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ABSTRACT
Hemiwicking has been introduced to describe the wetting state in which a liquid film surrounds a drop. To fully understand this special wetting state, we performed energy analysis and systematic lattice Boltzmann (LB) simulations on the wetting state through spreading liquid droplets on pillared hydrophilic substrates. Although the energy analysis shows that the hemiwicking is energetically unfavorable, droplets in stable hemiwicking are indeed observed in our LB simulations. This observation led us to conclude that we have obtained a result that is the same as the result obtained in the published experiment and theory: hemiwicking is dynamically trapped by the pinning of the imbibition front during invasion of the substrate texture by the liquid film. Our simulations show that the special wetting state is always found to emerge near the phase boundary between the liquid film and the Wenzel state. For the morphology of the droplet, strong deviation of the apparent contact angle from hemiwicking is observed when the contact line of the liquid imbibition film is close to the spherical caplike droplet. We also show that there exist at least two different kinetic pathways for the formation of hemiwicking, including spreading and evaporation.

INTRODUCTION

Controlling wettability of textured substrates is an important aspect of material science and surface chemistry, which has a variety of practical applications in industry, agriculture, and daily life. When a liquid droplet is in contact with a textured substrate, it can stay either in a Cassie state with gaseous pockets trapped inside substrate roughness or in a Wenzel state with liquid penetrating the roughness. Through coating substrates with properly designed roughness, sessile droplets on the substrates can display superhydrophobic2–4 or superhydrophilic behaviors.

Apart from the Wenzel and Cassie wetting states, intermediate situation hemiwicking5–7 can occur, in which the liquid forms a film on which the drop sits. Different from the Cassie and Wenzel states, hemiwicking includes liquid penetrating into the grooves of the surface texture and spreading toward the front of a droplet to form a liquid film surrounding the rest of the droplet. Several studies showed that spontaneous liquid penetration and the dynamics of subsequent spreading are crucially influenced by the microstructure of substrates.8–15 Thermodynamic analysis showed that for the occurrence of hemiwicking, a critical angle \( \theta_c \) of \( \cos \theta_c = (1 - f)/(r - f) \) should satisfy the requirement of \( \cos \theta_Y > \cos \theta_c \), where \( f \) is the solid fraction (i.e., the fraction of the solid/liquid interface below the drop), \( r \) is the roughness factor (i.e., the ratio of the actual surface over the projected one), and \( \theta_Y \) is the Young contact angle. The energies for the Wenzel state and hemiwicking...
are analyzed and compared as a function of texture parameters.\textsuperscript{29} When the textured substrate is sufficiently hydrophilic to allow the liquid to penetrate spontaneously into the substrate texture, the developed theory asserted that the drop would develop into hemiwickling.

The aforementioned reports provide valuable insights into understanding hemiwickling.\textsuperscript{37-39} Here, we perform energy calculations and systematic lattice Boltzmann (LB) simulations in order to prove the stability mechanism and formation pathway for hemiwickling. For the parameter space that was studied, hemiwickling is energetically unfavorable in comparison with either the complete wetting (liquid film) state or the Wenzel state. However, from LB simulations, we indeed observed the formation of hemiwickling, which was also observed from the aforementioned reports.\textsuperscript{37-39} The stability of the state is due to the pinning of the imbibition front, which stops the liquid imbibition from developing into a full film state. We also demonstrate that the contact angle of the rest droplet follows the prediction of the hemiwickling situation only when its contact line is far from the pinned imbibition front. We found different pathways for droplets achieving hemiwickling and mapped out the entire phase through a diagram in order to understand the role of contact line pinning in droplet formation.

**LATTICE BOLTZMANN METHOD**

We applied the three-dimensional multicomponent and multiphase Shan-Chen (SC) type lattice Boltzmann (LB) method\textsuperscript{40,41} based on a D3Q19 lattice.\textsuperscript{42} The LB method is a numerically robust technique for simulating interfacial phenomena, and, in particular, the SC model has been successfully used to study droplet impact on pillarated substrates,\textsuperscript{43} the effects of the entrapped bubbles on flow,\textsuperscript{44} impingement of liquid drops on dry surfaces,\textsuperscript{45} drag reduction of superhydrophobic surfaces,\textsuperscript{46} surface roughness-hydrophobicity coupling in channel flows,\textsuperscript{47} and the references mentioned in the recent review.\textsuperscript{48}

