Icephobic Performance of Superhydrophobic Coatings: A Numerical Analysis

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Abstract

Understanding the mechanism of cloud-sized particles impact and freezing is crucial to find viable solutions to prevent ice accumulation on critical aerodynamic surfaces such as aircraft wing or nacelle. It has been reported that superhydrophobic surfaces (SHS) have promising anti-icing properties due to their excellent water-repellent characteristics. However, the anti-icing performance of such surfaces has not been fully understood. A multi-region multiphase flow solver including phase change has been employed to model the icing of a micro-droplet as it impinges on a superhydrophobic substrate with a given thickness, texture, and solid material thermal properties. The Navier-Stokes equation expressing the flow distribution of the liquid and the gas, coupled with the volume of fluid (VOF) method for tracking the liquid-gas interface, was solved numerically using the finite volume methodology. The superhydrophobic morphology is modelled through series of micro-structured arrays with squared cross sectional pillars. As such, thermal contact resistance is directly modeled by inclusion of air pockets underneath the micro-droplet. Consequently, direct effect of surface topology and thermal properties on droplet maximum spreading diameter, penetration to the surface asperities, contact time, and the freezing onset have been investigated.

1. Introduction

Water droplet freezing has received increased attention in numerous fields more specifically in aviation industry to analyze the ice accretion mechanism due to the impact and solidification of cloud droplets. The resulting in-flight icing can take place on aircraft wing, tail, engine or instruments leading to decrease the aerodynamic performance which could result in a lack of control or loss of thrust and constitute a major safety and security issue. Hence, Understanding the physics governing the process of water droplet freezing on various surfaces would be necessary to develop more efficient anti and de-icing solution. Assuming a similar surface energy characteristics for ice and water, it has been a common approach to correlate the surface hydrophobicity to the ice adhesion. Consequently, Superhydrophobic surfaces (SHSs) have been considered as suitable icephobic candidates for their extraordinary capability of reducing adhesion with water,^{7,14,15,18,27} by minimizing droplet contact time and consequently reducing or eliminating ice formation.^{2,16}

Even though many experimental,^{20,23,25} numerical^{9,19} and analythical²¹ studies have focused on the impact and freezing of milliliter size droplets on superhydrophbic surfaces, less is known to investigate the behavior of micro-drops on such surfaces and considering the direct effect of surface asperities in their models. Because of the smaller mass of micro-droplets, freezing occurs in much shorter time which can deteriorates ice-phobicity of the surface faster. It has been found that there are several parameters directly or indirectly control the ice-phobicity of a surface. For instance, in rapid cooling process Qing Xu et al.,²⁸ found that the material properties of the substrate may change the retraction time, and the spreading diameter of the retracting droplet. Huang et al.,¹² studied freezing of water droplets on copper surface for wide range of surface temperatures. They showed that freezing time significantly changed by the wettability and the temperature of the surface.

Since the freezing of water droplets is a heat transfer driven process, several numerical simulations have been carried out.^{17,19} Blake et al.,³ studied the conduction of the heat inside a millimiter-size droplet when the surface was subjected to a constant surface temperature. However, the constant temperature boundary condition in their model cause the droplet to solidify faster than actual droplets. Schremb et al.,²⁴ recently presented a computational model based on a volume of fluid method for a non-isothermal two-phase flow applying for the simulation of water droplet

impingement on a dry cold surface. It was shown that the amount of heat transferred between the fluid and the wall is affected by the contact time and the area available for heat transfer, which are determined by the impact conditions.

