MATHEMATICAL MODELING OF PHOTOINDUCED THERMOCAPILLARY CONVECTION IN A LAYER OF TRANSPARENT LIQUID ON AN ABSORPTIVE SUBSTRATE.

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Ph.D. thesis abstract

Physical Phenomenon Introduction and Thesis Objectives

Currents generated by laser irradiation induced surface tension alterations gain increasing attention of engineering and scientific communities. Potential commercial applications include fluids laser diagnostics, certain areas of bioengineering and such technological process as ablation, cutting of metals and dielectrics, surface alloying, welding, surface cladding and others.

Laser irradiation opens new areas of potential use of thermocapillary convection phenomenon. Laser beam reflected from a fluid surface contains complete information on surface shape profile which in turn is dependent on convective processes occurring in fluid bulk. Hence, information on surface shape profile can be used to deduce selected physical parameters of the fluid layer and a substrate properties shall a liquid layer be thin enough. Remote probing allows studying aggressive and radioactive fluids as well as fluids under extreme conditions.

Photoinduced thermocapillary convection is dependent on a number of parameters of "a beam-liquid layer-substrate"system. Continuous efforts to reveal physical nature of the phenomenon resulted in a significant progress in the subject, however a complete picture is yet to be constructed. This work aims to elucidate photoinduced thermocapillary convection evolution in a layer of transparent fluid on an absorptive substrate, specifically to establish relation between a delay time τ and "a laser beam-liquid layer-substrate"system properties. An ultimate goal is to construct a numerical model describing physical processes occurring in a system during thermocapillary convection initiation and depression formation on a fluid surface.

Research Methodology

During the course of this study, the following mathematical methods were employed: solution of partial derivatives differential equations, solution of integral equations, numerical methods of integration, differentiation and nonlinear equations solution, MAC and VOF methods. A VOF method was modified to define a free surface as in a system under investigation.

Scientific Novelty

1. A model of heat propagation prior to convective currents initiation was proposed. The model was based on a hypothesis of separate heat flows into a liquid layer and a substrate. A transcendental equation defining a thermocapillary convection delay time was derived.

2. A thermocapillary convection delay time dependence on a liquid layer thickness, a laser beam intensity and other system parameters was analyzed for the first time.

3. A model describing heat and mass transfer during thermocapillary convection in a layer of transparent liquid on an absorptive substrate was constructed using VOF method as a basis. A special feature of this model is representation of a "liquid-gas"system as a continuous phase with varying parameters.

4. A complex of numerical codes was realized on a VOF method basis with an aim to create a complete hydrodynamic model of the process. Evolution of a fluid surface deformation was numerically modeled. Simulation results suggest that an upsurge of all thermocapillary convection process parameters is expected after a certain time of irradiation with laser beam where time of upsurge is a function of system properties. Experimental observations support modeling results.

Verification and approbation

Validity of the numerical simulations was checked against experimental data. Also, a model created in this work was applied to known-to-art problems and the results were compared to those reported by other authors.

Contents

In the Introduction, the problem under investigation is defined, a scientific value of the research topic is presented, objectives of the work are formulated, a short synopsis of thesis is given.

In the 1st Chapter, a literature review on thermocapillary convection and the most relevant models accounting for the process are presented. Experiments elucidating photoinduced thermocapillary convection phenomenon are described and areas of its potential application are defined. Major steps of mathematical modeling of the process are identified. Contemporary methods of numerical modeling of the systems with interphase and free boundaries are analyzed.

A temperature gradient required for thermocapillary convection initiation on free surface can be created by laser irradiation. In case of transparent fluid, radiation is absorbed by a substrate only thus rendering a heat source on a liquid-substrate interface. To make thermocapillary convection observable via reflected laser beam registration, fluid surface deformation must be induced which requires a certain temperature increase in a near-surface region. A time interval between irradiation start and this observable fluid surface deformation is defined as a thermocapillary convection delay time.

The 2nd Chapter concerns the initial stage of photoinduced thermocapillary convection. Cases with Rayleigh number below a critical value of 1100, for which a buoyancy effect can be neglected, are examined. A mathematical model describing heat flux from a source located on a substrate into a liquid layer is constructed.

A problem statement is detailed below.

