

Buoyancy driven dissolution of inclined blocks: Erosion rate and pattern formation

Supplemental material - Raw data

Caroline Cohen, Michael Berhanu, Julien Derr, and Sylvain Courrech du Pont*
Laboratoire Matières et Systèmes Complexes
Université de Paris, CNRS
Bâtiment Condorcet, 10 rue Alice Domon et Léonie Duquet,
75205 Paris cedex 13, France

We suppose a linear relationship between concentration C and density ρ of solutions :

$$C = \frac{\rho - \rho_0}{\rho_{\text{sat}} - \rho_0} C_{\text{sat}}, \quad (1)$$

where $\rho_0 = \rho(C = 0)$ and where ρ_{sat} and C_{sat} are the density and the saturation concentration, respectively.

For the calculations of the scaling laws for the experiments with salt, we use the measured values $\rho_0 = 997.12 \text{ kg.m}^{-3}$, $\rho_{\text{sat}} = 1197.3 \text{ kg.m}^{-3}$ and $\rho_s = 2348 \text{ kg.m}^{-3}$. We calculate $C_{\text{sat}} = 317.25 \text{ kg.m}^{-3}$ from the tabulated value of the solubility $S = 359.45 \text{ kg.m}^{-3}$ ($C_{\text{sat}} = S\rho_{\text{sat}}/(S + \rho_0)$). We use $D = 1.59 \times 10^{-9} \text{ m}^2.\text{s}^{-1}$ for the coefficient of diffusion of the saturated brine [1]. Figure 1 shows our measurements of the viscosity of salt solutions as a function of their density. Raw data of experiments are listed in table I. Figure 2 shows the dependency of the receding velocity on the inclination angle and the density of the outer bath. Figure 3 shows the dependency of the wavelength of incipient longitudinal stripes on the density of the outer bath.

Alkattan *et al.* measured the dissolution velocity set by the chemical kinetic of salt in water. They found $\alpha \simeq 5 \times 10^{-4} \text{ m/s}$ [2]. Thus, the characteristic thickness over which the dissolution is limited by diffusion is $D/\alpha \simeq 3.2 \times 10^{-6} \text{ mm}$. This is much smaller than the values of wavelengths we measure in our experiments, which justifies the diffusion limited approximation, *i.e.* to reduce the value of the concentration at the interface to the one of the saturation concentration.

For the experiment with plaster, the solution was tap water and the inclination was $\theta = 52 \text{ deg}$. We measured $\lambda_s = 2.25 \pm 0.4 \text{ mm}$ and $-\dot{h} = 2.97 \times 10^{-6} \pm 6.5 \times 10^{-7} \text{ mm/s}$. We use $D = 1 \times 10^{-9} \text{ m}^2.\text{s}^{-1}$ (from tabulations, [3]), $\rho_{\text{sat}} - \rho_0 = 1.8 \text{ kg.m}^{-3}$ (measured), $C_{\text{sat}} = 2.04 \text{ kg.m}^{-3}$ (tabulated), $\eta_f = 0.89 \times 10^{-3} \text{ Pa.s}$ (pure water), and $\rho_s = 1859 \text{ kg.m}^{-3}$ (measured) for the calculation of the scaling law.

Colombani and Bert measured the dissolution velocity set by the chemical kinetic of gypsum in water, they found $\alpha \simeq 2.7 \times 10^{-6} \text{ m/s}$ [3]. Thus, the characteristic thickness over which the dissolution is limited by diffusion is $D/\alpha \simeq 0.37 \text{ mm}$. It is (only) 6 times smaller than the wavelength value we measured, which barely justifies the dissolution limited approximation.

For the calculations of scaling laws for experiments with caramel, we use the measured values $\rho_0 = 997 \text{ kg.m}^{-3}$, $\rho_{\text{sat}} = 1450 \text{ kg.m}^{-3}$ and $\rho_s = 1540 \text{ kg.m}^{-3}$. $C_{\text{sat}} = 968.26 \text{ kg.m}^{-3}$ is calculated from the measured value of ρ_{sat} and the tabulated value of the sucrose solubility $S = 2004 \text{ kg.m}^{-3}$ ($C_{\text{sat}} = S\rho_{\text{sat}}/(S + \rho_0)$). Figure 4 shows our measurement of the viscosity of caramel solutions as a function of their density. We estimate the coefficient of diffusion of the saturated solution $D = 1.34 \times 10^{-12} \text{ m}^2/\text{s}$ from the tabulated value at zero concentration ($5.2 \times 10^{-8} \text{ m}^2/\text{s}$) that we extrapolate to the saturated solution using the Stokes-Einstein relation. The error may be large for this value. This would affect the comparison between experiments with caramel and other dissolving materials but it should not affect the scaling laws when comparing the different caramel experiments with each other. We have chosen to use the value of the saturated solution for the diffusion coefficient whatever the concentration of the bath solution. Raw data are listed in tables II, III, IV and V. Figure 5 shows the dependency of the receding velocity on the inclination angle and the density of the outer bath. Figure 6 shows the dependency of the wavelength of incipient longitudinal stripes and of the characteristic time to observe them on the inclination angle and the density of the outer bath.

