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THE MECHANISM OF NUCLEATE BOILING IN PURE LIQUIDS AND IN BINARY MIXTURES—PART IV

SURFACE BOILING

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Abstract—A favourable effect of the composition in binary mixtures with a more volatile component on the peak flux in surface boiling of subcooled liquids with free convection has been predicted by the "relaxation microlayer" theory, cf. Parts I–III. The theoretical values are in quantitative agreement with the author's experimental data on the systems water–methylethylketone and water–1-butanol.

Expressions for the peak flux and for the maximum bubble radius, frequency, growth and condensation rates, and lifetime have been derived in dependence on the wire superheating and on the degree of subcooling. An increase in the subsequent condensation rate in certain binary mixtures is predicted. At present, no experimental data on bubbles in subcooled mixtures are available, but the theoretical predictions for pure liquids are in reasonably good agreement with some preliminary data by the author and with data on water by Ellion for free convection and by Gunther for forced circulation.

The occurrence of a minimal critical expansion ratio for supersaturated vapour mixtures in presence of air in Wilson cloud chambers is explained by, and the numerical value of the minimum ratio can be calculated from, the new theory. The corresponding liquid concentration in the droplets is shown to coincide with the concentration of maximal slowing down of bubble growth and maximum peak flux density in superheated liquid mixtures. This is in good agreement with data by Flood on water–ethanol vapour and by Froemke, Bloomquist and Anderson on water–ethanol.

Finally, a note is added on the ultrasonic velocity and absorption in binary liquid mixtures.

1. INTRODUCTION ON THE EXPERIMENTAL PEAK FLUX IN SURFACE BOILING

THE AUTHOR'S relaxation microlayer theory [1, 2, 3] is extended to surface (or "local") boiling, which occurs in the same superheating range of the wall as in case of nucleate pool boiling, if the temperature of the bulk liquid $T_0 < T$ ("negative superheating"). The vapour bubbles condense shortly after the rapid initial growth. Even condensation of adhering bubbles occurs for sufficiently large degrees of subcooling $\theta_0^* = T - T_0$. The vaporized mass fraction at the heating surface approximates zero then. An exhaustion of the more volatile component in mixtures in the neighbourhood of a relatively large heating area is thus avoided completely.

The peak flux in surface boiling with free convection increases proportionally to $T - T_0$ to very considerable values (Fig. 1). The experimental peak flux in 4·1% methylethylketone and in 1·5% 1-butanol exceeds the corresponding value in water at arbitrary subcooling with 50 per cent. This ratio follows also from equation (19) of [3] for $\theta_0^* \gg \theta_{0.\max}$; $\theta_{0.\max}$ is independent of θ_0^*, cf. Section 2.3.

Carne [4] investigated the effect of composition on peak flux in subcooled toluene–acetone mixtures with forced circulation on an electrically heated vertical stainless steel tube. In mixtures with 50–80% acetone a maximal increase occurred, which exceeded corresponding to the value in acetone with 15 per cent and

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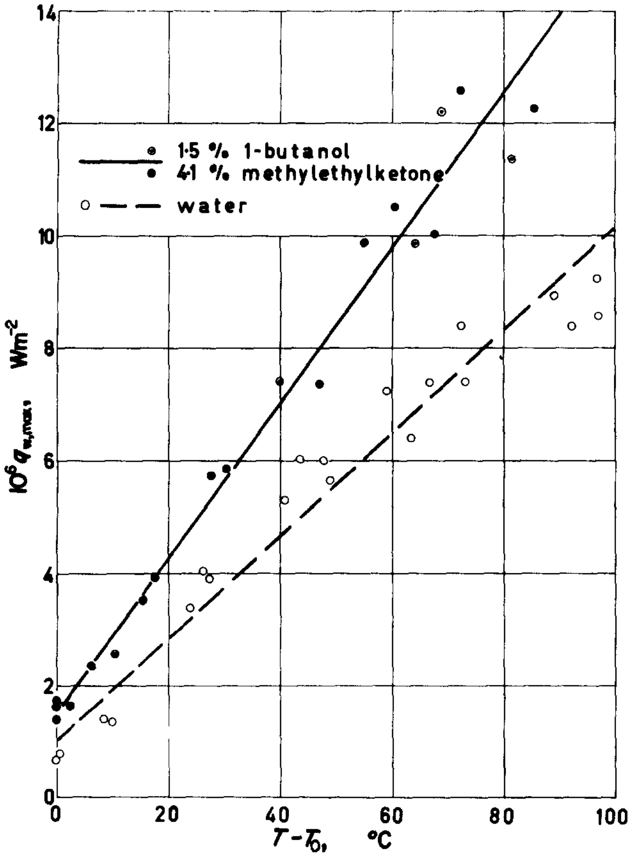

FIG. 1. *Water*, 1·5 wt.% *1-butanol* and 4·1 wt.% *methylethylketone.* Peak flux density in surface boiling with free convection in dependence on subcooling, at atmospheric pressure. The boiling points are 100, 97 and 88 degC, respectively. The temperature T_0 of the bulk liquid has been measured at a distance of 5 cm to the centre of the platinum heating wire.

the corresponding value in toluene with 53 per cent for stream velocities of 0–5 m/s and subcoolings of 0–60 degC.

2. SURFACE BOILING WITH FREE CONVECTION IN PURE LIQUIDS

2.1. *Bubble growth and maximum radius R_1^* in dependence on the degree of subcooling θ_0^**

It follows from equation (27) of [2] by taking $h_{w,co}$ independent of θ_0—i.e. the exponent in the right-hand side of (19) of [3] is taken as one here—that one can introduce now a superheated "effective relaxation microlayer" for bubble growth, the thickness of which amounts to (cf. Fig. 2 and equation (13) of [3]):

$$d^*_{0,p} = \frac{\theta_0}{\theta_0 + \theta_0^*} d_{0,p} = \left(\frac{12}{\pi}\right)^{\frac{1}{2}} \frac{\theta_0}{\theta_0 + \theta_0^*} (at_{1,p})^{\frac{1}{2}} = \left(\frac{12}{\pi}\right)^{\frac{1}{2}} (at^*_{1,p})^{\frac{1}{2}}. \quad (1)$$

The thickness $d_{0,p}$ of the entire relaxation microlayer is thus assumed to be independent of θ_0^*, but apparently only the excess enthalpy of the

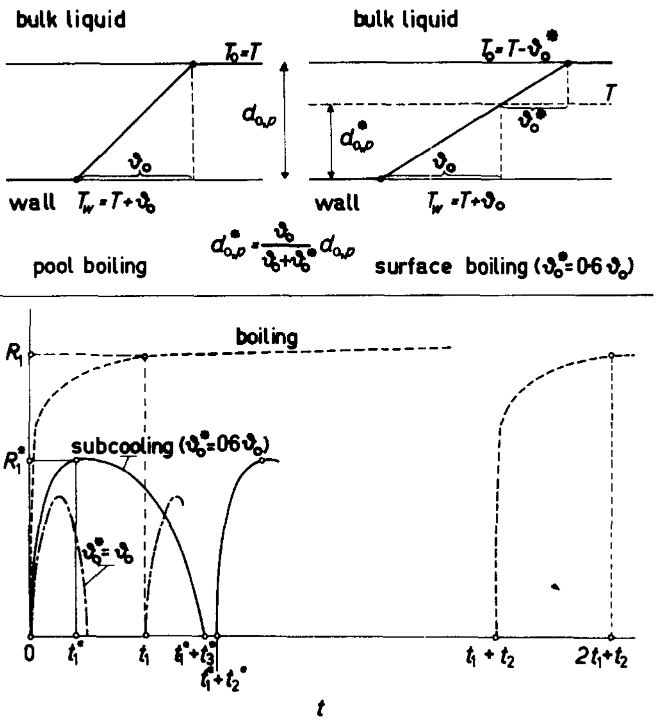

FIG. 2. *Arbitrary pure liquid.* Temperature drop in the equivalent conduction layer at the heating surface in nucleate pool boiling and in surface boiling. The entire layer with thickness $d_{0,p}$ is superheated in nucleate boiling, but only so the partial layer $d^*_{0,p}$ in surface boiling. Note that $d_{0,p} = 0{\cdot}79\, d_w$, cf. equation (27) of [2], has been drawn instead of d_w. Actually, the superheating of $d_{0,p}$ is assumed to be uniform, and the superheating of d_w is linear.

