

CHEMICAL ENGINEERING

UNIVERSITY of WASHINGTON

SEMINAR



Thomas Gill
**National Science Foundation Graduate Research
Fellow**
Stanford University

Engineering Electrodes and Electrolytes Toward Valuable Water Oxidation Products

1-2 pm PST Monday August 2nd, 2021
Zoom link is provided via email, or
contact dyss@uw.edu



Bio

Thomas is a National Science Foundation Graduate Research Fellow pursuing his Ph.D. in the Mechanical Engineering Department at Stanford University. His current research under professor Xiaolin Zheng focuses on systematically developing electrochemical systems for producing valuable products, especially hydrogen peroxide, from water. Generally, he is interested in studying electrochemical processes and mechanisms using spectroscopy and in establishing research approaches beyond electrocatalyst development for improving kinetics of emergent chemical reactions. Prior to his Ph.D., Thomas graduated from Villanova University in 2016 having studied mechanical engineering, math, and humanities. He is passionate about extending education to young people as a means of social mobility and bridging the gap between academic research and real-world applications.

Abstract

Electrochemical technologies have been a promising research area for both sustainable energy development and potable water provisions for decades. Recently, electrochemical oxidation of water has garnered a new focus, with much research aiming to selectively oxidize water to value-added products such as hydrogen peroxide. Providing dual function as a water purifying agent and an energy storage chemical, hydrogen peroxide production via water oxidation suffers some major drawbacks: first, it

requires high electric potentials to produce, rendering most catalyst materials unstable over longer durations necessary for implementation. Second, this reaction competes with the thermodynamically favored oxidation of water to oxygen gas, making overall selectivity and production rates low. Nearly all efforts to address these shortcomings have focused on developing new electrode and catalyst materials.

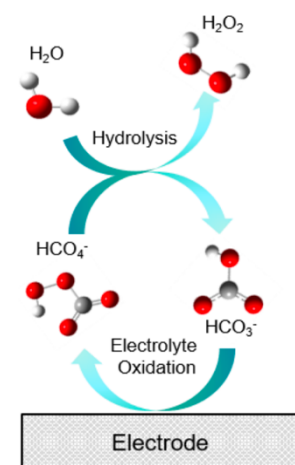


Figure 1. Schematic illustration of electrolyte oxidation, followed by reaction between the oxidized species and water to produce hydrogen peroxide.

In our work, we find that we can leverage the other side of the electrochemical interface, the aqueous electrolyte, to drastically improve the selectivity toward hydrogen peroxide without sacrificing stability or adding material cost. Contrary to the common assumption that the electrolyte is a chemically passive ionic conductor, we find that in carbonate-based electrolytes the electrolyte is oxidized, which likely leads to subsequent hydrogen peroxide production via reaction with water (**Figure 1**). Noting this, we engineer the electrolyte by controlling the amount of HCO₃⁻, CO₃²⁻, ionic strength, and cationic species to invoke an order of magnitude improvement in the hydrogen peroxide current density ($J_{H_2O_2}$) as compared to the most common electrolyte (2M KHCO₃, **Figure 2**). Notably, this comes with no depletion in electrocatalytic performance and is shown to be generalizable over a variety of electrocatalyst surfaces. Furthermore, the stability of the hydrogen peroxide generating system is improved under the optimized electrolyte conditions.

Together, the enhancements found through engineering the electrolyte present a means to drastically improve H₂O₂ production efficiency and offer a paradigmatic shift from the solid-state catalyst development which predominates our field. This method may also be generalized to other electrosynthesis applications as an inexpensive and facile means to improve performance.

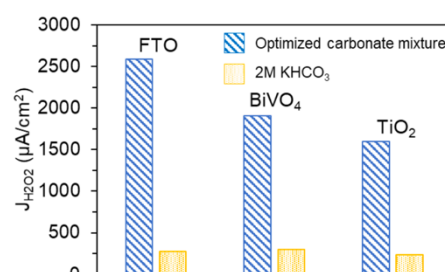


Figure 2. Order of magnitude improvement in current density toward H₂O₂ when substituting our optimized electrolyte (0.5M KHCO₃ + 3.5 K₂CO₃) for the commonly used 2M KHCO₃ on several metal oxides.