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Dynamics of Liquid Contact Line on Visco-Elastic Gels

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1. Introduction

Static and dynamics of wetting is still an active subject of research even on "hard" solid surfaces because of the need to patch up classical hydrodynamics near the contact line. Recently, the focus is directed to the complex situations in which the substrate is not an "ideal" solid, e.g., wetting with the substrate deformation (softness) [1-5] or with the volume exchange between the liquid and substrate (permeability) [6,7]. As a model system of such complex situations, we are focusing on the wetting on gels.

In the previous ECS2011 symposium, we have reported the experiment regarding the spreading and diffusing processes of water droplets on PAMPS-PAAM hydrogels in which we have observed a strong link between the swelling of gel and the transitions of wetting regimes [8,9]. In this conference, we are turning our attention to a different aspect of the gel complexity, i.e., the rheology of gels. We studied the dynamics of water sessile droplets advancing on poly(styrene-butadiene-styrene)(SBS)–paraffin gel substrates. As the SBS–paraffin gel is hydrophobic and there is no volume exchange between the drop and gel, the sole possible effect affecting the contact line behavior is the gel surface deformation due to the capillary force of liquid [10] moderated by the viscoelastic response of the material.

Unlike the case of wetting on an elastomer, the droplet contact line exhibits quite complex behaviors, i.e., the contact line shows two regimes of continuous advancing motion and one regime of stick-slip motion depending on the inflation rate and droplet radius. The stick-slip motion of the contact line was previously observed by Pu et al. during the wetting on thin polymer films [11]. Here, we found that on a SBS–paraffin gel, there are three different regimes, i.e., continuous, stick-slip, and another continuous motion [12]. We will discuss how the transitions of these contact line motions are characterized by the parameters such as the typical frequency of the contact line motion, and propose a possible mechanism. Finally, we will also present recent experiments of drying of colloidal suspension drops on the same gel, where the stick-slip motion is modified by particle deposition, which may be of importance for future processes of colloid deposition on soft surfaces.

2. Experimental Section

Poly(styrene-butadiene-styrene)(SBS)–paraffin gels were used for the gel substrates and distilled water (Milli-Q Integral; Millipore, USA) was used for the liquid. SBS powders (G1682; Kraton Polymers, USA) were dissolved in paraffin (Norpar15; ExxonMobil, USA) heated in a water bath at 90ºC. After SBS powders were completely dissolved, the solution was poured into a gel mold and was cooled down to ambient temperature. The gel mold consists of two glass plates separated by a rubber spacer. The dimension of gel samples were 70 mm in length, 20 mm in width and 3 mm in thickness. Gels of various SBS concentrations were prepared. The mass concentration of SBS was varied from c_{pol} = 8% to 25%. The rheology of the gel was measured by a strain controlled rheometer (Physica MCR 500; Anton Paar, Austria). Shear strain amplitude was set to 1%, and the experiments were conducted at frequencies from 10^{-4} Hz to10 Hz.

The inflation experiment of a water droplet on the gel was conducted as follows. A water droplet was placed on a gel by a micro-syringe. The syringe was connected to a motor syringe pump (Model 33; Harvard Apparatus, USA), and it supplies water to the droplet and inflate it at a constant volume rate q. From the side and top of the droplet, the shape of the droplet was monitored by 2 CCD cameras (Model A101fc and Model PLA1000; Basler, Germany) with magnification lenses (CCTV lens; Pentax, Japan). The same observation setup was used for the drying experiment of colloidal droplets. In the drying experiment, a droplet of volume

7 μl containing colloidal particles (Klebosol; d = 50 nm) was dried on a SBS-paraffin gel in a natural condition (T: 20ºC, H: 20%).

3. Result and Discussion

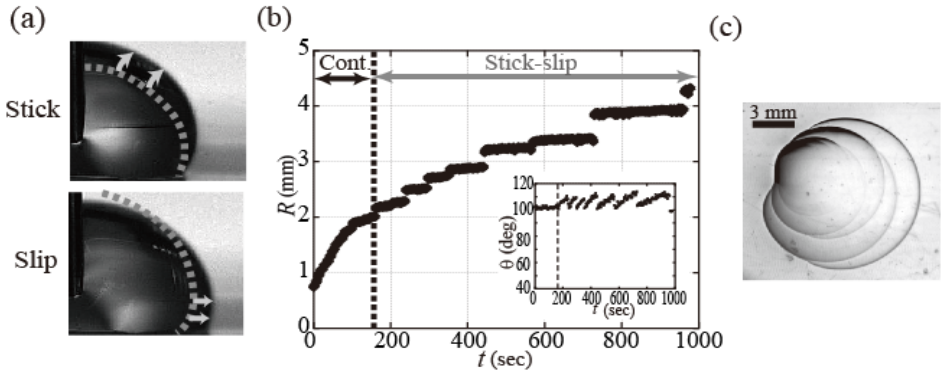


Fig.1 (a) Stick-slip behavior of the contact line observed in advancing droplet on SBS-paraffin gel of c_{pol} = 10% at an inflation rate q = 20 μl/min. (b) Plot of the radius and contact angle of the same droplet. (c) Multi-circular traces formed on the gel surface after the inflation experiment.

