Understanding and Controlling Electrochemistry for Electrolyzers and Batteries

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This talk addresses the electrochemical reactivity associated with electrolyzers and batteries. Relevant to electrolyzers we show that electrodeposition of CuAg or CuSn alloy films under suitable conditions yields high surface area catalysts for the active and selective electroreduction of CO_2 to multi-carbon hydrocarbons and oxygenates. Alloy films containing Sn exhibit greater efficiency for CO production relative to either Cu along or CuAg at low overpotentials. *In-situ* Raman and electroanalysis studies suggest the origin of the high selectivity towards C_2 products to be a combined effect of the diminished stabilization of the Cu_2O overlayer and the optimal availability of the CO intermediate due to the Ag or Sn incorporated in the alloy. Sn-containing films exhibit less Cu_2O relative to either the Ag-containing or neat Cu films, likely due to the increased oxophilicity of the admixed Sn. Incorporation of a polymer into the Cu electrodeposit leads to very active CO_2 reduction electrocatalysis due to pH changes at the electrified interface. Vibrational spectroscopy is used to evaluate the pH at the interface during electrolyzer operation.

Relevant to batteries, we discuss solid electrolytes (SEs) which have become a practical option for lithium ion and lithium metal batteries due to their improved safety over commercially available ionic liquids. The most promising of the SEs are the thiophosphates whose excellent ionic conductivities at room temperature approach those of commercially-utilized electrolytes. Hybrid solid-liquid electrolytes exhibit higher ionic conductivities than their bare solid electrolyte counterparts due to decreased grain boundary resistance, enhanced interfacial contact with electrodes, and decreased degradation at the interface. Spectroscopic and structural studies on these latter materials lead to new formulations and artificial SEI materials exhibiting advantageous properties.