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Generic features and puzzles of nucleate boiling

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ABSTRACT

At high reduced pressures extremely high nucleate boiling heat transfer coefficients (HTC) were measured. A single mechanism, which presents a consistent explanation of such HTCs, is very high intensity of liquid evaporation at the periphery of dry spots (nucleation sites) at the heated wall. Due to very small size the nucleation sites can be considered as point heat sinks. Between them convective heat transfer occurs, which in its turn is governed by the inherent mechanisms of boiling. The above two mechanisms comprise a total heat flux from the heated wall in nucleate boiling. The predicting equation, which determines heat flux in boiling via the wall superheat and liquid properties, has been developed with accuracy to two universal numerical factors fitted to the experimental data. Although the equation developed is found to be in good agreement with numerous experimental data for different liquids and in the wide range of reduced pressures and heat fluxes there exists a problem in nucleate boiling, which has not been understood to the full even qualitatively. This problem is the dependence of nucleation site density on the physical properties of the liquid and on the controlling parameters. Some new experimental results by Theofanous et al. [T.G. Theofanous, T.N. Dinh, J.P. Tu, A.T. Dinh, The boiling crisis phenomenon. Part I: Nucleation and nucleate boiling heat transfer, Exp. Therm. Fluid Sci. 26 (2002) 775–792; T.G. Theofanous, T.N. Dinh, J.P. Tu, A.T. Dinh, The boiling crisis phenomenon. Part II: Dryout dynamics and burnout, Exp. Therm. Fluid Sci. 26 (2002) 793–810.] and Qi et al. [Y. Qi, J.F. Klausner, R. Mei, Role of surface structure in heterogeneous nucleation, Int. J. Heat Mass Transfer 47 (2004) 3097–3107; Y. Qi, J.F. Klausner, Heterogeneous nucleation with artificial cavities, J. Heat Transfer 127 (2005) 1189–1196; Y. Qi, J.F. Klausner, Comparison of nucleation site density for pool boiling and gas nucleation, J. Heat Transfer 128 (2006) 13–20.] require revising the traditional views on a nature of the active nucleation sites in boiling. These results remind the old question: why can the nucleation sites arise at low superheats of the absolutely wettable surface? Obtaining theoretical equation for nucleation site density remains the most significant challenge in nucleate boiling theory.

1. Introduction

Our knowledge of boiling process was enlarged greatly during a life period of the elder generation of scientists, who continue to work actively in the field. This is enough to compare the textbooks or the review papers of the the 40s and 60s of the last century [1–4] with the modern review papers as [5–8]. However, a typical idea ordinarily presents in an introduction of many original and review papers: in spite of great efforts and essential progress "we have had limited success in the mechanistic modelling of boiling" [7].

The main reason is that we have no closed mathematical description of the nucleate boiling process, strictly speaking. Even in the case of a heated wall with infinite thermal conductivity, when there is no need to solve a conjugate problem of energy conservation, at present it is impossible to analyze actual two-phase turbulent flow with the interface of unknown shape. According to Theofanous [9] the interface in a two-phase flow presents an additional nonlinearity and requires considering such systems in their complexity, as a whole. In other words, we do not possess an analog of the Navier–Stokes equations in order to obtain instantaneous fields of velocity in liquid and vapour and their interface at an arbitrary time point. In distinction to single-phase flow, DNS for two-phase systems meets practically insuperable difficulties.

As it was discussed in detail in [8], in spite of some significant theoretical results, one has no grounds at present to speak of actual direct numerical simulation of boiling heat transfer. A strict theoretical study is possible only in relation to some particular ("elementary") processes of nucleate boiling. Progress in DNS of an individual vapour bubble growth and departure is very impressive, but this weakly affects the practical designing heat transfer equipment.

Now practical calculations of heat transfer with nucleate boiling are conducted on the basis of empirical or semiempirical equations. Number of these equations increases with time and practically has no limits: any new combination of a liquid, a surface material and geometry, pressure and heat flux range can require

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Nomenclature

B	nondimensional number according to Eq. (17)
$C_0,\ldots C_4$	numerical factors
c	specific heat, J/(kg K)
F	surface area of an equilibrium vapour bubble, m^2
G_R	mass flow rate per film width unit, kg/(m s)
H	curvature, m^{-1}
h_{LG}	latent heat of evaporation, J/kg
Ja	Jakob number
l_*	average distance between the nucleation sites, m
n_F	nucleation sites density, m^{-2}
p	pressure, Pa
Q	heat flux, W
q	heat flux density, W/m^2
R	vapour bubble radius, m
R_i	gas constant (individual), J/(kg K)
r	radial coordinate, m
T	temperature, K
t	time, s
U_∞ or W_0	characteristic velocity, m/s

Greek symbols

α	heat transfer coefficient, $W/(m^2\,K)$
Δ_0	thickness of a conductive liquid layer, m
Δp	Laplace pressure jump, Pa
$\Delta T = T_W - T_S$	wall superheat, K
$\Delta\Phi$	Gibbs energy variation, J
$\Delta\varphi$	difference of specific Gibbs potentials, J/kg
δ	liquid film thickness, m
θ	contact angle
λ	thermal conductivity, W/(m K)
ν	kinematic viscosity, m^2/s
ρ	density, kg/m^3
σ	surface tension, J/m^2

Subscripts

bi	boiling incipience
cr	critical
ds	dry spot
e	equivalent
G	gas (vapour)
hom	homogeneous
L	liquid
m	maximum evaporation rate
o	outer
R	refers to unit of interline length
S	saturation
w	wall
$*$	equilibrium

a new predicting equation for boiling heat transfer. As for nondimensional correlations for nucleate boiling modelling, in absence of closed mathematical description of the process there is no scientific ground to prove advantages or shortcomings of any such correlation (see [8] for more detailed argumentation). In the present author view, development of an approximate model of nucleate boiling heat transfer is the most suitable alternative to either unlimited generation of new empirical correlations or to attempts to realize the DNS of the process in absence of its strict mathematical description. The basic requirement to the approximate theory is revealing main features of the process modeled. Now our knowledge of nucleate boiling allows determining the main mechanisms of heat transfer. Furthermore, as it will be argued in the paper, one can say about particular, the most principal mechanism, which distinguishes the nucleate boiling from any other mode of convective heat transfer.

