Doctoral Dissertation Evaluation Review

Degree requested Doctor of Life Science Applicant's name MUHAMMAD ILYAS

Examiner:

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Title of Doctoral Dissertation

Synthesis of Large Area Bilayer-hybrid Gels and Exploration of Novel Functions
（大面積二分子膜ハイブリッドゲルの合成とその新機能探索）

Results of Evaluation of the Doctoral Dissertation (Report)

As a vital class of soft and wet materials, polymeric hydrogels have shown great potential as model substances to study the properties of biological tissues and also as biomaterials for developing artificial organs. Although the invention of tough hydrogels comparable to human cartilages greatly promotes such potential uses of hydrogels, however, conventional tough hydrogels have isotropic structure and properties, which is in contrast to those of natural biological tissues that have anisotropic structure and properties. Developing tough and anisotropic hydrogel-based materials similar to soft load-bearing tissues is strongly desirable but still challenging. Recently, novel soft bilayer based hydrogels have been successfully developed by inducing amphiphilic molecules that form rigid bilayers into conventional PAAm hydrogels. The new bilayer-hybrid gels are anisotropic in structure and show interesting functions such as structural color, toughness and stimuli responsive properties. This promises great potential of the bilayer-based hydrogels for diversified application such as soft color sensor in body. However, the synthesis of bilayer-hybrid gel in good quality and in large area is still challenging. Overcoming this problem is crucial for the in-depth investigation of their structure-properties as well as exploration of novel functions. This thesis aims to establish a method to fabricate high quality bilayer-based hydrogels thin films in large area and explore novel functions of bilayer-hybrid gels.

In this thesis, first, the author systematically studied the key factors that govern the structure of bilayer in the hydrogels and clarified that, the incubation and shear flow conditions for the precursor solution are important for controlling the quality of bilayer-hybrid hydrogels. Based on these findings, the author developed a synthesizing system that could well-control these conditions and obtained high quality bilayer-based hydrogels with good reproducibility, controlled thickness, and large area.

Then, the author systematically studied the structure-property relationship of the gels and discovered various interesting functions of the gel. Especially, the author controlled the water content by confined drying the anisotropic gel and found quite unique behavior of the bilayer-hybrid hydrogels with the change of water content. Specifically, the author revealed that the ductile-brittle transition of anisotropic bilayer-hybrid gel upon dehydration is quite different from that of the isotropic bulk PAAm gel. High quantity of bound water was found in the PAAm hydrogel layers of ~10 nm thick intercalated between bilayers, which governs the dynamics of PAAm and the mechanical properties of the anisotropic hydrogels, for the first time.

In conclusion, the author has made significant progresses not only in synthesizing, but also on understanding of the structure-properties relationship of anisotropic hydrogels. The result will contribute to new understanding in the anisotropic hybrid hydrogels and will give insight into understanding various biological tissues. Therefore, we acknowledge that the author is qualified to be granted a Doctorate of Life Science from Hokkaido University.