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Formation of hydrogel tubes through air-gap co-extrusion

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The extrusion of a polymer solution in air that falls into a gelling bath allows for forming hydrogel fibers while the air-gap prevents to clog the extruding nozzle. The co-extrusion of a polymer solution and a non-gelling solution in the core then leads to the creation of hydrogel tubes. Here, we use alginate, a natural polyelectrolyte, that turns into gel in presence of divalent cations, like calcium. These tubes having a semi-permeable wall will then be used as bio-reactors in which microbes can evolve in an environment where transport of nutrients, drugs and even cells can be tuned thanks to an interplay between convection and diffusion processes.

For low flow rates, drops are falling out of the nozzle having a millimeter size. However, a liquid bridge can be formed between the nozzle and the bath free surface when the gap between both is below a critical value (Fig. 1 a). The shape and stability of such a dynamic liquid bridge is shown to be a function of the fluid properties, mainly viscosity and surface tension, as well as the flow rate. For a water bath without any gelling agent, we observe that the polymer solution is unable to dynamically penetrate into the bath and spreads on the free surface instead, inertia is thus not efficient enough. However, a tube is formed when the alginate solution is extruded into a water bath containing calcium ions. This suggests that the sol-gel transition occurring at the interface between the two aqueous solutions leads to flow separation and thus the formation of a plunging jet that further solidifies. The size of the hydrogel tube is then a function of the liquid bridge shape, mainly tuned by the gap height, and more importantly of the flow rate. This later behavior is the signature of a competition between convection and diffusion during the solidification of the interface. The morphology of the inner wall of the tube can be tuned by adding, or not, a small amount of calcium ions into the core solution. Indeed, for a pure water core, polymers can diffuse towards the core while gelling occurs during diffusion of calcium ions from the outer medium, thus leading to a diffuse interface (Fig. 1 b). On the other hand, calcium ions from the core solution rigidify the inner interface that finally becomes sharper (Fig. 1 c).

This work is thus an illustration on how the interplay of hydrodynamics, mass transport and physicochemical reaction at interfaces can lead to the shaping of soft material.

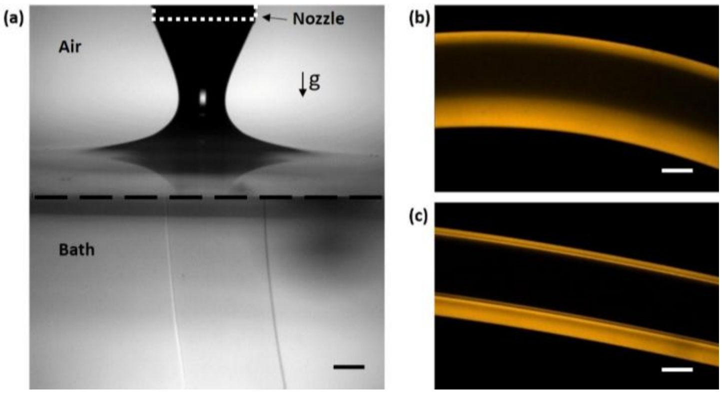

Figure 1: (a) Picture of the liquid bridge in air followed by the tube formation in the gelling bath. (b) and (c) Confocal microscopy images of hydrogel tubes marked with a fluorescent probe, without and with calcium in the core solution, respectively. Scale bars: 1mm.