Figure 1 (a) shows sequential pictures and (b) shows the plot of radius and contact angle of a water droplet being inflated on a SBS–paraffin gel of c_{pol} = 10% at an inflation rate q = 20 μl/min. During the inflation process, the droplet contact line exhibits continuous and stick-slip motions. At an early stage of the inflation process (t < 160 s) while the droplet radius R is still sufficiently small (R < 2 mm), the contact line advances continuously at a nearly constant contact angle ~ 100º. As R becomes large at a later stage (t > 160 s, R > 2 mm), the contact line starts the stick-slip motion, i.e., the droplet radius R stays at the same value during a certain moment then suddenly increases. Looking at the contact angle, once the contact line sticks, θ starts to increase. As θ reaches a critical value ~ 100º, the contact line slips forward. Successively, the contact line repeats this stick-slip motion.

For a quantitative analysis, we conducted the inflating experiments at various volume rates q ranging from from 1 μl/min to 200 μl/min. Figure 2 summarizes the behavior of contact line at various R and q. (i) At high q (200 μl/min), the contact line advances continuously with a constant contact angle during the whole inflation process. (ii) At intermediate q (20 μl/min), the contact line initially exhibits the continuous advancing motion. As the droplet radius reaches a critical value (R ~ 2.2 mm), the contact line starts the stick-slip motion. (iii) At low q (2 μl/min), the contact line exhibits the stick-slip motion even at a very early stage of the inflation. However, when the droplet radius increases, now the contact line stops the stick-slip motion and starts to advance continuously again.

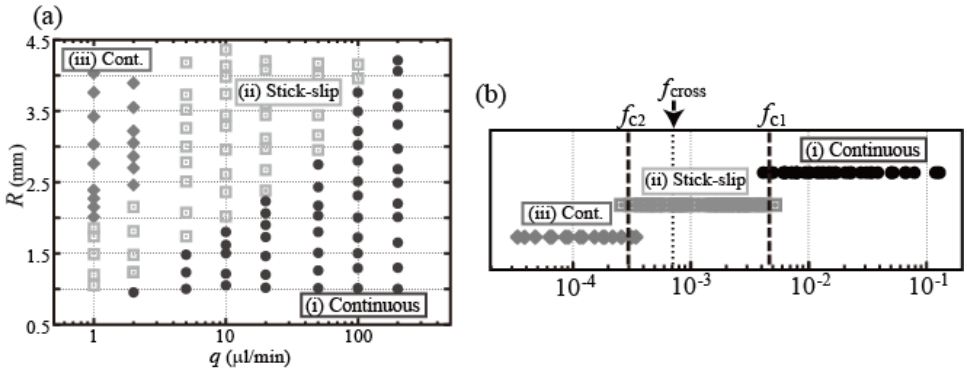


Fig.2 (a) Diagram of contact line behaviors as a function of R and q. (b) Replot of the data against the characteristic frequency f. The gel crossover frequency is also shown in the figure.

In order to compare the contact line behavior with the gel rheology, we further estimate the characteristic frequency of the contact line motion defined as:

$$f = \frac{v_a}{R} \approx \frac{q}{2\pi R^3} \quad (1),$$

where v_a is the apparent contact line velocity. In fig. 2 (b), we replot the diagram as a function of f. It is clearly observed that the three regimes of the contact line motions (i)–(iii) are separated by f, giving two critical frequencies which characterize the transition. From the comparison with the measurement of gel rheology, it is also observed that the crossover frequency f_{cross} where the storage and loss modulus correspond with each other $G''/G'=1$ is between these two critical frequencies. We have conjectured that the observed transitions of the contact line motions (continuous–stick-slip–continuous) are the consequence of the rheological property of the gel affecting the contact line dynamics. Depending on the frequency, the behavior of the contact line on gels shows both aspects of wetting on elastic solids and wetting on viscous liquid sheets. At an intermediate frequency close to f_{cross} where the gel behaves neither like a solid nor like a liquid, the stick-slip motion appears.

We also observed a slick-slip behavior, but now for a receding contact line in the drying process of colloidal suspension drops on SBS-paraffin gel. In the case of drying drops, the concentration of the colloidal particles largely affects the distance covered by contact line during the first jump. Figure 3 shows a plot of this distance d_{slip} normalized by the initial drop radius. At low particle concentration (0.001-0.1wt%), the droplet exhibits the larger slip with the increase of the concentration. At a high particle concentration (1-10wt%), the slip distance sharply decreases again. This result indicates that the addition of the colloidal particles increases the pinning force of the contact line; meanwhile it also contributes to the enhancement of the local dissipation near the slipping contact line.

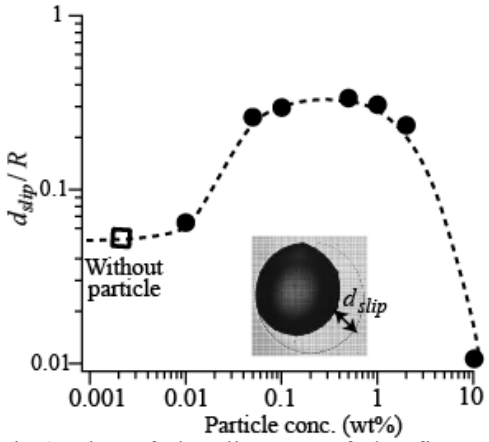


Fig.3 Plot of the distance of the first slip normalized by the drop radius.

4. Conclusion

Our results show that the dynamics of the contact line on visco-elastic gel substrates are quite different from those observed for general solid materials, especially for the transition between the continuous and stick-slip motions. The key factor here is that the capillary force at the contact line can make a local deformation on the gel surface, whose profile dynamically changes depending on the frequency of applied force. For the detailed analysis of the present phenomena, we have to analyze the precise profile of the gel deformation by solving the equation of the balance between the capillary force and visco-elastic resistance, which will be conducted as our future works.

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