Supplementary Materials: Supercooled Water Droplet Impacting Superhydrophobic Surfaces in the Presence of Cold Air Flow

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1. Surface Characterization

In current research three surfaces with different wettabilities were used for evaluation of supercooled droplet impacts behavior. Based on laser confocal microscopy, some important parameters have been extracted namely surface root mean square and average roughness. Furthermore, ratio of wetted area to based area which is well-known as a ϕ ratio is extracted form aforementioned technique. Aluminum and Teflon surfaces were purchased from Mcmaster carr, Boston, MA, USA. (Aluminum 6061, mirror like-finish and Teflon made with virgin Teflon PTFE resins). Detailed information of surface characteristic is presented in Table 1.1.

Table 1.1. Static contact angle (θ_S), advancing contact angle (θ_A), receding contact angle (θ_R), and contact angle hysteresis (θ_H), surface average roughness (R_a), and surface root mean square (R_q) and area ratio of surfaces (ϕ).



This information is become necessary for prediction of solid-liquid interface temperature.

2. Experimental Set up of Stagnation Cold Flow Generation

Stagnation flow is produced by a device which is called here droplet accelerator. As it was depicted from main context of paper, functionality of droplet accelerator is based on conversion of pressurized air (internal energy) to a coherent airstream of defined velocity (kinetic energy) by perforating air through tiny holes and finally passing through the convergent part of the nozzle. Based on the nozzle outlet diameter, different air speeds are produced. For nozzle outlet diameter up to 16.5 mm, maximum air stream velocity was about 30 m/s obtained from maximum inlet air pressure up to 7 atm.

As illustrated from Figure 2.1, generation of super cooled water and cold air flow has a relatively complex design and set up. Large volume flow rate of dry room temperature is cold down by an immediate cooling through a designed heat exchanger. Single-pass counter flow heat exchanger which is worked with liquid nitrogen (Les Gaz Spéciaux Megs, Montreal, QC, Canada) was designed to generate very cold air flow at exit pipe of heat exchanger. It is worth mentioning that latent heat of vaporization of liquid nitrogen is 200 kJ/kg, having amazing melting point up to -196 °C, make it possible to generate -80 °C air temperature at 30 cfm air flow rate. Therefore, simple calculation shows interesting cooling power up to 1.4 kW. Heat exchanger design was based on usage of small amount of liquid nitrogen which is transferred by a cryogenic transfer flex hose

with internal diameter of 0.5 inch and 36 inch long (Les Gaz Spéciaux Megs, Montreal, QC, Canada) from liquid nitrogen vessel.

Therefore an optimized length of helical copper pipe (50 feet with external diameter of 0.5 inch) was chosen to accomplish this aim. Heat exchanger is a Styrofoam box (Les Gaz Spéciaux Megs, Montreal, QC, Canada) with external dimensions of 19.5 inch length, 16.5 inch width, 15 inch height. Internal dimensions is 17.5 inch length, 14.5 inch width and 13 inch height. Small thermal conductivity of Styrofoam chamber (i.e., 0.033 w/m·K) makes it an ideal insulated heat exchanger when it works with liquid nitrogen. At the top part of heat exchanger, one pipe with external diameter of 10 cm is connected and completely sealed with Aluminum foil tape (General Purpose, 2" Wide and 004" Thick, Mcmaster-Carr, Boston, MA, USA). Finally, vaporized nitrogen exhaust pipe is connected to laboratory fume for safety precautions.

As presented in Figure 2.1, to conducting experiments, three different branches of cold air flow are required. First cold air line is directly connected with air intake of droplet accelerator's mixing chamber. Cold air flow rate is controlled by a brass ball valves (Mcmaster-Carr, Boston, MA, USA).



Figure 2.1. Schematic of experimental set up for simulation cold stagnation flow.

Second cold airline is used for icing chamber cooling. Icing chamber is used to provide cold air environment. All components of droplet impacts experiments are inserted inside of icing chamber. For reaching to this aim, Icing chamber has a dimension of 90 cm height, 55 cm length and 45 cm width and it is made of 5 sheets of optically clear cast acrylic Plexiglas (Mcmaster-Carr, Boston, MA, USA) with 0.25 inch thickness installed on 8 aluminum T-Slotted framing extrusion. To this end, third cold air line is used for cooling of another chamber which is termed here primary cooling chamber. This chamber provide controllable cold air temperature as low as –30 °C to cool down target substrates in a short period of time.

Surface temperature measurements of different objects were performed by thermal imaging photography. The IR thermometry has been used in this study to measure surface temperature of solid surfaces and supercooled water droplet as well [1]. In this study A320 IR camera and ThermoVision ExaminIR[®] software has been used in order to monitor surface temperature. For measuring surface and air temperature wider field of view is needed. In this case, 200 mm/75° lens with spatial resolution of 0.24 mm for 1 pixel is used to monitor surface temperature of solid substrate. IR camera can provide surface temperature distribution of water drop whether in room and supercooling condition. In IR thermography water drop is considered as an opaque object [1].

Calibration of IR camera was carried out by choosing emissivity factor of 0.96 to 0.97. Due to the fact that measurement of surface temperature of water drop needs higher spatial resolution, therefore 10 mm/90° lens with spatial resolution of 0.1 mm for 1 pixel has been used. Recording rate in all cases were 9 frames per seconds. It is worth mentioning that in well-insulated condition surface temperature at stagnation domain is similar to gas temperature, as depicted in Figure 2.2.



