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# Evolution of the interfacial shape in dissolutive wetting: Coupling of wetting and dissolution

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ABSTRACT

Interface evolution of a droplet on a dissolvable substrate is controlled by coupling of wetting and dissolution, which makes dissolutive wetting more complicated than wetting on inert substrate. In our experiments, xylene and polystyrene are used as solvent and solute, respectively. We record the continuous evolution of interfacial shape and measure change of geometrical parameters. Then, a model is built based on the Noyes-Whitney equation. The evolution mechanisms of interface and solute concentration under the combined actions of wetting and dissolution are analyzed. There are two stages in dissolutive wetting. The two stages are identified by the speed ratio of wetting and dissolution, which reflects the coupling between wetting and dissolution. The model can recover the evolution of the interfacial shape observed in the experiment. Our work may help to predict change of interfacial shape in dissolutive wetting, and the results provide a better understanding of dissolutive wetting which is a process that occurs in many important application areas.

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## 1. Introduction

Dissolutive wetting is a process in which a liquid wets and dissolves the substrate. It is a basic of reactive wetting. As for reactive wetting, the liquid have chemical reaction with the substrate by dissolution or producing reaction products at interface. The resulting reaction will strongly change the properties of interface, especially the contact line [1]. The early of technological interest in reactive wetting is focused on applications in joining processes between metals and ceramics [1,2]. However, dissolutive wetting is also ubiquitous and plays an important role in the fields of protection of optical components [3], digestion of fat [4] and steroids [5], printing [6], micro/nano-fluidics [7], fabrication of alloy nanowire [8,9], storage of carbon dioxide [10], and etc. In some applications, dissolutive wetting needs to be avoided. Even if some materials are durable such as Borosilicate glasses, there also exists very slow dissolutive wetting [11]. In some cases, dissolutive wetting is a much sought-after process. For example, etching that fabricates microstructures using silicon has been broadly applied in MEMS [12]. The design of drug also needs to take account of dissolutive wetting [13–16]. Predictions and control of the interfacial shape of dissolutive wetting in these applications are desirable. Not least motivated by these applications, the mechanisms that wetting interacts with dissolution come to attention of both chemistry and physics [17].

Compare with non-reactive wetting [18], dissolutive wetting involves many complex chemical and physical processes such as wetting, diffusion, and convection, which change components and their distributions in solvent. These processes change properties of solvent. Because of the complexity of the combined actions of wetting and dissolution, our fundamental understandings of dissolutive wetting mechanism relatively lag behind. For the theoretical investigations of dissolutive wetting, as far as we are concerned, there are three methods. One is thermodynamic method [19] For example, Yost et al. [20] used the early work of Onsager to calculate the evolution of contact line and contact angle. However, it was difficult to describe change of solid-liquid interface. The second method is fluid mechanics method. Warrant et al. [21] assumed solute was transported by diffusion primarily. However, they did not consider the influence of surface tension. The model of Braun et al. [22] was built according to lubrication theory. However, their model was so complex that the shape of interface could not be represented by a function. Alleborn et al. [23]. built a model to describe the interfacial shape near triple phase point in dissolutive wetting. The third method combines

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the N-S equations and thermodynamic theory. For example, Villanueva et al. [24] presented a multicomponent and multiphase model, and their model agreed with the experimental data successfully. However, the droplet was supposed to be thin and incompressible. In the above models, the surface tension of liquid was assumed as constant or brief function, and another important assumption was that the upper is spherical shape.

Pioneering studies also used simulated methods [25,26] to calculate the evolution of boundaries especially the Arbitrary Lagrangian-Eulerian (ALE) method. This method have outstanding capability to deal with large deformation and capture the interface [27]. In experimental study, the Cu-Si system and Bi-Sn system were used commonly. Protsenko et al. [28] conducted experiments on Cu-Si system to reveal the relation between weak concentration gradient and solid-liquid interface angle. Yin et al. [29] observed the initial concentration of liquid determined the speed of wetting. Previous experiments were conducted using opaque materials, so there were difficult to continuously record the process of interfacial evolution.

In this paper, we employed both experiment and theory to study the evolution of the interfacial shape in dissolutive wetting. In our studies, we use transparent xylene and polystyrene for experiments, measure the dissolutive wetting of small droplets on a dissolvable substrate, and discover the radius of droplet increases as $t^{0.11}$. Based on Noyes–Whitney equation, we develop a model to describe change of interface and concentration of droplet. Our model can reproduce the results of experiment. Base on this model, we analyze the competition speeds of wetting between dissolution, which decides the dissolutive wetting in two stages. We hope that our theoretical solution for the wetting liquid dissolves the substrate will shed light on the coupling of wetting and dissolution in dissolutive wetting. Our work can be further applied to the design of products of MEMS and medicine field, and to the protection of key components in equipment.

