Molecular dynamic approach to the study of the intense heat and mass transfer processes on the vapor-liquid interface

V Yu Levashov¹, P K Kamenov²

¹Institute of Mechanics, Moscow State University, Michurinskyi prospect 1, Moscow, 117192, Russia
²Low temperature department, National Research University Moscow Power Engineering Institute, Krasnokazarmennaya 14, Moscow 111250, Russia

e-mail:vyl69@mail.ru

Abstract. The paper is devoted to research of the heat and mass transfer processes on the vapor-liquid interface. These processes can be realized for example at metal tempering, accidents at nuclear power stations, followed by the release of the corium into the heat carrier, getting hot magma into the water during volcanic eruptions and other. In all these examples the vapor film can arise on the heated body surface. In this paper the vapor film formation process will be considered with help of molecular dynamics simulation methods. The main attention during this process modeling will be focused on the subject of the fluid and vapor interactions with the heater surface. Another direction of this work is to study of the processes inside the droplet that may take place as result of impact of the high-power laser radiation. Such impact can lead to intensive evaporation and explosive destruction of the droplet. At that the duration of heat and mass transfer processes in droplet substance is tens of femtoseconds. Thus, the methods of molecular dynamics simulation can give the possibilities describe the heat and mass transfer processes in the droplet and the vapor phase formation.

1. Introduction

There are various situations in which significantly nonequilibrium processes can be realized. Such processes can be observed, for example, during the metal hardening, in severe accidents at nuclear power plants, followed by the release of corium and molten steel into the coolant, the eruption of volcanoes with the ingress of heated magma into water, etc. In these cases the heated body immersed into subcooling liquid. As a result, a finite thickness vapor film can be formed on the hot object surface. The computational and theoretical modeling of such processes should be includes the analysis of the processes in the heater, in the vapor and in the liquid. Some approaches for these processes describing are presented in [1, 2]. At this modeling the initial vapor films thickness and formation time of vapor region are important questions that currently have no answers. Often the vapor film thickness is set arbitrarily and formation time of vapor region is assumed to be very small (much less film lifetime).

Other example of the non-equilibrium situations are the processes that can be realized as a result of high-power laser radiation absorption by a fluid or a solid droplet. A large number of experimental and theoretical works have been devoted to the study of the destruction of solids and liquid droplets under the action of laser radiation. In most publications the main attention is focused on the transmission and
interaction of radiation with droplet with taking into account optical effects. It is also noted that heat transfer mechanisms can be various depending on the droplet substance.

For example in [3] it is noted that in metals the laser radiation is absorbed by conduction electrons and as a result this energy is distributed between electrons over a time of the order of several femtoseconds and further, slowly converted to lattice vibration energy. Thus these processes should be described with taking into account the specific features of the processes of absorption of radiation and the propagation of heat in metals.

It should be noted that the characteristic features of these processes are small realization times and as a consequence, the macroscopic phenomenological approaches correctness is not justified in these cases. Therefore the main investigation method in this paper is the molecular-dynamic modeling method typical time of which is \(10^{-13}\)s. This method makes it possible to analyze fast and non-equilibrium processes.

2. Statement of the problem and solution results

2.1. Vapor liquid film

The statement of the problem for simulation of vapor film is shown in figure 1.

![Figure 1. Vapor film formation statement.](image)

The liquid film is modeled by a system of N particles located in a parallelepiped with dimensions \(L_x \times L_y \times L_z\). The interaction between i-th and j-th particles is described by the Lennard-Jones potential [4]. The argon parameters were used in the simulation. The simulation of the system by the molecular dynamics method is based on solving the second Newton’s law equations for each particle of system. It is assumed that the initial liquid temperature is \(T_0\). The liquid film is located on the heater surface directly. Heater temperature is \(T_H\).

The two different approaches were used for liquid or vapor atoms interaction with the heater surface: 1. The particles are reflected by a diffuse scheme; 2. The surface of the heater is modeled by a system of oscillating particles.

In the diffuse reflection model, the following algorithm is used. The velocity direction of particle that reflected from the heater surface is selected randomly (this velocity should be directed from the heater). The velocity magnitude is calculated from the following expression:

\[
\frac{m_i v_i^2}{2} = \frac{3}{2} k T_H
\]

where \(k\) – is the Boltzmann constant, \(T_H\) – is the heater temperature.