In this work, all the quantities used are dimensionless and described in LB units. A simulation box of 200 $\times$ 200 $\times$ 60 was adopted, and the substrate at the bottom of the box was decorated with square shaped pillars to model physical roughness. While the length ($L$) and width ($W$) of the pillars were both fixed to 1, the height ($H$) and spacing ($S$) between the pillars were varied to model different levels of roughness, as plotted in Fig. 1. For initial configurations, a microdroplet with a radius of 20 was placed on the solid surface that is made of material with an intrinsic Young contact angle of 62.2°. The bounce back boundary condition was adopted for the solid boundary. More details of the simulation method\textsuperscript{49,50} are given in the supplementary material.

**RESULTS AND DISCUSSION**

Energy analysis shows that hemiwickling is energetically unfavorable

Here, we first performed thermodynamic analysis on the stability of hemiwickling for droplets on the hydrophilic textured substrates, by comparing their energies with those corresponding to the Wenzel state and the complete wetting (liquid film) state, by a method similar to that used by Ishino et al.\textsuperscript{29} Note that a criterion of $\cos \theta_C < \cos \theta_Y$ for the spreading of the liquid film in textured substrates has already been proposed by Bico et al.\textsuperscript{37} This condition indicates that if $\cos \theta_C < \cos \theta_Y$, the Wenzel state is of a lower energy, meaning that the textured substrate remains dry, and the Wenzel state appears as indicated in Fig. 2(a). However, if $\cos \theta_C > \cos \theta_Y$, the Wenzel state has a higher energy, and a liquid film state becomes stable. This implies that the droplet can spread towards the front to achieve either the film state or hemiwickling, as shown in Fig. 2(a).

Now, we further compared the energy for the film state with hemiwickling to decide which one should be stable under the condition of $\cos \theta_C > \cos \theta_Y$. If the droplet finally develops into hemiwickling [Fig. 2(a)], its energy $E_{CP}$ can be described by

$$E_{CP} = \gamma S_{up} + (\gamma_{SL} - \gamma_{SV}) r S_{down} + [(r - f)(\gamma_{SL} - \gamma_{SV}) + (1 - f)\gamma] S_{film},$$

(1)

in which $\gamma_{SL}$, $\gamma_{SV}$, and $\gamma$ represent the interfacial tension for the solid-liquid, solid-vapor and liquid-vapor interfaces, respectively. In the equation, the surface area $S_{up} = 2\pi R^2_p (1 - \cos \theta_p)$ and $S_{down} = \pi R^2_p \sin^2 \theta_p$, where $R_p$ is the radius of the droplet in hemiwickling and $\theta_p$ is the corresponding apparent contact angle, as depicted in Fig. 2(a).

For the complete wetting state of the liquid film, the energy $E_f$ can be expressed as

$$E_f = [(r - f)(\gamma_{SL} - \gamma_{SV}) + (1 - f)\gamma] S_{film},$$

(2)

where the area $S'_{film} = V_0 (S_{down} + S_{film})/(V_0 - V_{CP})$ in which $V_0$ is the initial volume of the droplet and $V_{CP}$ represents the liquid volume above the top of the pillars for the droplet in hemiwickling, as depicted in Fig. 2(a). Substituting $S'_{film}$, $S_{up}$, and $S_{down}$ into Eqs. (1) and (2), we obtain

$$E_f - E_{CP} = \cos \theta_p \sin^2 \theta_p + 2 \cos \theta_p - 2 + E,$$

(3)

in which $E = V_{CP}/(V_0 - V_{CP})(S_{down} + S_{film})(r - f)(\gamma_{SL} - \gamma_{SV}) + (1 - f)\gamma$. Note that $\cos \theta_C > \cos \theta_Y$ is $\cos \theta_Y = (1 - f)(r - f)$ so that $E < 0$ and $E_f - E_{CP}$ always possesses a negative value [Fig. 2(c)], indicating that the liquid film state always has a lower energy than
hemiwicking. Therefore, from energy’s point of view, the final state of a droplet deposited onto textured hydrophilic substrates should be in either the Wenzel or the liquid film state, depending on $\cos \theta_Y > \cos \theta_C$ or $\cos \theta_Y < \cos \theta_C$. More importantly, hemiwicking never reaches neither the global minimum energy nor the local minimal, as shown in Fig. 2(c). In other words, hemiwicking is energetically unstable. This point is illustrated by Figs. 2(b) and 2(c), which are derived from the abovementioned energy analysis.

