

Article Heat and Mass Transfer Processes and Evaporation of a Liquid Droplet on a Structured Surface

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Abstract: The characteristics of water droplet heating and evaporation on structured hydrophobic and hydrophilic surfaces in the range of static contact angles from 73° to 155° were studied experimentally using high-speed video recording. Two fundamentally different technologies for applying coatings on a metal surface were used in comparison with the results on a polished surface. Microscopic studies were conducted to identify the features of the formed coatings. The wetting properties were characterized by means of the static contact angle and the contact angle hysteresis: on polished surface No. 1 (contact angle-73°, hysteresis-11°), on structured surface No. 2 (contact angle—125°, hysteresis—9°), and on structured surface No 3 (contact angle—155°, hysteresis—7°). The experimental dependences of the droplet evaporation rate on the different surfaces under normal conditions (ambient air temperature—293 K, atmospheric pressure, humidity—35%) were obtained. The evaporation regimes of droplets on the surfaces under study were identified. Water droplets evaporated in the pinning mode on surfaces No. 1 and No. 2. When a water droplet evaporated on surface No 3, the droplet was in the constant contact angle regime for \approx 90% of its lifetime. Based on the experimental data obtained, a two-dimensional model of conjugate heat and mass transfer was developed, which describes the heating and evaporation of a liquid droplet on structured hydrophobic and hydrophilic surfaces at a wide range of contact angles. Satisfactory agreement was obtained between the numerical simulation results and experimental data. Using the model, the fields of temperature, concentration and other key characteristics were established at different points in time. Recommendations for its application in the development of gas-vapor-droplet applications were formulated.

Keywords: hydrophobic surfaces; hydrophilic surfaces; superhydrophobic surfaces; liquid droplet; conjugate heat and mass transfer; evaporation; model

1. Introduction

The interaction of liquid droplets with solid surfaces is widespread in technical systems [1,2] and natural phenomena [3,4]. These interactions are implemented in the form of various regimes. The most interesting regimes are interactions with the formation of a liquid film on the surface [5], rebound of a droplet from it without breakup [6], and with the formation of secondary fragments [5,6]. In some technological directions, the interaction of liquid droplets with the surfaces of units, aggregates, and assemblies significantly changes the operational characteristics of their work [7–9]. Often, these changes critically affect the conditions for the safe operation of units, aggregates, assemblies, and systems [7–9]. One of the most important directions in this regard is associated with the development of affordable technologies for modifying the coatings of units, aggregates, assemblies, and devices that overheat during their operation [7,10]. This creates unsafe technological conditions. In this case, structured coatings are needed [10].

In order to impart unique near-surface properties, including the possibility of increased heat removal per unit area, various methods for modifying heating surfaces are being



Citation: Antonov, D.V.; Islamova, A.G.; Orlova, E.G. Heat and Mass Transfer Processes and Evaporation of a Liquid Droplet on a Structured Surface. *Energies* **2023**, *16*, 7505. https://doi.org/10.3390/en16227505

Academic Editors: Jose A. Almendros-Ibanez, Agustin Valera-Medina, Pavel Skripov and Aleksandr Pavlenko

Received: 8 September 2023 Revised: 10 October 2023 Accepted: 6 November 2023 Published: 9 November 2023



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developed: chemical etching [11], milling [12], additional coatings [13], laser processing [14], etc. Review articles devoted to the study of the influence of the properties [15,16] and geometry of surfaces [16] have been published. Compared to a polished surface, rough, microstructured (lithography method) and hybrid micro-/nano-engineered (lithography method and ZnO nanowire growth) surfaces have the potential to increase the critical heat flux by 15%, 42%, and 59% [17]. Additionally, the overall heat transfer coefficient in a cooling system based on spraying increases by 14%, 28%, and 42%, respectively [17]. It was shown that the evaporation rate increases on surfaces characterized by increased wettability due to spreading and convection [18]. The deposition of SiO_2 and carbon nanotubes on silicon surfaces led to an increase in the critical heat flux by 75.3% compared to the unmodified surface [17]. Microspray-based cooling on three nanotextured surfaces was studied [19]. It was found that the wettability of the surface and the liquid spreading were the reasons for the heat transfer enhancement [19]. When water boils on a nanoporous copper surface (the pore size is 50–200 nm), the wall overheating was found to decrease by 63%, and the heat transfer coefficient was found to increase by 173% compared to the polished surface [20]. Using the lithography method, fins 5 to 100 μ m wide and 5 to 50 μ m high were deposited on the silicon surface. This made it possible to increase the heat transfer coefficient by a factor of 1.4–1.7 compared to the polished surface [21]. It was substantiated that hydrophobic dots (260 µm in diameter) on a hydrophilic surface increase the critical heat flux and heat transfer coefficient by 65% and 100%, respectively [22]. Laser texturing of steel made it possible to increase the heat transfer coefficient by a factor of four compared to the untreated surface [23]. Using a femtosecond laser, self-organizing mound-like structures were fabricated on stainless steel surfaces. The treated surfaces demonstrated increased wettability and thermal conductivity (by a factor of 1.6 compared to the untreated surface) [24]. Narrow channels formed in closely spaced structures, which increased the capillary penetration of the liquid under the action of high heat fluxes, significantly increased the critical heat flux. It was shown that an increase in surface roughness (a change in the arithmetic mean surface profile from 0.528 μ m to 25.709 μ m) led to an increase in the critical heat flux from 176 kW/m^2 to 317 kW/m^2 [25]. The use of a capillary-porous coating was found to lead to a significant increase in the heat transfer up to 3.5 times when water boiled at low heat fluxes [26].

