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Investigations of the Importance of the Redox Environment in LPMO-Supported Bioconversion of Pretreated Lignocellulose

Chaojun Tang

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Fakultetsopponent: Professor Héctor A. Ruiz
Food Research Department, School of Chemistry, Autonomous University of Coahuila, Mexico

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Author
Chaojun Tang

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Abstract
Achieving high sugar yields in enzymatic saccharification of cellulose is a critical step in biochemical conversion of pretreated lignocellulosic biomass. An oxidoreductase, lytic polysaccharide monooxygenase (LPMO), has recently gained attention for its potential to act synergistically with conventional hydrolytic enzymes catalyzing deconstruction of cellulose. This investigation has focused on LPMO-supported enzymatic saccharification, exploring how process conditions, particularly the redox environment, affect saccharification, fermentability, and the chemistry of liquid and solid phases of pretreated biomass. Inclusion of LPMO in cellulolytic enzyme preparations warrants a reevaluation of industrial process configurations, especially in terms of oxygen supply and aeration strategies. However, the impact of aeration has not been well understood and the aim of the investigation has been to shed light on that gap of knowledge. The impact of aeration and the roles of lignin and water-soluble lignin-degradation products as reductants in LPMO-supported enzymatic saccharification were investigated. Aeration greatly improved saccharification, and both lignin and water-soluble lignin-degradation products supported LPMO catalysis. The benefits of aeration in LPMO-supported saccharification were weighed against the negative effects associated with high-solids loadings in the range 12.5-17.5% (w/w). The positive effects of aeration were larger than the negative effects of high-solids loadings. Introduction of a pre-hydrolysis phase with elevated temperature and aeration prior to fermentation was compared with a conventional approach based on simultaneous saccharification and fermentation. Pre-hydrolysis with aeration resulted in increased glucan conversion, but, unexpectedly, also in poor fermentability. The effects of aeration and of treatments with laccase and sulfite on inhibitors and fermentability were further investigated. Chemical analysis and fermentation experiments showed that both aeration and laccase treatment can result in a decrease of some inhibitors and in an increase of others. The investigation provides more knowledge regarding LPMO-catalyzed saccharification of cellulose and regarding process designs for efficient bioconversion of lignocellulosic biomass.

Keywords
Lignocellulose bioconversion, lytic polysaccharide monooxygenase (LPMO), lignin, cellulose, enzymatic saccharification, yeast

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