Agricultural & Biological Engineering

Thesis Defense

Speaker: Jeff de Kozlowski
Title: Development of a Starch-Based Mussel-Mimetic Adhesive Polymer

Major Professor(s): Bernard Tao
Date: Friday, October 11, 2013
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Abstract:
Mussel-mimetic adhesive polymers have gained lots of attention for their strong adhesive strength, moisture resistance, and unique ability to crosslink. These properties are mainly attributed to the high content of catecholic 3,4-dihydroxyphenylalanine (DOPA) in mussel adhesive proteins. While there has been success in creating mussel-mimetic synthetic polymers, less effort has been given to create a renewable, green, biocompatible counterpart. This thesis explores the possibilities of starch-based mussel-mimetic adhesives. Carboxymethyl starch (CMS) of various molecular weights and degree of substitution (DS) was synthesized and subsequent conjugation of dopamine to these polymers by 1-Ethyl-3-(3-dimethylaminopropyl)carbodiimide was investigated. The polymers suffered from very low substitution and easily precipitated from solution as an insoluble product. Lap-shear strengths of the CMS-dopamine conjugates failed to exceed those of unmodified CMS.

In search of another path to starch-catechol conjugates with higher DS_{catechol}, 1,1’-carbonyldiimidazole was employed for direct conjugation of bis-O-protected 3,4-dihydroxybenzoic acid (DHBA) to unmodified starch. High DS was achieved with 3,4-dimethoxybenzoic acid, but demethylation techniques were incompatible with starch and its esters. Phenylboronic acid was then employed as an easily removable diol protecting group for DHBA, but the complex was apparently not stable enough in solution for selective activation of the carboxylic acid group of PBA-DHBA by either CDI or TosCl. Further screening of different protecting groups or a new coupling chemistry is needed to fully assess the possibilities of starch-catechol conjugates of high DS_{catechol}.

Application:
This work sheds light on the inherent difficulty of creating a mussel-mimetic starch adhesive and lays out considerations for future studies in this area. Successful synthesis of a mussel-mimetic starch adhesive would have a variety of applications as a green, moisture-resistant, biocompatible, and strong adhesive.