

**Position-dependent rates of film growth in drying colloidal suspensions on tilted air-water interfaces**

Kohei Abe <sup>\*,a</sup> and Susumu Inasawa <sup>\*,a,b</sup>

<sup>a</sup>*Graduate School of Bio-Application and Systems Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Naka-Cho, Koganei, Tokyo, 184-8588, Japan.*

<sup>b</sup>*Department of Applied Physics and Chemical Engineering, Tokyo University of Agriculture and Technology, 2-24-16 Naka-Cho, Koganei, Tokyo, 184-8588, Japan.*

\*E-mail: [kohei.abe@oist.jp](mailto:kohei.abe@oist.jp) and [inasawa@cc.tuat.ac.jp](mailto:inasawa@cc.tuat.ac.jp)

Table S1 List of  $A$  for various conditions for which  $H$ ,  $T$ , suspension (particle diameter  $d$ ), or  $\varphi_0$  is changed.

For all of the drying experiments,  $W$  and  $\theta_0$  are equally set to be approximately 5 mm and 45°, respectively

	Suspension	$H$ [mm]	$T$ [°C]	$\varphi_0$ [-]	$A$ [-]
Standard condition	KE-W30	0.1	20	0.10	2.1
	KE-W30	0.05	20	0.10	2.1
	KE-W30	0.2	20	0.10	2.1
	KE-W30	0.1	5	0.10	2.7
	KE-W30	0.1	40	0.10	2.1
	Snowtex-OL	0.1	20	0.11	2.7
	KE-W10	0.1	20	0.09	2.1
	MP-2040	0.1	20	0.25	2.9
	KE-W30	0.1	20	0.033	2.9
	KE-W30	0.1	20	0.020	2.9

