Stability of multiphase liquid media

Anatoliy Pavlenko 1,2,3, Hanna Koslak 1 and Anna Słowak 1
1 Kielce University of Technology, Faculty of Environmental, Geomatic and Energy Engineering, Poland;
2 Kielce ul. Jarzębinowa 6/57, Poland.
3 Email: apavlenko@tu.kielce.pl

Abstract. The processes of spontaneous boiling of superheated liquids under conditions when the liquid appears in a no equilibrium metastable state are one of the greatest interest, since studying the kinetics of a metastable state dysfunction can contribute to the creation of new high-intensity technologies in the creation of emulsions. In this paper it has been studied the destruction process of oil-film emulsions of the water-oil type with account of both the thermal and dynamic aspects of the problem.

1. Nomenclature

\( p \) - pressure, Pa; \( T \) - temperature, \(^\circ\)C; \( R \) - radius, m; \( w \) - where, m/s; \( \tau \) - time, s; \( \sigma \) - the surface tension at the boundary (interface) of the oil-steam, N/m; \( \mu_{oil} \) - the coefficient of the dynamic viscosity of the oil, Pa\(\cdot\)s; \( \rho_{oil} \) - is the oil density, kg/m\(^3\); \( T_v \) - vapor temperature, K (Kelvin); \( c_v \) - specific heat capacitance of vapor, J/(kg\(\cdot\)K); \( m_v \) - vapor mass, kg; \( V_v \) - vapor volume, m\(^3\); \( Q \) - amount of heat, W; \( A \) - square meter, m\(^2\); \( \alpha_{ev} \) - coefficient of evaporation; \( B \) - absolute gas constant, J/(mole\(\cdot\)K); \( \rho_w \) - water density, kg/m\(^3\); \( \Delta m \) - mass-flux density, kg/(m\(^2\)s); \( \lambda_w \) - water thermal conductivity, W/(m\(\cdot\)K); \( L(T) \) - heat of vaporization, J/kg; \( T_r \) - reduced temperature; \( M_v \) - molecular weight of vapor, kg/mole; \( \alpha_w \) - temperature conductivity coefficient of oil and water, m/s\(^2\);

\[ B = 8,31441 \pm 0,00026 \] - universal gas constant, J/(mol\(\cdot\)K) index number; \( R_3 \) - boundary (radius) of the volume of water; \( R_4 \) - the boundary (radius) of the vapor region; \( R_5 \) - radius oil shell

2. Introduction

Most of the carried researches studies the dynamics of growth of vapour bubbles which occur either in the liquid or on a superheated surface [1-3]. A peculiarity of emulsion boiling media is that formation of a vapour phase occurs on the surface of liquid interface if one liquid (for example, oil) is a superficially active substance or there is a superficially active substance in the emulsion (in multicomponent emulsions). The process is based on a decrease in tension at the phase boundary between the dispersive medium (water) and the dispersion phase (oil) [1, 3-6]

Experimental data [1, 3] indicate that the process of a new phase (vapour) formation is initiated by an internal thermal impact on the surface of the oil particles. Thus, for example the preheated emulsion will be in the super saturation state of the thermo labile-water phase (the water is superheated against the saturation temperature at a given pressure), with a sudden pressure decay, i.e. it will contain an excessive amount of heat [7-10]. This heat excess is expended on the work of vapour layer formation and further vaporization. The effect of oil films is ambiguous: on the one hand, when
heated, they play the role of thermal resistance, thus, slowing down the heating process; on the other hand, they are the heat carriers which can be used for the formation and growth of the vapour phase and also they are superficially active substance, which contributes to achieving lower water super saturation temperatures, i.e. early boiling [5]. The consideration of oil thermal effect plays an important role when regarding heat and mass transfer processes during the boiling of emulsion media. Theoretical description and analysis of heat and mass exchange processes in emulsions, associated with the presence of oil and oil films, allows us to study more in detail the process itself, as well as to offer optimal parameters and characteristics for the processes of obtaining and processing a fine emulsion. As an example, we can consider the proposed homogenization technology, the essence of which is as follows. A pre-heated coarse emulsion (average droplet diameter of 50 μm) to a temperature higher than the boiling point of the low-boiling liquid (for example, water in the water-oil emulsion) under pressure that is higher than the saturation pressure is sprayed in the thermal chamber. With a sharp decrease in pressure, the microdroplets of water evaporate. We thus consider a drop of emulsion that flies in a thermal chamber and is a core of water covered with oil. This drop is surrounded by air. The thermodynamic processes that occur when the coarse emulsion is sprayed and when the temperature and pressure in the emulsion are equalized lead to a substantial increase in the dispersity of the emulsion. In our experiments, we obtained the average size of a thermally treated drop of water in an emulsion - 5-10 microns. Known mechanical methods of homogenization do not allow to achieve this level of homogenization. The theoretical problem considered in this paper makes it possible to determine, using the mathematical model, the main energy and dynamic parameters of this technology.

