【Grant-in-Aid for Scientific Research(S)】
Science and Engineering (Chemistry)

Title of Project : Proof of the Universality of the Sacrificial Bond Principle for Toughening of Hydrogels and Creation of Tough Functional Hydrogels with Varieties of Sacrificial Bonds

Jian Ping Gong
(Hokkaido University, Graduate School of Advanced Life, Professor)

Research Area : Polymer Science
Keyword : Polymer, Hydrogel, Toughness, Biomaterials

【Purpose and Background of the Research】

Hydrogels are intriguing soft and wet materials showing excellent biocompatibility and low surface friction. However, applications of hydrogels are tightly restricted due to their mechanical weakness. To solve this problem, we have created double network (DN) gels with excellent robustness comparable to that of cartilages and industrial rubbers. We have discovered that the extra-ordinary toughness of DN gels is due to the internal fracture of the brittle component of the DN gels, which prevents the crack propagation. Effect of brittle component can be called "sacrificial bond". This "sacrificial bond principle" for toughness is similar to the proposed mechanism for fracture of bone. Based on these researches, we have advocated the universality of the sacrificial bond principle and introduced a new strategy in designing other tough materials, that is "purposely introducing an easily fractured structure to make the material as a whole mechanically tough," which is completely different from the existing common method for designing high-strength materials. In this study we intend to introduce several sacrificial bonds such as covalent, ionic and hydrophobic bond to gels and demonstrate that fracture of various sacrificial bonds increase toughness of gels dramatically. Additionally, we synthesize tough and functional gels by applying this principle to various functional polymers and apply them widely such as medical materials.

【Research Methods】

We prove that not only covalent bonds like DN gels, but also physical bonds can be used as sacrificial bonds. Some examples are shown in below. To introduce sacrificial ionic interaction, both anion and cation moieties are introduced to side chain of gel networks. As a result, polyion complex forms between polymer chains. In this case, high toughness can be realized by a single network, different from DN gels. Additionally, as ionic interaction is reversible, broken ionic bonds are recovered with time. We also investigate the relationship between structure or recovering speed of sacrificial bonds and self-healing or impact absorption properties of gels. To introduce sacrificial hydrophobic interaction, lamellar bilayer structure is introduced to gels. Our preliminary experiment has succeeded to synthesize the gels with 3000 layers of well-oriented lamellar structure. Such gels showed not only toughness derived from sacrificial bonds but also emergent properties such as swelling anisotropy and brilliant structural color. In future, we investigate the functions of well-oriented lamellar structure as sacrificial bonds and anisotropic properties (swelling, permeability, and mechanical response).

【Expected Research Achievements and Scientific Significance】

Creation of tough and functional gels, which really can be applied as materials, is expected by applying our method to various functional polymers. These gels will attract much attention as medical and industrial materials. Moreover, our Sacrificial Bonds Principle has potentials to become guiding principle for toughening of materials including solid materials such as plastics and to initiate material innovation.

【Publications Relevant to the Project】

1. Jian Ping Gong, "Why are double network hydrogels so tough?" *Soft Matter* **2010**, 6, 2583.
2. Md. Anamul Haque, Gen Kamita, Takayuki Kurokawa, Kaoru Tsujii, Jian Ping Gong, "Unidirectional Alignment of Lamellar Bilayer in Hydrogel: One-Dimensional Swelling, Anisotropic Modulus, and Stress/Strain Tunable Structural Color," *Adv. Mater.* **2010**, 22, 5110.

【Term of Project】 FY2012-2016
【Budget Allocation】 209,600 Thousand Yen

【Homepage Address and Other Contact Information】

http://altair.sci.hokudai.ac.jp/g2/index.html
gong@mail.sci.hokudai.ac.jp