Thus, the energy analysis raises a fundamental question: how is hemiwicking stable while theoretical analysis shows that the special state never reached a minimum energy when compared to the Wenzel or the film states? We performed lattice Boltzmann (LB) simulations to answer this question.

**LB simulations prove that hemiwicking is stabilized by the pinning of the imbibition front**

We first simulated droplet spreading on textured substrates that were decorated with regularly distributed pillars with a height of $H = 7$ on a certain region. Depending on the pillar spacing, $S$, different final wetting states were observed: the Wenzel state and the liquid film state [Figs. 3(a) and 3(b)], which were in agreement with the above theoretical analysis.

Here, we noticed that hemiwicking was not observed under this condition. According to the theory in our paper, hemiwicking should be observed when the condition $\cos \theta_Y > \cos \theta_C$ and $\theta_Y > \theta_C$ is satisfied, in which $f = W^2/(W + S)^2$ and $r = ((W + S)^2 + 4HW)/(W + S)^2$. For the condition of $H7S5$, $\theta_C = 56.25^\circ$ ($f = 0.028$, $r = 1.778$). We can conclude that the Wenzel state should be observed in the case of $H7S5$ due to $\cos \theta_Y < \cos \theta_C$ ($\theta_Y = 62.2^\circ$), which agrees with our simulation result [Fig. 3(a)]. We obtain $\theta_C = 62.51^\circ$ ($f = 0.04$, $r = 2.12$) for the condition of $H7S4$, which leads to $\cos \theta_Y$ being almost equal to $\cos \theta_C$. From the theoretical analysis, this condition is on the boundary between the Wenzel state and hemiwicking or the film state. We calculated the value of $\theta_C = 60.26^\circ$ for $H7S4$. Hence, we should observe the Wenzel state or hemiwicking for $H7S4$ ($\theta_Y > \theta_C$). However, the film state is observed from our simulation results [Fig. 3(a)], which means errors exist in our simulation. We suggest that the errors originated from the simulation method, which is hardly consistent with the theory.

However, if we fixed $S = 3$ but increased $H$ from 1 to 5, the transition of wetting states is different [Figs. 3(c) and 3(d)]: the final state of the deposited droplets changed from the Wenzel to film state. More interestingly, a wetting state for the deposited droplet, called hemiwicking, was observed between the liquid film state and the Wenzel state [see, e.g., the case of $H3S3$ in Fig. 3(d)]. For this wetting state, the droplet takes a hat-like shape with a liquid film surrounding it. Thus, our results provide direct computer simulation evidencing of hemiwicking. The simulated results also indicate that the stability of the wetting states is strongly related to the height and spacing of the pillared array (roughness), which was consistent with the experimental results.

We then performed a systematic simulation study of the effect of substrate roughness, and a phase diagram for the corresponding final wetting states is given in Fig. 4(a). In general, the Wenzel state is observed at a substrate with weak roughness, i.e., large pillar spacing and a short pillar height. In contrast, the liquid film forms on the substrate with enhanced roughness, featured with small pillar spacing and high pillar height. Hemiwicking was observed in several cases corresponding to different $H$ and $S$, always at the boundary between the film and Wenzel states [see the phase diagram shown in Fig. 4(a)]. However, we must point out that the appearance of hemiwicking from LB simulations is inconsistent with the energy
A detailed inspection on the formation of the film state indicates that, if and only if the advancing contact line of the penetrating liquid for reaching the Young contact angle of the bottom surfaces can arrive at the neighboring pillar, or geometrically $\theta_Y < \theta_G$ with $\theta_G = \arccot(S/H)$ [Fig. 5(a)], film spreading continues until complete formation of the film state, which is similar to the experimental observation. Otherwise, the contact line motion during spreading stops by the constraint of the Young contact angle. This kind of contact line pinning caused by $\theta_Y > \theta_G$ is called the geometry pinning effect [Fig. 5(a)], which is an essential component to achieve hemiwicking. Whether the advancing film is pinned or not during droplet spreading decides if the final state is hemiwicking or a liquid film. We thus proposed that this kind of pinning effect of imbibition is responsible to the stability of hemiwicking. Note that mechanisms of contact line pinning on regular arranged pillars are complex and are discussed experimentally and theoretically. In our case, we found that the interface can be pinned on the top or side edges of the pillars [Fig. 5(a)], which is in agreement with the previous studies.