3. Mathematical framework

The research objective is to consider a drop of emulsion (Figure 1), which consists of a drop of water covered with a layer of oil at the moment of emerged steam layer. At the initial time this system is under some pressure (for example, \( p_0 > p_{\text{atm}} \)) and at a temperature (for example, \( T_0 = T_s(p_0) \)). At some point of time the system is in the zone of low pressure \( p_4 \) (\( p_4 < p_0 \)). Thus, the growth of steam volume and water vaporization will occur due to the fact that the water is superheated relative to the pressure \( p_4 \). The movement of the oil-vapor interface will occur under the influence of the pressure difference inside the vapor volume \( p_a \) and pressure on the surface of the oil layer \( p_4 \).

In this case the whole system will be heated by air with the temperature \( T_{\text{air}} \).

![Figure 1](image.png)

**Figure 1.** The calculated model of vapor phase growth (\( R_2 \) is the radius of the water-vapor interface, \( R_3 \) is the radius of the oil-vapor interface, and \( R_4 \) is the radius of the oil-air interface).

Most methods for calculating the radial motion of a liquid at the interface with a spherical enclosure use the Rayleigh equation [2,4].

Considering the absence of any gas in the vapor volume, i.e. The vapor region consists only of water vapor and contains no vaporized oil, taking into account the capillary forces and shear stresses, the equation for the speed of the radial motion of the oil-vapor interface with the oil film of outer \( R_4 \) radius becomes:
\[
\frac{dw_3}{d\tau} = \frac{p_3 - p_4 - 1.5\rho_{oil}w_3^2}{\rho_{oil}R_3} + \frac{w_3^2[1 + R_3/R_4 + (R_3/R_4)^2]}{2R_3}.
\]

where \( w_3 \) is the speed of the oil-vapor interface (boundary), m/s; \( \tau \) – time, s; \( p_3 \) is the pressure at the oil-vapor interface, Pa;

As can be seen from equation (1), the growth rate of a bubble in a bounded volume always exceeds its growth rate in an infinite volume by the amount determined by the second term on the right side of the given equation, which is always positive. It is clear that as \( R \to \infty \), equation (1) can be written as:

\[
\frac{dw_3}{d\tau} = \frac{p_3 - p_4 - 1.5\rho_{oil}w_3^2}{\rho_{oil}R_3} \quad \text{(1a)}
\]

Equation (1a) is the Rayleigh equation for a vapor bubble that is in infinite volume;

\[
p_3 = p_\infty - 2\sigma_3/R_3 - 4\mu_{oil}w_3/R_3, \quad \text{(1b)}
\]

\( p_\infty \) – the pressure inside the vapor volume, Pa; \( \sigma_3 \) – the surface tension at the boundary (interface) of the oil-steam, N/m; \( \mu_{oil} \) – the coefficient of the dynamic viscosity of the oil, Pa·s; \( p_4 \) – the pressure at the oil-air interface, Pa; \( \rho_{oil} \) is the oil density, kg/m³.