An easy way of estimating a lower bound of the characteristic dissolution velocity α of a material consists in measuring the rate of dissolution of a body of this material in a water flow as fast as possible. Thereby, the thickness of the

* sylvain.courrech@univ-paris-diderot.fr

ρ_b (kg.m ⁻³)	θ (deg)	$-\dot{h}$ (mm/s)	λ_s (mm)	C_b (kg.m ⁻³)	C_f (kg.m ⁻³)	η_f ($\times 10^{-3}$ Pa.s)
997.12	6	$6.35 \times 10^{-3} \pm 2 \times 10^{-4}$		0	158.63	1.23
997.12	11.5	$5.18 \times 10^{-3} \pm 5 \times 10^{-4}$		0	158.63	1.23
997.12	21.4	$5.13 \times 10^{-3} \pm 1 \times 10^{-4}$		0	158.63	1.23
997.12	29.6	$5.67 \times 10^{-3} \pm 1 \times 10^{-4}$		0	158.63	1.23
997.12	45.4	$4.2 \times 10^{-3} \pm 4 \times 10^{-4}$		0	158.63	1.23
997.12	60.1	$4.21 \times 10^{-3} \pm 3 \times 10^{-4}$		0	158.63	1.23
997.12	70.8	$3.5 \times 10^{-3} \pm 1.6 \times 10^{-4}$		0	158.63	1.23
997.12	74.5	$3 \times 10^{-3} \pm 4 \times 10^{-4}$		0	158.63	1.23
997.12	79.8	$3.23 \times 10^{-3} \pm 3.5 \times 10^{-4}$		0	158.63	1.23
997.12	86.8	$2.12 \times 10^{-3} \pm 2.4 \times 10^{-4}$		0	158.63	1.23
999.8	30	$5.67 \times 10^{-3} \pm 1.1 \times 10^{-4}$		4.25	160.75	1.24
1025	30	$3.59 \times 10^{-3} \pm 1.8 \times 10^{-4}$		44.18	180.72	1.31
1065	30	$2.08 \times 10^{-3} \pm 1 \times 10^{-4}$		107.58	212.41	1.43
1108	30	$1.29 \times 10^{-3} \pm 1.9 \times 10^{-4}$		175.73	246.49	1.58
1152.6	30	$3.85 \times 10^{-4} \pm 7.7 \times 10^{-5}$		246.41	281.83	1.76
1178	30	$1.53 \times 10^{-4} \pm 8.3 \times 10^{-6}$		286.66	301.96	1.87
1188.4	30	$1.65 \times 10^{-5} \pm 2 \times 10^{-6}$		303.15	310.2	1.92
1197	0	$2.56 \times 10^{-6} \pm 5 \times 10^{-7}$		316.78	317.01	1.96
1179.6	82.7	$8.37 \times 10^{-5} \pm 5 \times 10^{-6}$		289.2	303.2	1.88
1179	5	1.27×10^{-4}		288.25	302.75	1.88
1172.7	84.2	$1.32 \times 10^{-4} \pm 5 \times 10^{-6}$		278.26	297.76	1.85
1171.9	29.3	$2.3 \times 10^{-4} \pm 5 \times 10^{-5}$		277	297.12	1.84
1105.8	69.8	$1.02 \times 10^{-3} \pm 5 \times 10^{-5}$		172.24	244.74	1.57
1020.15	80.3	$2.24 \times 10^{-3} \pm 1 \times 10^{-4}$		36.5	176.87	1.3
1012.3	21	$3.89 \times 10^{-3} \pm 1 \times 10^{-4}$	0.56 ± 0.2	24.06	170.65	1.27
1182.4	45		1.55 ± 0.3	293.64	305.4	1.89
1192.6	45		2.4 ± 0.4	309.8	313.53	1.94
1192.2	82		3.2 ± 0.7	309.17	313.21	1.94
997	70		0.61 ± 0.3	0	158.53	1.23
1105.6	70		0.8 ± 0.1	171.92	244.59	1.57
1119.2	70		0.915 ± 0.1	193.48	255.36	1.62
1134.8	70		1.09 ± 0.05	218.2	267.72	1.69
1164.4	70		1.21 ± 0.15	265.11	291.18	1.81

