DMR-0097202 Solid-State NMR Analysis of Chain Packing and Dynamics in Polycarbonates

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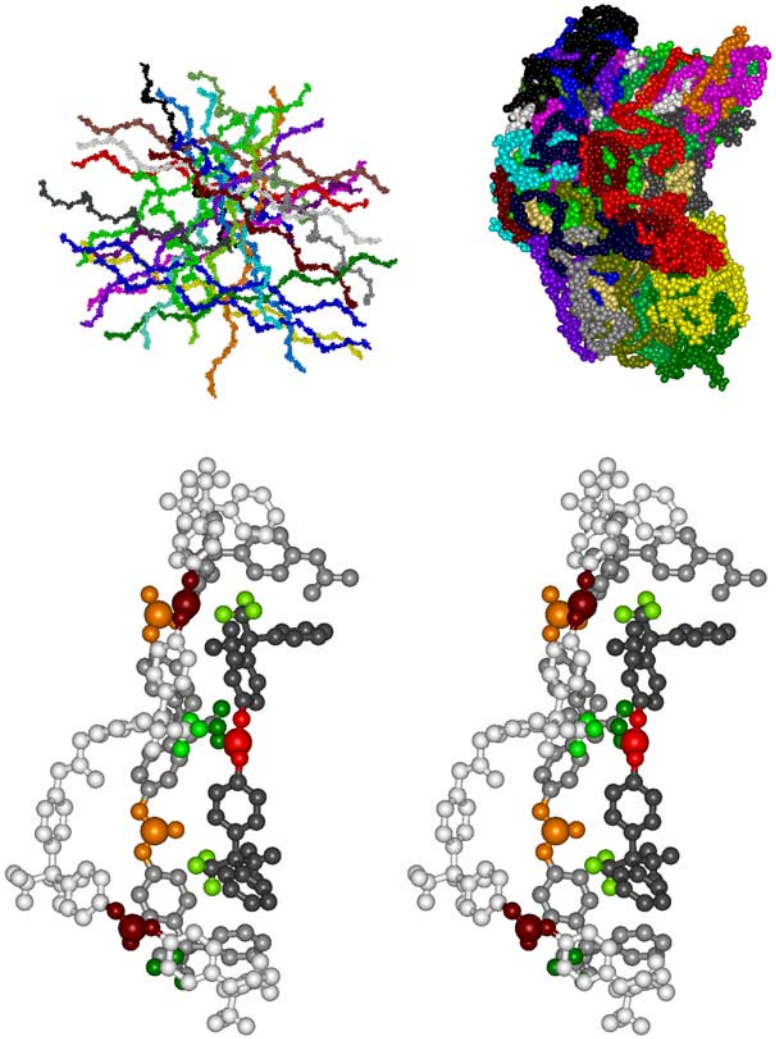

We have used rotational-echo double resonance (REDOR) solid-state NMR to measure the orientation and distance between two polycarbonate chains, one labeled with ^{13}C and the other with ^{19}F. These measurements show unambiguously a preference for the isopropylidenes of one chain to be proximate to the carbonates of the nearest-neighbor chain. We have now been able to account for this preference using a "collapsed-chain" model. The model results from molecular dynamics calculations performed on a collection of extended 25-mer chains (top left), including evolution first in vacuum and then under a pressure constraint to reach the experimental bulk density (top right, expanded distance scale). Even with no imposed REDOR restraints, local order results, with the chains folded such that isopropylidenes are proximate to carbonates (stereo view at bottom). An attractive feature of this modeling is that there is no starting bias.

Broader Impact

Polycarbonates are candidates for holographic thin films. They are mechanically tough, have high glass-transition temperatures, and can be synthesized with photosensitive sidechains. The characterization of their packing and local dynamics by REDOR and modeling should help in the development of new and useful data-storage materials.

Outreach and Education

Polymers and materials science are good topics to use in reaching out to inform the community about modern technology. Polymers are intuitive and do not require the background necessary to understand structural biology, for example. The PI is an active participant in a formal education training program for St. Louis K-8 science teachers. He applies the basic ideas of mass spectrometry (see figure below) and nuclear magnetic resonance to familiar objects like plastics (especially impact-resistant polycarbonates!).

A demonstration of the principles of a mass spectrometer. A magnet deflects a steel ball rolling down an inclined cardboard plane. The magnet is taped below the cardboard