

Molecular Electrocatalysis for Hydrogen Evolution and Environmental Remediation

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Electrocatalytic proton reduction to form dihydrogen (H_2) is an effective way to store energy in the form of chemical bonds. In this talk, I will discuss the applicability of a main-group-element-based tin porphyrin complex as an effective molecular electrocatalyst for proton reduction. A PEGylated Sn porphyrin complex displayed high activity and high selectivity in acetonitrile. The maximum turnover frequency (TOF_{max}) for H_2 production was obtained as $1,099\ s^{-1}$. Spectroelectrochemical analysis, in conjunction with quantum chemical calculations, suggest that proton reduction occurs via an Electron-Chemical-Electron-Chemical pathway. This study reveals that the tin porphyrin catalyst serves as a novel platform for investigating molecular electrocatalytic reactions and provides new mechanistic insights into proton reduction. In this talk, I will also discuss a separate study of electrocatalytic per- and polyfluoroalkyl substances (PFASs) mineralization using a copper(I) catalyst. Groundwater reservoirs contaminated with PFAS need purifying remedies. Perfluorooctanoic acid (PFOA) is the most abundant PFAS in drinking water. Although different degradation strategies for PFOA have been explored, none of them disintegrates the PFOA backbone rapidly under mild conditions. I will present a molecular copper electrocatalyst that assists in the degradation of PFOA up to 93% with 99% defluorination rate within 4 h of cathodic controlled-current electrolysis. Free F^- , CF_3COO^- , CF_3H , and CF_4 were detected as fragmented PFOA products along with the evolution of CO_2 using gas chromatography, ion chromatography, and gas chromatography-mass spectrometry techniques, suggesting comprehensive cleavage of C–C bonds in PFOA.