Equation (1a) is part of the Rayleigh equation (1) and characterizes the pressure that occurs in the vapor region and acts on the inner surface of the oil film at the interface between water vapor and oil. This is an important parameter, since it allows you to determine the force acting on the film and the level of dynamic effects that arise. From a technological point of view, this parameter means whether the structure of the emulsion, the fragment of which is shown in Figure 1, will be preserved or it will be destroyed. In this case, we can determine the level of energy impact on the structure, sufficient for its destruction (grinding). This is the proposed technology of homogenization of various liquid media. Thus, equation (1) is the Rayleigh-Plessse equation for a bounded spherical volume.

Without phase transitions at the interface, we can write:

\[
\frac{dR_3}{d\tau} = w_3. \quad \text{(2)}
\]

Equation (2) allows to determine the speed of movement of the interface between the water vapor and oil phase. This parameter is also necessary for estimating the stability of the emulsion to determine the critical radius of the interface with increasing vapor volume.

To determine the temperature inside the vapor volume, we can use the first law of thermodynamics, which after differentiation takes the form:

\[
\frac{dT_n}{d\tau} = \frac{1}{c_n m_n} \left[ Q - c_n T_n \Delta m A_2 - p_n \frac{dV_n}{d\tau} \right], \quad \text{(3)}
\]

where \( T_n \) – vapor temperature, K (Kelvin); \( c_n \) – specific heat capacitance of vapor, J/(kg·K); \( m_n \) – vapor mass, kg; \( V_n \) – vapor volume, m³; \( Q \) is the amount of heat, W; \( A_2 = 4\pi R_2^2 \) – square meter, m².

We consider mass and heat transfer through the water-vapor interface in terms of the molecular-kinetic theory, which allows to estimate more accurately the kinetics of phase transitions on the liquid-gas surface [2].

We assume that the temperature of the vapor volume \( T_n \) is not equal to the temperature of the water-vapor interface \( T_2 \), i.e. the process will occur if there is a certain temperature jump at the interface. In this case the mass transfer intensity at the phase transition and heat-flux density between the vapor and the water-vapor interface [2-4] are equal to:

\[
\Delta m = 0.25\alpha_{m}\left[\rho_2 U_2(T_2) - \rho_n U_n(T_n)\right], \quad \text{(4)}
\]
\[ q = 0.25 \rho_n c_n U_n (T_2 - T_{n}) + \Delta m c_n T_2, \]  
(5)

where \( U(T) = \frac{8BT}{\pi M_n} \) - arithmetic mean rate of thermal motion of vapor molecules at a temperature \( T \), m/s; \( \alpha_n \) - coefficient of evaporation; \( B \) – absolute gas constant, J/(moleꞏK); \( \rho_2 \) – water density, kg/m\(^3\); \( \Delta m \) – mass-flux density, kg/(m\(^2\)s).

Since the water cooling or its heating is caused by heat exchange with vapor and transfer of latent heat of vaporization, then taking into account the heat flux resulting from the temperature difference of the water drop center \( T_1 \) and the temperature of the water-vapor interface \( T_2 \) \([2, 7]\) we can write:

\[ \frac{2 \lambda_w}{R_2} (T_1 - T_2) = q + \Delta m L(T_2), \]  
(6)

where \( \lambda_w \) – water thermal conductivity, W/(mꞏK); \( L(T) \) – heat of vaporization, J/kg.

The solution of this equation allows us to determine the unknown temperature \( T_2 \).

Equation (3) also includes unknown quantities: the vapor mass \( m_n \), its specific heat capacitance \( c_n \), pressure \( p_n \) and volume \( V_n \), as well as the quantity of heat \( Q \).

The vapor mass depends on the mass flow and is determined by:

\[ \frac{dm_n}{d\tau} = \Delta m A_2. \]  
(7)

The volume of the vapor layer is determined by the difference between the volume \( \frac{4}{3} \pi R_3^3 \) and water volume \( \frac{4}{3} \pi R_2^3 \):

\[ V_n = \frac{4}{3} \pi [R_3^3(\tau) - R_2^3(\tau)]. \]  
(8)

We consider the specific heat capacitance to be constant.

