Doctoral Dissertation Evaluation Review

Degree requested: Doctor of Soft Matter Science Applicant's name: LIAO Hongguang

Examiner :

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Title of Doctoral Dissertation

Design of Underwater Adhesive Hydrogels through Bioinspired Data-Driven Strategy
（バイオインスパイアードなデータ駆動型戦略による水中接着性ハイドロゲルの設計）

Results of Evaluation of the Doctoral Dissertation (Report)

The development of underwater adhesives represents a cutting-edge frontier in materials science, with the potential to enable transformative advancements across a wide range of fields. Unlike adhesion in dry environments, achieving rapid and robust bonding in aqueous conditions remains a significant challenge due to the presence of interfacial hydration layers, which inhibit key interactions such as van der Waals forces, electrostatic attractions, and hydrogen bonding. A promising strategy for developing underwater adhesives is to draw inspiration from natural systems. Although substantial progress has been made through bioinspired design and synthetic approaches, the field remains in its early stages, offering considerable opportunities for further discovery, mechanistic understanding, and performance optimization.

In this study, the author employed data-driven methodologies in a novel way to facilitate the design of underwater adhesives, significantly advancing materials development. The author developed a data mining strategy to extract adhesion-related sequence features from a comprehensive adhesive protein database and derived 180 bioinspired formulations to guide the preparation of underwater adhesive hydrogels. Two major sequence-level features potentially contributing to protein adhesive performance were identified. Statistical analysis revealed that adhesive proteins exhibit statistically random sequence heterogeneity, with average block lengths typically shorter than three, which may play a crucial role in their robust adhesion process. Further pairwise frequency analysis identified specific functional class pairings within the sequences, suggesting an underlying order beneath the sequence disorder that may contribute to adhesion performance. Based on these observations, the author then proposed a formulation strategy using six functional monomers to statistically replicate the adhesive sequence features through ideal random copolymerization. A total of 180 hydrogels were synthesized based on formulations derived from data mining, forming the foundation for a comprehensive training dataset. Using various machine learning optimization models, the author conducted three iterative rounds of formulation prediction and experimental validation. This led to the discovery of a serials of super-adhesive hydrogel exhibiting an adhesion strength exceeding 1 MPa, far surpassing the performance of previously reported underwater adhesive hydrogels and elastomers.

In conclusion, the author presents novel findings on applying a data-driven strategy to accelerate hydrogel design. A successful framework was proposed that integrates data mining, bioinspired experimental design, and machine learning to create high-performance underwater adhesive hydrogels. Using this framework, the author optimized formulations from an initial dataset of 180 bioinspired hydrogels, achieving a remarkable improvement in adhesive strength, with a maximum value exceeding 1 MPa. This work offers a promising pathway for developing next-generation functional soft materials.

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