Figure 2.2. Surface temperature measurement using IR thermography low temperature condition.

3. Numerical Simulation of Impinging Air Flow

In order to characterize air profile through nozzle of droplet accelerator numerical simulation of one-phase gas flow has been carried out. Modeling of the geometry was accomplished with ICEM CFD with almost 400,000 tetrahedral meshes, as depicted in Figure 3.1, mesh resolution in the vicinity of nozzle throat was 500 um in order to capture air profile precisely. Numerical simulation was carried out by non-commercially use of ANSYS-FLUENT 14. Three boundary condition namely velocity inlet, wall and pressure outlet have been used for simulation purposes. Different turbulent models were evaluated through available option in software. Here, results of standard $k - \varepsilon$ model are presented. Simulation results show a good agreement with that of experimental measurements carried out with Pitot tube around core of nozzle which is a matter of interest in current study. As can be seen from Figure 3.1, uniform air velocity up to 10 m/s is observed which is used for quantitative evaluation of present study.



Figure 3.1. Mesh generation using ICEM CFD 14. Almost 400,000 tetrahedral meshes have been used for proper capturing of air jet velocity profile. Figure (**a**–**c**) represent droplet accelerator's nozzle mesh structure, contours of velocity magnitude and velocity vectors, respectively.

4. Analysis of Falling Droplet Exposed to Airflow with Different Velocity and Temperature

In order to properly evaluate wetting dynamics of impacting supercooled water drop in the presence of cold stagnation flow, thermal analysis of falling drop is carried out in extremist cold condition (i.e., -30 °C with air velocity up to 10 m/s) to understand how much it would effect on droplet temperature. Two important dimensionsless number namely Biot and Fourier has been used for droplet temperature distribution evaluation. Analysis of moving drop through air is divided in to two domains, as illustrated in Figure 4.1a.



Figure 4.1. Schematic of producing stagnation cold flow (a); Imposed air velocity on moving drop through cold air (b).

Calculation of Biot number based on terminal velocity shows uniform temperature distribution as it was less than critical Biot number in domain 1. (i.e., 0.08 < 0.1). Indeed, this value is even smaller than 0.08 as droplet velocity in domain 1 is varied from 0 to almost terminal velocity.

$$Bi = \frac{h_g l_c}{k_d} = \frac{h_g D_d}{6k_d} \tag{4.1}$$

For domain 1 and 2, Biot number is calculated based on forced convective heat transfer coefficient as illustrated in following formula:

$$Re = \frac{\rho_g D_d (U_g - U_d)}{\mu_g} \tag{4.2}$$

$$Pr = \frac{\mu_g c_{p,g}}{k_g} \tag{4.3}$$

Spalding number =
$$B = \frac{C_{p,v}(T_g - T_d)}{H_v}$$
 (4.4)

$$Nu = \left(2 + 0.6Re^{\frac{1}{2}}Pr^{\frac{1}{3}}\right)(1+B)^{-0.7}$$
(4.5)

$$h_g = \frac{Nu.k_g}{D_d} \tag{4.6}$$

where k_g is thermal conductivity of cold air at -30 °C. Nusselt number Nu is presented by modified Ranz-Marshall correlation for liquid drop [2]. Property of air at wide range of temperature from 20 °C to -30 °C is illustrated in Table 4.1 [3,4].

Temperature	re Density Dynamic Viscosity		Thermal Conductivity	Specific Heat	Thermal Diffusivity	
(k)	(kg/m ³)	(10 ⁻⁶ Pa·s)	(10 ⁻³ W/m·K)	(J/kg·K)	(10 ⁻⁶ m ² /s)	
240	1.47	15.54	21.45	1005.5	14.49	
250	1.41	16.06	22.26	1005.4	15.67	
260	1.35	16.57	23.05	1005.5	16.87	
290	1.21	18.05	25.38	1006.0	20.72	

Table 4.1. Dry air properties at different temperatures.

Analysis of Biot number in domain 2 shows that it passes critical Biot number (i.e., 0.1) and becomes as large as 0.17 due to larger convective heat transfer coefficient. On the other hand, calculation of another important dimensionless number, Fourier number ($\tau = \frac{4\alpha t}{D_a^2}$), shows very small value up to 0.00045. it demonstrates that only interface of liquid water drop is affected by

incoming cold air flow. Therefore, when wetting dynamic analysis is carried out, heat penetration length is calculated. As presented in main context of paper, this value becomes as small as 0.1 mm highlighting that there is a viscosity jump up to 8 folds at supercooled water drop interface.

