Study reveals how polymers relax after stressful processing

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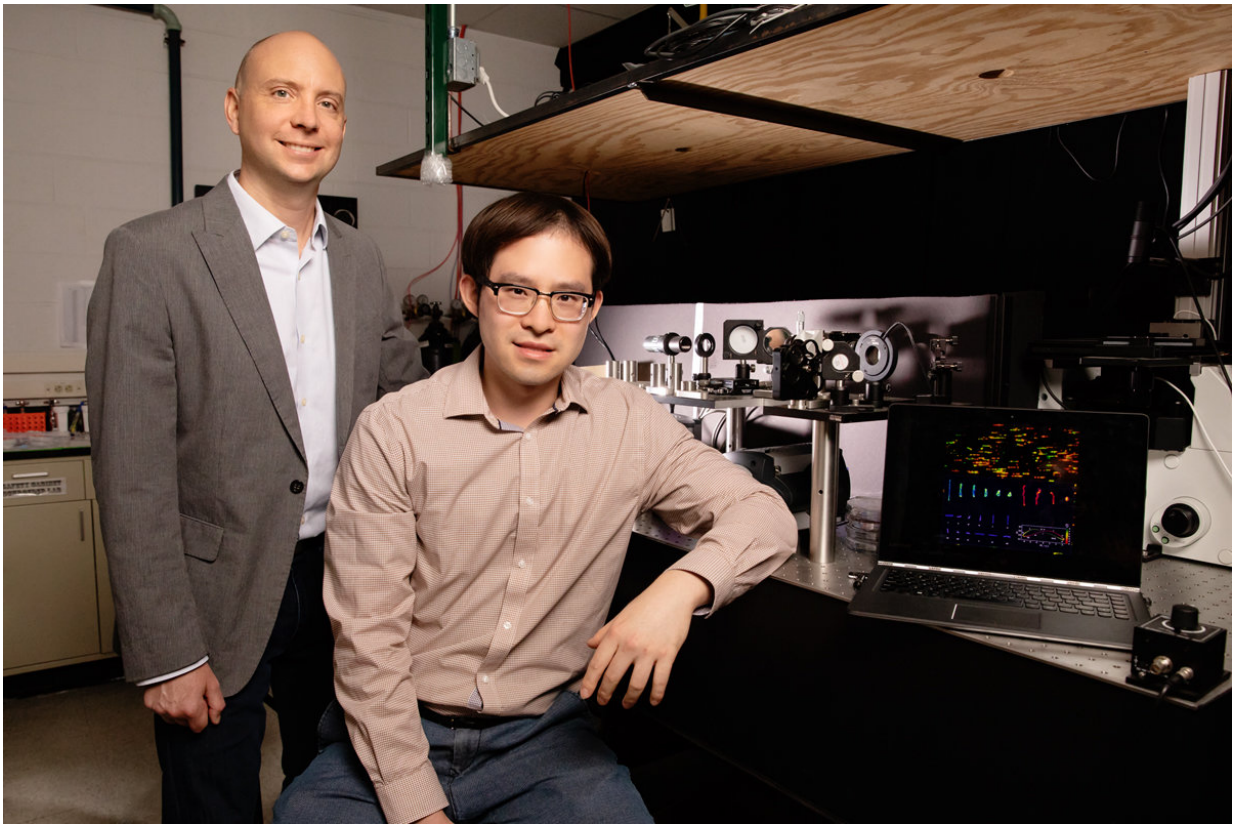

Chemical and biomolecular engineering professor Charles Schroeder, left, and graduate student Peter Zhou have found that single polymers -- acting as individuals -- work together to give synthetic materials macroscopic properties like viscosity and strength. Credit: L. Brian Stauffer

The polymers that make up synthetic materials need time to de-stress

after processing, researchers said. A new study has found that entangled, long-chain polymers in solutions relax at two different rates, marking an advancement in fundamental polymer physics. The findings will provide a better understanding of the physical properties of polymeric materials and critical new insight to how individual polymer molecules respond to high-stress processing conditions.

The study, published in the journal *Physical Review Letters*, could help improve synthetic materials manufacturing and has applications in biology, mechanical and materials sciences as well as condensed matter physics.

"Our single-molecule experiments show that polymers like to show off their individualistic behavior, which has revealed unexpected and striking heterogeneous dynamics in entangled polymer solutions," said co-author Charles Schroeder, a professor of chemical and biomolecular engineering and faculty member of the Beckman Institute for Advanced Science and Technology at the University of Illinois at Urbana-Champaign. "A main goal of our research is to understand how single polymers—acting as individuals—work together to give materials macroscopic properties such as viscosity and toughness."

Using a technique called single-molecule fluorescence microscopy, researchers can watch—in real time—as individual polymer molecules relax after the stretching, pulling and squeezing of the manufacturing process. "Imagine looking into a bowl of cooked spaghetti and watching the motion of a single noodle as the bowl is mixed," Schroeder said.

"We found that the polymers exhibit one of two distinct relaxation modes," said co-author and graduate student Yuecheng (Peter) Zhou. "One group of polymers relaxed via a single decaying exponential rate and the other group showed a two-phase process. That second population undergoes a very quick initial retraction followed by a slow relaxation.

The existence of two different molecular populations was unexpected and not predicted by classic theory."

This study worked with high molecular-weight DNA because it serves as an ideal model of other types of synthetic organic polymers, the researchers said.

"We chose DNA as our model polymer because it is a very large molecule and the chains are big enough to image in our microscope," Schroeder said. "They are also all the same weight, which provided us with a very clean, well-defined system for data analysis."

The researchers found that the percentage of the molecular subpopulation that exhibits the two-phase relaxation behavior increases as the overall polymer concentration increases in the entangled solutions.

"We are not certain why the single-mode relaxation or fast-retraction mode seems to be concentration-dependent, but it may have to do with enhanced interpolymer friction—the more polymers, the higher the chance they will interact, especially out of equilibrium," Zhou said. "We are working with theorists here at the University of Illinois to better explain the single-mode and two-mode relaxation phenomena."

The team is excited to bring new insight to the understanding of how complex fluids flow and how they are processed and manufactured, especially with polymers that are subjected to intense stress, such as the fluids that are used for 3-D printing.

More information: "Dynamically heterogeneous relaxation of entangled polymer chains" *Physical Review Letters* (2018). journals.aps.org/prl/abstract/ … ysRevLett.120.267801

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