

Acid-Stable
Electrocatalysts for the
Solar Production of Fuels

Thesis by
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In Partial Fulfillment of the Requirements for
the degree of
Doctor of Philosophy

The logo for the California Institute of Technology (Caltech), featuring the word "Caltech" in a bold, orange, sans-serif font.

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ABSTRACT

Sunlight is one of the few renewable resources that can meet global energy demand. Unfortunately, while solar energy has grown in the past few years, several economic and scientific constraints have hindered mass adoption. One of the main obstacles solar energy faces is the lack of economically competitive storage technologies. Artificial photosynthesis is a potential solution in which solar energy is directly converted into energy dense chemical bonds that can be easily stored and transported.

One impediment facing the commercialization of artificial photosynthesis is the use of expensive and rare precious metals as catalysts. This dissertation focuses on the achievements of the past five years in characterizing novel, earth-abundant, acid-stable hydrogen evolution catalysts. While nickel alloys have long been known as catalysts for the hydrogen evolution reaction in basic media, it has only been in the past decade that earth abundant catalysts that are stable in acidic media have been reported. These discoveries are critically important as the many proposed artificial photosynthetic devices require the use of acidic media.

In this dissertation we examine two families of hydrogen evolution catalysts: transition metal chalcogenides (namely molybdenum and cobalt selenide) as well as transition metal phosphides (cobalt phosphide). In addition to the electrochemical characterization of these catalysts, spectroscopic characterizations were performed in order to carefully

examine the chemical compositions of these catalysts before, after and during the hydrogen evolution reaction. This analysis elucidated both chemical, and structural changes that occurred after the catalysts had been subject to the hydrogen evolution reaction conditions.

The final chapter in this thesis delves into the techno-economic realities of energy transportation via different fuels. Due to the strong interest in renewable energy, several future energy transportation scenarios, including 100% grid electrification and widespread installation of hydrogen pipelines, have been proposed. In order to get a fuller understanding of such potential infrastructure alternatives, we report their differing energy transportation costs.

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