

LOCAL TEMPERATURE FLUCTUATIONS IN SATURATED POOL BOILING OF PURE LIQUIDS AND BINARY MIXTURES

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Abstract—A direct experimental verification of the common physical background underlying the various bubble growth theories and Van Stralen's "relaxation microlayer" theory for the mechanism of nucleate boiling is given.

For this purpose, instantaneous local temperature measurements with a thin-wired thermocouple have been synchronized with high speed motion pictures of vapour bubbles generated at active nuclei in close proximity to the hot junction.

The thermal boundary layer is shown to be pushed away from the wall during rapid initial bubble growth. A subsequent heating of this layer occurs during the waiting time between the detachment of a bubble and the generation of the succeeding bubble. The dew point of vapour in the bubble space is uniform and approximates the saturation temperature at ambient (atmospheric) pressure.

The mechanism results in large local temperature fluctuations in pure liquids. In contradistinction to this behaviour, in binary mixtures containing a small fraction of the more volatile component, the temperature dips, which are observed by passage of the hot junction from the superheated liquid into the vapour space, are limited to considerably lower values. Obviously, this phenomenon proves the existence of a decreased "effective superheating", which is caused by an increase of the dew point in comparison with the boiling point of the liquid of the original composition.

NOMENCLATURE

a ,	$k/\rho_1 c$, liquid thermal diffusivity [m^2/s];	D_e ,	external diameter of thermocouple [μm or m];
c ,	liquid specific heat at constant pressure [$\text{J}/\text{kg } ^\circ\text{C}$];	D_w ,	diameter of heating wire [μm or m];
C_1 ,	$= R/\vartheta_0 t^{\frac{1}{2}}$, bubble growth constant [$\text{m}/\text{s}^{\frac{1}{2}} ^\circ\text{C}$];	G_d ,	$= (a/D)^{\frac{1}{2}}(C_{1,m}/C_{1,p})(c/l)\vartheta_0$, vaporized mass diffusion fraction for individual bubble;
$d_{o,p}$,	$= (12/\pi)^{\frac{1}{2}}(at_1)^{\frac{1}{2}}$, initial (maximal) thickness of relaxation microlayer in pure liquid [μm or m];	Ja ,	$= (\rho_1 c/\rho_2 l)\vartheta_0$, Jakob number for pure liquid;
$d_{w,co}$,	thickness of equivalent thermal conduction layer for convection [μm or m];	Ja_0 ,	$= (C_{1,m}/C_{1,p})Ja = (\rho_1 c/\rho_2 l)(\vartheta_0 - \Delta T)$, modified Jakob number for binary mixture;
D ,	mass diffusivity of more volatile component in less volatile component [m^2/s];	k ,	liquid thermal conductivity [$\text{W}/\text{m } ^\circ\text{C}$];
		l ,	latent heat of vaporization [J/kg];
		Δp ,	excess pressure in expanding spherical cavity [$\text{kg}/\text{m s}^2$];
		q_w ,	heat flux density at heating surface [W/m^2];

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$q_{w, bi}$	difference between total heat flux density at surface and convective contribution [W/m^2];	ϑ_0	initially uniform liquid superheating (for free bubble) or average superheating of heating surface [$^{\circ}\text{C}$];
$q_{w, \max}$	peak flux density in nucleate boiling [W/m^2];	$\vartheta_0 - \Delta T$	$= (C_{1,m}/C_{1,p})\vartheta_0$, effective uniform liquid superheating for free bubble in binary mixture [$^{\circ}\text{C}$];
R	instantaneous (equivalent) bubble radius [m];	$\delta\vartheta_0$	local liquid superheating at distance z above heating wire with $q_w = 0$ [$^{\circ}\text{C}$];
R_1	$= R(t_1)$, bubble departure radius [m];	$\Delta\vartheta_0$	local liquid superheating at distance z above heating wire with convective heat flux density [$^{\circ}\text{C}$];
t	time [ms or s];	ν	frequency of bubble formation on nucleus [$1/\text{s}$];
Δt	time interval [ms or s];	ρ_1	liquid density [kg/m^3];
t_1	bubble departure instant [ms or s];	ρ_2	saturated vapour density [kg/m^3];
T	boiling point of pure liquid [$^{\circ}\text{K}$];	τ	response time of thermocouple [ms or s];
$T(R, t)$	instantaneous temperature at bubble boundary [$^{\circ}\text{K}$];	Subscripts	
$T(x)$	boiling temperature of liquid at bubble boundary in binary mixture [$^{\circ}\text{K}$];	m ,	value in binary mixture;
$T(x_0)$	boiling temperature of original liquid in binary mixture [$^{\circ}\text{K}$];	p ,	value in pure liquid.
$T(y)$	$= T(x)$, dew temperature of saturated vapour in binary mixture [$^{\circ}\text{K}$];	Numerical values for water at atmospheric boiling point	
ΔT	$= T(y) - T(x_0)$, temperature difference between dew temperature of vapour in bubble and boiling temperature of original liquid in binary mixture [$^{\circ}\text{C}$];	a ,	$= 16.9 \times 10^{-8} \text{ m}^2/\text{s}$;
x	mass fraction of more volatile component in liquid at bubble boundary in binary mixture;	c ,	$= 4216 \text{ J}/\text{kg } ^{\circ}\text{C}$;
x_0	mass fraction of more volatile component in original liquid in binary mixture;	$C_{1,p}$,	$= 24 \times 10^{-4} \text{ m}/\text{s}^{\frac{1}{2}} ^{\circ}\text{C}$;
y	mass fraction of more volatile component in vapour of binary mixture;	Ja ,	$= 2.995 \vartheta_0$;
z	distance between hot junction of thermocouple and upper side of heating wire [μm or m].	k ,	$= 0.6825 \text{ W}/\text{m } ^{\circ}\text{C}$;
Greek symbols		l ,	$= 22.56 \times 10^5 \text{ J}/\text{kg}$;
$\vartheta(t)$	instantaneous uniform superheating of relaxation microlayer [$^{\circ}\text{C}$];	ρ_1 ,	$= 958.4 \text{ kg}/\text{m}^3$;
$\vartheta(z, t)$	instantaneous local superheating recorded with thermocouple [$^{\circ}\text{C}$];	ρ_2 ,	$= 0.598 \text{ kg}/\text{m}^3$.
		Numerical values for water-2-butanone ($x_0 = 4.1 \times 10^{-2}$) at atmospheric boiling point ($362 ^{\circ}\text{K}$)	
		$(a/D)^{\frac{1}{2}}$,	$= 13.2$;
		$C_{1,m}$,	$= 6 \times 10^{-4} \text{ m}/\text{s}^{\frac{1}{2}} ^{\circ}\text{C}$;
		D ,	$= 9.7 \times 10^{-10} \text{ m}^2/\text{s}$;
		Ja_0 ,	$= 0.749 \vartheta_0$;
		ΔT ,	$= 0.75 \vartheta_0$;
		$\Delta T/G_a$,	$= 180 ^{\circ}\text{C}$.
		THE SCOPE of the present investigation is to verify directly, by using a suitably chosen experimental technique, the validity of the hypotheses,	

which form the common physical background of the various bubble growth theories, cf. [1,2]. Also, the basis of the Van Stralen "relaxation microlayer" model theory [3-5] for the mechanism of nucleate boiling will be checked.