Heat \( Q \) consists of heat which comes from the liquid (water) or comes into it, as well as from the heat that comes from the oil film:

\[ Q = qA_2 + \frac{4\pi \lambda_{oil}}{1} \left\{ \frac{1}{R_3(\tau)} - \frac{1}{R_2(\tau)} \right\}, \]  
(9)

where \( \lambda_{oil} \) – oil heat conductivity, W/(mꞏK).

The vapor pressure depends on both its temperature \( T_n \) and density \( \rho_n \).

For calculation we take the equation of Redlich-Kwong in Wilson’s modification \([11]\), which gives the smallest relative error in the definition \( p_n = f(T_n, \rho_n) \). Then:

\[ p_n = \frac{BT_\nu \rho_n}{M_n} \left[ \frac{1}{1 - b\rho_n} \cdot \frac{\delta_a \cdot b}{M_n + b} + A \right], \]  
(10)

where \( A = 1 + (1.57 + 1.62 \theta)(T_{\nu}^{-1} - 1) \); \( \theta \) - acentric factor; \( T_\nu = T/T_{cr} \) - reduced temperature; \( M_n \) - molecular weight of vapor, kg/mole; \( b = \frac{\delta_b BT_{cr}}{p_{cr}} \); \( \delta_a = 0.4274802327 \); \( \delta_b = 0.08664035 \); \( T_{cr}, p_{cr} \) - thermal critical point and pressure.
Assuming homogeneity \(p_n\) and \(T_n\) in the vapor volume, as a consequence, the density of the vapor phase in the vapor volume \(\rho_n\) is homogeneous, i.e. \(d\rho_n / dR_n = 0\), the function of changing of the vapor density in time can be represented in the following way:

\[
\frac{d\rho_n}{d\tau} = \frac{1}{V_n} \left( \frac{dm_n}{d\tau} - \rho_n \frac{dV_n}{d\tau} \right).
\]

(11)

Substituting the obtained values \(\rho_n\) from (11) and \(T_n\) from (3) into (10) we can determine the vapor pressure at any time.

The thermal side of the problem is to write the energy equation for oil and water, as well as boundary conditions. Equations of thermal conductivity for oil and water are:

\[
\frac{\partial T(r, \tau)}{\partial \tau} = a_oil \left( \frac{\partial^2 T(r, \tau)}{\partial r^2} + \frac{2}{r} \frac{\partial T(r, \tau)}{\partial r} \right) - w_r \frac{\partial T(r, \tau)}{\partial \tau},
\]

\[
-w_r \frac{\partial T(r, \tau)}{\partial \tau},
\]

\[
\frac{\partial T(r, \tau)}{\partial \tau} = a_w \left( \frac{\partial^2 T(r, \tau)}{\partial r^2} + \frac{2}{r} \frac{\partial T(r, \tau)}{\partial r} \right),
\]

\[
\tau > 0, \quad 0 < r < R_3, 
\]

(12)

where \(a_{oil}\), \(a_w\) – temperature conductivity coefficient of oil and water, m/s².

Initial data:

\[
T(r, 0) = T_0, \quad 0 < r < R_4.
\]

(14)

Boundary condition with account of the equation (6):

\[-\lambda_{oil} \frac{\partial T}{\partial r} \bigg|_{r=R_a} = \alpha (T_{air} - T_a); \]

\[
T_3(\tau) = T_n; \quad -\lambda_w \frac{\partial T}{\partial r} \bigg|_{r=0} = 0,
\]

(15)

where heat-exchange coefficient according to [5] is equal:

\[
\alpha = \lambda_{air} / R_4.
\]

(16)

\(\lambda_{air}\) – thermal conductivity of air, W/(m·K).

