COALESCENCE OF A DROPLET CLUSTER SUSPENDED OVER A LOCALLY HEATED LIQUID LAYER

A. A. Fedorets,1 I. V. Marchuk,2,3,* & O. A. Kabov2,4

1Tyumen State University, Semakova Street 10, Tyumen, 625003, Russia
2Institute of Thermophysics, Russian Academy of Sciences, prosp. Lavrentyev 1, Novosibirsk, 630090, Russia
3Novosibirsk State University, Pirogova 2, Novosibirsk, 630090, Russia
4Tomsk Polytechnic University, Lenin Avenue 30, Tomsk, 634050, Russia

*Address all correspondence to I. V. Marchuk, E-mail: marchuk@itp.nsc.ru

Experimental results on the coalescence of a dissipative structure “droplet cluster” obtained by means of high-speed thermal imaging are presented. It is found out that coalescence of a single cluster droplet with a liquid layer can generate a capillary wave on the interphase, and propagation of this wave causes coalescence of the whole cluster during several thousands of a second. Cluster coalescence is accompanied by a temperature jump on the interphase. With cluster restoration, the surface temperature of a liquid layer returns gradually to the initial level. The interphase temperature under the droplet cluster can be both lower and higher than the interphase temperature without this cluster, i.e., the cluster can not only initiate both heat and mass transfer between liquid and gas, but also deteriorate it.

KEY WORDS: levitating drops, evaporation, thermocapillary convection, capillary wave, thermal imaging, temperature jump on the interphase

1. INTRODUCTION

Many engineering processes and laboratory experiments include liquid layer heating by an intensive localized heat source on a substrate or by a powerful localized radiation beam. For instance, this situation occurs at cooling of micro-electronic equipment, and locally the heat flux density can be 1 kW/cm² (Mahajan et al., 2006). Boiling, evaporation, thermocapillary convection, and break of liquid layers are studied at local heating [e.g., see Nepomnyaschy et al. (2002), Zaitsev and Kabov (2007), Andreev et al. (2008), Marchuk (2009)].

At intensive local heating and evaporation of water and some other liquids (glycerin, benzyl alcohol, ethylene glycol), the spatially ordered structure “droplet cluster” can be observed. This is a structure of dozens or hundreds microdroplets levitating parallel to the horizontal liquid-gas interphase at a distance comparable to the droplet diameter, Fig. 1. The droplet cluster was detected both at radiation of a thin liquid layer by a light beam (the area of $8 \times 10 \text{ mm}^2$) (Fedorets, 2004) and at heating of a liquid layer from the substrate by a heater with diameter of 1 mm (Fedorets, 2005). The local character of interphase heating as well as the open character of the system (i.e., its ability to remove efficiently heat fluxes and vapor flows from the heated region of interphase) are principally important for generation and stable existence of the droplet cluster. Some analogy with this effect is observed with the Leidenfrost effect, when droplets are held up by a layer of vapor over a heated surface due to intense evaporation of the drops, but in our case the condensation of vapor is on the drops, because the drops grow in time.

There is the threshold liquid temperature in the zone of heating, below which the cluster can not be formed, i.e., the droplets precipitated on the liquid surface coalesce with the layer. Under atmospheric pressure and air temperature $T_g$ of about 20–25°, water surface temperature $T_S$ in the zone of local heating should not be lower than 50° (Fedorets,
It is determined in Arinshtein and Fedorets (2010) that the vertical temperature gradient in the gas medium above the locally heated region can be \( \sim 30 \text{ K/mm} \). If the temperature is relatively high, a pattern of precipitated condensate microdroplets with the size of \( \sim 10 \mu\text{m} \) in the form of continuously changing light irregular polygons can be observed above the liquid surface in a cup of tea or coffee (Schaefer, 1971). Transformation from uniform to local heating leads to a qualitatively new phenomenon.