The heating and evaporation of sessile droplets have been thoroughly studied [27]. The wide array of applications for commercial and industrial purposes, as well as the interests of fundamental research, are the factors that draw the focus of numerous researchers to these issues [27,28]. The most interesting effects of droplet evaporation for certain applications are recorded on superhydrophobic and superhydrophilic surfaces, which have received significant interest in recent decades [27,28]. The mechanisms of sessile droplet heating and evaporation established during mathematical modeling were presented in [27,29–33]. In [27,32], known models of heating and evaporation of sessile droplets were reviewed. In [27], the problem of conjugate heat and mass transfer in a droplet placed on a substrate in an air environment was solved numerically. The impacts of several factors—namely, the boundary layer thickness, substrate temperature, surface wettability, temperature and humidity of air environment—on heat and mass transfer in evaporating droplets were studied using the volume of fluid (VOF) method in [32,34–38], the lattice Boltzmann method in [33,39,40], and molecular dynamics modeling in [41]. In particular, the evaporation characteristics of a sessile droplet under forced convection were studied in [32]. In addition, the lifetimes of sessile droplets on heated surfaces were calculated [32]. The mathematical model provides a highly precise prediction of the lifetime of a droplet evaporating under forced convection. The primary constraint of the model [32] resides in the presumption of the quasi-stationarity of heat and mass transfer in direct numerical simulation to mitigate computational expenses. The Marangoni effects on droplets on superhydrophobic surfaces were described in [38,42], including an analytical solution for thermocapillary flow as a function of stream and vorticity. The work [42] compared the simulation and experimental results, while also providing an estimation of the effective heat transfer coefficients. In [41], a model based on molecular dynamics methods was developed to explain the wetting properties of a three-dimensional droplet placed on a textured surface.

Unfortunately, there are still not enough achievements in the field of intensifying heat removal from a heated surface to solve urgent technological problems in the direction of increasing the efficiency of power equipment. The problems associated with the heat transfer of droplets on structured surfaces are of particular interest. To solve them, fundamentally new models of conjugate heat and mass transfer are required that make it possible to predict the characteristics of heat and mass transfer and phase transformations when varying the input parameters over a wide range. The lack of relevant models motivated the present study. The purpose of this work is to develop, based on experimental data, a model of conjugate heat and mass transfer, considering the evaporation of a liquid droplet on a solid surface with contact angles varied over a wide range (from a hydrophilic to a hydrophobic state). This model is necessary to predict the characteristics of the heating and evaporation of liquid droplets with different component compositions on modified surfaces.

2. Experimental Research

2.1. Materials

An aluminum–magnesium alloy (wt. %: Mg 3.2–3.8; Mn 0.3–0.6; Si 0.5–0.8; Ti up to 0.1; Cu up to 0.1; Zn up to 0.2; Fe up to 0.5; Al—the rest) was used in the experiments in the form of plates 30×30 mm in size and 1 mm thick. This alloy is widely used in aviation, automotive and shipbuilding. Three types of substrates were prepared with different characteristics in terms of the metal surfaces, differing in the roughness (texture) and wetting properties.

Surfaces named as No. 1 were polished according to a proven procedure [43] using diamond pastes (ACM 40/28 HOM, ACM 14/10 HOM, ACM 5/3 HBOM, ACM 1/0 HBOM) and felt materials. After the polishing procedure, the surfaces were cleaned of the products of the diamond pastes and felt materials in an ultrasonic bath successively in ethyl alcohol and ultrapure water. Cleaning characteristics: operating frequency 35 kHz, cleaning time in each medium 5 min, medium temperature corresponded to the laboratory (299–231 K). After cleaning, the surfaces were dried naturally for at least 24 h in an air environment corresponding to laboratory conditions. In order to form hydrophobic properties, the surfaces named as No. 2 and No. 3 were modified by nanosecond laser radiation (ytterbium pulsed fiber laser (IPG Photonics, Moscow, Russia)) followed by low-temperature annealing. The essence of this approach to hydrophobization is the adsorption of hydrocarbon compounds (non-polar compounds) from the environment [44,45] onto the developed roughness activated for the deposition of non-polar compounds. Differences in the textures and wetting properties of the surfaces were obtained by varying the beam linear speed, number of lines, and the number of laser beam passes. A more detailed description of the methods for obtaining the studied surfaces is presented in [46–48].

The static contact angle (θ) and hysteresis (θ_{Δ}) were determined for a 5 µL water droplet on surfaces No. 1–3 from shadow images obtained using the shadow optical method equipment implemented in the DSA 25S (Kruss, Germany) setup and the Tangent-1 method in ADVANCE 1.16.0 software, which is recommended in the range of contact angles from 1° to 180°, with an accuracy $\pm 0.1^{\circ}$. In addition, the measurements in this work were conducted until the contact angle reached a value of less than 6°. When determining θ and θ_{Δ} , the experiments were repeated at least three times under identical conditions. The error in determining the static contact angle and the contact angle hysteresis did not exceed 5%. According to the measurement results, it was found that surface No 1 was hydrophilic ($\theta \approx 73^{\circ}$, $\theta_{\Delta} \approx 11^{\circ}$) and surfaces No. 2 and 3 were hydrophobic ($\theta \approx 125^{\circ}$, $\theta_{\Delta} \approx 9^{\circ}$ and $\theta \approx 155$, $\theta_{\Delta} \approx 7^{\circ}$, respectively). Note that the classification of hydrophilic and hydrophobic surfaces in this paper was based on the assumption that a material is considered hydrophilic if the contact angle is less than 90°, and vice versa, it is considered hydrophobic if the contact angle is greater than 90° [49–51]. The contact angle hysteresis was obtained using the inflating and deflating a sessile drop method [52], and assuming that the droplet volume is 5 μ L (the droplet contact radius is less than the capillary constant of distilled water, which is 3.8 mm), the liquid flow rate determining the contact line speed of 0.05 μ L/s was taken according to a prior recommendation [53]. The recorded advancing and receding contact angles on polished surface S1 were 76° and 65°, respectively. The texture microrelief was studied by means of scanning electron microscopy using a Hitachi S-3400N microscope (Hitachi, Tokyo, Japan). To evaluate the roughness, an Olympus Lext OLS4100 laser scanning microscope (Olympus NDT, Tokyo, Japan) was used. The arithmetic mean deviation Sa of surface No. 1 was 0.31 μ m, No. 2 was 2.3 μ m, and No. 3 was 16.30 μ m. However, the recorded roughness parameters were not taken into account in the modeling due to the accepted assumptions about static contact angles, which indirectly are the basic indicators of wettability, and also taking into account the surface energy and its roughness. Figure 1 presents SEM images of the surfaces.





