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Size matching effect on Wenzel wetting on fractal surfaces

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ABSTRACT

Materials with fractal structures may show distinct features of wetting. Here we report the characteristics of wetting of liquid caps on fractal surfaces in the Wenzel wetting regime. It shows that the relationship between the apparent and Young's contact angles is associated with the size-matching degree or discrepancy between the liquid caps and substrate. An explicitly fractal-dimension-and size-related equation is developed to bridge over the apparent contact angle and the Young's contact angle, and validated by the experimental results of water wetting on nanostructured fractal surfaces reported in the literature. The features of the Wenzel wetting of liquid caps on fractal surfaces are also discussed. This work enables us to understand the wetting regimes on fractal surfaces, and may pave a path for the rational uses of fractal structures to control liquid wetting.

Introduction

Understanding the wetting mechanisms of liquid drops on solid substrates is crucial to address technological and scientific issues in surface, material, chemical, electrical, bio-and civil engineering. To date, a large number of experimental, theoretical and computational studies have been conducted to advance the knowledges of wetting on solid surfaces with discovering the underlined characteristics and mechanisms step by step [1–5]. However, there remain having gaps between experimental observations and theories for the wetting of liquids on extremely complex substrates structured, for example, in the pattern of self-similarity (fractal structure) [6–13].

Fractals are objects that are structured in the patterns of self-similarity at every scale [14] or over a range of length scales [15]. Those structures can be characterised by a noninteger value, named fractal dimension D [14]. For the materials with fractal structures, the wetting behaviors may be significantly different from those with simple and ordinary-ordered structures (even being rough). For example, the researchers from the "Kao" institute reported sigmoid plots of cosine of the apparent contact angle between liquid drops and fractal surfaces to that of the Young's contact angle [11,12]. Those behaviors can not be captured by any single wetting model. Experimental results about liquid wetting on multi-fractal hierarchical single-walled carbon nanotube films [16] suggested that the multi-fractal hierarchical structure may be a switch to change the hydrophility or hydrophobility of materials. Davis et al. [10] found that the apparent contact angles of water drops on a flat surface in 66.8° suddenly decrease to zero for those on a fractal structure. The authors failed to explain the observations by the classic Wenzel wetting regime [17]. It thus raises a key question that needs to be answered: how a fractal structure influences the wettability of a surface? This question may have been partially answered by various thermodynamic analyses on complete and/or partial wetting of liquids on 2D or 3D multi-scale hierarchical structures [18–24]. However, those models are generally very complex and only involve some specific surface structures, such as, the hierarchically pillared surfaces [18,19,21,22]. Therefore finding a simple and easy-to-use model accounting for the fractal effect with generally scientific significances and broad applications remains a challenging task. Indeed, recent work by Li et al. [20] has briefly shown a function that describes the relationship between apparent contact angle and Young's contact angle, which is only a specific case of wetting on fractal surfaces. Furthermore, due to the limited fractal scales of real materials and the size of liquid drops, it also generates another question: do the size-matching degrees between the fractal regions and drop volumes play a part? The answer is still remaining to be explored. A recent theoretical study [25] suggested that the superhydrophobicity of randomly rough substrates are intimately related to the size-dependent Wenzel roughness parameter. As to the fractal surfaces, the wetting mechanisms and the influential factors remain to be advanced.

In the present study, we investigate the wetting behaviors of fractal surfaces in the physical constraints of the Wenzel wetting model. We focus on the scale (or size) consistency or discrepancy between liquid caps and fractal surfaces, which is, however, often undervalued, but possibly influences the hydrophilicity or hydrophobicity of the surfaces and the relationships between apparent and Young's contact angles [25]. This work may help us to clarify the physical regimes accounting

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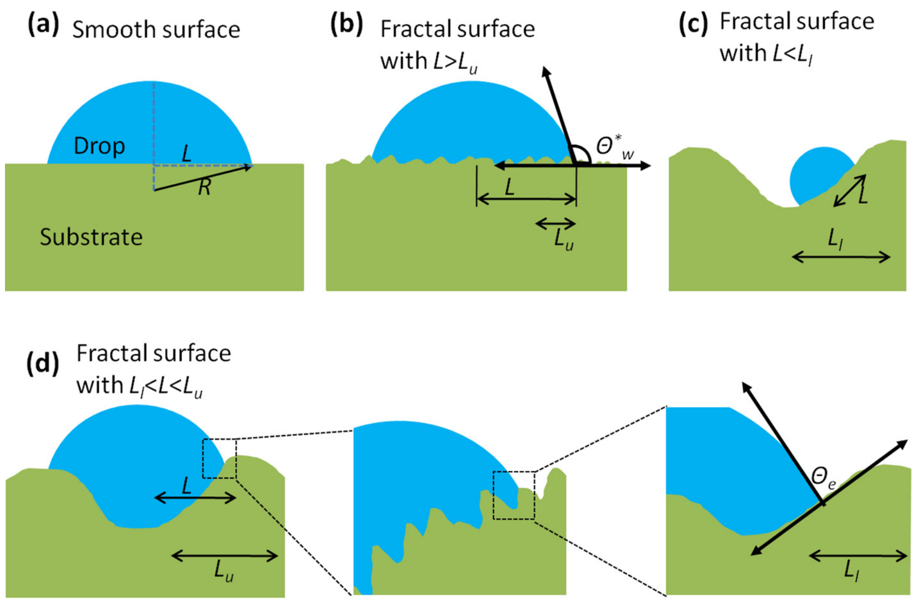

