学 位 論 文 の 要 約

博士の専攻分野の名称　博士（理　学）　氏　名　　孫　　桃林

学 位 論 文 題 名

Synthesis of Novel Hydrogels from Linear Polyampholytes and Analysis of Their High Toughness and Viscoelasticity

(線形両性電解質高分子による新規ハイドロゲルの合成とその高靭性と粘弾性機能の解析)

Hydrogels attract great attention as biomaterials as a result of their soft and wet nature, similar to that of biological tissues. Recent inventions of several tough hydrogels show their potential as structural biomaterials, such as cartilage. Any given application, however, requires a combination of mechanical properties including stiffness, strength, toughness, damping, fatigue resistance and self-healing, along with biocompatibility. This combination is rarely realized.

In this study, the author presents a new class of tough and viscoelastic hydrogels from polyampholytes that can be tuned to change multiple mechanical properties over wide ranges. These physical hydrogels are synthesized by random copolymerization of oppositely charged ionic monomers around the charge balance point at high concentration. The randomness of charges forms multiple ionic bonds with a wide distribution of strengths, through inter and intra-chain complexation. The ionic bonds play two roles for the mechanical properties of the hydrogels: as strong bonds and weak bonds, different from other hydrogels in which they are used as weak bonds. The strong bonds serve as permanent crosslinks to maintain the shape of the gel, whereas the weak bonds perform several mechanical functions simultaneously: enhance the fracture resistance by bond rupture and therefore toughen the materials; enhance the shock absorbance by generating high internal friction; enhance the fatigue resistance and self-healing by bond re-formation. Accordingly, the physical polyampholyte hydrogels become tough in analogy to double-network hydrogels although they have a different topological structure: the strong bonds form a primary network and the weak bonds form a sacrificial network.

As all of the ion groups form ionic bonds, the polyampholyte gels have a supramolecular structure, and contain 50-70 wt% of water at an equilibrium state, which is much less than that of conventional hydrogels that usually have a high water content (>80 wt%). They are strongly viscoelastic and have a high toughness (fracture energy of 4,000 J/m^2), 100% self-recovery and high fatigue resistance.

Some gels showed partial self-healing after cutting and solvent induced shape-memory effect. Young's modulus and damping ability can be tuned over a wide range by choosing the proper ion combination. In addition, the hydrogels are non-toxic and anti-fouling to cell adhesion. Many studies on the properties of polyampholytes, both the solution and the chemically crosslinked hydrogels, have been reported. This is the first report on the possibility of using polyampholytes as structural biomaterials. This polyampholyte approach has advantages for practical applications of hydrogels: it is general and directly applicable to a variety of ionic monomer combinations with specific functions; it increases the choices of tough hydrogels with desirable combinations of mechanical properties; the one-step polymerization process is easy to scale up for mass production.