200 µm



Figure 1. Cont.



Figure 1. SEM images of the surfaces: (a)—polished aluminum alloy (surface No. 1); (b)—hydrophobic aluminum alloy (surface No. 2); and (c)—superhydrophobic stainless steel (surface No. 3).

2.2. Experimental Technique

Experimental studies of water droplet heating and evaporation on the studied surfaces were conducted using the DSA 25S setup and the HC4210 humidity control chamber included in it (Figure 2). The chamber was used to maintain the constant temperatures of the surface (T_{sub}) and medium (T_g) equal to 293 K, as well as constant humidity (35%). A 5 μ L droplet was placed on the surface at an arbitrarily chosen point using an automatic dosing system of the DSA 25S setup. The initial droplet temperature was equal to 293 K. Using a high-speed video camera, the water droplet evaporation was recorded. The shooting speed was 50 fps at a resolution of 1200×1200 pix. The experiments on water droplet evaporation were conducted at least five times. The droplet lifetime (τ_h) was determined from a video recording of the evaporation dynamics. It was determined that the systematic error of τ_h was no more than 4%. At the same time, it was previously established [54] that the maximum relative error in determining the geometric characteristics of an evaporating liquid droplet using the software of the DSA installation does not exceed 5%.



(a)

Figure 2. Cont.



Figure 2. Scheme (**a**) and photo image (**b**) of the experimental setup: 1—light source; 2—high-speed video camera (image type—monochrome; maximum resolution 1920×1080 pix; maximum shooting speed— 4×10^4 fps); 3—droplet; 4—surface; 5—isolated zone; and 6—stand.

The video fragments obtained from the results of the experiments were processed using the ADVANCE 1.16.0 software of the DSA 25S setup, as well as using the authors' software codes in MatLab (Figure 3).



Figure 3. Typical images presenting processing procedure video frames in the MatLab software 2023a.

In the MatLab software package, the received video frames were converted into a sequence of 8-bit black and white images. At the next stage, the obtained images were binarized; the binarization threshold was selected for each series separately. The droplet radius expressed in pixels was determined by equating it to the radius of a circle with the same area as the area on the binarized image that the droplet occupied. While processing, any change in the contact angles, volume, contact diameter and droplet radii in time were recorded. The droplet radii R_d at each time step were analyzed as $(3 \times V_d/(4 \times \pi))^{1/3}$, where V_d is the droplet volume. The contact diameter was assumed to be the distance between extreme points on the surface in the case of a binarized image. This approach allows determining reliably the contact line geometric dimensions. Moreover, the systematic

errors of the contact diameter and droplet radii were more than 0.02 mm. The random error Δ_r was estimated as:

$$\Delta_{\mathbf{r}} = t(\alpha, n_{\mathbf{m}})S,\tag{1}$$

where $t(\alpha, n_m)$ is the Student's coefficient, depending on the number of measurements n_m and confidence level α , taken as equal to 0.95; *S* is the root squared deviation.

Figure 4 presents typical frames of water droplets evaporating under natural conditions on polished and hydrophobic surfaces.



Figure 4. Typical frames demonstrating water droplets evaporating on the studied surfaces ($T_{sub} = T_g \approx 293$ K and humidity 35%) with varying surface wettability: (**a**)— $\theta \approx 73^\circ$; (**b**)— $\theta \approx 125^\circ$; and (**c**)— $\theta \approx 155^\circ$.

It was found that during the evaporation of water droplets on polished (No. 1) and hydrophobic (No. 2) surfaces, the pinning mode was observed most of the time due to the high adhesion to the surface. The contact diameter remained constant and the contact angle decreased. At a certain point in time (when more than $\approx 85\%$ of the liquid was evaporated), the surface tension forces began to prevail over all the other forces (viscosity, gravity, friction, etc.) and the contact area began to shrink, and the depinning mode set in. During the evaporation of a water droplet on the hydrophobic surface (No. 3), the liquid droplet was in the constant contact angle mode for $\approx 90\%$ of its lifetime, and its diameter decreased. After reaching this time, the values of the contact diameter and the droplet diameter decreased monotonically.

The behavior of a droplet evaporating on smooth hydrophilic and hydrophobic surfaces has been widely discussed in the literature. In particular, in [55], water droplet evaporation was studied by varying the wettability of surfaces (the static contact angles varied from 10° to 150°). Satisfactory agreement with Popov's diffusion model [56] was substantiated for the evaporation of a droplet on a superhydrophobic surface in the pinning mode. It was shown that the diffusion-only model can predict the characteristics of droplet evaporation on a hydrophobic surface [57]. Gleason and Putnam [58] reported forced contact line pinning in evaporation experiments. The results were compared with those obtained on the basis of the diffusion model developed based on Popov [56] and Deegan et al. [59]. When solving the problems of droplet evaporation on surfaces with a small hysteresis and roughness, and also when realizing the constant contact angle mode, known evaporation models that take into account only diffusion do not allow for increasing the reliability of theoretical studies. In [30], it was shown that the model based only on vapor diffusion predicts with sufficient accuracy the total evaporation time of droplets on a hydrophobic surface. However, in the case of a superhydrophobic surface, the vapor diffusion model significantly overestimates the evaporation rate. As a result of experimental studies, we found that the surfaces used can be conditionally divided into three types: smooth hydrophilic—surface No. 1, rough hydrophobic—surface No. 2 (during evaporation the contact line is pinned), and rough hydrophobic—surface No. 3 (contact diameter decreases). Thus, this work will consider the agreement of the obtained model, which includes the equations for energy, vapor diffusion during the evaporation of a droplet in a gas—air medium, and transfer, with the experimental results when implementing various interaction modes. Note that in this paper, the experimental results are presented only under natural conditions, which is sufficient for validating the developed numerical model, since under natural conditions, as a rule, all the subtle effects of the droplet interaction with the surfaces are manifested, including the effects of cooling, Marangoni convection, natural convection, etc. In the future, it is planned to conduct additional studies in wide temperature ranges in order to refine the model. The established effects and quantitative data from the experiments were used to formulate a physical and mathematical model for the heating and evaporation of a liquid droplet on a modified surface.