Droplets come to the cluster from the gas environment, where they nucleate and undergo the first stage of condensation growth. Initially these droplets are the fog droplets, generated at vapor condensation, which mix with cold air, while moving up from the heated surface. If the typical size of fog droplets for water is \( \sim 10 \mu\text{m} \) (Raist, 1984), the size of cluster droplets is about 50–100 \( \mu\text{m} \) (Fedorets, 2005). The spatially ordered structure close to the structure of the plane hexagonal lattice is typical for the cluster, Fig. 2. The cluster has clearly defined boundaries, and its shape is determined by the geometry of the locally heated region of the liquid surface (Fedorets, 2004, 2005). The cluster droplets are subject to continuous condensation growth, the rate of which is proportional to heating power; at this, the linear dependence of area \( S \) of the spherical droplet surface on time is observed, Fig. 3. Usually, the droplets of cluster have almost the same diameter, Fig. 2(a), but the sizes of simultaneously levitating droplets can differ significantly because the moment of droplet penetration into the cluster is random, Fig. 2(b). At this, the height of levitation \( H \)
and droplet diameter $D$ are connected by inverse dependence, $H \approx 1/D$. Geometrical sizes of cluster droplets were measured in detail in Fedorets (2005, 2011), Shavlov et al. (2011), and Shavlov and Dzhumandzhi (2012).

It is shown experimentally in Fedorets et al. (2011) that cluster droplets levitate above the liquid surface, Fig. 2(c), because of the influence of the vapor-air flow generated by intensive liquid evaporation (the Stokes mechanism of levitation). The question of the mechanism of droplets' interaction (mutual attraction at large distances and repulsion at small distances) is still unsolved. The similarity of the structure of dust plasma crystals, intensively studied in Thomas et al. (1994) and Fortov et al. (2004), and structure of the droplet cluster are discussed in Shavlov and Dzhumandzhi (2010). An assumption of similar interaction mechanisms in dust crystals and droplet clusters is considered. According to Shavlov and Dzhumandzhi (2010), the Coulomb forces, related to discharge accumulation in cluster droplets and on the interphase, are three orders less than the droplet weight.

According to investigations, this phenomenon is well reproduced in experiments of different authors, and it does not require expensive equipment for its implementation. The droplet cluster has some useful properties; for instance, the small size of droplets located close to the interphase, and cluster keeping after disconnection of heat load (~10 s).

The above stated can soon lead to practical application of this phenomenon, e.g., for determination of the physical properties of liquid. In Fedotets (2008), cluster microdroplets have been already used as the natural tracers for flow visualization in the gas phase directly near the interphase. Understanding the mechanism of droplet cluster formation can allow the conclusion about possible formation of the ordered structures of microdroplets in thundercloud and fog, and these structures can achieve some specific properties such as surface tension and shear viscosity (Shavlov and Dzhumandzhi, 2010). Moreover, the cluster generates the droplets, naturally suspended in the atmosphere, with a significant temperature difference at the upper and lower boundaries (Arinshtein and Fedorets, 2010), which gives the unique possibility for investigation of evaporation and condensation as well as thermocapillary convection in the microscale.

Nevertheless, the possible role of the above-mentioned evaporation, condensation, and thermocapillary convection in the mechanism of droplet cluster formation is as yet unclear. An adequate mathematical model of this phenomenon

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**FIG. 3:** Changes in diameter (1), spherical surface area (2), and mass (3) of cluster droplets in the process of their condensation growth. Values $D_0 = 21.6 \mu m$, $S_0 = 1470 \mu m^2$, and $m_0 = 5.3$ ng correspond to the moment of measurement beginning, $t = 0$. Water layer, cluster induced by the metal heater of 1 mm diameter built into the substrate [according to (Fedorets, 2011)].
has not been developed until now. The mechanism and conditions of natural break of the cluster have not been studied in detail. In the current research, we present experimental results on coalescence of the droplet cluster with the layer; these data were obtained with application of high-speed thermal imaging. The effect of droplet cluster on the processes of heat and mass transfer between liquid and gas medium was studied.