TABLE I. Raw data of the receding velocity \dot{h} and of the first observed wavelength λ_s of the longitudinal stripes at the bottom interface of inclined blocks of salt into saline solutions of various densities. We measure the density ρ_b of the solution and the inclination θ of the block. Measurement uncertainties are $2-3 \text{ kg.m}^{-3}$ for ρ_b and $1-2 \text{ deg}$ for θ . We calculate the concentration C_b of the solution from the density ρ_b . The characteristic concentration C_f in the solute layer is taken halfway between the concentration of the solution and the saturation concentration $C_{\text{sat}} = 317 \text{ kg.m}^{-3}$. C_{sat} is calculated from the measured density of the saturated brine $\rho_{\text{sat}} = 1197.3 \text{ kg.m}^{-3}$ and the tabulated value of the solubility (359.45 g of salt for one liter of water). The characteristic viscosity η_f inside the solute layer is calculated from the characteristic density ρ_f (fig. 1). Experiments were done at room temperature ($\sim 22^\circ\text{C}$).

solute boundary layer is very thin and the system approaches the regime where the dissolution is limited by the chemical kinetic. Doing so with a magnetic stirrer, we found $5 \times 10^{-6} \text{ m/s}$ as a rough lower bound for α . Taking the value of the coefficient of diffusion at zero concentration $D = 5.2 \times 10^{-8} \text{ m}^2/\text{s}$ gives an upper bound value of D/α of $1 \times 10^{-2} \text{ m}$ which is larger than the wavelengths we observe. Taking the value of the coefficient of diffusion at the saturation concentration $D = 1.34 \times 10^{-12} \text{ m}^2/\text{s}$ gives an upper bound value of D/α of about $2 \times 10^{-7} \text{ m}$, which is much smaller than the values of wavelengths we measure on caramel blocks.

-
- [1] V. Vitagliano and P. A. Lyons, Diffusion coefficients for aqueous solutions of sodium chloride and barium chloride, *Journal of the American Chemical Society* **78**, 1549 (1956).
[2] M. Alkattan, E. H. Oelkers, J.-L. Dandurand, and J. Schott, Experimental studies of halite dissolution kinetics, 1 the effect of saturation state and the presence of trace metals, *Chemical Geology* **137**, 201 (1997).
[3] J. Colombani and J. Bert, Holographic interferometry study of the dissolution and diffusion of gypsum in water, *Geochimica et cosmochimica acta* **71**, 1913 (2007).

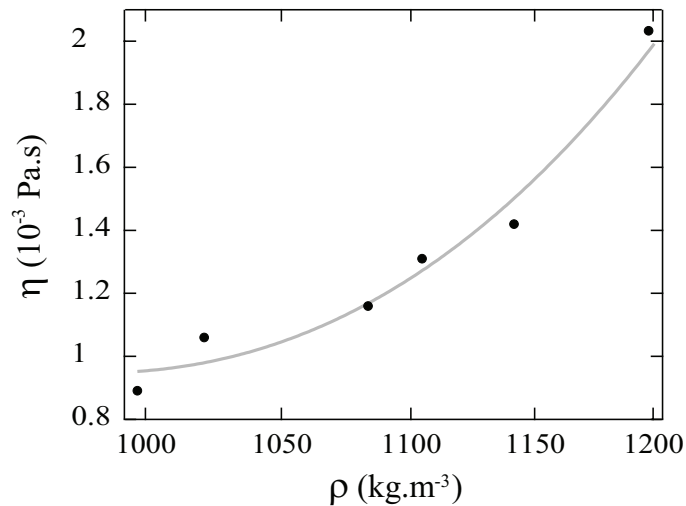


FIG. 1. Dynamic viscosity η of table salt solutions as a function of the solution density ρ at $T = 22^\circ C$. The grey curve is the polynomial fit we use to calculate the viscosity: $y = 22.471 - 0.043753x + (2.2236 \times 10^{-6})x^2$.

θ (deg)	$-\dot{h}$ ($\times 10^{-3}$ mm/s)
0.5	3.47 ± 0.03
20	3.64 "
43	2.92 "
73	2.89 "
88	1.13 "
31	3 "
33	3.06 "
38.5	3.17 "
51	2.75 "
51	3.39 "
62	2.78 "
63	2.28 "
66	2.67 "
69	2.17 "
74	1.86 "
78	2.08 "
82	1.81 "
88	1.08 "

TABLE II. Raw data of the receding velocity \dot{h} at the bottom interface of caramel blocks into water with different inclination angles θ . The concentration of the outer solution is zero. For the calculations we take the density of the solution $\rho_b = 997.3 \text{ kg.m}^3$ and the characteristic concentration and viscosity inside the solute layer to be $C_f = 484.13 \text{ kg.m}^3$ and $\eta_f = 1.46 \times 10^{-2} \text{ Pa.s}$. The characteristic viscosity η_f inside the solute layer is calculated from the characteristic density ρ_f (fig. 4).

