

# Xingda Lecture

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**Title:** Enzymatic bioelectrocatalysis: From metabolic pathways to metabolons

**Abstract:** Oxidoreductase enzymes have been employed for almost 5 decades in biosensors and for energy conversion in the form of biofuel cells. However, most enzymatic bioelectrodes in the literature utilize complex biofuels (e.g. glucose), but only partially oxidize the complex biofuel via the use of a single enzyme (i.e. glucose oxidase or glucose

dehydrogenase). This presentation will detail the use of enzyme cascades at bioanodes for deep to complete oxidation of substrates to improve performance (current density and power density), but will focus on the importance of forming metabolons for substrate channeling in multi-enzyme cascades. These enzyme cascades will include natural metabolons (i.e. the Krebs' cycle) and artificial metabolons utilizing DNA and peptides as scaffolds. It will discuss the importance of structural orientation of enzymes and enzyme complexation in enzymatic cascades for efficient bioelectrocatalysis.

**Short resume:** Dr. Shelley Minteer is a USTAR Professor in both the Departments of Chemistry and Materials Science and Engineering at the University of Utah. She received her PhD in Analytical Chemistry at the University of Iowa in 2000 under the direction of Professor Johna Leddy. After receiving her PhD, she spent 11 years as a faculty in the Department of Chemistry at Saint Louis University before moving to the University of Utah in 2011. She also was a Technical Editor for the Journal of the Electrochemical Society and is now an Associate Editor for the Journal of the American Chemical Society. She has published greater than 250 publications and greater than 350 presentations at national and international conferences and universities. She has won several awards including the Luigi Galvani Prize of the Bioelectrochemical Society, the Missouri Inventor of the Year, International Society of Electrochemistry Tajima Prize, Fellow of the Electrochemical Society, and the Society of Electroanalytical Chemists' Young Investigator Award. Her research interests are focused on electrocatalysis and bioanalytical electrochemistry. She has expertise in biosensors and bioelectronics.

## Selected Publications

- R.D. Milton, R. Cai, S. Sahin, S. Abdellaoui, B. Alkotaini, D. Leech, and S.D. Minteer, "[The In Vivo Potential-Regulated Protective Protein of Nitrogenase in Azotobacter vinelandii Supports Aerobic Bioelectrochemical Dinitro-gen Reduction In Vitro,](#)" Journal of the American Chemical Society, 2017, 139, 9044-9052.
- R.D. Milton and S.D. Minteer, "[Direct Enzymatic Bioelectrocatalysis: Differentiating Between Myth and Reality,](#)" Journal of the Royal Society Interface, 2017, 14, 20170253.
- L. Xia, K. Nguyen, Y. Holade, H. Han, K. Dooley, P. Atanassov, S. Banta, and S.D. Minteer, "[Improving the Performance of Methanol Biofuel Cells Utilizing an Enzyme Cascade Bioanode with DNA Bridged Substrate Channeling,](#)" ACS Energy Letters, 2017, 2, 1435-1438.