2. EXPERIMENTAL METHODS

A cylindrical duralumin cuvette was used for generation of the droplet cluster, Fig. 4. A copper rod of round cross section \( R_{\text{heater}} = 0.5 \) mm with a length of 5 mm is glued in the center of the cuvette bottom; this rod serves as the heating element. The rod is heated by electric current passing through the nichrome wire wound around the lower part of the rod. The gap between the heater and the cuvette is filled by epoxy resin with relatively low heat conductivity, which allows us to have a localized heat source on the cuvette bottom.

Experiments were carried out with degassed distilled water containing natural microadmixtures of surfactants in the cuvette exposed to the air in the laboratory room. In all experiments, the layer thickness was \( h_0 = 380 \pm 10 \) μm, and it was determined by the volume of liquid poured into the cuvette directly before the experiment (with the use of Lenpipet Stepper dispenser, tip of 5 ml, relative dosing error of \( \pm 0.3\% \)). Liquid at the layer periphery was in thermodynamic equilibrium with the ambient air, whose temperature \( T_{\text{g0}} \) was \( 22 \pm 1 \)°. The electric power of the heater was measured discretely; it took one of the following values: 0.31, 0.39, 0.49, 0.60, or 0.72 ± 0.005 W. At this, the temperature stabilized on the interphase above the heater center \( T_{S_{\text{max}}} \) and took one of five following values: 78.1, 82.6, 86.6, 90.1, or 92.8 ± 0.2°, respectively.

3. METHOD OF TEMPERATURE MEASUREMENT ON THE INTERPHASE

To register temperature distribution on the interphase, the thermal imager itanium 570 M was used (spectral range of 3.7–4.8 μm, matrix of 640 × 512, measurement accuracy of \( \pm 1\% \), NETD < 25 mK). The accuracy of absolute temperature measurements is \( \sim 0.5 \) K (1% of the scale range), the accuracy of measurements of temperature differences is defined by sensitivity, which is defined by a parameter called NETD (noise equivalent temperature difference). For IR camera Titanium 570M, NETD is less than 25 mK. The lens “L0120 – MW x1 2.0 Jade” for the scale of 1:1 was applied; at this, the pixel size was 15 × 15 μm. For this lens, the image field flatness is ± 0.3 mm. It is quite enough for simultaneous temperature measurements of droplets and liquid surface because the distance between the layer surface

![Image of experiment scheme](image-url)

**FIG. 4:** Scheme of experiment.
and top of the droplet less than 0.3 mm. Measurement frequency varied from 25 to 1500 Hz. At high frame frequency (>115 Hz), not the whole matrix was scanned, but only the window with the size of 132 × 132 pixels. The method of temperature measurements on liquid surfaces with high temperature gradients is presented in Kabov et al. (1996). In particular, the questions related to radiation of the inner liquid layers and arising measurement errors, which can be significant for such semitransparent liquids as alcohols, were considered. The high absorption coefficient of water, α = 34.7 mm⁻¹ within the spectral range, registered by the thermal imager, allows an assumption that the average temperature of a thin surface layer of liquid is measured (the layer thickness is <10 μm). The water emissivity in the spectral range, registered by the thermal imager, is within 9.75–9.85. Factory calibration of the setup was used for recalculation of the measured intensity of IR radiation to the values of temperature with taking into account the value of water emissivity coefficient equal to 0.98.

An example of temperature distribution over the interphase is shown in Fig. 5 for different heater powers. Here, the spatial coordinate is rated by heater radius R_{heater}, and the temperature is rated by T_{S\text{max}} in the region of interphase near the heater center, individual for every dependence,

\[ T_{S\text{max}} = \frac{1}{4} \left[ T_{S\text{max}(1)} + T_{S\text{max}(2)} + T_{S\text{max}(3)} + T_{S\text{max}(4)} \right] \]  

(1)

where T_{S\text{max}(1)}, T_{S\text{max}(2)}, T_{S\text{max}(3)}, and T_{S\text{max}(4)} are the maximal temperatures on the interphase in each of four rectangular fragments of the image, located symmetrically relative to the heater center (two fragments are shown in Fig. 5, the size of each fragment is 9 × 9 pixels). Expression (1) allows proper determination of T_{S\text{max}} for every value of the heater power, even in the presence of cluster, whose droplets partially overlap the interphase and have the lower temperature.