Bottom curves represent the bubble growth and the departure or the maximum radius in dependence of time. A slow subsequent growth after departure occurs in nucleate boiling, a rapid condensation in surface boiling. In the last case, the behaviour at two different ratios of liquid subcooling to wall superheating is shown.

superheated part with thickness $d^*_{0,p}$ can be used for bubble growth.

It is seen from (1), that:

$$(t^*_{1,p})^{\frac{1}{2}} = \frac{\theta_0}{\theta_0 + \theta_0^*} t^{\frac{1}{2}}_{1,p}, \tag{2}$$

whence the maximum radius is given by:

$$R^*_{1,p} = \frac{b}{e} C_{1,p}\,\theta_0 (t^*_{1,p})^{\frac{1}{2}} = \frac{\theta_0}{\theta_0 + \theta_0^*} R_{1,p}$$
$$= \frac{b}{e} C_{1,p} \frac{\theta_0^2}{\theta_0 + \theta_0^*} t^{\frac{1}{2}}_{1,p}. \tag{3}$$

The ratio

$$\frac{d^*_{0,p}}{R^*_{1,p}} = \frac{d_{0,p}}{R_{1,p}} \tag{4}$$

is thus independent of θ_0^*. Both $(t^*_{1,p})^{\frac{1}{2}}$, $d^*_{0,p}$ and $R^*_{1,p}$ decrease for increasing θ_0^*; the relation of these quantities to θ_0^* is hyperbolical.

2.2. *Subsequent condensation of the bubble*

Condensation of the bubble occurs after R_1^* has been reached. For the "condensation constant" in an initially uniformly subcooled pure liquid the following expression:

$$C^*_{1,p} = C_{1,p} = \left(\frac{12}{\pi}\right)^{\frac{1}{2}} \frac{a^{\frac{1}{2}}}{\rho_2 l/\rho_1 c} \tag{5}$$

can be deduced from equation (19) of [1], i.e. the effects of inertia and surface tension on bubble behaviour can be neglected. The heat diffusion is directed now away from the bubble centre, whence a surrounding subcooled microlayer is warmed up, cf. Section 2.3. The equation (5) can also be derived from the Scriven theory [5, 6, 7]; at the bubble boundary one has the condition:

$$\begin{aligned}\theta_0 &= \frac{\rho_2 l}{\rho_1 c}\varphi(0, \beta)\\ &= \frac{\rho_2 l}{\rho_1 c} 2\beta^3 \exp \beta^2 \int_{\beta}^{\infty} \zeta^{-2} \exp(-\zeta^2)\, \mathrm{d}\zeta. \end{aligned} \tag{6}$$

The effect of radial convection on bubble growth has been neglected in (6) by taking $\varepsilon = 0$, which is allowed for relatively large values of θ_0 [5, 7, 8], i.e. (6) is identical with an expression in Bruijn's theory [9, 6]. For subcooling, i.e. for negative superheating, (6) still holds, as:

$$-\theta_0 = \frac{\rho_2 l}{\rho_1 c}\varphi(0, -\beta) = -\frac{\rho_2 l}{\rho_1 c}\varphi(0, \beta).$$

For relatively large

$$\beta = C_{1,p}\theta_0/2a^{\frac{1}{2}}, \varphi(\varepsilon, \beta) \to \left(\frac{\pi}{3}\right)^{\frac{1}{2}} \beta$$

[5, 6, 7], which yields (5), i.e. (10) of [1].

Analogously to the bubble growth at a heating surface, cf. equations (21) and (27) of [1], the condensation can be described by (cf. [6] and Fig. 2):

$$R^*_p(t) = R^*_{1,p} - b\, C^*_{1,p}\theta^*_0 t^{\frac{1}{2}} \exp -(t/t^*_{3,p})^{\frac{1}{2}}.$$

The physical interpretation of this equation is based on a gradually decreasing condensation rate on account of a simultaneous heating of an adjacent liquid microlayer due to the liberated heat of condensation. Actually, however, the bubble boundary, especially at the base, will be covered gradually with flowing liquid with an increasing subcooling; the following equation will yield more realistic predictions:

$$\begin{aligned}&R^*_p(t) = bC^*_{1,p}\theta^*_0\{(t^*_{3,p})^{\frac{1}{2}} - t^{\frac{1}{2}}\} \exp\\ &\quad -\frac{(t^*_{3,p})^{\frac{1}{2}} - t^{\frac{1}{2}}}{(t^*_{3,p})^{\frac{1}{2}}} = \frac{b}{\mathrm{e}} C^*_{1,p}\theta^*_0\{(t^*_{3,p})^{\frac{1}{2}} - t^{\frac{1}{2}}\}\\ &\quad \exp\left(\frac{t}{t^*_{3,p}}\right)^{\frac{1}{2}} = \frac{b}{\mathrm{e}} C^*_{1,p}\theta^*_0 (t^*_{3,p})^{\frac{1}{2}}\\ &\quad \exp\left(\frac{t}{t^*_{3,p}}\right)^{\frac{1}{2}} - \frac{b}{\mathrm{e}} C^*_{1,p}\theta^*_0 t^{\frac{1}{2}} \exp\left(\frac{t}{t^*_{3,p}}\right)^{\frac{1}{2}}.\end{aligned} \tag{7}$$

One has thus:

$$\frac{\mathrm{d}R^*_p}{\mathrm{d}t^{\frac{1}{2}}} = -\frac{b}{\mathrm{e}} C^*_{1,p}\theta^*_0 \left(\frac{t}{t^*_{3,p}}\right)^{\frac{1}{2}} \exp\left(\frac{t}{t^*_{3,p}}\right)^{\frac{1}{2}},$$

whence $(\mathrm{d}R^*_p/\mathrm{d}t^{\frac{1}{2}})_{t=0} = 0$ and $(\mathrm{d}R^*_p/\mathrm{d}t^{\frac{1}{2}})_{t=t^*_{3,p}} = -bC^*_{1,p}\theta^*_0$, i.e. the maximum condensation rate occurs at the final stage of the bubble lifetime. Equation (7) is similar to the behaviour during the preceding bubble growth, cf. equation (21) of [1]:

$$R_p(t) = bC_{1,p}\theta_0 t^{\frac{1}{2}} \exp -\left(\frac{t}{t^*_{1,p}}\right)^{\frac{1}{2}}.$$

One has in this case:

$$\frac{\mathrm{d}R_p}{\mathrm{d}t^{\frac{1}{2}}} = bC_{1,p}\theta_0 \left\{1 - \left(\frac{t}{t^*_{1,p}}\right)^{\frac{1}{2}}\right\} \exp -\left(\frac{t}{t^*_{1,p}}\right)^{\frac{1}{2}},$$

whence

$$(\mathrm{d}R_p/\mathrm{d}t^{\frac{1}{2}})_{t=0} = bC_{1,p}\theta_0$$

and

$$(\mathrm{d}R_p/\mathrm{d}t^{\frac{1}{2}})_{t=t^*_{1,p}} = 0.$$

It follows from (7) in combination with (5) and (3), that

$$R^*_{1,p} = \frac{b}{\mathrm{e}} C_{1,p}\theta^*_0 (t^*_{3,p})^{\frac{1}{2}} = \frac{b}{\mathrm{e}} C_{1,p}\theta_0 (t^*_{1,p})^{\frac{1}{2}}. \tag{8}$$

The lifetime or the instant $t^*_{1,p} + t^*_{3,p}$, at which the bubble has disappeared entirely, is thus given by the condition (Fig. 2):

$$\left(\frac{t^*_{3,p}}{t^*_{1,p}}\right)^{\frac{1}{2}} = \frac{\theta_0}{\theta^*_0}. \tag{9}$$

According to equation (13) of [2], the succeeding bubble on the same nucleus in surface boiling is initially generated at the instant $t^*_{1,p} + t^*_{2,p}$, where

$$t^*_{2,p} = 3t^*_{1,p}, \tag{10}$$

whence the bubble frequency on a nucleus

$$\nu^*_p = \frac{1}{4t^*_{1,p}} = \left(1 + \frac{\theta^*_0}{\theta_0}\right)^2 \frac{1}{4t_{1,p}} \tag{11}$$

increases rapidly for increasing θ^*_0.

