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Study on Triblock Copolymers: From Relaxation Dynamics of Solution to Toughening Mechanism of Physical Double Network Hydrogels

(トリブロック共重合体に関する研究：溶液緩和のダイナミクスから ダブルネットワーク物理ゲルの高靭性化原理まで)

Hydrogels composed of three-dimensional polymer network and abundant of water are a class of typical soft and wet matters, which are used for numerous applications including force-sensitive adhesives, scaffolds of tissue engineering, gas filtration membranes, drug-controlled release and ionic conductive media. However, traditional hydrogels are mechanically weak, which limits their application severely. Recently, several tough and self-recoverable hydrogels were developed by introducing physical association as sacrificial bonds to dissipate energy, which widely extend the scope of applications even as structural biomaterials, like artificial cartilage and ligament. Despite great success in creation of tough and self-recoverable hydrogels, the developments generally relied on an empirical and trial-and-error approach as the understanding of structure and toughening mechanism of those gels is still unclear.

This thesis focuses on revealing the structure and underlying toughening mechanism of tough and self-recoverable hydrogels. Recently developed hydrogels based on self-assembly of ABA type triblock copolymer were chosen as a model system, which have been demonstrated to possess excellent mechanical property and self-recoverable capability. These gels are prepared in sequential steps: (i) firstly the triblock copolymer poly(butyl methacrylate)-*b*-poly(methacrylic acid)-*b*-poly(butyl methacrylate), PBMA-PMAA-PBMA, was dissolved in dimethylformamide (DMF) to form precursor solution; (ii) then the precursor solution undergoes a solvent exchange process along with self-assembly of endblock PBMA into micelles acting as cross-linking to form B gels; (iii) finally the second polymer, polyacrylamide (PAAm) was introduced to form sacrificial hydrogen bonds between the carboxy group of PMAA and amino group of PAAm, and to generate the tough and self-recoverable hydrogels, named as B-DN gel. In this work, we systematically studied the preparation process and the structure-property relationship of those gels, including relaxation dynamic of precursor solution, contribution of physical crosslinks and trapped entanglements in B-gels, the effect of molecular parameters on self-assembly structure of B-gels, and the structure-property relationship of B-DN gels. Benefiting from the thoroughly understanding of chain structure and self-assembly structure in gelation process and final gels, we successfully developed thin but ultra-tough and 100% recoverable hydrogel membranes with high pH sensitivity, excellent mechanical properties and biocompatibility, which is urgently needed in many fields but rarely realized in hydrogels before.