Fig. 1. Schematic illustration of scale-associated Wenzel wetting on different surfaces: (a) a drop being contact with a smooth surface without the size-matching effect; (b) a drop being contact with a fractal surface with the characteristic size of the drop larger than the upper bound of the fractal region, $L > L_u$; (c) a drop being contact with a fractal surface with the characteristic size smaller than the lower bound of the fractal region, $L < L_l$; and (d) a drop being contact with a fractal surface with the characteristic size in the fractal region, $L_l < L < L_u$.

for the anomalous experimental observations reported in the literature with better understandings of the size-matching effect on the Wenzel wetting behaviors of fractal surfaces.

Size-dependent wetting model

Generally, the wetting of a surface by a liquid phase can be governed by the Wenzel, the Cassie-Baxter and the mixed Wenzel-Cassie-Baxter regimes in company with or without a dynamic impregnation process [5,17,27,28]. For a rough surface, although the Cassie-Baxter state may be more robust [29], the steady Wenzel state of the wetting between the liquid drops and the rough surfaces is frequently observed [10]. Within the Wenzel wetting regime, a liquid phase impregnates the interstices and surfaces of a rough substrate with complete contact (Fig. 1), which is the most distinct feature from the Cassie-Baxter wetting regime. So, the apparent contact angle between the liquid and the rough surface in the Wenzel regime (θ_w^*) is linked to the equilibrium contact angle (θ_e, or the Young's contact angle) via the Wenzel equation [17],

$$\cos\theta_w^* = \lambda\cos\theta_e \tag{1}$$

where λ is a surface roughness factor, the ratio of the actual area of the rough surface in contact with the liquid to the geometric projected area.

In general, $\lambda \geqslant 1$ for any real rough surface. This indicates that increasing the roughness of a surface can enhance its hydrophilicity or hydrophobicity consistently. This functional enhancement may be promoted by the surfaces in multi-hierarchical structures (or fractal structures) [6,7,9,11,12,19,29]. Based on a series of experimental studies, the researches in the "Kao" institute [11,12] proposed a Wenzel-type model to relate the apparent contact angle between liquids and fractal substrates to the Young's contact angle, which gives,

$$\cos\theta_w^* = \left(\frac{L_u}{L_l}\right)^{(D-2)}\cos\theta_e \tag{2}$$

where L_u and L_l are the upper and lower bounds of the fractal measurement, D is the fractal dimension. Typical scaling range of fractal regions spans only 0.5–2 decades, $L_u/L_l \in (10^{0.5}, 10^2)$ [15]. This model can roughly catch the data when $\cos\theta_e \in (-0.5, 0.5)$ [5,11,12], and the surface roughness factor remains constant, which means the physical bases of Eq. (2) are not beyond those of Eq. (1).

Now let us consider more general cases of the Wenzel wetting of liquids on fractal surfaces:

- For a plate and smooth surface, the roughness factor is equal to unit, and thus the apparent contact angle is identical to the Young's contact angle, $\cos\theta_w^* = \cos\theta_e$; see Fig. 1(a). Here the possible effects of contact line and deformation of substrate are not considered [30].
- For a fractal surface with the upper bound L_u lower than L (defined as the characteristic size of the (projected) contact area between the liquid cap and the fractal surface; see Fig. 1(b)), the roughness factor is identical to Eq. (2) developed in Refs. [11,12]. This is practical because the size of the water drops used in Refs. [11,12] (in millimetre) is larger than the upper bound of the fractal surfaces (in micrometer) by around three orders of magnitude.
- If the characteristic length of the contact area L, by contrast, is even smaller than the lower bound of the fractal surface L_l, the roughness factor reduces to 1 and the surface is identical to the smooth one; see Fig. 1(c). This means that the roughness loses its function to impact the apparent contact angle. It is noteworthy that this case can be rarely observed by experiments because the lower bounds of fractal surfaces are generally in nano scales [16,31–33] and the liquid drops are in micro scales or above [10–12]. Note that in such thin scales it is unclear whether or not the Wenzel wetting is applicable. However, the special case may be favoured for sorption on a virtual surface or heterogeneous nucleation on a fractal surface as nuclei are generally very small [34–36].
- For the case where the characteristic length of the contact area L is between the upper and lower bounds of the fractal surface ($L_l < L < L_u$; see Fig. 1(d)), the roughness factor is directly associated with L and D. The relationship between the apparent contact angle and the Young's contact angle can be expressed by,

$$\cos\theta_w^* = \left(\frac{L}{L_l}\right)^{(D-2)}\cos\theta_e, \quad L_l < L < L_u \tag{3}$$

Eq. (3) remains following the same form of Eq. (2), but shows its own characteristics; see Section "Result and discussion" for detailed discussion.