In addition, reducing roughness can increase the free energy barrier for heterogeneous nucleation which results in significant freezing delay. Hao et al.,¹⁰ focused on the freezing delay and the freezing time of sessile droplet on smooth, micro-structured and micro/nano-structured surfaces. Their experimental data and theoretical analysis showed that the surface roughness plays a crucial role in nucleation time. In addition, they found that smooth surfaces with roughness smaller than the size of the critical ice nuclei have a larger freezing delay than superhydrophobic surfaces with hierarchical structures. Visser et al.,²⁶ studied water microdroplet impact at velocities ranging from 1 to 100 m.s^-1 on smooth solid surface for droplet diameters ranging from 12 to 100μ m which can cover transition between surface tension and viscosity-dominated spreading of the droplet. They illustrated the importance of thin boundary layer over the surface in which majority of viscous dissipation occurs.

Time scale for micro-droplet impact spreading on superhydrphobic surface is very short, on the order of microseconds which makes it fairly difficult to visualize experimentally. More importantly, because of the optical limitation inside the droplet, experimental analysis of small micro-droplets (i.e. diameter range between 40-100 μ m) impact on rough surfaces is complicated to control. In addition, understanding the effect of surface topology and thermal properties of the substrate are of utmost importance for fabrication of functional ice-phobic surfaces. Therefore, in this study we employed a 3D computational analysis to evaluate the effect of two different substrate materials accompanied with surface roughness on freezing process of 30 μ m droplets impingement.

1.1 Framework

The objective of this study is to develop a three-dimensional conjugate heat transfer (CHT) module based on Open-FOAM package utilities, to simulate the complicated thermal process of a cloud-size droplet icing phenomena on superhydrophobic surfaces. This procedure is based on a continuous exchange of boundary conditions at fluid/solid including water-solid and air-solid interfaces. It couples together the Navier-Stokes flow solver, the Eulerian droplet impingement module, a liquid to solid phase change module and the solid conduction module simultaneously.



Figure 1: Hierarchical overview of the implemented solver

The conjugate heat transfer method iteratively solves the thermal interactions between fluid and solid domains. The coupled method can directly calculate the heat transfer loads at fluid-solid interfaces instead of using empirical correlations and its results is more accurate than empirical methods if the flow field is refined enough.

1.2 Governing equations

The model is formulated to simulate the impact of a micro-droplet on a superhydrophybic substrate at the instant the droplet comes into contact with the surface and proceeding until the onset of freezing is captured. Formulation of

transport equations for incompressible, laminar flow of Newtonian fluids, phase change, and conjugate heat transfer with the solid surface are as described in the following section.

Continuity equation

In the freezing of a micro-droplet, mass transfer from one phase to another is a local phenomena and does not change the global continuity equation,

$$\frac{\partial \rho}{\partial t} + \vec{\nabla}.(\rho \vec{V}) = 0. \tag{1}$$

Energy Equation

The heat transfer between water, ice, gas and the substrate is addressed using an energy conservation equation based on the enthalpy formulation as follow,

$$\frac{\partial \rho H}{\partial t} + \vec{\nabla} . (\rho \vec{V} H) = \vec{\nabla} . \left[\vec{\nabla} (kT) \right], \tag{2}$$

where ρ is the density. C_p , the heat capacity, k the thermal conductivity, V_0 and ΔT are the characteristic velocity and temperature, respectively. Since the Eckert number ($Ec = V_0^2/C_p\Delta T \ll 1$), representing the ratio between the kinetic energy to the enthalpy is too small, the viscous dissipation is neglected.

The enthalpy of the material is computed as the sum of the sensible heat (h) and the latent heat (ΔH),

$$H = \underbrace{h_{ref} + \int_{Tref}^{T} C_p dT}_{h} + \underbrace{\alpha \gamma L_f}_{\Delta H}, \tag{3}$$

where, h_{ref} is the reference enthalpy which is zero for pure water, T_{ref} , the reference temperature when the phase change starts. In order to account for phase change during the freezing, the specific sensible enthalpy is extended by the latent heat of fusion L_f . This latent heat should be multiplied by the liquid-solid fraction, γ , within the computational cells and this fraction satisfies:

$$\gamma = \begin{cases} 0 & ice \\ 1 & wate \end{cases}$$

Therefore the energy equation can be formulated in terms of the temperature by inserting in Eq.2,

$$\frac{\partial(\rho C_p T)}{\partial t} + \vec{\nabla}.(\rho C_p \vec{V}T) = \vec{\nabla}.\left[\vec{\nabla}(kT)\right] \underbrace{-L\left[\frac{\partial \rho \gamma}{\partial t} + \vec{\nabla}.\left(\rho \vec{V}\gamma\right)\right]}_{S_H},\tag{4}$$

where, S_H is a source term accounting for the phase change.

