学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

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)

Hydrogels are a class of soft and water-rich polymer materials with wide-ranging applications in biomedicine, soft robotics, tissue engineering, and other fields requiring biocompatibility, flexibility, and high-water retention. However, traditional chemically crosslinked single network (SN) hydrogels are inherently brittle due to their heterogeneous network structures. Stress tends to concentrate at defects or crack tips, leading to catastrophic failure under relatively low stress and strain. This defect sensitivity severely limits the utility of SN hydrogels in applications demanding high strength and toughness.

Double network (DN) hydrogels offer a transformative solution to these challenges. By combining two interpenetrating networks with contrasting properties—a brittle, highly crosslinked first network and a soft, stretchable second network—DN gels achieve remarkable toughness, with fracture energies several orders of magnitude higher than their single network counterparts. The exceptional toughness arises from a unique energy dissipation mechanism: the first network fractures sacrificially under stress, dissipating energy, while the second network maintains structural integrity and prevents catastrophic failure. Despite extensive experimental and theoretical studies on the relationship between their network structures and macroscopic mechanical properties, comprehensive and systematic studies on the relationship between critical mechanical parameters and network structures remain scarce. Meanwhile, the key factors influencing the internal fracture behavior of the first network have yet to be fully elucidated. In particular, the effect of relative strength, topology, chemistry, and extensibility of the two networks on the overall mechanical performance of DN hydrogels requires further clarification. Addressing these issues is of significant theoretical and practical importance for advancing our understanding of the mechanical behavior of DN hydrogels and optimizing their design.

This dissertation aims to deepen the fundamental understanding of the tensile mechanics and fracture behavior of DN hydrogels by focusing on two key questions: (1) How do the topological and chemical structures of the first network affect the tensile behaviors of DN gels? (2) How does the extensibility of the second network influence the fracture behavior and effective crack length of the first network? By addressing these questions, this work seeks to establish a comprehensive framework for predicting and tailoring the mechanical properties of DN gels, enabling their optimization for various advanced applications.

To achieve these objectives, a systematic experimental approach was employed.

1. In Chapter 3, the topological and chemical structures of the first network were varied by controlling monomer type, concentration, and crosslinking density. The resulting DN gels were characterized to uncover universal scaling relationships governing their mechanical behaviors, such as yielding, necking, strain-hardening, and fracture. The chemical effects were further studied using hydrophobic polyelectrolyte-based first networks to assess the influence of chemical composition on mechanical performance.

2. In Chapter 4, the effective crack length of the first network and its fracture modes were analyzed in detail. DN hydrogels with varying relative strengths between the two networks were prepared by systematically tuning the crosslinking densities of both the first and second networks. The role of the second network's extensibility in crack tip stress concentration reduction, and fracture mode transition was investigated.

This research provides critical insights into the mechanics of DN hydrogels, with contributions summarized as follows:

1. Universal Scaling Laws. A set of scaling relationships between mechanical parameters (e.g., yield stress, plateau stress, fracture stress) and the topological structure of the first network was established, offering predictive power for the design of DN gels with desired mechanical properties.

2. Effective Crack Length Regulation. The study demonstrates that the second network significantly reduces the effective crack length of the first network, enhancing its fracture resistance. This highlights the importance of the second network's extensibility in determining the overall toughness of DN gels.
3. Fracture Mode Control. The findings reveal a transition in the fracture mode of the first network, from localized necking dominated fracture to macroscopically uniform deformation, driven by the relative strength of two networks and the extensibility of the second network.
4. Practical Implications. The insights gained offer guidance for tailoring DN hydrogel properties through network design, paving the way for their application in advanced technologies requiring exceptional mechanical performance.

This dissertation is structured as follows:

In **Chapter 1**, the research background, objectives, strategy, significance, and outline of this thesis are discussed.

In **Chapter 2**, the fundamental properties and mechanical limitations of hydrogels, along with various strategies to overcome these limitations, are reviewed. The chapter also discusses the concept and mechanical behavior of DN hydrogels, identifies existing research gaps, and outlines the strategy employed in this study.

In **Chapter 3**, the effects of topological and chemical structures in the first network on the tensile behaviors of DN gels are explored. This chapter establishes scaling laws to describe the relationships between the first network's structure and the tensile mechanics and investigates how the chemical composition and topology of the first network influence the macroscopic mechanical performance.

In **Chapter 4**, the role of the second network's extensibility in controlling the tensile behavior of DN gels is investigated. This chapter focuses on how the second network modulates the effective crack length in the first network, emphasizing the underlying mechanisms of stress delocalization and the critical interplay between the two networks.

In **Chapter 5**, conclusions of the whole dissertation are summarized.