The speed of the water-vapor interface can be found from the equation:

\[
w_2 = \frac{dR_2}{d\tau} = \frac{\Delta m}{\rho_2(T_2)}.
\]

(17)

The thickness of the vapor layer is:

\[
\delta_n = R_2(T) - R_3(T).
\]

(18)

The thickness of the oil film is determined by:

\[
\delta_{oil} = R_4(T) - R_3(T).
\]

(19)

The criterion for the final calculation is the critical thickness of the oil film at which it collapses [5]. Equations (1) - (19) must be supplemented with equations of the dependences of thermo physical parameters on temperature.

4. Numerical modeling

Presentation of the equations of the dynamics of the change in the volume of the vapor region in the form of ordinary differential equations from one variable substantially simplifies the numerical
solution procedure. All the parameters defined depend only on time. Simple algebraic equations are used to estimate the speed, acceleration, and pressure in a fluid.

The solution of the system of differential equations was carried out by the Runge-Kutta method of the fifth order using the Pascal language. To do this, the Fortran subroutine RKF45, developed specifically for solving the Cauchy problem for ordinary differential equations, was used [2]. This subroutine is one of the best universal implementations of the Runge-Kutta methods, which has increased accuracy and flexibility.

Since the basic programs for solving problems in this study are written in the Pascal language, we have translated the RKF45 subroutine from Fortran, for which it was developed, into the Pascal language for coordination with work programs. The subroutine works with automatic step selection and automatic error control and allows you to set the desired level of accuracy. The local error in the calculations does not exceed $10^{-6}$, which is quite acceptable for such, albeit effective, but qualitatively simple subroutine.

The use of the Runge-Kutta method and the use of an effective subroutine for its implementation make it possible to perform calculations on a PC with a minimal amount of computer time. In addition, the chosen method makes it easy to operate with computer programs.

5. Results and discussion

To solve the equations of the mathematical model, we used the integral method [2]. The calculations are performed for an emulsion drop which consists of a water drop of $R_2 = 99 \, \mu m$, a vapor layer of 1 $\mu m$ thickness and an oil film of $\delta_{oil} = 15 \, \mu m$ thickness. At the initial time this system has the same temperature equal to $T_0 = 180 ^\circ C$, the vapor is saturated, i.e. is at atmospheric pressure of $p_0 \approx 10 \, atm$. At some point in time this system is in the zone of low $p = 1 \, atm$ and at the same time it is heated by air at a temperature of $T_{air} = 300 ^\circ C$.

The calculations were carried out using the method of finite differences with explicit selection of variables. The results are shown in Figure 2-8.