As can be seen in Fig. 5, in the whole working range of the heater power, the heat field on the interphase is not undergone to qualitative reconstructions. The heat source is localized well, and a drastic drop of temperature occurs.
in the heat-insulating resin layer around the heater. In the area of cluster existence \((r/R_{\text{heater}} < 1)\), the temperature decreases gradually with distance from the heater center; at this, the temperature difference within this area does not exceed 5% of \(T_{S_{\text{max}}}\).

4. RESULTS AND DISCUSSION

Spontaneous coalescence of the droplet cluster can be easily observed and traced by any continuous video record of this phenomenon. However, coalescence is so fast that on a video record with the standard frame frequency of 25 fps, this phenomenon is observed as “instantaneous” disappearance of the cluster. Details of the process can be observed only on a video with frequency of at least 1000 frames per second, Fig. 6. It was found out that the whole cluster “disappears” in 3 ms as a result of coalescence of a single droplet initiator (in the frame before coalescence beginning, it is outlined by a solid line).

Droplets coalesce with the layer leaving characteristic “cold” traces on the interphase, Fig. 6. The concentric symmetry and propagation velocity of the front of cluster destruction \(\sim 70 \text{ cm/s}\) prove that coalescence is caused by the capillary wave on the interphase. In approximation of a thin layer, phase velocity \(\nu\) of the capillary waves is determined by classical relationship (Landau, 1987),

\[
\nu = \sqrt{\frac{2\pi\sigma}{\lambda\rho}}
\]

where \(\sigma\) and \(\rho\) are surface tension and density of liquid, and \(\lambda\) is the wavelength.

The principle possibility for capillary wave formation, propagating with a velocity of \(\sim 70 \text{ cm/s}\), exists in the whole range of values of surface tension of water with surfactant admixtures \((\sigma = 30–74 \text{ mN/m})\). The detailed investigation of parameters of the wave process on the interphase, causing cluster coalescence, relates to application of specialized experimental equipment, and it is beyond the framework of the current research.

High-speed thermal imaging allowed registration of the effect of droplet cluster on the heat and mass transfer processes on the liquid-gas interphase, Fig. 7. Instantaneous cluster coalescence is accompanied by an expressed temperature jump on the interphase. With cluster restoration, the temperature of the surface layer returns gradually to the initial level. The nomenclature \(T_{S_{\text{max}}(d)}\), used in the diagram, indicates average values of \(T_{S_{\text{max}}}\) during the last four seconds before the moment of droplet cluster coalescence, i.e., it is the equilibrium temperature of interphase under the droplet cluster. Below, the main attention will be paid to such a parameter as temperature jump \(\Delta T_{S_{\text{max}}}\) on the interphase at the moment of cluster coalescence,

\[
\Delta T_{S_{\text{max}}} = T_{S_{\text{max}}0} - T_{S_{\text{max}}(d)}
\]

where \(T_{S_{\text{max}}0}\) is the average value of \(T_{S_{\text{max}}}\) during the first two seconds after droplet cluster coalescence. Factors influencing the choice of the time length of averaging \(T_{S_{\text{max}}}\) are different with or without a cluster. With droplet cluster, the process is quite steady state during measurement, and extended \((4 \text{ s})\) interval averaging \(T_{S_{\text{max}}(d)}\) increases

FIG. 6: Effect of “spontaneous” coalescence of the droplet cluster. The coalesced first drop is circled by the solid line. The dashed line shows an approximate position of the capillary wave front.
FIG. 7: Temperature jump of the interphase due to droplet cluster coalescence. Temperature $T_{S \, \text{max} \, 0} = 86.6^\circ$, separate video fragments used for diagram construction are shown additionally.

the accuracy of measurements. Restoration of the cluster begins immediately after the coalescence, according to the extent of increasing the area of the cluster increase and its effect on the temperature of the surface layer. By definition, $T_{S \, \text{max} \, (0)}$ characterizes the surface layer in the absence of drops, which is rightly only the first 2–3 s after the coalescence. Hence, the averaging interval $T_{S \, \text{max} \, (0)}$ is equal to 2 s.

