学　位　論　文　の　要　約
Summary of Doctoral Dissertation

博士の専攻分野の名称	博士（生命科学）	氏　名
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学　位　論　文　題　名
Title of Doctoral Dissertation

Study on frictional properties of zwitterionic hydrogels
（両性イオン性ゲルの表面摩擦に関する研究）

Polyzwitterionic materials, which have both cationic and anionic groups in each repeating unit of the polymer, are drawing great attention as biomaterials due to their excellent anti-biofouling properties and biocompatibility. Recently a zwitterion based tough double network (DN) hydrogel has been developed by using poly(*N*-(carboxymethyl)-*N*,*N*-dimethyl-2-(methacryloyloxy) ethanaminium, inner salt) (PCDME). The DN hydrogel using the zwitterionic polymer as the second network exhibits excellent anti-biofouling properties in addition to high mechanical strength and toughness. This promises a great potential of the zwitterion-based hydrogels as better candidate for bio-application, for example, as coating materials of low-friction biomedical devices and implants. Understanding the surface frictional properties of the zwitterion hydrogels is indispensable for these potential applications. In this thesis, the surface sliding friction of zwitterionic hydorgel PCDME was systematically studies. It has been revealed that zwitterionic PCDME hydrogel remains stable of its swelling and mechanical properties over a wide range of pH and ionic strength, which makes it suitable for frictional study under different conditions. The thesis consists of 5 chapters. In **Chapter 1**, some general introductions are discussed. **Chapter 2** enlightens a review on friction of hydrogel and advancement of finding their mechanism.

In **Chapter 3** the effect of macroscopic contact between the gel and substrate is explored. The frictional tests were carried out in aqueous solution using a rheometer with parallel plate geometry and the interfacial contact during the measurement was monitored by a laboratory-made optical system, which gives information on the evolution of gel contact to the glass during sliding motion. The frictional stress on glass was found to be high in water and it showed a strong dependence on interfacial contact. The real time contact images showed that pre-existing water layer at the interface facilitates forced wetting and reduces frictional contact area, which in tern reduces the friction. Again, a very weak dependence on pressure was found as long as the two sliding surfaces were in complete contact. The results might be helpful for understanding the lubrication mechanism of soft materials.

In **Chapter 4** the influence of surface chemistry of both surfaces are explored. Hydrogels with different swelling degree were prepared by modifying the crosslinking density and were swelled in aqueous solution of various pH and ionic strength. The results performed in solutions with varied ionic strength revealed that the high friction on glass substrates has an electrostatic origin. The electrostatic potential measurement showed that the PCDME gels have an isoelectric point at pH 8.5. Since the glass substrates carrying negative charges in pure water, the gel and the glass have electrostatic attraction in water. Study on the effect of pH has shown that below pH 8.5, attraction between the positively charged gels and negatively charged glass gives high friction, while above pH 8.5, the electrical double layer repulsion between two negatively charged surfaces gives low friction. A series of frictional tests carried out with hydrogels with different swelling in water at pH 6.8 showed that the friction is related to the swelling degree through the surface polymer chain density of the hydrogels. Measurement of surface potential of these hydrogels revealed that in water at pH 6.8 the electrostatic potential governed by the surface polymer chain density of hydrogel creates the attractive force during sliding and frictional stress is linearly related to the surface potential. However, glass surface contains a plenty of negative charges originated from SiO^- groups. The charge density on the glass surfaces was modified by coating them with hydrophobic silane binding agent in various extents. The measurement of contact angle of water to the glass substrates with varied hydrophobic coating and calculation of their charge density on the surface showed that the hydrophobic coating reduces the exposed charges on the surface. Frictional tests carried out with these glasses showed that the frictional stress is linearly related to the charge density on the glass surface. From these results, it is concluded that although the PCDME gels behave like neutral gels in the bulk properties, their surface properties sensitively change with pH and ionic strength of the medium. The study on the friction of zwitterionic hydrogels might be helpful for understanding and finding new approaches for designing low-friction biomedical devices and implants.

In **Chapter 5** the concluding remarks are included based on the overall work.