Mathematical simulation for non-equilibrium droplet evaporation

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Abstract: -Investigations of acute problems of heat and mass exchange accompanied by phase transitions need adequate modeling of evaporation, which is extremely important for the curved surfaces in the presence of strong heat fluxes. Working cycle of heat pipes is governed by the active fluid evaporation rate. Combustion of most widely spread hydrocarbon fuels takes place in a gas-phase regime. Thus, evaporation of fuel from the surface of droplets turns to be one of the limiting factors of the process as well. In the present paper processes of non-equilibrium evaporation of small droplets were investigated theoretically. The rate of droplet evaporation is characterized by a dimensionless Peclet number (Pe). A new dimensionless parameter I characterizing the deviation of phase transition from the equilibrium was introduced, that made it possible to investigate its influence on variations of the Peclet number and to determine the range of applicability for the quasi-equilibrium model. As it follows from the present investigations accounting for non-equilibrium effects in evaporation for many types of widely used liquids is crucial for droplets diameters less than 100 microns, while the surface tension effects essentially manifest only for droplets below 0.1 micron.

Key-Words: - Phase transition, heat flux, evaporation, non-equilibrium, diffusion, mathematical simulation

1 Introduction

Investigations of acute problems of heat and mass exchange accompanied by phase transitions need adequate modeling of evaporation, which is extremely important for small droplets and sprays [1-4]. Combustion of hydrocarbon fuels takes place mostly in a gas-phase regime. Thus, evaporation of fuel from the surface of droplets turns to be one of the limiting factors of the process [5-7].

Mathematical models for individual droplets evaporation incorporated in polydispersed mixtures modeling, are usually based on the assumptions of the equilibrium character of phase transitions.⁵ Comparison of theoretical and experimental data shows that this assumption being undoubtedly valid for large droplets and flat surfaces, brings to essential errors for small droplets [4, 10, 11].

The aim of the present study is to develop a mathematical model for the non-equilibrium evaporation of droplets and to determine the applicability limits for the existing quasi-equilibrium models.

2 Mathematical model

We regard an axis-symmetrical problem in the coordinate system with the center coinciding with the center of a droplet, the zone $x > x_w$ occupied by a gas

mixture, the zone $0 < x < x_W$ - by liquid, $x = x_W$ being the phase interface.

The system of equations for the gas mixture above the interface $(x > x_w)$ has the form:

$$\frac{\partial \rho}{\partial t} + \frac{1}{x^{k}} \frac{\partial \rho v x^{k}}{\partial x} = 0, \qquad (1)$$

$$\frac{\partial \rho Y_{i}}{\partial t} + \frac{1}{x^{k}} \frac{\partial \rho Y_{i} v x^{k}}{\partial x} = \frac{1}{x^{k}} \frac{\partial}{\partial x} x^{k} \rho D_{i} \frac{\partial Y_{i}}{\partial x},$$

$$i = 1, \dots N$$

$$\frac{\partial \rho h}{\partial t} + \frac{1}{x^{k}} \frac{\partial \rho v h x^{k}}{\partial x} = \frac{1}{x^{k}} \frac{\partial}{\partial x} x^{k} \lambda \frac{\partial T}{\partial x} +$$
(3)

(2)

$$+\frac{1}{x^{k}}\frac{\partial}{\partial x}\left(x^{k}\rho T\sum_{i=1}^{N}D_{i}c_{pi}\frac{\partial Y_{i}}{\partial x}\right)$$

$$p = \rho \frac{R}{m}T = \rho RT \sum_{i=1}^{N} \frac{Y_i}{m_i} = const , \quad (4)$$