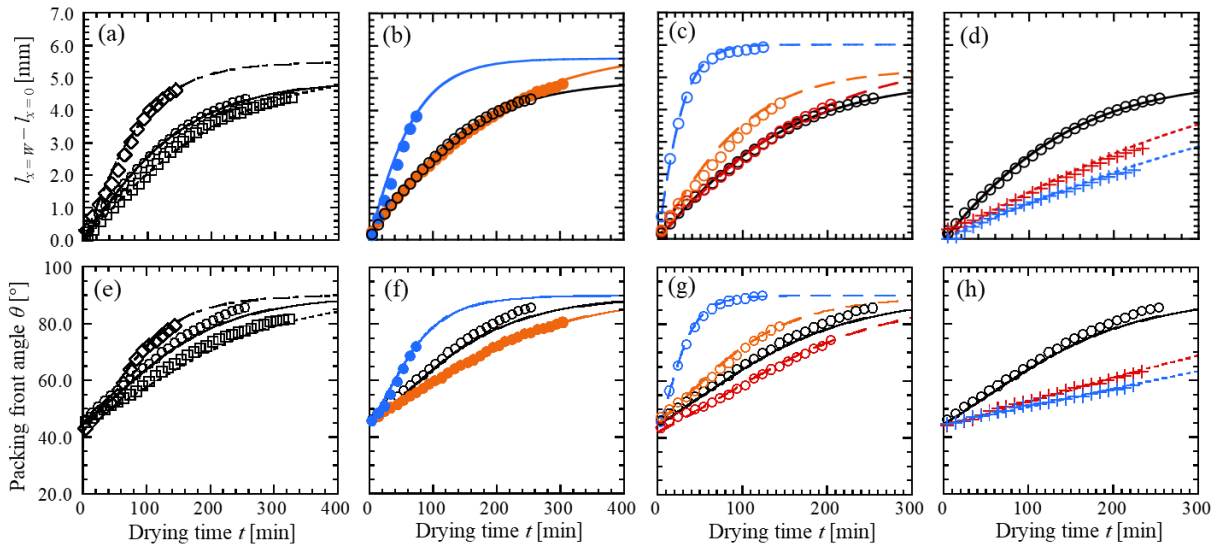


Figure S1 Time evolution of (a–d)  $l_x = w - l_{x=0}$  (e–h)  $\theta$  for a series of experiments listed in Table S1 whereas the lines show mathematical models calculated from Eq. (5) and (7). (a, d)  $H = 0.05$  mm (black open diamonds and black chain line) and  $0.2$  mm (black open squares and black dotted line). (b, f)  $T = 5$  °C (orange solid circles and orange solid line) and  $40$  °C (blue solid circles and blue solid line). (c, g)  $d = 62$  nm (Snowtex-OL, orange open circles and orange broken line),  $98$  nm (KE-W10, red open circles and red broken line) and  $2.9 \times 10^2$  nm (MP-2040, blue open circles and blue broken line). (d, h)  $\varphi_0 = 0.033$  (red plus signs and red dotted line) and  $0.020$  (blue plus signs and blue dotted line). Black open circles and black solid line in each figure show the experimental results and the model calculations at the standard condition ( $H = 0.1$  mm,  $T = 20$  °C,  $d = 3.3 \times 10^2$  nm and  $\varphi_0 = 0.10$ ). In the model calculation,  $A$  in Table S1 is used for each calculation.

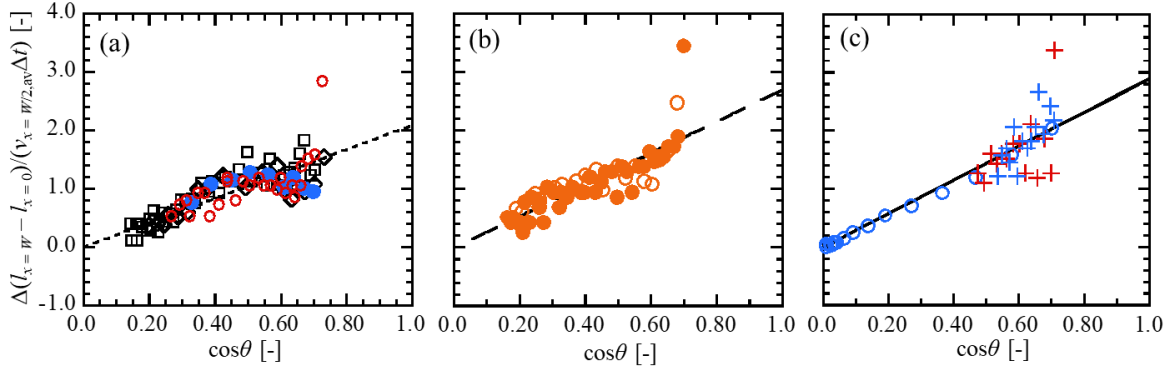


Figure S2 Normalized  $\Delta(l_{x=w} - l_{x=0})/\Delta t$  by  $v_{x=w/2,av}$  in terms of  $\cos\theta$  for the data in Fig. S1. Time interval  $\Delta t$  is set to be 16 min for the samples of  $\varphi_0 = 0.033$  and 0.020, and 8 min for the other samples. Each symbol corresponds to that in Fig. S1 and these data are summarized by different  $A$  values as listed in Table S1. (a) Samples of  $H = 0.05$  mm (black open diamonds),  $H = 0.2$  mm (black open squares),  $T = 40$  °C (blue solid circles) and  $d = 98$  nm (red open circles). (b) Samples of  $T = 5$  °C (orange solid circles) and  $d = 62$  nm (orange open circles). (c) Samples of  $d = 2.9 \times 10^2$  nm (blue open circles),  $\varphi_0 = 0.033$  (red plus sign) and  $\varphi_0 = 0.020$  (blue plus sign). Dotted, broken, and solid lines in (a), (b), and (c) correspond to Eq. (3) in which  $A$  is set to be 2.1, 2.7, and 2.9, respectively.

