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Title:

Viscoelastic and Covalently Cross-Linked Hydrogels Inspired by Mussels

Abstract:

The mechanical holdfast of the mussel, the byssus, is a collection of viscoelastic threads that irreversibly attach to a surface. Because mussels inhabit turbulent intertidal zones, the adhesives employed by mussels are extremely robust and able to function under water. The proteins that compose these permanent anchors are especially enriched in metal ions and DOPA, a catecholic amino acid. While the presence of metal ions has been largely underappreciated, DOPA is well-studied and thought to play roles in both the adhesion and cohesion of the byssus. Due to this interesting combination of features, the byssus has served as the inspiration for materials ranging from those that offer insight into mussels and byssi to those that can be applied to biomedical fields. I will describe two different classes of materials. First, by processing hydrogels composed of catechol-terminated PEG and Fe^{3+} through a mussel-mimetic acidic-to-alkaline pH change, mechanically tough materials based on a covalent network fortified by sacrificial Fe^{3+} -catechol coordination bonds were produced. These findings offer the first direct evidence of Fe^{3+} -induced covalent cross-linking of catechol polymers and reveal the potential importance of the pH dependence and mechanical role of Fe^{3+} -catechol interactions in mussel byssi. Second, I will describe the synthesis and characterization of mechanically tough zero- or negative-swelling mussel-inspired surgical adhesives based on catechol-modified poly(propylene oxide)-poly(ethylene oxide) block copolymers. Catechol oxidation at or below room temperature rapidly resulted in a chemically cross-linked network, with subsequent warming to physiological temperature inducing volumetric reduction and mechanical toughening. These two sets of hydrogels offer a small sampling of the rich variety of materials that have been inspired by mussels.