A layer of a transparent liquid of a thickness h_0 is supported on a solid substrate. A "liquid layer-substrate"system is heated by Gaussian laser beam. Liquid is transparent and laser radiation is absorbed by a substrate from where generated heat propagates into both liquid and a substrate. A substrate is assumed to be a semi-infinite solid body in comparison to a liquid layer thickness. Diametrical dimensions of the system are assumed to be infinite. Until temperature change on a liquid layer surface is insufficient to produce a surface tension gradient, heat propagation through liquid is conductive (thermal expansion is neglected). The following equations are valid for cylindrical coordinate system:

$$\frac{1}{\kappa_l}\frac{\partial T_l}{\partial t} = \frac{1}{r}\frac{\partial T_l}{\partial r} + \frac{\partial^2 T_l}{\partial r^2} + \frac{\partial^2 T_l}{\partial z^2}, \qquad 0 < z < h_0; \tag{1}$$

$$\frac{1}{\kappa_s}\frac{\partial T_s}{\partial t} = \frac{1}{r}\frac{\partial T_s}{\partial r} + \frac{\partial^2 T_s}{\partial r^2} + \frac{\partial^2 T_s}{\partial z^2}, \quad -\infty < z < 0; \tag{2}$$

$$0 \le r < \infty$$

with initial and boundary conditions

$$T_l = T_s = T_0 \qquad \text{at} \quad t = 0; \tag{3}$$

$$\frac{\partial T_l}{\partial z} = 0 \qquad \text{at} \quad z = h_0; \tag{4}$$

$$T_l = T_s, \quad k_l\frac{\partial T_l}{\partial z} - k_s\frac{\partial T_s}{\partial z} = -H_0 \exp\left(-\frac{r^2}{a^2}\right) \qquad \text{at} \quad z = 0, \tag{5}$$

where $H_0 = P/\pi a^2$ — heat flow density, P — laser beam power, a — beam radius, k — heat conductivity, κ — thermal diffusivity, indices l and s denote properties corresponding to liquid and substrate respectively.

Finding solutions for heat conductance problems for an infinite solid plate and half-space is a current task. The challenge is to distribute heat flows from a source located on a phase boundary (5). It was hypothesized that it is possible to obtain heat distribution coefficients for a simplified system and then apply them to a more complex problem. Such coefficients were obtained from a problem concerning heat conductance in two semi-infinite spaces with different thermal properties sharing a planar heat source on the interface. Then boundary conditions (5) are changed:

$$\frac{\partial T_l}{\partial z} = -\frac{1}{k_l}\frac{k_l\sqrt{\kappa_s}}{k_l\sqrt{\kappa_s}+k_s\sqrt{\kappa_l}}H_0\exp\left(-\frac{r^2}{a^2}\right), \tag{6}$$

$$\frac{\partial T_s}{\partial z} = -\frac{1}{k_s}\frac{k_s\sqrt{\kappa_l}}{k_l\sqrt{\kappa_s}+k_s\sqrt{\kappa_l}}H_0\exp\left(-\frac{r^2}{a^2}\right) \quad \text{at } z=0. \tag{7}$$

At such boundary conditions, the initial problem can be divided into two separate ones. The temperature field can be defined through a parabolic equation with Neumann boundary condition.

$$T_s(r,z,t)-T_0 = \frac{P\sqrt{\kappa_l}}{4\pi\sqrt{\pi\kappa_s}(k_l\sqrt{\kappa_s}+k_s\sqrt{\kappa_l})}\int\limits_0^t \frac{1}{(t-\tau+t_{0s})\sqrt{t-\tau}}\times$$
$$\times\exp\left(-\frac{r^2}{4\kappa_s(t-\tau+t_{0s})}\right)\exp\left(-\frac{z^2}{4\kappa_s(t-\tau)}\right)d\tau \tag{8}$$

$$T_l(r,z,t)-T_0 = \frac{P\sqrt{\kappa_s}}{4\pi\sqrt{\pi\kappa_l}(k_l\sqrt{\kappa_s}+k_s\sqrt{\kappa_l})}\int\limits_0^t \frac{1}{(t-\tau+t_{0l})\sqrt{t-\tau}}\times$$
$$\times\exp\left(-\frac{r^2}{4\kappa_l(t-\tau+t_{0l})}\right)\sum_{n=-\infty}^{\infty}\exp\left(-\frac{(z+2nh_0)^2}{4\kappa_l(t-\tau)}\right)d\tau \tag{9}$$

Solutions (8) and (9) account for heat propagation before to convective processes are initiated. These expressions satisfy the second formula in fitting condition (5), yet temperature equality is not precisely observed. The maximum difference $\left|\Delta T_l - \Delta T_s\right| / \min\{\Delta T_l, \Delta T_s\}$ during this stage of the process does not exceed 5%. These expressions are instrumental for a quick calculation of the media temperature profile before thermocapillary convection initiation.