The diagram of dependence between $\Delta T_{S \, \text{max}}$ and temperature on the interphase under the central part of the heater (data correspond to five discrete levels of heater power with other things being equal) is shown in Fig. 8. IR cluster images in parts of a second before coalescence are shown additionally there. It is obvious that the droplet diameter increases significantly with a rise of heater power. The cluster effect on the surface layer temperature can be reproduced stably; at this, the amplitude of the registered temperature jumps exceeds significantly the error level of the used thermal imager. Every point in Fig. 8 corresponds to 150 measurements. In sum, we can distinguish following new experimental facts: (i) characteristic value $\Delta T_{S \, \text{max}}$ makes up the tenths of a degree; (ii) duration of the period of temperature restoration on the interphase after cluster coalescence is measured by tens of seconds, Fig. 7; (iii) the interphase temperature under the droplet cluster can be both higher and lower than the interphase temperature without a cluster.

The possible physical mechanisms, explaining the above-mentioned facts, can be as follows: (i) the sequences of “instantaneous” injection of relatively cold cluster droplets into the heated liquid layer; (ii) the flows, mixing liquid in the layer, initiated by droplet coalescence; (iii) the processes of heat and mass transfer in cluster droplets, which actively effect the air-vapor jet above the locally heated region of the layer.

Actually, when coalescing with the layer, the cluster droplets generate the local areas on the interphase with the temperature, decreased by several tenths of a degree; these areas can be clearly seen in Fig. 6. The heat effect depends on the temperature and volume of injected liquid. With an increase in the temperature of the surface layer from 78 to 93$^\circ$, the volume of a single droplet at the moment of cluster coalescence increases approximately by a factor of 22; however, on the contrary, the number of droplets, Fig. 8, decreases by 20 times. Dependence of the volume of injected liquid $V = nV_1$ on the temperature of the surface layer is shown in Fig. 9 ($n$ is the number of coalescing droplets,
FIG. 8: Dependence of amplitude and sign of the temperature jump on the interphase on the liquid temperature in the zone of local heating of the layer.

$V_1$ is the volume of a single droplet). This dependence is not monotonous: the knee corresponds approximately to $90^\circ$, i.e., the temperature at which the temperature jump at coalescence changes its sign, Fig. 8. The volume of injected fluid is less than one order of magnitude of the layer volume and thermal footprint relaxes in a few milliseconds, Fig. 6. Moreover, the cluster droplets are always colder than the liquid layer [i.e., this mechanism cannot explain the experimental fact of $T_{S,\text{max}}$ increase after cluster destruction (Fig. 7)].

Local areas with the decreased temperature on the interphase can cause there thermocapillary convection, which can mix the liquid. Arising shear stress $\tau_{\text{sur}}$,

$$\tau_{\text{sur}} = \frac{\partial \sigma}{\partial T} \cdot \frac{\partial T}{\partial x}$$

removes liquid from more heated zones of liquid surface to less heated ones [e.g., see Nepomnyaschy et al. (2002) and Andreev et al. (2008)]. The assumption of the fact that coalescence of cluster droplets can initiate convective mixing of the layer was checked experimentally, Fig. 10. Liquid above the heater was mixed by a needle, briefly dipped into the layer. The intensity of this mechanical effect should exceed significantly the sequences of cluster coalescence. As a result of mechanical mixing of the layer, the temperature on the interphase decreases by $>6^\circ$; however, the heat field restored during two seconds. It is important to note that temperature on the interphase restores after cluster destruction one order slower; this requires $\sim20$ s, Fig. 7.

Correlation between dependence $T_{S,\text{max}(t)}$ and formation of the droplet cluster, Fig. 7, proves a significant effect of the dissipative structure on liquid evaporation and heat and mass transfer in the gas medium near the interphase.
FIG. 9: Dependence of liquid volume injected into the layer at cluster coalescence on the interphase temperature, rated by the volume of liquid layer region with the area equal to the heater area ($V_{\text{heater}} = \pi R_{\text{heater}}^2 h_0$).

