

# Controllable Structure-Dependent Water Response by Protein Block Copolymers

Jacob Kronenberg<sup>1\*</sup>, Yeojin Jung<sup>2,3\*</sup>, Jason Chen<sup>1</sup>, Maria Jinu Kulapurathazhe<sup>1</sup>, Xi Chen<sup>2,3</sup>, Raymond Tu<sup>2,3</sup>, Jin Kim Montclare PhD<sup>1,4,5,6,7</sup>

\* J.B.K. and Y.J. contributed equally.

1 New York University Tandon School of Engineering, Department of Chemical and Biomolecular Engineering, Brooklyn, New York, United States

2 City College of New York, Advanced Science Research Center, New York, New York, United States

3 City College of New York, Department of Chemical Engineering, New York, New York, United States

4 New York University, Department of Chemistry, New York, New York, United States

5 New York University College of Dentistry, Department of Biomaterials, New York, New York, United States

6 New York University Grossman School of Medicine, Department of Radiology, New York, New York, United States

7 New York University Tandon School of Engineering, Department of Biomedical Engineering, Brooklyn, New York, United States

## Abstract

Natural water-responsive (WR) biological materials are abundant, having independently evolved in places such as wheat awns, pinecones, and bacterial spores. WR biomaterials are of interest for applications as high-energy actuators, which can be useful in soft robotics or for capturing energy from natural fluctuations in humidity. Recent work on WR protein materials has shown that  $\beta$ -sheet structure correlates with WR energy density, but the design parameters for water response in proteins remain poorly understood. Here we design, synthesize, and study CEC protein block-copolymers consisting of two  $\alpha$ -helical domains derived from cartilage oligomeric matrix protein coiled-coil (C) flanking an elastin-like peptide domain (E). We use these protein materials to create water-responsive actuators whose energy densities outperform mammalian muscle by two orders of magnitude and match those of spider silk actuators, the highest found in nature. To elucidate the effect of structure on water response, CEC was compared to a variant, CEC<sub>L44A</sub>, in which a point mutation disrupts the  $\alpha$ -helical structure of the C domain. Surprisingly, CEC<sub>L44A</sub> outperformed CEC, showing higher energy density and less susceptibility to wear after repeated cycling. This is likely because CEC<sub>L44A</sub> is softer than CEC allowing for greater expansion during water absorption as well as lower brittleness. These results suggest that rigid, highly structured domains are not necessary for strong water response.