On Figure 2-8 can be seen the radii oscillations of interfaces, temperatures, and vapor pressures, which corresponds to physics of the real process. The time up to the oil film destruction to is $\approx 1.23 \cdot 10^{-2} \, s$. Figure 2 shows the time variation of interfaces temperatures of the water-vapor $T_2$, oil-air $T_a$, vapor $T_n$, and drop-water centers $T_i$. Obvious temperature oscillations are seen $T_2$, $T_n$, $T_i$ in the time interval $\tau < 3.7 \cdot 10^{-3} \, s$, where the vapor temperature $T_n$ is higher than the temperature of water-vapor interface which indicates its superheated state. This overheating is a consequence of the additional amount of heat coming from the oil film as a result of its heat exchange with the surrounding air. The highest superheat is observed during the time period $\tau \approx (0.05 \div 0.15) \cdot 10^{-3} \, s$, which in its turn leads to the increase in the mass flux according to Figure 6. In general, there is a consistency in the change of vapor and mass flow temperature, which indicates a significant role of the heat flux from superheated vapor to the water-vapor interface as a result of its heating from the oil film. The decrease $T_n$ leads to the corresponding decrease of mass flow, which, starting at time $\tau > 4 \cdot 10^{-3} \, s$, decreases steadily. As can be seen in Figure 2, at the initial moment of vapor enclosure expansion the temperature $T_{air}$ increases insignificantly, after which its sharp decrease occurs practically to the temperature $T_n$. The first peak in vapor temperature rise ($\tau \approx 0.02 \cdot 10^{-3} \, s$) is coherent with a sharp decrease in the radius of the oil-vapor interface (Figure 4) and, as a consequence, an increase in the oil film thickness (Figure 5). Accordingly, the vapor pressure increases Figure 3. Oscillations of the temperature of the water-vapor interface are a consequence of the corresponding oscillations of the mass flow, the vapor temperature and its density. As shown in Figure 2-8, the resulting oscillations of temperatures, pressures, interfaces radii, and motion speed of these interfaces are the decaying oscillations. So the temperatures $T_2$, and $T_n$ tend to certain values, respectively, $T_0 \approx 100 ^\circ C$ and
≈102 °C, and after \( \tau \approx 4 \cdot 10^{-3} \) s these fluctuations are not very noticeable. The vapor pressure reflects the combined effects of temperature and vapor density on the further development of the process, and, as can be seen from Figure 3, at the beginning of vapor volume growth it is observed a significant reduction in pressure \( p_2 \) to a pressure which is lower \( p_4 \), as a result of which the movement of the oil-vapor interface is carried out to the reduction of vapor volume (Figure 4), as shown in Figure 8 by negative values of the motion speed \( \omega_3 \). This decrease in pressure \( p_2 \) indicates that the mass flow with the heat flow from the oil to the vapor is not able to compensate for the dynamic effect of expansion of the vapor volume, which leads to a decrease in temperature and vapor density. The vapor pressure tends to a certain value, which is slightly higher than the environmental pressure \( p_4 \). After the setting of this specific pressure (with small oscillations), it can be seen that the change of \( R_2 \) and \( R_3 \) radii, the oil film thickness \( \delta_n \) and the vapor layer \( \delta_n \) (Figures 4 and 5) occurs steadily, and the speed of the oil-vapor interface (Figure 8) tends to some constant value.

\[ T_2, T_1, T_5 \]

**Figure 2.** Changes of interfaces temperatures and water center in time.

\[ P_2 \times 10^3 \, \text{Pa}, \text{Fig.} \]

**Figure 3.** Change of vapor pressure in time.

\[ R_2, R_3, \delta_n, \mu m \]

**Figure 4.** Change of radii of the interfaces between oil-vapor, water-vapor and the thickness of the vapor layer in time.

\[ \delta_{oil}, \mu m \]

**Figure 5.** Dependence of oil film thickness on time.
Figure 6. Change in time of mass flow.

Figure 7. Dependence of speed motion of the water-vapor interface on time.

Figure 8. Speed motion change of the oil-vapor interface in time.

The speed of the water-vapor interface \( w_2 \) (Figure 7) has the same time behaviour with the mass flow (Figure 6), which indicates an insignificant role of changes in the water density with temperature on the process nature. The temperature of the center of the water drop \( T_1 \) (Figure 2) for \( \approx 1 \text{ ms} \) remains constant and equals to the initial temperature of the system (the inertial period), after which it decreases steadily, asymptotically approaching to \( T_2 \) temperature.

6. Conclusions
The consideration of dynamic forces plays a decisive role in the growth of the vapor layer and allows to study this process more accurately and in details. Using only the thermal side of the problem as for the growth of the vapor phase \([5]\), one could only assume and initially set the temperature of the water-vapor interface, but using the molecular-kinetic theory and taking dynamic forces into account, it is possible to determine the temperature at each point of the calculation. The principal difference
from the thermal problem, in which it was assumed that the vapor is always saturated and is at the same temperature throughout the growth interval of the vapor phase, is the change in the vapor density and its overheating. Therefore, we can conclude that it is necessary to solve both the dynamic and the thermal problems.

The most interesting point is the determination of the heat flux that comes into the system or is taken away from it, taking into account the change in Nusselt criterion and the dynamic effect from the medium, which can lead to the premature oil film destruction when the considered emulsion drop is in motion in the medium.

The proposed theoretical approach to the consideration of this problem, as well as the proposed mathematical model, allows one to modify the problem for various physical models, for example, a drop of water can be surrounded by an oil film (or several films of various liquids) and all this is surrounded by water. There may be other modifications of the object in question.

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