Condensation of vapour bubbles, which adhere meanwhile to the heating surface, is predicted to be possible only for $t^*_{3,p} < t^*_{2,p}$, i.e. for sufficiently large subcoolings $\theta^*_0 > (1/\sqrt{3})\theta_0 = 0{\cdot}58\ \theta_0$ (Fig. 2) For relatively small subcoolings $\theta^*_0 \leqslant 0{\cdot}58\,\theta_0$, the bubbles must depart from the wall in order to avoid a premature coalescence of succeeding bubbles. Some preliminary experimental results of the author on water yielded, that at $\theta_0 = 20$ degC, the bubbles depart from the heating surface for $\theta^*_0 \leqslant 16$ degC (Fig. 3), i.e. for $\theta^*_0 \leqslant 0{\cdot}80\ \theta_0$.

It may be noticed here, that during the condensation of a liquid drop, which is adhering to a wall, the wetting parameter B (cf. Section 2.3 of [1]) has to be replaced by $1 - B$. The bubble growth parameter b, which occurs in equations (7) and (8), is related to B and to the microlayer parameter b^* according to:

$$b = \frac{b^*}{\{B^2(3 - 2B)\}^{\frac{1}{2}}}. \tag{12}$$

For $B = 0{\cdot}75$ (bubble growth), (12) yields: $b = 1{\cdot}12\,b^*$, whence $b = 0{\cdot}84$ in case of heat supply towards the entire bubble boundary ($b^* = B$). For $B = 0{\cdot}25$ (dropwise condensation): $b = 3{\cdot}45\,b^*$. However, $b^* = 0{\cdot}25$ in case of heat

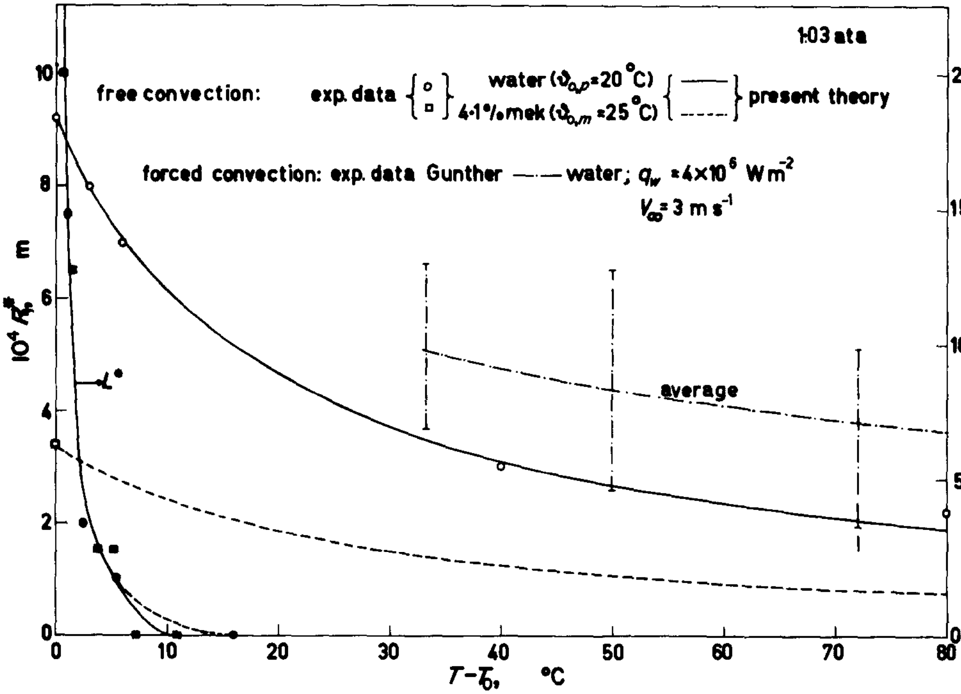

FIG. 3. *Water and* 4·1 wt. % *methylethylketone.* Comparison of theoretical maximum bubble radius in surface boiling according to equation (3) with some preliminary experimental data, partly deduced from high speed motion pictures taken at a rate of 10000 frames per s.

Average data of Gunther for forced circulation exceed the theoretical values with approximately 68 per cent (Table 2). The vapour bubbles collapse, meanwhile being adhered to the heating wire, at experimental subcoolings, exceeding 16 degC in water (———— ●) and 9 degC (— — — ■) in the mixture.

removal through the entire vapour–liquid interface, whence $b = 0{\cdot}86$. These values differ only slightly, i.e. the same value of b can be taken in case of dropwise condensation.

The reader is referred to the Appendix for a comparison with some experimental data on subcooled water by Ellion [11, 10, 12] for free convection and by Gunther [13, 14] for forced circulation.

The experimental data in Fig. 3 at low subcoolings of water are deduced from the photographs (Figs. 4 and 5).

Gunther's data (Table 2) show, that $C^*_{1,e} < C_{1,e}$ for large θ_0^*. More accurately, $bC^*_{1,e} < bC_{1,e}$ then; i.e. according to (12) for constant B during condensation, the microlayer parameter b^* decreases for increasing θ_0^*. This means, that cold liquid covers only the lowest part of the bubble boundary during the decreasing condensation time.

In case of bubble growth at a heating surface, it follows from equations (35) of [1] and (13) of [2], that $d_{0,p} = (12/\pi)^{\frac{1}{2}} (at_{1,p})^{\frac{1}{2}} = (4/\pi)^{\frac{1}{2}} (at_{2,p})^{\frac{1}{2}}$, whence $d_{0,p}^2 = 0{\cdot}32\,(4at_{2,p})$; the average initial superheating of the relaxation microlayer amounts thus to $0{\cdot}72\ \theta_0$. The value 0·72 agrees exactly with the experimental value b_e, cf. the appendix to [1]; i.e. one may take $b = 1$ in the growth equation for the equivalent radius (Section 2.3.2 of [1]) and replace θ_0 by $0{\cdot}72\ \theta_0$. One has thus for $B = 0{\cdot}75$: $b^* = (27/32)^{\frac{2}{3}} = 0{\cdot}89$, for $B = 0{\cdot}50$ (hemispherical bubbles according to Forster [10]): $b^* = (1/2)^{\frac{2}{3}} = 0{\cdot}63$ and for $B = 1{\cdot}00$ (spherical bubbles): $b^* = 1{\cdot}00$.

Note, that the peak flux density is independent of the value of b, cf. [2, 3] and Section2.3.

