Doctoral Dissertation Evaluation Review

Degree requested Doctor of Soft Matter Science Applicant's name LU FEIXUE

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Title of Doctoral Dissertation

Study on Effects of Network Topology and Chemistry on Tensile Behaviors of Double Network Gels
（網目構造のトポロジーと化学構造がダブルネットワークゲルの引張力学特性に及ぼす影響に関する研究）

Results of Evaluation of the Doctoral Dissertation (Report)

Hydrogels, celebrated for their high-water content and flexibility, have found extensive applications in biomedicine, soft robotics, and tissue engineering, owing to their biocompatibility and tunable mechanical properties. However, traditional single network (SN) hydrogels are inherently brittle, experiencing catastrophic failure under low stress due to stress concentration at defects and crack tips. To address these limitations, double network (DN) hydrogels have emerged as a groundbreaking solution. By combining a brittle, highly crosslinked first network with a soft, extensible second network, DN hydrogels exhibit exceptional toughness and strength, opening new avenues for advanced material applications. The remarkable toughness of DN gels is attributed to the sacrificial bond principle, however, realizing this mechanism requires that the relative strength (quantified by strand density) and relative stretchability (characterized by maximum stretch ratio) of the second network be greater than those of the first network. To date, the quantitative relationship between the relative strength and stretchability of the two networks and the tensile behavior of DN gels has not been fully established.

In this dissertation, building upon the current understanding of DN hydrogel mechanics and the internal fracture processes of the first network, the author systematically modified the topological and chemical structures of the first network while maintaining the second network constant, synthesizing a series of DN gels with varying different strand density ratios. The author elucidated how the strength balance between the two networks influences the uniaxial tensile behavior of DN gels and established quantitative relationships between tensile curve patterns and chain density ratio. Furthermore, universal scaling relationships were identified between the strand density of the first network and key tensile parameters such as yield stress, yield stretch ratio, plateau stress, hardening stretch ratio, strain-hardening degree, and fracture stress. This dissertation revealed that the mechanical behavior of DN gels is primarily governed by the topological structure, with only a minimal influence from the chemical structure of the first network. The quantitative relationships at the yield point indicated that it corresponds to the first network reaching its average stretch limit. The relationships between strain-hardening degree, fracture stress, and strand density demonstrated that even though the first network is fragmented in the hardening region, it functions as sliding crosslinking points for the second network, significantly enhancing the mechanical performance of DN gels. This research not only deepened the understanding of the fracture process of the first network in DN gels but also provided a comprehensive framework for predicting and tailoring the mechanical properties of DN gels, facilitating their optimization for a range of advanced applications.

Additionally, this dissertation explored how the extensibility of the second network influences the internal fracture behavior of the first network in DN gels. The author demonstrated that the second network

significantly reduces the effective crack length of the first network by mitigating stress concentrations at crack tips, thereby increasing the force and energy required to fracture the first network. This dissertation further revealed that as the second network becomes stiffer and less extensible, the tensile process of DN gels transitions from localized yielding to macroscopically uniform deformation. These findings underscore the critical role of the second network in controlling the fracture behavior of the first network and optimizing the performance of DN gels.

This dissertation not only advanced the fundamental understanding of the mechanical mechanisms governing DN hydrogels but also introduced innovative design strategies for developing functional hydrogels with high toughness, efficient energy dissipation, and controllable fracture behavior. The universal scaling relationships established in this dissertation offer predictive power for optimizing DN hydrogels for a variety of advanced applications, from biomedical devices to soft robotics. Therefore, we acknowledge that the author is qualified to be granted a Doctorate of Soft Matter Science from Hokkaido University.