3. Physical and Mathematical Model

Figure 5 presents the solution domain of a two-dimensional axisymmetric model of conjugate heat and mass transfer, which describes the heating and evaporation of a liquid droplet on structured hydrophobic and hydrophilic surfaces at a wide range of contact angles. The main driving factor of heat transfer in this paper is evaporation. The cases of droplet evaporation on different surfaces (with different contact angles) under normal conditions (temperature of the external gas-air medium 293 K, atmospheric pressure and humidity 35%) are considered. It was assumed that the vapor concentration at the droplet interface was in local equilibrium as determined by the saturated vapor density, which depends on the surface temperature. The parameters of the unstructured computational grid: the number of elements was 25,000, the quality of the elements was 0.9. The number of grid elements increased until the difference as a result of calculations did not exceed 1%. In each element, the approximating function type was chosen. In the first approximation, it is legitimate to use a linear approximation (the first-degree polynomial). The approximating function is zero when outside its element. The solution to the heat and mass transfer problem was obtained by evaluating the functions at the boundaries of the elements. The coefficients of the approximating functions were calculated from the equality condition of the values of neighboring functions at the boundaries between elements [60,61]. An illustration of the mesh independence study is presented in Figure 6.



Figure 5. Scheme of the solution domain (**a**) and grid (**b**) for a two-dimensional axisymmetric problem of conjugate heat and mass transfer during heating and evaporation of a liquid droplet.



Figure 6. Mesh independence study of the dynamics of the change in the normalized droplet volume with time ($U_a = 0 \text{ m/s}$, $T_{sub} = T_g = T_0 = 293 \text{ K}$, surface No 2 and humidity of 35%): 1— $N_m = 20,000$ mesh elements; 2— $N_m = 25,000$ mesh elements; and 3— $N_m = 30,000$ mesh elements.

The system of non-stationary differential equations included the energy equation with initial and boundary conditions (2)–(8), the equation of vapor diffusion during the evaporation of a droplet in a gas–air medium with initial and boundary conditions (9)–(14) and the transfer equations with initial and boundary conditions (15)–(20) with the corresponding initial and boundary conditions. When implementing this system of equations in COMSOL Multiphysics, the following modules were used to track the evaporation front: Two-Phase Laminar Flow, Transport of Diluted Species, Heat Transfer in Solids and Fluids, Nonisothermal Flow, and Moving Mesh. The solution convergence criteria in COMSOL Multiphysics was based on the relative tolerance, which was chosen as 10^{-6} .

The temperature distribution in space and time was described by the non-stationary energy equation:

$$\frac{\partial T}{\partial t} + \left(u\frac{\partial T}{\partial r} + w\frac{\partial T}{\partial z}\right) = \kappa \left(\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) + \frac{\partial^2 T}{\partial z^2}\right) + \mu\Phi,$$
(2)

where *T*—temperature, *t*—time, *u* and *w*—velocity projections along the *r* and *z* axes, respectively, and κ , *k*, *c*, μ and ρ —thermal diffusivity, thermal conductivity, heat capacity, dynamic viscosity, and fluid density in the cell, respectively.

The dissipative function Φ in a cylindrical coordinate system was determined using the equation:

$$\Phi = 2\left(\frac{\partial u}{\partial r}\right)^2 + 2\left(\frac{u}{r}\right)^2 + 2\left(\frac{\partial w}{\partial z}\right)^2 + \left(\frac{\partial u}{\partial r} + \frac{\partial w}{\partial z}\right)^2 \tag{3}$$

Initial conditions:

$$T|_{t=0} = T_0 = T_g. (4)$$

Boundary conditions:

$$\left. \frac{\partial T}{\partial r} \right|_{r=0} = 0 \text{ on the symmetry axis,}$$
(5)

$$2\pi Rk_{\rm d} \frac{\partial T}{\partial R}\Big|_{R=R_{\rm d}} = h(T_{\rm eff} - T_{\rm s}) \text{ on the water droplet/gas boundary,}$$
(6)

$$-h\left(\frac{\partial T}{\partial z}\right)\Big|_{z=0} = 0 \text{ on the bottom wall,}$$
(7)

$$T|_{r=20R_{d0}} = T_g$$
 and on the outer boundary of the solution area, (8)

where $R = \sqrt{r^2 + z^2}$, $T_{\text{eff}} = T_{\text{g}} + \frac{\rho_{\text{d}}L_{\text{d}}R_{\text{d}}}{h}$, *h*—heat transfer coefficient, $R_{\text{d}(\text{e})} = J_{\text{d}(\text{e})}/\rho_{\text{d}}$ —evaporation rate in m/s, $J_{\text{d}(\text{e})} = M_{\text{d}}n \cdot \left(-D \cdot \left(\frac{\partial c}{\partial r} + \frac{\partial c}{\partial z}\right)\right)$ —mass evaporation rate in kg/(m²·s), M_{d} —molar mass of water, *n*—normal vector to the droplet surface, *D*—water vapor diffusion coefficient, *c*—molar volume concentration of water vapor in mole/m³, L_{d} —vaporization heat of water, and T_{g} —gas temperature.

The distribution of components (water vapors and air) in the gas–air medium was described by differential Equation (9). This equation was used to determine the water vapor concentrations above an evaporating droplet.

$$\frac{\partial Y_{i}}{\partial t} + \left(u\frac{\partial Y_{i}}{\partial r} + w\frac{\partial Y_{i}}{\partial z}\right) = D\left(\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial Y_{i}}{\partial r}\right) + \frac{\partial^{2}Y_{i}}{\partial z^{2}}\right),\tag{9}$$

where $i \ge 1$, *D*—water vapor diffusion coefficient, *Y*—mass concentration, i = 1—water, and i = 2—air.

