**PUBLICATIONS**


**DISSERTATION DEFENSE**

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Sub 168
Department of Chemistry and Biochemistry

*“Contributors to catalytic bias revealed in the investigation of an [FeFe]-hydrogenase model system”*

**Graduate Committee**
Dr. John Peters (Research Advisor)
Dr. Brian Bothner
Dr. Joan Broderick
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Dr. Paul King (National Renewable Energy Laboratory)
ABSTRACT

The need for food, fuel, and pharmaceuticals has been increasing at a growing rate as the world’s population increases and lifestyles improve. All of these needs are highly energy dependent, and, to a significant degree, rely on an inefficient use of fossil fuels. In order to break free of this dependence, new understanding is required for how to efficiently generate the products humanity needs. Here, a model system of two closely related [FeFe]-hydrogenases, CpI and CpII, is employed in order to understand how biology is able to efficiently control the formation of reduced products, in order to further delineate the limits of control, and the extent to which biology may be co-opted for technological needs. CpI, one of nature’s best catalysts for reducing protons to hydrogen gas, is compared to CpII, which functions catalytically to oxidize hydrogen to protons and electrons. Oxygen sensitivity, midpoint potentials, catalytic mechanisms, and catalytic bias are explored in-depth using electron paramagnetic resonance, Fourier Transform Infrared spectroscopy, and protein film voltammetry. CpI and CpII have been found to function under different metabolic conditions, and key amino acids influencing their distinct behavior have been identified. The conduit arrays of hydrogenases, which direct electrons to or from the active site, have been found to have distinct midpoint potentials in CpII compared to CpI, effectively reversing the favored electron flow through CpII in comparison to CpI. In order to probe the contributions of the protein framework on catalysis, analysis of site-specific amino acid substituted variants have been used to identify several determinants that affect the H-cluster environment, which contributes to the observed differences between CpI and CpII. This has resulted in a deeper understanding of the hydrogenase model system and the ability to directly influence catalytic bias. Thus, the work presented here represents key progress towards developing unidirectional catalysts, and demonstrates the possibility of targeted, rational design and implementation of unidirectional catalysts.

BIOGRAPHICAL NOTES

Academic Preparation:

2008-2012 Calvin College, Grand Rapids, MI (May 2012)
Bachelor of Science in Biochemistry
Research: Visualizing the activation of glucose transporter 1
Advisor: Dr. Eric Arnoys

Graduate Studies

Field of Study: Biochemistry

Teaching and Outreach Activities

2013 General Chemistry Lab TA, Montana State University (Jan.-May)
2012 General Chemistry Lab TA, Montana State University (Sep.-Dec.)

Awards

2016 Best Poster, 11th International Hydrogenase Conference
2015 First Prize, MSU Graduate Student Research Summit
2015 MSU Graduate Student Competitive Research Competition
2012 Presidential Graduate Scholarship

Poster Presentations

July, 2016 11th International Hydrogenase Conference, “Structural determinants of catalytic bias in [FeFe]-hydrogenases as revealed by potentiometric EPR spectral deconvolution” [updated]

June, 2016 Northwest Crystallography Conference, “Structural determinants of catalytic bias in [FeFe]-hydrogenases as revealed by potentiometric EPR spectral deconvolution”

October, 2015 Montana State University Graduate Student Research Summit, “[FeFe]-Hydrogenases as a Model System for the Study of Catalytic Bias”

March, 2015 22nd West Coast Protein Crystallography Workshop, “Structural and Biochemical Characterization of a Highly Thermostable Mercuric Reductase from Metallosphaera sedula”