Therefore, we have, in principle, three different final states for droplet spreading on textured hydrophilic surfaces. The Wenzel state will be present when $\cos \theta_Y < \cos \theta_C$. For cases with $\cos \theta_Y > \cos \theta_C$, either the film state or hemiwicking could be obtained, depending on whether the pinning of the imbibition front exists or not.

According to this point, we added the geometry pinning effect [$\theta_Y > \arccot(S/H)$] into the energy analysis and thus obtained a new
phase diagram of wetting states [Fig. 4(b)], in which hemiwicking appears now. Note that hemiwicking is still not energetically favorable, but it is the pinning effect that stabilizes it. Then, we performed extensive simulations, and the obtained phase diagram, shown in Fig. 4(b), is used for comparison with theoretical analysis. In general, the wetting states from both energy analysis and LB simulations are in good agreement. This agreement again confirms that the contact line pinning effect is responsible for the appearance of hemiwicking. Importantly, all hemiwicking appears at the boundary separating the regime of the Wenzel state and that of the film state, as shown in the phase diagram of Fig. 4(b).

In the present work, we also show that there exists another situation that causes contact line pinning. When a flat substrate is coated only with a small patch of roughness [Fig. 5(b)], the boundary of the rough patch provides a strong pinning effect that is called the boundary pinning effect. Different from the geometry pinning effect, the boundary pinning effect induces hemiwicking much more effectively. In fact, if we added boundary pinning into energy analysis, all the stable film states, as shown in Fig. 4(b), would change into hemiwicking.

For the case with boundary pinning, however, the final state that is either hemiwicking or liquid film is also found to be related to the droplet volume. As shown from our simulation results [Fig. 5(c)], hemiwicking caused by the boundary pinning effect would disappear if we enlarge the patch size of roughness or decrease the droplet size: the hemiwicking would turn to the liquid film state immediately, as indicated in Fig. 5(c). In both cases, all liquid was used up to wet the roughness, without any liquid left for forming the droplet. Note that the geometry pinning effect and boundary pinning effect show different dependence on the droplet size. If the geometry pinning effect exists, a high energy barrier stops the liquid from advancing and stabilizes hemiwicking, showing a weaker volume dependence when compared to the boundary pinning effect. When the boundary pinning effect takes effect, either the film state or hemiwicking appears, depending strongly on the droplet volume: for spreading of a large droplet, the liquid left over after wetting the roughness forms a droplet in a caplike shape, sitting on the film. However, for small droplets, all the liquid is used up to wet the roughness, forming the liquid film state. This situation is, in principle, the same as that which exhibits no pinning effect, for which the liquid film state is formed.

More importantly, in the case of boundary pinning, a relatively large droplet would always develop into hemiwicking for a variety of patch sizes for substrate roughness. This mechanism is also shown in Fig. 6(c). The observation indicates again that hemiwicking is not stabilized energetically, but it is the strong pinning effect that makes it happen.

**Contact angle for droplets in the hemiwicking**

We also measured the evolution of the contact angle of droplets spreading on the surface coated with an H3S3 pillar array, the substrate that can generate hemiwicking (Fig. 3). In general, after a droplet reaches its stable state, the apparent contact angle from simulations roughly agrees with the theoretical value from the Cassie penetrating equation,
\[ \cos \theta = 1 - (f - f \cos \theta_f), \]
as demonstrated in Fig. 6(a). We also changed the size of the patch decorated with H3S3 pillars to obtain a different hemiwicking situation [Fig. 6(c)] and measured the corresponding contact angle from LB simulations [Fig. 6(b)]. It is found, however, that the contact angle for a different hemiwicking situation is not necessarily the same.
In fact, there exist two contact lines for a droplet in hemiwicking: one is for the liquid imbibition film and the other is for the caplike droplet. Figure 6(c) indicates that as long as the two contact lines are far from each other, the measured contact angle would finally reach a critical value, which agrees roughly with the theoretical prediction of the hemiwicking case. However, when the two contact lines are close to and affect each other, the deformed contact lines change the droplet shape, featured with a larger and anisotropic contact angle.

