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Bénard-Marangoni instability driven by moisture absorption

SANGWOO SHIN¹, IAN JACOBI^{1,2} and HOWARD A. STONE¹

¹ *Department of Mechanical and Aerospace Engineering, Princeton University - Princeton, NJ 08544, USA*

² *Faculty of Aerospace Engineering, Technion-Israel Institute of Technology - Haifa 32000, Israel*

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Abstract – Glycerol is a viscous liquid widely used in industry and known for its strong hygroscopic nature. While this unusual property has been well documented from the perspective of solution chemistry, its impact on the mechanical properties of glycerol remains largely unknown. Here, we report a spontaneous Bénard-Marangoni instability in pure glycerol driven by absorption of water vapor. Even under standard laboratory conditions, ambient humidity is sufficient to drive distinct Bénard-Marangoni convection cells for hours. We describe the mechanism of this instability in terms of the vapor transport process and competition between solutal and thermal Marangoni forces, and provide insight into potential uses and impact of this aspect of dynamics driven by moisture absorption in various common settings.

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Marangoni convection is a flow driven by surface-tension gradients, which result from interfacial inhomogeneities in either temperature or solute concentration. Within a thin liquid layer, the resulting interfacial flow can induce convection cells, which appear as a distinct stationary pattern on the interface [1]. Although the direct effect of solutal concentration gradients on surface tension is often much stronger than indirect thermal effects [2], the two mechanisms are not mutually exclusive. In some cases the presence of different chemical solutes can itself alter the temperature distribution in the system by means of reactions or the chemical heat of solution. These solutal heating effects can then either accelerate the solutal instability or damp it, depending, respectively, on whether energy is chemically released or absorbed [3,4]. Moreover, even uniform heat generation within a solutal Marangoni system can initiate [5,6] or further destabilize [7,8] the interfacial flows. Marangoni convection results in a significant change in the mass transport properties of binary liquid systems. In applications from falling thin films [9,10] to mixing between liquid layers [11], changes in velocity and temperature distributions due to Marangoni convection can result in vastly different transport behaviors.

A surprisingly common binary liquid pair that is susceptible to a variation of this Bénard-Marangoni (BM) instability is the water-glycerol system. Glycerol solutions are widely found in industry, in everything from pharmaceutical co-solvents to natural fermentation and food

production processes [12], and are commonly used in a variety of laboratory experiments, from basic fluid mechanics to protein crystallization [13]. One of glycerol's useful properties is its strong hygroscopic nature. Not only is glycerol highly soluble in water, glycerol naturally absorbs water vapor from the air under standard laboratory temperature and humidity conditions. This chemical property of glycerol is well known [14], and glycerol is often exploited as a natural humectant or dessicant [12], but the hygroscopic property can also result in unintentional glycerol-water solutions when atmospheric exposure causes undesired water uptake. Nevertheless, the fluid mechanical consequences of the solutions have not been considered, despite their significant implications for applications. In this study, we report the presence of a spontaneous BM instability within the water-glycerol system, explain its physical mechanism, and examine its implications for fluid transport in a variety of practical scenarios.

Materials and methods. – Glycerol ($\geq 99.5\%$) and sodium dodecyl sulfate ($\geq 98.5\%$) were purchased from Sigma-Aldrich. The BM convection cells were visualized via Schlieren photography using a digital camera (D5100, Nikon) and a concave mirror ($f = 500\text{ mm}$, Thorlabs). A macro lens equipped with a long hood was used, which effectively screens out the refracted light. A small number density of $2\text{ }\mu\text{m}$ polystyrene particles (Polysciences) was added to the glycerol to visualize the pathlines of the

