The University of Fribourg / Faculty of Sciences and Medicine / Department of Chemistry

Exploring and Exploiting Phase Segregation in Hydrogen-Bonded Supramolecular Polymers

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Hydrogen bonded supramolecular polymers represent a large subdivision of supramolecular polymers under the focus of the scientific community. Hydrogen bonds are relatively weak, and using supramolecular motifs having low number of binding sites and thus low association constants, lead to low melt viscosities. In this approach, phase segregation can be used to improve the solid state mechanical properties. In addition, when the conditions are favorable, it is possible to observe the crystallization of the hard phase containing the supramolecular motif, which further improves the mechanical properties. The present thesis reports materials synthesized *via* this approach and which are based on a low association constant supramolecular motif, *i.e.,* the hydrogen-bonded motif formed by isophthalic acid (IPA) and a complementary pyridine (Py) unit. This thesis deals both with main-chain (chain extension of an IPA-terminated telechelic with bipyridines) and side-chain (chain extension of side-chain polymers bearing IPA pendant groups with bipyridines) supramolecular polymers displaying IPA-Py reversible links.

The first study of this thesis focuses on understanding the influence that subtle variations within the structure of the IPA-Py supramolecular motif have on the nanoscale phase segregation and crystallization of main-chain supramolecular polymers. The application of these materials as adhesives is investigated in the second study of this thesis. The last study of this thesis presents different approaches followed to insert IPA units as side-groups in polymers and mixing the resulting polymers with bipyridines with the aim of synthesizing supramolecular networks. Finally, general conclusions of these studies are drawn at the end of this thesis and an outlook of possible follow-up studies is presented.

Jury:

Prof. Dr. Christoph Weder (thesis director and supervisor)
Dr. Lucas Montero de Espinosa (internal expert)
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Prof. Dr. Michael Mayer (president of the jury)