HOKKAIDO UNIVERSITY

Title	Study on mechanoradical generation and application in azo-crosslinked double network hydrogels [an abstract of dissertation and a summary of dissertation review]
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Doctoral Dissertation Evaluation Review

Degree requested Doctor of Soft Matter Science Applicant's name WANG ZHIJIAN

Examiner:

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Title of Doctoral Dissertation

Study on mechanoradical generation and application
in azo-crosslinked double network hydrogels
（アゾ架橋ダブルネットワークゲルにおけるメカノラジカル
の発生と応用に関する研究）

Results of Evaluation of the Doctoral Dissertation (Report)

Mechanochemistry is an active field. Most of the studies in polymer mechanochemistry aim at developing optical mechanophores for force detection in polymer materials. Designing mechano-induced chemical reactions and utilizing reaction products to impart new functions to polymer materials are a frontier topic. Recently, a promising strategy of using mechanoradicals generated in the double network (DN) hydrogels for developing new mechanoresponsive polymer materials has been proposed.

In this study, the author aims at increasing the mechanoradical concentration and thereby enhancing the polymerization efficiency in DN hydrogels. For this purpose, the author incorporated a weak azoalkane crosslinker into the first network of DN gels as sacrificial bond and compared the mechanoradical concentrations in azo-crosslinked DN gels and conventional DN gels crosslinked by methylene bisacrylamide. The author clarified that the azoalkane crosslinker indeed works as efficient sacrificial bond to generate remarkably enhanced mechanoradicals. In the azoalkane-crosslinked DN gels, the concentration of mechanoradicals can reach a maximum of ~220 μM, which is 5 times that of the traditional DN gels. The high mechanoradical concentration greatly alleviates harsh anaerobic environment for mechanoradical applications and accelerating the polymerization reaction rate.

Then, the author demonstrated two interesting and important applications of mechanoradicals using azo-crosslinked DN gels. One is for the control of hydrogel morphogenesis. The author utilized the force-induced mechanoradicals to trigger the formation of a new network inside the deformed gels. This new network conferred plasticity to the elastic DN gel, which maintained its force-induced shape deformation, along with an increase in volume and strength. With this plasticity, the author regulated sheet-shaped DN hydrogels into various three-dimensional shapes at ambient temperature by applying existing shaping processes such as blowing and drawing.

Another application of mechanoradicals is for crack resistance enhancement. The author utilized the mechanoradicals generated in the crack tip to form a new network in the crack tip. This new network reinforced the crack tip and thus improved the stretchability of the notched DN gels. The author systematically performed the single-edge notched test, pure shear test, and tearing test to demonstrate the above enhancement effect. The author also found the interesting rate-dependent tensile behavior through the application of mechanoradicals.

In conclusion, the author developed an approach to increase the mechanoradical concentration in DN gels and demonstrated new ideas to utilize the mechanoradicals for force-induced morphogenesis and self-toughening. These achievements greatly contribute to the development of mechanochemistry in polymer material fields.

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