During the course of this investigation, temperature fluctuations in boiling or subcooled water have been reported by Bonnet, Macke, Morin and Salomon [6] and by Séméria and Flamand [7]. Some results of the present study are in good agreement with those obtained by these workers. The other information described here, was hitherto unknown in the literature.

1. THEORETICAL BUBBLE GROWTH

1.1. Free bubbles

A more comprehensive survey of the various theories concerning the growth rate of free, spherically symmetric growing vapour bubbles in initially uniformly superheated pure liquids and binary mixtures has been published recently [1, 2]. Only the most essential feature of the physical model and some important conclusions are discussed here.

Isothermal motion. An asymptotic approximation for the radius of an expanding spherical cavity with constant excess pressure Δp in a nonviscous incompressible liquid follows from the dynamic Rayleigh equation of motion [8]: $R \cong (2\Delta p/3\rho_1)^{1/2}t$. The Rayleigh equation can be derived by differentiation of the energy equation in combination with continuity.

Pure liquids. According to Foster and Zuber [9], and Plesset and Zwick [10], an expression for the growth rate of a vapour bubble follows by extending the right-hand side of the Rayleigh equation with a surface tension term and an evaporation term. The driving pressure term is transformed into a superheating term by using the Clausius-Clapeyron equation. The dynamic and surface tension terms are negligible after an initial stage of 0.1-1 ms, whence the remaining time-independent superheating term is balanced by the evaporation term; i.e. asymptotic bubble growth is no longer governed by hydrodynamics, but isobaric and determined only by heat diffu-

sion towards the bubble boundary to satisfy the requirement of energy necessary for evaporation. Thermodynamic equilibrium at the bubble boundary is postulated, and the vapour temperature, which will be uniform due to the large thermal diffusivity of vapour, is shown to approximate the saturation value at ambient pressure. This is in accordance with Prüger's experimental results [11] at a vapour-liquid interface during stationary evaporation of a superheated liquid without ebullition.

The resulting asymptotic approximation for the bubble radius:

$$R_p(t) \cong \left(\frac{12}{\pi}\right)^{1/2} \frac{(k\rho_1 c)^{1/2}}{\rho_2^{1/2}} \vartheta_0 t^{1/2} = \left(\frac{12}{\pi}\right)^{1/2} \times Ja(at)^{1/2} = C_{1,p} \vartheta_0 t^{1/2} \quad (1)$$

was also derived by Scriven [12] by extending the heat conduction equation for spherical symmetry to establish the effect of radial convection resulting from unequal phase densities. The boundary and initial conditions are determined by taking initially uniform superheating and from $T(R, t) = T$. Obviously, the various theories have the same physical background.

Binary mixtures. Bubble growth in superheated binary mixtures is slowed down due to the analogous mass diffusion of the more volatile component, according to Van Wijk, Vos and Van Stralen [13], Scriven [12], Bruijn [14], Van Stralen [15], and Skinner and Bankoff [16]. Van Stralen's modification [1, 2, 15] shows the physical equivalence of the various theories: the dew point of the vapour in the bubble space is increased with an amount ΔT with respect to the boiling temperature of the original liquid.

As a consequence, the occurrence of a minimal bubble growth rate (corresponding with a maximal value of $\Delta T/G_d$) has been predicted at a certain low concentration of the more volatile component, especially in case $D \ll a$. This composition can be deduced from equilibrium data only.

The asymptotic approximation for the bubble radius:

$$\begin{aligned}
 R_m(t) &\cong \left(\frac{12}{\pi}\right)^{\frac{1}{2}} J a_0(at)^{\frac{1}{2}} = C_{1,m} \vartheta_0 t^{\frac{1}{2}} \\
 &= C_{1,p} (\vartheta_0 - \Delta T)t^{\frac{1}{2}} = \left(\frac{12}{\pi}\right)^{\frac{1}{2}} \\
 &\quad \times \frac{a^{\frac{1}{2}}}{(\rho_2/\rho_1)\{l/c + (a/D)^{\frac{1}{2}}\Delta T/G_d\}} \vartheta_0 t^{\frac{1}{2}} \quad (2)
 \end{aligned}$$

is obviously a generalization of equation (1) and based on the introduction of a modified Jakob number

$$J a_0 = \frac{\vartheta_0 - \Delta T}{\vartheta_0} J a = \frac{\rho_2 c}{\rho_2 c} (\vartheta_0 - \Delta T). \quad (3)$$

The "effective superheating" is related to ϑ_0 as follows:

$$\begin{aligned}
 \vartheta_0 - \Delta T &= \vartheta_0 - \{T(x) - T(x_0)\} \\
 &= \vartheta_0 - \{T(y) - T(x_0)\} = \{T(x_0) + \vartheta_0\} \\
 &\quad - T(y) = \frac{C_{1,m}}{C_{1,p}} \vartheta_0 \quad (4)
 \end{aligned}$$

e.g. for the mixture with $x_0 = 4.1 \times 10^{-2}$ 2-butanone (methyl ethyl ketone) in water $C_{1,m} = 0.25 C_{1,p}$, cf. [1-5], whence $\vartheta_0 - \Delta T = 0.25 \vartheta_0$.

1.2. Adhering bubbles in nucleate boiling

The van Stralen "relaxation microlayer" theory [3-5] for the mechanism of nucleate boiling is essentially based on a periodic local removal of the superheated convective thermal boundary layer due to the initially rapid growth of succeeding vapour bubbles, which are generated at active nuclei on the heating surface. The initial thickness of the relaxation microlayer $d_{o,p} = (12/\pi)^{\frac{1}{2}}(at_1)^{\frac{1}{2}}$ in water amounts to 60 μm , and differs only slightly from the value of the equivalent conduction layer $d_{w,co} \approx 50 \mu\text{m}$ in convection [3-5]. The relaxation microlayer, the superheating of which is assumed to be uniform, is cooled during the bubble growth time according to:

$$\vartheta(t) = \vartheta_0 \exp - (t/t_1)^{\frac{1}{2}}.$$

The layer is superheated again by heat transmission at the wall during the waiting time between succeeding bubbles. The transient heat-

conduction formalism for semi-infinite bodies is used to describe this process. The heat flux density at the wall is shown to depend mainly on the thermal properties of the liquid, cf. [3-5].

1.3. Local temperature fluctuations

Thermal boundary layer. Apparently, the relaxation microlayer theory predicts the occurrence of relatively large temperature fluctuations in the local superheating $\vartheta(z, t)$ of the thermal boundary layer at the heating surface. A maximal amplitude $\vartheta_0(1 - 1/e) = 0.63 \vartheta_0$ can be expected.

Contrarily, the Moore and Mesler "evaporation microlayer" model [17] does not involve the occurrence of any fluctuations of the local liquid temperature at all, as the observed sudden temperature dips at the the contact area between wall and boiling liquid are ascribed to thermal properties of the heating material only. The thickness of the hypothetical evaporation microlayer does not exceed $(c\vartheta_0/1)d_{o,p} \approx 2 \mu\text{m}$.