However, there exists such a problem in nucleate boiling, which has not been understood to the full even qualitatively. This problem is the dependence of nucleation site density on the physical properties of the liquid and of the heated surface and on the controlling parameters. Some new experimental results by Theofanous et al. [10,11] and Qi et al. [12–14] require revising the traditional views on a nature of the active nucleation sites in boiling. These results raise some new questions. First, what is a nucleation site at the nanoheater surface [10,11], where there are no cavities with a size larger or equal to an equilibrium nuclei radius? Second, what is a reason of a great difference in nucleation site densities at the brass surface and at the stainless steel surface with practically the same roughness characteristics [12,14]? One can say that these questions turned us to the other, rather old question: why can the nucleation sites arise at low superheats on the absolutely wetted surface? Or, in particular, why does boiling incipience in helium occur at so small superheat?

This objectively makes the problem of nucleate boiling heat transfer open for new investigations and discoveries. According to the author view the problem of nucleation is the main one in creating the boiling heat transfer theory.

2. Principal mechanisms of nucleate boiling heat transfer

2.1. HTC in boiling

Nucleate boiling being the most important boiling regime is characterized by very strong nonlinear heat flux dependence on wall superheat. Different empirical correlations reflect this dependence as $q \sim \Delta T^3$ ($\alpha \sim q^{2/3}$), or $\alpha \sim q^{0.7}$ ($q \sim \Delta T^{10/3}$). In experiments with boiling of high-molecular highly wetting liquids (fluorocarbons, refrigerants), especially on the very smooth heated surfaces essentially steeper dependence $q(\Delta T)$ is observed. This means that in nucleate boiling a concept of heat transfer coefficient (HTC) has no physical meaning. Estimating in general “new heat transfer” by Adiutori [15] as rather vain, we have to agree with him in this case. Really, regularities of nucleate boiling heat transfer must be expressed by means of direct interconnection of the heat flux and the wall superheat, without using HTC. The latter has actual physical meaning and is useful practically, when this is a parameter of hydrodynamic nature. In particular, the HTC has to keep its value in the limit: $q \to 0$, $\Delta T \to 0$. It is clear that in boiling this limit has no sense, because boiling incipience occurs at the definite (finite) values of q (and ΔT).

The above argumentation does not mean an attempt to get a “prohibition” of the HTC using in nucleate boiling analysis. This quantity can be practically convenient and can be used, in particular, as a tool of comparison of intensity of boiling heat transfer with other types of convective heat transport. But it is necessary to understand clearly that primary mechanisms of boiling heat transfer are expressed through the heat flux and the wall superheat, the HTC being only an auxiliary, subservient parameter in this process.

Nevertheless, there are a lot of empirical or semiempirical equations, which are destined for HTC prediction. The majority of numerous generalized correlations for boiling heat transfer are built by one way or another on the basis of analogy with single-phase convection. A typical equation connects the dimensionless HTC (Nusselt number) with the modified Reynolds number, the Prandtl number and some specific nondimensional numbers suggested by the particular investigator. The Reynolds number comprises a characteristic length and a characteristic velocity. In many cases the Laplace constant is used as a former scale, although now it is quite clear that there is no reason for such a choice [16–18]. As for the characteristic velocity there are two actual scales in nucleate boiling. The first one is an average rate of evaporation:

$$W_0 = q/(h_{LG} \cdot \rho_G) \tag{1}$$

which often used in the Reynolds number for modelling of boiling heat transfer. The other actual velocity in nucleate boiling is a bubble growth rate. This value is ordinarily used in the more sophisticated approaches to the problem. Labuntsov in 1959 and 1963 convincingly argued that “external” hydrodynamics does not affect heat transfer in the developed nucleate boiling [16], so there are no grounds to use any other scale for velocity here.

Due to decrease of vapour specific volume with pressure increase the both above velocities become very small at high reduced pressures. At the same time HTC increases continuously with pressure. This means that an analogy with single-phase convection in principle can not explain extremely high values of HTC in pool boiling at high reduced pressures. Table 1 presents several examples of the measured values of heat transfer coefficient in nucleate boiling at high reduced pressures. Very different liquids are considered. Three first lines correspond to boiling occurred on the outer surface of horizontal cylinders (tubes). Besides heat transfer coefficient, average rate of evaporation and vapour bubble growth rate, the values of thickness of so-called equivalent liquid film ($\delta_e = \lambda/\alpha$) are given. The bubble growth rate is calculated according to the Labuntsov [16] formula:

$$R = \sqrt{12\frac{\lambda\Delta t t}{h_{LG}\rho_G}}, \tag{2}$$

at the growth time $t = 1$ ms, i.e. at the moment close to nucleation, when a bubble grows quickly. It is noteworthy that a bubble growth rate in uniformly superheated liquid calculated according to the Plesset–Zwick equation for Jacob number $Ja < 1$ is essentially lower than according to (2). The last column in the Table 1 gives the values of the liquid velocity U_∞, which can provide the same heat transfer intensity in the case of external cross-flow of the cylinder (by means of single-phase convection), the well-known equation by Zhukauskas being used in the calculations.