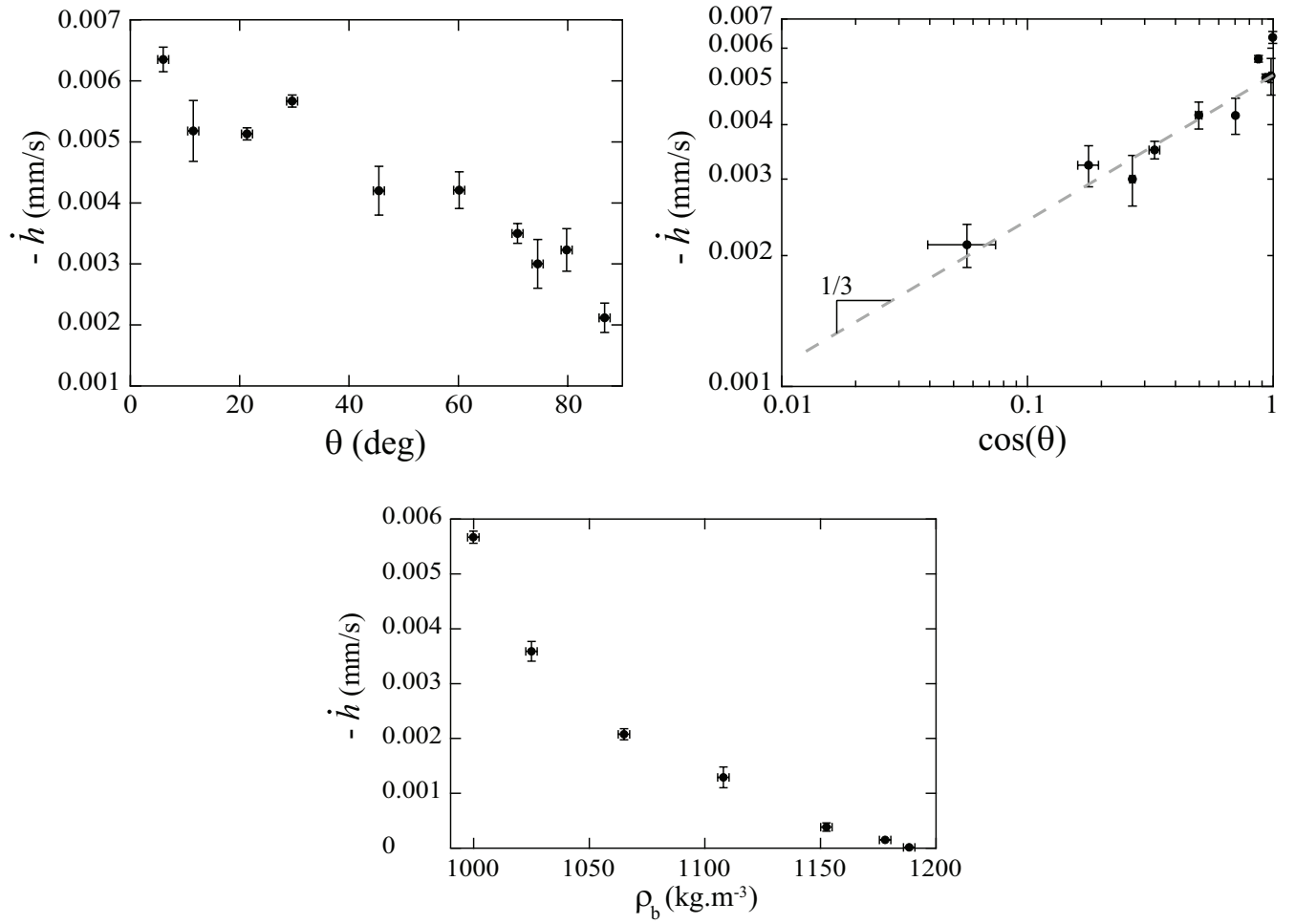


FIG. 2. Receding velocity \dot{h} of the bottom interface of salt blocks in tap water as a function of the inclination angle θ (top left), as a function of $\cos(\theta)$ with logarithmic axes (top right) and inclined of 30 deg as a function of the density ρ_b of the saline solution of the outer bath (bottom).