Initial conditions:

$$Y|_{t=0} = Y_0 \tag{10}$$

where index 0—initial conditions. It was assumed that Y_0 is determined by the initial humidity in a room (30%).

Boundary conditions:

$$\left. \frac{\partial Y}{\partial r} \right|_{r=0} = 0 \text{ on the symmetry axis,}$$
(11)

 $Y = Y_{\text{sat}}$ on the water droplet/gas boundary, (12)

$$-D\left(\frac{\partial Y}{\partial z}\right)\Big|_{z=0} = 0 \text{ on the bottom wall,}$$
(13)

$$|Y|_{r=20R_{d0}} = Y_{inf} = Y_0$$
 on the outer boundary of the solution area, (14)

where Y_{sat} —mass *concentration* of water vapors under saturation conditions.

The recirculation of convective flows was described by a system of equations consisting of the continuity Equation (15) and Equations of motion (16) and (17) in the r and z directions.

$$\frac{\partial \rho}{\partial t} + \frac{\rho}{r} \left[\frac{\partial (ru)}{\partial r} + \frac{\partial (rw)}{\partial z} \right] = 0.$$
(15)

$$\frac{\partial u}{\partial t} + u\left(\frac{\partial u}{\partial r}\right) + w\left(\frac{\partial u}{\partial z}\right) = -\frac{1}{\rho}\left(\frac{\partial p}{\partial r} + \mu\left(\left(\frac{\partial u}{\partial r} + \frac{\partial u}{\partial z}\right) + \left(\frac{\partial u}{\partial r} + \frac{\partial u}{\partial z}\right)^T\right)\right) + \rho g + F_{\rm st},\tag{16}$$

$$\frac{\partial w}{\partial t} + u\left(\frac{\partial w}{\partial r}\right) + w\left(\frac{\partial w}{\partial z}\right) = -\frac{1}{\rho}\left(\frac{\partial p}{\partial z} + \mu\left(\left(\frac{\partial w}{\partial r} + \frac{\partial w}{\partial z}\right) + \left(\frac{\partial w}{\partial r} + \frac{\partial w}{\partial z}\right)^{\mathrm{T}}\right)\right) + \rho g + F_{\mathrm{st}},\tag{17}$$

where $\rho = \rho(p_{ref}, T_{ref})$ —fluid density, *p*—pressure, μ —dynamic viscosity, *g*—gravitational constant, superscript *T*—transposition, subscript ref—reference value, and *F*_{st}—surface tension force.

Initial conditions:

$$u|_{t=0} = 0, \ w|_{t=0} = 0, \ p = p_{g} \text{ in gas,}$$

$$p = p_{g} + \frac{2\sigma}{R_{d0}} \text{ in droplet,}$$
(18)

where $p_g = 101,325$ Pa and σ —surface tension of water.

Boundary conditions:

$$\left(\frac{\partial u}{\partial r}\right)\Big|_{r=0} = 0, \left(\frac{\partial w}{\partial r}\right)\Big|_{r=0} = 0 \text{ on the symmetry axis,}$$
(19)

$$u|_{R=R_{d}} = 0, w|_{R=R_{d}} = 0$$
 (no slip) on the solid surface. (20)

At the droplet/gas interface, the dependence of the surface tension on the temperature was set, resulting in Marangoni recirculation (21):

$$\sigma(T)| = \sigma_0 - \left|\frac{\mathrm{d}\sigma}{\mathrm{d}T}\right|(T - T_0).$$
(21)

In contrast to the models [62,63], the convective and interfacial transport was additionally taken into account, and the possibility of varying the contact angles over a wide range was provided. An adaptive time step was used and was set in the range of 0.000001–0.5 s. The Deformed Geometry block used the Yeoh smoothing type to represent the movement of the evaporation front. Computer characteristics: processor Intel(R) Xeon(R) CPU E5-2697 v3 @ 2.60 GHz 2.60 GHz (number of processors—2), installed RAM 128 GB. The typical computation time was from 2 to 5 h.

Table 1 presents the properties of water and air used in the calculations [64].

Table 1. Properties of water and air used in the calculations [64].

Material	Physical Properties
	$M_l = 0.018015 \text{ kg/mol};$
	$ ho_l = 325 \cdot 0.27^{-[1-(rac{T}{647.13})]^{0.23}} \text{ kg/m}^3;$
	$C_l = \left(-2.2417 \cdot 10^4 + 876.97 \cdot T - 2.5704 \cdot T^2 + 2.4838 \cdot 10^{-3} \cdot T^3\right) / 18.015 \text{ J} / (\text{kg} \cdot \text{K});$
	$k_l = -0.35667 + 5.057 \cdot 10^{-3} \cdot T - 6.1071 \cdot 10^{-6} \cdot T^2 \text{ W}/(\text{m} \cdot \text{K});$
Water Air	$\mu_l = 10^{-11.6225 + 1949/T + 0.021641 \cdot T - 0.00001599 \cdot T^2} \cdot 0.001 \text{ Pa} \cdot \text{s};$
	$L = \frac{54000 \cdot (1 - T / 647.13)^{0.34}}{18.015} \text{ J/kg;}$
	$P_{sat} = 10^{8.05573 - 1723.6425/(T - 40.07)} \cdot 133.322387415$ Pa;
	$D_v = (-0.29890 + 0.0016253 \cdot T + 0.00000075 \cdot T^2) \cdot 0.0001 \text{ m}^2/\text{s}.$
	$M_a = 0.028951 \text{ kg/mol};$
	$C_a = \left(-0.000000048325 \cdot T^3 + 0.000013106 \cdot T^2 - 0.0051373 \cdot T + 29.643\right) / 0.028951 \text{ J} / (\text{kg} \cdot \text{K});$
	$\mu_a = \left(0.00000090437 \cdot T^3 - 0.00037287 \cdot T^2 + 0.70077 \cdot T + 4.5608\right) \cdot 10^{-7} \text{ Pa} \cdot \text{s};$
	$k_a = \begin{pmatrix} 0.0000000017429 \cdot T^3 - 0.000000054199 \cdot T^2 + \\ +0.00010311 \cdot T - 0.00038603 \end{pmatrix} W/(m \cdot K).$

Figure 7 presents typical temperature fields and velocity vectors of the convective flows for three cases corresponding to the input parameters of the experiments. The results of these experiments are presented in Figure 4.