The 4th line of the table presents the typical value of HTC in helium pool boiling at atmospheric pressure on the copper horizontal surface. In this case a comparison with single-phase convection is given for a jet of liquid helium impacting from a nozzle of 3 mm in diameter. As is seen in all the above cases the actual velocities in boiling at high reduced pressures are several orders of magnitude lower than the velocity of single-phase liquid, which needs in order to get the HTC values observed in the experiments with boiling. The equivalent film thickness is also impressed: it is difficult to conceive so thin conductive liquid layer covering an entire surface. In some cases the calculated values of δ_e are less than a characteristic size of the surface roughness.

The last line in Table 1 relates to flow boiling of carbon dioxide in a submillimetric channel. This is well-known that forced convection does not affect heat transfer at developed nucleate boiling. In the experiments of paper [22] heat fluxes were rather small (or moderate), but at high reduced pressure HTCs in boiling are much greater than in single-phase convection even in microchannels. In order to obtain in single-phase flow those heat transfer coefficients, which have been measured in flow boiling one has to support tenfold higher flow velocity than it was in the experiments [22] (For this case U_∞ is an average liquid velocity in the tube).

Thus, at high reduced pressures actual flow velocities in nucleate boiling are extremely low in order to provide the observed high intensity of heat transfer by means of any possible mechanism of single-phase convection. It is necessary to point out that the above consideration refers not only to those models that use qualitative arguments for choice of nondimensional numbers, which interdependence is established by fitting to the experimental data. So-called “mechanism-based correlations” [6] are factually also based on the analogy with single-phase convection. The particular approaches discussed in [6] supposed that the bubbles act like a pump removing the superheated liquid from the heated wall, this liquid being replaced with the cold one after bubble departure. The equation obtained accounts for contribution of transient conduction, natural convection between the growing bubbles and microlayer evaporation underneath the bubbles. Dhir [6] believes that the problem of development of this type correlation is in impossibility to obtain physically based equations for nucleation site density, bubble departure diameter and bubble release frequency. However, the considered approach in itself provokes objection. First, the model relates to the isolated bubbles regime, which is a very small part of the entire nucleate boiling regime (Zuber [23] as long ago as in 1964 sceptically estimated attempts to build up a generalized correlation, basing on analysis of the isolated bubbles regime). Second, at high reduced pressures bubble release frequency is extremely low, natural convection being negligible practically always in boiling. It is easy to understand that none of the mechanisms considered explains so high values of HTC, which are observed at high reduced pressures and are presented in Table 1.

Nonetheless, it would be completely unjustified to regard without necessary respect to the works of our predecessors. Their experimental results, their ideas on the process mechanisms, both confirmed and refuted by the succeedent investigations, constituted the basis of a modern understanding of nucleate boiling regularities. In this connection it is noteworthy to mention that the Rohsenow correlation (1952) for nucleate boiling heat transfer was not the earliest one as it is stated in [6]. In the USSR by then several correlations have been published, some of them being better physically founded than the Rohsenow formula (see, for example, [24]).

2.2. Evaporation at the interline as a generic feature of nucleate boiling

There exists a peculiarity of nucleate boiling, which distinguishes it from any other mode of convective heat transfer. This

Table 1
Rounded-off experimental values of HTC in nucleate pool and flow boiling and characteristic velocities.

Data source	Liquid	Reduced pressure	Heater	q (kW/m^2)	α (kW/m^{2}K)	δ_e (μm)	W_0 (m/s)	$\frac{dR}{dt}$ (m/s)	U_∞ (m/s)
[19]	Water	0.665	Tube, $d_o = 6$ mm	830	415	1.57	0.008	0.0052	90
[19]	Ethanol	0.775	Tube, $d_o = 5$ mm	350	140	0.79	0.009	0.0046	88
[20]	R-134a	0.798	Tube, $d_o = 25.4$ mm	60	160	0.313	0.0033	0.0018	274
[21]	Helium	0.445	Flat horizontal surface	4.0	10.0	1.96	0.0116	0.0083	12.8
[22]	Carbon dioxide	0.78	In-tube boiling, $d_i = 0.8$ mm, $G = 280$ kg/m^{2}s	10	16.6	4.9	0.00034	0.0021	3.6

is availability of interlines, i.e. the boundaries of contact of three phases. Stimulus to understanding the role of evaporation along the interline in boiling was given objectively by the work [25], in spite of an absence of a direct mention of this effect. In this work it was shown first that in nucleate boiling a local heat flux under the growing vapour bubble can be two orders of magnitude higher than the average one at the heated wall. A hypothesis on liquid microlayer evaporation as a reason of such peak heat fluxes suggested in [25] has been confirmed soon by direct observation in some special experiments. The experiments revealed also that at the center of the bubble base a dry spot exists. Now this seems to be quite natural. As an active nucleation site is an area of direct vapour–solid contact, consequently the dry spot does exist and its boundary is the line of the three phases contact.

As at the interline itself a liquid film thickness tends to zero, the local heat flux at the isothermic heated surface formally tends to infinity, if the temperature at the liquid–vapour interface is assumed to be equal to T_S. With such assumptions the problem of liquid evaporation in the vicinity of the interline was solved in [26,27]. In order to avoid of a heat flux divergence at the interline the authors used the heat flux calculated on the basis of the kinetic theory of gases as the limiting quantity. This heat flux for the conditions considered in [26,27] (water at 10 MPa) is as high as 10^{10} W/m^2 (probably, only under these unrealistic conditions it is possible to find a certain foundation of vapour recoil effect in boiling; at the above peak heat flux even in copper the temperature gradient must be 2.5×10^7 K/m, i.e. 25,000 K/mm!).