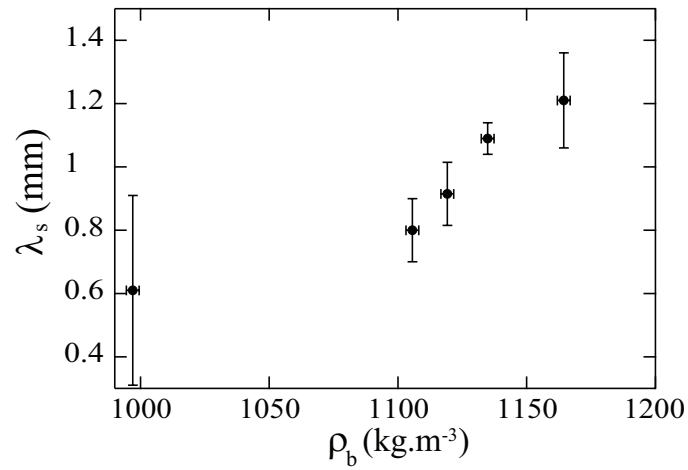


FIG. 3. Top: First observed wavelength λ_s of longitudinal patterns as a function of the density of the saline solution of the outer bath. The inclination angle of blocks is $\theta = 30$ deg.

ρ_b (kg.m ⁻³)	θ (deg)	$-\dot{h}$ ($\times 10^{-3}$ mm/s)	C_b (kg.m ⁻³)	C_f (kg.m ⁻³)	η_f (Pa.s)
1000	73	1.79	0	484.07	0.0149
1000	73	2.89	0	484.07	0.0149
1000	75	2.66	0	484.07	0.0149
1001	73	1.39	0	484.1	0.015
1100	73	0.836	219.66	593.961	0.0348
1190	73	0.364	412.16	690.209	0.116
1250	73	0.166	540.49	754.375	0.442
1310	73	0.0706	668.82	818.541	1.78
1320	73	0.0617	690.21	829.235	2.38
1050	73	0.973	112.72	540.489	0.0218
1100	75	0.927	219.66	593.961	0.0348
1150	75	0.822	326.60	647.432	0.0627
1200	75	0.619	433.55	700.904	0.137
1250	75	0.25	540.49	754.375	0.442
1300	75	0.151	647.43	807.846	1.41
1320	75	0.151	690.21	829.235	2.38
1403	75	0.00278	867.73	917.997	20.07

TABLE III. Raw data of the receding velocity \dot{h} at the bottom interface of inclined caramel blocks into sucrose-water solutions. Last line corresponds to an inclined caramel block into a water-caramel solution. The uncertainties are about 20% of the measured values. The characteristic viscosity η_f inside the solute layer is calculated from the characteristic density ρ_f (fig. 4).

ρ_b (kg.m ⁻³)	θ (deg)	λ_s (mm)	T_s (s)	C_b (kg.m ⁻³)	C_f (kg.m ⁻³)	η_f (Pa.s)
1432	73		321300 \pm 100	929.76	949.01	32.38
1410	73		143700 \pm 100	882.71	925.48	22.78
1403	73	4.45 \pm 0.8		867.73	918	20.07
1356	73	3.2 \pm 0.4		767.21	867.73	5.8
1303	70	1.57 \pm 0.29	1590 \pm 10	653.85	811.06	1.5
1274	71	1.26 \pm 0.2	390 \pm 10	591.82	780.04	0.765
1229	75	0.99 \pm 0.44	150 \pm 10	495.57	731.92	0.273
1155	75	0.63 \pm 0.2	30 \pm 10	337.3	652.78	0.0669
1108	72	0.52 \pm 0.15	20 \pm 10	236.77	602.52	0.0381
1075	72	0.55 \pm 0.18	15 \pm 5	166.19	567.23	0.0268
1041	70	0.45 \pm 0.24	10 \pm 5	93.47	530.86	0.0203
1006	72	0.41 \pm 0.16	1 \pm 1	18.61	493.44	0.0155
1000	75	0.44 \pm 0.17	1 \pm 1	0	484.07	0.0149
1000	60	0.4 \pm 0.2		0	484.07	0.0149

TABLE IV. Raw data of the first observed wavelength λ_s of the longitudinal stripes and the characteristic time T_s to observe them on inclined caramel blocks into water-caramel solutions of various densities ρ_b . We measure the density ρ_b of the solution and the inclination θ of the block. We calculate the concentration C_b of the solution from the density ρ_b . The characteristic concentration C_f in the solute layer is taken halfway between the concentration of the solution and the saturation concentration $C_{\text{sat}} = 968.13$ kg.m⁻³. C_{sat} is calculated from the measured density of the saturated solution $\rho_{\text{sat}} = 1450$ kg.m⁻³. Experiments were done at room temperature ($\sim 22^\circ\text{C}$). The characteristic viscosity η_f inside the solute layer is calculated from the characteristic density ρ_f (fig. 4).