Integrals (8) and (9) can yield temperature at any point in the system after integration over variable τ. To accomplish this, the author developed a "Delay Time"algorithm and implemented it in C++ code which realizes numerical integration of expressions (8) and (9) and calculates temperature fields in a "liquid layer-substrate"system.

A term "triggering temperature permutation" $(\Delta T)_{TC}$ is introduced defined as a maximum temperature gradient on a free surface at the moment of thermocapillary convection initiation. Experiments show that delay time is a function of a layer thickness (h_0) square. This relation is valid only in case when the $(\Delta T)_{TC}$ value for a specific fluid is constant and independent of a laser beam power and spot size. It is possible to construct in an implicit relation of delay time τ_d on a laser beam power:

$$F(\tau_d, P) = (\Delta T)_{TC}, \tag{10}$$

where $F(\tau_d, P) = \Delta T(0, h_0, \tau_d) = T_l(0, h_0, \tau_d) - T_0$ — the left part of equation (9) and power P is variable. Calculations of $(\Delta T)_{TC}$ values for a particular fluid are based on a reference $(\Delta T)_{TC}$ value for 0.81 mm thick layer of fluid irradiated with 20.9 mWt related to conducted experiments.

Figure 1(a) shows experimental values (dots) of delay time as a function of a laser beam power for butanol layers of $h_0 = 0.33$ (top), 0.57, 0.81, 1.41, and 2.01 (bottom) mm; calculated trends for the systems with identical parameters are represented by solid lines. As evident from the figure, calculated trends are in a good agreement with experimental data. A significant description was observed only for thicker layers with h_0 approaching 2 mm. This comparison supports a hypothesis of $(\Delta T)_{TC}$ being independent of a laser beam power P.

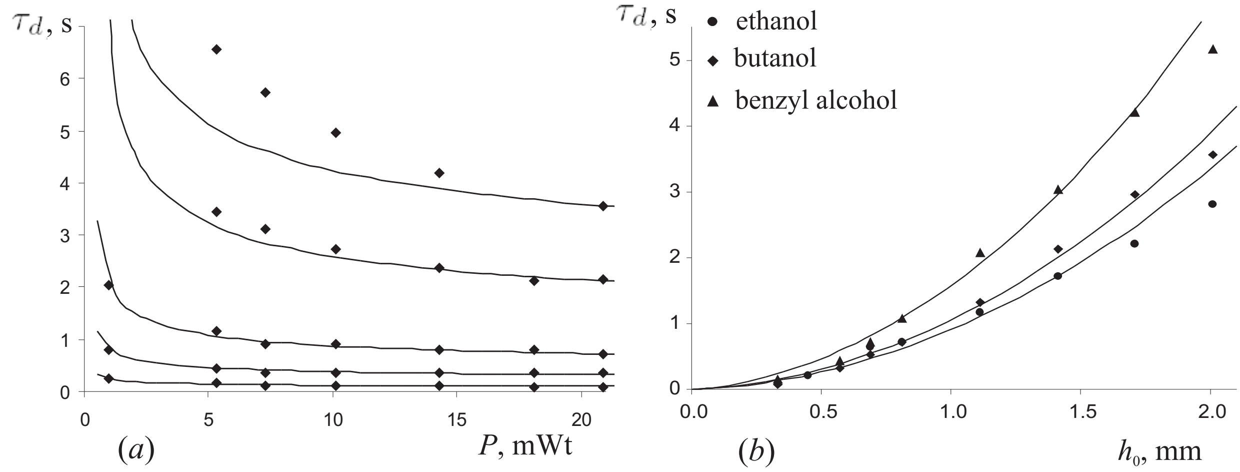


Рис. 1: Delay time τ_d — system parameters relationship: P — laser beam power, H_0 — liquid layer thickness. Calculated trends are represented by solid curves, experimental values by dots.

