Revealing the pattern between frontal polymerization and natural convection

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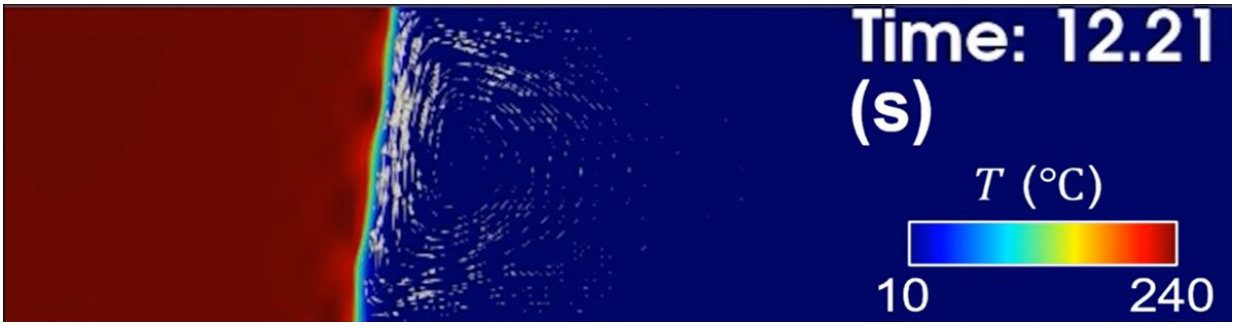

Revealing the pattern between frontal polymerization and natural convection. Credit: The Grainger College of Engineering at University of Illinois Urbana-Champaign

A self-propagating chemical reaction can transform a liquid monomer into a solid polymer, and the interaction between the propagating front and the reaction's natural convection leads to patterns in the resulting solid polymeric material. New University of Illinois Urbana-Champaign work has shown how the coupling between natural convection and frontal polymerization leads to those observed patterns.

This research was led by a unique team of researchers: Materials Science and Engineering professor Nancy Sottos, Aerospace Engineering professor Philippe Geubelle, and Mechanical Science and Engineering professor Leonardo Chamorro. A paper describing this research was

recently published in *Physical Review Letters*.

Thermoset polymers and composite materials are used in a wide range of industries, but producing such materials requires their being cured at high temperatures in a slow and highly energy intensive process. Frontal polymerization to cure the materials is an attractive alternative approach that is significantly faster and more energy efficient.

In frontal polymerization, a self-propagating chemical front converts a liquid monomer into a solid polymer through a reaction that generates a significant amount of heat. Monomers are a simple class of molecular "building blocks" that can react to form larger molecules that are polymers. All of the energy needed to create the polymer is contained within the monomer itself, and to harness that energy, only a small stimulus is needed to kick off the reaction.

Because of instabilities, that self-propagating front doesn't always move uniformly. Although it is ideal for the front to move smoothly and at a constant speed for applications like composite manufacturing and 3D printing, Geubelle says, "We're actually very interested in these instabilities because they allow us to generate patterns in the material. That's very exciting, because for some materials, these instabilities can lead to very different properties for the material."

Geubelle explains that the team's goal was "to understand, experimentally and computationally, the interaction between the front that is propagating in the monomer bath and the convection that takes place ahead of it, and how the interaction between the two can lead to patterns in the material."

To visualize and characterize the polymerization front and the recirculation ahead of the front, the team had to design a clever mold that would allow them to make observations through both the top and the

side. They constructed and used a glass mold that allowed for the observation of the front from the top and for a laser beam to enter from the side.

They then used particle image velocimetry (PIV) to characterize the velocity field. To use PIV, they needed to seed the fluid with small "tracer" particles that would follow the flow and would be tracked by a camera and illuminated by a laser sheet to visualize the patterns in the material. Chamorro says that particle selection was one of the challenges of this work. The team tried various kinds of particles before settling on silver-coated glass particles.

They were able to show that as the front propagates and transforms the liquid monomer into the solid polymer, the energy released generates convection. Convection is a process where heat is transferred by the movement of a heated fluid. Like water in the ocean, when a fluid is heated, it expands, and due to buoyancy, the hotter fluid rises because it is less dense, and colder fluid replaces it by sinking to the bottom because it is more dense. This process continues, creating a recirculating flow.

The polymerization process gives off a lot of heat, resulting in temperatures over 350° F. That heat generated during the transformation goes to the top of the surface. The researchers showed that this was a buoyancy-driven process, and that the recirculation associated with the heat of the reaction, along with the effect of gravity, leads to the formation of the patterning observed in the material and to the impact on the polymerization front. Thanks to the recirculation, the front tends to be inclined rather than perfectly vertical. That inclined front can result in a different speed or cooling effect and even a different patterning effect.

Sottos says the experiments revealed that the recirculation not only creates patterns inside the material that affects the material's properties,

but "it also creates surface patterns on the top of the material as well, because the monomer is getting pushed by the recirculating flow."

The revealed mechanisms of the interaction between the polymerization front and the induced natural convection, and the resulting patterning, represent a deepened understanding of frontal polymerization that may be helpful in the future manufacture of polymeric materials.

Other authors on this work include Yuan Gao (postdoc, Beckman Institute and Aerospace Engineering); Justine Paul (graduate student, Beckman Institute and Material Science and Engineering); Manxin Chen (undergraduate student, Beckman Institute and Aerospace Engineering); and Liu Hong (graduate student, Mechanical Science and Engineering).

More information: Y. Gao et al, Buoyancy-Induced Convection Driven by Frontal Polymerization, *Physical Review Letters* (2023). DOI: 10.1103/PhysRevLett.130.028101

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