Fixed thermocouple. Eventual local fluctuations in $\vartheta(z, t)$ can be measured directly using a thin-wired thermocouple with the hot junction placed in the thermal boundary liquid layer at the heating wall.

A direct consequence of the theoretical model, cf. equations (1, 2, 4), is that such a small thermocouple shows a sudden temperature dip if the hot junction is instantaneously covered with saturated vapour instead of superheated liquid.

The amplitude should equal ϑ_0 in case of a free bubble in a pure liquid. Contrarily, in 4.1 wt % 2-butanone, this amplitude should be limited to a much smaller value i.e. $0.25 \vartheta_0$. For adhering or released bubbles generated in nucleate boiling, correspondingly lower amplitudes, but with the same ratio of 0.25, are predicted, cf. equation (5).

2. APPARATUS AND EXPERIMENTAL PROCEDURE

The experimental setup of the boiling vessel and the high speed motion picture framing camera is shown in Fig. 1. A comprehensive description of the photographic technique has been presented previously [2, 18, 19].

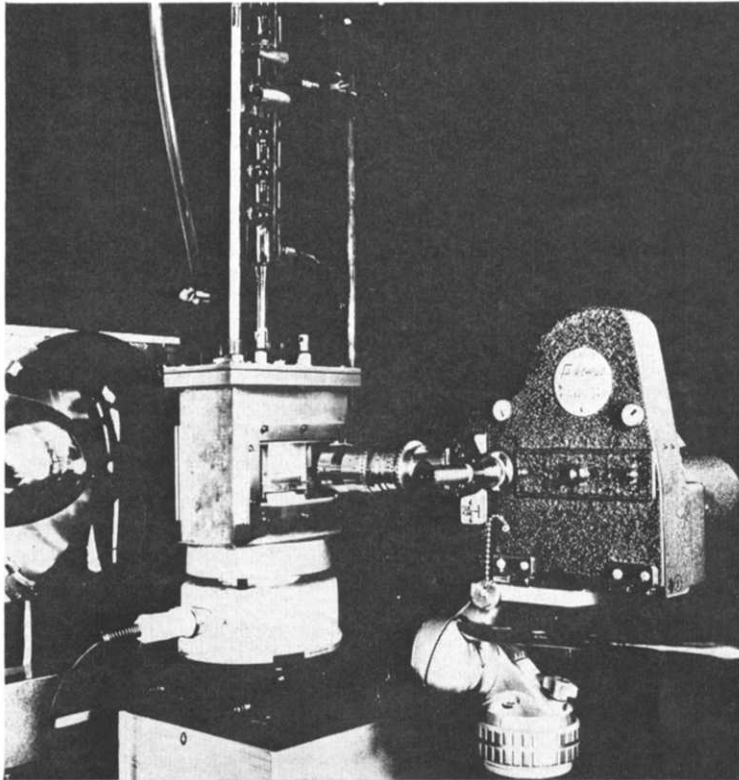


FIG. 1. Experimental setup of boiling vessel, high speed motion picture camera and light source for background lighting.

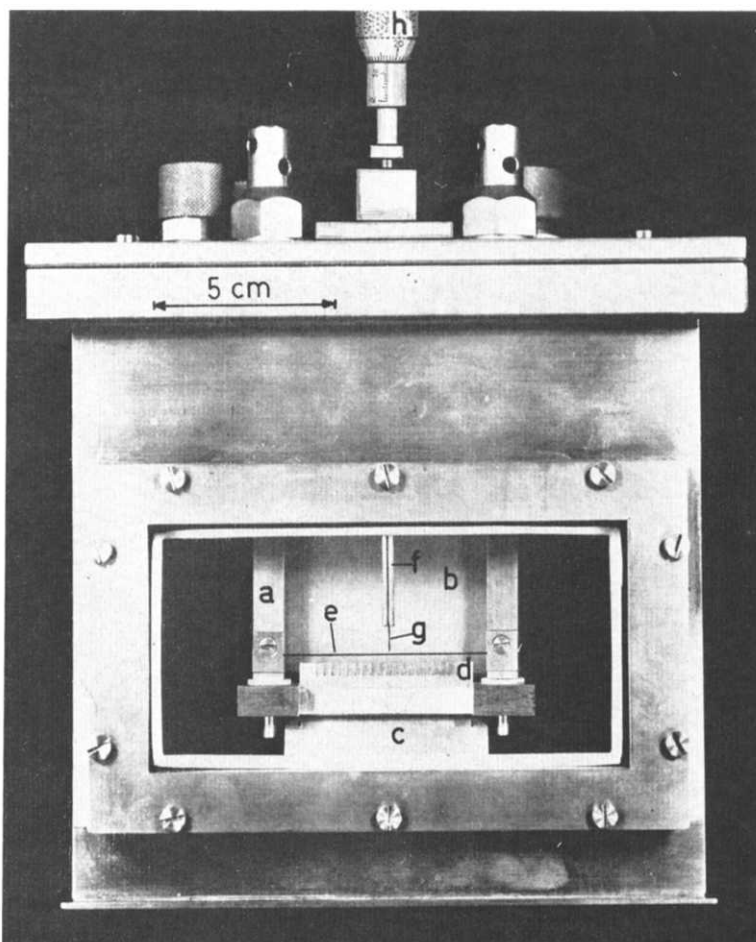


FIG. 2. Boiling vessel provided with thermocouple and screw micrometer. A counter balancing spring and several other accessories have been omitted.

a: nickel-coated brass electrode;

b: ground glass;

c: screen to keep ascending bubbles from bottom plate out of vision;

d: 2-mm scale for evaluation of bubble dimensions;

e: platinum heating wire;

f: glass capillary

g: thermocouple;

h: screw micrometer.

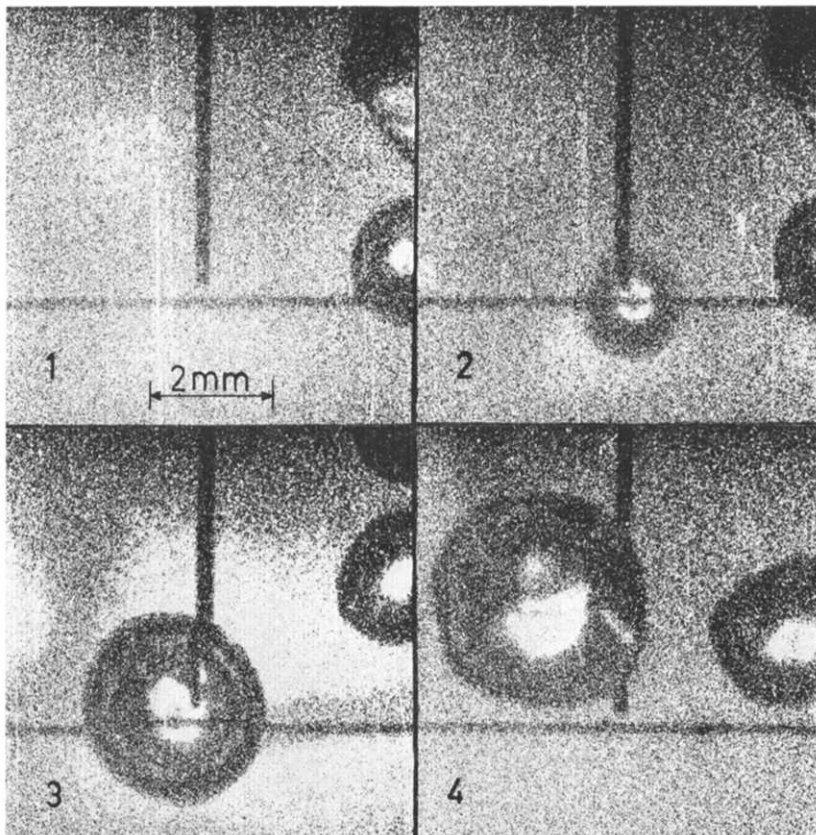


FIG. 5. *Water*. Photographs taken from simultaneous high-speed motion picture for distance $z=100\ \mu\text{m}$. Numbers of photographs correspond with instants indicated in Fig 4.