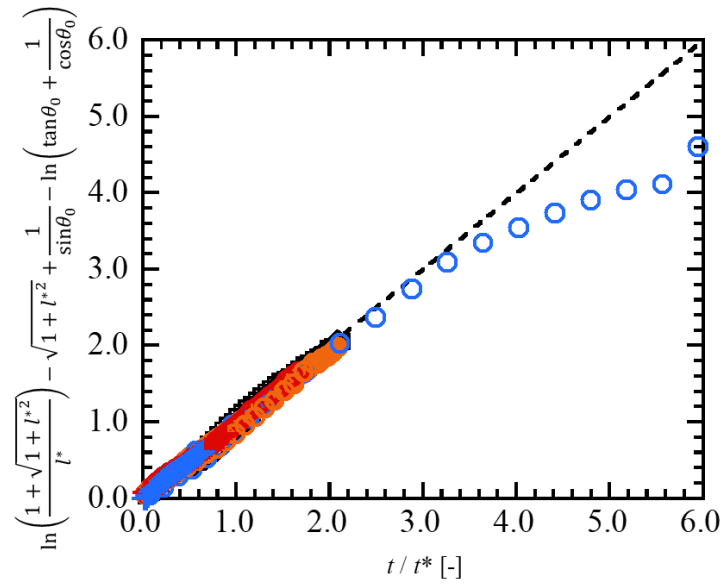


Figure S3 Correlation between the left-hand and right-hand sides in Eq. (5) calculated from the experimental data in Fig. S1(a-d). Dotted line indicates the theoretical line expressing Eq. (5). Each symbol corresponds to that in Fig. S1 and  $A$  is set for each drying experiment as listed in Table S1.

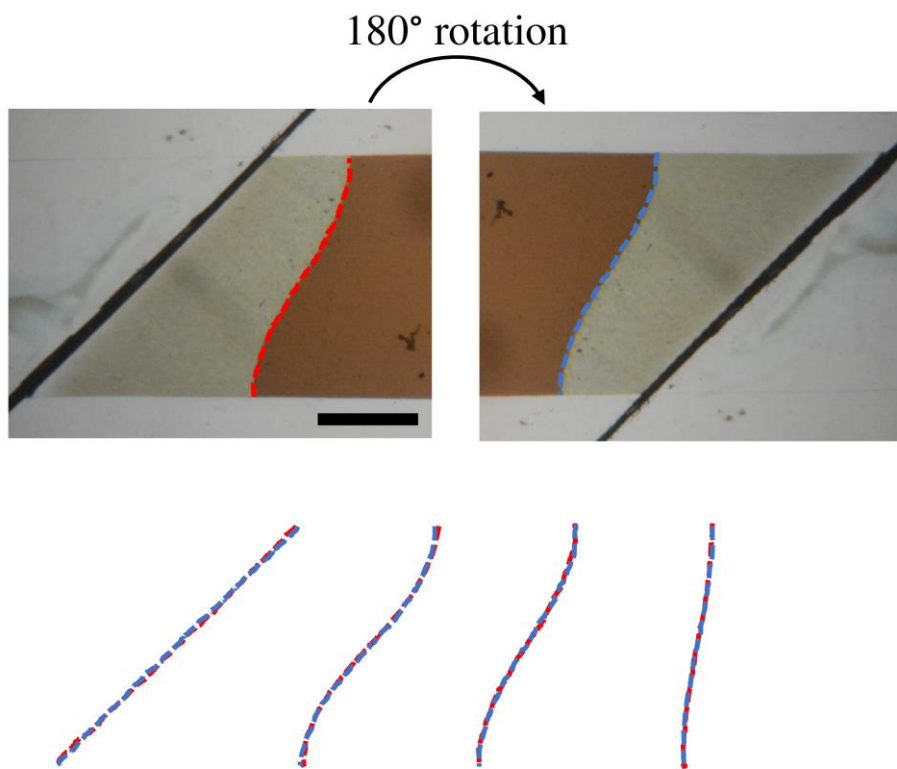


Figure S4 (Top) Shape of packing front at  $t = 120$  min before (red dotted line) and after rotation (blue dotted line) by  $180^\circ$ . The drying interface of the cell,  $\theta_0$ , was set as  $45^\circ$ . Scale bar shows 2 mm. (Bottom) Superimposition of the two dotted lines at  $t = 0, 80, 120, 240$  (Left to right).

### Movie S1

Growth of the packed film in the drying cell in which the drying interface is set to be perpendicular to spacers ( $\theta_0 = 92^\circ$ ). 0.1 s in the movie corresponds to 1 min in real observation.

### Movie S2

Growth of the packed film in the drying cell in which the drying interface is set to be tilted with an angle of  $45^\circ$  to the spacer ( $\theta_0 = 45^\circ$ ). 0.1 s in the movie corresponds to 1 min in real observation.