Non-equilibrium processes in multiphase and multicomponent microscale systems

V V Kuznetsov
Institute of Thermophysics Siberian Branch of Russian Academy of Sciences
1, Academician Lavrentyev Av., Novosibirsk, 630090, Russian Federation
E-mail: vladkuz@itp.nsc.ru

Abstract. The current trend towards miniaturization of the devices in various fields of technology has led to the creation of a wide range of heat exchangers with a size from units to hundreds of microns. The analysis of the processes of non-equilibrium phase change in microsystems was performed for high rate of heat and mass fluxes. A new approach for the prediction of the flow boiling and condensing heat transfer in microchannels accounting the suppression of nucleate boiling, two-phase forced convection and non-equilibrium evaporation of a thin liquid film is proposed and verified using experimental data. Numerical study of the explosive boiling in a metastable liquid was performed based on the mathematical model of nucleation that account for thermal non-equilibrium bubble growth model.

1. Introduction
Due to a rapid growth of the applications that require transfer of the large amount of heat in the limited space, more and more attention has recently been paid to the development of microscale systems where the non-equilibrium phase transition occurs. Two-phase micro heat exchangers offer ample opportunities for engineering applications such as evaporators or condensers of power and cryogenic units as well as MEMS devices [1]. Published methods for predicting the heat transfer with flow boiling and condensing are based either on certain modification of models for conventional tubes [2] or on specially developed models [3], but their application in various heat-exchange regimes was hardly a success. A promising direction in the application of microsystems is the MEMS devices based on controlled explosive vaporization of the liquid micro volumes such as thermal inkjet printheads [4]. The characteristics of explosive vaporization on a flat microheater were considered, for example, in [5], but the initial stage of an explosive evaporation is poorly understood because of its short duration.

The aim of the present study is analysis of the mechanisms of non-equilibrium phase change in microsystems and proposing new approaches for the prediction of flow boiling and condensing heat transfer as well as explosive vaporization that account for the phenomena becomes essential for high rate of heat and mass fluxes.

2. Non-equilibrium phase change in microscale systems
The non-equilibrium phase change in microscale systems is characterized by high intensity of the heat and mass transfer when the non-equilibrium phenomena becomes important for prediction of the fluxes on the vapor-liquid interface. The important processes when non-equilibrium effects can be essential, is
the evaporation and condensation on the interface of thin liquid film. The dominant mechanism of resistance to heat transfer for this film is the thermal conduction but the waves on the interface due to shear stress in interface can considerably enhance the heat transfer.

For calculation of the heat transfer coefficient for liquid film without waves, the assumption that the temperature at the liquid-vapor interface equals the saturation temperature of the vapor is usually used. This assumption becomes wrong for high mass fluxes in the interface. Therefore, when analyzing evaporation or condensation for thin liquid film, it is necessary to pay attention on correct formulation of the thermal boundary conditions at the vapor–liquid interface. The simple approach that account for alteration of the interfacial temperature from the saturation temperature during phase change in a flat surface was proposed in [6] using the Maxwell velocity distribution function for the vapor molecules. If the evaporation and condensation coefficients are equal and Clausius-Clapeyron equation can be applied (small temperature difference), this approach provides the linear dependence of the heat flux on the excess temperature of the phase boundary as follows:

\[
q_{non} = \frac{2\chi}{2 - \chi} \frac{h_{lg}^2}{\sqrt{2\pi R_{gas} T_{sat}^{3/2}}} \rho_{gas} \left( T_i - T_{sat} \right),
\]

where \(q_{non}\) is the heat flux in the interface, \(T_i\) is the temperature of interface, \(T_{sat}\) is the saturation temperature, \(\chi\) is the condensation coefficient, \(h_{lg}\) is latent heat of vaporization, \(\rho_{gas}\) is the vapor density and \(R_{gas}\) is the gas constant. Using equation (1) the heat transfer coefficient at the liquid-vapor interface due to non-equilibrium phase change can be obtained as follows:

\[
h_{non} = \frac{2\chi}{2 - \chi} \frac{h_{lg}^2}{\sqrt{2\pi R_{gas} T_{sat}^{3/2}}} \rho_{gas}.
\]

The total heat transfer coefficient for non-equilibrium evaporation or condensation for thin liquid film is defined for the temperature difference \(T_{ev} - T_{sat}\) and can be written as follows:

\[
\frac{1}{h_{ev}} = \frac{1}{h_{non}} + \frac{1}{h_{film}}.
\]

This means that when studying processes of evaporation or condensation on the microscale it has to be taken into account that the temperature of a vapor–liquid interface for one-component liquid is not exactly equal to the saturation temperature.