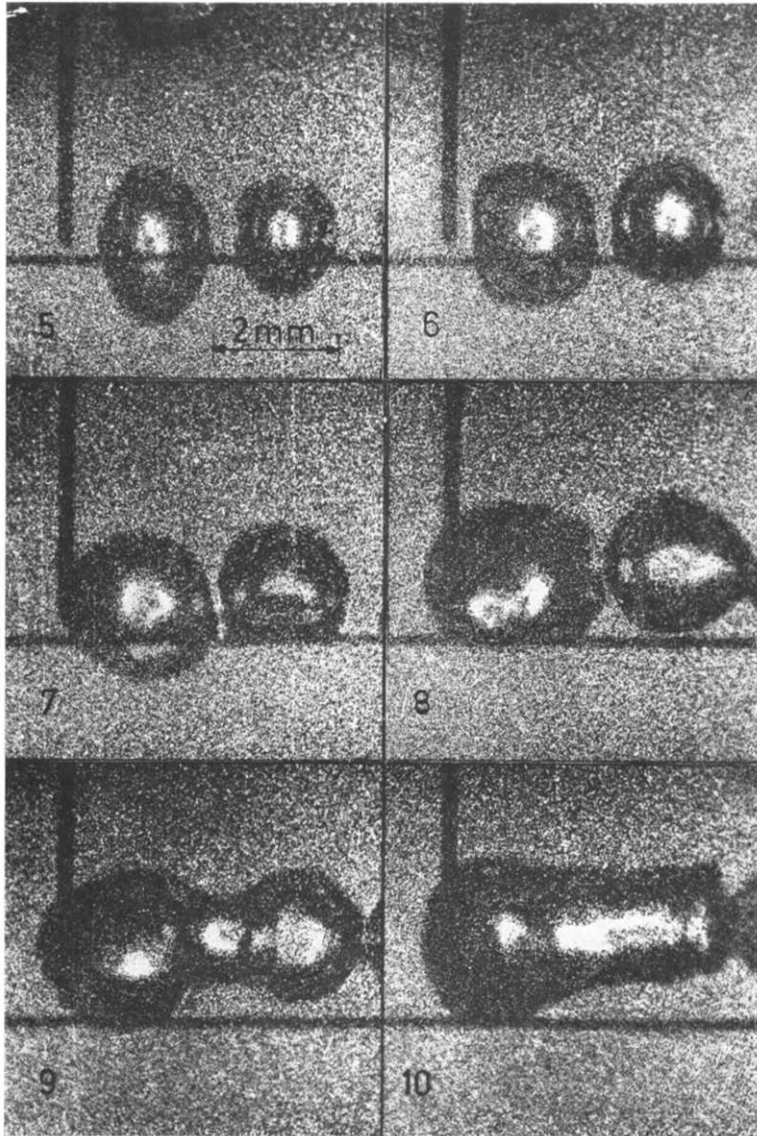


FIG. 6. *Water*. Photographs taken from simultaneous high-speed motion picture for distance $z=100\ \mu\text{m}$. Numbers of photographs correspond with instants indicated in Fig. 4.

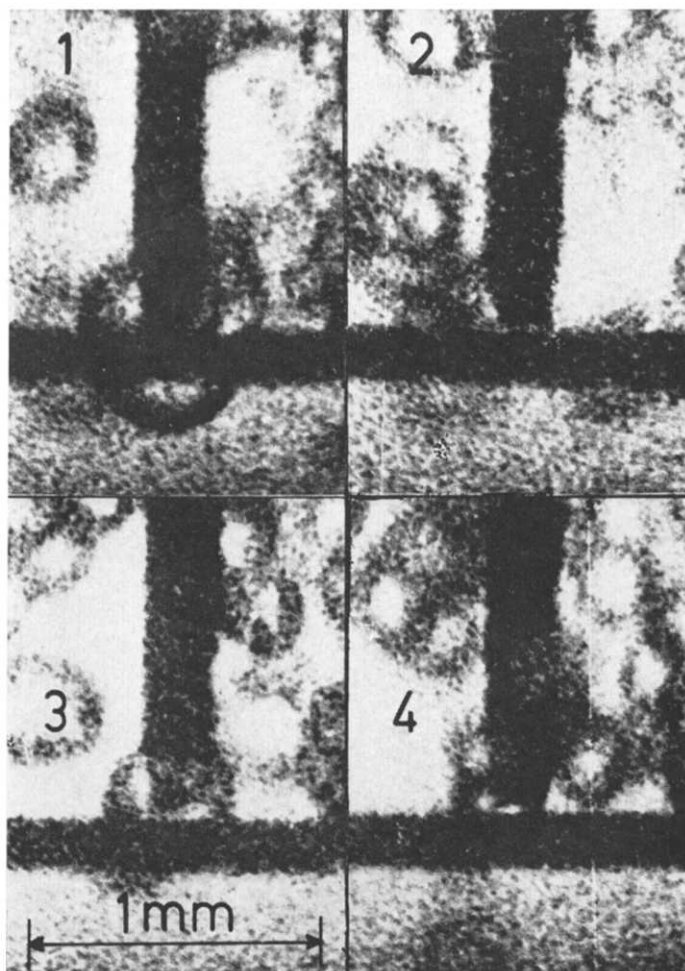


FIG. 8. *Water-2-butanone* ($x_0 = 4.1 \times 10^{-2}$). Photographs taken from simultaneous high-speed motion picture for distance $z = 30 \mu\text{m}$. Numbers of photographs correspond with instants indicated in Fig. 7.

A Thermocoax chromel–alumel thermocouple (external diameter of the SS 304 sheath: $D_t = 250 \mu\text{m}$, wire diameter: $50 \mu\text{m}$) with TM hot junction is carried vertically through the cover plate of the vessel by using a polytetrafluoroethylene (Teflon) insulation (Fig. 2).

The response time (or time constant) τ of this thermocouple for a step change in temperature amounts to 1.7 ms ,* τ is proportional to $D_t^{\frac{3}{2}}$. This theoretical value, which can easily be derived from the Newton cooling law, is in good agreement with the experimental value following from data obtained in the laboratories of Sodern, the manufacturer. The condition $\tau < t_1$ is satisfied, cf. [3].