5. Drag Force Analysis

Associated drag force of moving liquid drop through air flow is approximated by following formula which assumes it maintained spherical shape through entire domain. Calculation of drag force [5], as illustrated in Equation (5.1), is important as droplet impact velocity with and without air flow must be equal in order to have a same droplet Weber and Reynolds number. Aforementioned dimensionless numbers are used for predicting maximum spreading diameter of impacting water drop.

$$C_D = \frac{24}{Re} (1 + 0.150Re^{0.681}) + \frac{0.407}{1 + \frac{8710}{Re}} Re < 2 \times 10^5$$
(5.1)

Analysis of droplet moving through air shows that if relative Weber number (i.e., $\frac{\rho_{air}(U_g - U_d) D_d}{\sigma_{lv}}$) become less than 3. Droplet shape is almost spherical. For spherical object drag force coefficient become 0.41 for Weber and Reynolds number up to 88 and 1890, respectively. However, it was confirmed that drag coefficient is increased about 15% to that of pure spherical solid object for liquid water drop [6]. Therefore, associated drag force due to imposed air flow is illustrated by Equation (5.2):

$$F_{Drag} = \frac{1}{2} \rho_{air} (U_{air} - U_d)^2 \ 1.15 C_D \frac{\pi D_d^2}{4}$$
(5.2)

Calculated amount of impose drag force for lowest temperature (i.e., -30 °C) becomes 1.29×10^{-4} N. Therefore, increased velocity is 0.07 m/s for almost 5 ms exposure time in domain 2, as depicted in Figure 4.1.

Carefully measurement of droplet impact velocity accompanied with air flow with high speed imaging experiment shows that impinging droplet velocity is almost identical or even slightly smaller with that of still air. This phenomenon can be referred to effect of very small air suction through the small pipe connected to droplet generator system. It can be more argued that based on results of numerical simulation reverse velocity is also observed and it has a small magnitude. By choosing very small suction velocity along with appropriate exposure time (i.e., 88 ms based on terminal velocity of free falling drop from 0.15 distance to substrate) almost impinging velocity become similar with that of still air having a good agreement with experimental measurements results. To this end, all aforementioned calculations were for highest air velocity up to 10 m/s. Indeed, when air velocity becomes lower, deviation of impinging droplet velocity with that of still air is insignificant (i.e., completely locates in precision error of experiments).

6. Heat Transfer Analysis of Target Substrates Contacting with Cold Air Flow

Calculation of Biot number of substrates contacting with cold air flow is carried out by usage of forced convective heat transfer coefficient of impinging air flow calculated based on modified correlation of Nusselt number [7]. In this correlation, Reynolds number is calculated based on impinging air jet velocity and diameter of impinging area. Therefore, Maximum spreading diameter for each surface has been used as a characteristic length for calculation of Reynolds number. Figure 6.1, shows schematic of contacting cold air flow on a substrate.

$$Re = \frac{\rho_g \, D_{\max} \, U_{g,jet}}{\mu_g} \tag{6.1}$$

$$Pr = \frac{\mu_g \ c_{p,g}}{k_g} \tag{6.2}$$

$$Nu = 0.966 \ Re^{0.46} \ Pr^{\frac{1}{3}} \tag{6.3}$$

$$h_g = \frac{Nu.k_g}{D_{\text{max}}} \tag{6.4}$$

$$Bi = \frac{h_g l_c}{k_{substrate}}$$
(6.5)

Calculation of convective heat transfer coefficient was based on maximum air jet velocity to evaluate the worst case scenario, which largest Biot number even more than critical value (i.e., 0.1), might be happened. Therefore, for aluminum substrate with thermal conductivity of 148 (w/m·K), maximum spreading diameter about 8.5 mm and thickness of 2.1 mm (thickness of aluminum plate) it becomes as low as 0.002.

For Teflon substrate due to low thermal conductivity it becomes slightly larger than critical value up to 0.47. Therefore, we need to calculate Fourier number for flat surface (i.e., $\tau = \frac{\alpha t}{L_c^2}$). For maximum period of 100 ms, which is sufficient time of spreading and retraction phase of drop impacts dynamics in current study, and thickness up to 1 mm. it becomes as small as 0.016. Therefore, refereeing to Heisler charts, core temperature of substrate become almost equal to surface temperature. (i.e., 0.98 Substrate temperature). Same procedure for superhydrophobic surface is carried out which shows Biot number (i.e., 0.075, thickness of coating is 0.2 mm) smaller than critical Biot number value (i.e., 0.1).



Figure 6.1. Schematic of impinging air flow on different target substrates.

7. Stagnation Flow Analysis Based on Classical Homann Flow Approach

The air jet leaving the outlet of the droplet accelerator generates a stagnation flow which rapidly transforms to an axisymmetric radially outward flow, Reference [8] as illustrated in Figure 7.1.



Figure 7.1. Schematic of stagnation air flow on different target substrates, classical Homann flow [8].

Using principle equations, it is possible to find associated shear force imposed to liquid–water interface. Using the similarity analysis and variable $\eta = z \sqrt{\frac{2a}{v}}$, velocity components along with

associated shear and normal force (i.e., pressure force), satisfying the continuity equation in radial direction, can be expressed as follows:

$$V_r = arf'(\eta) \tag{7.1}$$

$$V_z = -\sqrt{2av} f(\eta) \tag{7.2}$$

$$\tau_{rz(z=0)} = \mu(\frac{\partial V_r}{\partial z} + \frac{\partial V_z}{\partial r})$$
(7.3)

$$P_s = \frac{1}{2} \rho_{air} V_z^2 \pi r^2 \tag{7.4}$$

As depicted from Equation (7.1), radial velocity is increased from stagnant point to maximum spreading diameter as a function of *r*. therefore; associated shear force is increased by increasing maximum spreading diameter. Consequently, it is possible to say that imposed shear force on a droplet impacting on a hydrophilic surface is higher than superhydrophobic surface due to larger maximum spreading diameter.