ρ_b (kg.m ⁻³)	θ (deg)	λ_s (mm)	T_s (s)	C_b (kg.m ⁻³)	C_f (kg.m ⁻³)	η_f (Pa.s)
1000	74	0.39 ± 0.14		0	484.07	0.0149
1000	75	0.38 ± 0.18		0	484.07	0.0149
1000	76	0.44 ± 0.24	1 ± 1	0	484.07	0.0149
1320	75	2.1 ± 0.4	1800 ± 100	690.21	829.24	2.38
1230	74	0.76 ± 0.25		497.71	732.99	0.279
1295	75	1.54 ± 0.76	420 ± 10	636.74	802.5	1.26
1140	75	0.51 ± 0.25		305.22	636.74	0.0559
1040	75	0.4 ± 0.24	8 ± 2	91.33	529.8	0.0201
1100	75	0.59 ± 0.23	8 ± 2	219.66	593.96	0.0348
1080	75	0.48 ± 0.2		176.88	572.57	0.0281
1220	73	0.71 ± 0.32		476.32	722.29	0.218
1275	74	1.1 ± 0.4	270 ± 10	593.96	781.11	0.784
1300	76	1.64 ± 0.56	510 ± 10	647.43	807.85	1.41
1200	75	0.85 ± 0.46		433.55	700.9	0.137
1210	73	0.64 ± 0.43		454.94	711.6	0.17
1285	75	1.275 ± 0.43	400 ± 20	615.35	791.81	0.998
1197	75	0.79 ± 0.31		427.13	697.7	0.13
1235	76	0.73 ± 0.25		508.41	738.33	0.315
1249	75	0.91 ± 0.51		538.35	753.31	0.43
1310	76	1.84 ± 0.74	1650 ± 20	668.82	818.54	1.78
1305	74	1.64 ± 0.46	960 ± 20	658.13	813.19	1.57
1240	75	0.97 ± 0.51	115 ± 10	519.1	743.68	0.354
1255	75	0.935 ± 0.33		551.183	759.72	0.49
1319	76	1.93 ± 0.57		688.07	828.17	2.31
1280	77	1.17 ± 0.43	320 ± 10	604.66	786.46	0.885
1260	76	1.09 ± 0.51	150 ± 20	561.88	765.07	0.547
1215	77	0.69 ± 0.39	85 ± 5	465.63	716.95	0.192
1150	85	0.77 ± 0.41	160 ± 10	326.6	647.43	0.0627
1150	80	0.66 ± 0.29	65 ± 5	326.6	647.43	0.0627
1150	75	0.62 ± 0.25	45 ± 5	326.6	647.43	0.0627
1150	60	0.49 ± 0.15	24 ± 4	326.6	647.43	0.0627
1150	45	0.45 ± 0.28	18 ± 2	326.6	647.43	0.0627

TABLE V. Raw data of the first observed wavelength λ_s of the longitudinal stripes and the characteristic time T_s to observe them on inclined caramel blocks into sucrose solutions of various densities ρ_b . We measure the density ρ_b of the solution and the inclination θ of the block. We calculate the concentration C_b of the solution from the density ρ_b . The characteristic concentration C_f in the solute layer is taken halfway between the concentration of the solution and the saturation concentration $C_{\text{sat}} = 968.13 \text{ kg.m}^{-3}$. C_{sat} is calculated from the measured density of the saturated solution $\rho_{\text{sat}} = 1450 \text{ kg.m}^{-3}$. Experiments were done at room temperature ($\sim 22^\circ\text{C}$). The characteristic viscosity η_f inside the solute layer is calculated from the characteristic density ρ_f (fig. 4).

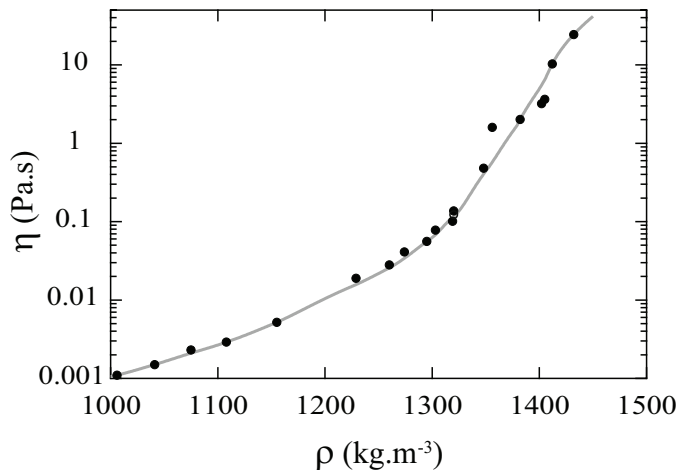


FIG. 4. Dynamic viscosity η of caramel solutions as a function of the solution density ρ at $T = 21^\circ\text{C}$. The empty symbol ($\rho = 1320 \text{ kg.m}^{-3}$) is a measurement for a sucrose solution. The grey curve is a polynomial fit used to calculate η_f .

