



Title	A Novel Polyampholyte Hydrogel Based on Dynamic and Reversible Sacrificial Bonds : Toughness and Self-healing [an abstract of dissertation and a summary of dissertation review]
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学位論文審査の要旨  
Doctoral Dissertation Evaluation Review

博士の専攻分野の名称 博士 (生命科学) 氏名 Abu Bin Ihsan  
Degree requested: Doctor of (Life Science) Name:

審査担当者	主査 / Chief examiner	教授	龔 劍萍
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	副査 / Associate examiner	准教授	黒川 孝幸

学位論文題名  
Title of Doctoral Dissertation

A Novel Polyampholyte Hydrogel Based on Dynamic and Reversible Sacrificial Bonds:  
Toughness and Self-healing

(動的・可逆な犠牲結合を用いた新規両性イオン性ハイドロゲル：その高い靱性と自己修復特性)

博士学位論文審査等の結果について (報告)

Results of Evaluation of the Doctoral Dissertation (Report)

Polyampholytes are polyelectrolytes carrying randomly distributed opposite charges. A neutral polyampholyte of equal amounts of opposite charges, due to intra-chain ionic bond formation, forms a globule structure and precipitates in dilute aqueous solution of low ionic strength. Chemically cross-linked polyampholyte hydrogels deswell near the equal charge composition. Recently, polyampholytes have been discovered to form tough physical hydrogels by employing the ionic bond as a reversible sacrificial bond that breaks and reforms dynamically. Owing to the random distribution, the opposite charges on the polyampholytes form multiple ionic bonds of wide distribution in strength. The strong bonds work as permanent cross-linkers, imparting the elasticity of the hydrogel. The weak bonds are fragile and they break under stress to dissipate energy, serving as reversible sacrificial bonds.

In this study, the author attempt to design polymer hydrogels of multiple high mechanical performances, including a proper modulus, high strength and toughness, high fatigue resistance, and self-healing, based on dynamic reversible sacrificial bond. A novel tough and self-healing polyampholyte hydrogel based on strong bond/weak bond concept has been developed by random copolymerization of two oppositely charged monomers, sodium *p*-styrenesulfonate (NaSS) and acryloyloxethyltrimethylammonium chloride (DMAEA-Q); and systematic investigation on the behavior of this supramolecular system, viz., poly(NaSS-*co*-DMAEA-Q) without and with a slight chemical cross-linking has been performed. A phase diagram of mechanical strength has been constructed in the space of monomer concentration  $C_m$  and cross-linker density  $C_{MBAA}$ , and very tough phase has been discovered. These tough polyampholyte hydrogels exhibit excellent healing efficiency (~ 84% on average and maximum 99%) at room temperature without any external stimuli. The effects of healing temperature, aging time, and the modulus of the gels on the healing efficiency have been clarified. The self-healing mechanism is revealed.

In conclusion, this work may give insight to design supramolecular hydrogels with high toughness and self-healing efficiency. The novel polyampholyte hydrogel have a great potential as a novel tough and self-healing material. Therefore, we acknowledge that the author is qualified to be granted the Doctorate of (Life Science) from Hokkaido University.