New approach to form non-equilibrium structures

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Although most natural and synthetic processes prefer to settle into equilibrium—a state of unchanging balance without potential or energy—it is within the realm of non-equilibrium conditions where new possibilities lie. Non-equilibrium systems experience constant changes in energy and phases, such as temperature fluctuations, freezing and melting, or movement. These conditions allow humans to regulate their body temperature, airplanes to fly, and the Earth to rumble with seismic activity.

But even though these conditions exist naturally and are required for the most basic life, they are rarely understood and difficult to find in synthetic materials.

"In equilibrium thermodynamics, we know everything," said Northwestern University's Igal Szleifer. "Non-equilibrium thermodynamics is an old subject, but we don't have a complete set of rules for it. There are no guidelines."

Szleifer is the Christina Enroth-Cugell Professor of Biomedical Engineering and professor of chemical and biological engineering in Northwestern's McCormick School of Engineering and Applied Science, professor of chemistry in the Weinberg College of Arts and Science, and professor of medicine at the Feinberg School of Medicine.

Szleifer, his postdoctoral fellow Mario Tagliazucchi, and Emily Weiss, the Irving M. Klotz Research Professor of Chemistry at Weinberg, have

developed a new technique for creating non-equilibrium systems, which will bring scientists closer to understanding the fundamentals of the mysterious topic. Their work is described in the paper "Dissipative self-assembly of particles interacting through time-oscillatory potentials," which was featured in the June 23 issue of the *Proceedings of the National Academy of Sciences.*

Past research has shown that theoretical, non-equilibrium particle structures can self-organize when continuously injected with energy, but strategies for injecting energy were limited.

"Think about us as humans," Szleifer said. "For us to be alive, we need to use energy all the time. In order to do that, we have to be out of equilibrium. We are trying to understand non-equilibrium assembly systems, so we have to give them energy."

Using models and simulations, Tagliazucchi, Weiss, and Szleifer found that they could give equilibrium systems energy by using a mixture of pH-responsive particles. Varying pH levels flipped the electric charges of the particles, causing them to oscillate and create the energy needed to assemble into non-equilibrium structures.

"By controlling the structure of the material, we can control its properties as well," Szleifer said. "The moment you stop the oscillations, the structure disappears."

The oscillatory method has allowed Szleifer and his collaborators to create novel structures that are impossible to find in equilibrium conditions. He said scientists could potentially determine how they want particles to interact and then tailor oscillations to lead to that outcome.

"For a number of years, my group has tried to find rules for self assembly," Szleifer said. "This is building toward that. We want to make

guidelines for experimentalists."

Provided by Northwestern University

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