There is a special treatment to be performed when solving the enthalpy formation (Eq.2) which requires a scheme for the enthalpy to be carried out at each outer iteration of the energy equation.⁴ This scheme is used to ensure boundness and convergence. In the present study, to avoid the classical numerical ad hoc treatment for solving the energy enthalpy-based equation and due to the severe transition for water freezing, an approximate analytical function of the water-ice fraction is proposed and expressed as follows:

$$\gamma = \frac{1}{2} [1 + \tanh(k_l(T - T_{ref}))],$$
(5)

where k_l is an adjusting parameter indicating the severity of phase transition. The liquid fraction in the freezing liquid dependenting on temperature indicates that phase change can occur in a very narrow temperature range as for water or any pure substance.

Heat transfer within solid domain can be described by Fourier's law defining the conductive heat flux, q, proportional to the temperature gradient ($q = -k.\vec{\nabla}T$). For a time-dependent problem, the temperature field in an immobile solid is governed by the following heat equation,

$$\rho_s C_{ps} \frac{\partial T_s}{\partial t} = \vec{\nabla} . (k_w \vec{\nabla} T_s). \tag{6}$$

Liquid Fraction Equation

The liquid-gas interface is implicitly determined by the field volume fraction for every computational cell. The advection of interface is based on an existing multiphase solver (interFoam) which utilizes Volume of Fluid (VOF),¹¹ an eulerian interface capturing method. The transport equation accounting for the advection and compression of the interface is as follow,

$$\frac{\partial \alpha}{\partial t} + (\vec{V}\vec{\nabla})\alpha + \vec{\nabla}.(\vec{V}_r\alpha(1-\alpha)) = 0, \tag{7}$$

where, $\vec{V_r}$ is the vector of relative velocity, and the last term is called artificial compression term contains the compression velocity which is computed to avoid smearing of the phase interface.²²

$$\vec{V_r} = n \min\left[C_a \frac{|\phi|}{|S_f|}, \max\left(\frac{|\phi|}{|S_f|}\right)\right],\tag{8}$$

where, C_a , n, ϕ , and S_f , are the compression coefficient, unit normal flux, face volume flux and cell face area vector, respectively. The interface unit normal \vec{n} is computed by taking the gradient of smoothed volume fraction, α , at the cell faces,

$$\vec{n_f} = \frac{\vec{\nabla}\alpha}{|\vec{\nabla}\alpha| + \delta},\tag{9}$$

where, δ is a small number in order to stabilize the calculation in regions outside the transition region where, $|\vec{\nabla}\alpha| \rightarrow 0$. In this study, $\delta = 10^{-8}$ is used for the entire simulations. The main benefit of such formulation is in the possibility of capturing the sharp interface region. The continuum surface force method[?] is used to model surface tension as a body force \vec{F}_b that acts only on interfacial cells,

$$\vec{F_b} = \sigma k \vec{\nabla} \alpha \quad and \quad k = -\vec{\nabla} \cdot \left(\frac{\vec{\nabla} \alpha}{|\vec{\nabla} \alpha|}\right),$$
(10)

where, σ is the surface tension, and k is the mean curvature of the free surface.