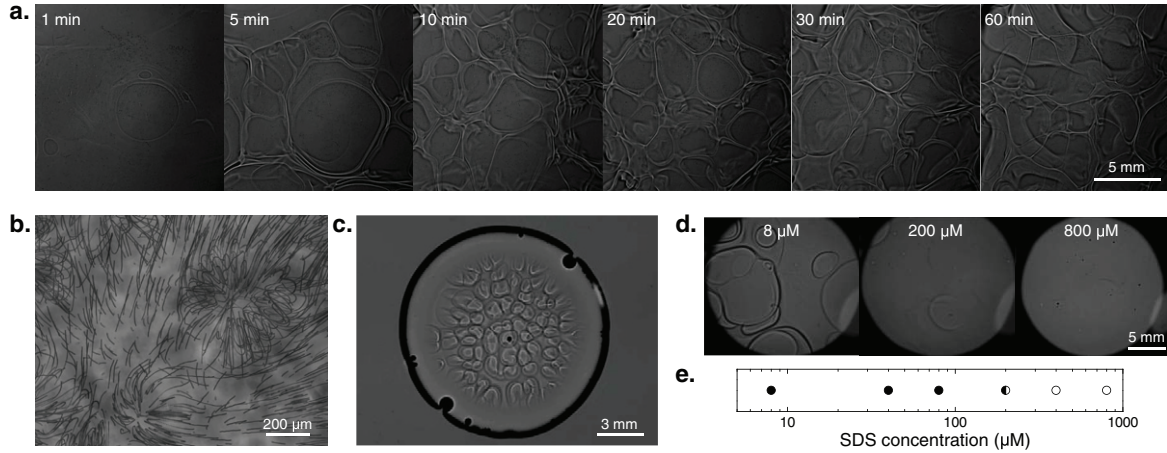


Fig. 1: Spontaneous formation of Bénard-Marangoni convection cells in glycerol. (a) Time lapse of convection cells from a pool of glycerol (fluid depth ≈ 1 cm) imaged by Schlieren photography (supplementary video [movie1.mp4](#)). (b) Reconstructed optical microscope image of particle pathlines in a droplet of glycerol by stacking sequential images (total time elapsed = 40 s). (c)–(e) Surface-tension-driven convection cells. (c) Convection cells in a droplet of glycerol still appear even when the system is oriented upside-down (imaged by Schlieren photography, supplementary video [movie2.mp4](#)). (d), (e): influence of surfactant (sodium dodecyl sulfate) concentration on the formation of convection cells. (d) Schlieren images from pools of glycerol with different surfactant concentrations (see supplementary video [movie3.mp4](#)). (e) A plot showing that the formation of convection cells (unstable, filled circle) are being suppressed (stable, open circle) as the concentration exceeds a threshold value (half-filled circle).

convection cells using an optical microscope (DMI4000B, Leica), whereas $20\text{ }\mu\text{m}$ polyamid particles (Dantec Dynamics) were used to measure the flow speed using a digital camera (uEye, IDS). The temperature of the glycerol surface was measured using an infrared thermometer (E40, FLIR). The mass of the glycerol was measured using a computer-connected digital balance (Practum 124-1S, Sartorius). Flow speed, temperature and mass measurements were conducted in a humidity-controlled closed chamber, where the relative humidity (RH) is controlled via a water source and a dry air source. RH was measured using a digital hygrometer (VWR).

For the fluid mechanical experiments, a cone-and-plate rheometer (Physica MCR 301, Anton Paar) was used to measure the viscosity of the glycerol. A digital spectrometer (USB2000+, Ocean Optics) was used to measure the thickness of the falling glycerol film. Thin glass capillary tubes (Drummond Scientific) were used to conduct capillary rise experiments. All of the experiments were conducted in a humidity-controlled chamber at room temperature.