8. Finding Solid-Liquid Interface Temperature

In this section, model of transient conduction heat transfer from droplet to cold substrate is presented. We have used same methodology to that of Bahadur et al. [9] for finding transient interface temperature and temperature distribution when in contact with different solid material from hydrophilic to superhydrophobic surfaces. While previous model was derived for textures superhydrophobic surfaces, using confocal image microscopy technique which provide surface morphology information (e.g., average roughness and ratio of wet to base area, ϕ ratio,), same procedure which was used for textured superhydrophobic surface is applied for coated or uncoated surfaces. Furthermore, one modification on transient interface temperature formula presented by of Bahadur et al. [9] was carried out. In fact, predicted temperature contradicted with physical state, as it becomes colder than substrate at initial condition. Therefore, another derivation of transient interface temperature. Transient conduction equation was solved for each domain with same boundary conditions at interface location. First domain involves substrate-post-air domain and second one is droplet, as depicted in Figure 8.1, which is similar to that of Bahadur et al. [9].

It is worth mentioning that convection boundary condition at top of spread drop was not considered for solving transient heat condition equations. In fact, penetrated heat length [10], $\delta_h \approx (\alpha_w t_c)^{\frac{1}{2}}$, during spreading and retraction time for SHS, which is about 14 ms, is 42 µm. Furthermore, for aluminum and Teflon substrate for spreading and retraction time up to 100 ms, which is a sufficient time for evaluation of stagnation flow on impacting drop, heat penetration length becomes 112 µm which is almost 5 folds smaller than thickness of spreading water droplet at the maximum spreading diameter. Therefore, effect of convective heat transfer due to cold air flow on solid-liquid interface temperature during small period of time (i.e., 100 ms) can be neglected. In following, clear derivation of transient conduction for both sub domains are presented which results in deriving analytical model of solid-liquid interface temperature prediction.

Transient heat conduction equation for domains illustrated in Figure 8.1 is presented like following:

$$\frac{\partial T_{pa}(x,t_c)}{\partial t_c} = \alpha_{pa} \frac{\partial^2 T_{pa}(x,t_c)}{\partial x^2}, \text{ for region between } 0 < x < h.$$
(8.1)

$$\frac{\partial T_w(x,t_c)}{\partial t_c} = \alpha_w \frac{\partial^2 T_w(x,t_c)}{\partial x^2}, \text{ for region between } x > h.$$
(8.2)

In equation above, T_{pa} , is temperature of post-air domain and T_w is water temperature, h is average rougness of surface and t_c is total contact time of liquid drop on surface. α_{pa} and α_w are thermal diffusivity of post air domain and water, respectively, as presented by Bahadur et al. [9].

The boundary and initial condition for each domain is presented in following. Therefore, we need two boundaries and one initial condition for each domain.



Figure 8.1. Schematic of two domains involved in model of transient heat conduction for finding interface temperature. Effect of air thermal resistance is modeled in the average thermal diffusivity of post-air domain.

Boundary and initial condition for post-air domain are like following:

First boundary condition:
$$T_{pa}(0,t) = T_{substrate}$$
, $x = 0$ (8.3)

Second boundary condition:
$$T_{pa}(h,t) = T_w(h,t)$$
, $x = h$ (8.4)

First initial condition:
$$T_{pa}(x < h, 0) = T_{substrate}$$
, $t = 0$ (8.5)

Boundary and initial condition for water domain are like following:

First boundary condition:
$$T_w(h,t) = T_{pa}(h,t)$$
, $x = h$ (8.6)

Second boundary condition:
$$T_w(\infty, t) = T_{droplet}, x = infinity$$
 (8.7)

First initial condition:
$$T_w(x > h, 0) = T_{droplet}$$
, $t = 0$ (8.8)

It is clear that subsequent solution of two transient conduction equation become similar at interface, as it illustrated from boundary conditions (8.4) and (8.6) which are the same.

By defining:

$$\theta_{pa}(x,t) = T_{pa}(x,t) - T_{drop} \tag{8.9}$$

$$\theta_w(x,t) = T_w(x,t) - T_{drop} \tag{8.10}$$

Due to the fact that there are infinity boundary conditions, the best way for solving partial differential equation is defining similarity variable which make PDE to ODE like below:

$$\eta_{pa} = \frac{x}{2\sqrt{\alpha_{pa}t_c}}, \eta_w = \frac{x}{2\sqrt{\alpha_w t_c}}$$
(8.11)

Inserting Equations (8.9)–(8.11) to Equations (8.1) and (8.2) and by definition of chain derivatives following second order ODE are obtained.

$$\frac{d^2\theta_{pa}}{d\eta_{pa}{}^2} + 2\eta_{ns}\frac{d\theta_{pa}}{d\eta_{pa}} = 0$$
(8.12)

$$\frac{d^2\theta_w}{d\eta_w^2} + 2\eta_w \frac{d\theta_w}{d\eta_w} = 0$$
(8.13)

Equations (8.12) and (8.13) can be solved by defining variable *S* like below:

$$\frac{d\theta_{pa}}{d\eta_{pa}} = S_{pa'} \frac{d\theta_w}{d\eta_w} = S_w \tag{8.14}$$

These substitutions make second order equation in to a first order equation which can be solved analytically by method of separation of variables as illustrated in Equations (8.15) and (8.16).

