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Exploring mussel byssus fabrication with peptide-polymer hybrids: Role of pH and metal coordination in self-assembly and mechanics of histidine-rich domains

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Abstract

Mussel byssal threads are biopolymeric fibers that possess high toughness and self-healing capacity owing to a hierarchical arrangement of collagenous protein building blocks known as preCols. To investigate the proposed role of histidine-rich domains (HRDs) at preCol termini in the rapid self-assembly and properties of mussel byssus, we functionalized 4-arm star-shaped polyethyleneglycol (star-PEG) molecules with peptides based on an evolutionarily-conserved HRD sequence motif. PEG-HRD hybrid molecules instantly and reversibly form physical hydrogels without metal ions, using a simple pH trigger mimicking the natural assembly process. Spectroscopic analysis indicates that gelation is mediated via a conformational transition into ??-sheet crystalline structure, comprised of multiple peptide groups that cross-link the star-PEGs. Additionally, metal ions introduced during gelation form His-metal coordination bonds within the network and increase viscoelastic damping of the gels, as proposed for native fibers. These findings help disentangle the different roles of pH and metal coordination during natural byssus formation/function and may have important implications for generating novel metallopolymeric materials with hierarchical structure.

Keywords: mussel byssus; self-healing; supramolecular self-assembly; peptide; metal coordination

Introduction