Spontaneous BM instability induced by water absorption. – When pure glycerol is brought into contact with air, we observe unsteady, periodic patterns on the surface of glycerol (fig. 1). The patterns form within a minute of the exposure to a normal laboratory atmosphere (temperature $\approx 24.5^\circ\text{C}$, relative humidity $\approx 40\%$, pressure ≈ 1 atm), and persist for hours (fig. 1(a), supplementary video [movie1.mp4](#)). By adding small polystyrene particles (diameter $\approx 2\text{ }\mu\text{m}$) to the glycerol and visualizing their pathlines, we confirm that the patterns are

convection cells (fig. 1(b)). Furthermore, the convection cells appear even when the system is oriented upside-down, which reverses any effect of buoyancy (fig. 1(c), supplementary video [movie2.mp4](#)), and thus indicates that the instability is driven by surface-tension gradients and not buoyancy. This conclusion is verified by the addition of trace amounts of surfactant (sodium dodecyl sulfate) to the glycerol, altering the interfacial properties and drastically affecting the presence of the convection cells. As the amount of surfactant is increased, the motion of the cells tends to slow down until a threshold concentration of surfactant is added ($\approx 200\text{ }\mu\text{M}$) at which point the convection cells are completely suppressed (figs. 1(d) and (e), supplementary video [movie3.mp4](#)). The surface-tension-dependent behavior of the convection cells indicates that the glycerol exhibits a self-initiating BM instability under normal laboratory conditions.

The surface-tension gradient driving this instability originates in the strong hygroscopic nature of glycerol. As glycerol absorbs water vapor from the air, a water-glycerol solution is formed at the interface, providing the basis for a solutal Marangoni flow. The surface tension of glycerol has a positive dependence on water content [14], so as water is absorbed locally at the interface, gradients in surface tension naturally arise. A local negative perturbation in the surface tension drives an outward flow towards higher surface tension, which, by continuity, draws upward fresh, undiluted (low surface tension) glycerol from beneath the interface, thus sustaining the instability.

The water vapor content of the air therefore controls the onset and magnitude of the instability. The dependence of the flow speed of the convection cells on the relative

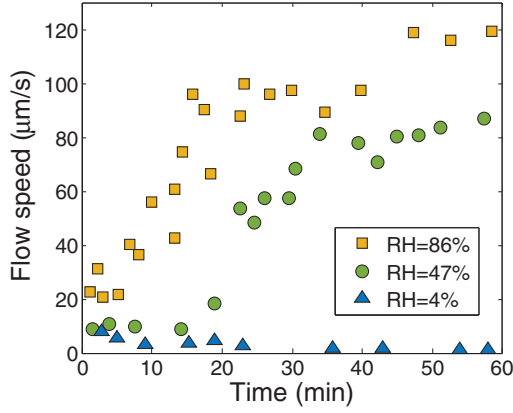


Fig. 2: (Colour online) Flow speed of the convection cells for various relative humidities (RH). Polyamid tracer particles with diameter of $20\mu\text{m}$ were employed to measure the flow speed of the convection cells from a pool of glycerol (fluid depth $\approx 1\text{ cm}$).

humidity (RH) of the atmosphere is shown in fig. 2. At low RH, a very small initial flow is observed due to the presence of water vapor that came from the atmosphere during the loading of the sample. However, this initial flow quickly subsides as the RH reaches a steady state within several minutes. Beyond some threshold RH, the flow within the convection cells begins and gradually accelerates for 20–30 minutes towards a steady-state speed, which ultimately persists for hours.

