

Interfacial Water Confinement Couples Hygroactuation and Mechanical Robustness in Free-standing Biocomposites

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Water-responsive materials convert water chemical potential gradients into mechanical work, but combining large actuation with structural robustness remains a key challenge. Here, we develop free-standing silk-cell biocomposite films that couple the hygroscopic response of autoclaved *Bacillus subtilis* cells with the mechanical strength of regenerated silk fibroin. Using a scalable process combining blade casting and water-assisted hot pressing, an optimized 75:25 silk-cell composite achieves a maximum water-responsive stress of 55.0 MPa, free strain of 14.8%, and rapid dehydration and hydration time constants of 2.4 and 3.7 s, respectively. The composite also exhibits a Young's modulus of 6.8 GPa, tensile strength of 120.2 MPa, and toughness of 2.5 MJ m⁻³. Mechanistic analysis indicates that hot-press-induced interfacial densification reduces mobile water and improves stress transfer between the cellular and silk phases. These results reveal a processing-structure-property relationship in which control of interfacial water enables efficient coupling between molecular-scale hydration changes and macroscopic mechanical output, providing a scalable strategy for robust hygroscopic biocomposites and dehydration-induced actuating materials.

Keywords: *Bacillus subtilis* cell, regenerated silk fibroin, biocomposites, interfacial water, hot-press, water-responsive materials