For a sinusoidal variation of the liquid temperature, the thermocouple temperature oscillates around the same average value. For the investigated bubbles, generated at a relatively low frequency ν (water: large bubbles with departure radius $R_{1,p} = 15 \times 10^{-4} \text{ m}$, $\nu = 15 \text{ s}^{-1}$, cf. figures 4–6; 4.1 wt % 2-butanone: $R_{1,m} = 2.5 \times 10^{-4} \text{ m}$, $\nu = 30 \text{ s}^{-1}$, cf. Figs. 7 and 8, the peaks of the thermocouple lag the peaks of the liquid temperature by a time interval [20]:

$$\frac{\arctan 2\pi\nu\tau}{2\pi\nu}, \quad (6)$$

which differs only slightly (3%) from 1.7 ms .

The damping of the amplitude is given by a factor of:

$$\frac{1}{\{1 + (2\pi\nu\tau)^2\}^{\frac{1}{2}}}, \quad (7)$$

resulting in a decrease of only 2 per cent in water and 10 per cent in the binary mixture. These values may still be lowered considerably by diminishing the diameter of the thermocouple, cf. Section 3.3. For the purpose of the present investigation, however, the thermocouple is sufficiently small, cf. Sections 1.3 and 3.1. The heat-conduction error of the thermocouple is left out of consideration.

The distance z of the hot junction of the thermocouple to the surface of the electrically heated horizontal platinum wire (diameter $D_w = 200 \mu\text{m}$), which acted as a heating surface, could be adjusted accurately to $2 \mu\text{m}$ by using a special screw micrometer design. Mechanical contact ($z = 0$) between this junction and the upper side of the heating wire could be detected electrically. The construction permits small horizontal displacements of the thermocouple in two orthogonal directions, one of which is parallel to the axis of the heating wire. In this way, the hot junction could be placed arbitrarily close to an active nucleus on the wire, generating vapour bubbles.

The camera is provided with two neon bulbs, one for time marking and the other one for event marking. Two pulse generators (1000 Hz, $30 \mu\text{s}$) were used. The pulses of one generator were supplied to the event mark entry on the camera, and simultaneously, via power amplification, to a separate 8000 Hz light-beam galvanometer channel on a Visicorder. A instantaneous switching on of this circuit at a previously fixed instant ensures an exact synchronization of simultaneous phenomena recorded both on the motion picture (taken at a final rate of 6000 frames per second) and on the oscilloscript paper. Apparently, the latter must be corrected for the time lag due to the response of the thermocouple.

The cold junction of the thermocouple was immersed in melting ice. Approximately 90–95 per cent of the e.m.f. of the thermocouple to the saturation temperature of the liquid was compensated by a time-independent negative voltage taken from a mercury battery. Voltages were measured with a digital voltmeter, accurately to $1 \mu\text{V}$, which corresponds to 0.025°C . The remaining signal was amplified a factor of 2000 on two Accudata III cascaded stabilized amplifiers with differential input.

The data were recorded with a 1650 Hz light-beam galvanometer channel. Only shielded coaxial cables were used, resulting in a negligible electrical phase shift (approximately

* For a velocity $\dot{R}_p = 1 \text{ m/s}$ in water; 3.4 ms for $\dot{R}_m = \frac{1}{2}\dot{R}_p$ in 4.1 wt % 2-butanone; $170 \text{ ms} = 100 \tau_p$ in water vapour, cf. [20].

0.02 rad) in the thermocouple circuit. The bottom plate of the boiling vessel was heated from below by means of a Bunsen burner to avoid electromagnetic induction. The adjustable power of the heating wire was supplied by a storage-battery for the same reason. The final noise recorded on the Visicorder could thus be restricted to 0.25°C . The chopper frequency of the amplifiers (380 Hz) is visible in the experimental temperature curves (Figs. 3, 4 and 7).

caused by the response of the thermocouple has been established in evaluation of the instants of the photographs in Figs. 5, 6 and 8. No correction has been made on the amplitude of the observed local temperature fluctuations due to the heat-conduction error of the thermocouple, cf. also equation (9). The distances $z = 3 \mu\text{m}$, $30 \mu\text{m}$ and $100 \mu\text{m}$ in nucleate boiling have been chosen in consideration of the initial thickness $d_{o,p} = 60 \mu\text{m}$ of the relaxation microlayer in water, cf. Section 1.2.

3. EXPERIMENTAL RESULTS

The time lag of approximately 1.7–3.4 ms

3.1. Water

Convection. Local temperature fluctuations at

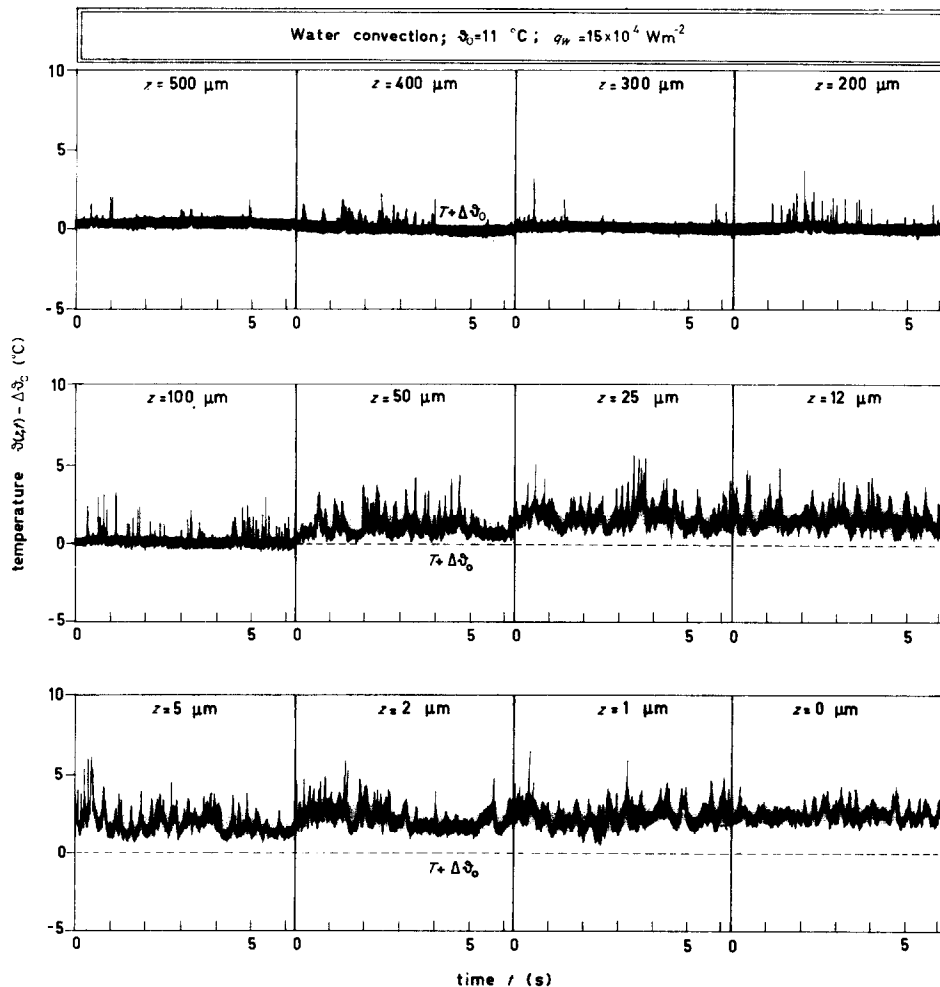


FIG. 3. Water. Local temperature fluctuations in convection at various distances of hot junction of thermocouple to upper side of heating wire ($z = 500\text{--}0 \mu\text{m}$).