An approach to modelling of evaporating liquid film at the interline region developed by Wayner and his co-workers [28–31] seems to be essentially more sound. According to Wayner a thin liquid film near the interline comprises the regions of a nonevaporating adsorbed film, an evaporating film with essential influence of the London–van der Waals forces on liquid evaporation, and an intrinsic meniscus. In the latter region the London–van der Waals forces are negligible, the film thickness is much less than in the outer part of the meniscus; this means that the intrinsic meniscus presents an area of extremely high intensity of evaporation. All the above regions are extremely small in comparison with any other linear scales commonly used in heat transfer analysis. According to the Wayner's investigations a width of the entire interline region is an order of 1 μm or less.

Consequently, a nucleation site is the dry spot and along its boundary a very narrow zone of extremely intensive evaporation exists. Bearing in mind in addition that a typical size of the nucleation site is equal approximately to an equilibrium vapour bubble radius R_* one can consider the site as a point heat sink at the heated surface. At the space between the nucleation sites convective heat removal from the surface occurs. Basing on the above ideas the present author in 1988 [18] have suggested that the total heat flux from the wall is the sum:

$$q = q_1 + q_2, \tag{3}$$

where q_1 and q_2 are correspondingly heat fluxes due to intensive evaporation at the dry spot boundary and due to convection to liquid between the nucleation sites. Very small fraction of dry spots area in relation to the total heated surface area indicates in favour of possibility to use a simple additive law in this case (see Fig. 1).

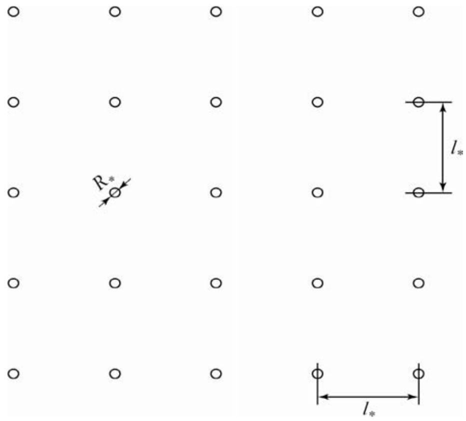

Fig. 1. Schematic of nucleation sites distribution.

At that time the main objective of the author was simply to improve the Labuntsov's models of boiling heat transfer [16]. Labuntsov was the first who has developed a theoretical model of nucleate boiling in 1963 (see [16, p. 104]). His approximate theory of developed nucleate boiling heat transfer is based on an assumption that this process is governed by its internal mechanisms and does not depend on "external hydrodynamics". Nucleation and initial very fast growth of vapour bubbles can be characterized by a definite pulsating velocity, which is analogous in its part to a friction velocity in single-phase turbulent convection. Its averaged value is determined on the basis of equating of the total kinetic energy resulting from simultaneously growing bubbles to the kinetic energy resulting from liquid pulsating motion with this velocity. The velocity discussed determines a thickness of a liquid layer at the wall, which in its turn determines heat flux density. According to the content of the analysis this heat flux is the "convective" heat flux q_2, and Labuntsov for the first time obtained an equation for this heat flux by direct way basing on the approximate mechanistic model. This equation is as follows

$$q_2 = C_1 \frac{\lambda^2 \Delta T^3}{\nu \sigma T_s} \tag{4}$$

with a numerical constant C_1 of an order of magnitude of 10^{-3}.

The other heat transfer mechanism due to direct liquid evaporation in the growing bubbles was not determined in the Labuntsov's studies on a basis of any mechanistic model. In his later work of 1972 [16, p. 138] this effect was accounted for by means of the empirical function of vapour and liquid densities ratio. It is clear that the limited possibilities of convection to remove high heat fluxes at high reduced pressures discussed in the previous section are valid also in this case. Although the Labuntsov's final equation for nucleate boiling heat transfer is in rather good agreement with numerous experimental data, his model does not give a physically grounded quantitative explanation of extremely intensive heat transfer in nucleate boiling at high reduced pressures.

Now it is possible to state that the heat flux resulted from intensive liquid evaporation in the vicinity of the dry spot boundaries, which initially has been considered by the present author as the additional one, factually presents the main mode of heat removal and is really a generic feature of nucleate boiling. So if one imagines that the circles presenting the nucleation sites in Fig. 1 tends to be the points, the two mechanisms of heat removal from the wall can be considered as practically the independent ones from each other, and the both heat fluxes in (3) being related to the entire area of the heated surface.

The above discussion, undoubtedly, essentially affects the conceptual view on nucleate boiling principal mechanisms. However, the basic steps of analysis and the final equation for HTC remain quite similar to those published in [18]. The problems of boiling incipience on the solid surface and of nucleation sites density prediction are the most complex in boiling as it will be discussed below. But at the present stage of our knowledge a simple approach by Labuntsov [16] seems to be the most suitable from a practical viewpoint. Assuming that a commercial heated surface presents cavities of any size that corresponds to ordinary observed wall superheats and employing dimension analysis one obtains the following correlation for nucleation sites density

$$n_F = C_0 R_*^{-2}, \tag{5}$$

where an equilibrium radius of vapour bubble is determined by the Laplace equation:

$$R_* = 2\sigma/\Delta p, \tag{6}$$

C_0 is a numerical factor of an order of magnitude 10^{-8}–10^{-7}.