The system of equations for multi-component fluid $(0 < x < x_W)$ looks as follows:

$$\frac{\partial \hat{\rho}}{\partial t} + \frac{1}{x^k} \frac{\partial \hat{\rho} v x^k}{\partial x} = 0, \qquad (5)$$

$$\frac{\partial \hat{\rho} Y_i}{\partial t} + \frac{1}{x^k} \frac{\partial \hat{\rho} Y_i v x^k}{\partial x} = \frac{1}{x^k} \frac{\partial}{\partial x} x^k \hat{\rho} D_i \frac{\partial Y_i}{\partial x}, \quad (6)$$

$$i = 1, \dots N$$

$$\frac{\partial \widehat{\rho} \widehat{h}}{\partial t} + \frac{1}{x^k} \frac{\partial \widehat{\rho} v \widehat{h} x^k}{\partial x} = \frac{1}{x^k} \frac{\partial}{\partial x} x^k \lambda \frac{\partial T}{\partial x}, \qquad (7)$$

where ρ is density of the mixture; v - velocity; Y_i mass concentration of the *i*-th component; m_i - its molar mass; D_i - diffusion coefficient; λ - heat conductivity; T - temperature; $h = c_p T = \sum_{i=1}^{N} c_{pi} Y_i T$ specific enthalpy of the mixture; c_{pi} - specific heat capacity of the component at constant pressure; k=0,1,2 correspond to the cases of plane, cylindrical and spherical symmetry respectively. The specific volume for fluid mixture $1/\hat{\rho} = \sum_{i=1}^{N} Y_i / \rho_i + L\Delta V_x$, and the specific enthalpy is $\hat{h} = \sum_{i=1}^{N} h_i Y_i + L\Delta h_x$,

where $L\Delta V_x, L\Delta h_x$ are specific extra volume and extra enthalpy for the solution.

The boundary conditions at phase interface $x = x_w$ are:

$$(\rho v)_g = (\rho v)_l = \dot{m}, \qquad (8)$$

$$(\rho v Y_i)_g - (\rho D_i \frac{\partial Y_i}{\partial x})_g = (\rho v Y_i)_l - (\rho D_i \frac{\partial Y_i}{\partial x})_l = \dot{m}_i$$
$$\sum_{i=1}^N h_{Li} \dot{m}_i = \left(\lambda \frac{\partial T}{\partial x}\right)_g + \left(\lambda \frac{\partial T}{\partial x}\right)_l + \dot{m} L \Delta h_x,$$

where subscripts g,l denote the values of parameters in gas and liquid phases respectively; h_{Li} is the specific enthalpy of phase transition. The modified Hertz-Knudsen equation for non-equilibrium evaporation will serve as an additional boundary condition:

$$p_{i} = p_{e}X_{i} = p_{i}^{*}(T_{W}) - \frac{1}{\delta_{i}}\sqrt{\frac{2\pi RT}{m_{i}}}\dot{m}_{i},$$
 (9)

where X_i is the molar concentration of the *i*-th component; p_i is the partial pressure of the *i*-th component above the interface; δ_i is the accommodation coefficient; $p_{i}^{*}(T_{w})$ is the equilibrium vapor pressure for the *i*-th component at a temperature T_W , which could be determined from the Clausius-Clapeyron equation or simplified its solutions:

$$\frac{p_0}{p_i^*} = \exp\frac{h_{Li}m_i}{R} \left(\frac{1}{T_W} - \frac{1}{T_0}\right).$$
 (10)

The boundary conditions at infinity $(x \rightarrow \infty)$ are the following:

$$T = T_e, Y_i = Y_{ie}, i = 1, \dots N .$$
(11)

The boundary conditions in the center $(x \rightarrow 0)$ are:

$$\frac{\partial T}{\partial x} = 0, \frac{\partial Y_i}{\partial x} = 0, i = 1, \dots N .$$
(12)

To solve the system (1)-(7) along with the boundary conditions (11)-(12) one needs to pose initial distributions for temperature and concentrations, which would not contradict the boundary conditions.

3 Unsteady-state solution

Numerical simulations of evaporation of droplets in the atmosphere being the mixture of perfect gases was undertaken based on the mathematical model (1)-(12). Investigations of a number of substances showed that the essentially unsteady-state stage at the beginning of the process comes to a quasi-steady stage. The solutions based on an equilibrium and non-equilibrium evaporation models differ for both stages of the process. On decreasing the radius of droplets the difference between equilibrium and nonequilibrium models increases, which agrees qualitatively with the conclusions derived from experiments [4].



Fig. 1. Mass flux versus time variation for evaporation of freon-11 droplet $d=0.5 \mu m$ for different initial concentrations Y_{e} of freon-11 in the atmosphere.