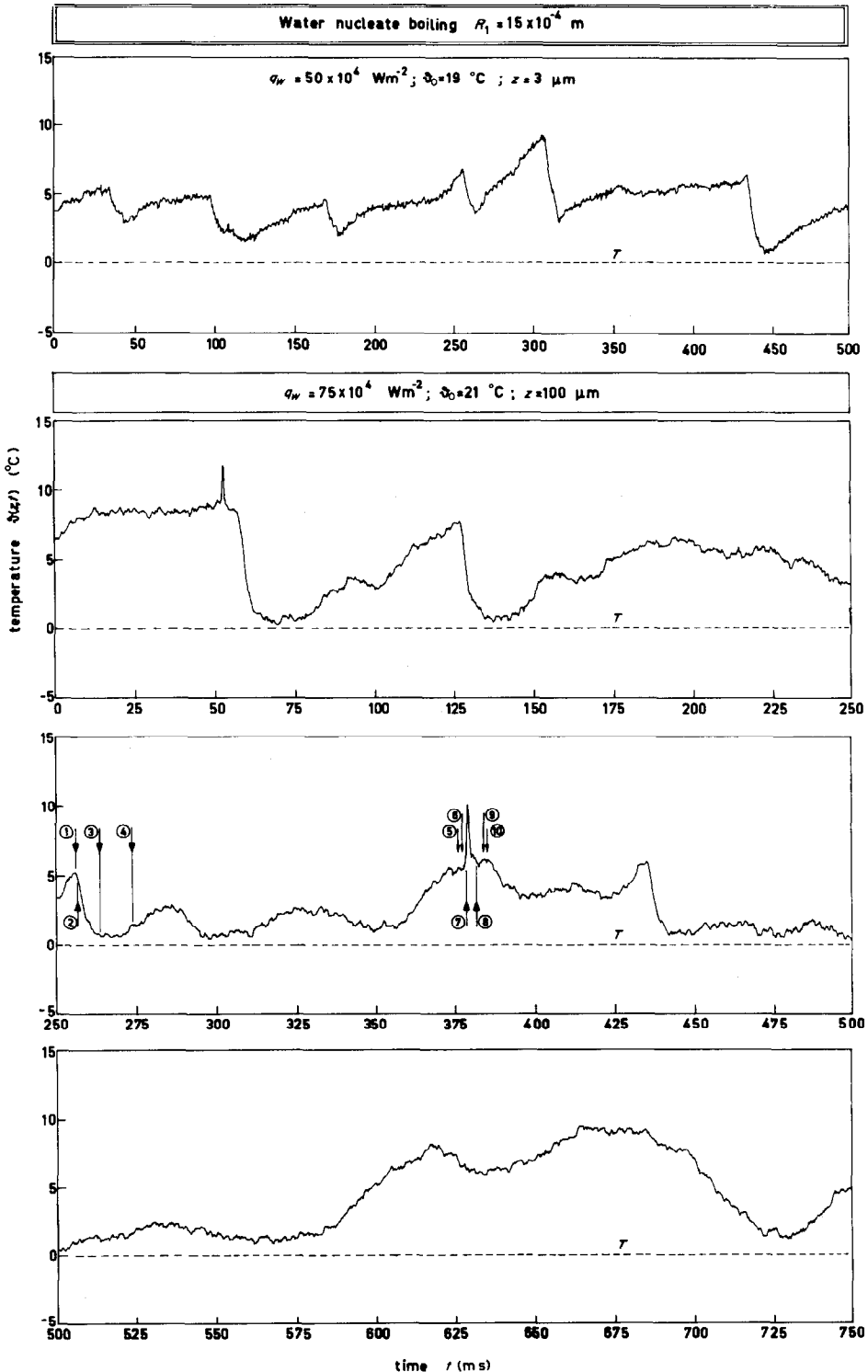


FIG. 4. Water. Local temperature fluctuations in nucleate boiling at two different distances of hot junction of thermocouple to upper side of heating wire ($z = 3$ μ and 100 μm). Cf. Figs. 5 and 6.

the rear of the heating wire up to 3 degC occur in convection without ebullition on the wire (Fig. 3). The convection is here a combination of natural and forced convection, as slightly superheated liquid (at local temperature $T + \delta\vartheta_0$), due to convection currents ascending from the externally heated bottom plate of the vessel, is flowing normally to the horizontal heating wire ($q_w = 15 \times 10^4 \text{ Wm}^{-2}$) with a velocity of approximately $5 \times 10^{-2} \text{ m/s}$.

The distance z between the hot junction of the thermocouple and the upper tangent plane to the wire of circular cross-section is varied from 500–0 μm . Only slow temperature fluctuations with periods of the order of 0.1–1 s and amplitudes up to 2°C are observed. Both the average amplitude and frequency increase with diminishing distance. These fluctuations are due to eddies at the rear of the wire, which travel, apparently, over a distance of approximately $5 \times 10^{-2}/1 = 5 \times 10^{-2} \text{ m}$ for a period of 1 s.

The average temperature ($T + \Delta\vartheta_0$ for $z = 500 \mu\text{m}$, i.e. outside the equivalent conduction layer at the wire) should be expected to increase up to $T + \vartheta_0$ for $z = 0$. Actually, the measured value is considerably lower, which may be due to the following causes: (a) the heat-conduction error, (b) only a small central area of the hot junction touches the upper side of the heating wire, and (c) possibly, the plane through the centre of the hot junction and the axis of the wire deviates from the vertical direction.

Nucleate boiling. Local temperature fluctuations up to 11 degC occur both for $z = 3 \mu\text{m}$ and 100 μm in nucleate boiling on the heating wire (Fig. 4). The concerning nucleus generates relatively large bubbles ($R_{1,p} = 15 \times 10^{-4} \text{ m}$). The heat flux density ($q_w = 80 \times 10^4 \text{ Wm}^{-2}$) of the wire for $z = 100 \mu\text{m}$ amounted to 95 per cent of the peak value.

The most important other experimental data are:

- (i) The frequency of the fluctuations is

determined by the behaviour of nearby vapour bubbles.

- (ii) An instantaneous temperature decrease (up to 8 degC in 6 ms, i.e. 3.5 times the response time of the thermocouple) occurs before the hot junction is suddenly covered with vapour, which is replacing the preceding superheated liquid.
- (iii) Generally, the dew temperature of vapour in the bubble space is uniform and approximates the saturation value at ambient (atmospheric) pressure.
- (iv) A continuous heating of stagnant liquid is observed during the waiting time between succeeding bubbles.
- (v) During adherence, a growing vapour bubble is pushing a surrounding superheated "relaxation microlayer" away from the wall.