2.3. *The peak flux density*

It follows from (5) of [3], that, if k, ρ_1 and c are assumed to be independent of temperature:

$$q^*_{w,bi,p,\max} = \frac{1}{\pi^{\frac{1}{2}}}(k\rho_1 c)^{\frac{1}{2}} \frac{1}{(t^*_{1,p,\max})^{\frac{1}{2}}}(\theta^*_{0,\max} + \theta_0)\left(1 - \frac{1}{\mathrm{e}}\right)$$

$$= \frac{1}{\pi^{\frac{1}{2}}}(k\rho_1 c)^{\frac{1}{2}} \frac{1}{t^{\frac{1}{2}}_{1,p,\max}} \frac{(\theta_{0,\max} + \theta_0^*)^2}{\theta_{0,\max}}\left(1 - \frac{1}{\mathrm{e}}\right), \tag{13}$$

and, from (4) of [3], that the contribution of the direct vapour formation is expressed by:

$$q^*_{w,b,p,\max} = \frac{1}{2}\left(\frac{3}{\pi}\right)^{\frac{1}{2}} (k\rho_1 c)^{\frac{1}{2}} \frac{1}{(t^*_{1,p,\max})^{\frac{1}{2}}}\theta_{0,\max}\left(1 - \frac{1}{\mathrm{e}}\right)$$

$$= \frac{1}{2}\left(\frac{3}{\pi}\right)^{\frac{1}{2}} (k\overset{*}{\rho}_1 c)^{\frac{1}{2}} \frac{1}{t^{\frac{1}{2}}_{1,p,\max}}(\theta_{0,\max} + \theta_0^*)\left(1 - \frac{1}{\mathrm{e}}\right) \simeq \frac{\theta_{0,\max}}{\theta_{0,\max} + \theta_0^*} q^*_{w,bi,p,\max}, \tag{14}$$

whence

$$q^*_{w,bi,p,\max} - q^*_{w,b,p,\max} = \frac{\theta_0^*}{\theta_{0,\max} +^* \theta_0^*} q^*_{w,bi,p,\max}. \tag{15}$$

The direct vapour production is subtracted here on account of the periodical condensation, i.e. formally. the superheated part of the relaxation microlayer is warmed up by the heat of condensation, and not by the heating surface, cf. (7). The wall has to warm up only the remaining part of the microlayer (with thickness $d_{0,p} - d^*_{0,p}$), cf. Fig. 2, which is periodically pushed away by the growing bubbles. This behaviour differs strikingly from that in saturated pool boiling, where $q_{w,b,p,\max}$ is removed from the wall by the ascending bubbles.

One has thus in comparison to nucleate pool boiling:

$$\frac{q^*_{w,bi,p,\max} - q^*_{w,b,p,\max}}{q_{w,bi,p,\max}} = \frac{\theta_0^*}{\theta_{0,\max} + \theta_0^*}\left(\frac{\theta_{0,\max} + \theta_0^*}{\theta_{0,\max}}\right)^2 = \frac{\theta_0^*}{\theta^2_{0,\max}}(\theta_{0,\max} + \theta_0^*) \tag{16}$$

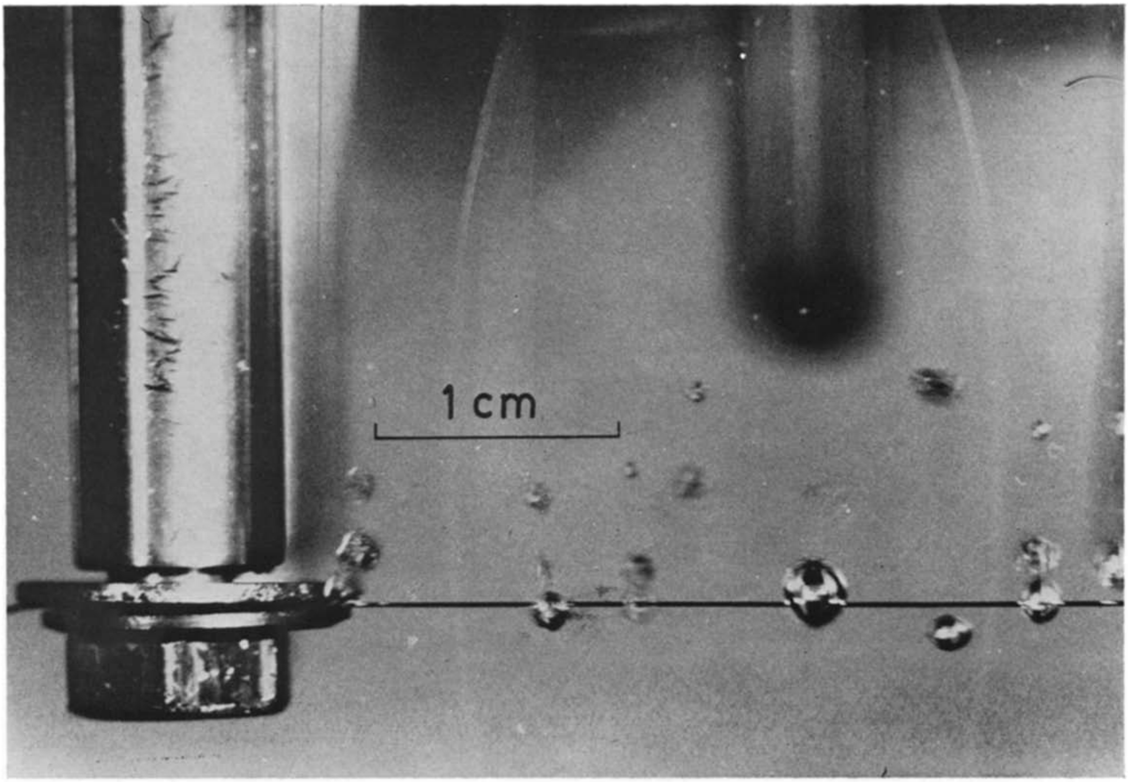

FIG. 4. *Water*. Surface boiling with free convection at atmospheric pressure, $\theta_0^* = 3$ degC, $q_w = 25 \times 10^4\ \mathrm{W/m^2}$. Columns of vapour bubbles are generated at the heating wire and condense in the neighbourhood of the wire.