The threshold RH can be predicted by estimating the Marangoni number based on RH values. The Marangoni number is defined as $\text{Ma} = (\partial\gamma/\partial c)c_v d/(\mu D_m)$ [15], where $\partial\gamma/\partial c$ is the surface-tension gradient with respect to the water content ($\approx 0.23\text{ mN}\cdot\text{m}^{-1}\cdot\text{M}^{-1}$ [14,16]), c_v is the water vapor concentration, d is the depth of the container ($\approx 1\text{ cm}$), μ is the viscosity of the glycerol, and D_m is the molecular diffusivity of the glycerol ($\approx 1.4\times 10^{-11}\text{ m}^2\text{ s}^{-1}$ [17]). The water vapor concentration c_v is related to the RH by $c_v = (\text{RH}/100) \cdot p_{ws}/(R_v T M_w)$, where p_{ws} is the saturation water vapor pressure, R_v is the specific gas constant for water vapor, T is the temperature, and M_w is the molar mass of water. At $T = 24.5^\circ\text{C}$, $\text{Ma} \approx 92.5$ and ≈ 7.9 at $\text{RH} = 47\%$ and 4% , respectively. Pearson predicted that the critical Marangoni number $\text{Ma}_c = 48$ for the onset of the BM instability [18], which is between the Ma values we obtained at $\text{RH} = 47\%$ and 4% . From the above estimates, the threshold RH, where $\text{Ma} = \text{Ma}_c$, is predicted at $\text{RH} \approx 24\%$.

Heat release during water absorption. – The water uptake by glycerol not only modifies the surface tension of the resulting solution, it also produces an exothermic heat of solution ($\approx 5.8\text{ kJ mol}^{-1}$ [14,19]). We used infrared thermography to observe the temperature increase at the interface due to this energy release (fig. 3(a)) and the temperature map itself exhibits the same cellular pattern as the solutal instability. The fresh, undiluted

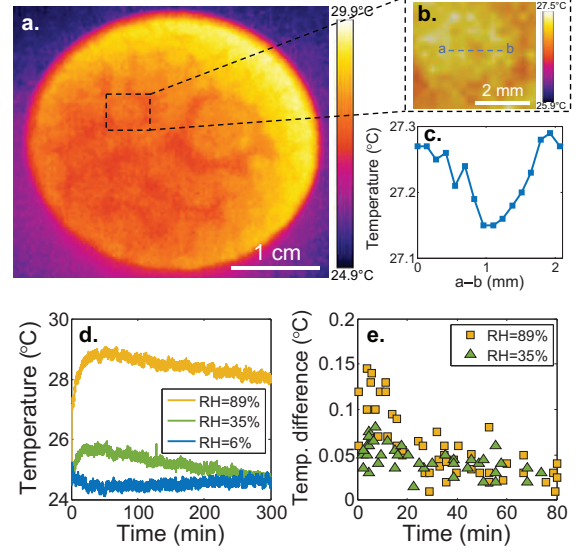


Fig. 3: (Colour online) Temperature change of the glycerol solution upon exposure to the air. (a) Infrared thermal image of the glycerol surface 6 minutes after being exposed to the air. (b) Close-up image of a single convection cell where the flow is directed radially outward from the center. (c) Temperature profile along line a–b in (b), indicating a temperature difference of $\approx 0.15^\circ\text{C}$. (d) Area-averaged surface temperature change *vs.* time for different RH. (e) Temperature difference within convection cells *vs.* time under different RH.

glycerol drawn up from beneath the interface is colder than the interfacial glycerol-water mixture, and thus a temperature gradient is formed at the interface, with a magnitude as high as 0.15°C , depending on the RH (figs. 3(b)–(e)). However, the surface tension of glycerol has a negative dependence on the temperature [14], therefore the temperature gradient competes against the concentration gradient, so that the thermal effects tend to suppress the solutal instability. We conclude that the solutal Marangoni force initiates and maintains the observed instability by means of the hygroscopic uptake of water vapor, whereas the thermal Marangoni force due to the heat of solution acts to damp the motion.

The damping effect of the heat of solution is most prominent only during the initial period of the instability, which results in very slow convection cells (fig. 2). Although the heating of the glycerol lasts for hours (fig. 3(d)), the local temperature gradients are observed only for the initial tens of minutes (fig. 3(e)). Therefore, the instability is initially suppressed by the significant temperature gradient. After ≈ 10 minutes, the temperature gradient within the convection cells starts to decay, which leads to a gradual acceleration of the flow until a steady state is reached.