The results of numerical modelling of freon-11 droplets evaporation under the condition of maintaining constant radius of droplet bv introducing a necessary source term in the center are shown in Fig.1 in the form of the plots of mass flux from a unit surface versus time. Accommodation coefficient was determined based on the experimental results presented in [12]. As it is seen from Fig.1 the process comes to a quasi-steady regime within several seconds. The results of numerical modelling for freon droplets of different radii showed that the smaller was the radius - the faster evaporation process came to a quasi-steady regime.

The sensitivity of results on non-equilibrium evaporation to variation of droplets radii and other parameters brought us to the necessity to search for a universal dimensionless parameter, which could characterize the deviation of the evaporation process from an equilibrium one. To obtain an analytical solution we regard the simplified problem of a steadystate evaporation of a single droplet under nonequilibrium conditions.

4 Steady-state solution for nonequilibrium evaporation

Regard the steady-state problem of nonequilibrium evaporation of uni-component uniformly heated liquid droplet of a constant radius. The governing system of equations takes the form:

• $x > x_W$

$$\frac{d}{dx}(\rho v x^2) = 0, \qquad (13)$$

$$\frac{d}{dx}(\rho Y_i v x^2) = \frac{d}{dx}(x^2 \rho D_i \frac{dY_i}{dx}), \quad (14)$$
$$i = 1, \dots N$$

$$\frac{d}{dx}(\rho v h x^2) = \frac{d}{dx}(x^2 \lambda \frac{\partial T}{\partial x}) + \frac{d}{\partial x}(x^2 \sigma T \sum_{i=1}^{N} D_i c_i \frac{dY_i}{\partial x})$$
(15)

$$+\frac{1}{dx}\left(x^{2}\rho T\sum_{i=1}^{N}D_{i}c_{pi}\frac{1}{dx}\right)$$

$$R_{T}=R^{T}\sum_{i=1}^{N}Y_{i}$$
(10)

$$p = \rho \frac{R}{m} T = \rho RT \sum_{i=1}^{n} \frac{T_i}{m_i} = const , \quad (16)$$

• $0 < x < x_W$

$$Y_{Nl} = 1, Y_{il} = 0, i = 1, ..., N - 1, T = T_W \quad (17)$$

• $x = x_W$

$$(\rho v)_g = (\rho v)_l = \dot{m} , \qquad (18)$$

$$\dot{m}Y_{ig} = (\rho D_i \frac{dY_i}{dx})_g, \qquad (19)$$

$$\dot{m}(Y_{Ng}-1) = (\rho D_N \frac{dY_N}{dx})_g,$$
 (20)

$$h_L \dot{m} = \left(\lambda \frac{dT}{dx}\right)_g,\tag{21}$$

$$p_e X_i = p_i^*(T_W) - \Delta p_i, \qquad (22)$$

where $\Delta p_i = \frac{1}{\delta_i} \sqrt{\frac{2\pi RT}{m_i}} \dot{m}_i$ for the non-equilibrium

model of phase transitions, and $\Delta p_i = 0$ - for an equilibrium one. The boundary conditions at infinity $(x \rightarrow \infty)$ take the form:

$$T = T_e, Y_i = Y_{ie}, i = 1, ..., N.$$
(23)

As it follows from the equation (13) and the boundary condition (18) the following integral is valid within the gas phase:

$$\rho v = \dot{m} \frac{x_W^2}{x}.$$
 (24)

The equation (15) could be converted into the following form

$$\frac{d}{dx}(\rho vhx^{2}) = \frac{d}{dx}\left(x^{2}\frac{\lambda}{c_{p}}\frac{\partial T}{\partial x}\right) + \frac{d}{dx}\left(x^{2}\lambda T\sum_{i=1}^{N}(Le_{i}-1)\frac{dY_{i}}{dx}\right),$$
(25)

which shows that for gases $(Le_i = \rho D_i c_{pi} / \lambda \approx 1)$ the last term in the right hand side of the equation could be neglected.