S8 of S16

$$\frac{dS_{pa}}{d\eta_{pa}} + 2\eta_{pa}S_{pa} = 0 \tag{8.15}$$

$$\frac{dS_w}{d\eta_w} + 2\eta_w S_w = 0 \tag{8.16}$$

$$S_{pa} = \frac{d\theta_{pa}}{d\eta_{pa}} = C_{pa} \exp(-\eta_{pa}^2)$$
(8.17)

$$S_w = \frac{d\theta_w}{d\eta_w} = C_w \exp(-\eta_w^2)$$
(8.18)

Therefore, solution of Equations (8.17) is defined based on error function illustrated in Equations (8.19)–(8.26).

$$\theta_{ps}(\eta) - \theta_{ps}(0) = \int_0^{\eta} C_{ps} \exp\left(-\eta_{ps}^2\right) d\eta$$
(8.19)

$$\theta_{ps}(\eta) - \theta_{ps}(0) = a_{ps} erf(\eta_{ps})$$
(8.20)

which a_{pa} , is new integral constant defined by applying associated boundary conditions.

By substitution of Equations (8.9)–(8.20):

$$T_{ps}(\eta) - T_{ps}(0) = a_{ps} erf(\eta_{ns})$$
(8.21)

However, as it depicted from Equations (8.4) and (8.6), which are the same, at x = h and t = 0, $T_{ps}(\eta) = T_{drop}$ and $T_{ps}(0) = T_{substrate}$, therefore, following equation is derived:

$$T_d - T_{sub} = a_{ps} erf(\infty) \xrightarrow{yields} a_{ps} = T_d - T_{sub}$$
 (8.22)

Transient temperature profile in post-air domain is presented like Equation (8.23) by recalling Equation (8.21) and knowing $T_{ps}(0) = T_{substrate}$.

$$T_{ps}(\eta) - T_{ps}(0) = T_d - T_d + a_{ps} erf(\eta_{ps})$$
(8.23)

Therefore by substitution Equation (8.22) in (8.23), Transient conduction equation for post-air domain is derived, as illustrated in Equation (8.26) which, $erfc(\eta)$, is a complementary error function.

$$T_{pa}(\eta) = T_d + T_{sub} - T_d + (T_d - T_{sub})erf(\eta_{pa})$$
(8.24)

$$T_{pa}(\eta) = T_d + (T_{sub} - T_d)(1 - erf(\eta_{pa}))$$
(8.25)

$$T_{pa}(\eta) = T_d + (T_{sub} - T_d) erfc(\eta_{pa})$$
(8.26)

Same as aforementioned procedure for solving transient conduction equation of post-air domain, solution of Equation (8.18) is also defined based on error function. Transient conduction equation in water domain can be presented by integrant splitting in Equation (8.27).

$$\theta_w(\eta) - \theta_w(h) = \int_h^{\eta} C_w \exp(-\eta_w^2) \, d\eta \tag{8.27}$$

$$\theta_{w}(\eta) - \theta_{w}(h) = \int_{h}^{0} C_{w} \exp(-\eta_{w}^{2}) \, d\eta + \int_{0}^{\eta} C_{w} \exp(-\eta_{w}^{2}) \, d\eta$$
(8.28)

$$\theta_w(\eta) - \theta_w(h) = -a_w \operatorname{erf}(h_w) + a_w \operatorname{erf}(\eta_w)$$
(8.29)

which a_{w} , is new integral constant defined by applying associated boundary conditions.

From Equation (8.7), $x \to \infty, \eta \to \infty, T_w(\infty) = T_{drop}$, and from Equation (8.10),

$$\theta_w(\infty) = T_{drop} - T_{drop} = 0 \tag{8.30}$$

It is worth stressing that after defining similarity variable in Equation (8.11), variables in Equation (8.10) (i.e., *x* and *t*) are substituted with one variable defined η .

Furthermore, from Equation (8.10), water temperature at $\eta = h$, is presented like following:

$$\theta_w(h) = (T_w(h) - T_{drop}) \tag{8.31}$$

Inserting Equations (8.30) and (8.31) into (8.29), following useful equation is derived.

$$-(T_w(h) - T_{drop}) = -a_w \operatorname{erf}(h) + a_w = a_w [1 - \operatorname{erf}(h)] = a_w \operatorname{erf}(h_w)$$
(8.32)

Form boundary conditions (8.4) or (8.6) temperatures at interface of two boundaries are similar; $T_w(h) = T_{pa}(h)$. Therefore, by recalling Equation (8.26), following equation is emerged.

$$T_w(\mathbf{h}) = T_d + (T_{sub} - T_d) erfc(h_{pa})$$
(8.33)

$$T_w(\mathbf{h}) - T_d = (T_{sub} - T_d) erfc(h_{pa})$$
(8.34)

Recalling Equation (8.32) and using Equation (8.34) following formula are derived.

$$-[(T_{sub} - T_d)erfc(h_{pa})] = C_w erfc(h_w)$$
(8.35)

$$a_w = -[(T_{sub} - T_d)]\frac{erfc(h_{pa})}{erfc(h_w)}$$
(8.36)

