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Title:	Synthesis and Processing Strategies to Tune the Film Structure and Optoelectronic Properties of Non-Planar Molecular Semiconductors
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Abstract:	<p>Molecular semiconductors have generated significant interest for their potential use in lightweight and mechanically flexible electronic devices. Yet, predicting how new molecular semiconductors will perform in devices remains a challenge because devices are comprised of polycrystalline thin films of molecular semiconductors, and charge transport in these films depends greatly on the details of their microstructure whose heterogeneities can span multiple length scales. The microstructure typically evolves during deposition, and thus developing organic electronics not only hinges on the success of materials discovery, but also on the ability to fine-tune deposition and processing parameters to access the thin-film structure most conducive for charge transport. This thesis explores chemical modification of a non-planar organic semiconductor, contorted hexabenzocoronene, cHBC, to tune its optoelectronic properties and processing strategies to induce structural changes in thin films. We primarily explore fluorine- and chlorine-substitution at the peripheral aromatic rings of cHBC to lower its energy levels and optical bandgap, and we demonstrate such halogenated derivatives as electron acceptors in organic solar cells. Substitution with these larger atoms also increases cHBC's intramolecular steric hindrance, providing access to an alternative molecular conformation with an order of magnitude higher solubility and systematic shifts in absorption and emission characteristics. cHBC's non-planarity provides an added dimension of tunability as it frustrates crystallization during deposition, producing amorphous films that can be subsequently crystallized with post-deposition processing. Decoupling structural development from deposition allows us to fabricate transistors from differently treated cHBC films and elucidate the effects of changes in film structure on charge transport, as measured by the field-effect mobility. With different processing, the extent of cHBC oriented out-of-plane can be increased, which results in up to an order-of-magnitude increase in mobility. Also by solvent-vapor annealing with different solvents and applying solvent-vapor and thermal annealing in alternating sequences, three polymorphs of cHBC can be accessed whose mobilities differ by up to an order-of-magnitude. Films of fluorinated cHBC derivatives can also access two of these polymorphs depending on the solvent employed during solvent-vapor annealing. With a data-mining approach, we identify the solvent properties dictating polymorph selection to help guide processing for this family of compounds.</p>
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