10^{-2} .

It may be worth noticing, that in general the interaction between the relaxation microlayer and the evaporation microlayer has not been taken into account in the Appendix. This is due to a present lack on appropriate experimental information in literature. However, this neglect has no influence on the order of magnitude of substantial quantities, neither on the validity of the statements. In this respect, the reader is referred to the forthcoming experimental results and a comparison of those with theoretical predictions [41].