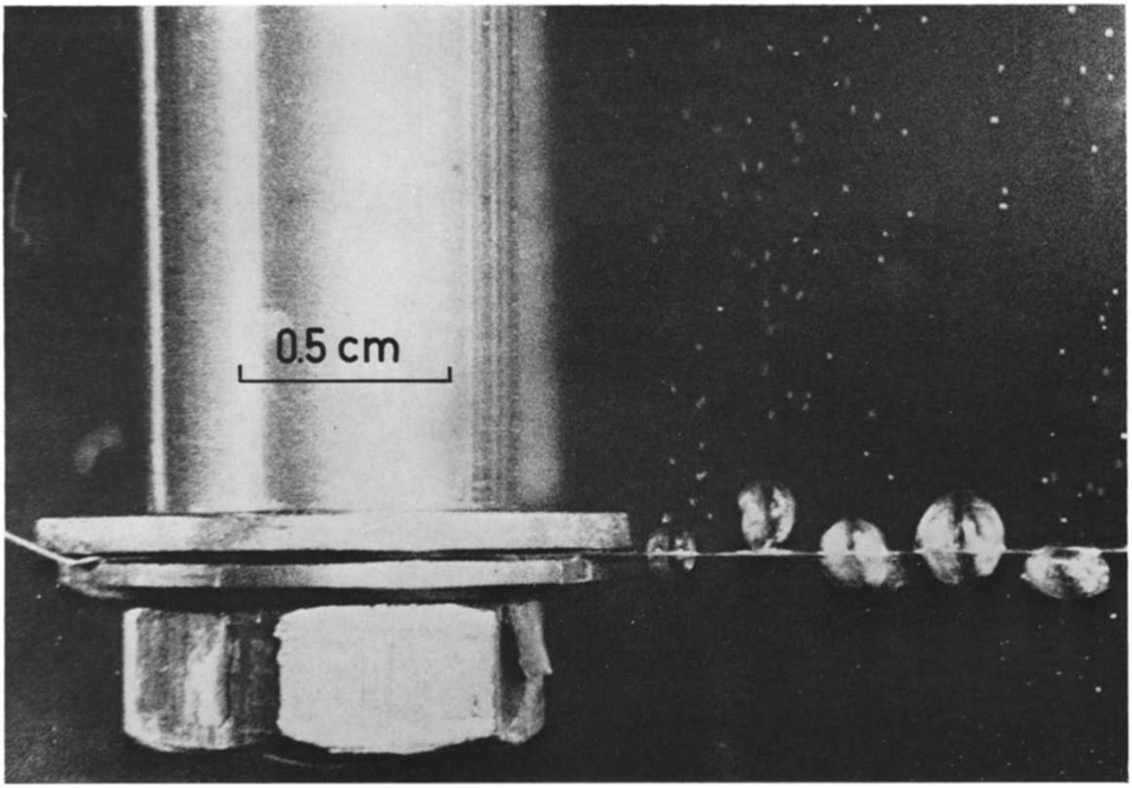

FIG. 5. *Water*. Surface boiling with free convection at atmospheric pressure, $\theta_0^* = 6$ degC, $q_w = 37 \times 10^4$ W/m^2. The vapour bubbles condense now more rapidly due to the increased θ_0^*.

However, small bubbles remain and ascend slowly to the liquid-level surface. It is believed, that these small bubbles acted previously as "equilibrium bubbles", necessary for initial vapour formation.

or, in combination with equation (14) of [3]:

$$\frac{q^*_{w,p,\max}}{q_{w,p,\max}} = \frac{\left\{\dfrac{\theta_{0,\max} + \theta_0^*}{\theta_{0,\max}} + 0{\cdot}88\,\dfrac{\theta_0^*}{\theta_{0,\max}^2}(\theta_{0,\max} + \theta_0^*)\right\} q_{w,co,\max}}{(1 + 0{\cdot}88)\,q_{w,co,\max}} = \frac{\theta_{0,\max} + \theta_0^*}{1{\cdot}88\,\theta_{0,\max}}\left(1 + 0{\cdot}88\,\frac{\theta_0^*}{\theta_{0,\max}}\right) \tag{17}$$

e.g. for water at room temperature $\theta_{0,\max} = 22$°C (independent of the degree of subcooling, cf. [15]) and $\theta_0^* = 80$°C, whence $q^*_{w,p,\max} = 11{\cdot}1\ q_{w,p,\max}$, in good agreement with the experimental data (Fig. 1).

3. SURFACE BOILING IN BINARY MIXTURES

3.1. *Growth and condensation of vapour bubbles*

The validity of equation (3) for the maximum bubble radius can be extended to the case of vapour bubbles in binary mixtures with a more volatile component by taking $C_1 = C_{1,m}$. The maximum radius occurring at constant θ_0 and θ_0^* shall thus be reduced considerably in comparison to the value in pure liquids, i.e. to 25 per cent in 4·1% methylethylketone compared to water.

However, the subsequent condensation rate is not slowed down in mixtures, but, contrarily, increased in certain mixtures. This follows from the expression governing the growth of free, spherical bubbles in initially uniformly superheated mixtures at relatively low pressures [5, 6, 7]:

$$\theta_0 - \Delta T = \frac{\rho_2 l}{\rho_1 c}\varphi(0, \beta), \tag{18}$$

where

$$\Delta T = \left(1 - \frac{C_{1,m}}{C_{1,p}}\right)\theta_0. \tag{19}$$

(18) is the corresponding extension of equation (6) to mixtures, cf. [6, 7]. The "effective superheating" of the liquid at the bubble boundary amounts now to $\theta_0 - \Delta T$. Analogously, in condensation the "effective subcooling" with respect to the bubble boundary equals $\theta_0^* + \Delta T$, where

$$\Delta T = \left(1 - \frac{C_{1,m}}{C_{1,p}}\right)\theta_0^* \leqslant \theta_0^*, \tag{20}$$

whence

$$\frac{C^*_{1,m}}{C^*_{1,p}} = \frac{C^*_{1,m}}{C_{1,p}} = \frac{\theta_0^* + \Delta T}{\theta_0^*} = 2 - \frac{C_{1,m}}{C_{1,p}}$$

$$= 2 - \frac{1}{1 + \left(\dfrac{a}{D}\right)^{\frac{1}{2}}\dfrac{c}{l}\dfrac{\Delta T}{G_d}} = \frac{1 + 2\left(\dfrac{a}{D}\right)^{\frac{1}{2}}\dfrac{c}{l}\dfrac{\Delta T}{G_d}}{1 + \left(\dfrac{a}{D}\right)^{\frac{1}{2}}\dfrac{c}{l}\dfrac{\Delta T}{G_d}}. \tag{21}$$

The condensation rate in mixtures is thus maximally increased up to a factor 2 in comparison to the main component, i.e. in 4·1% methylethylketone [1, 2]: $C_{1,m} = 6 \times 10^{-4}$ m/s$^{\frac{1}{2}}$ degC, $C_{1,p} = C^*_{1,p} = 24 \times 10^{-4}$ m/s$^{\frac{1}{2}}$ degC in water, whence $C^*_{1,m} = 42 \times 10^{-4}$ m/s$^{\frac{1}{2}}$ degC is predicted for bubbles in surface boiling at atmospheric pressure.

In case of bubbles generated at a heating surface, the factor 2 in (21) is reduced to 1 + (e − 1)/e = 1·63, cf. equation (54) of [2]. In the extreme case, equation (9) is extended to:

$$\left(\frac{t^*_{3,m}}{t^*_{1,m}}\right)^{\frac{1}{2}} = \frac{C_{1,m}}{C^*_{1,m}}\frac{\theta_0}{\theta_0^*} = \frac{C_{1,m}}{C_{1,p}}\frac{C^*_{1,p}}{C^*_{1,m}}\frac{\theta_0}{\theta_0^*}$$

$$= \frac{\theta_0 - 0{\cdot}63\,\theta_0}{\theta_0} \times \frac{\theta_0^*}{\theta_0^* + 0{\cdot}63\,\theta_0^*}\frac{\theta_0}{\theta_0^*}$$

$$= \frac{0{\cdot}37}{1{\cdot}63}\frac{\theta_0}{\theta_0^*} = 0{\cdot}23\,\frac{\theta_0}{\theta_0^*}, \tag{22}$$

whence $t^*_{3,m}$ should decrease maximally up to a factor 20 at a constant ratio θ_0/θ_0^* in comparison to pure liquids.

Extremely high condensation rates are predicted in certain mixtures for large θ_0^*. Actually, however, these values will be decreased, as the instantaneous superheating at the instant $t^*_{1,m}$ of the liquid at the bubble boundary amounts to 0·63 θ_0 instead of 0·63 θ_0^*. It follows from equation (8) of [2], that $t^*_{2,m}/t^*_{2,p} = (C_{1,m}/C_{1,p})^2$, i.e. also a very considerable decrease of the waiting time in the subcooled mixtures is predicted.

The author's future research program includes experimental investigations on this subject. Until now, only some data by Ellion [11] on a mixture with a nonvolatile wetting agent (water–aerosol), which are reported by Sabersky [12], are known in the literature. It is easy to extend the theory for binary mixtures with a more volatile component to the case of a nonvolatile component [6, 7]. However, at present Ellion's data cannot be compared with the theoretical predictions, as the corresponding θ_0 is not reported in [12].

