

When it comes to polymer fragility, size does matter

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Polymers are very large molecules consisting of thousands, even millions, of atoms bonded together in a repeating pattern similar to a chain. They make up many of the things around us we consider part of our everyday lives, from bottles and tires to airplanes and medical devices. Understanding what gives polymers their unique properties is helpful in developing new functional materials for various current and future technologies. A multinational team of researchers have brought together expertise and experimental results over years to help to explain the extremely strong temperature dependence of viscoelastic properties of polymers melts, a mystery that had thus far evaded explanation.

Fragility index is a parameter that quantifies how fast the material transforms from a solid to a liquid with temperature increase. For many years the higher fragility of polymers as compared to small molecules has been well documented. Many polymers exhibit fragility index approximately 1.5 times higher than even the most fragile, small molecular liquids and until now, there has been no clear answer as to why this is the case.

By combining a number of tools and techniques, a team of researchers from the U.S., Italy and China was able to find a more complete picture of the glass transition phenomenon in polymers and to point out where the polymers differ from small molecular liquids. The researchers explain their findings this week in *The Journal of Chemical Physics*.

"We worked on this problem with our colleagues for a long time and



though our paper with the similar title, 'Why many polymers are so fragile?' was published in 2007, we could only formulate the problem, we had no answer," explained Alexei P. Sokolov, a research scientist at Oak Ridge National Laboratory and professor of Chemistry and Physics at the University of Tennessee. "Over the years we accumulated many <u>experimental results</u> obtained by many different techniques (this is why the paper has so many authors) on a model <u>polymer</u> polystyrene to come up with this idea." This provided the broad view of many polymer specific properties needed to figure out what was missing. Using polystyrene with various chain lengths, researchers correlated many of their properties to their fragility and demonstrated that these correlations work for short chains but progressively fail when the length, i.e. the number of repeated units or segments, increases. The work may finally resolves this puzzle.

The researchers realized that the segmental (also called structural) relaxation in the case of polymers presents the relaxation of only a small part of the molecule. For polymers the complete molecular scale relaxation happens only on a much longer time scale that corresponds to chain relaxation. They show that analysis of the chain relaxation instead of relaxation of segments restores all the correlations characteristic for non-polymeric systems. This discovery leads to a new way of looking on the problem.

What does this mean for the polymers that are part of our daily life?

"Our work has broader implications, because similar mechanisms may account for rather high fragility of other complex systems in soft condensed matter," Sokolov said. "Whether this will help to make better polymers remains to be seen, but it should help in the design of polymers with the desired viscoelastic properties."

More information: "Why many polymers are so fragile: a new



perspective," The Journal of Chemical Physics, DOI: 10.1063/1.4964362

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