



Title	Study on the Brittle-Ductile Transition of Double Network Hydrogels [an abstract of dissertation and a summary of dissertation review]
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学位論文審査の要旨
Doctoral Dissertation Evaluation Review

博士の専攻分野の名称 博士 (生命科学) 氏名 Saika Ahmed
Degree requested: Doctor of (Life Science) Name:

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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)

博士学位論文審査等の結果について (報告)

Results of Evaluation of the Doctoral Dissertation (Report)

In recent years, double network hydrogels have drawn great attention due to their extra-ordinary toughness in the range of living tissues, in spite of having larger water content. 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 network. Indeed, DN gels are regarded as the toughest synthetic hydrogel reported so far, at the optimal compositions of the two networks. In the optimized toughest DN gel, the molar ratio of the 1st to 2nd network is necessarily 1:20. However, the toughness of DN gels exhibits a sharp decrease below a certain critical concentration of the 2nd network at a fixed 1st network concentration. This transition is termed as the ‘brittle-ductile transition’.

This thesis focuses on how the brittle DN gel changes into a tough one by increasing the amount of ductile component; that is, the 2nd network, and describes, in detail, difference of the fracture processes of these brittle and ductile DN gels. By comparing the molecular structures of the individual 1st and 2nd network gels, the true key mechanical parameter that governs the brittle-ductile transition is found to be the fracture stress (strength) ratio of the two networks. This ratio is related to the number density ratio of elastically effective polymer strands of the 2nd network to the 1st network, where the inter-network topological entanglement makes dominant contribution to the 2nd network strand density. When the strand density ratio is lower than a critical constant value, the 2nd network fractures right after the fracture of the 1st network, and the gels become brittle. When the ratio is higher than that constant, only the first network fractures. As a result, the brittle 1st 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. In this thesis, the author also introduced a new method to directly

observe the fracture of brittle and ductile type DN gels in support of the above mentioned hypothesis. Tensile and pure shear force was applied to create tensile and tearing fracture of DN gels and the resulting birefringence pattern was macroscopically observed using crossed polarizing films. The ductile DN gel exhibited strong birefringence during stretching at the damaged zone, while the brittle one did not. This strong birefringence indicates the force transfer from the 1st to the 2nd network followed by damage in the sacrificial 1st network. If the 2nd network polymer strand concentration (strength) is enough high, the force is effectively transferred to the 2nd network, inducing orientation of the 2nd network chains and strong birefringence. However, if it is not enough, the transferred force induces immediate 2nd 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 this work.

In conclusion, these explanations offer a deeper insight into the fracture mechanism of double network gels and provide essential information to design tough materials based on the double network concept. Therefore, we acknowledge that the author is qualified to be granted the Doctorate of (Life Science) from Hokkaido University.