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Study for critical roughness based on interfacial energy



PHYSIC

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ABSTRACT ARTICLE INFO Keywords: Solid-liquid contact state between water droplets and Superhydrophobic surface (SHS) is mainly determined by Superhydrophobicity the microstructures of a surface. Composite and non-composite wetting states (NCWS) even together with their Hierarchy mixture are generally regarded as basic states. For an SHS, a composite wetting state (CWS) is more preferred Wetting state due to water droplet easier to slide. Therefore, determining a critical roughness to form, then to maintain such a Transition state has profound significance for fabrication of SHS. For this reason, we propose a thermodynamic metho-Adhesion work dology based on a three-dimensional (3-D) pillar model, integrating wetting states (WS) with interfacial energy, Contact angle to obtain the corresponding critical roughness for the reference of micromachining. By calculating, we found that the critical roughness for a transition from NCWS to CWS changes with inherent materials from aspect of

1. Introduction

The secret of the "lotus effect" having been revealed, promotes the appearance of artificial superhydrophobic surface (SHS) [1-3], and the study of new surface science on superhydrophobicity. Theoretically, people often described the system of both droplet and solid surfaces from the following several aspects, e.g., static or dynamic wetting feature (for instance, intrinsic or apparent contact angles (APCA), contact angle hysteresis (CAH), sliding angles (SA) [4-13] along with the three-phase contact line) [4,5,14-16]; composite or non-composite wetting states (NCWS) along with transition between them; surface roughness together with its influence on the superhydrophobicity [4,7-9,11,17-20]; surface or interface free energy (FE) including FE barriers, adhesion work (W_a), and spreading coefficient [10,13]. Actually, considering the interplay from the solid-liquid interface, it is more significant to study the wetting states (WS) together their transition, and to explain the superhydrophobicity from the surface FE viewpoint.

The SHS (CA > 150°, CAH < 10° or SA < 5°) creating a large CA and little CAH, results in the reduced adhesion or friction efficiently, hence has ideal water–repellence property, and acquires widespread application in industry, such as glass coatings, bio–microfluidics, pesticides, anti–corrosion [21–25]. Moreover, it would be possibly used for anti–icing adhesion of high–voltage Cable and aircraft surfaces in the

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Received 9 April 2018; Received in revised form 20 April 2018; Accepted 29 April 2018 Available online 04 May 2018 0921-4526/ © 2018 Elsevier B.V. All rights reserved. near future [26–29]. Especially, for the SHS, when a droplet is deposited on it, if the WS appears to be a composite, the adhesion from the solid–liquid interface is largely reduced, thus leading towards a large CA and small CAH. Consequently, we may study the SHS from the angle of WS or interfacial FE. However, up to now, there is no still enough study targeting to the integration of the WS and the interfacial FE.

adhesion work; furthermore, if given hydrphobic materials, such roughness is also given and not relevant to the fractal dimension of an SHS. Therefore, the hydrophobic materials determine the transition. Meanwhile, we also

proved the above finds by interfacial energy and contact angle (CA) varying with the roughness.

Microscopic observations show that the CAH results mainly from the surface heterogeneities and microscopic roughness, even droplet velocity, especially that of three phase contact line [5,11,13,30–35]. Interestingly, for the SHS, the roughness is more a key factor to create the hydrophobicity than a chemical composition (especially when the materials is given), e. g., largely decreasing the effect of chemical compositions on the hydrophobicity in the CWS, or causing large CAH in the NCWS [5,8,9,36,37]. Furthermore, in some cases, certain roughness pattern possibly creates a transition from CWS to NCWS, or a possibility of making high-contact-angle rough surface from low-contact-angle materials [7,13,31,38,39]. Obviously, such transition has real value. Therefore, researchers studied it from different angle, i.e., FE barrier [10,40-42], CAH [31,40], surface tension and relevant experiment [19,43-51], etc., and found different critical roughness for a transition. However, these studies are only limited to both the outside behavior (i.e., pressure, vibrating, electrical voltage, evaporating) and the macroscopic performance (i.e., CA, CAH, surface FE) of the droplets-surfaces system, rather than from the real solid-liquid interface energy. Furthermore, current studies are mostly restricted in the transition from CWS to NCWS, instead of its invers. Thus little research has been devoted to the integration of the WS and the W_a . Actually, for the SHS, study of how to create CWS and maintain it by surface micro/nano design has more real significance. Although our former study also mentioned the W_a for finding the critical roughness factor [52,53], it is only primary thought, far from perfection, without consideration of the transition state from the APCA and interfacial FE angles, when the roughness changes. For our part, now that the definition of the WS is based on the contact between water droplet and the solid surface, we think that, when the roughness changes, it is still reasonable to describe the WS along with its transition from point of the W_a . By doing so, the obtained critical roughness can meet with the requirement of micromachining.