Discussion. A more comprehensive description and discussion of experimental data is following now:

(A) Figure 5 gives a demonstration of the results (i–iv). Photograph:

- No. 1: hot junction of thermocouple is immersed in superheated liquid;
- No. 2: hot junction covered with vapour, preceding temperature decreases very rapidly with a rate of approximately 100°C s^{-1} ;
- No. 3: vapour temperature approximates the saturation value. The actually measured temperature will err on the high side now, as the thermocouple leads are acting as a local heat source in this case. The total temperature decrease amounts to $\vartheta(z, t)$;
- No. 4: hot junction covered with liquid, which is gradually warmed up.

(B) Figure 6 demonstrates the results (iv) and (v). Photograph:

- No. 5: vapour bubble is generated on nucleus at some distance to hot junction;

- No. 6: boundary of this growing bubble is moving in close proximity to hot junction;
- No. 7: hot junction is passed by superheated boundary layer surrounding moving bubble interface. This causes the occurrence of an instantaneous peak in the local temperature;
- No. 8: hot junction instantaneously in vapour causes a preceding decrease in temperature;
- No. 9: hot junction instantaneously in superheated liquid causes a gradual increase in temperature;
- No. 10: hot junction in vapour again is accompanied by a simultaneous preceding decrease in temperature.

(C) The gradual heating, cf. result (iv), during $t = 585\text{--}618$ ms ($\Delta t = 33$ ms) for $z = 100$ μm of the liquid layer invested between the bottom of the thermocouple and the upper side of the heating wire will be approximated here by taking the layer to be stagnant (Fig. 4). The solution of the transient heat-conduction equation for a semi-infinite liquid body (initially at temperature T) with the plane boundary $z = 0$ kept suddenly (at $t = 0$) at constant temperature $T + \vartheta_0$ [21]:

$$\vartheta(z, t) = \vartheta_0 \operatorname{erfc} \frac{z}{2(at)^{\frac{1}{2}}} = \vartheta_0 \left\{ 1 - \operatorname{erf} \frac{z}{2(at)^{\frac{1}{2}}} \right\} \quad (8)$$

yields: $\vartheta(10^{-4}, 33 \times 10^{-3}) = 0.33 \vartheta_0 = 6.9$ degC. This value exceeds the experimental value of 6.6°C only slightly. Apparently, the heat-conduction error of the thermocouple, which errs on the low side now, is nearly negligible, cf. also Section 2.

The average local heat flux density at the heating surface amounts to (cf. Part II of [3]):

$$q_{w,bi} = \frac{2k\vartheta_0}{(\pi a \Delta t)^{\frac{1}{2}}}, \quad (9)$$

whence $q_{w,bi} = 35 \times 10^{-4}$ Wm^{-2} , i.e. 44 per cent of the nucleate boiling peak flux density. This value is also in good agreement with van Stralen's relaxation microlayer theory, which predicts a ratio of $0.88/1.88 = 0.47$, cf. [3-5].

The fundamental assumption of the validity of the formalism for transient heat conduction during the waiting time, is thus also justified.

The local superheating decreases during $t = 618\text{--}635$ ms owing to the presence of a vapour bubble, which is generated on a nucleus at some distance to the hot junction of the thermocouple.

3.2. Water-2-butanone ($x_0 = 4.1 \times 10^{-2}$)

Convection. Figure 7 shows temperature fluctuations measured at $z = 30$ μm both for convection and nucleate boiling. In case of $\vartheta_0 = 0$, $q_w = 0$, whence a time-independent local liquid temperature $T(x_0) + \delta\vartheta_0$ is recorded. The (slight) local superheating $\delta\vartheta_0$, which amounts to $0.1\text{--}1$ degC, is caused only by convection currents ascending from the heated bottom plate of the vessel. Apparently, in this case the value of the Reynolds number (35) is too low to generate eddies at the rear of the wire.

The pattern of the slow temperature fluctuations with a maximal amplitude at 1.5 degC occurring in convection are similar to those in water, as should be expected.

Nucleate boiling. The theoretical temperature jump in this mixture, which is caused by passing the hot junction from superheated liquid into saturated vapour, amounts to only 25 per cent of the corresponding value in water, cf. Sections 1.1-1.3 and equation (7).

Actually, local temperature fluctuations up to only 1.5°C are recorded in nucleate boiling at a heat flux density, which amounts to 45 per cent of the peak value (Fig. 7). The bubble radius at departure ($R_{1,m} = 2.5 \times 10^{-4}$ m) amounts to 17 per cent of the value for the investigated bubbles in water.

The observed temperature jump, which is recorded by passing the hot junction of the thermocouple from superheated liquid into vapour, amounts to only 1 degC, in fair agreement with the theoretical prediction.

Photographs. Figure 8 demonstrates this extremely important result. Photograph:

No. 1: hot junction of thermocouple in vapour with dew temperature $T(y) = T(x) =$

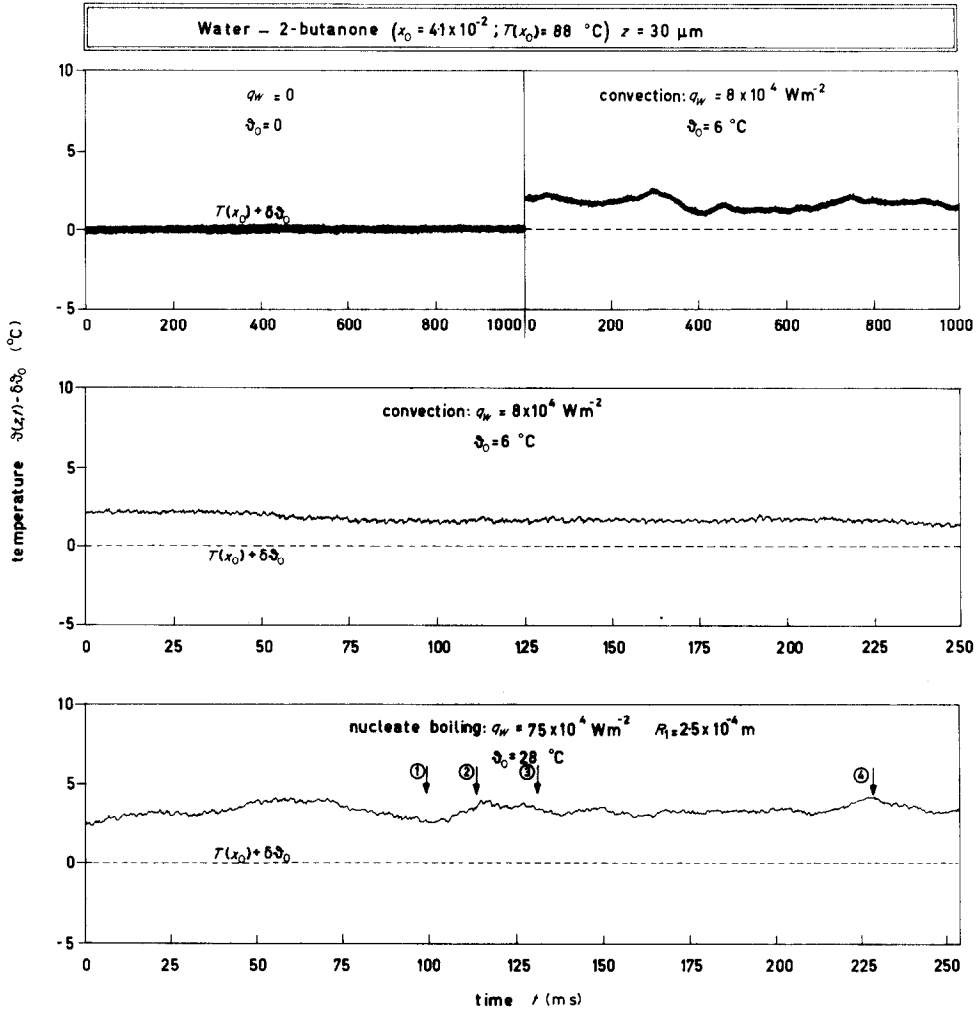


FIG. 7. Water-2-butanone ($x_0 = 4.1 \times 10^{-2}$). Local temperature fluctuations at rear of heating wire, for $q_w = 0$ and in convection, and for nucleate boiling, respectively ($z = 30 \mu\text{m}$), cf. Fig. 8.

