








Chemical flowers: Buoyancy-driven instabilities under modulated gravity during a parabolic flight

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Fluid displacements occur in a wide variety of applications such as CO₂ sequestration [1], H₂ storage [2], and soil remediation or reactive transfer in porous media [3] where they can appear in diverse levels of complexity. The understanding of the flow pattern and the derivation of relevant scalings for such displacements is of great importance for the above applications. Here we study experimentally a buoyancy-driven instability arising during the horizontal displacement of a fluid by another miscible fluid of different density in a Hele-Shaw (HS) cell, a fluid flow setup that allows us to investigate quasi-two-dimensional flows. It consists of two plexiglass plates separated by a thin gap of height h , where fluid is injected from a point in the middle of the cell. The displacement leads to a complex three-dimensional flow pattern [4,5].

As illustrated in Fig. 1, due to the unstable density stratification in the HS gap, a Rayleigh–Taylor-like mechanism triggers an instability which generates characteristic convective rolls perpendicular to the displacement direction [visible as fine radial stripes in Fig. 3(a)]. Rayleigh–Taylor convection refers to the sinking of a denser fluid that lies on top of a less dense one because of buoyancy. The

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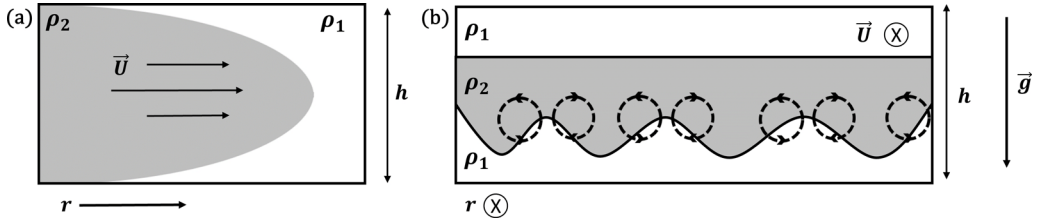


FIG. 1. Sketch of a miscible displacement of a fluid of density ρ_1 by a fluid with higher density ρ_2 : (a) perpendicular view and (b) transversal view.

dynamics depends on the system's Rayleigh number $Ra = (\rho_2 - \rho_1)gh^3/\mu D$, where ρ_1 and ρ_2 are the densities of the two solutions, g is the acceleration experienced by the system, μ is the mean dynamic viscosity of the two miscible solutions, and D is the diffusion coefficient of the species contributing to the density gradient [4,5].

To investigate the instability further and to assess the effect of gravity on it, we performed experiments onboard a parabolic flight aircraft (Air Zero G, Novespace, Bordeaux, France). Such parabolic flights offer acceleration modulations between micro- g (i.e., approximately equal to $10^{-2}g$) and $1.8g$. Hence, parabolic flights offer the possibility to investigate systems under weightlessness, thus eliminating any buoyancy effects. In addition, during the sequence of flown parabolas, almost twice the earth's gravitational acceleration is temporarily reached (hyper- g). We conducted experiments in a HS cell setup with radial injection. The setup and the procedures used in the experiments are described in detail by Stergiou *et al.* [6]. A schematic of the employed experimental setup and a photo of the parabolic flight equipment is shown in Fig. 2.

We used two colorless reactant solutions as the two miscible liquids of different density. Upon reacting, a colored product forms in their mixing zone, visualizing the instability. A white LED light array illuminated the HS cell reactor, while a monochrome camera recorded the experiment from the top. Prior to the flight, we filled the Hele-Shaw cell reactor with an aqueous 0.03 M KSCN solution ($\rho_1 = 999.6 \text{ kg m}^{-3}$). During the flight, we injected radially an aqueous 0.03 M FeNO_3 ($\rho_2 = 1007.4 \text{ kg m}^{-3}$) solution which contained 0.1 M HNO_3 as well. When the two solutions came in contact, a red product formed (FeSCN^{2+}). The developing flow patterns translated into spatial patterns of the product concentration distribution.

We have observed that the onset time of the instability and the ordered spatial distribution of the convective rolls depend on the gap height h , the injection flow rate Q , and the acceleration magnitude during the parabolic flight. In Fig. 3 we show that the instability is absent under micro- g injection, whereas it is well developed in normal $1g$ acceleration, proving the buoyant nature of the instability. Note that for the same duration t , the reaction front has moved farther in the Hele-Shaw

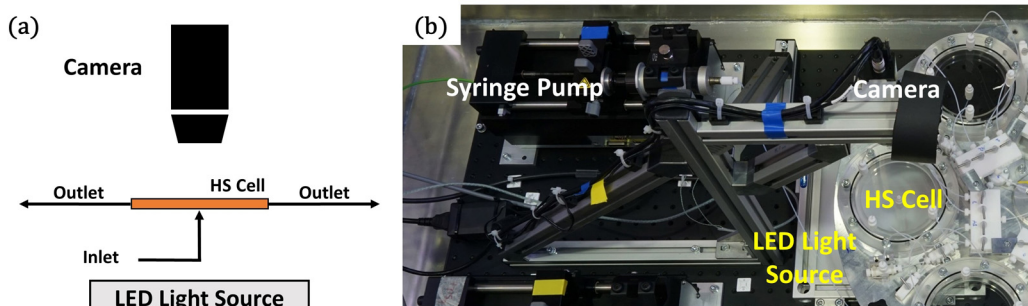


FIG. 2. Experimental setup: (a) schematic representation and (b) setup used for the parabolic flights.