On introducing the following dimensionless variables

$$z = \frac{x}{x_W}, \chi_h = \frac{h}{h_e}, \chi_L = \frac{h_L}{h_e}, \chi_i = Y_i, Pe = \frac{\dot{m}x_W}{\rho D}$$

the system of equations (14), (15) takes the form:

$$\frac{d\chi_j}{dz} = \frac{d}{dz} \frac{z^2}{Pe} \frac{d\chi_j}{dz}, i = 1, \dots, N, h.$$
(26)

The boundary conditions for z=1 are

$$\chi_{iW} = \frac{1}{Pe} \frac{d\chi_{ig}}{dz} \bigg|_{W}, \quad \chi_{NW} - 1 = \frac{1}{Pe} \frac{d\chi_{Ng}}{dz} \bigg|_{W}, \quad (27)$$

$$\chi_L = \left(\frac{1}{Pe} \frac{d\chi_h}{dz}\right)_W,$$
 (28)

$$X_N = X_N^* - \varsigma_N \,, \tag{29}$$

where
$$\zeta_N = \frac{\Delta p_N}{p_e} = Pe \cdot I_N \cdot \sqrt{\frac{c_{pe}m_e}{c_{pN}m_N}} \chi_{hW}$$
,
$$I_N = \frac{D_{eN}}{\delta_N x_W} \sqrt{\frac{2\pi}{R_e T_e}}.$$
(30)

Due to our derivations the deviation of the process from the equilibrium one could be characterized by a dimensionless parameter I_N .

The boundary conditions at infinity $(z \rightarrow \infty)$ are:

$$\chi_h = 1, \chi_i = Y_{ie}, i = 1, \dots, N.$$
(31)

$$\xi(z) = \int_{z}^{+\infty} \frac{Pe}{z^2} dz \Longrightarrow \frac{d}{dz} = -\frac{Pe}{z^2} \frac{d}{d\xi}, \quad (32)$$

which varies in the range $(\xi_W, 0)$

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$$z \Longrightarrow \xi (1,+\infty) \Longrightarrow (\xi_W,0), \quad \xi_W = \int_1^{+\infty} \frac{Pe}{z^2} dz ,$$

one could transform the equation (26) as follows

$$\frac{d\chi_j}{d\xi} = -\frac{d^2\chi_j}{d\xi^2}.$$
 (33)

The solution of (33) is the following:

$$\chi_{j} = A_{j} + B_{j} e^{-\xi}, j = 1, ..., N, h, \quad (34)$$

where A_j, B_j can be determined from the boundary conditions.

$$Y_{jW} = Y_{je}e^{-\xi_{W}}, j = 1,..., N - 1;$$

$$Y_{NW} = 1 - (1 - Y_{Ne})e^{-\xi_{W}};$$

$$\chi_{hW} = 1 + \chi_{L}(1 - e^{\xi_{W}}).$$
(35)

Substituting (35) into the boundary condition (29) one obtains the equation determining the dimensionless evaporation rate ξ_W or the Peclet number *Pe*:

$$Y_{NW} \frac{m_{W}}{m_{N}} = X_{N}^{*}(\chi_{hW}) - Pe \cdot I_{N} \cdot \sqrt{\Gamma_{eW} \frac{m_{W}}{m_{N}} \chi_{hW}};$$

$$m_{W} = \left(\sum_{i=1}^{N} \frac{Y_{iW}}{m_{i}}\right)^{-1}; \Gamma_{eW} = \frac{\gamma_{e}(\gamma_{W}-1)}{\gamma_{W}(\gamma_{e}-1)}; \gamma = \frac{c_{p}}{c_{v}};$$

$$X_{N}^{*} = \frac{p_{0}}{p_{e}} \exp\left[\chi_{L} \frac{m_{W}}{m_{N}} \frac{\gamma_{W}}{(\gamma_{W}-1)} \left(\frac{1}{\chi_{h0}} - \frac{1}{\chi_{hW}}\right)\right].$$
 (36)

The last expression for the phase equilibrium function could be also substituted by the data from the tables on the thermophysical properties of substances [13].