$T(x_0) + \Delta T$ above $\delta\vartheta_0$, assuming thermodynamic equilibrium at bubble interface. The temperature has been decreased with an amount $\vartheta(z, t) - \Delta T = 0.25 \vartheta(z, t)$;

- No. 2: hot junction is superheated liquid at temperature $T(x_0) + \vartheta(z, t)$ above $\delta\vartheta_0$;
- No. 3: hot junction in vapour again;
- No. 4: small bubble occurring at bottom of thermocouple causes again a small temperature decrease of $0.25 \vartheta(z, t)$.

3.3. Thermocouple with fast response time

Some additional experiments have been carried out with a thin-wired (70 μm) thermocouple with a hot junction diameter of 70 μm . The sheath around the lower part of the thermocouple had been removed. The response time of this thermocouple amounts to only 0.25 ms. Essentially, the results were similar to those obtained with the 2 ms-thermocouple. A steeper negative slope of the $\vartheta(z, t)$ -curve for constant z occurred by passing the hot junction suddenly

from superheated water into vapour: up to 3.5 degC/ms instead of 1.3 degC/ms, cf. Section 3.1.

4. CONCLUSIONS

Water. The results (ii) and (iii) of Section 3.1. justify the common hypotheses, on which the various bubble growth theories for pure liquids are based, cf. Sections 1.1 and 1.3. The remaining results are in good agreement with the physical background of van Stralen's relaxation micro-layer model for the mechanism of nucleate boiling, cf. Sections 1.2. and 1.3. The temperature fluctuations occurring in and nearby the thermal boundary layer at the wall are in favour with this theory, and in contradistinction to the Moore and Mesler [17] model.

4.1 wt % 2-butanone. Obviously, the occurrence of an increase ΔT in the dew point of vapour is shown to exist, i.e., the fundamentals of the various bubble growth theories for binary mixtures are justified experimentally.

The sudden temperature dips occurring exactly before the hot junction is passed into the bubble space, must be attributed to the liquid, and not to the vapour on account of the 100 times higher response time in the latter, cf. footnote in Section 2.

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Résumé—On donne une vérification expérimentale directe de l'arrière-plan physique général qui est à la base des différentes théories de croissance de bulle et la théorie de Van Stralen de la "microcouche de relaxation" pour le mécanisme de l'ébullition nucléée.

Dans ce but, les mesures de température locale instantanée avec un thermocouple à fils fins ont été synchronisées avec des films à grande vitesse de bulles de vapeur engendrées sur des germes actifs très voisins de la jonction chaude.

On montre que la couche limite thermique est repoussée loin de la paroi pendant la croissance initiale rapide de la bulle. Un chauffage postérieur de cette couche se produit pendant le temps d'attente entre le détachement d'une bulle et la naissance de la bulle suivante. Le point de rosée de la vapeur à l'intérieur de la bulle est uniforme et très voisin de la température de saturation à la pression ambiante (atmosphérique).

Ce mécanisme aboutit à de grandes fluctuations de la température locale dans les liquides purs. En opposition avec ce comportement, les baisses de température dans les mélanges binaires contenant une faible fraction du composant le plus volatil, qui sont observées lorsqu'on fait passer la jonction chaude du liquide surchauffé dans l'espace contenant la vapeur, sont limitées à des valeurs considérablement plus faibles. Ce phénomène prouve évidemment l'existence d'une diminution de la "surchauffe effective" qui est produite par une augmentation du point de rosée en comparaison avec le point d'ébullition du liquide de la composition originale.

Zusammenfassung—Eine direkte experimentelle Bestätigung des allgemeinen physikalischen Hintergrundes, der den verschiedenen Blasenwachstumstheorien und der van Stralen'schen "Entspannungs-Mikroschicht" Theorie für den Mechanismus des Blasensiedens unterliegt, wird angegeben.

Dazu wurden gleichzeitig Messungen der momentanen örtlichen Temperatur mit einem feindrahtigen Thermolement und Hochgeschwindigkeitsaufnahmen der Dampfblasen, die an aktiven Keimen in unmittelbarer Nähe der heissen Lötstelle entstanden, durchgeführt.

Es zeigt sich, dass die thermische Grenzschicht während des plötzlichen anfänglichen Blasenwachstums von der Wand weggestossen wird. Die Grenzschicht erwärmt sich dann während der Wartezeit zwischen dem Ablösen einer Blase und der Entstehung der nächsten wieder.

Der Taupunkt des Dampfes im Blasenvolumen ist einheitlich und nähert sich der Sättigungstemperatur bei Umgebungs-(Atmosphären)-Druck.

Dieser Mechanismus hat in reinen Flüssigkeiten grosse örtliche Temperaturschwankungen zur Folge. Im Gegensatz zu diesem Verhalten bleiben in binären Gemischen, die einen kleinen Anteil einer leichter flüchtigen Komponente enthalten, die Temperaturabsenkungen, die beim Übergang der heissen Lötstelle von der überhitzten Flüssigkeit in den Dampfraum beobachtet werden, auf wesentlich kleinere Werte beschränkt. Offensichtlich beweist dieses Phänomen die Existenz einer verminderten "effektiven Überhitzung", die von einer Erhöhung des Taupunktes im Vergleich zum Siedepunkt der Flüssigkeit mit der ursprünglichen Zusammensetzung verursacht wird.

Аннотация—Приводятся результаты проверки общих физических основ различных теорий роста пузырьков и теории «релаксационного микрослоя» Ван Стралена для пузырькового кипения. С этой целью измерялись значения локальных температур с помощью микротермопар, синхронизированных со скоростной киносъемкой пузырьков пара, образующихся вблизи от горячего спая. Показано, что тепловой пограничный слой отрывается от стенки во время быстрого начального роста пузырька. Последующий подогрев этого слоя происходит в период между отрывом пузырька и образованием следующего. Точка росы пара в пузырьке одинакова и приближается к температуре насыщения при атмосферном давлении. В чистых жидкостях этот механизм приводит к большим колебаниям локальной температуры. В противоположность этому в бинарных смесях, содержащих небольшую долю летучих компонент, падение температуры, наблюдаемое при прохождении через горячий спай, ограничивается довольно малыми значениями. Очевидно, это свидетельствует о существовании пониженного «эффетивного перегрева», вызванного ростом точки росы по сравнению с точкой кипения жидкости первоначального состава.