Summary of Doctoral Dissertation

Degree requested: Doctor of Soft Matter Science Applicant's name: Duangkamon VIBOONRATANASRI

Title of Doctoral Dissertation

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)

With the global rise in aging populations and life expectancy, musculoskeletal disorders—particularly injuries to load-bearing soft tissues such as tendons and ligaments—are becoming increasingly prevalent. Traditional treatments, especially for severe tissue damage, often rely on surgical grafts, which are associated with several limitations, including donor site morbidity, immune rejection, and limited tissue availability. Synthetic alternatives, such as artificial tendons and ligaments made from materials like 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, especially those derived from natural polymers, offer a promising alternative due to their high-water content and biocompatibility. In particular, porous hydrogels have demonstrated improved performance compared to conventional hydrogels in promoting nutrient diffusion and tissue integration, key aspects in regenerative medicine.

This dissertation focuses on developing a composite material that integrates a polyacrylamide/carboxymethyl cellulose (PAAm/CMC) porous hydrogel with PET fabric as a potential scaffold for artificial tendons and ligaments. 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. 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. Additionally, the morphology of the PC gel was that of an elastic sponge (i.e., it fills with water and solvent and returns to its original shape after being squeezed). The mechanical properties and shape of PC2.5 were not significantly different when tested in different solutions (pure water and PBS). Regarding the cyclic compression test, the modulus of PC2.5 decreased slightly after 100 compressions under wet conditions. Finally, PC2.5 exhibited slower drug release than NP, despite its components being similar to those of NP. The release behavior of PC2.5 was best described using Korsmeyer–Peppas model with a predominant mechanism of Fickian diffusion. Neither PC2.5 nor NP was toxic to cells. 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.