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Summary of Doctoral Dissertation

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Degree requested　Doctor of Life Science　Applicant's name　Saika Ahmed

学 位 論 文 題 名
Title of Doctoral Dissertation

Study on the Brittle-Ductile Transition of Double Network Hydrogels
（ダブルネットワークゲルの脆性-延性転移に関する研究）

In recent years, hydrophilic polymers, especially their cross-linked forms, known as hydrogels, have emerged out as an extremely promising material in biological and pharmaceutical applications. Synthetic hydrogels also offer a great potential to replace damaged biological tissues and cartilages due to their high water content, up to many times of their weight and their outstanding biocompatibility. But, most of the conventional gels fail to keep up with the expectation due to their poor mechanical properties, such as low strength, toughness, crack resistivity and lack of self-healing properties. Many recent approaches to synthesize strong and tough gels have been successful. Among them, double network hydrogels have grabbed a great deal of attention due to their extra-ordinary toughness, in spite of having larger water content similar to living tissues.

Double Network (DN) gel is a type of interpenetrating network (IPN) gel comprised of two networks having completely opposite features. The first network is poly(2-acrylamido-2-methylpropanesulfonic acid) or PAMPS, a polyelectrolyte, hence rigid, dilute, substantially cross-linked and brittle network; while the second network is polyacrylamide or PAAm, neutral, flexible, concentrated, loosely cross-linked and ductile. Indeed, DN gels are regarded as the toughest synthetic hydrogel reported so far; as they can exhibit high extensibility (~2000% of original length), strength (tensile fracture stress~5 MPa) and toughness (tearing fracture energy ~2000 J/m^2) at the optimal compositions of the two networks. In the optimized toughest DN gel, the molar ratio of the 1^{st} to 2^{nd} network is necessarily 1:20.

The toughness of DN gels has been found to exhibit a sharp decrease below a certain critical concentration of the 2^{nd} network at a fixed 1^{st} network concentration. This transition is termed as the 'brittle-ductile transition'. In this thesis, I focus on how the brittle DN gel changes into a tough one by increasing the amount of ductile component; that is, the 2^{nd} network and tried to differentiate the fracture processes of these brittle and ductile DN gels. Prior to the main topic, in **Chapter 1**, I put some general introductions. **Chapter 2** presents a review on hydrogels, their structures and properties, some recent progresses on hydrogel system, finally I discuss on double network hydrogels and their fracture mechanisms.

In **Chapter 3**, I studied on several series of DN gels by changing both the 1^{st} and 2^{nd} network concentrations. By comparing the molecular structures of the individual 1^{st} network and 2^{nd} network gels, I found that the true key mechanical parameter that governs the brittle-ductile transition is the fracture stress ratio of the two networks. This ratio is related to the density ratio of elastically effective polymer strands of the second network to the first network, where the inter-network topological entanglement makes dominant contribution to the second network strand density. When the strand density ratio is lower than a critical constant value, the 2^{nd} network fractures right after the fracture of the 1^{st} network, and the gels are brittle. When the ratio is higher than that constant, only the first network fractures. As a result, the brittle 1^{st} network serves as sacrificial bonds, imparting toughness of DN gels. The study also confirms that the load transfer between the two networks is via inter-network topological entanglement. This result provides essential information to design tough materials based on the double network concept.

In **Chapter 4,** I introduced a new method to directly observe the fracture of brittle and ductile type DN gels in addition to the mechanical tests. I applied tensile and pure shear force to create tensile and tearing fracture of DN gels and observed the resulting birefringence pattern using crossed polarizing films. Interestingly, I found that the ductile DN gel exhibited strong birefringence during stretching at the damaged zone, while the brittle one did not. This presence or absence of the strong birefringence indicates the force transfer from the 1^{st} to the 2^{nd} network due to damage in sacrificial PAMPS network during the fracture process, in agreement with the discussion in Chapter 3. If the 2nd network concentration (strength) is enough high, the force is effectively transferred to the 2^{nd} network, inducing orientation of the 2^{nd} network chains and strong birefringence. However, if it is not enough, the transferred force induces immediate 2^{nd} network fracture, resulting in negligible birefringence. The observations from tensile and pure shear fracture test clearly revealed different fracture processes for brittle and ductile DN gels and thereby, supported the proposed brittle-ductile fracture mechanism in Chapter 3.
In **Chapter 5**, I ended the dissertation with some concluding remarks on the summary of my works.