To verify τ_d dependence on h_0^2 the following calculations were performed. As in the previous case, one experiment for $(\Delta T)_{TC}$ determination was performed on 0.81 mm thick liquid layer. A constant value of $(\Delta T)_{TC}$ obtained in the experiment was used to construct a theoretical relationship of τ_d and a layer thickness h_0 $F(\tau_d, h_0) = (\Delta T)_{TC}$ for the three fluids with different viscosity. Figure 1(b) presents theoretical curves (solid lines) and experimental values (dots) of τ_d, h_0 relationship for different fluids irradiated with 20.9 mWt laser beam. As for a previous fit, significant discrepancy (<15%) is observed for $h_0 > 1.8$ mm.

In the 3rd Chapter, a thermodynamic model is constructed. In contrast to a classic system of balance equations, proposed model accounts for Marangoni forces acting on a free surface which are incorporated into Navier-Stocks equations. In this work, such system of equations was derived and a solution algorithm based on VOF method was implanted in a numerical code. Since numerical solution can be realized only in a finite space, schemes of departure from the infinite boundaries based on dimensionless variable parameters analysis were developed. Another challenge in a boundary description part arises from a requirement of a precise definition of the deformed and thus unstable gas-liquid interphase. To adapt this parameter to the developed numerical model, a special technique known as a Volume of Fluid (VOF) method based on a piecewise linear interface construction was applied.

To define heat- and mass transfer processes in the system, a set of balance equations is required. In this system, the fluid is assumed to be uncompressible and viscous. At small temperature variations, all system parameters remain constant except surface tension which is linearly dependent on temperature.

To define an interphase boundary in the VOF method, a special function F is introduced. This function characterizes fluid concentration in each calculated space cell, with $F = 1$ in liquid phase, $F = 0$ in gas phase, and $0 < F < 1$ for cells located on the interphase boundary. Function F field is changed during fluid movement according to the following transport equation:

$$\frac{\partial F}{\partial t} + u\frac{\partial F}{\partial r} + v\frac{\partial F}{\partial z} = 0 \tag{11}$$

At the phase boundary, phase physical properties discontinuity and surface tension forces are present:

$$\vec{f_s} = \sigma\varkappa\hat{n} + \nabla_T\sigma \tag{12}$$

where $\varkappa = 1/R_1 + 1/R_2$ — a free surface curvature, $\hat{n}$ — a unit vector normal to the free surface and directed inwards the liquid phase. The equation of continuity holds valid for the entire numerical space including phase boundary.

Fluid and surrounding gas are considered as a continuous liquid phase with the properties linearly dependent on function F:

$$\begin{aligned} \mu &= F\mu_l + (1-F)\mu_g \\ \rho &= F\rho_l + (1-F)\rho_g \end{aligned} \tag{13}$$

where indices l and g designate properties to liquid and gas respectively. A dynamic condition (12) is valid for the phase boundary and is incorporated into Navier-Stocks equation. With all assumptions made, a set of balance equations for this system can be written as following:

$$\frac{1}{r}\frac{\partial ru}{\partial r} + \frac{\partial v}{\partial z} = 0 \tag{14}$$

$$\begin{aligned} \frac{\partial u}{\partial t} + u\frac{\partial u}{\partial r} + v\frac{\partial u}{\partial z} = -\frac{1}{\rho}\frac{\partial p}{\partial r} + \frac{1}{\rho}\left\{\frac{\partial}{\partial r}\left(\mu\frac{1}{r}\frac{\partial ru}{\partial r}\right) + \frac{\partial}{\partial z}\left(\mu\frac{\partial u}{\partial z}\right)\right\} + \\ + \left\{\sigma\varkappa n_r - \gamma\left(\frac{\partial T}{\partial r} - n_r\left(n_r\frac{\partial T}{\partial r} + n_z\frac{\partial T}{\partial z}\right)\right)\right\}\frac{|\nabla F|}{\rho} \end{aligned} \tag{15}$$

$$\frac{\partial v}{\partial t}+u\frac{\partial v}{\partial r}+v\frac{\partial v}{\partial z}=-\frac{1}{\rho}\frac{\partial p}{\partial z}+\frac{1}{\rho}\left\{\frac{1}{r}\frac{\partial}{\partial r}\left(\mu r\frac{\partial v}{\partial r}\right)+\frac{\partial}{\partial z}\left(\mu\frac{\partial v}{\partial z}\right)\right\}+$$
$$+\left\{\sigma\varkappa n_z-\gamma\left(\frac{\partial T}{\partial z}-n_z\left(n_r\frac{\partial T}{\partial r}+n_z\frac{\partial T}{\partial z}\right)\right)\right\}\frac{|\nabla F|}{\rho}+g\beta(T-T_0) \tag{16}$$
$$0\leqslant r<\infty,\quad 0\leqslant z<\infty$$

where β is thermal expansion coefficient. In contrast to known in the filed balance equations (Hirt and Nicols 1981, Meyer 1999), equations derived in this work allow account for surface tension changes due to the temperature gradient on a free surface.