Obviously, the heat flux due to liquid evaporation at the boundaries of the dry spot can be determined as

$$q_1 = Q_{ds} n_F,$$

where Q_{ds} is heat removal rate at a dry spot. This quantity was derived on the basis of an approximate model of the interline region. Using the results by Wayner [28–31] one can present a schematic of the liquid film meniscus in the vicinity of the dry spot as it is shown in Fig. 2. A model of the liquid film at the interline region is inevitably strongly idealized. According to the estimations of [18] the liquid film thickness at the zone of intense evaporation δ_m is an order of 10^{-8}–10^{-7}m at ordinary conditions of nucleate boiling. These estimations coincide with the further numerical simulations. The paper [32] was, probably, the first numerical simulation of heat transfer in the so-called “micro-region” that is in the thin liquid film at the boundary of a nucleation site at the heated surface. Later Stephan and his co-workers have developed this approach [33], in particular for binary mixture boiling. In [33] the maximum local heat flux in propylene/propane mixture boiling is obtained at the distance about 2×10^{-7} m from the boundary of adsorbed (non-evaporating) liquid film, where the film thickness is about 1×10^{-8} m. It is clear that for such linear scales any actual heated surface can hardly be considered as the smooth one. The above scales are less than the height of micro-roughness in the experiments of [10,11], where the special nanoscale surface has been used. This consideration gives additional arguments in favour of an approximate model of the process.

The approximate model of [18] considers the same characteristic regions of the thin liquid film at the vicinity of a dry spot as later were strictly analyzed in [32]. However, the aim of the approximate model was not the exact solution for some particular conditions, but obtaining a general correlation between the controlling parameters and the thermophysical properties of a liquid. According to the Wayner’s studies liquid flow in the film governed mainly by the capillary forces, or more exactly by the gradient of the film surface curvature. As vapour pressure is uniform and the meniscus curvature H decreases from its maximal value $1/R_*$ at the interline till to $1/\delta_o$ at the outer part of the meniscus, the pressure gradient in the liquid is directed from the dry spot boundary to the thick part of the film. Assuming that the above curvature variation occurs at the short distance $\Delta r \approx \delta_m$, one obtains for the pressure gradient in the liquid film:

$$dp/dr \sim \sigma/(R_* \delta_m). \tag{7}$$

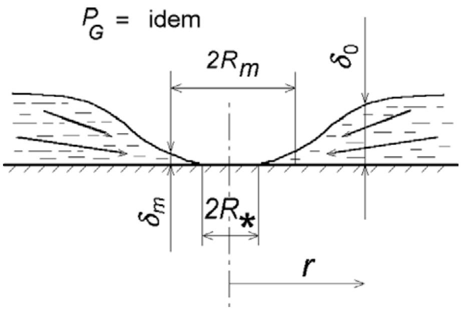

Fig. 2. Schematic of liquid film meniscus at the dry spot boundary.

This expression is the only one, which is obtained by means of physical estimations. For the known pressure gradient there exists an analytical solution for radial liquid flow in the film. This solution gives for liquid flow rate per the film width unit:

$$G_R = (dp/dr)(\delta_m^3/\nu) \sim \sigma\delta_m^2/(R_*\nu). \tag{8}$$

If all liquid, which is supplied to the interline is evaporated, a heat flux per the film width unit corresponds to the above value G_R:

$$q_R = \lambda\Delta T = G_R h_{LG}. \tag{9}$$

From the two last equations an important expression for the liquid film thickness in the region of strong evaporation follows:

$$\delta_m = C_2(\lambda\Delta T R_* \nu/(\sigma h_{LG}))^{1/2}. \tag{10}$$

Assuming a linear law of the liquid film variation with radial coordinate in the strong evaporation zone one obtains for heat sink per a dry spot:

$$Q_{ds} \sim \lambda\Delta T\delta_m. \tag{11}$$

From (5), (10), and (11) an equation for the heat flux due to evaporation in the interline region follows:

$$q_1 = C_3\left(\frac{\lambda\Delta T}{R_*}\right)^{3/2}\left(\frac{\nu}{\sigma h_{LG}}\right)^{1/2} \tag{12}$$

Eq. (4) was obtained by Labuntsov on the basis of the expressions for an isolated bubble growth rate (2) and for nucleation site density (5). It is reasonable to use a more general approach in order to obtain an equation for the convective heat flux. An average distance between the nucleation sites is obviously

$$l_* = n_F^{-1/2} = (1/\sqrt{C_0})R_* \approx 10^4 R_*.$$

Assuming that the convective heat flux is determined by the thickness of a conductive liquid layer $\varDelta_0$, which depends on liquid viscosity and on characteristic scales of length and velocity, one gets:

$$\varDelta_0 \sim (\nu R_*/W_0)^{1/2}. \tag{13}$$

In Eq. (13) an analogy with a laminar boundary layer was used, the average distance between nucleation sites and a mean evaporation rate (1) being the length and velocity scales. This equation is valid both for the isolated and coalesced bubbles boiling regimes; in the former case it gives an effective thickness of the conductive liquid layer, in the latter case $\varDelta_0$ is a liquid macrofilm thickness. Thus, the convective heat flux in boiling is expressed as follows:

$$q_2 = C_4\lambda\Delta T[q/(\nu R_* h_{LG}\rho_G)]^{1/2}. \tag{14}$$

One can see that this equation transforms into Eq. (4), if the total heat flux q coincides with the convective one and the pressure jump Δp at the bubble interface is expressed through ΔT by means of the Clapeyron–Clausius equation.

In general case substituting Eqs (12) and (14) into Eq. (3), one gets a quadratic equation in relation to the total heat flux q. After its solution interpretation of a quantity R_* is made. The pressure jump in Eq. (6), as it was shown in [18], in general case can be presented as follows:

$$\Delta p = h_{LG}\rho_G\Delta T(1 + h_{LG}\Delta T/(2R_i T_S^2))/T_S. \tag{15}$$

As is seen, at high wall superheats ΔT this equation differs essentially from the formula, which is deduced from the Clapeyron–Clausius equation. As the convective heat flux q_2 is significant at rather low reduced pressure, when high wall superheats are observed, in Eq. (14) R_* is calculated according to (6) and (15). At high reduced pressures nucleation site density is very high and a contribution of the first constitutive of heat flux becomes dominant. Under these conditions the wall superheat is commonly small, therefore in (12) R_* is determined with using the Clapeyron–Clausius equation, i.e. the expression in brackets in (15) is assumed to be equal to unit. Bearing in mind this consideration the final equation for nucleate boiling heat transfer can be presented as follows:

$$q = 3.43 \times 10^{-4} \frac{\lambda^2 \Delta T^3}{\nu \sigma T_s} \left(1 + \frac{h_{LG} \Delta T}{2 R_i T_s^2}\right) \left(1 + \sqrt{1 + 800B} + 400B\right), \tag{16}$$

where

$$B = \frac{h_{LG} (\rho_G \nu)^{3/2}}{\sigma (\lambda T_s)^{1/2}}. \tag{17}$$

Two numerical factors in (16) were determined from the best fitting to the experimental data.