The solutions (34) providing flow parameters distribution around evaporating droplet then take the form:

$$Y_{i} = Y_{ie}e^{-\zeta};$$

$$Y_{N} = 1 - (1 - Y_{Ne})e^{-\xi};$$

$$\chi_{h} = 1 - \chi_{L}e^{\zeta_{W}}(1 - e^{-\zeta});$$

where ξ_W could be obtained from solution (35), (36). To determine the $\xi(z)$ function analytically one could assume that $\rho D = const / \rho$, which could be a good approximation for constant pressure problems [6]. Then the equalities are valid

$$\frac{\rho D}{\rho_e D_e} = \frac{\rho_e}{\rho} = \frac{h}{h_e} \frac{Rc_{pe}}{c_p R_e} = \chi_h \frac{\gamma - 1}{\gamma} \frac{\gamma_e}{\gamma_e - 1},$$

which neglecting the γ variation in space provide the following formula

$$\frac{\rho D}{\rho_e D_e} = \chi_h \,. \tag{37}$$

Formula (37) allows to express Peclet number in the following way

$$Pe = \frac{\dot{m}x_W}{\rho D} = \frac{\dot{m}x_W}{\rho_e D_e} \frac{\rho_e D_e}{\rho D} = \frac{Pe_e}{\chi_h},$$

which being substituted into (32) provides an equation:

$$\frac{d\xi}{dz} = -\frac{Pe_e}{\chi_h z^2}, \quad \xi(1) = \xi_W. \quad (38)$$

Substituting the solution $\chi_h(\xi)$ and integrating the equation (38) one obtains analytical formula providing the link between *z* and ξ :

$$z = \frac{Pe_e}{\xi + \chi_L e^{\xi_W} (1 - \xi - e^{-\xi})},$$

$$Pe_e = (1 - \chi_L e^{\xi_W}) \xi_W + \chi_L (e^{\xi_W} - 1) =$$

$$= \xi_W - \chi_L \xi_W^2 / 2 - O(\xi_W^3).$$

The set of equations (35), (36) allows to determine the unknown Peclet number in non-equilibrium phase transitions as a function of the following parameters:

$$\frac{m_i}{m_N}, \chi_L, \frac{p_0}{p_e}, Y_{ie}, \Gamma_{eW}, X_N^*(\chi_{hW}), \quad I_N$$

The first six of them characterize the media and the boundary conditions. The seventh parameter I_N characterizes the deviation of the system from the equilibrium state. The value I_N is zero for the case of equilibrium, and it grows remaining positive($I_N > 0$) with the increase of the deviation from the equilibrium.

5 The role of non-equilibrium effects

To investigate the influence of non-equilibrium effects in phase transitions Peclet numbers for droplets evaporation were determined for different values of I_N parameter (Fig.2). The values of the other governing parameters were assumed to be the following

$$\frac{m_i}{m_N} = 4.75; \ \chi_L = 0.54; \ \frac{p_0}{p_e} = 1; \ Y_{Ne} = 0.1, \ \Gamma_{eW} = 1, \ \chi_{h0} = 0.9$$

The Fig. 2 gives the plots of Peclet numbers, obtained within the frames of non-equilibrium (Pe_{ne}) and quasi-equilibrium (Pe_{eq}) models. The value of $|\delta Pe|$ is also plotted in Fig. 2 to characterize the relative deviation of Peclet number from its equilibrium value $(\delta Pe = (Pe_{ne} - Pe_{eq})/Pe_{eq})$. The Peclet number for non-equilibrium evaporation could be determined by the formula

$$\frac{Pe}{Pe_{eq}} = \Psi_{S}(I_{N}, \frac{m_{i}}{m_{N}}, \chi_{L}, \frac{p_{0}}{p_{e}}, Y_{ie}, \Gamma_{eW}, X_{N}^{*}(\chi_{hW})), (39)$$

wherein the value of the Ψ_s – *factor* as a function of I_N parameter for the chosen values of other governing parameters is shown in Fig. 2. The equilibrium Peclet number as obtained from the guasisteady solution has the following form:

$$Pe_{eq} = \log_e \left(1 + \frac{c_{pe}T_e - c_{pW}T_W}{h_L} \right). \quad (40)$$

The results of comparison of non-equilibrium and quasi-equilibrium Peclet numbers for droplet evaporation show that the rate of evaporation strongly depends on the value of the parameter I_N . On increasing I_N Peclet number decreases rapidly as compared with the equilibrium evaporation; the deviations of the evaporation rates from those predicted by the equilibrium model turn to be larger and larger. For $I_N < 1$ the divergence of equilibrium and non-equilibrium solutions $|\delta Pe|$ does not exceed 4%, for $I_N = 10$ the deviations increases up to 25%, for $I_N > 100$ the deviation surpasses an order of magnitude. Thus in order to have adequate estimates for small droplets evaporation rate one needs to use the nonequilibrium model for $I_N > 1$.