Figure 7. Temperature fields and velocity vectors of the convective flows for three surfaces ((**a**)—No. 1, (**b**)—No. 2, (**c**)—No. 3) corresponding to the parameters of the experiments in Figure 4.

When analyzing the temperature and convection velocity fields in Figure 7, the following key patterns were highlighted:

(i) The lowest temperature values correspond to the upper near-surface layer of the droplets throughout the entire evaporation time and over the entire range of contact angles. This is due to the fact that an intense heat sink is formed in the direction of a droplet's upper free surface relative to its lower surface in contact with the heated solid surface. A nonmonotonic decrease in temperature from the lower to the upper near-surface layer was recorded due to the intensification of the convective heat and mass transfer. Under the conditions considered, it was convection that significantly affected the temperature distributions in the droplet volume.

(ii) The highest temperatures are observed near the contact of the droplet with the surface, provided that the temperature of the latter is kept constant (i.e., with compensation for the heat sink due to the droplet evaporation). By varying the values of the input parameters, it was found that in order to ensure the balanced heating of the droplet under conditions of intense evaporation and convective heat and mass transfer, it is necessary to maintain the heat flux from the heated surface quite flexibly. If the heat fluxes are small, then almost all of the supplied thermal energy will be spent on the endothermic phase transition. If the heat flux is high enough, intense evaporation will lead to an increase in the thickness of the vapor buffer zone between the droplet's lower surface and the heated substrate. By controlling the fields of temperature and convection rate, it is possible to provide a mode of the monotonic decrease in the droplet size during evaporation.

(iii) The Marangoni effect appears due to the temperature difference on the droplet's surface. The Marangoni effect leads to the transfer of substance along the interface between two media due to the presence of a surface tension gradient. When this effect is manifested, a toroidal vortex appears in the droplet. During evaporation, a vapor buffer zone is formed around the droplet, which is involved in the transformation of the temperature field above the droplet and inside it. During modeling, the influence of the near-surface layer characteristics on these effects was established.

(iv) Over time, the droplet size decreases due to the movement of the heating and evaporation fronts in the direction of the deep layers. The nonlinearity of the dependence of the main characteristics of this process on time increases with an increase in the substrate temperature. The longer this process takes, the weaker the nonlinearities become. This is due to the droplet's temperature reaching certain asymptotic values due to the high role of the heat capacity and vaporization heat of water. If the values of these parameters for a liquid (for example, for combustible and easily flammable compositions) are small relative to water, and the boiling point is higher, then the temperature of the liquid droplet grows quite rapidly almost throughout the entire heating time.

The established patterns, even under natural environmental conditions, indicate the significant non-uniformity of the temperature fields in the volume of droplets and in their small vicinity in a gas-air medium. They play an important role in practical applications (cooling surfaces with sprays and aerosols, drying droplets, etc.). In particular, assumptions regarding the uniform temperature field of a droplet are often formulated when predicting the characteristics of its heating and evaporation. Especially often, such assumptions are formulated when estimating the evaporation rates of liquids on surfaces in the form of a supplied spray. The calculations performed show that the corresponding estimates are justified only in the region of small droplet sizes (less than 0.1 mm) and high rates of energy supply to them from the surface. Accordingly, the duration of complete evaporation (the term "lifetime" is used) is less than 1 s. The larger the droplet size and the lower the supplied heat flux, the greater the importance of considering the effects of droplet spreading and changes in the contact angle with the surface, and it is also necessary to take into account the processes of liquid boiling and the formation of a vapor buffer zone between the surface and liquid. In this case, the additional effects of the heat transfer of a liquid with a solid surface consist of the detachment of both the entire volume of the liquid from the surface in the form of the Leidenfrost effect [39,65,66] and small volumes of liquid in the puffing and micro-explosion modes [67–69]. The identified effects are characterized by the detachment of liquid fragments from the parent initial droplet. These fragments have different sizes and temperatures. Their detachment from the parent droplet leads to a shortterm and abrupt change in the energy balance in the near-surface layers of the droplets. These droplets, as a rule, are in a metastable state. Modeling of heat and mass transfer under such conditions is important to conduct with simultaneous consideration of hydroand gas-dynamic processes and control of the volume of liquid and vapor. Accounting for the number, volume, speed, and trajectory of secondary fragments, which are called child droplets, is of particular difficulty. Accounting for all these factors and processes in the model requires large computing power and leads to a nonlinear increase in the duration of the calculations. Therefore, it is important to consistently connect certain effects in the models, taking into account existing ideas about the studied processes from experiments.

Figure 8 presents trends that show the rate of change in the normalized droplet volume for the three surfaces used. From the view point of the importance of applications in technologies, the main pattern recorded in the experiments was a decrease in the evaporation rate of the droplets with an increase in the contact angle. This is mainly due to a decrease in the specific amount of thermal energy supplied to the droplet from a solid surface due to the lower values of the droplet contact diameter and the absence of additional flows in the gas–air medium. It is shown that the liquid evaporation rate changes several times due to the modification of the material surface. In this case, it is possible to change the behavior of the dependence of the rate on the time and changing heat flux. This factor is very important in controlling the heat exchange between a liquid and a solid surface. Quite often, it is required to adaptively change the heat transfer conditions when the liquid film thins in order to prevent surface drying or, conversely, excessive growth of the liquid film's thickness on the surface. The importance of these processes increases with the transition to micro- and nanosized liquid droplets and films due to the significant increase in the heat and mass transfer rates in a liquid and gas–air medium. The developed model allows for organizing the transition to the appropriate size ranges. When compared with the obtained experimental data in the next section, we present certain limits of the applicability of the developed model. Note that the changes in the normalized volume on the three surfaces have different slopes due to the shape change effect. For example, for the hydrophobic substrate area of the droplet's contact with gas above the area of the droplet's contact with a solid wall.