Recalling Equation (8.29) and using Equation (8.10) we can write like following:

$$T_w(\eta) - T_w(h) = -a_w \operatorname{erf}(h_w) + a_w \operatorname{erf}(\eta_w)$$
(8.37)

On the other hand from Equation (8.32), (i.e., $T_w(h) = T_{drop} - a_w erfc(h_w)$), and substitution into Equation (8.37), following equations are emerged.

$$T_{w}(\eta) = T_{drop} - a_{w}[erfc(h_{w}) + erf(h_{w}) - erf(\eta_{w})] = T_{drop} - a_{w}[1 - erf(\eta_{w})] = T_{drop} - a_{w}[erfc(\eta_{w})]$$
(8.38)

$$T_w(\eta) = T_{drop} + \left[(T_{sub} - T_d) \right] \frac{erfc(h_{pa})}{erfc(h_w)} \cdot erfc(\eta_w)$$
(8.39)

Therefore interface temperature can be obtained by choosing $\eta = h$ in Equation (8.39), as it illustrated in Equation (8.40). Therefore, transient temperature distribution within water drop is also presented in Equation (8.41).

$$T_{w-interface}(h, t_c) = T_{drop} + \left[(T_{sub} - T_d) \right] erfc \left(\frac{h_{pa}}{2\sqrt{\alpha_{pa}t_c}} \right)$$
(8.40)

$$T_w(x_w, t_c) = T_{drop} + [(T_{sub} - T_d)] \frac{erfc\left(\frac{hp_a}{2\sqrt{\alpha_p a t_c}}\right)}{erfc\left(\frac{h_w}{2\sqrt{\alpha_w t_c}}\right)} \cdot erfc\left(\frac{x_w > h}{2\sqrt{\alpha_w t_c}}\right)$$
(8.41)

It is worth mentioning that complementary error function is a reducing function which become equal to 1 when \rightarrow 0. However, height at interface line, which has a small length scale compare to thickness of spread drop (i.e., average roughness length scale), is not exactly zero which leads to complementary error function become smaller than 1. Neglecting this term cause a significant physical conflict in terms of designated temperature value which becomes even colder than substrate temperature totally contradicted with initial and boundary conditions.

Form Equation (8.40) it is possible find interface temperature of an impacting supercooled water on different solid surfaces. As illustrated from Figure 8.2, aluminum substrate shows a faster convergence to substrate temperature compare with Teflon and Superhydrophobic surface due to very small average roughness (see Table 1.1) and higher thermal diffusivity of post-air domain. Furthermore, from Equation (8.41) temperature distribution inside of water can be obtained, as depicted in Figure 8.3.



Figure 8.2. Transient interface temperature of an impacting supercooled water drop on solid surfaces. Droplet and substrate temperatures are –5.5 °C and –30 °C, respectively.



Figure 8.3. Transient temperature distribution of an impacting supercooled water drop on cold superhydrophobic surface. Droplet and substrate temperature are –5.5 °C and –30 °C, respectively.

9. Results of Super Cooled Droplet Impact on Cold Aluminum Substrate

Figure 9.1 shows sequential images of supercooled water droplet impacts on aluminum substrate maintained at –30 °C. Furthermore, quantitative analysis of temporal spreading diameter which is also illustrated in Figure 9.2. Corresponding droplet diameter, droplet impact velocity, Weber, Reynolds, Stefan, and Prandtl number are 88, 1890, 0.40, 17.51, respectively. It is exposed to various air velocities ranging from 0 to 10 m/s. Classical nucleation theory, as illustrated in main context of paper, emphasis that nucleation rate has an exponential relation with Gibbs free surface energy. By lowering surface temperature, energy barrier regarding phase change is further decreased compare to previous cases causing an immediate freezing at maximum spreading diameter, as it illustrated in Figure 9.1, In terms of wetting dynamics, cooled air flow does not shows a observable change in impacting droplet but it promotes homogeneous nucleation at gas-liquid interface, as discussed in main context of paper. After this phase, droplet start freezing and almost it shapes becomes more blurry up to 25 ms.



Figure 9.1. Sequential images of supercooled water droplet impacting on aluminum substrate maintained at -30 °C.



Figure 9.2. Temporal evolution of impacting supercooled water droplet on cold aluminum substrate at various air velocities. Droplet and substrate temperature are –5.5 °C and –30 °C, respectively.

10. Results of Super Cooled Droplet Impact on Cold Teflon Substrate

Figure 10.1 shows sequential images of an impinging super cooled water droplet with mean value temperature up to -5.5 °C impacting on a cold Teflon substrate with temperature maintained at -30 °C. Corresponding droplet diameter, droplet impact velocity, Weber, Reynolds, Stefan, and Prandtl number are 88, 1890, 0.40, 17.51, it is exposed to various air velocity ranging from 0 to 10 m/s.

In terms of maximum spreading diameter no difference was seen respected to previous cases highlighting that droplet reaches to maximum wetting diameter at 3 ms which is not affected by ice growth layer during such a short time of spreading phase. Striking phenomena was observed when relaxing time (i.e., time at maximum spreading diameter) is significantly increased. Droplet receding angle almost become zero signaling that capillary force become insignificant. Viscous force both internal and within contact line is dramatically increased proposing harsher viscous dissipation at this moment. In terms of thermal transport phenomenon, larger cooling rate both by increasing larger temperature gradient and cooling time increase the probability of phase change at this moment. Quantitative evaluation of temporal spreading diameter, which is illustrated in Figure 10.2, shows that instantaneous pinning or freezing happened at 3 ms and retraction phase completely eliminated. Both heterogeneous and homogeneous nucleation become dominant as by lowering surface temperature, energy barrier regarding phase change decreases. After that liberation of latent heat of fusion [1] controls freezing front of ice-water-interface.