- S. Abdellaoui, M. Snow, I. Matanovic, A. Stephens, P. Atanassov, and S.D. Minteer, "[Hybrid Molecular/Enzymatic Catalytic Cascade for Complete Electro-oxidation of Glycerol Using a Promiscuous NAD-dependent Formate Dehydrogenase from \*Candida boidinii\*](#)," ChemComm, 2017, 53, 5368 – 5371.
- Y. Liu, D.P. Hickey, J.Y. Guo, E. Earl, S. Abdellaoui, R. Milton, M.S. Sigman, S.D. Minteer, and S. Calabrese Barton, "[Substrate Channeling in an Artificial Metabolon: A Molecular Dynamics Blueprint for an Experimental Peptide Bridge](#)," ACS Catalysis, 2017, 7, 2486–93.
- M. Grattieri, N.D. Shivel, I. Sifat, M. Bestetti, and S.D. Minteer, "[Sustainable Hypersaline Microbial Fuel Cells: Inexpensive Recyclable Polymer Supports for Carbon Nanotube Conductive Paint Anodes](#)," ChemSusChem, 2017, 10, 2053–2058.
- K. Knoche, K. Hasan, E. Aoyama, and S.D. Minteer, "[Role of Nitrogenase and Ferredoxin in the Mechanism of Bioelectrocatalytic Nitrogen Fixation by the Cyanobacteria \*Anabaena variabilis\* SA-1 Mutant Immobilized on Indium Tin Oxide \(ITO\) Electrodes](#)," Electrochimica Acta, 2017, 232, 396–403.
- J. Kitt, D. Bryce, S.D. Minteer, and J.M. Harris, "[Raman Spectroscopy Reveals Selective Interactions of Cytochrome c with Cardiolipin that Correlate with Membrane Permeability](#)," Journal of the American Chemical Society, 139(10), 3851–3860.
- K. Hasan, R. Milton, M. Grattieri, T. Wang, M. Stephanz, and S.D. Minteer, "[Photobioelectrocatalysis of Intact Chloroplasts for Solar Energy Conversion](#)," ACS Catalysis, 2017, 7, 2257–2265.
- C. Serov, D.P. Hickey, M. Cook, S. Robinson, S. Barnett, S.D. Minteer, M.S. Sigman, and M. Sanford, "[A Physical Organic Approach to Persistent, Cyclable, Low-Potential Electrolytes for Flow Battery Applications](#)," Journal of the American Chemical Society, 2017, 139, 2924–2927.
- R.D. Milton, R. Cai, S. Abdellaoui, D. Leech, A.L. De Lacey, M. Pita, and S.D. Minteer, "[Bioelectrochemical Haber–Bosch Process: An Ammonia-Producing H<sub>2</sub>/N<sub>2</sub> Fuel Cell](#)," Angewandte Chemie, 2017, 56(10), 2680–2683.
- R.D. Milton, S. Abdellaoui, D. Dean, L. Seefeldt, D. Leech, and S.D. Minteer, "[Nitrogenase bioelectrocatalysis: heterogeneous ammonia and hydrogen production by MoFe protein](#)," Energy and Environmental Science, 2016, 9, 2550–2554.
- L. Pelster and S.D. Minteer, "[Mitochondrial Inner Membrane Biomimic for the Investigation of Electron Transport Chain Supercomplex Bioelectrocatalysis](#)," ACS Catalysis, 2016, 6, 4995–4999.
- K. Knoche, D.P. Hickey, R.D. Milton, C. Curchoe, and S.D. Minteer, "[Hybrid Glucose/O<sub>2</sub> Biobattery and Supercapacitor Utilizing a](#)

- [Pseudocapacitive Dimethylferrocene Redox Polymer at the Bioanode,](#)” ACS Energy Letters, 2016, 1, 380–385.
- B. Bulutoglu, K.E. Garcia, F. Wu, S.D. Minteer, and S. Banta, “[Direct evidence for metabolon formation and substrate channeling in recombinant TCA cycle enzymes,](#)” ACS Chemical Biology, 2016, 11, 2847–2853.
  - T. Wang and S.D. Minteer, “[Effect of Riboflavin Metabolites on Mitochondrial Electrochemistry,](#)” Journal of the Electrochemical Society, 2016, 163(13), H1047–1052.
  - C. Tapia, R.D. Milton, G. Pankratova, S.D. Minteer, H. Akerlund, D. Leech, A. De Lacey, M. Pita, and L. Gorton, “[Wiring of Photosystem I and Hydrogenase on an Electrode for Photoelectrochemical H<sub>2</sub> Production using Redox Polymers for Relatively Positive Onset Potential,](#)” ChemElectroChem, 2017, 4, 90–95.
  - Y. Holade, D.P. Hickey, and S.D. Minteer, “[Halide-Regulated Growth of Electrocatalytic Metal Nanoparticles Directly on Carbon Paper Electrodes,](#)” Journal of Materials Chemistry A, 2016, 4, 17154–17162.
  - D. P. Hickey, K.L. Knoche, K. Albertson, C. Castro, R.D. Milton, and S.D. Minteer, “[Improving O<sub>2</sub> Reduction at an Enzymatic Biocathode: Mimicking the Lungs,](#)” ChemComm, 2016, 52, 13299–13302.
  - T. Wang, R.D. Milton, S. Adellaoui, D.P. Hickey, and S.D. Minteer, “[Laccase Inhibition by As<sup>3+</sup>/As<sup>5+</sup>: Determination of Inhibition Mechanism and Preliminary Application to a Self-Powered Biosensor,](#)” Analytical Chemistry, 2016, 88, 3243–3248.
  - R.D. Milton, T. Wang, K. Knoche, and S.D. Minteer, “[Tailoring Biointerfaces for Electrocatalysis,](#)” Langmuir, 2016, 32, 2291–2301.
  - I. Wheeldon, S.D. Minteer, S. Banta, S. Calabrese Barton, P. Atanassov, and M. Sigman, “[Substrate channeling as an approach to cascade reactions,](#)” Nature Chemistry, 2016, 8, 299–309.
  - D.P. Hickey, D. Schiedler, I. Matanovic, P. Doan, P. Atanassov, S.D. Minteer, and M. Sigman, “[Predicting Electrocatalytic Properties: Modeling Structure–Activity Relationships of Nitroxyl Radicals,](#)” Journal of the American Chemical Society, 2015, 137, 16179–16186.