

Design and Characterization of Biomimetic Adhesive Materials

M. Jane Brennan

Major Professor: Julie C. Liu

School of Chemical Engineering
Purdue University

When we engineer new materials, nature provides us with a wealth of inspiration, often in the form of proteins. The blue mussel *Mytilus edulis* and sandcastle worm *Phragmatopoma californica* produce adhesive proteins that help them to adhere in wet, turbulent environments. The frog *Notaden bennetti* secretes a sticky, proteinaceous emulsion that helps it defend against predators; the velvet worm bombards a similar protein onto its prey to prevent its escape. Mammals and insects produce remarkably elastic proteins to support highly repetitive motions. This work describes the design, production, and characterization of several biomimetic materials inspired by natural adhesive proteins.

First, we evaluated the cytotoxicity of a mussel-mimetic polymer, poly[(3,4-dihydroxystyrene)-co-styrene]. This polymer was previously shown to be strongly adhesive with strengths similar to a variety of commercial glues. To investigate the versatility of the polymer for biomedical applications, we evaluated the polymer cytotoxicity by assessing the viability, proliferation rate, and morphology of fibroblasts cultured with polymer. We demonstrated that the polymer is highly cytocompatible and is therefore a promising material for applications in which biological contact is necessary.

Next, we investigated the adhesive capabilities of a system of recombinant elastin-like polypeptides (ELPs) as well as the extrinsic and intrinsic factors influencing their adhesion strengths. We found that pH, concentration, and crosslinking did not affect the adhesion strength, whereas moisture was a critical factor. When comparing protein designs, amino acid composition did not affect the strength as significantly as protein structure and length. Finally, our adhesive proteins exhibited comparable strengths to commercially available protein-based glues. These results have strong implications for the general understanding and future design of proteinaceous adhesives.

Finally, we developed an elastin-based mussel-mimetic adhesive. The ELP is easily over-expressed and purified from *E. coli*. Following enzymatic conversion to produce adhesive DOPA residues, the protein demonstrated strong cytocompatibility, enhanced adsorption to glass, significant dry adhesion, and moderate adhesion in a humid environment. Additionally, the protein possesses a reversible phase transition behavior that can be tuned to physiological conditions. Upon transitioning, the protein forms a protein-rich coacervate phase that can provide measurable adhesion strength after being applied underwater. This novel material has potential applications as a surgical adhesive or a scaffold for tissue engineering.