The physical properties of the liquid, gas and ice such as viscosity, μ , thermal conductivity, k, heat capacity, C_p , and the density, ρ , of the mediums are calculated based on the weighted average of, α , in the computational domain,

$$\phi = \alpha(\gamma \phi_l + (1 - \gamma)\phi_s) + (1 - \alpha)\phi_g \quad where, \quad \phi \in \{k, C_p\},\tag{11}$$

$$\phi = \alpha \phi_l + (1 - \alpha) \phi_g \quad where, \quad \phi \in \{\rho\}, \tag{12}$$

where, the subscripts *l* and *g* denote the liquid and gas phases, respectively. In VOF method the phase fraction scalers tracer, α , takes values between 0 and 1. When a cell is empty with no traced fluid inside, the value of α is zero; when the cell is full, α is equal to 1; and when there is a fluid interface in the cell, $0 < \alpha < 1$.

Momentum Equation

The momentum equation expressing flow distribution of the liquid and the gas, coupled with the Volume of Fluid (VOF) method for tracking the interface between the liquid and the gas are solved numerically using the following equation,

$$\frac{\partial \rho \vec{V}}{\partial t} + \vec{\nabla} . (\rho \vec{V} \vec{V}) = -\vec{\nabla} p + \nabla . (\mu \nabla \vec{V}) + \vec{F}_b + \underbrace{\alpha \beta \vec{V}}_{S_V}$$
(13)

where, \vec{V} is the velocity vector, p is the pressure, $\vec{F_b}$ is the body force, α is the volume fraction and S_V is the source term accounting for the viscosity change of the liquid droplet as it freezes. Carman-Kozney formulation⁵ used to adjust the coefficient β from zero for liquid to a large value as the liquid freezes,

$$\beta = \frac{\chi (1 - \gamma)^2}{\gamma^3 + \epsilon} \tag{14}$$

where, ϵ is a small value to avoid division by zero.

Surface Wettability

The wettability or hydrophobicity effect at the substrate are taken into account by implementation of Kistler's dynamic contact angle model.¹³

$$\theta_d = f_H [Ca + f_H^{-1}(\theta_e)], \tag{15}$$

where, Ca is the capillary number, f_H is the Hoffman's function and is defined as,

$$f_H = \arccos\{1 - 2\tanh[5.16[\frac{x}{1 + 1.31x^{0.99}}]^{0.706}]\}$$
(16)

in the above equation (Eq.15) the equilibrium contact angle θ_e is replaced by either the advancing contact angle, θ_A , or the receding contact angle, θ_R , depending on the direction of the velocity vector at the contact line or the static contact angle, θ_s , if the contact line velocity is zero. Among no-slip models, Kistler's correlation¹³ have been extensively used to investigate droplet dynamics, and proven to be in good agreement with the experimental observation.⁸ In addition, it is shown that this model is capable to capture the relevant physics of microdroplet-substrate interaction and provides good agreement with experiment.¹

2. Geometrical Configuration and Meshing

The computational domain size of $100 \ \mu m \times 100 \ \mu m \times 400 \ \mu m$ is used for the 3D simulation of the impinging droplet onto a cold substrate with a given thickness. The mesh dependency test is performed based on the jumping velocity of droplet on superhydrophobic surface. The fluid domain is discretized by about 5 million, 8 million, and 12 million Hexahedral mesh with adaptive refinement at the interfacial cells. As the change of droplet jumping velocity in 8 million and 12 million mesh was minimal, the domain composed of 8 million fluid region mesh is applied for this study. According to Attarzadeh and Dolatabadi,¹ for droplets smaller than 70 um direct modeling of surface roughness is crucial to simulate the dynamic of micro-droplet on a superhydrophobic surface. Superhydrophobicity is modeled as a series of micro-structured pillars with squared cross section and squared patterns. The static, advancing and receding contact angles of 111°, 120°, and 86° respectively are imposed over the surface of the pillars.



Figure 2: 3D computational domain with surface configurations (left), three coating morphologies used in this study (right), a) 2w3p, b) 2w4p, c) 2w5p

The substrate thickness of 30 μ m is extracted from the domain. Series of hydrophobic pillars are implanted on top of the solid surface with 2 μ m height and width but varying spacing between two adjacent pillars (1, 2, and 3 μ m). The material properties of the substrate is given in Table1.