Fig. 2. Peclet number variation versus I_N parameter for equilibrium and non-equilibrium evaporation.

To have adequate data for small droplets $(I_N > 1)$ evaporation rates it is necessary to use the non-equilibrium model.

6 Quasi-steady solution for a single droplet evaporation

of Investigations droplet unsteady-state evaporation show that for small droplets flow parameters in gas phase reach their quasi-steady values very quickly. Thus the quasi-steady approximation regarding droplet evaporation could be used, which means that its radius decreases in time much slower than gas flow parameters come to a steady state. Thus it is assumed, that the rate of droplet evaporation for each time moment could be determined based on the obtained steady non-equilibrium solution. Then the decrease of the droplet radius can be determined by the following formula:

$$\frac{dx_w}{dt} = -\frac{\dot{m}}{\rho_{\rm vir}}.$$
(41)

On introducing dimensionless variables

$$r = \frac{x_W}{x_{W0}}, \quad \tau = \frac{t}{t_0}, \quad t_0 = \frac{\rho_{NI} x_{W0}^2}{2\rho_e D_e P e_{eq}} \quad (42)$$

equation (41) takes the form

$$\frac{dr}{d\tau} = -\frac{\dot{m}t_0}{\rho_{NI}x_{W0}} = -\frac{Pe}{r}\frac{\rho_e D_e t_0}{\rho_{NL}x_{W0}^2},\qquad(43)$$

where x_{W0} is the initial radius, t_0 is the characteristic time for droplet evaporation under equilibrium conditions.



Fig. 3. Droplet dimensionless radius variation versus time for different initial values of I_{Ne} parameter characterizing deviation of the system from the equilibrium.

Fig. 3 presents the comparison of $r(\tau)$ functions (trajectories) obtained for different initial

values of *I* parameter (
$$I_e = \frac{I}{\delta_N}$$

 $\frac{D_e}{\sqrt{x_{W0}}} \sqrt{\frac{2\pi}{R_e T_e}}$ characterizing initial deviation of the system from an equilibrium state. Increasing the initial deviation from the equilibrium brings to a decrease of the slope of the curve and increases the actual time for

droplet evaporation. Fig. 4 illustrates the dimensionless mass flux from the surface of a droplet

$$\dot{\mu} = \frac{\dot{m}(x_w)x_{w_0}}{\rho_e D_e} = \frac{Pe(r(\tau))}{r(\tau)}$$
(44)

as a function of dimensionless radius r (Fig. 4a) and dimensionless time τ (Fig. 4b). It is seen from the figures that the values of mass flux provided by nonequilibrium model have always a final limit on $r \rightarrow 0$. That is the major qualitative difference between the non-equilibrium model and equilibrium ones. The mass flux reaches its maximum by the end of evaporation $(r \rightarrow 0)$. The maximal values of the mass flux decrease on increasing the deviation from the equilibrium (Increasing I). The time for droplet evaporation increases essentially on increasing the deviation from the equilibrium.

7 Conclusions

The results of investigation showed that the difference between quasiequilibrium and non-equilibrium solutions increase on decreasing droplet radius, decreasing accommodation coefficient and on increasing diffusion coefficient.

The dimensionless criterion was developed characterizing the deviation of small liquid droplet evaporation process from the equilibrium, which permits to determine applicability limits for equilibrium models.

It was demonstrated that non-equilibrium models more adequately describe final stages of droplet evaporation. Those models are free from the common drawbacks of all quasi-equilibrium models, because non-equilibrium approach allows to avoid non-physical growth of the evaporation rate on decreasing droplet radius.

Accounting for non-equilibrium effects produces an influence on the integral characteristics of droplet evaporation process. The lifetime for single evaporating droplet could be several times longer under non-equilibrium conditions as compared with equilibrium ones.

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Fig. 4. Dimensionless mass flux variation versus dimensionless radius (a) and time (b) for different initial values of non-equilibrium parameter I_{Ne}