Abstract of Doctoral Dissertation

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Study on Porous and Tough Hydrogels for Biomedical Engineering Applications
（バイオメディカルエンジニアリング応用のための多孔質かつ高靭性ハイドロゲルに関する研究）

As global populations age and life expectancy increases, the prevalence of musculoskeletal disorders, particularly injuries to load-bearing soft tissues such as tendons and ligaments, is expected to rise significantly. Current treatment modalities, especially in cases of severe tissue damage, rely heavily on surgical grafts, which are limited by issues such as donor site morbidity, immune rejection, and restricted tissue availability. Artificial ligaments and tendons fabricated from synthetic polymers, such as polyethylene terephthalate (PET), offer mechanical advantages; however, they often lack bioactive features, such as high water content and controlled drug delivery capabilities, which have led researchers to focus on enhancing their bioactivity.

Hydrogels, particularly those based on natural polymers, present a promising approach to addressing these limitations due to their high water content and biocompatibility. Among these, porous hydrogels have demonstrated superior performance compared to conventional hydrogels in terms of mass transfer and tissue integration, which are critical parameters for regenerative medicine.

This dissertation aims to fabricate a composite material composed of polyacrylamide/carboxymethyl cellulose (PAAm/CMC) porous gel and PET fabric, serving as a model for artificial ligaments and tendons. Firstly, we introduce a facile one-pot strategy that combines CMC with acrylamide (AAm) under cryogelation and low-intensity UV irradiation to achieve covalent bonding and a high polymerization yield. The resulting polyacrylamide/carboxymethyl cellulose (PAAm/CMC) porous gels were systematically evaluated for their chemical, physical, thermal, and drug-release properties, with a focus on the effects of AAm concentration and polymerization temperature (frozen vs. room temperature). Notably, the cryogel synthesized with 2.5 M AAm (PC2.5) exhibited significantly enhanced mechanical properties—that is, an 8.4-fold increase in tensile modulus and a 26-fold increase in toughness—compared with the non-cryo gel. Moreover, PC2.5 demonstrated excellent cyclic compression stability in water and phosphate-buffered saline (PBS), with less than 10% reduction in modulus after 100 cycles. These increases in the mechanical properties of PC2.5 are attributed to the formation of macropores with high polymer density and high crosslinking density at the pore walls. PC2.5 also showed slower drug release in PBS and good cytocompatibility. Finally, we successfully fabricated a composite material combining porous gel and PET fabric (PCC), which exhibited a significant increase in stiffness, over 700-fold, compared to porous gel without PET fabric, while maintaining the macroporous structure. These findings highlight the potential of PAAm/CMC porous gel composites for biomedical engineering applications, particularly in the development of artificial ligaments and tendons.