It should be noted that heat and mass transfer for thin liquid film are complex phenomena and many factors need to be account not included in equation (3). Among the factors, that causes the local deviation from equilibrium are the Laplace pressure in curved interface and Van der Waals forces. In case of multicomponent liquid, the vapor and liquid temperatures at the interface are dependent on mixture composition and the impact of non-equilibrium phase change is not so essential.

The non-equilibrium in the vapor phase will affect the evaporation rate during bubble growth in metastable liquid. The initial stage of the bubble growth is characterized by high heat flux in the bubble wall due to thin thermal boundary layer. The non-equilibrium evaporation changes the interface temperature in compare with equilibrium temperature that corresponds to the pressure inside of the bubble. For a relatively small mass fluxes, the pressure in a vapor can be presented as follows [7]:

\[
p_{sat}(T_i) - p_{sat}(T_{gas}) = \frac{2\pi R_{gas} T_i}{1.67} j_i,
\]

where \(p_{sat}(T)\) is the equilibrium pressure for given temperature and \(j_i\) is the mass flux in interface. Using the Clausius-Clapeyron equation, the equation (4) can be presented as follows:

\[
q_i = h_{non} \left( T_i - T_{gas} \right),
\]

\[
h_{non} = \frac{1.67 h_{lg}^2 \rho_{gas}}{T_i^{3/2} \sqrt{2\pi R_{gas}}}.
\]
The total heat transfer coefficient for non-equilibrium evaporation $h_{ev}$ depends on heat transfer coefficient for the bubble wall $h_{nab}(t)$ that is decreased in time due to thermal boundary layer growth.

3. Flow boiling and condensing heat transfer in microchannels

The reliable model for prediction of the flow boiling heat transfer in microchannels should account for the determining mechanisms of heat transfer selected in [8] as follows: nucleate boiling heat transfer is essential for the high heat flux, low vapor quality, moderate and high reduced pressures and relatively small mass flux; nucleate boiling suppression occurs in the annular flow if the liquid film thickness becomes sufficiently small; forced convection heat transfer is essential at high mass flux and vapor quality; evaporative heat transfer in the annular flow becomes essential for high mass flux and small transverse size of the channels due to formation of a thin liquid film; evaporative heat transfer in the elongated bubble flow is essential at small heat flux and vapor quality. To account the contribution of nucleate boiling, forced convection and non-equilibrium evaporation for thin liquid film in the general equation for two-phase heat transfer coefficient $h_{tp}$, it is suggested to use the method proposed in [9]:

$$h_{tp}^2 = (h_{con} F)^2 + (h_{nab} \Phi_{sup} S)^2 + (E \phi \phi_{ev})^2$$

(6)

Here $h_{con}$, $h_{nab}$ and $h_{ev}$ are the forced convection, nucleate boiling and liquid film evaporative heat transfer coefficients, $F$ and $S$ are the forced convection enhancement and nucleate boiling suppression:

$$F = \left[1 + x \Pr \left(\rho_{liq}/\rho_{gas} - 1\right)\right]^{0.35}, \quad S = \left(1 + 0.055 \rho_{liq} a_{16}^{-1}\right)^{-1}$$

(7)

$\Phi_{sup}$ is the boiling suppression factor in the liquid film [8], parameter $\phi$ equals to the part of the channel perimeter, where the liquid film exists (without the area of menisci). To account the effect of thermophysical properties of the working fluid and channel dimensions on forced convection heat transfer, the suitable correlation for heat transfer coefficients can be used for laminar and turbulent liquid flow [9].

Nucleate boiling suppression for a thin liquid film is accounted by boiling suppression factor in the liquid film $\Phi_{sup}$ as the multiplier for nucleate boiling suppression term $S$ in equation (6). For the round tubes, the boiling suppression factor is not varied along the channel perimeter, therefore $\Phi_{sup} = \Phi_{sup0}$, $\Phi_{sup0} = \tanh^2(2.5 \times 10^{-5} \theta_{sup}^2)$ and $\phi = 1$. For the rectangular microchannels, it is necessary to take into account the absence of nucleate boiling suppression in the menisci area. The relative size of this area for a three-sided heating rectangular duct is determined by $\alpha_m = 6R_m/(2a+b)$, $\Phi_{sup} = (\alpha_m + (1-\alpha_m)\Phi_{sup0})$ and $\phi = 1 - \alpha_m$. The correct selection of nucleate boiling model in equation (6) is important for reliable heat transfer prediction. For microchannel heat sink made by precise milling, the nucleate boiling model [10] is recommended. The evaporation factor $E$ corresponds to the relative cross-sectional areas along the flow occupied by the continuous gas core. For the elongated bubble flow and transition flow, evaporation factor can be estimated from homogeneous flow model $E = (1 + \rho_{gas}(1-x)/\rho_{liq}x)^{1/2}$. For the annular flow, gas core is continuous and $E = 1$.