Heat conservation equation for the liquid is

$$\frac{\partial T}{\partial t}+u\frac{\partial T}{\partial r}+v\frac{\partial T}{\partial z}=\frac{1}{c_p\rho}\left\{\frac{1}{r}\frac{\partial}{\partial r}\left(kr\frac{\partial T}{\partial r}\right)+\frac{\partial}{\partial z}\left(k\frac{\partial T}{\partial z}\right)\right\} \tag{17}$$
$$0\leqslant r<\infty,\quad 0<z<\infty$$

Energy conservation equation for the substrate is

$$\frac{\partial T_s}{\partial t}=\kappa_s\left\{\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T_s}{\partial r}\right)+\frac{\partial^2 T_s}{\partial z^2}\right\} \tag{18}$$
$$0\leqslant r<\infty,\quad -\infty<z<0$$

where k — heat conductivity, c_p — specific heat capacity, κ — thermal conductivity.

Equations (11), (14), (15)–(18) are principal for describing thermocapillary convection phenomenon in axially symmetric representation, and these equations are solved numerically. For closure of the equations, it is necessary to formulate initial and boundary conditions. At the initial time, the liquid is stationary and there is no pressure or temperature gradient present in the system. No slip and impermeability conditions are observed at the walls

$$u=v=0,\quad \text{at } z=0$$

Since the liquid layer is assumed fully transparent, all energy of the laser beam is absorbed by the substrate. Then, a thermal contact condition is valid for the substrate-liquid interface:

$$T-T_s=0,\quad k\frac{\partial T}{\partial z}-k_s\frac{\partial T_s}{\partial z}=-H_0\exp\left(-\frac{r^2}{a^2}\right),\quad \text{at } z=0 \tag{19}$$

where H_0 — heat intensity of a laser beam (which is conditioned to fade out in infinite space).

For equations discretization, a classic Marker And Cell (MAC) method is applied to 2D cylindrical coordinates system. To achieve best convergence, the space is divided into rectangular cells of $\Delta r\times\Delta z$ dimensions. In these cells, values of functions (p,T,F) and physical properties (μ,ρ,k,c_p) are defined in the centers and velocities on the side midpoints. Coordinates in the cells are defined as $r_i = (i - 0.5)\Delta r$ и $z_j = (j - 0.5)\Delta z$ (for the sake of brevity, finite-difference scheme is not shown in this abstract).

In the 4th Chapter, numerical codes used for the phenomenon modeling are detailed. Velocity, temperature and pressure fields are constructed. The problem for a thermocapillary convection delay time is solved numerically for the liquid with permanently changing certain physical properties.

There is no well-defined criterion for qualifying convection starting point. Hence, the author proposed a criterion based on the analysis of differential equations approximation errors. Calculated delay time values are compared to the experimental results. Developed mathematical model allows rapid calculation of delay times for different systems with variable parameters which would be a very involved process if determined experimentally. Also, free surface deformation profiles during thermocapillary convection were calculated which yet could not be resolved experimentally. Such profiles could be used for the fluid properties evaluation if physical analysis is complicated or unavailable.

In VOF method, an interphase boundary position calculation based on a function F fractal field, which is defined at each time step from the transport equation (11). This method is allows to account for a large-scale interface perturbances such as injection, droplet fall, bubble surfacing. However, free surface deformation during thermocapillary convection is sufficiently low to apply a kinematic scheme for the interphase boundary definition. This scheme is based on the assumption that in absence of evaporation from the surface, the surface film velocity is equal to the fluid velocity in a layer adjacent to the free surface. Therefore, in a dimensionless form, surface shape for the axially symmetrical case can be defined as:

$$\frac{\partial h}{\partial t} + u\frac{\partial h}{\partial r} = v, \tag{20}$$

where $h = h(r, \tau)$ — a free surface shape which is undisturbed at $t = 0 : h(r, 0) = h_0$. Its discrete approximation is:

$$h_i^\bullet = h_i + \Delta t\left(\overline{v}^\bullet_{f\,i} - \overline{u}^\bullet_{f\,i}\frac{h_{i+1} - h_{i-1}}{2\Delta r}\right) \tag{21}$$

where u_f and v_f are free surface velocities.