Fig. 3 is reproduced from the original paper [18]. The calculated curve is built in accordance with the Eq. (16) in the form of dependence of nondimensional HTC $\tilde{\alpha} = \frac{\alpha}{q^{2/3}} \left(\frac{\nu \sigma T_S}{\lambda^2}\right)^{1/3} \left(1 + \frac{h_{LG} \Delta T}{2 R_i T_S^2}\right)^{-1/3}$ on the nondimensional parameter B determined according to (17). About 3000 experimental points for 12 different liquids (water, ethanol, methane, ethane, ethylene, nitrogen, benzene, and different refrigerants) were used for comparison. More than 91% of the points lie within the range ±35% from the calculated line. The parameter B is a strong function of reduced pressure. The experimental data presented in Fig. 3 relate to reduced pressures at very wide range $p/p_{cr} = 1.8 \times 10^{-4}$–0.94. As it has been already mentioned, when the reduced pressure increases the nucleation sites density increases also and a contribution of heat flux q_1 becomes the predominant one in the total heat flux.

Certainly, the Eq. (16) describes nucleate boiling heat transfer at some "average" conditions. The state of [34] on possible twofold difference in HTC in boiling at the same wall superheat and the different heated surface microstructure remains true independently to any nucleate boiling model. This explains the inevitable scatter of experimental points in relation to the predicting equation. As was discussed in [18], some tendencies in the experimental data can be connected with an effect of thermophysical properties of the heated wall substance. An influence of thermal activity factor $(\rho c \lambda)_w^{1/2}$ of the wall material on nucleate boiling heat transfer is especially remarkable for cryogenic liquids. In Fig. 3 the experimental values of HTC in nitrogen boiling on copper surface are usually higher than the predicted ones, and they lie below the predicted line when boiling on stainless steel wall is studied. For this reason the equation (16) agrees with the experimental results in hydrogen and helium nucleate boiling only when the heated wall with high thermal conductance (copper and argentum) was used in the experiments. Detailed analysis of cryogenic liquids boiling heat transfer is given and the specific predicting equations for these liquids are proposed in [35]. The Eq. (16) does not account for an influence of the wall thermophysical properties on nucleate boiling heat transfer. But it reflects well the reduced pressure effect on HTC variation for all cryogenic liquids, as it was convincingly shown in [17].

During rather long time after the Eq. (16) derivation many new experimental data were compared with prediction according to it. In particular, the results for hydrocarbons and fluorocarbons have been tested. Certainly, in some cases, especially in boiling at the heated walls with properties strongly different in comparison to ordinary commercial surfaces the deviation of experimental and predicted values of HTC occurs to be rather essential. But in general the comparison gave the confirmation of rather good predicting capability of the equation.

In the last few years a substantial upswing in the interest is observed in studying nucleate boiling of carbon dioxide. This relates both to pool [36] and to flow boiling [37]. Fig. 4 presents a comparison of the calculations according to Eq. (16) with the experimental data by the authors of [38] on carbon dioxide pool boiling. The experiments were conducted at rather low for CO_2 reduced pressures $p/p_{cr} = 0.1$ and 0.19, if one remembers that reduced pressure at the triple point is 0.07 for this substance. The horizontal tubes from copper, stainless steel and aluminum with outer diameter of 16 mm and different surface treatment were used as heaters. As is seen from the figures, the experimental points are distributed near the computed curves with typical for nucleate boiling scatter, the higher experimental HTCs being observed for surfaces with higher thermal activity $(\rho c \lambda)_w^{1/2}$ and higher roughness.

Regularities of nucleate boiling remain the dominant ones at flow boiling under the conditions of rather high heat fluxes. As was demonstrated in [39], the Eq. (16) describes well the experimental results on heat transfer in boiling of water and refrigerant R-113 in tubes and rather unique data on boiling heat transfer of nitrogen in impinging jets. At flow boiling the region of heat fluxes with commensurable effects of single-phase convection and nucleate boiling is rather narrow. HTC in this region is successfully predicted with a help of a simple interpolation between the HTC values calculated with using the reliable equations for single-phase convection and nucleate boiling. The Eq. (16) is a good candidate for nucleate boiling contribution prediction. Very recently the experimental data on carbon dioxide flow boiling were compared with calculations according to Eq. (16). At rather low flow qualities ($x < 0.3$) good agreement of the data and the predictions was revealed both for ordinary (6–10 mm) and for very small (0.6–2 mm) channels.