## 2. Experiment procedures

### 2.1. Dissolutive wetting experiments

In the experiments, polystyrene and xylene are chosen to be solute and solvent, respectively. The polystyrene substrates are polished and cleaned by ultrasonic cleaning. Fluorescent agent is added to the xylene according to mass ratio of 800:1 to enhance the clarity of interface. The fluorescent agent will become green under the excitation of white light. The experimental process of dissolutive wetting is observed and recorded by using dark field mode of a microscope (KH-8700, Hirox). The relative influences of gravity and surface tension in spreading are measured by the Bond number $\mathrm{Bo} = \rho g R_0^2/\gamma$, where $\rho$ is the density of the liquid, $g$ is the gravitational acceleration, $\gamma$ is the surface tension and $R_0$ is initial radius of droplet. When the initial radius is much less than 1.87 mm, effect of gravity on the shape of the droplet can be ignored. In order to rule out the effects of gravity, a droplet with the volume of 0.7 ± 0.015 μl whose radius is about 0.54 ± 0.15 mm is produced by microsyringe and deposited on the polystyrene surface using a micropipette. During the experiments, the temperature always maintains at 23 °C, and the pressure 101.3 kPa. In standard condition, we obtain the average rate of evaporation of xylene is $1.1\times10^{-3}$ g·min$^{-1}$·m$^{-2}$ by experiments. The mass loss ($3 \times 10^{-9}$ g/min) result from evaporation is much less than the total liquid mass ($5.46 \times 10^{-4}$ g). Therefore, the influence of xylene's evaporation to droplet is negligible. Other data about the properties of solute and solvent is listed in the Table 1. The experimental pictures are analyzed by software (SCA202 V.3.61.4, DataPhysics). The data of geometrical parameters is the average five experimental results.

**Table 1**
The properties of solute and solvent.

| Solvent | Density (kg·m$^{-3}$) | Viscosity ($10^{-4}$ Pa·s) | Surface tension ($10^{-2}$ N·m$^{-1}$) | Rate of evaporation ($10^{-3}$ g·min$^{-1}$·m$^{-2}$) |
|---|---|---|---|---|
| Xylene | 840 | 7.5 | 3.0 | 1.1 ± 0.5 |

| Solute | Density (kg·m$^{-3}$) | Diffusion coefficient in xylene ($10^{-10}$ m$^2$·s$^{-1}$) |
|---|---|---|
| Polystyrene | 1050 | 6.05 |

### 2.2. Verification test

The relative influences of convection and diffusion in flow field are measured by the Pelect number $Pe = vL/D$, where $v$ is characteristic velocity, $L$ is characteristic length, $D$ is the diffusion coefficient. In this case, $Pe \sim 10^{-2}$. According to the Pelect number, we cannot ruled out the influence of convection completely. So we use a concise and district method to determine the influence of convection to mass transfer in our case, which compare the dissolution process of any point (point A as an example) that near the origin and far away from the contact line (see Fig. 3a) with that of the verification test (see Fig. 1d).

The verification test is a 1D dissolution which can ignore the influence of wetting and convection. The test device is a thin glass tube, and the lower part of glass tube is filled with polystyrene, and the xylene is poured into tube from the upper part of glass

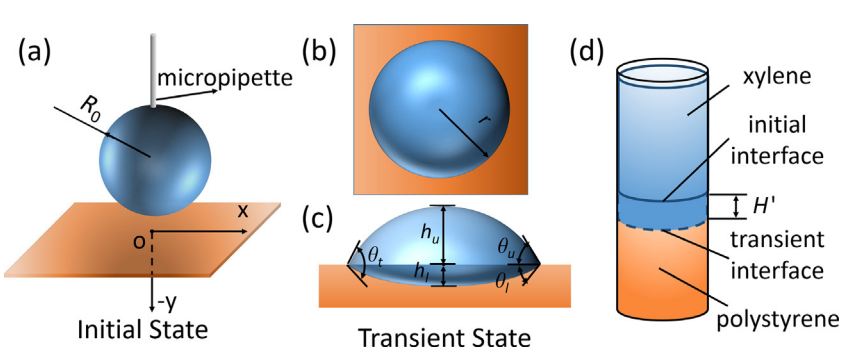


**Fig. 1.** Illustrations of (a–c) dissolutive wetting, and (d) verification tests. Dissolutive wetting: (a) at the initial state, a xylene droplet (blue) with radius of $R_0$ is placed on the surface of polystyrene (orange). (b) Top and (c) side views of transient state in dissolutive wetting. The symbols $r$, $h$ and $\theta$ are the instant radius, height and contact angle, respectively. The subscript $u$, $l$ and $t$ represent upper, lower and total, respectively. Verification tests: (d) the bottom of glass tube is filled with the polystyrene (orange), and then the xylene (blue) is poured into tube from the top. Evolution of solid-liquid interface (the distance between initial interface and transient interface) is noted as $H'$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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