Since explicit scheme is used for numerical calculation, the algorithm stability requires fulfillment of certain conditions for time increments Δt: Kourant-Friedrich-Levi condition for the velocity $\Delta t \leqslant \Delta t_{CFL} = \Delta r/V$, where V — the maximum velocity $\mid u^\bullet_{i+1/2,j} \mid$, $\mid v^\bullet_{i,j+1/2} \mid$; thermal conductivity $\Delta \leqslant \Delta_{th} = (\Delta r)^2/2\kappa$, viscosity $\Delta t \leqslant \Delta t_{vis} = \rho(\Delta r)^2/2\mu$, and surface tension $\Delta\tau \leqslant \Delta\tau_{s.t.} = \sqrt{(\rho_l + \rho_g)/4\pi\sigma_0}(\Delta r)^{1.5}$.

As stated above, a thermocapillary convection process causes free surface deformation with a depression formed on fluid free surface. Figure 2 shows a free surface profile during thermocapillary convection development and at stationary conditions. The model accounts

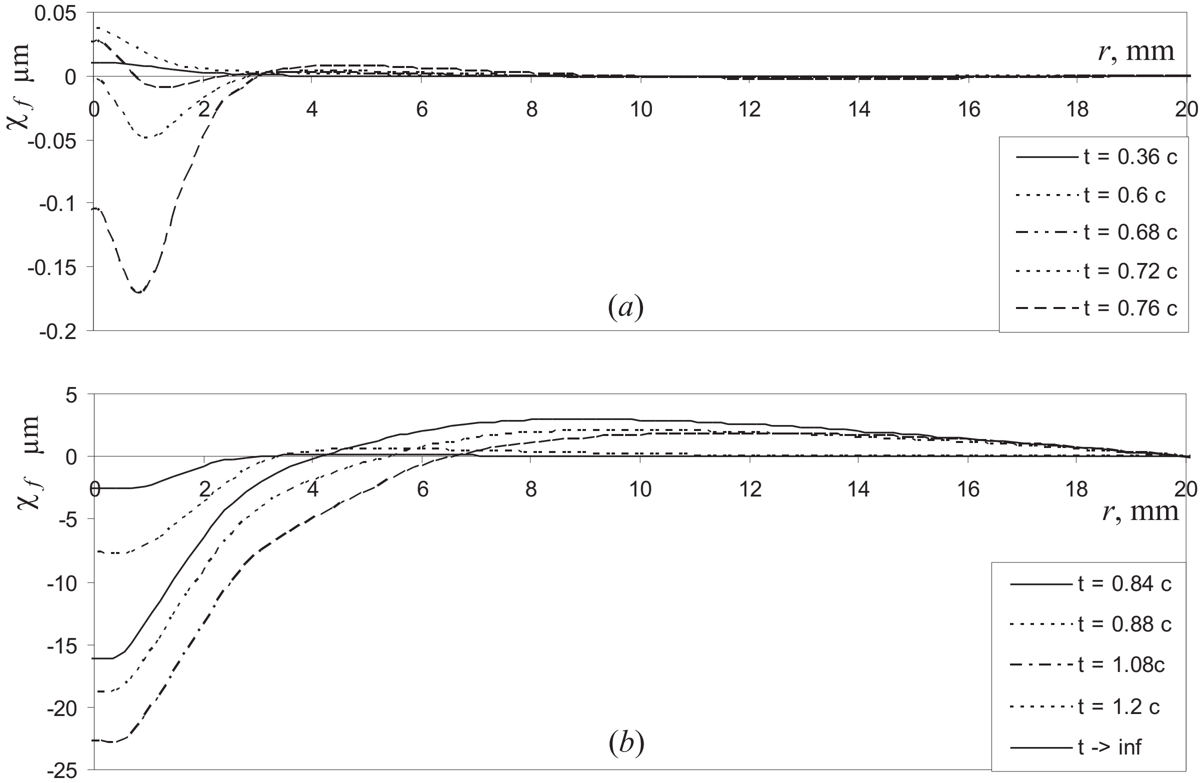
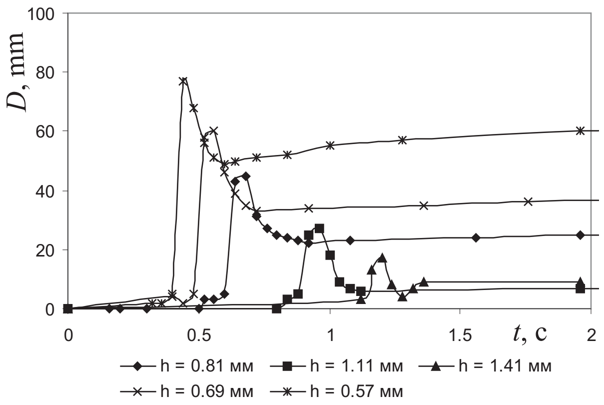