In a model with oscillating particles, there is no need to specify the direction and modulus of the velocity of the reflected particle. The value of the heater temperature is calculated as follows in this case. It is assumed that particles simulating a solid heater can perform harmonic oscillations. At the first stage, oscillations were considered only along the \(OZ\) axis. The velocity of a particle that has such harmonic oscillations can be represented in the following form:
\[ V(t) = V_m \cdot \sin(\omega \cdot t + \varphi) \]  
(2)

where \( V_m = A \omega \) – is maximum velocity, \( A \) – is oscillation amplitude, \( \omega \) – is oscillation frequency, \( t \) – is time, \( \varphi \) – is initial oscillation phase.

The average velocity value of the oscillating particle in the period half can be obtained from the following expression:

\[ \mathcal{V} = \frac{\int_0^{\tau/2} V_m \cdot \sin(\omega \cdot t) \cdot dt}{\tau/2} = \frac{2V_m}{\pi} = \frac{2A \cdot \omega}{\pi} \]  
(3)

where \( \tau \) – is harmonic oscillation period. The initial phase of the oscillations is assumed zero (\( \varphi = 0 \)) in (2).

The oscillation frequency was coordinated with the surface temperature using the mean velocity (3) and expression (1) in which the quantity \( V_j \) is replaced by \( \mathcal{V} \) from (3). The oscillating particles velocity distribution is set as Maxwellian function with temperature \( T_H \).

It should be noted here that surface particles move along strictly defined trajectories. In this case their velocities and coordinates are determined exclusively by the harmonic oscillation equations and do not change as a result of interaction with the liquid or vapor phase.

At the initial simulation stage the liquid system is brought into the equilibrium state with given temperature. In this paper the following simplest method is used. The liquid film temperature can be calculated from the following expression:

\[ \sum_{i=1}^{N} \frac{m_i V_i^2}{2} = \frac{3}{2} kT \]  
(4)

The temperature \( T \) calculated from (4) can be differ from the required temperature - \( T_0 \), i.e. in general case \( T \neq T_0 \). Hence the values of all particles velocities should be increased for case \( T < T_0 \) or decreased for case \( T > T_0 \). The scaling factor is introduced as follows:

\[ \text{coeff} = \sqrt[4]{\frac{T}{T_0}} \]  
(5)

This process is finished if after several iterations with the scaling of the velocities the system temperature calculated by expression (4) differs by not more than 3% from the set temperature \( T_0 \).

After this preliminary stage is completed the liquid film with a predetermined temperature \( T_0 \) located above the surface of the heater with a temperature \( T_H \) is obtained (figure 1).

The following conditions was considered: liquid temperature \( T_0 = 85.5 \text{K} \); the heat surface temperature was assumed to be 140K and 300K. Some modeling results are presented in figures 2-3.

The spatial distribution of liquid particles for diffusion reflection model and surface temperature 300K is presented in figure 2. The heater is marked by a red line in this figure. The presented picture corresponds to the time moment \( 3.0 \cdot 10^{-10} \text{s} \). At this figure the dimensions of the computation domain is shown in \( \sigma \) units (for argon \( \sigma = 3.4 \cdot 10^{-10} \text{m}, \varepsilon = 120K \)). As can be be seen from this figure the particles concentration (density) is small near the heater (range from zero to \( 7\sigma \)). This region can be considered as a vapor film region.
A similar computational experiment was carried out for a surface temperature of 140K. At all calculations the main criterion by which vapor film can be determined is the density of the medium. The variation of density over time for a region located at a distance of $5\sigma$ from the surface of the heater is a plot in figure 3. At this figure the results obtained for two different heater temperatures are presented. Time is given in relative units. The time scale is $t_s \approx 3.0 \times 10^{-13} \text{s}$.

For the model of oscillating particles the formation of vapor near a heated surface is observed also.

2.2. Interaction of laser radiation with droplet

Another strongly non equilibrium process observed in this paper is the phenomena of the high-power laser radiation interaction with liquid droplet.

In this case, heat is released in a limited region of the liquid. As a result an intensive evaporation of a droplet of matter can occur, as well as explosive destruction. The duration of the heat and mass transfer processes taking place during this process is tens of femtoseconds. The characteristic scale of time of molecular dynamics modeling is the period of lattice vibrations that is equal approximately to $\sim 10^{-13}\text{sec}$. Thus, using the methods of molecular dynamics simulation, heat transfer in a droplet and formation of a vapor near the surface of the droplet can be described. In the paper [5] the value of the specific power of the radiation incident on the particle approximately is $I_0 \approx 2 \text{GW}/\text{sm}^2 \approx 2 \times 10^4 \text{GW}/\text{m}^2$.