3.2. *The peak flux density*

For sufficiently large subcooling $\theta_0^* \gg \theta_{0,\max}$, the driving temperature difference $\theta_0^* + \theta_{0,\max} \simeq \theta_0^*$, whence equation (19) of [3] predicts a 50 per cent higher peak flux in certain mixtures in comparison to the pure less volatile component at the same θ_0^*. This value is in quantitative agreement with the experimental data for 4·1 % methylethylketone and 1·5 % 1-butanol in water (Fig. 1).

A very high peak flux of 15 × 10^6 W/m^2 occurs in these mixtures at atmospheric pressure and at a subcooling of 90 degC, in comparison to 10 × 10^6 W/m^2 in water. This value exceeds the peak flux in boiling water with a factor 20, and the maximum peak flux in water, boiling at the elevated pressure of 75 ata (Fig. 3 of [3]) with a factor 5.

Even higher values were obtained by Gunther [13] by application of forced circulation to water under elevated pressures, e.g. 36 × 10^6 W/m^2 at $V_\infty = 12$ m/s, $p = 8$ ata and $\theta_0^* = 140$ degC. The experimental data of this worker agreed with an empirical equation:

$$q^*_{w,p,\max} \sim V_\infty^{\frac{1}{2}}\,\theta_0^*,$$

cf. equation (20) of [3] and Fig. 1. The highest peak fluxes to water, which are known in the literature, were obtained by Gambill and Greene [16] in vortex flow (170 × 10^6 W/m^2). Probably, even an increase of this value shall be made possible by utilization of suitable binary mixtures.

4. CONDENSATION OF SUPERSATURATED BINARY VAPOUR MIXTURES

4.1. *Experimental investigations*

The existence of subcooled vapour has been demonstrated by Volmer and Flood [17, 18] for water, by Flood and Tronstad [19] for heavy water, and by Froemke, Bloomquist and Anderson [20] and Flood [21] for water–methanol and water–ethanol mixtures, respectively. These investigations were of practical importance for utilization in the Wilson cloud chamber. A supersaturation of the initially saturated vapour occurs rapidly by means of adiabatic expansion, which causes a sudden decrease of the temperature with an amount θ_0^*.

The minimum expansion ratio (corresponding with the "critical subcooling") was determined, for which dropwise condensation occurs spontaneously in the absence of nuclei, i.e. ions or dust particles. The charged particles were previously removed by the application of an electrostatic field.

The mixtures condense more easily, i.e. at a lower critical expansion ratio in comparison with the behaviour of the pure components, e.g. for water–ethanol in air a minimum expansion ratio (1·10, i.e. 11·6 degC temperature drop) occurs in vapour containing 35 mol % (60 wt. %) ethanol. This corresponds with an equilibrium composition of the liquid in the droplets of 8·5 mol % (20 wt. %) ethanol. The critical ratio amounts to 1·27 (temperature drop 27·3 degC) in water and to 1·17 (temperature drop 16·3 degC) in ethanol.

Strikingly, the nucleate boiling peak flux shows a maximum, exactly at the same liquid concentration, and $\Delta T/G_d$ shows also a maximum at approximately the same composition, viz. at 6 mol % (14 wt. %) ethanol, cf. [15].

The corresponding liquid composition in the nucleus can be calculated from equilibrium data, or (if not available) from the vapour pressure curves [21] using the Thomson equation [22] for the increase in vapour pressure at a convex liquid surface with a small radius of curvature (corresponding with a lowering of the boiling point). If even partial vapour pressure curves for the mixture at low temperatures are not known, the Duhem–Margules equation has to be used for this purpose [20].

More recent measurements of the minimum expansion ratio for water–ethanol vapour mixtures in air give a lower value for water (1·22, corresponding with a temperature drop of 23 degC), but the previous value (1·17) in ethanol [23]. The minimum (1·12) in water–ethanol vapour mixtures occurs at a temperature drop of 13 degC. This minimum can further be lowered to 1·07 by using a monatomic gas (argon) instead of air [23].

The critical supersaturation S can be calculated from the experimental data by the expression:

$$R_gT_0 \ln S = R_gT_0 \ln \left(\frac{p_{2,R_0^*}}{p_{2,\infty}}\right)^{x_M} \left(\frac{p_{1,R_0^*}}{p_{1,\infty}}\right)^{1-x_M} = \frac{2\sigma}{R_0^*} V', \tag{23}$$

where V' denotes the molar volume of the mixture in the droplet.

According to Volmer's nucleation theory [17, 18] $R_0^* \sim (T_0/\sigma)^{\frac{1}{2}}$, whence

$$\ln S \sim \left(\frac{\sigma}{T_0}\right)^{\frac{3}{2}} V'. \tag{24}$$

4.2. *Explanation by the present theory*

According to the present theory, one has to distinguish between the mass fraction of the more volatile component x at the boundary of the droplet and the fraction x_0 of the liquid inside the droplet. For 20 wt. % ethanol $C_{1,m} = 16 \times 10^{-4}$ m/s$^{\frac{1}{2}}$ degC, whence $C_{1,m}/C_{1,p} = 0{\cdot}67$ and $C^*_{1,m}/C^*_{1,p} = 1{\cdot}33$. The experimental data yield: $\theta^*_{0,m} + \Delta T = \theta^*_{0,p}$, whence $\Delta T = 23 - 13 = 10$ degC, or $\Delta T/\theta^*_{0,p} = 0{\cdot}43$. It follows thus in combination with equation (21), that the experimental data give $C^*_{1,m}/C^*_{1,p} = 1{\cdot}43$, which is in reasonable agreement with the theoretical value.

The occurrence of a minimal expansion ratio in cloud chambers with supersaturated vapour mixtures can thus be explained satisfactorily with the theory. The coincidence of the corresponding liquid composition at the minimum expansion ratio with the composition of maximal slowing bubble growth (and maximal peak flux) is obvious now. The "coalescence" theory [20], which is based on a hypothetical formation of molecule-aggregates in vapour mixtures, is shown here to be unreliable.

It may be noticed, that: (i) the growth of the droplets is stopped readily on the account of the rapidly decreasing θ^*_0 due to the heat of condensation, which is not removed in this case; (ii) the theoretical maximum radius of the drops in the vapour mixture of minimal critical expansion ratio exceeds the value in the pure vapour mixtures, in agreement with experimental data [24]; (iii) the formation of condensation nuclei in vapour mixtures may occur more easily, as the required energy is proportional to the surface tension constant, cf. [17, 18] and (24); (iv) the critical expansion ratios in absence of an electrostatic field were approximately 2 per cent lower due to drop formation on charges particles, in agreement with the extended Thomson equation, cf. [23]; (v) probably, the application of water–methylethylketone vapour mixtures in cloud chambers (and in dropwise condensation at the cooling surface in condensers) will be more advantageous with respect to a lowering of the critical expansion ratio.

5. NOTE ON THE ULTRASONIC VELOCITY AND ABSORPTION IN BINARY LIQUID MIXTURES

5.1. *Velocity*

Finally, the interesting phenomenon of the occurrence of a maximum ultrasonic velocity in binary mixtures may be brought to notice [25]. Ernst [26] observed no maximum, but a monotonous curve, in ideal mixtures (which obey Raoult's law), but a maximum at a low concentration of the more volatile component in nonideal mixtures. The concentration of maximum velocity in water–ethanol agrees again with the concentration of minimum bubble growth rates. This phenomenon is not yet clearly understood [27], but is generally attributed to a decreased association of water molecules due to the addition of ethanol.

5.2. *Absorption*

The following conclusions can be drawn from a survey on the behaviour of ultrasonic sound waves in liquids by Bergmann [25]. The modified frequency-independent absorption coefficient $\alpha'' = \alpha'/(v')^2$ for ultrasonic waves is mainly dependent on the dissipative effect of the kinematic viscosity η/ρ_1, viz. $\alpha'' \sim \eta/\rho_1(v')^3$, according to Stokes [28]. The contribution to α'' due to heat conduction is relatively small (0·1 per cent in water and 5 per cent in organic liquids) and has thus been neglected here. One should expect now, that a maximal v' at a certain concentration of a binary liquid mixture corresponds with a minimal α''.