Figure 10.1. Sequential images of supercooled water droplet impacting on Teflon substrate maintained at -30 °C.



Figure 10.2. Temporal evolution of impacting supercooled water droplet on cold Teflon substrate. Droplet and substrate temperature are –5.5 °C and –30 °C, respectively.

11. Results of Super Cooled Droplet Impact on Cold Superhydrophobic Substrate

Figure 11.1 shows sequential images of impacting supercooled water droplet on a superhydrophobic surface along with quantitative evaluation of temporal spreading diameter. Droplet and substrate temperatures are -3.5 °C and -20 °C, respectively. Effect of stagnation pressure (i.e., normal force) completely suppressed droplet jetting phenomenon. As depicted in Figure 11.1a, at 8 ms droplet height was reduced up to 37% at 10 m/s air speed to that of still air case. Additionally, droplet contact time was reduced about 30% which can be related to presence of

thin moving layer of air converting partial slip to full slip condition results in droplet contact time reduction. In fact, surface and air temperature are not sufficiently low to generate heterogeneous and homogeneous nucleation, as the critical temperature for crystallization process was determined about –24 °C [11] If droplet be pinned on surface (i.e., due to crystallization process) stagnation flow might have contradictory results which is presented in following.

Figure 11.2 shows sequential images of impacting supercooled water droplet on superhydrophobic surface maintained at -30 °C which is below critical temperature of heterogeneous nucleation (i.e., -24 °C). Droplet cannot be retracted when cold stagnation flow presents which is related to effect of both shear and normal force of incoming air flow and severe reduction of capillary force. In fact surface tension is reduced up to 3.5-folds to that of room temperature water drop by presence of induced homogeneous nucleation due to forced evaporation cooling. Therefore, solidified wetting diameter is increased up to two folds to that of still air condition.



Figure 11.1. Sequential images of supercooled water droplet impacts on superhydrophobic substrate (**a**); temporal evolution of spreading diameter (**b**). Droplet and substrate temperature are -3.5 °C and -20 °C, respectively.



Figure 11.2. Sequential images of supercooled water droplet impacts on superhydrophobic substrate maintained at –30 °C.

12. Liquid Properties of Water-Glycerol Solution Simulating Viscosity of Supercooled Water at Different Temperature

Values illustrated in Table 12.1 has been interpolated from references [12–15].

Table 12.1. Property of water-glycerol solution at different weight ratio along with super cooled water property at different temperatures.

% Weight of Glycerol in Water	Density of Water-Glycerol Solution (kg/m³)	Dynamic Viscosity of Water-Glycerol Solution (m·Pas)	Surface Tension of Water-Glycerol Solution (N/m)	Dynamic Viscosity of Water at Different Temperature (m·Pas)	Surface Tension of Water at Different Temperature (mN/m)	Density of Water at Different Temperature (kg/m³)
0	998.2	1.002	72.8	1.002 @ 20 °C	72.8@ 20 °C	998.2@ 20 °C
22.3	1052.8	1.93	70.8	1.93 @−2 °C	76@-2°C	999.7@−2 °C
23.8	1056.7	2.04	70.7	2.04 @-3.5 °C	76.2@−3.5 °C	999.5@−3.5°C
26	1062.4	2.205	70.5	2.205 @−5.5 °C	76.5@−5.5 °C	999.1@−3.5°C
38.2	1094.5	3.5	69.7	3.5 @−16 °C	78.5@-16°C	996.1@−16 °C
47.8	1120.4	5.5	68.7	5.5 @−24 °C	81.1@-24°C	984.3@−24 °C

13. Prediction of Maximum Spreading Diameter

Maximum spreading diameter was evaluated based on two models. First, energy based model proposed by Passandideh-Fard et al. [16] and second one is analytical hydrodynamic model presented by Roisman [17]. While for superhydrophobic surface experimental results were also compared with scaling law analysis and correlation presented by Clanet et al. [18]. In energy based model near wall boundary layer thickness was used for prediction of maximum spreading diameter instead of thickness of splat which overestimates maximum spreading diameter. Furthermore, effect of surface wettability was considered which provides a comparison for wide range of surface wettabilities. Typically, when Weber and Reynolds numbers are large enough, predicted maximum spreading values agrees well with experimental results. This model is presented in Equation (13.1) which is based on advancing contact angle during spreading phase.

$$\zeta = \frac{D}{D_{max}} = \sqrt{\frac{We + 12}{3(1 - \cos\theta_a) + 4\frac{We}{\sqrt{Re}}}}$$
(13.1)

The second model is based on direct usage of continuity and momentum equations. In this analytical hydrodynamic model proposed by Roisman [17] effect of rim edge on maximum spreading diameter was also considered. Velocity distribution in viscous region was derived based on analytical self-similar solutions which satisfy Navier-Stocks equation. Finally, based on some experimental evidence a semi-empirical engineering correlation was proposed to predict maximum spreading diameter, as illustrated in Equation (13.2):

$$D_{\max} \approx 0.87Re^{\frac{1}{5}} - 0.4Re^{\frac{2}{5}}We^{-\frac{1}{2}}$$
(13.2)

In this correlation effect of surface wettability was not considered as it was observed that if Weber and Reynolds number are high enough maximum wetting diameter is independent of surface wettability [19].