The equation (6) can be used also for prediction of the condensing heat transfer excluding the boiling term. In this equation, the heat conduction through liquid film is the important mechanism of heat transfer for high mass fluxes if nucleate boiling is suppressed. For the elongated bubble flow, heat transfer coefficient with account of non-equilibrium temperature jump for evaporation and condensation can be calculated according to (3) using the equation for film thickness [11]:

$$h_{film} = \left(\lambda_{liq}/\delta_{ph}\right), \delta_{ph}/D_h = 0.67Ca^{0.5}/\left(\left[1 + 3.35Ca^{0.5}\right]\right)$$

(8)

Here, $\delta_{ph}$ is the liquid film thickness, $D_h$ is the hydraulic channel diameter, $Ca = u_b \mu_{liq}/\sigma$ is the capillary number defined via bubble velocity $u_b$, liquid viscosity and surface tension.

For annular flow in rectangular microchannel, the results of numerical calculations of the heat transfer according to the model [12] were used. This model allows calculating evolution of the interface shape deformed by capillary forces due to liquid accumulation near the channel corners and
evaporation. The interfacial shear stress is calculated according to [13]. After that, the heat problem for film and wall areas is solved to determine the distribution of local transfer coefficient along the channel perimeter as well as the dependence of averaged heat transfer coefficient on balance vapor quality. Figure 1 shows the interface shape in rectangular channel with cross-section of 335x930 μm during refrigerant R134a condensation for vapor quality 0.645 and mass flux 380 kg/m²s. Here ξ is the distance from the channel angle. The initial film has uniform thickness and data in figure 1 correspond to distance from channel inlet equals to 0.6 m. As seen, the condensation in rectangular channel is characterized by ultra thin liquid film near the channel corner where the non-equilibrium temperature jump can be essential. Another surface shape is observed for evaporation in annular flow when film rupture occurs near the channel corners [12]. In this case, the heat transfer coefficient averaged along the channel perimeter is determined by local heat flux near the contact line and can essentially exceed the value of heat transfer coefficient for condensation. For microchannel with cross-section of 335x930 μm², the heat transfer coefficient for evaporated ruptured film of R134a is estimated as 11 kW/m².

![Figure 1. Interface shape in rectangular channel with cross-section of 335x930 μm during refrigerant R134a condensation for vapor quality 0.645 and mass flux 380 kg/m²s.](image)

![Figure 2. Heat transfer coefficient vs. vapor quality for qw = 22.3 kW/m². Points are data [9], lines are calculations using [2] and equation (6).](image)

The comparison of experimental data [9] for saturated flow boiling of R-134a in copper microchannel heat sink with predictions according to equation (6) is shown in figure 2. In experiments, the microchannel plate with twenty-one channels having cross-section of 335x930 μm² was used. The effect of vapor quality on heat transfer coefficient is presented in figure 2 for average mass flux G=380 kg/m²s, saturation temperature 24.2 °C and heat flux qw = 22.3 kW/m². Points in figure 2 show the experimental data from [9], the calculation according to model [2] is presented as dashed line. The solid line in figure 2 shows the calculation according to equation (6) that reasonably fits the experimental data.

**4. Explosive boiling in microscale systems**

The MEMS control systems, such as ink jet printers, optical switches and valves, use the explosive vaporization of metastable liquid. Homogeneous nucleation becomes dominant for explosive vaporization unlike the conventional boiling incipience. The proposed model of explosive vaporization is based on the theory of homogeneous nucleation [14] with account of non-equilibrium in vapor phase. In the nucleation model, frequency of nucleation is given by the work of critical nucleus formation \( W^*_c \):

\[
J = N_f B \exp\left(-W^*_c/kT\right), \quad W^*_c = \psi(\theta)16\pi\sigma^3/3(\rho_{sat} - \rho_{liq})^3(1-\rho_{sat}/\rho_{liq}).
\]

Here \( N_f \) is the number of molecules per a unit volume, \( B \) is the kinetic coefficient, \( \theta \) is the wettabiliy angle. The low wettability of solid surface decreases the work of critical nucleus formation.
The total number of bubbles nucleated in the volume of liquid with non-uniform temperature in time $\tau$ is determined as the integral on the superheated liquid volume and time:

$$n(\tau) = S_0 \int_0^\infty (1 - S_{\text{gas}}(t)) \int J_{\text{hom}}[T_{\text{liq}}(z, t), p_{\text{liq}}] dz dt$$

(10)