Suppose that the pulse duration is 100fs, and the dimensionless radius of the particle is $R_0 = 12.0$. The value $R_b = 5.2\text{A}$ is used as the base parameter for the distance.

As noted in [3] the interacting non equilibrium thermal, photochemical and photomechanical processes that occur during the interaction of laser radiation with matter are complex. So, it is very difficult and sometimes impossible to formulate closed mathematical models in terms of continuum mechanics. For example, the characteristic time of heat propagation in a droplet of radius $R$ by mechanism of thermal conductivity can be estimated as:

$$\tau = \frac{\rho c_p R^2}{\lambda}$$

(6)

For a drop of liquid argon with a radius $R = R_0 R_b = 12.0 \cdot R_b = 6.24 \text{nm}$ this value is $\approx 9.5 \cdot 10^{-9} \text{\text{s}}$.

It can be seen from this estimates the characteristic time of heat transfer due to thermal conductivity exceeds by five orders of magnitude the time of exposure to radiation ($\sim 100\text{fs} \approx 10^{-5}\tau$ where $\tau \approx 9.5 \cdot 10^{-9}\text{\text{s}}$ (see above)). At the same time, the available experimental data [25] show that
explosive fragmentation of liquid droplets can occur in a time much less than the characteristic thermal conductivity time. Thus, to describe the process of interaction of laser radiation with a drop at times comparable to the duration of the laser pulse, it is necessary to use approaches different from traditional ones. A drop of 6.24 nm contains 28873 particles.

We note that in the present study, effects associated with the focusing of the radiation incident on the drop and other optical effects will not be taken into account. The formulation of the problem for describing the process of interaction of radiation with a drop of liquid is shown in figure 5. Two cases of absorption of laser radiation are considered. In the first case, the energy that comes to the drop is absorbed in the painted part of the sphere (figure 5A), and in the second case - in the cylindrical region inside the drop (figure 5B).

![Figure 5. Laser radiation absorption models.](image)

To analyze the results of calculations the simulated area was divided into several layer. The values of the temperature were determined from the velocities of the particles contained in each layer. The center of the drop is at the point with coordinates \((0,0,0)\).

The change in droplet temperature over time for Case A and Case B is shown in figures 5-6. The numbers near the lines accord to the value of the time. The time scale is \(t_\infty \approx 3.3 \times 10^{-13}\) s. It can be seen from the figure that before the arrival of the laser pulse, the temperature everywhere in the drop is approximately equal to 87K (the time moment \(\tau^* \approx 54.0\)). After absorption of radiation, which has come to the right surface of the drop (\(\tau^* \approx 55.3\)), the surface temperature increases. In this case, the depth of "warming up" and the surface temperature depend on the chosen absorption model. So, for Case A, the surface temperature rises to 240 K and the heat from the right boundary (coordinate 12 in figure 5) penetrates to the coordinate \(-4\), that is, the region \(-4 < r^* < 12\) warms up. At the same time for "Case B", the droplet temperature rises to 190K, and the droplet region \(0 < r^* < 12\) warms up. Obviously, this is due to the fact that for "Case A" the absorption surface is larger than for "Case B". Thus, the model of the transmission and absorption of radiation by the droplet substance is important at considering this process. After cessation of exposure to radiation, the temperature of the droplet decreases and approaches the initial value (moments of time \(\tau^* \approx 57.5, \tau^* \approx 77.5\)). Obviously, this phenomenon is associated with the process of evaporative cooling.
It is interesting to note that, in the cases considered above, the liquid drop practically does not change its shape.

3. Conclusion
Methods of molecular dynamics simulation have been used to study the vapor film formation near the heater. The two different approaches were applied for liquid or vapor atoms interactions with the solid surface. The quantitative data for the vapor phase formation rate are obtained for two temperatures of the heater.

The processes occurring inside the liquid droplet as result impact of the high-power laser radiation have been considered. Two different models of laser radiation absorption are presented. The temperature distributions inside the droplet are given for different time moments and different absorption models.

Acknowledgments
This work is supported by the Russian Foundation for Basic Researches (project No. 17-08-00805)

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