Contrarily, however, the experimental data yield a maximal α''. This is apparently due to the dominating influence of η, e.g. in water ethanol mixtures at a temperature of 20 degC, η shows a maximum at 45 wt. % ethanol, which exceeds the value in water with a factor 2·8 and the value in ethanol with a factor 2·4 [15]. The maximal increase of v' amounts to approximately 10 per cent.

Jellinek [22] collected data from the literature, which report a lowering of the isothermal compressibility of water at room temperature due to an addition of NaCl with 80 per cent. This should correspond with an increase of v' of 40 per cent. A considerably higher v' in sea water has actually been observed [27].

6. SURVEY

For practical utilizations, aqueous binary mixtures may be advantageous with respect to the increased peak fluxes, both in nucleate pool boiling (at atmospheric pressure up to a factor 3 in comparison to water, at high pressures up to a factor 2) and in surface boiling (up to a factor 1·5). The smaller bubbles are more regular and more spherical and the formation of large vapour slugs is avoided due to a diminished tendency for coalescence. The peak fluxes are practically independent of the orientation of the heating surface. The most favourable concentration can be deduced from equilibrium data and both the behaviour of the bubbles and the values of the peak fluxes are predicted by the present theory, in quantitative agreement with experimental data.

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APPENDIX

Comparison With Experimental Data

1. *Surface boiling with free convection*

The author's preliminary data on the maximum bubble radius in water are in quantitative agreement with the theoretical equation (3), cf. Fig. 3. At $\theta_0 = 20°\text{C}$ and $\theta_0^* = 80°\text{C}$, $t^*_{3,e}/t^*_{1,e} = \frac{1}{14}$, in good agreement with the value $(\frac{1}{16})$ following from equation (9). Experimental data on bubbles in subcooled water with free convection of Ellion [11], cf. also Sabersky [12], are here also compared with the theoretical equation (3), (cf. Table 1):

$$R^*_{1,p} = \frac{b}{\mathrm{e}} C_{1,p} \theta_0 (t^*_{1,p})^{\frac{1}{2}}.$$

Only those experimental data are taken into consideration, which are also compared by

Table 1. *Comparison of equation* (3) *with data of Ellion* (*for water*). *The values of b are determined from the condition*: $R^*_{1,t} = R^*_{1,e}$

θ_0	θ_0^*	$10^3(t^*_{1,e})^{\frac{1}{2}}$	$10^4 R^*_{1,e}$	$10^4 R^*_{1,t}$ (for $b = 0.72$)	b
(degC)	(degC)	($s^{\frac{1}{2}}$)	(m)	(m)	
28	83	16	2·68	2·80	0·69
27	62	19	3·29	3·20	0·70
24	40	21·5	3·81	3·22	0·85

Table 2. Water in surface boiling with forced circulation according to experimental data of Gunther. These data on bubbles are taken at a constant $q_w = 4 \times 10^6$ W/m², *at a constant (atmospheric) pressure and a constant flow velocity* $V_\infty = 3$ m/s. T_0 = *mixed outlet temperature here. The values of* θ_0 *have not been reported, but estimated from boiling curves for water. The theoretical equation (9) is in good agreement with the experimental data. This holds also reasonably well for equation (8), i.e.* $C_{1,p} = C^*_{1,p}$ *in pure liquids, cf. the column at the right-hand side.*

Gunther's values of $R^*_{1,p}$ *exceed the theoretical values with 68 per cent (Fig. 3), and hence also the experimental data for free convection. The largest discrepancy occurs at high subcoolings, and may thus be due to a premature bubble coalescence, which can be caused by the geometrical arrangement (vertical, electrically heated metal strip in a narrow channel), cf. Section 5 of* [3]

Vapour bubble	θ^*_0 (degC)	Θ_0 (degC)	$\frac{\theta_0}{\theta^*_0}$	$10^{-6}\, q_{w,\max}$ (W/m²)	$10^2\, (t^*_{1,e})^{\frac{1}{2}}$ ($s^{\frac{1}{2}}$)	$10^2\, (t^*_{3,e})^{\frac{1}{2}}$ ($s^{\frac{1}{2}}$)	$\left(\frac{t^*_{3,e}}{t^*_{1,e}}\right)^{\frac{1}{2}}$	$10^4\, R^*_{1,e}$ (m)	$10^2\, C^*_{1,e}$ ($m/s^{\frac{1}{2}}$ degC)	$10^4\, R^*_{1,t}$ ($b = 0{\cdot}72$) (m)	$\frac{C_{1,e}}{C_{1,t}}$	$\frac{C^*_{1,e}}{C_{1,e}} = {}^{\prime}\frac{\theta_0}{\theta^*_0}\Big)\, \frac{t^*_{3,e}}{t^*_{1,e}}{}^{\frac{1}{2}}$
maximum	33·3	25	0·75	4·8	1·73	1·10	0·63	6·6	0·72			
minimum		17	0·51		1·41	0·94	0·67	3·7	0·47			
average		21	0·63		1·60	0·95	0·59	5·1	0·64	3·55	1·44	1·07
maximum	50	26	0·52	7·1	1·52	0·76	0·50	6·5	0·65			
minimum		15	0·30		1·27	0·78	0·62	3·0	0·31			
average		20	0·40		1·36	0·71	0·52	4·4	0·50	2·60	1·69	0·77
maximum	72	22	0·31	11·7	1·32	0·70	0·53	5·1	0·40			
minimum		9	0·13		0·95	0·39	0·41	1·5	0·21			
average		18	0·25		1·22	0·51	0·43	3·8	0·41	2·00	1·90	0·58

Forster [10] to check his equation:

$$2^{\frac{1}{2}} R^*_{1,p} = 0{\cdot}46 \frac{(k\rho_1 c)^{\frac{1}{2}}}{\rho_2 l} \frac{\theta_0}{\theta_0^*} (\theta_0 + \theta_0^*)(t^*_{1,p})^{\frac{1}{2}}, \tag{25}$$

where the factor $2^{\frac{1}{2}}$ arises from the conversion of hemispheres into equivalent spheres. For large $\theta_0^* \gg \theta_0$, this expression (25) is simplified to:

$$R^*_{1,p} = 0{\cdot}37 \frac{(k\rho_1 c)^{\frac{1}{2}}}{\rho_2 l} \theta_0 (t^*_{1,p})^{\frac{1}{2}} \tag{26}$$

where the constant (0·37) was determined empirically to fit Ellion's data. The constant in equation (3) amounts to

$$0{\cdot}26(12/\pi)^{\frac{1}{2}} = 0{\cdot}51 \text{ for } b = 0{\cdot}72,$$

cf. [1], which agrees reasonably well with (26), but only for large superheatings. It may be noticed, that Forster's equation fails to predict the bubble radius R_1 for pool boiling, i.e. for $\theta_0^* = 0$. A further disadvantage of Forster's theory in comparison to the present treatment is the impossibility to predict $q^*_{w.\max}$ in dependence on the degree of subcooling.

Good agreement is obtained for $\bar{b} = 0{\cdot}75$, i.e. practically the value for bubbles in boiling. The numerical value of $(t^*_{1,p})^{\frac{1}{2}}$ can be determined from equation (2), as $\overline{t^{\frac{1}{2}}_{1.p}} = 7{\cdot}0 \times 10^{-2}$ s$^{\frac{1}{2}}$ for pool boiling (Section 4·3 of [2]), whence $(t^*_{1.p})^{\frac{1}{2}} = \{20/(20 + 70)\}\, 7{\cdot}0 \times 10^{-2} = 1{\cdot}55 \times 10^{-2}$ s$^{\frac{1}{2}}$ at $\theta_0^* = 70$ degC. Ellion observed a value of $t^*_{1.p} = 3{\cdot}0 \times 10^{-4}$ at this subcooling, whence $(t^*_{1.p})^{\frac{1}{2}} = 1{\cdot}73 \times 10^{-2}$ s$^{\frac{1}{2}}$, i.e. in good agreement with the present theory.