Estimation of the flow speed. – We can estimate the steady-state flow speed (fig. 2) within the convection cells by scaling arguments. Assuming a quasi-steady flow structure, the stress balance between the viscous shear stress and the Marangoni stress at the glycerol-air

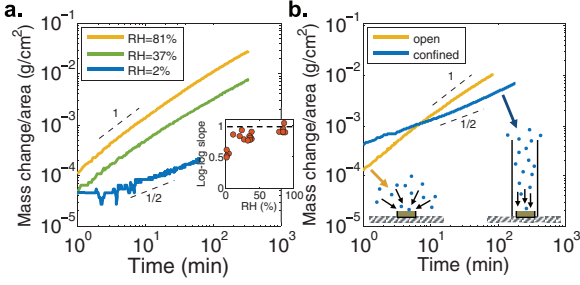


Fig. 4: (Colour online) Mass change of glycerol due to water vapor absorption. (a) Mass change per unit area *vs.* time (log-log scale) in an open system for different RH. The inset shows the slope of a log-log plot at one hour *vs.* RH. (b) Mass change per unit area *vs.* time (log-log scale) for different system configurations. Measurements are taken either on an open space (yellow curve) or in a confined space (blue curve).

interface scales as $\mu u / \ell_{\perp} \approx \Delta\gamma / \ell_{\parallel}$, where u is the characteristic flow speed, γ is the surface tension of the glycerol, $\Delta\gamma$ is the surface-tension change that drives the flow, ℓ_{\perp} is the length scale perpendicular to the glycerol surface, and ℓ_{\parallel} is the length scale parallel to the glycerol surface. Provided that the convection cells are not geometrically confined, *i.e.*, the depth of the container (≈ 1 cm) is larger than ℓ_{\perp} , then both length scales should be identical owing to the isotropic nature of Marangoni-type convection cells, *i.e.*, $\ell_{\perp} \approx \ell_{\parallel} = \ell$ [20]. This assumption is verified experimentally from the cross-sectional view of the convection cells and ℓ is taken as a typical radius of a convection cell (≈ 2 mm). The characteristic flow velocity can then be represented as $u = \alpha \Delta\gamma / \mu$, where α is a dimensionless prefactor of $\mathcal{O}(1)$ that accounts for the change of fluid properties due to water absorption; α will be treated in detail below.

The surface-tension difference $\Delta\gamma$ due to the solutal effect of the water uptake is obtained by estimating the concentration, c_0 , of the water at the surface of the glycerol assuming that the water molecules diffuse into the glycerol with a constant flux condition at the interface. This assumption is reasonable given that the rate of water vapor uptake is more or less linear over time above moderate RH conditions (fig. 4), and is discussed in detail below. Then, with j as the mass flux (slope of fig. 4(a)) and D the effective diffusion coefficient of water in glycerol, the water concentration at the interface with a boundary condition $\partial c / \partial x|_{x=0} = j / (DM_w)$ is given by $c_0(t) = 2j / M_w (t / (\pi D))^{1/2}$, where the distance x is measured normal to the interface [21]. On the time scale of convection along the interface, *i.e.*, $t = \ell / u$, the mass flux j produces concentration of the water at the interface as $c_0 = 2j / M_w (\ell / (\pi u D))^{1/2}$.

The effective diffusion coefficient in the system is governed by the mass transport behavior of the convection system. Mass transport can be characterized using the Péclet number, $Pe = u\ell / D_m$. In particular, mass transport across convection cells in the limit of high Péclet

number is known to exhibit a significant dispersion, where the effective diffusion coefficient is increased by a factor of $Pe^{1/2}$ [22,23]. In our case, owing to the high viscosity of glycerol, the BM convection cells quickly induce counter-rotating vortices that propagate along the depth direction much further than the thickness of the individual BM cells, which allows us to use the $Pe^{1/2}$ dispersion enhancement in our estimate for the effective diffusion process in the water-glycerol system. Therefore, by letting $D \approx Pe^{1/2} D_m = (u\ell D_m)^{1/2}$, we obtain the surface tension difference $\Delta\gamma \approx c_0 (\partial\gamma / \partial c)$ as

$$\Delta\gamma = \frac{2j}{M_w} \left(\frac{\ell}{\pi^2 u^3 D_m} \right)^{1/4} \frac{\partial\gamma}{\partial c}. \quad (1)$$