Рис. 2: Thermocapillary surface deformation profile calculated for 1.5 mm n-octane layer irradiated with 20.9 MWt laser beam: (a) initial hump formation (a) followed by a dimple development (b).

for fluid movement due to thermal expansion and gravitation. The former leads to the formation of a small hillock at the initial stage of the process which is illustrated in Figure 2(a). At certain point, the fluid diffluence and a depression is formed as shown in Figure 2(b). From these profiles it can be seen that the maximum depth of the depression occurs some time into its formation but not at the stationary regime. For example, for 0.81 mm n-octane layer this maximum occurs at 1.08 sec into the laser irradiation. Later on the surface raises and the profile becomes stationary.

As established in the study performed by the Liquid Microgravitational Technologies laboratory, the diameter of the thermocapillary response is a function of a maximum inclination angle of the deformed free surface. Figure 3 shows simulated plots for a thermocapillary convection response diameter (left) and a maximum tangent of the interphase inclination angle (right) versus time for n-octane layers of different thickness. The plots reproduce all experimentally observed trends. Therefore, the model accounts for the three main stages observed during thermocapillary convection progression: a delay, an upsurge and a stationary regime setting.

Highlight of the project



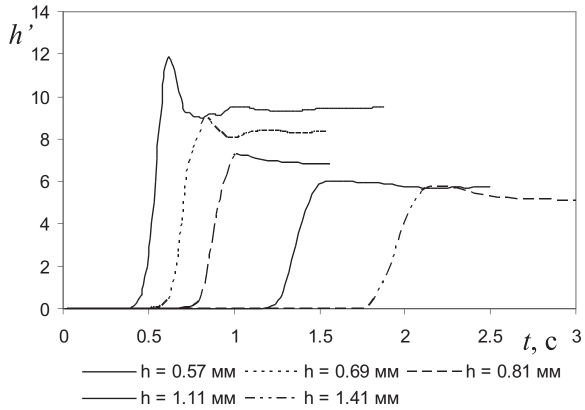


Рис. 3: Progression of a thermocapillary response diameter (left) and of a tangent of a maximum inclination angle of the free surface (right). Calculations are performed for the layers of different thickness listed under the plots.

1. A model of heat propagation occurring prior to convective flows initiation was developed. Heat distribution between the fluid and the substrate was quantified. Based on this model, a thermocapillary convection delay time was calculated and its relation to the fluid layer thickness and laser beam intensity was established.

2. The numerical code "Delay Time"calculating the temperature profiles for the "liquid layer-substrate"system, the initiation temperature, and the delay time was implemented in C++.

3. It was proposed for the first time to utilize a novel method for solving free boundaries problems (VOF method) for in the thermocapillary convection modeling. A set of balance equations for a free surface under dynamic conditions with acting Marangoni forces was derived.

4. Based on VOF method, a thermohydrodynamic model for thermocapillary convection in a layer of transparent fluid on an absorptive substrate was developed. Heat propagation through the liquid layer and the substrate was described. Interphase boundary was defined through a kinematic interfacial condition and once it is defined, a liquid fraction filed F was calculated at each increment.

5. The "Thermocapillary Convection"set of numerical codes was developed to realize constructed mathematical model. This modeling package allows calculation of the temperature, pressure, and velocity fields as well as a free surface deformation profile.

6. The model describes thermocapillary convection onset and is in a good qualitative agreement with experimental results.