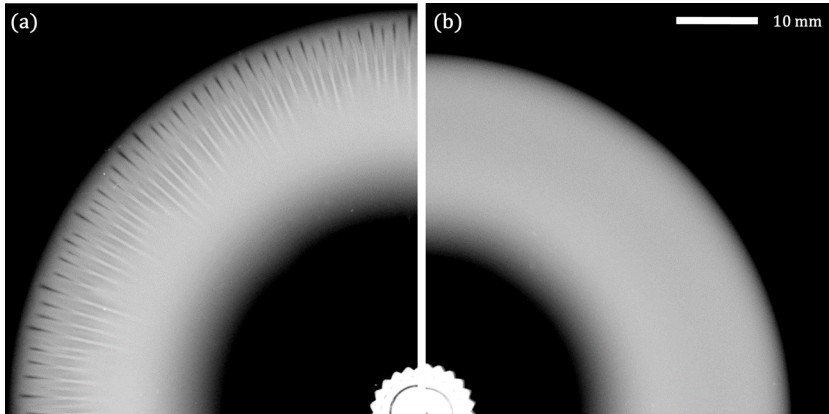


FIG. 3. Comparison of two displacement experiments ($Q = 8.22$ mL/min and $h = 0.6$ mm) at $t = 25$ s under different acceleration values, but otherwise identical experimental parameters: (a) 1g and (b) micro-g.

cell for the ground case because of the contribution of buoyant convection. Here t indicates the time elapsed after the injection started.

As the radial injection progresses in time, the patterns become more complex as a result of the competition between convection, diffusion, reaction, and the velocity decrease that is caused by the radial flow. The convective rolls [Fig. 1(b), visible as fine radial stripes in Fig. 3(a)] are separated by a characteristic wavelength while they move farther from the injection point [4,5]. Hence, new convective rolls start forming to compensate for the dissipation of the preexisting rolls as they travel towards the outer region of the Hele-Shaw cell.

Modulating the gravity level during displacement increases the instability's complexity. In Fig. 4 the injection starts in the absence of gravity [Fig. 4(a)]; later on the gravity level increases from micro-g to 1.8g [Fig. 4(b)], resulting in the emergence of dotted patterns resembling a typical Rayleigh–Taylor instability. As the acceleration level stabilizes to 1g [Fig. 4(c)] and the injection carries on, the convective rolls form again out of the initial pattern as seen in Fig. 4(b). During other experiment runs, an even larger variety of patterns emerged when multiaxial acceleration (i.e., acceleration along two different axes due to the nature of the parabolic flight maneuver [6]) was applied to the Hele-Shaw cell.

This work has provided insights in a previously investigated buoyancy-driven instability [4,5]. In particular, the effect of hyper-g and multiaxial acceleration on the pattern formation was revealed.

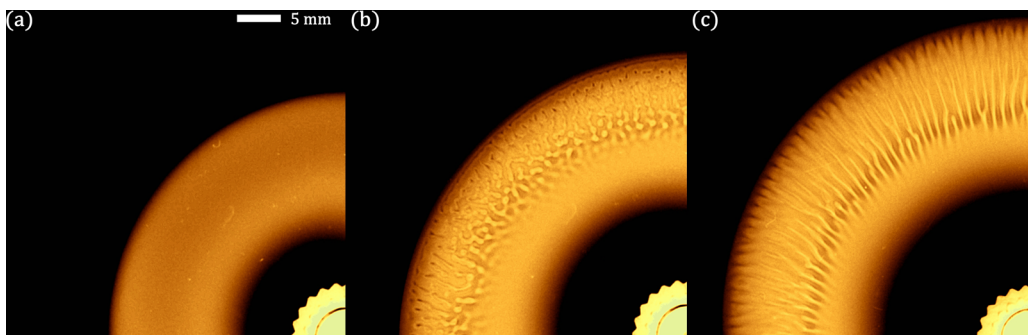


FIG. 4. Evolution of an injection experiment ($Q = 3$ mL/min and $h = 1.0$ mm) under modulating gravity: (a) micro-g, (b) 1.8g, and (c) 1g.

The observation of the system under micro-g confirmed that no instability develops in the absence of buoyancy effects. Future studies under continuous microgravity with longer experimental duration will provide the opportunity to document quantitative trends and scalings during the displacement of miscible reactions fronts.

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