Figure 8. The dynamics of the change in the normalized volume of a droplet on three surfaces: 1—surface No. 1, 2—surface No. 2, and 3—surface No. 3. The calculation parameters are similar to those used in the experiments (experimental results are shown in Figure 4).

In the future, the developed model can be explicated by taking into account the slope of the surface, the presence of certain depressions and protrusions on it, as well as the synergistic effects when a set of droplets with a variable distance between them is placed on it. Under these conditions, the decisive role is played by the collective and synergistic effects, as characterized by the multidirectional influence of the factors. The main effects for a single droplet were taken into account in the development of the model presented in this paper. The group effects of the mutual influence of neighboring droplets on each other play an important role when the distances between them are less than 5–7 initial diameters [70–73]. Moreover, this mutual influence is enhanced with an increase in the heat fluxes to the surface of droplets, their mutual arrangement with vapor leakage from the surface of one droplet to neighboring ones, transformation of the surfaces of droplets and change in their shape (oblate or elongated ellipsoids), their swirling, as well as increased thermal diffusivity, reduced viscosity and surface tension of liquids. These effects for a set of droplets located in a heated gas–air flow have been studied experimentally and numerically. The main results are given in [74–77].

4. Experimental Data vs. Modeling and Discussion

In this section, the results of the performed mathematical modeling are compared with the experimental data obtained on surfaces with significantly different contact angles.

Figure 9 presents the data on the dynamics of the change in the normalized droplet volume over time predicted by the present simulations and the results of the experimental measurements for $T_{sub} = T_g = 293$ K and humidity of 35% for three cases (surface No. 1, surface No. 2, surface No. 3). It is substantiated that a satisfactory agreement is achieved between the theoretical and experimental results at the initial heating stage (at t < 500 s). The difference between the predicted values based on the performed experiments and mathematical modeling increases with an increase in the droplet lifetime. This is mainly due to the fact that, with time, the droplet size becomes commensurate with the scales of the textured surface due to liquid evaporation. At the final stage of the calculations, taking into account the identified features, it is advisable to use models based on molecular modeling methods. In addition, an important contribution to the difference between the experimental and theoretical data is made by the presence in the experiments of external disturbing influences (additional convective flows in the gas-air medium), which lead to the entrainment of vapors from the droplet's surface and an increase in the convective heat transfer coefficient and the mass transfer coefficient. As a result, the droplet evaporation rate increases. The same patterns also explain the increase in the degree of discrepancy between the results of the mathematical modeling and experiments with an increase in the contact angle and, accordingly, the droplet free surface area. When these additional effects are taken into account in the course of the calculations, the discrepancy between the results of the experiments and simulations is reduced by 20-30% (dotted curves in Figure 9). It is important to note that the developed model deterministically responds to the identified factors and effects. This is very important, since it provides the possibility of distinguishing between the main and secondary effects and factors and balancing between the complication of the model and the computer performance during calculations. Note that a comparison with published data on droplet evaporation on solid walls was performed in our earlier paper [63].

The results of comparing the values of the evaporation rates in experiments and simulations demonstrate the feasibility of the correct account of the convective heat transfer coefficients, mass transfer, and the corresponding heat flux supplied to the gas droplet. It is also important to take into account the structure of a solid surface when solving problems of droplet evaporation at the micro- and nanoscales.



Figure 9. Cont.



Figure 9. The dynamics of the change in the normalized droplet volume with time predicted by the present simulations (solid curves are base model ($U_a = 0 \text{ m/s}$) (BM), dashed curves are corrected model ($U_a = 0.3 \text{ m/s}$) (CM)) and the results of experimental measurements (EXP) for $T_{sub} = T_g = T_0$ = 293 K and humidity of 35% for three cases (surface No. 1 (**a**), surface No. 2 (**b**), surface No. 3 (**c**)).

The performed numerical simulation showed that the slope of the curves and the behavior of the change in the single-strength droplet volume over time can be quite significantly changed by correcting the values of several input parameters. The initial volume and the droplet shape, the temperature and properties of the substrate surface, and the properties of the liquid have the greatest influence. The intensive spreading of a droplet over the surface can be caused both by the properties of the surface and the liquid, and by the initial droplet volume. And vice versa, the preservation of the droplet's shape in the form of a hemisphere depends on the same factors. It is possible to ensure the output of the curves in Figure 9 at certain asymptotic values (when the liquid spreads over the surface and prolonged evaporation) and the continuous almost monotonic nature of their variation (with a small droplet volume, hydrophobic surface, and high heating rate). These indicated effects substantiated the need to obtain a database of approximation expressions that make it possible to predict the separate and collective contribution of input parameters to the integral characteristics of heat and mass transfer during heating and

evaporation of droplets on specially prepared surfaces. Such a database can be formed using the developed mathematical model.

5. Practical Importance and Applications

In this section, the integral temporal characteristics of evaporation in the form of the droplet lifetime τ_h on the three used surfaces (Figure 1) were predicted using mathematical modeling in a wide range of liquid volumes (from 0.001 µL to 10 µL). These volumes correspond to the characteristic droplet sizes in a number of technological areas based on the interaction of the droplet flows and liquid sprays with the surfaces of units, aggregates, assemblies, and systems, which significantly affect the operational characteristics of their work. The corresponding simulation results and the derived approximation expressions are presented in Figure 10. As expected, the lifetime of droplets increases with an increase in their volume. The larger the contact angle (as in the case of surface No. 3), the higher the values of the total lifetimes of droplets for identical initial droplet volumes. This is due to a change in the fraction of the heat fluxes supplied to the droplet from the gas and the solid surface, which was also confirmed by estimates using one-dimensional models [63].



Figure 10. The results of modeling the integral time characteristics of evaporation in the form of lifetimes $\tau_{\rm h}$ depending on the initial droplet volume on the three studied surfaces (solid curve is surface No. 1, dashed curve is surface No. 2, dotted curve is surface No. 3) for $T_{\rm sub} = T_{\rm g} = T_0 = 293$ K and humidity of 35%.

