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Title of Doctoral Dissertation

Fast Underwater Adhesion of Hydrogels by Multi-Scale Design
(マルチスケール設計によるハイドロゲルの高速水中接着)

Adhesion has long been studied, and early efforts mainly focused on the contact between stiff materials. Recently the emerging great progress on soft materials has been made, and adhesion that involves a soft material especially soft and wet materials start attracting attention. Hydrogel, one kind of typical soft and wet material, has promising applications in diverse areas, especially wet environments including tissue engineering, wound dressing, bio-medical devices, and underwater soft robotics. When it comes to adhesion in these wet environments, many of the mechanical properties which showed significant effects on the strength of adhesion, are compromised. Understanding the underwater contact behaviors of hydrogels will help design adhesives for various applications in wet or underwater environments. In this thesis, the hydrogels with dynamic bonds are chosen as underwater adhesives to be studied and designed. And the results have revealed that hydrogels with dynamic bonds can achieve outstanding underwater adhesion performance by promoting draining efficiency of water film at the contact interfaces and choosing polymers with proper dynamic bonds.

In Chapter 3, we have developed hydrogels showing fast, strong, and reversible underwater adhesion based on a multi-scale design. The success is due to the synergetic effect brought about by integrating macroscopic surface engineering and the tough hydrogels with dynamic bonds. The surface grooves not only accelerate water drainage and prevent water trapping, but also delay crack propagation during detachment. Specifically, the discontinuous contact pattern leads to independent detachment of contacts, which requires re-initiation of the crack for each contact. The splitting of contact also leads to an increase in the compliance of the contact point, which significantly enhances the bulk deformation of the gel. The dynamic bonds of the gel not only form reversible bridges at the interface to show reversible adhesion, but also dissipate a significant amount of energy in bulk during deformation. Smaller feature sizes lead to stronger underwater adhesion but poorer reversibility as the self-recovery time increases with the deformation at debonding. Such trade-off relations, determined by the adhesion strength, the modulus, and the self-recovery kinetics of the hydrogels, should be considered when designing the size of the surface features. This research could be used in some hydrogels applications requiring fast and reversible adhesion in wet environments or underwater, such as re-usable sheets for wound dressing, temporary adhesives for tissue healing, and anti-slippery gloves for wall-climbing robotics. The proposed method is simple but effective, and suitable for large-scale manufacturing with feature size dimensions of several millimeters.

In Chapter 4, the underwater contact adhesion behaviors between soft hydrogels were investigated. It was found that softness of the materials would affect the bonding formation and debonding energy. Rigid hydrogels got a poor contact due to the poor conforming ability to the substrate, while soft hydrogels might easily get water trapped at the interface, resulting in the reduce of the real contact area and increase of initial flaws. A well softness balance between the contact surfaces become quite important in underwater contact formation. Further study on the contact element's size showed that the smaller contact element demonstrated the ability to form a faster and better contact underwater as well as the ability to be much more insensitive to the crack initiation and competent to delay crack propagating. Moreover, the surface hydrophobic nature of the hydrogels may also improve the water draining efficiency to form a better underwater contact adhesion than hydrophilic ones. Investigations on these adhesion behaviors will help to get a better understanding the contact formation and debonding mechanism underwater, and then using this knowledge to design synthetic adhesives on soft substrates such as tissue, skin.
In Chapter 5, we have developed a porous hydrogels which showed instant underwater contact on diverse non porous plates as well as a high debonding energy dissipated by suction. The success is due to the

synergetic effect brought by connective porous structure and viscoelastic properties of the matrix with dynamic bonds. The connective pores not only accelerate the water drainage at the interfaces but also dissipate a large amount of energy by suction. The dynamic bonds induced the matrix a strong viscoelastic properties enabling the large residual deformation after removing the applied force, which results in the sealing of the porous hydrogels adhesives on the substrate. The porous hydrogels with high surface bonding efficiency and suction of the bulk make them available for many potential applications, such as effective underwater adhesives-attaching sensors, beacons under the waterline, stopping watery leaks, in medicine, repairing wet tissues, wound dressing. The obtained results might provide a better perception to understand the frictional properties of soft and wet materials like biological tissues, which is informative for designing low-friction biomaterials.