学 位 論 文 の 要 約

博士の専攻分野の名称　博士（理　学）　氏　名　　尹　　海燕

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 the 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.

In this study I attempt to synthesize novel polyzwitterionic hydrogels with high mechanical strength toward their application as mechanically-robust biomaterials. Prior to the main topic, in chapter 2, I synthesized the 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. In addition, it was confirmed that the PCDME hydrogel shows excellent anti-biofouling property.

Based on these fundamental researches, in chapters 3 and 4, I synthesized novel polyzwitterionic hydrogels with high mechanical strength. In chapter 3, I applied the double network (DN) concept to prepare the tough polyzwitterionic hydrogels. Using PCDME gels as the second network of the DN gels, I have successfully fabricated the tough polyzwitterionic DN gels. These DN gels showed excellent mechanical properties, similar to those of conventional DN gels. It has been also revealed that the polyzwitterionic DN gels still keep excellent anti-biofouling properties.

In chapter 4, based on the fact that the single network PCDME hydrogel in strong acid condition shows property of positively charged polyelectrolyte, I combined the PCDME with negatively charged polyelectrolyte at the strong acid condition to obtain mechanically-strong polyion complex hydrogels. I confirmed that these two kinds of polymers actually form interpolymer ionic bond, and the resulting hydrogels showed excellent mechanical properties and, furthermore, 100% self-recovery upon internal damage.

Furthermore, in chapter 5, I 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 to the first network. I clarified that adjustment of molar ratio of the two networks is crucially important for obtaining desired mechanical properties of the polyzwitterionic hydrogels.

These results demonstrate that the novel tough and biocompatible polyzwitterionc hydrogels have a high potential as a novel soft and wet biomaterials.