3. Fluid-Solid Coupling

In this study, the partition method has been used to couple the temperature gradient between solid and fluid regions. Unlike the monolithic approach which uses a single matrix to solve the governing equation for both regions simultaneously, partitioned approach seeks to utilize existing fluid and solid solutions and couple these solvers at the interface

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of the two domains. The fluid solver uses a separate set of equations, variables and mesh than the solid solver. The coupling happens at the boundary $(\lambda_f \& \lambda_s)$ where the temperature from the fluid updates the heat flux at the boundary of the solid which is then solved to determine gradient temperature at the interface boundary (Neumann boundary condition). This updated boundary is then used as a Dirichlet boundary condition to solve the fluid domain. At the coupling interface λ the following condition must be satisfied,

$$\begin{cases} T_{\lambda \to f} = T_{\lambda \to s} = T_{\lambda} \\ k_f \left(\frac{\partial T_f}{\partial n_f} \right) = k_s \left(\frac{\partial T_s}{\partial n_s} \right) \end{cases}$$
(17)



Figure 3: Geometrical configuration of the computational domain, a) fluid and solid b) fluid and solid separated, 3) fluid and solid coupling patches.

where, the subscript f and s corresponds to the fluid and solid neighboring cells near the interface (λ) .

Table 1. Material properties.				
	$\rho(\frac{kg}{m^3})$	$K(\frac{kg.m}{s^3.T})$	$C_p(\frac{m^2}{s^2 \cdot T})$	$\nu(\frac{m^2}{s})$
Air	1	0.02	1000	$1.78*10^{-5}$
Water	1000	0.6	4200	$1.0*10^{-6}$
TiO ₂	4230	11	690	-
Al	2700	205	900	-

Table 1: Material properties

4. Results and Discussion

The effect of surface roughness on the dynamic and transient heat transfer of the micro-droplet with the solid surface has been examined. Figure 4 shows the impact of a room temperature micro-droplet on a textured (2w3p) superhydrophobic surface (TiO₂, Al). The droplet and surface are assumed to be in thermal equilibrium and above freezing temperature $(T = 17^{\circ}C)$. The droplet $(D = 30\mu m)$ was initialized close to the substrate at the instant of impact with 1.6 $m.s^{-1}$ terminal velocity. The exposed area of the substrate is coated with sub-micron thin layer of Stearic acid which has the intrinsic contact angles of $\theta_S = 110^{\circ}$, $\theta_A = 120^{\circ}$, $\theta_R = 86^{\circ}$ which by nature is not superhydrophobic. According to Cassie-Baxtar model⁶, physical roughness reduces energy barrier between the substrate and the droplet. It has been observed that roughness can increase the apparent contact angle above 150°. Because of the reduced surface energy of the surface, droplet relatively spread less comparing to that on smooth surface and was able to detach after 22 μs . Since the substrate and droplet are in thermal equilibrium above freezing temperature, the thermal properties of neither TiO₂ nor Al changes the hydropdynamic of impact, while it solely depends on the physical properties of the surface in contact with droplet.

Figure 5 shows the impact of a room temperature micro-droplet on a $-5^{\circ}C$ Al substrates similar to the previous case which was micro-fabricated using laser patterning (2w3p) and coated with Stearic acid. Comparing two figures 5 and 4, it is evident that the dynamic of droplet changed after $t = 13\mu s$. Because of the heat transfer between droplet and the substrate, droplet showed no tendency to fully recoil and consequently remains on the surface.

Figure 6 illustrates a similar impact scenario but on Titanium dioxide, TiO_2 . Even though hydrodynamic of droplet found to be same on Al and TiO_2 , different outcome was seen. The different arises when the droplet reach its maximum



Figure 4: Time lapse impact of a room temperature micro-droplet ($V = 1.6m.s^{-1}$) on textured (2wp3) superhydrophobic surface.