In scaling law analysis, which was just used for evaluation of superhydrophobic surface, it was claimed that P factor (i.e., $We/Re^{0.8}$) determines dynamics of wetting. For an impacting drop with p < 1, maximum wetting diameter is just influenced by capillary force and it is independent of viscosity and can be approximated by $D_{\text{max}}/D_o = 0.9We^{0.25}$. While for p > 1 dynamic of drop followed by viscous force and represented by $D_{\text{max}}/D_o = 0.9Re^{0.2}$. In current study p vary from 0.11 to 0.21. As it was observed experimentally correlation of Clanet et al. [18] predicts maximum spreading diameter very well for superhydrophobic surface in room temperature condition.

References

- 1. Alizadeh, A.; Yamada, M.; Li, R.; Shang, W.; Otta, S.; Zhong, S.; Ge, L.; Dhinojwala, A.; Conway, K.R.; Bahadur, V.; et al. Dynamics of ice nucleation on water repellent surfaces. *Langmuir* **2012**, *28*, 3180–3186.
- 2. Levi-Hevroni, D.; Levy, A.; Borde, I. Mathematical modeling of drying of liquid/solid slurries in steady state one-dimensional flow. *Dry. Technol.* **1995**, *13*, 1187–1201.
- 3. Touloukian, Y.; Saxena, S.; Hestermans, P. *Thermophysical Properties of Matter: The TPRC Data Series*; Purdue University, West Lafayette, IN, USA, 1975; Volume 11.
- 4. McQuillan, F.; Culham, J.; Yovanovich, M. *Properties of Dry Air at One Atmosphere*; University of Waterloo–Microelectronics Heat Transfer Laboratory Report UW MHTL; University of Waterloo–Microelectronics Heat Transfer Laboratory: Waterloo, ON, Canada, 1984
- 5. Brown, P.P.; Lawler, D.F. Sphere drag and settling velocity revisited. *J. Environ. Eng.* **2003**, *129*, 222–231.
- 6. Luxford, G. Experimental and Modelling Investigation of the Deformation, Drag and Break-Up of Drizzle Droplets Subjected to Strong Aerodynamics Forces in Relation to SLD Aircraft Icing. Ph.D. Dissertation, Cranfeild University, Cranfeild, UK, 2005.
- 7. Kobus, C.; Shumway, G. An experimental investigation into impinging forced convection heat transfer from stationary isothermal circular disks. *Int. J. Heat Mass Transf.* **2006**, *49*, 411–414.
- 8. Sahoo, B.; Labropulu, F. Steady homann flow and heat transfer of an electrically conducting second grade fluid. *Comput. Math. Appl.* **2012**, *63*, 1244–1255.
- 9. Bahadur, V.; Mishchenko, L.; Hatton, B.; Taylor, J.A.; Aizenberg, J.; Krupenkin, T. Predictive model for ice formation on superhydrophobic surfaces. *Langmuir* **2011**, *27*, 14143–14150.
- 10. Maitra, T.; Tiwari, M.K.; Antonini, C.; Schoch, P.; Jung, S.; Eberle, P.; Poulikakos, D. On the nano engineering of superhydrophobic and impalement resistant surface textures below the freezing temperature. *Nano Lett.* **2013**, *14*, 172–18210.
- 11. Eberle, P.; Tiwari, M.K.; Maitra, T.; Poulikakos D. Rational nanostructuring of surfaces for extraordinary icephobicity. *Nanoscale* **2014**, *6*, 4874–4881.
- 12. Speedy, R.J. Thermodynamic properties of supercooled water at 1 atm. J. Phys. Chem. 1987, 91, 3354–3358.
- 13. Hacker, P.T. *Experimental Values of the Surface Tension of Stjpercooled Water;* National Aeronautics and Space Administration: Houston, TX, USA, 1951.
- 14. Hallett, J. The temperature dependence of the viscosity of supercooled water. *Proc. Phys. Soc.* **1963**, *82*, 1046–1050.
- 15. Glycerine Producers' Association. *Physical Properties of Glycerine and Its Solutions*; Glycerine Producers' Association: New York, NY, USA, 1963.
- 16. Pasandideh-Fard, M.; Qiao, Y.; Chandra, S.; Mostaghimi, J. Capillary effects during droplet impact on a solid surface. *Phys. Fluids* **1996**, *8*, 650–659.
- 17. Roisman, I.V. Inertia dominated drop collisions. II. An analytical solution of the Navier–Stokes equations for a spreading viscous film. *Phys. Fluids* **2009**, *21*, 052104.
- Clanet, C.; Béguin, C.; Richard, D.; Quéré, D. Maximal deformation of an impacting drop. J. Fluid Mech. 2004, 517, 199–208.
- 19. Rioboo, R.; Marengo, M.; Tropea, C. Time evolution of liquid drop impact onto solid, dry surfaces. *Exp. Fluids* **2002**, *33*, 112–124.



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