Designing soft materials that mimic biological functions

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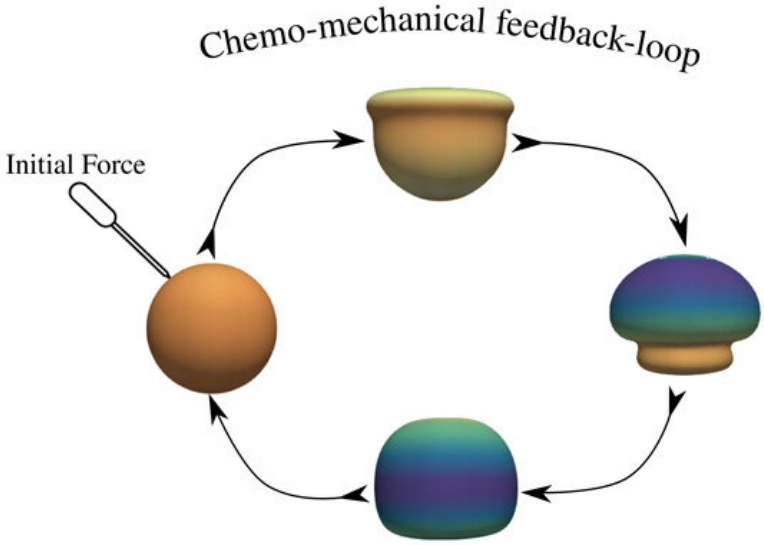

Credit: Northwestern University

Northwestern Engineering researchers have developed a theoretical model to design soft materials that demonstrate autonomous oscillating properties that mimic biological functions. The work could advance the design of responsive materials used to deliver therapeutics as well as for robot-like soft materials that operate autonomously.

The design and synthesis of materials with biological functions require a delicate balance between structural form and physiological function. During embryonic development, for instance, flat sheets of embryonic cells morph through a series of folds into intricate three-dimensional structures such as branches, tubes, and furrows. These, in turn, become dynamic, three-dimensional building blocks for organs performing vital functions like heartbeat, nutrient absorption, or information processing by the nervous system.

Such shape-forming processes, however, are controlled by chemical and mechanical signaling events, which are not fully understood on the microscopic level. To bridge this gap, researchers led by Monica Olvera de la Cruz designed computational and experimental systems that mimic these biological interactions. Hydrogels, a class of hydrophilic polymer materials, have emerged as candidates capable of reproducing shape changes upon chemical and mechanical stimulation observed in nature.

The researchers developed a theoretical model for a hydrogel-based shell that underwent autonomous morphological changes when induced by chemical reactions.

"We found that the chemicals modified the local gel microenvironment, allowing swelling and deswelling of materials via chemo-mechanical stresses in an autonomous manner," said de la Cruz, Lawyer Taylor Professor of Materials Science and Engineering at the McCormick School of Engineering. "This generated dynamic morphological change, including periodic oscillations reminiscent of heartbeats found in living systems."

A paper, titled "Chemically Controlled Pattern Formation in Self-oscillating Elastic Shells," was published March 1 in the journal *PNAS*. Siyu Li and Daniel Matoz-Fernandez, postdoctoral fellows in Olvera de la Cruz's lab, were the paper's co-first authors.

In the study, the researchers designed a chemical-responsive polymeric shell meant to mimic living matter. They applied the water-based mechanical properties of the hydrogel shell to a chemical species, a chemical substance that produces specific patterned behavior—in this case, wave-like oscillations—located within the shell. After conducting a series of reduction-oxidation reactions—a chemical reaction that transfers of electrons between two chemical species—the shell generated microcompartments capable of expanding or contracting, or inducing buckling-unbuckling behavior when mechanical instability was introduced.

"We coupled the mechanical response of the hydrogel to changes in the concentration of the chemical species within the gel as a feedback loop," Matoz-Fernandez said. "If the level of chemicals goes past a certain threshold, water gets absorbed, swelling the gel. When the gel swells, the chemical species gets diluted, triggering chemical processes that expel the gel's water, therefore contracting the gel."

The researchers' model could be used as the basis to develop other soft materials demonstrating diverse, dynamic morphological changes. This could lead to new drug delivery strategies with materials that enhance the rate of diffusion of compartmentalized chemicals or release cargos at specific rates.

"One could, in principle, design catalytic microcompartments that expand and contract to absorb or release components at a specific frequency. This could lead to more targeted, time-based therapeutics to treat disease," Li said.

The work could also inform the future development of soft materials with robot-like functionality that operate autonomously. These 'soft robotics' have emerged as candidates to support chemical production, tools for environmental technologies, or smart biomaterials for

medicine. Yet the materials rely on external stimuli, such as light, to function.

"Our material operates autonomously, so there's no external control involved," Li said. "By 'poking' the shell with a chemical reaction, you trigger the movement."

The researchers plan to build on their findings and further bridge the gap between what's possible in nature and the science lab.

"The long-term goal is to create autonomous hydrogels that can perform complex functions triggered by clues as simple as a local mechanical deformation," Olvera de la Cruz said.

More information: Siyu Li et al, Chemically controlled pattern formation in self-oscillating elastic shells, *Proceedings of the National Academy of Sciences* (2021). DOI: 10.1073/pnas.2025717118

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