Figure 5: Time lapse impact of a micro-droplet impact ($V = 1.6m.s^{-1}$, $T = 17^{\circ}C$) onto a cold Al textured substrate ($T = -5^{\circ}C$).

spreading length and there is an uneven exchange of heat between surface and droplet cause droplet freezes ans stick to the substrate.

Figure 7 shows the mechanism in which the ice is formed inside the micro-droplet as it impinges on Al substrate. It has been shown that there is an exchange of heat between the droplet and the substrate since the beginning of the impact ($t = 2\mu s$). The temperature contour within the solid bulk, and the droplet interface are presented individually. The white nuclei shows the onset of icing. At ($t = 5\mu s$) the temperature of liquid and solid are at freezing temperature and the latent heat starts releasing. During this time, the temperature of liquid remains unchanged where the substrate heats up until the phase change is completed. At ($t = 7\mu s$) the entire latent energy of the cells are released from the peripheral region of the liquid droplet and icing nuclei starts forming. For water, the ratio of latent heat to sensible heat is about an order of magnitude greater than other materials. This means that the discontinuity at the freezing front for water is much more severe when phase change starts. Because of the large ratio of sensible heat to latent heat of the water, there is a severe transition between water to ice thermal properties. With current methodology, the severity of the freezing front from water to ice has been well captured. At ($t = 20\mu s$) the triple point is pinned on the surface and prevents droplet to complete its retraction stage. From this moment beyond, droplet is stuck on the surface and the ice layer keeps growing. The heat transfer rate on TiO₂-2w3p was relatively slower comparing to that on Al which cause droplet to bounce off before the entire energy of latent is released. This mechanism can even be empowered by changing the morphology of the surface.

The maximum wetted diameter becomes larger as the post spacing length is increased between adjacent pillars. Relative quantitative information is given in figure 8. The maximum spreading diameter for droplets in collision with Al and TiO₂ on w2p3 is same. However, due to the freezing phenomena on Al surface, the triple point of the droplet



Figure 6: Time lapse impact of a micro-droplet impact ($V = 1.6m.s^{-1}$, $T = 17^{\circ}C$) onto a cold TiO₂ textured substrate ($T = -5^{\circ}$).



Figure 7: Time lapse sequence of the freezing front dynamic inside the micro-droplet on a ($V = 1.6m.s^{-1}$, $T = 17^{\circ}C$) a cold Al substrate

pinned to surface and preventing droplet to complete retraction stage. We also examined the wetted area on w2p4 and w2p5. However, these two patterns hydrodynamically are not water repellent and droplet impales into the surface cavities, and breaks down the hydrophobicity of the surface.

5. Conclusion

In this paper a volume of fluid (VOF) based multi-region conjugate heat transfer model is developed to undertake microdroplet impact on textured surfaces accompanied by continues exchange of heat between solid and fluid including phase



Figure 8: Temporal evolution ($T = tV_0/D_0$) of the spreading diameter ($\beta = D/D_0$) along the lattice axis direction after water droplet impact on different surfaces topologies.

change phenomena. The model is based on the enthalpy formulation of the energy equation. Coupling the momentum equation and contact angle model enabled us to directly model the effect of wall contact resistance through inclusion of surface asperities into the model. Further conclusions are drawn as follow,

- 1. The method used in this paper can simulate the in-cloud icing of droplet impingement on superhydrophobic surface accompanied with modeling of surface asperities under different conditions and surface morphology.
- 2. The textured patterns has a great influence on the hydrodynamic of micro-droplet impact. Meanwhile, these patterns (roughness) can also change the contact resistance of the surface.
- 3. For substrates with high thermal diffusivity, ice growth initiates from the periphery of the droplet in the maximum spreading stage, while for low thermal diffusivity, ice growth occurs from the center point of impact.

Future work will address the extension of the model to account for the impact of a supercooled water droplet as well as the density change of the liquid droplet during freezing process.

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