Here $S_0$ is the surface area of the heater and $S_{\text{gas}}(t)$ is surface coverage by vapor bubbles. The important feature of explosive vaporization is generation of the pressure wave in the vicinity of the heater surface [15]. The pressure in a vicinity of the heater can be determined using the model of accelerated motion of the flat piston with an effective diameter equivalent to the surface area of the heater. Let $z_{\text{gas}}(\tau) = V_{\text{gas}}(\tau) \cdot (1 - p_{\text{gas}}/p_{\text{liq}})/S_0$ is the effective external coordinate of the bubbles layer during nucleation and growth of the bubbles, where $V_{\text{gas}}(\tau)$ is the total vapor volume, accumulated during time $\tau$. Then the pressure in a vicinity of the heater, averaged over its surface, can be determined as follows [15]:

$$m_p \frac{d^2 z_{\text{gas}}}{d \tau^2} = S_0 (p_{\text{liq}} - p_{\text{e}}) \quad m_p = 8 \rho_{\text{liq}} \eta^3 / 3 \pi^{3/2}.$$

(11)

Here $p_{\text{liq}}$ is the pressure in a vicinity of the heater, $p_{\text{e}}$ is the pressure in surrounding liquid, $m_p$ is the effective piston mass, $l$ is the heater size. The total surface of the heater covered by semi-spherical bubbles can be represented as the integral over time:

$$S_{\text{gas}}(\tau) = S_0 \int_0^\tau (1 - S_{\text{gas}}(t)) \int J_{\text{hom}}[T_{\text{liq}}(z, t), p_{\text{liq}}] dz dt$$

(12)

Here $S_{\text{gas}}(t)$ is the area of the heater surface occupied by vapor bubbles, $S_0(t, t)$ is the area of bubble projection nucleated at time $t$.

The self-consistent model of explosive boiling in metastable liquid includes equations (9) – (12), bubble growth equation and Fourier equations for the heater and liquid. The bubble growth equation for compressible liquid is as follows:

$$\left[1 - \frac{2 \alpha_{\text{liq}}}{c_s} \right] \frac{d c_{\text{liq}}}{d \tau} + \left[1 - \frac{4 \alpha_{\text{liq}}}{c_s} \right] \frac{3 \sigma_{\text{liq}}^2}{2 \rho_{\text{liq}}} - \frac{R_b}{\rho_{\text{liq}} c_s} \frac{d (p_{\text{liq}} - p_{\text{e}})}{d \tau} = \frac{p_{\text{liq}} - p_{\text{e}}}{\rho_{\text{liq}} c_s}$$

(13)

Here $\alpha_{\text{liq}}$ is the radial liquid velocity at the bubble wall, $c_s$ is the sound velocity, $p_{\text{liq}} = p_{\text{liq}}(T) - 2 \sigma/R_b$ is the pressure in liquid at the bubble wall. To determine vapor pressure in the bubble, equation (4) is used.

The dependence of the bubble wall velocity on time for single bubble in water near superheated limit is shown in figure 3. The calculation was done for stepwise water heating described in [15] at $q_{\text{eff}} = 732.3$ MW/m$^2$ and atmospheric pressure. As seen, the maximum velocity is observed at $10$ ns after...
bubble incipience, and is high then 40 m/s. After that, cooling of the liquid in a bubble wall reduces the rate of bubble growth.

Figure 4 shows the comparison of calculation result for heater surface coverage by vapor bubbles on time for explosive boiling of water with the experimental data [15]. The calculation was done for stepwise water heating at $q_{\text{eff}} = 732.3$ MW/m$^2$ and atmospheric pressure. In calculation, the reduction of critical nucleation work on the silicon-carbide surface was taken into account. As seen, the proposed model that includes non-equilibrium model of the bubble growth predicts well the experimental data.

5. Conclusions
The analysis of the processes of phase change in microsystems shows that non-equilibrium temperature and pressure jumps on the interface become essential for high heat and mass fluxes. Numerical calculations of the evaporation and condensation for annular flow in rectangular microchannel allows calculating evolution of the interface shape deformed by capillary forces due to liquid accumulation near the channel corners and evaporation. A new approach for the prediction of the flow boiling and condensing heat transfer in microchannels, taking into account the suppression of nucleate boiling, two-phase forced convection and the non-equilibrium evaporation of a thin liquid film correspond well to the experimental data for flow boiling in microchannel heat sink. The non-equilibrium in the vapor phase will affect the bubble growth in single component liquid that is characterized by high heat and mass fluxes on the bubble wall. In case of multicomponent liquid, the vapor and liquid temperatures at the interface are dependent on mixture composition and the impact of non-equilibrium phase change is not so essential. The accounting non-equilibrium model of the bubble growth in the mathematical model of self-consistent nucleation in the induced pressure field allows to predict more accurate the rate of bubble growth after nucleation. The numerical calculations of explosive boiling show that calculation results correspond well to the experimental data obtained for water.

Acknowledgments
This research was supported by the Federal Agency for Scientific Organizations of Russia (sections 2, 3) and Comprehensive Program of Basic Research of the SB Russian Academy of Sciences on Integration Interdisciplinary Studies, project 5.3 (section 4).

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