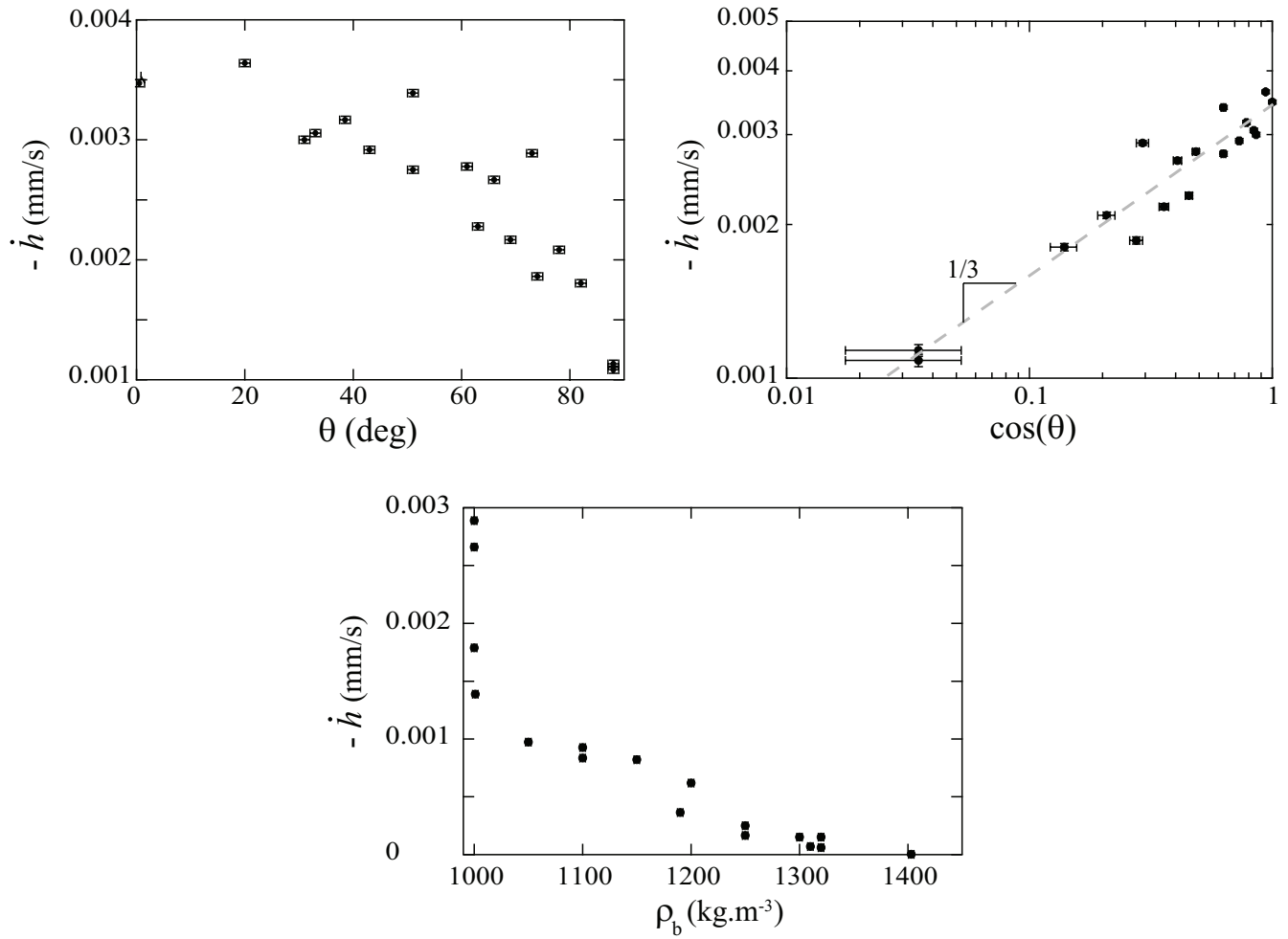


FIG. 5. Receding velocity \dot{h} of the bottom interface of caramel blocks in tap water as a function of the inclination angle θ (top left), as a function of $\cos(\theta)$ with logarithmic axes (top right) and inclined of an angle ranging between 73 and 76 deg as a function of the density ρ_b of the sucrose - water solution of the outer bath (bottom).

