

# Liquid-Phase, Acid-Base Catalytic Conversion of Biomass to Fuels and Chemicals

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The prevalence of “light” (C1-C3) hydrocarbons obtained from shale gas requires us to find alternative sources of larger molecules suitable for producing commodity and specialty chemicals. Moreover, production of these species cannot be fully decarbonized, because the final products contain embodied carbon. Biomass is an attractive source of de-fossilized carbon, and it natively contains the oxygen needed to produce high-value chemicals. Indeed, significant oxidation is a feature of most renewable carbon resources, and a key challenge for producing renewable chemicals is to selectively de-functionalize biomass, CO<sub>2</sub>, and other such feedstocks.

In this presentation, we will discuss a strategy for converting highly oxygenated biomass species into high-value chemicals and fuels, following a platform chemical approach. In one example, we take advantage of the chirality of the C5 carbon of glucose to produce enantiomerically pure (S)-3-hydroxy- $\gamma$ -butyrolactone (HBL), a key precursor in the synthesis of a variety of crucial pharmaceuticals.  $\beta$ -D-Glucose is first converted to a labile intermediate we refer to as ‘trione’ by whole-cell enzyme oxidation by pyranose-2-oxidase and dehydration by a modified aldose-2-ulose dehydrase. Trione is then upgraded to a chiral glycolate ester of 3,4-dihydroxybutyric acid via a retro-Aldol reaction catalyzed by weak homogeneous bases (e.g., sodium bicarbonate or piperazine). Finally, the glycolate ester is acidified with HCl, leaving (S)-HBL (with glycolic acid as a byproduct). The overall process occurs at ambient temperature in the aqueous phase and yields (S)-HBL from glucose at 88% yield. Preliminary techno-economic analysis suggests the process is viable compared to existing alternatives.

In another example, we develop mechanistic insight into base-catalyzed ethanol dehydrogenation (as a forerunner to C-C bond formation ultimately producing butadiene). Reaction kinetics measurements are combined with in situ spectroscopy and site titrations to identify the importance of Mg-O Lewis acid-base site pairs for achieving high selectivity. This insight is then used to guide development of a highly porous magnesium oxide, synthesized from Mg-MOF-74 via thermolysis. The activity of the MOF-derived MgO catalyst was evaluated using the Knoevenagel condensation of benzaldehyde with malononitrile as a model reaction. The rate obtained using the MOF-derived material was a factor of 5 greater than that for a traditional MgO. XRD analysis indicates smaller MgO crystallites in the MOF-derived MgO, and the higher activity observed over this catalyst was attributed to a higher density of basic sites, as probed by CO<sub>2</sub>-TPD and DRIFTS.

A key feature that unifies these examples is the impact of the solvent on not only the activity of the catalyst but also the selectivity to desired reaction products. We will conclude our presentation by highlighting recent efforts to disentangle how the identity of the solvent impacts catalytic reaction rates.



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Prof. Thomas J. Schwartz earned BS degrees in Chemical Engineering and Biological Engineering from the University of Maine and a PhD at University of Wisconsin under the supervision of Prof. James Dumesic. Schwartz joined the Department of Chemical and Biomedical Engineering at the University of Maine in 2015 as an Assistant Professor and was promoted to Associate Professor in 2021. His research group seeks to develop a molecular-level understanding of processes that occur on catalytic surfaces used for the conversion of carbon-based feedstocks (e.g., biomass, petroleum, natural gas, waste polymers) to chemicals and fuels. Since 2023 he has also been the Associate Director of the Forest Bioproducts Research Institute, where he helps lead a larger team pursuing scale-up of chemical processes, including “Thermal DeOxygenation” for producing SAF and marine diesel. He is active in the ACS Division of Catalysis Science & Technology, the AIChE Catalysis and Reaction Engineering Division, and the North American Catalysis Society, and he serves on the Early Career Editorial Board for the Journal of Catalysis and serves as an editor for Scientific Reports. Schwartz received an ACS PRF Doctoral New Investigator award in 2016, an NSF EPSCoR RII Fellowship in 2020, and the NSF CAREER Award in 2021. He has been honored by the University of Maine with the College of Engineering Early Career Research Award in 2018 and the Pulp and Paper Foundation’s Joseph M. Genco Award in 2020.