For the purpose of finding the critical roughness to form the CWS (or to realize the transition), and providing a reference for fabrication of an SHS (for micromachining technology, e.g., lithography, metalassisted chemical etching, grinding, nanoimprint lithograhpy) to obtain a roughness higher critical value used for maintaining such CWS, our research mainly focused on three aspects of creative work as follows: Firstly, a dependence relationship only between the microstructure and the interfacial FE was established by modeling a 3-D SHS with multi-scale, but without considering the external incitation, for example, mechanical/magnetic/thermal/electrostatic/even gravity effect. Secondly, the critical roughness was obtained by equaling the W_a of both CWS and NCWS. To the end, we also demonstrated the rationality and reliability of the used methodology by equaling solid-liquid interfacial FE, or APCA [31], between CWS and NCWS, when the transition from NCWS to CWS occurs.

2. A thermodynamic methodology to compute the Wa

2.1. Some basic hypothesis

For illustrative purpose, we make some assumptions or hypothesis to simplify the theoretical analysis for the real SHS.

- Considering the capillary length, negligible gravity, the shape of a waterdroplet may be regarded as an ideally spherical segment [54–56]. Consequently we study a droplet with a macroscale of 1–2 mm being much larger than the characteristic scale of the surface configuration.
- Relationships between the W_a and the microstructure are established based on the same materials and the environmental conditions (e.g., temperature, humidity, pressure, acidity).
- 3) To avoid a debate on Wenzel and Cassie theories [16], we only limit our computation to the equations of Cassie–Baxter's and Wenzel's, and do not need to consider the other WS, i.e., gecko state, rose petal effect [36,57].

2.2. Young's equation along with its modified

For an ideal smooth surface (atomically flat, chemically homogenous, isotropic, insoluble, non-reactive, and non-deformed) (Fig. 1), Young's equation (1) is used to describe the relationship between intrinsic CA and surface tension,

$$\gamma^{sa} = \gamma^{sl} + \gamma^{la} \cos \theta_{\rm Y} \tag{1}$$

For a real solid surface, if necessarily establishing the relationship between the interface tension and the APCA, we may modify Young's equation. As a modified form, it may be denoted as follows,

$$\gamma_r^{sa} = \gamma_r^{sl} + \gamma^{la} \cos \theta_A \tag{2}$$

unlike equation (1), where γ_r^{sa} , γ_r^{sl} respectively represents the surface tension of the real solid–air, solid–liquid interfaces, rather than those of



Fig. 1. A droplet on the ideal and smooth surface with homogenous, flat and rigid structure, where θ_Y is the intrinsic CA only used for Young's model, γ^{sa} , γ^{sl} , γ^{la} are the surface tension of solid–air, solid–liquid, and liquid–air interfaces, respectively. The equation (1) connects the interface tension with the intrinsic CA, suggesting the key role of the interface tension (as property parameter) to the intrinsic CA. if such a surface is not ideal and smooth, the corresponding parameters must be substituted by real ones, i.e., θ_Y by θ_A (real APCA), γ^{sa} , γ^{sl} by γ^{sa} , γ^{sl} (real solid–air and solid–liquid interfacial tension) respectfully.

ideal solid surface; γ^{la} keeps constant for purity liquid at constant environment (i.e., pressure and temperature); θ_A represents the real APCA (Cassie or Wenzel CA, the corresponding state of which has the minimal surface FE) rather than intrinsic CA (θ_y), thus θ_A supersedes θ_y in equation (1). Otherwise, such modification has no foundation. As long as the droplet is in the equilibrium state, the above equation always can be established according to the equilibrium conditions of forces.

2.3. Young-Dupre equation along with its modified

For the solid–liquid interfaces (Fig. 1), its adhesion work (denoted as lowercase w) from unit area may be denoted as below,

$$w_a = \gamma^{sa} + \gamma^{la} - \gamma^{sl} \tag{3}$$

according to Young's equation, the above equation may be simplified as follows,

$$w_a = \gamma^{la} (1 + \cos \theta_y) \tag{4}$$

namely Young–Dupre equation, which is only applied to the homogenous and smooth surface, thus θ_{γ} is the intrinsic CA.

For the real rough solid surface, we also obtain a similar simplified form,

$$w_a = (\gamma_r^{sa} + \gamma^{la} - \gamma_r^{sl}) = \gamma^{la}(1 + \cos\theta_A)$$
(5)

Thus the normalized adhesion work $\left(w_{ai}\right)$ of the unit area may be denoted in the following,

$$w_{ai} = \frac{w_a}{\gamma^{la}} = (1 + \cos \theta_A) \tag{6}$$

where the simplified Young–Dupre equation establishes a link between the w_a of the real solid–liquid interface and the equilibrium APCAs, subscript i stands for surface fractal dimension. Note that θ_A represents the real APCA rather than the intrinsic CA.

2.4. Determination of normalized total adhesion work (W_a)

When a droplet is dropped onto the SHS, the droplet will be dew it. At constant temperature and pressure, we can find the total normalized total adhesion work W_a with respect to γ^{la} in the equilibrium state based on Eq. (6):

$$W_a = (1 + \cos \theta_A) \times \pi \times (R_1 \sin \theta_A)^2$$
⁽⁷⁾

enlightened by the above equation, given R_1 of droplet radius, if the APCA (θ_A) is also given, we can find the corresponding normalized W_a .

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