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Author(s)	尹, 海燕
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学 位 論 文 審 査 の 要 旨

博士の専攻分野の名称 博士 (理 学) 氏 名 尹 海 燕

主 査 教 授 龔 劍 萍
審査担当者 副 査 教 授 玉 置 信 之
副 査 准教授 黒 川 孝 幸 (生命科学院)

学 位 論 文 題 名

Synthesis of High Strength Polyzwitterionic Hydrogels: from Polyion Complex to Double Network Structure
(両性イオン高分子を用いた高強度ハイドロゲルの合成—ポリイオンコンプレックス構造からダブルネットワーク構造へ)

Polyzwitterionic materials have both cationic and anionic groups in the single polymeric repeat unit. Such special structure leads to unique biological properties; *i.e.* biocompatibility and anti-biofouling property due to formation of hydration layer on their surface, and making ionic complex with DNA toward gene carriers. Thus, biomedical application of polyzwitterions has been the subject of numerous studies. Especially, polyzwitterionic hydrogels have attracted much interest as artificial soft tissue due to their mechanical compatibility (modulus matching) with other soft tissues. However, conventional polyzwitterionic hydrogels were so brittle mainly due to their highly water swollen structure, which greatly limited their applications as biomaterials.

This study, for the first time, reports successful synthesis of novel polyzwitterionic hydrogels with high mechanical strength toward their application as mechanically-robust biomaterials. In order to fabricate high mechanical strength of polyzwitterionic hydrogels, firstly author confirms the properties of single network novel polyzwitterionic hydrogel using N-carboxymethyl-N,N-dimethyl(methacryloyloxy) ethanaminium, inner salt (CDME). Although the single network PCDME hydrogels contain both anionic and cationic groups, they behave like neutral hydrogels, confirmed by the relationship between the Young's modulus and swelling degree as well as their behavior in NaCl aqueous solution. On the other hand, these hydrogels behave like polyelectrolyte in acidic environment. However, single network PCDME hydrogel is mechanically too weak to be applied as load-bearing biomaterials. Therefore, author applied two methods to fabricate the mechanically strong PCDME hydrogel based on properties of the single network PCDME hydrogel which shows property of positively charged polyelectrolyte in strong acid condition and neutral property in water. One method is that the PCDME was combined with negatively charged polyelectrolyte at the strong acid condition to obtain mechanically strong polyion complex hydrogels. Other method is applying the double network (DN) concept to prepare the tough polyzwitterionic hydrogels. Furthermore, author revealed that the properties of the interpenetrating network hydrogels consisting of negatively charged polyelectrolyte as the first network and PCDME as the second network changes from polyion complex-like ones to double network-like ones with increasing molar ratio of PCDME repeat unit to that of PAMPS first network. Author clarified that adjustment of molar ratio of the two networks is crucially important for obtaining desired mechanical properties of the polyzwitterionic hydrogels.

In conclusion, the author has developed different structure of tough hydrogels such as polyion complex and double network structure from one novel zwitterionic polymer, which have potential application as biomaterials, such as cartilages and blood vessel. Therefore, we acknowledge that the author is qualified to be granted the doctorate of science from Hokkaido University.