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(Photo)Electrochemical Conversion for Sustainable Fuels and Chemicals

1-2 pm PST Monday July 12th, 2021 Zoom link is provided via email, or contact <u>dyss@uw.edu</u>



Bio

Elizabeth Corson is a TomKat Center Postdoctoral Fellow in Sustainable Energy working with Prof. Will Tarpeh in the department of Chemical Engineering at Stanford University. She is currently investigating electrochemical nitrate reduction to recover ammonia from wastewater. Elizabeth was a NSF Graduate Research Fellow at the University of California, Berkeley where she completed her Ph.D. in Chemical Engineering with Prof. Bryan McCloskey. She conducted her dissertation research on plasmon-enhanced electrochemical carbon dioxide reduction at the Joint Center for Artificial Photosynthesis at Lawrence Berkeley National Lab. Her research demonstrated that light coupled with an applied bias could selectively enhance carbon dioxide reduction products while simultaneously suppressing undesired hydrogen evolution. While at UC Berkeley she received the Outstanding Graduate Student Instructor Award and went on to complete her Certificate of Teaching and Learning in Higher Education and become a Graduate Remote Instruction Innovation Fellow. She also served as the co-chair of Expanding Your Horizons, hosting an annual, day-long conference with hands-on STEM workshops for 350 middle school girls. Prior to graduate school, she worked in industry for four years as a Research Associate and Quality Assurance Manager at Air Liquide. Originally from Iowa, Elizabeth received her B.S. in Chemical Engineering from the Illinois Institute of Technology in Chicago.

Abstract

(Photo)electrochemical conversion can be used to transform putative wastes into valuable products, using light and potential bias as driving forces. Unlike traditional thermochemical

processes, electrochemical conversion can occur in a wide range of temperatures and pressures (including ambient), can use solely renewable electricity, and can be performed in modular reactors that permit small- through large-scale operation. The potential impact is great: we can address climate change by treating CO2 as a feedstock rather than a waste, and improve our energy independence by offsetting the use of fossil fuels for thermochemical processes and creating renewable fuels (Fig. 1). To make this vision a reality we must address the key challenges of electrochemical conversion: a) low selectivity, which requires costly separation; and b) low efficiency, which increases the operating costs from electricity.



Figure 1: Create fuels and chemicals from water, CO_2 , electricity, and sunlight. Adapted from ref. 1.

I address these challenges through the use of nanostructured plasmonic electrodes. Nanofeatures can influence the catalytic properties of electrodes for both electrochemical and photoelectrochemical systems. In dark electrochemistry, nanostructures increase the catalytic surface area and can expose more edge and low-coordination sites which have different catalytic behavior than those found on planar electrodes. In photoelectrochemistry, plasmon decay generates hot electrons that can be transferred selectively to an unoccupied electronic state of a surface molecule. Simultaneously, the enhanced electric fields can alter the electronic coupling with surface-adsorbed molecules. These catalytic and plasmonic effects can be tuned by changing the size, shape, and proximity of nanofeatures on the electrode surface, while applying a bias allows us to adjust the energy level of the charge carriers.

To study plasmon-enhanced CO_2 reduction, I developed a front-illuminated, temperature-controlled electrochemical cell that enabled precise gas and liquid product analysis^{.2} Using this cell, I reported the first instance of plasmonic promotion of CO_2 reduction at a voltage-biased cathode: illumination of the silver cathode selectively enhanced CO_2 reduction products while simultaneously suppressing undesired H₂ evolution.³

To understand the plasmonic mechanisms driving this selectivity change, I conducted an in situ ATR– SEIRAS (attenuated total reflectance–surface-enhanced infrared absorption spectroscopy) study under dark and illuminated conditions.⁴ I concluded that the light was enhancing the desorption of CO from the surface through the temporary transfer of a plasmonically excited hot electron. I also measured an increase in the strength of the symmetric CO₂ stretch of bicarbonate in the light, likely caused by the plasmonically enhanced local electric field, increasing the local pH and suppressing H₂ formation. To build on this work, I combined the plasmonic properties of silver with the catalytic properties of copper.



At low overpotentials, CO was promoted in the light while H₂ was suppressed, and at high overpotentials ethylene, methane, formate, and allyl alcohol were enhanced upon illumination.⁵

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- [3] E. B. Creel et al., ACS Energy Lett., 2019, 4, 1098.
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- [5] E. R. Corson et al., Chem. Commun., 2020, 56, 9970.