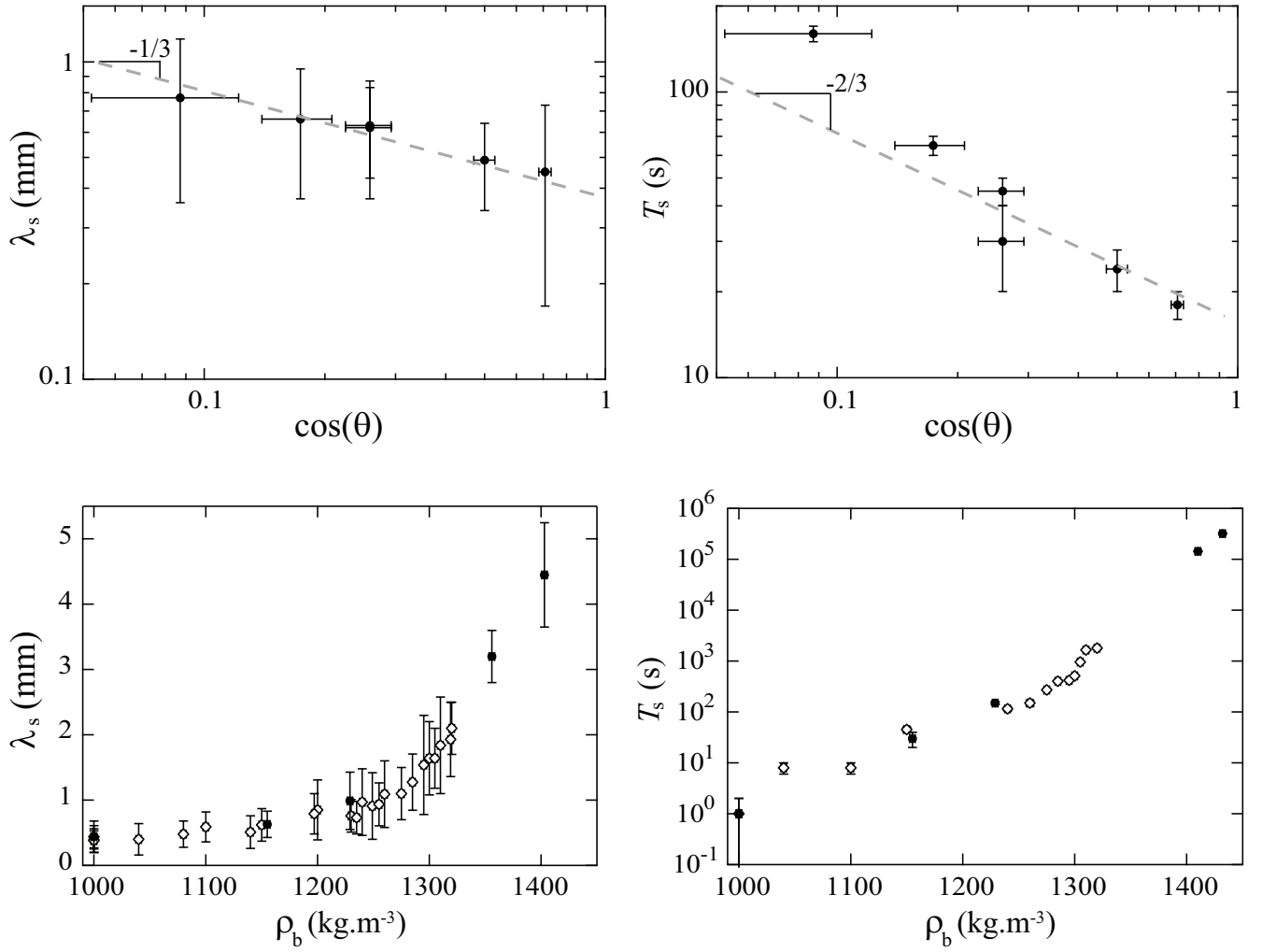


FIG. 6. Top: First observed wavelength λ_s of longitudinal patterns (left) and characteristic time T_s to observe them (right) as a function of the inclination $\cos(\theta)$ with logarithmic axes. The density of the sucrose - water solution is $\rho_b = 1150 \text{ kg.m}^{-3}$. Bottom: First observed wavelength λ_s of longitudinal patterns (left) and characteristic time T_s to observe them (right) as a function of the density ρ_b of the outer bath. The inclination angle ranges between 73 and 76 deg. The outer bath is either a caramel - water solution (plain circles) or a sucrose - water solution (empty diamonds).