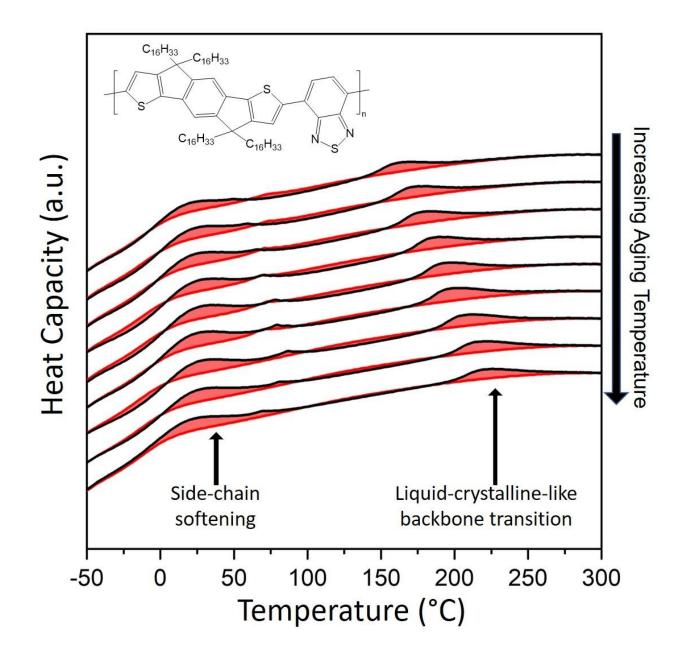
Understanding the impact of polymer chain rigidity on the assembly, structure, and photo-physical processes

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The field of polymer-based electronics has witnessed major developments in the past few years that have led to systems of vastly improved charge transport- and energy-harvesting properties. This progress can be predominantly attributed to synthetic efforts in the form of the creation of new materials, which often comprise backbones of a significantly more rigid nature than the first generation polymer semiconductors and most bulk commodity plastic. Moreover, many semiconducting polymer frequently lack significant long-range order, but it is hypothesized that they may exhibit liquidcrystalline-like behavior because of their hairy-rod nature. To understand the polymer phase behavior, how it relates to chemical design and how it dictates important optoelectronic features, we use fast scanning calorimetry to identify the glass transition and possible liquid-crystalline-like transitions, as well as side-chain softening regimes, using physical aging signatures and focusing on poly(indacenodithiophene)s and other next-generation semiconducting polymers. By pairing the thermodynamic data with 2D coherent excitation spectroscopy, we can relate information of the very local polymer order (backbone torsion, structural dynamics introduced by side-chain softening) on exciton coherence and charge transport properties. This approach allows us to gain insights on the role of polymer assembly and solid-state structure on energetic disorder and photophysical characteristics towards the delivery of important structure-property interrelations for design of fourth generation semiconducting plastics for organic solar cells, plastic electronics, wearable sensors, and beyond.



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