Finally, we obtain the characteristic steady-state flow speed u as

$$u = \left(\frac{16}{\pi^2} \frac{\alpha^4 j^4 \ell}{\mu^4 M_w^4 D_m} \left(\frac{\partial\gamma}{\partial c} \right)^4 \right)^{1/7}. \quad (2)$$

As mentioned, here we must account for variations in viscosity and diffusivity due to the water absorption, which are included in the factor α . However, from eq. (2), viscosity effects are only significant since $u \sim \mu^{-4/7} \sim D_m^{-1/7}$. This indicates that, as a first approximation, α represents the ratio between the viscosity of pure glycerol μ and the changed viscosity of the glycerol solution due to water absorption μ_c . Using the above estimates, $c_0 \approx 2.2$ M at $t = 1$ h, then the corresponding viscosity of a glycerol-water mixture is $\mu_c \approx 0.3$ Pa \cdot s [12], hence $\alpha \approx 3.5$. With this value, we obtain $u \approx 120 \mu\text{m s}^{-1}$ and $58 \mu\text{m s}^{-1}$ for RH = 81% and 37%, respectively, which are in good agreement with the experimental observations at the late times of the instability shown in fig. 2.

Relaxation oscillation in BM instability. – Although the instability persists as a steady state for hours in terms of cell size and flow speed, the convection cells themselves are unstable such that they eventually fade away and new cells are subsequently developed, exhibiting a relaxation-oscillation mode. This behavior usually occurs when the forcing that drives and sustains the convection cells is insufficient to maintain a steady flow, which causes an episodic relaxation and re-generation process [20,24]. Since the forcing driving the instability in the glycerol-water system is the uptake of water vapor, the presence of a relaxation-oscillation mode implies that the instability dynamics are vapor transport limited.

We can now examine the vapor transport more carefully by modeling the mass change of glycerol over time due to water uptake. Because the convection cells in the glycerol do not induce any significant advective motion of the adjacent air layer ($Pe_v = u\ell / D_v \ll 1$ in the air layer, where $D_v \approx 10^{-5}$ m²/s is the diffusion coefficient of water vapor in air), we assumed earlier that the vapor transport is driven primarily by diffusion. We can then model the

vapor transport from a semi-infinite volume of air with vapor concentration c_∞ towards a finite, shallow bath of glycerol as a hemispherical radial diffusion process. We express the mass flux of water vapor on the surface of a sphere of radius a as $\partial c / \partial r|_{r=a} = \Delta c / a + \Delta c / (\pi D t)^{1/2}$, where $\Delta c = c_\infty - c_0$ is the concentration difference between the atmosphere (c_∞) and the interface [21]. Then the mass increase Δm is the time integral of the vapor flux, which yields $\Delta m \propto (\Delta c / a)t + (2\Delta c / (\pi D)^{1/2})t^{1/2}$. At low RH, the second term dominates, giving rise to $t^{1/2}$ variation of mass, which we measure in experiments (fig. 4(a)). As RH increases, the time dependence becomes linear, implying that the steady state is reached quickly. In contrast to the hemispherical diffusion in an open atmosphere, a cylindrically confined system was constructed experimentally to mimic 1D linear diffusion and it was found to exhibit a $t^{1/2}$ scaling at high RH, which further confirms the diffusive nature of vapor transport toward the air-glycerol interface (fig. 4(b)).

