



HOKKAIDO UNIVERSITY

Title	Study on Force-Induced Polymerization and Growth in Double-Network Hydrogels with Vascular-Like Monomer Supply [an abstract of dissertation and a summary of dissertation review]
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Doctoral Dissertation Evaluation Review

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Examiner :

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Title of Doctoral Dissertation

Study on Force-Induced Polymerization and Growth in Double-Network Hydrogels with Vascular-Like Monomer Supply
(血管様モノマー供給を有するダブルネットワークゲルにおける力誘発重合と成長に関する研究)

Results of Evaluation of the Doctoral Dissertation (Report)

Double-network (DN) gels offer an excellent platform for mechanochemical reactions due to their ability to generate mechanoradicals from the fracture of the brittle polymer network. This process efficiently transfers mechanical energy to the chemical bonds of the brittle network, producing mechanoradicals that initiate the polymerization of pre-loaded monomers and reconstruct the materials.

However, sustaining mechanoradical polymerizations in DN gels requires immersing the DN hydrogel in monomer solutions to replenish consumed monomers, limiting their practical applications. To address this issue, the author aimed to create a self-sustaining system with independent monomer complementation, eliminating the constraints of the under-solution system.

Drawing inspiration from nature's vascular structures, the author integrated channel structures within double-network hydrogels. The channel in the DN gel functions as a delivery pathway, similar to blood vessels in the human body or vascular tissues in plants. This vascular-like flow allows solvents or solutions to pass through the gel, replenishing chemicals and water, maintaining stable water content, and facilitating continuous self-strengthening in dry environments through hours of cyclic mechanical training.

The internal circulatory flow from the channel enables the channel-integrated DN (c-DN) hydrogel to sustain itself during mechanoradical polymerizations. The diffusion of monomers into permeable c-DN gels allows mechanochemical growth to be easily adjusted by varying the monomer concentration and the duration of solution flow through the channel. This direct control of mechanical growth and corresponding variations in chemical components is vital. The adjustable duration of monomer supply in the channel also helps to decipher the interplay between mechanoradical-initiated polymer growth and self-strengthening.

Using Raman spectroscopy, the author identified correlations between the kinetics of polymer growth and mechanical strengthening. The distinct difference between the increasing trend of polymer content and the mechanical parameters was examined, with the author discussing this unique polymerization behavior from the perspective of the possible long lifetimes of mechanoradicals tethered to a polymer network. Additionally, quantifying both monomer diffusion and in-situ polymerization areas revealed that the lifetime of network-trapped mechanoradicals is much longer than that of small radicals in solution.

In conclusion, the author presents new findings on mechanochemical reactions in DN gels, including extremely long-lived radicals and delayed mechanical strengthening. These insights contribute to a more detailed understanding of long-lasting macroradicals in the DN gel system. Based on the self-sustainable channel structure, mechanochemical reactions in c-DN hydrogels could enhance applications such as real-time monomer conversion monitoring and mechano-chemical reactors.

Therefore, we acknowledge that the author is qualified to be granted a Doctorate of Soft Matter Science from Hokkaido University.