FIG. 10: Process of restoration of the heat field after mechanical mixing of the layer.

Perhaps a key role is played by the following features of the cluster: (i) active condensation growth of droplets increases the gradient of vapor concentration near the interphase, which intensifies layer evaporation; (ii) the evident temperature gradient on droplet surfaces makes probable development of thermocapillary flows in these droplets, which can effect the structure of the vapor-air jet above the heated region of the layer [see also Arinshtein and Fedorets (2010)]. Data obtained with the help of the thermal imager allow an estimate of the temperature gradient on the surface.
of large droplets, Fig. 11. Points in distribution below the dashed line correspond to the regions of droplet surface (near its top), whose inclination toward the focal plane of the lens does not exceed 35 deg. The temperature in these regions is measured correctly, and it follows from distribution that the temperature gradient along the droplet surface is $\sim 30^\circ/\text{mm}$ ($1.4^\circ/0.045 \text{ mm}$). The estimation of the temperature gradient in the gas phase over the heated zone of the liquid also gives very high values, up to 40 K/mm.

Under the conditions of experiment performed at $T_{S\text{ max}} \sim 90^\circ$, the qualitative reconstruction of heat and mass transfer processes in the gas medium near the interphase occurs. The interdroplet distances increase significantly together with the mass of droplets, which can levitate above the interphase. With an increase in $T_{S\text{ max}}$ from 86.6$^\circ$ to 92.8$^\circ$, a fourfold growth of droplet mass is observed at the moment of cluster coalescence, whereas the heater power increases by $<1.5$ times from 0.49 to 0.72 W. The effect of the droplet cluster on the layer temperature also changes principally. When $T_{S\text{ max}} < 90^\circ$, cluster formation decrease the temperature on the interphase, which indicates intensification of evaporation or convective heat transfer between liquid and gas phase. This effect becomes more intensive with a rise of heater power, Fig. 8 (the positive temperature jump at coalescence of the droplet cluster). If $T_{S\text{ max}} > 90^\circ$, we can observe the reverse tendency, and at $T_{S\text{ max}} = 92.8^\circ$, maximal within the studied range, the temperature on the interphase under the droplet cluster increases, which proves heat transfer deterioration. This issue requires further study, but the most likely cause of the slight deterioration of heat transfer is the conversion of steam-air flow around the drop cluster associated with the development of thermocapillary flow in large droplets (Arinshtein and Fedorets, 2010). These flows can generate around droplets’ toroidal vortices that disimprove the outflow of hot gas from the surface layer.

5. CONCLUSION

Here, we present the results of experimental investigation of the mechanism of natural collapse of the spatially ordered structure, consisting of tens of microdroplets levitating parallel to the horizontal liquid-gas interphase at a distance comparable to the droplet diameter. These results were obtained via high-speed thermal imaging. It is found out that the reason for destruction of the whole “droplet cluster” can be coalescence of a single cluster droplet with the liquid

FIG. 11: Radial distribution of temperature over the surface of droplets with the diameter above 160 $\mu$m. Marker numbers 1–3 correspond to droplet numbers in the IR image of the cluster; marker 4 — distribution averaged by three droplets.
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Cluster coalescence occurs during 3 ms. Concentric symmetry of the wave of cluster destruction and velocity of destruction front of $\sim 70$ cm/s prove that the process of cluster destruction relates to generation of the capillary wave on the interphase, caused by the fall of the first droplet. It is assumed that the amplitude of the capillary wave exceeds the height of droplet levitation $H$ or is comparable to it.

It is determined that instantaneous coalescence of a cluster is accompanied by a temperature jump on the interphase; this jump equals tenths of a degree. With cluster restoration, the temperature of the layer surface returns gradually to the initial level. The period of temperature restoration is measured by tens of seconds. The interphase temperature under the droplet cluster can be both higher and lower than the interphase temperature without cluster, i.e., the cluster can both intensify and deteriorate heat and mass transfer between liquid and gas.

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