Modeling the vapor transport as a diffusive process helps to explain the presence of the transport-limited relaxation-oscillation mode, since the hygroscopic strength of the glycerol rapidly depletes water vapor near the interface, thereby creating a diffusion layer through which the transport is limited. The hemispherical diffusion geometry also rationalizes why the temperature of the glycerol at the edge of the container rises faster than at the center, as clearly seen in fig. 3(a).

Impact of water absorption on various fluid mechanical experiments. – Not only does the mass uptake of water serve as the driving force behind the instability in the water-glycerol system, it also presents its own significant fluid mechanical consequences, which last as long as the glycerol is not fully saturated by water: as long as the water uptake persists, the solution’s properties continue to change. This response has striking implications for many experiments that rely on glycerol’s mechanical properties, in particular its high viscosity [25–28]. To illustrate this impact, we consider the sensitivity of a number of common experimental configurations to hygroscopically driven changes in water concentration.

In a conventional cone-and-plate rheometer for viscosity measurements, the fluid in the gap between the plates is exposed to the atmosphere for a sustained measurement period. Figure 5(a) shows that, for moderate RH, even a thin gap (≈ 0.5 mm) allows vapor absorption that significantly affects the measured viscosity value over time.

Many industrial coating processes rely on gravity-driven thin-film drainage from a vertical flat plate. For constant mechanical properties, the thickness of the draining liquid varies as $t^{-1/2}$ [29], which is in a good agreement with the results at low RH (fig. 5(b)). However, as RH increases, rapid vapor absorption makes the viscosity a time-dependent variable, and alters the time-dependent response. This time-dependent viscosity is also critical in capillary rise experiments where the initial rise rate

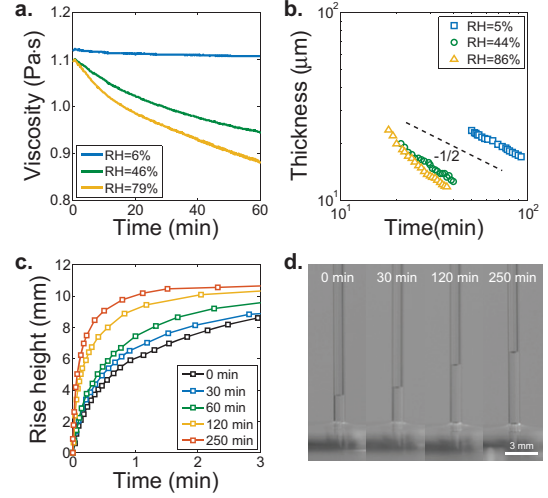


Fig. 5: (Colour online) Some classical fluid experiments with glycerol involving direct exposure to the air. (a) Viscosity measurements with a cone-and-plate rheometer under different RH. (b) Thickness change of glycerol from a vertical plate upon drainage due to gravity under different RH. (c), (d): capillary rise with the same glycerol solution at high RH (RH = 81%), but with different initial times of exposure to the air prior to experiments. (c) Plot of the capillary rise height *vs.* time. (d) Optical images of the capillary rise at 10 seconds for different initial exposure times (see supplementary video [movie4.mp4](#)).

depends on the viscosity of the fluid [30]. Figures 5(c) and (d) (supplementary video [movie4.mp4](#)) demonstrate how the initial rise rate increases with the duration of prior exposure of the glycerol to air.

Conclusion. – We have observed the spontaneous formation of a Bénard-Marangoni instability in a water vapor/glycerol system; The instability is driven by the diffusion-limited, hygroscopic absorption of water vapor and is damped, for early times, by the exothermic heat of solution. The mass uptake of water in this system not only generates an interfacial instability, but significantly alters the mechanical properties of the system in ways that can significantly impact many classical experimental techniques but also offer new opportunities for exploiting a simple, time-dependent binary fluid system. Moreover, this instability is not limited to glycerol, but is also possible for other types of hygroscopic fluids having exothermic heat of solution such as ethylene glycol, which may impact their performance and long-term stability in heat transfer applications [31].

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