2. *Forced circulation*

Gunther [13], cf. also Rohsenow [14], has taken high speed motion pictures at a rate of 20000 frames per s of vapour bubbles in subcooled water, generated on a vertical, electrically heated metal strip in a narrow transparent channel. A survey of some of his data is given in Table 2.

The most important result is, that the growth and condensation constants in a pure liquid have the same value, which is in fair agreement with the present theory, cf. Section 2.2.

Extrapolation of Gunther's data to the high subcooling of 100 degC gives for water maximal values of $(m^*_{\max}/A_w)v^*_{\max} \simeq 2{\cdot}5 \times 10^{10}$ bubbles/sm^2. Such astronomical population densities are also predicted by the present theory, viz. at nucleate pool boiling:

$$(m_{\max}/A_w)v_{\max} = 70 \times 10^4 \times \frac{1}{4(4{\cdot}09 \times 10^{-2})^2} = 1{\cdot}1 \times 10^8 \text{ bubbles/sm}^2,$$

cf. Table 3 of [2] and the appendix of [3]. At a subcooling $\theta_0^* = 100$ degC (and at $\theta_0 = 20$ degC), both $m^*_{\max}/A_w = 1/A_i^*$ and $v^* = 1/(4t_1^*)$, are increased in the ratio $\{(\theta_0 + \theta_0^*)/\theta_0\}^2 = 25$, as these quantities are proportional to $1/t_1^*$, cf. equation (14) and (19) of [2] and equation (2). Consequently, under these conditions $(m^*_{\max}/A_w)v^*_{\max} = 1{\cdot}1 \times 10^8 \times 625 = 6{\cdot}9 \times 10^{10}$ bubbles/sm^2. As a matter of fact, this value following from the present theory even exceeds Gunther's prediction. Quantitative agreement, however, is obtained by taking his larger maximum bubble radius into consideration, which causes a $(1{\cdot}68)^2$ smaller $m^*_{\max}/A_w$, cf. Table 2.

Résumé—Un effet favorable de la composition dans les mélanges binaires avec un constituant plus volatil sur le flux maximal dans l'ébullition de surface de liquides sous-refroidis avec convection naturelle a été prévue par la théorie de la "microcouche de relaxation" des parties I à III. Les valeurs théoriques sont en accord quantitatif avec les résultats expérimentaux de l'auteur sur les systèmes eau–méthyléthylcétone et eau–1-butanol.

Des expressions pour le flux maximal et pour le rayon maximal, la fréquence de croissance, les vitesses de condensation et la durée de vie des bulles ont été obtenues en fonction de la surchauffe du fil et du degré de sous-refroidissement. Une augmentation de la vitesse de condensation dans certains mélanges sous-refroidis n'est disponible mais les prévisions théoriques pour des liquides purs sont en accord raisonnable avec certains résultats préliminaires obtenus par l'auteur et avec des résultats sur l'eau obtenus par Ellion avec convection naturelle et par Gunther avec circulation forcée.

L'existence d'un rapport de détente critique minimal pour des mélanges de vapeur sursaturée en présence d'air dans des chambres de Wilson est expliqué et la valeur numérique du rapport minimal peut être calculée à partir de la nouvelle théorie. La concentration correspondante coïncide avec la concentration du ralentissement maximal de la croissance des bulles et du maximum maximorum de la densité de flux dans les mélanges liquides surchauffés. Ceci est en bon accord avec les résultats de Flood sur la vapeur d'eau et de méthanol et de Froemke, Bloomquist et Anderson sur l'eau et le méthanol.

Finalement, une note additionnelle traite de la vitesse et de l'absorption des ultrasons dans les mélanges liquides binaires.

Zusammenfassung—Mit der "relaxation microlayer" Theorie (Teil I–III) war ein bemerkenswerter Einfluss der Zusammensetzung eines binären Gemisches mit einer stark flüchtigen Komponente auf den maximalen Wärmestrom beim Oberflächensieden in unterkühlten Flüssigkeiten vorausgesagt worden.

Die theoretischen Werte stimmen mit Versuchsergebnissen des Autors für die Systeme Wasser–Methyläthylketon und Wasser–1-Butanol quantitativ überein.

Es wurden Ausdrücke für den maximalen Wärmestrom, den grössten Blasenradius, die Frequenz, die Wachstums- und Kondensationsgeschwindigkeit und die Lebensdauer, in Abhängigkeit von der Unterkühlung der Flüssigkeit und der Überhitzung des Heizdrahtes, abgeleitet. Ein Anwachsen der Kondensationsgeschwindigkeit bei gewissen binären Gemischen, wird vorausgesagt. Gegenwärtig sind keine Versuchsergebnisse über Blasen in unterkühlten Mischungen verfügbar, doch stimmen die theoretischen Voraussagen für reine Flüssigkeiten mit vorläufigen Versuchen des Autors und mit Versuchen bei freier Konvektion von Ellion und mit Versuchen bei erzwungener Konvektion von Gunther, recht gut überein. Mit der neuen Theorie wird das Auftreten eines minimalen kritischen Expansionsverhältnisses für übersättigte Dampfgemische in Gegenwart von Luft in Wilson'schen Nebelkammern erklärt. Der numerische Wert des kleinsten Verhältnisses kann mit dieser Theorie berechnet werden. Es wird gezeigt, dass die entsprechende Konzentration zusammenfällt mit der Konzentration, bei der das Blasenwachstum am stärksten abnimmt, und bei der in überhitzten Flüssigkeitsgemischen die maximale Wärmestromdichte auftritt.

Das ist in guter Übereinstimmung mit Werten von Flood für Wasser-Äthanoldampf und mit Werten von Froemke, Bloomquist und Anderson für Wasser-Methanol.

Abschliessend werden Bemerkungen über Überschallgeschwindigkeit und über Absorption in binären Flüssigkeitsgemischen gemacht.

Аннотация—В частях 1–3 с помощью теории «релаксации микрослоя» рассчитано положительное влияние состава бинарных смесей с более легким компонентом на максимум теплового потока при поверхностном кипении недогретой жидкости со свободной конвекцией. Теоретические расчеты количественно согласуются с экспериментальными данными автора для систем вода–метил-кетон и вода–бутанол.

Выражения для максимального теплового потока и максимального радиуса пузырька, роста частоты и скорости конденсации, а также времени жизни выводятся в зависимости от перегрева проволоки и степени недогрева. Рассчитано увеличение скорости конденсации для некоторых бинарных систем. В настоящее время не имеется экспериментальных данных по пузырьковому кипению недогретых жидкостей, однако теоретические расчеты для чистых жидкостей находятся в хорошем соответствии с некоторыми предварительными данными автора, а также с данными Элисона по кипению воды при свободной конвекции и данными Гунтера для воды при вынужденной циркуляции.

С помощью новой теории объясняется наличие минимального критического коэффициента расширения для перегретых смесей пара при наличии воздуха в камере Вильсона, а также рассчитано числовое значение коэффициента расширения. Показано, что соответствующая концентрация совпадает с концентрацией максимального замедления роста пузырьков и максимальной критической плотности теплового потока в перегретых жидких смесях. Это находится в хорошем соответствии с данными Флуда для системы вода–пары этанола и с данными Фромке, Блумквиста и Андерсона для воды–метанола.