The established behavior of the trends in Figure 10 substantiates the exit of the values of the droplet lifetime to certain asymptotic values. This effect is due to an increase in the role of the liquid spreading over the surface with an increase in the volume. When a certain critical volume is exceeded, the liquid spreading becomes decisive and does not allow for increasing the layer thickness on the substrate surface. The longitudinal (with respect to the substrate surface) dimensions of the droplet increase. This leads to an increase in the contact area between the liquid and the substrate and, accordingly, to an increase in the evaporation rate. The droplet lifetime practically does not change under such conditions. This behavior persists until the volume of the parent droplet reaches a value at which it will be in contact with neighboring ones on the substrate, and the effects of their coagulation with the formation of a single liquid film will not appear. The corresponding effects become even more interesting when the gas–air flow impinges on the

droplets in the longitudinal and transverse directions with respect to the substrate surface. This is typical for technological processes with flow around the structural elements of motive systems, taking into account the effects of their irrigation with liquids (for example, during the movement of cars, trains, aircraft and other equipment, taking into account the irrigation of their surfaces with droplets of fog and rain, particles of ice, snow, pollutants and materials). The role of heat and mass transfer processes and phase transformations is enhanced. These conditions are important in practical applications and are of considerable interest for the further development of the model presented in this paper.

It is advisable, considering the need to ensure the possibility of performing calculations on a PC with different performance characteristics, to adhere to the concept of simplifying models and using known reliable experimental data obtained using advanced software and hardware stands. High requirements for the speed and accuracy of recording the key characteristics of heat and mass transfer and hydro- and gas dynamics in systems with micro- and nanosizes significantly limit the list of methods that can solve the tasks. For example, to register the convection rates, temperature and heat fluxes in liquid and gaseous media, as well as the size and shape of droplets, it is advisable to use micro particle image velocimetry, particle tracking velocimetry, two-color laser-induced phosphorescence, planar laser-induced fluorophore, shadow photography and high-speed systems of video recording with the tracking of dynamic objects and automatic processing of video signals. Examples of using these systems for droplet applications are given in [78–80]. The presence of video and photo frames with illustrations of the main processes from experiments allows for formulating more complete physical and mathematical models, as well as prioritizing the list of the most and least strongly influential factors on the heat and mass transfer and phase transformations. These data are the rationale for the simplifications and assumptions in the development of predictive models that are created for engineering calculations and in the fundamental study of effects, processes and phenomena.

6. Conclusions

(i) Unique experimental data are presented on the characteristics of the water droplet evaporation on heated structured surfaces obtained using nanosecond laser texturing followed by low-temperature annealing. The dependences of the evaporation rates under natural conditions on each of the substrates were determined. The evaporation regimes were identified for water droplets on hydrophilic and hydrophobic surfaces. It was found that the surfaces used can be divided into three types: smooth hydrophilic—surface No. 1, rough hydrophobic—surface No. 2 (during evaporation the contact line is pinned), and rough hydrophobic—surface No. 3 (contact diameter decreases). Thus, this work examines the agreement of the model with the experimental results when implementing various interaction regimes. An experimental base has been prepared for the development of fundamentally new and existing models of heat and mass transfer, which describe the conditions for the interaction of liquid droplets with hydrophobic and hydrophilic surfaces.

(ii) A mathematical model was developed for the heating and evaporation of a liquid droplet on a solid surface with contact angles varying over a wide range (from a hydrophilic to a superhydrophobic state). The model considers convective and interfacial transfer to describe the corresponding recirculation of liquid layers and transformation of the temperature field, and it also provides satisfactory prediction characteristics of the heating and evaporation of droplets at a wide range of contact angles (from 70° to 160°), in contrast to the previously developed one-dimensional and two-dimensional models without considering these effects [63].

(iii) Using the developed mathematical model, we predicted the characteristics of the heating and evaporation of water droplets with different volumes (from 0.001 μ L to 10 μ L) on the obtained structured surfaces. The corresponding approximation expressions were obtained. The satisfactory agreement between the numerical simulation results and known experimental data was substantiated. The maximum deviations did not exceed

5%. The limits of applicability and perspective directions of development of the created mathematical model were described.

(iv) Among the unsolved problems, the most interesting for further study are controlling the evaporation of droplets from surfaces not only through texturing the surfaces but also through the use of additives and impurities to liquids, taking into account the macro- and microtexture of surfaces when describing the dynamics of droplet evaporation, formulating new approaches to the evaporation of liquid droplets, and the creation of combined methods for controlling heat transfer between a droplet and a solid wall.

Author Contributions: Conceptualization, D.V.A.; writing-original draft, review and editing, D.V.A., A.G.I. and E.G.O. All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported by the Russian Science Foundation (project no. 23-73-30004).

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

С	Heat capacity, J/(kg·K)
2	Molar volume concentration of water vapor, mole/m ³
W	Water vapor diffusion coefficient, m ² /s
F _{st}	Surface tension force, N
<i>र</i>	Gravitational constant, m/s ²
h	Heat transfer coefficient, $W/(m^2 \cdot K)$
$J_{d(e)}$	Mass evaporation rate, $kg/(m^2 \cdot s)$
k	Thermal conductivity, W/(m·K)
L _d	Vaporization heat of water, J/kg
М	Molar mass of water, kg/mol
n	Normal vector to the droplet surface
Psat	Saturation vapor pressure, Pa
b	Pressure, Pa
$R_{d(e)}$	Evaporation rate, m/s
r, z	Labels of axes, m
S	Root squared deviation
Г	Temperature, K
u, w	Velocity projections along the r and z axes, m/s
Y	Mass concentration
Y_{sat}	Mass concentration of water vapors under saturation conditions
Greek	
к	Thermal diffusivity, m ² /s
μ	Dynamic viscosity, Pa·s
р	Fluid density in the cell, kg/m^3
σ	Surface tension of water, N/m
Φ	Dissipative function, $1/s^2$
Index	
C	Initial conditions
1	Water
2	Air
d	Droplet
eff	Effective
g	Gas
i	Number of material

- 9
- 1 Liquid
- ref Reference value
- v
 - Vapor

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