**The formation pathways for the hemiwicking**

Our simulations also show that there exist at least two distinct pathways for the formation of hemiwicking. The first pathway is droplet spreading, during which the contact line of the imbibition front is pinned by the roughness to induce the special wetting state. A typical example for the pathway is shown in Fig. 7(a) (on the substrate with an H3S3 pillar array). The spreading process can be described as follows. A spherical droplet was initially placed on the top of the structured substrate and spread until \( t = 11\,000 \). At \( t = 11\,000 \), the spreading stopped as the droplet front was pinned by the wetted pillars, forming a shape of an octagon. Note that no liquid film emerged during \( t = 0\)–\( 11\,000 \). The droplet as a reservoir would progressively shrink in order to form a liquid film surrounding the droplet. When geometry pinning took effect, the film growth stopped, with completely forming the hemiwicking.

The second pathway to form hemiwicking is via evaporating droplets initially in the Wenzel state [Fig. 7(b) with H4S3 pillars]. First, the front of a droplet in the Wenzel state was pinned by the pillars (60 × 60) in the boundary region and then evaporation started. Hemiwicking was formed after a period of time of liquid evaporation, with the front of droplet receding while maintaining the liquid film. Since most liquids evaporate easily at normal temperature and pressure, we think this path would frequently appear in reality.

In the pathway of droplet evaporating, hemiwicking comes initially from the Wenzel state, as a result of the geometry pinning effect. In other words, for the pathway of droplet evaporating, it is the Wenzel states [see, e.g., Fig. 4(b)] that would change into hemiwicking with the help of contact line pinning. This is different from the pathway of liquid spreading, in which the pinning effect changes the stable film states into hemiwicking, as demonstrated in Fig. 4(b).

**Hemiwicking on substrates with random distributed roughness**

In reality, physical roughness or chemical heterogeneity is not always regularly distributed on substrates as in the aforementioned models. To mimic such real surfaces on which physical roughness is not periodically ordered, we generated several realizations of rough substrates with randomly distributed pillars. There are a total of 2500 pillars of H3S3 or H4S3 that were randomly distributed on the
substrate. Note that the same number of pillars on the substrates of the same area was employed as in the uniform distribution discussed above, which ensures the same roughness factor for comparison. The simulation results show that hemiwick [Fig. 8(a)] and the film state [Fig. 8(b)] were obtained for $H3S3$ and $H4S3$ substrates, respectively, which is in good agreement with uniform distribution. Again, it is the contact line pinning that causes the appearance of hemiwick. This observation not only confirms the main conclusions of this work but also implies that hemiwick would be a common wetting state.

**CONCLUSIONS**

In the present work, we employed theoretical analysis and 3D lattice Boltzmann simulations to study the wetting states for droplets on hydrophilic textured substrates. We performed energy analysis to show that hemiwick is energetically unfavorable and has not even reached local or global energy minimal. In other words, hemiwick is energetically unstable.

Then, our LB simulations on the wetting states of droplets demonstrated that there indeed exists hemiwick for droplets on textured substrates with uniformly or randomly distributed pillars. For this kind of a wetting state, droplets take a hat-like shape with a liquid film surrounding them. This wetting state is found to follow the properties of the hemiwick model, and thus, our results provide direct computer simulation evidence of hemiwick. More importantly, we demonstrated that the special wetting state is thermodynamically unfavorable but in fact dynamically trapped by pinning of the imbibition front during the liquid film invading the substrate texture. For the contact angle of the droplet in hemiwick, strong deviation of the apparent contact angle from the hemiwick model is observed when the contact line of the liquid imbibition film is close to the spherical caplike droplet.
For formation of the special wetting state, our simulations show that hemiwicking is always found to emerge near the phase boundary between the liquid film and the Wenzel state. We also present two different routes to obtain hemiwicking from the regime in which either the liquid film or Wenzel state is thermodynamically stable in the absence of the pinning effect. Since liquid evaporating and droplet spreading occur commonly at normal temperature and pressure, different kinetic pathways of generating hemiwicking imply that this special wetting state could be easily found in nature.

**SUPPLEMENTARY MATERIAL**

See the supplementary material for details of the lattice Boltzmann method.

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