# Doctoral Dissertation Evaluation Review

Degree requested Doctor of Life Science Applicant's name YANAN YE

Examiner:

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Title of Doctoral Dissertation

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)

Results of Evaluation of the Doctoral Dissertation (Report)

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 interaction 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 very limited.

This thesis focuses on revealing the structure and underlying toughening mechanism of tough and self-recoverable hydrogels. The tough hydrogels recently developed 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. In this work, the author systematically studied the preparation process and the structure-property relationship of those gels, including relaxation dynamic of precursor solution, topological contribution of physical crosslinks and trapped entanglements in the triblock copolymer gels (B-gels), the effect of molecular parameters on self-assembly structure of B-gels, and the structure-property relationship of double network gels with the B-gels as the first network (B-DN gels). The author revealed the B-DN gels having hierarchical structure. Furthermore, based on the structure evolution during deformation, the author clarified that the high toughness of self-recoverable B-DN gels is a synergistic effect of multi-step structure breakage to dissipate energy. In addition, the author explored the application of such tough physical hydrogels by developing ultra-tough and thin (10-100 μm) B-DN gel membranes. The membranes exhibit excellent mechanical properties superior to those of various biological membranes, biocompatibility, and postoperative anti-adhesive property, foreshadowing their potential use as substitutes of biological membranes or as postoperative anti-adhesive membrane.

In conclusion, using B-DN physical gels as a model system, the author has revealed the hierarchical structure of B-DN gels originating from coexistence of hydrophobic interaction and hydrogen bonding, and clarified the toughening mechanism of B-DN gel based on a multi-step structure breakage. These works will help to understand the structure and toughening mechanism of other physical tough hydrogels, to design next generation tough materials, and to guide practical application.

Therefore, we acknowledge that the author is qualified to be granted a Doctorate of Life Science from Hokkaido University.