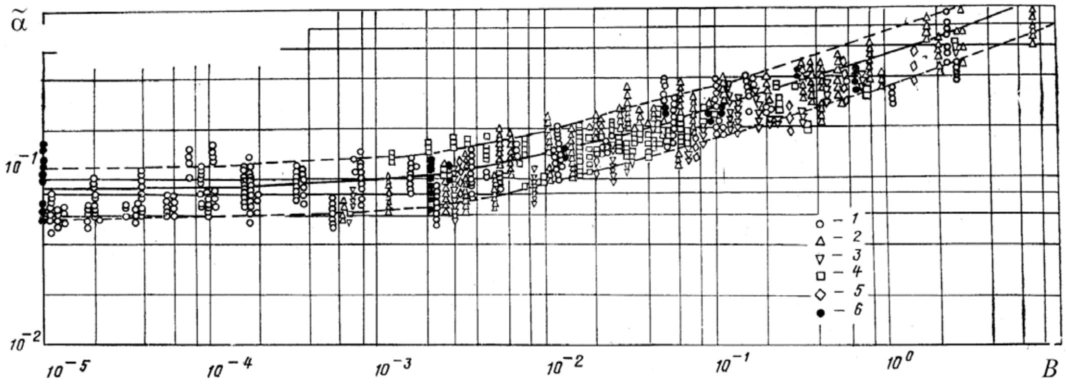

Fig. 3. Comparison of calculations according to Eq. (16) with the experimental data on nucleate boiling heat transfer: 1 – water; 2 – ethanol; 3 – benzene; 4 – refrigerants; 5 – nitrogen; 6 – methane, ethane, ethylene. The figure is reproduced from [18].

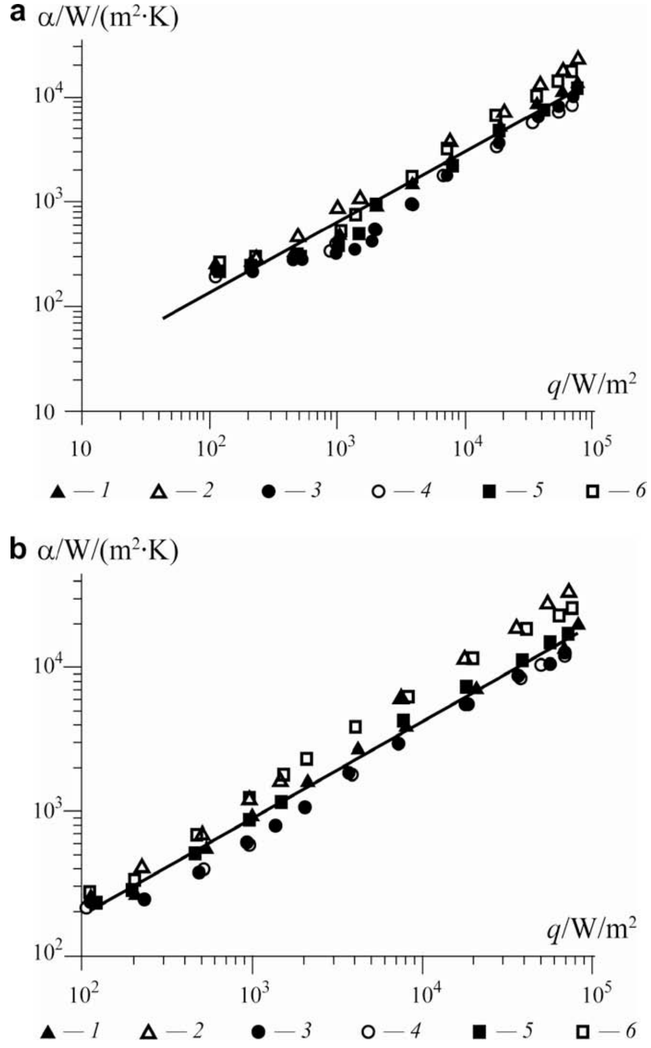

Fig. 4. Calculated and measured HTCs at carbon dioxide pool boiling: lines – calculations according to Eq. (16), points – experiments of [38]; (a) $p = 0.738$ MPa, (b) $p = 1.4$ MPa; 1, 2 – copper grinded and sandblasted tubes, 3, 4 – stainless steel differently grinded tubes, 5, 6 – aluminium grinded and sandblasted tubes.

3. Nucleation and nucleation site density

An ability of the Eq. (16) to correlate very different experimental data on nucleate boiling heat transfer can be considered as evidence that the underlying approximate theory adequately describes the main features of the process. But it does not mean that this theory closes the problem. On the contrary, the significance of some unanswered issues becomes clearer. Undoubtedly, the main of such issues is dependence of nucleation site density on controlling parameters and properties of a liquid and a heated surface. The very simple formula (5) proposed by Labuntsov and used by the present author is dimensionally correct and has a rather clear interpretation for the commercial surfaces. The disadvantage of this correlation consists not only in its discrepancy from some experimental measurements. (Their majority has been conducted at the limited ranges of heat flux and pressure variation and on the surfaces, which greatly differ from those corresponding to technical applications). It seems to be more significant that the experiments with boiling of high-molecular highly wetting liquids (fluorocarbons, refrigerants) demonstrate essentially steeper dependence $q(\Delta T)$ than commonly used one in the models and correlations: $q \sim \Delta T^3$. In these experiments boiling incipience (T_{bi}) is usually shifted to the region of rather high wall superheats. At the same time, Eq (5) predicts the continuous increase of n_{F} with ΔT at any wall superheat $\Delta T > 0$. There is some reason in the idea of using the temperature difference $T_{\text{w}} - T_{\text{bi}}$ instead ordinary ΔT in calculations of R_* (see [35]). However, now the equations for predicting boiling incipience are essentially less reliable than the equations for nucleate boiling HTC.

It should be noted that at present the problem of nucleation and nucleation site density is investigated energetically and rather widely. Recently the main results of complex and coordinated investigations of the German scientists were published in the special issue of the International Journal of Thermal Sciences [20,40–42]. Among other things very important data on short term and long term nucleation site densities were presented [40,20,42]. It was established that the number of simultaneously acting sites can be of an order of magnitude less than the total number of the sites, which become active for a long time observation. This is especially true for high reduced pressures, when a number of continually acting sites is rather small. Very fine and sophisticated measurements of [41] present the significant information on local surface temperature and void fraction variation during nucleation and bubble growth on the as small linear scales as several micrometers. But more fundamental questions arise due to the papers [10–14] mentioned in the Introduction.

