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Structure and Properties of Physical Hydrogels from Polymer Adsorption on Bilayer
Membranes

（二分子膜へのポリマー吸着による物理ハイドロゲルの構造と物性）

〈Title of Doctoral Dissertation submitted for the degree〉

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Conventional hydrogels are isotropic in structure and weak in mechanical strength. Recent studies on double network hydrogels have shown that by introducing rigid and brittle sacrificial bonds to soft and ductile matrix, the hydrogels become tough by dissipate energy during deformation. The innovative developments of tough hydrogels have greatly expanded the potential applications of this material in various fields, including soft actuators, artificial organs, regenerative medicine, etc. For these potential uses, the next challenge is to develop tough hydrogels with anisotropic super-structures of macroscopic scale, as like biological tissues. A promising strategy is to incorporate self-assembled structures into hydrogels. The self-assembled structures, usually being more rigid and brittle than polymer networks, can rupture at loading and reform at unloading. Therefore, they not only impart various anisotropic properties, but also have potential as reversible sacrificial bonds to make the soft materials tough and self-healing.

Directional molecules, for example, lipid molecules, can self-assemble into various meso-scale super-structures in aqueous solution, notably lamellar structure and vesicles. However, usually it is very difficult to build these meso-scale structures into the macroscopic scale, especially in hydrogels. Some recent works have succeeded in developing hydrogels with macro-scale super-structures. An interesting example is the dodecyl glyceryl itaconate/acrylamide (DGI/AAm) system. The gel, consisting of poly(dodecyl glyceryl itaconate) (PDGI) that form lamellar bilayers and chemically cross-linked polyacrylamide (PAAm), exhibits many unique functions that are substantially superior to conventional hydrogels, such as structure color, unidirectional swelling, high toughness, and self-healing. Despite these excellent functions, the structure formation mechanism of superstructure hydrogel and the interaction between the bilayer and the polymer in the hydrogel are still unknown. Considering the vulnerable nature of the bilayer membranes that are easily disturbed by subtle changes in the environment conditions, it is an interesting puzzle why bilayers form macro-scale, mono-domain lamellar structure in PAAm hydrogel. Furthermore, it is also attractive to design a new method for the precisely control of the lamellar structure. Currently, DGI monomers are polymerized simultaneously with AAm by the one-step polymerization method, in which the formed PDGI membranes are only decided by the self-assembling and hard to control.

The aim of this thesis is to elucidate how the bilayer membranes and polymer form superstructure hydrogels and how the two components affect the properties of the hydrogel. The thesis consists of 5 chapters. Chapter 1 is the general introduction of the thesis. Chapter 2 is the background about the previous works of DGI/AAm system. In chapter 3, I focus on the linear polymer system without using any chemical cross-linker to reveal the interaction between PDGI bilayer and PAAm. I have

systematically studied the structure evolution of the precursor monomer solution, the as-prepared gel, and water-equilibrated gel. I elucidated, for the first time that, the PAAm chains adsorb on the bilayer surfaces via hydrogen bond to form physical hydrogels. The adsorption of polymer causes the isotropic/anisotropic bilayer structure transition through the excluded volume effect. Corresponding to this structure change, the gels showed unique swelling behaviors. Furthermore, the physical gels showed very large stretchability and self-healing properties. The hydrophobic associations of bilayers serve as sacrificial bonds to dissipate mechanical energy, and the self-similar adsorption of polymer imparts elasticity to the gel.

In chapter 4, I successfully developed a novel two-step polymerization method based on the self-assembling of PDGI in DMSO/water mixed solvent to control the bilayer and polymer individually. The properties of hydrogels prepared by this method, are comparable to that prepared by one-step polymerization. Based on this method, hydrogels with porous lamellar membranes can be prepared by the copolymerization of DGI with AAm. The mechanical properties of the porous hydrogel can be improved by the copolymerization of DGI with crosslinker MBAA.

In chapter 5, concluding remarks are summarized.

This work is helpful to design tough materials with specific macroscopic structures. Besides, lipid bilayers are widely existent in organisms acting as structural compartments of cell and organelle membranes. The deformation of cells strongly correlates to their functions. For example, pathological changes especially for cancer, always accompany with large cell deformation. This work also gives new insight to understand the large and reversible deformation mechanism of biological cells from the viewpoint of the interaction between phospholipid bilayers and the adjacent cytoskeleton of the cells.