Title	Study on facile fabrication of super strong cellulose hydrogel with anisotropic hierarchical fibrous structure [an abstract of dissertation and a summary of dissertation review]
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Abstract of Doctoral Dissertation

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Title of Doctoral Dissertation

Study on facile fabrication of super strong cellulose hydrogel with anisotropic hierarchical fibrous structure
(異方的階層繊維構造を持つ超高強度セルロースハイドロゲルの簡便な製造に関する研究)

It's been widely studied that the extreme mechanical property of connective tissues like tendon and ligaments result from the highly aligned and hierarchical fibrous structure. Besides the extreme strength, the toughness of those connective tissues are also excellent. Preparation of finely organized fibrous structure which can optimize the strength and toughness is really challenging for hydrogel scientists.

Previously, my lab developed a facile method called Drying in Confined Condition method, simple as DCC method, which can effectively produce highly aligned and hierarchical fibrous structure on alginate hydrogels, the resulting strength can reach the human ligaments level, but the extreme strength of achilles tendon (~80MPa) is still far from reaching. And since alginate hydrogel is not stable in saline solution due ionic cross-linking, which would limit the application prospect.

Hence in this study, I choose cellulose as raw material, and focus on preparation of super strong cellulose hydrogel through DCC method. We successfully improved the mechanical strength of purely cellulose based hydrogel to human achilles tendon level, and meanwhile there is no sacrificing in toughness. Through multiple characterization method, I confirmed the high orientation degree through different scales and the way of fibrils aggregation play important role in the improved mechanical performance.

In **chapter 1**, general introduction and outline of the dissertation are discussed.

In **chapter 2**, a brief review on the strong and tough hydrogels, strengthen and toughening strategies are introduced. Meanwhile, brief introduction on the DCC method and the validity of this method are discussed. The effect of DCC method on different polymers are compared.

In **chapter 3**, the ideal environmental condition on preparation of DCC-Cellulose hydrogels are discussed.

In **chapter 4**, extremely strong and purely cellulose-based cellulose gels are prepared, The high strength and toughness of the DCC-E gels were realized by optimizing the cellulose fibril arrangement from nanoscale to macroscale, which was done by selection of an appropriate solvent used for cellulose regeneration. Parallel aggregated fibrous structures observed in the DCC-E gels are thought to play a central role in the enhancement of both toughness and strength.

In **chapter 5**, the toughening mechanism of DCC-Cellulose hydrogel have been explored through

SEM and cycling tests. It's been clarified that untwisting of hierarchical fibrous structure under loading is the main mechanism for energy dissipation. And abundant hydrophobic interaction between fibrous structure have been confirmed through immersing the samples in urea solution to observation the change of fibrous structure and compare the hysteresis change in cycling tests.

In **chapter 6**, by using DCC process as training method, we successfully further improved the mechanical property of cellulose hydrogel. By various characterization, we confirmed that during repetitive DCC process, cellulose fibrils become more aggregated together. And by giving a extreme pretrain in the second DCC process, we find the fracture stress is hard to breakthrough 90 MPa.

In **chapter 7**, conclusions of the dissertation are summarized.