In very interesting and informative investigations [10,11] the nucleation sites were found not to be the cavities at the heated surface. But this decisively established experimental fact is revealed for a very peculiar condition of a nanoscale heater, when nucleate boiling starts at superheats more than 20 K in water at atmospheric pressure. This observation on one hand poses the question of what is a nucleation site on such a smooth surface, but on the other hand it cannot disprove earlier directly observed in many experiments an actual part of the surface cavities in nucleation. The above mentioned studies [20,40,42] also confirmed the surface cavities significance in nucleation. The results of [10,11] provoke a more general question: what is a reason of boiling incipience at rather small superheats in the case of absolutely wettable surfaces?

According to [16] the Gibbs energy variation during a vapour bubble formation on the heated surface is expressed as follows:

$$\Delta\Phi = V\rho_G\Delta\varphi + \sigma F[1 + (F_w/F)(1 - \cos\theta)]. \tag{18}$$

In this equation $\Delta\varphi$ is difference of the specific Gibbs potentials of vapour and liquid, V is a volume of vapour phase formed, F and F_w are correspondingly an entire surface area of the vapour nucleus and area of vapour/solid contact, θ is a contact angle. In a superheated liquid the first term of Eq. (18) is negative; the second one is positive and presents factually an energetic barrier for development of a viable vapour nucleus connected with new interface formation. As is clear, at absolutely wettable solid surface the second term in the square brackets of (18) is equal to zero. This means that in this case the solid wall does not present any advantages for nucleation in comparison with liquid volume. The recent work [43] presents, probably, the only example, when this conclusion is confirmed experimentally. In this paper boiling incipience of fluorocarbon FC-72 occurs at the homogeneous superheat limit for this liquid. The microheaters 0.27 × 0.27 mm coated with silicon dioxide were used in this study.

But it is well known [35] that liquid helium incipience occurs at the very low superheats (about 0.02 K), which are incommensurably less than the superheat corresponding to the homogeneous nucleation, $\Delta T_{hom} = 0.53$ K. Labuntsov [16] pointed out that for nucleation it is enough to have a very small portion of the solid surface with poor wettability, the linear scale of this portion must be equal to an equilibrium size of vapour nucleus R_*. (For helium at $\Delta T = 0.02$ K this is about 0.1 μm). But what are these nonwettable portions at the wall in the case of helium boiling or on the nanoheater in the experiments of [10,11]? Now we have no answer.

Enigmatic results were observed in [12–14]. A great difference in nucleation site densities at the brass surface and at the stainless steel surface with practically the same roughness and wettability characteristics does not find any explanation. As it was mentioned above, HTC in boiling is affected by the thermal activity factor of the heated wall material. In [35] this is connected with the different cooling effects of microlayer evaporation on the different materials. But in [12] a gas nucleation technique was used; gas bubbles nucleation was caused by depressurization, so that no unsteady thermal conductivity processes occurred. Nonetheless, nucleation site density in water on the brass surface was $2.5 \times 10^6\ \mathrm{m}^{-2}$, while on the stainless steel surface only $3.4 \times 10^5\ \mathrm{m}^{-2}$ at practically the same wettability and surface roughness.

The experiments on nucleation at boiling and at depressurization [13,14] at the same surfaces also revealed some surprising regularities. For water the nucleation site density in boiling was less than at the gas nucleation technique. But for ethanol, which wettability is very high, especially in relation to the brass surface, nucleation site density was very low in the gas experiments. At the same time in boiling the nucleation site density was rather high at the moderate wall superheats. On the silicon surface, which is absolutely wetted by ethanol, its boiling incipience required as high wall superheat as 60 K, gas bubbles did not formed at depressurization at all [13].

Only some of the above enigmatic observations can be qualitatively explained on the basis of the thermodynamic Eq. (18). In [14] it is reported about new very important observations obtained by atomic force microscopy [44,45]. On hydrophobic surface (glass with 0.5 nm rms roughness) closely packed nanobubbles with pancake shape are observed, their height and base diameter being about 20–30 and 200 nm correspondingly [44]. But such nanobubbles have not been formed on the hydrophilic surfaces [45]. These results also can be considered as agreeable with the Eq. (18), as at poor wettability even at the small area very low wall superheats are required for nucleation. But how can nucleus arise on the highly wettable surface in the case of ethanol or helium? And what is the reason of the great difference in nucleation on the brass and stainless steel surfaces or at the same surface during boiling and depressurization? Without finding the answers on these questions it is impossible to believe that we have understood the main mechanisms of nucleate boiling to the full.

4. Concluding remarks

1. Presence of dry spots at the heated wall and strong liquid evaporation along their boundaries distinguishes nucleate boiling from any other mode of convective heat transfer. This is a generic feature of nucleate boiling and the only reason of extremely high heat transfer intensity at high reduced pressures.
2. Development of an approximate model of nucleate boiling heat transfer is the most suitable alternative to either unlimited generation of new empirical correlations or to attempts to realize the DNS of the process in absence of its strict mathematical description. The approximate theory proposed by the present author adequately describes the main features of the process modeled; the final equation for boiling heat transfer is in a good agreement with the majority of available experimental data for different liquids and in the wide range of reduced pressures.
3. Nucleation on the heated wall and nucleation site density dependence on the controlling parameters and liquid/surface properties are not understood to the full at present. Without an answer to the main question, why the nucleation sites can arise at low superheats on the absolutely wetted surface, it is impossible to believe that nucleate boiling theory has been created.

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