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　Jamil Ahmed
Degree requested: Doctor of （Life Science）　　Name:

審査担当者	主査 / Chief examiner	教　授	龔　剣萍
Examiners	副査 / Associate examiner	教　授	芳賀　永
	副査 / Associate examiner	特任教授	佐々木　直樹
	副査 / Associate examiner	准 教 授	黒川　孝幸

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）

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)

Zwitterionic polymers are polyelectrolytes that contain cations and anions in the same repeating unit. Recently, the zwitterionic polymers have been found to have excellent anti-biofouling properties and non-toxicity, which made them suitable for many bio medical applications. Thus, these polymers have drawn a great attention of many scientists. Until now studies have been focused on zwitterionic polymers and there is very few studies on zwitterionic hydrogels. 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 good 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. 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.

In this thesis, the surface sliding friction of zwitterionic hydorgel PCDME was systematically studied. 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. It was found that at full contact the friction is mainly due to the elastic stretching of the adsorbed polymer chain. With the obtained knowledge of the effect of interfacial contact, the influence of surface chemistry of both surfaces was 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 of different swelling degrees 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. However, through the modification of the negatively charged glass substrate with hydrophobic silane binding agent at various extents, calculation of charge density on their surfaces and friction study of the hydrogel against them, it was found that the frictional stress is linearly related to the charge density on the glass surface.

In conclusion, the author has found 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 friction of the gels is governed by the electrostatic interaction. These new findings on the sliding friction of zwitterionic hydrogels will be helpful for understanding and finding new approaches for designing low-friction biomedical devices and implants. Therefore, we acknowledge that the author is qualified to be granted the Doctorate of Life Science from Hokkaido University.