**Innovative Strategies for Nonconventional Water Splitting and Beyond**

**Abstract:** One of the challenges to realize large-scale H2 production from water electrolysis is the lack of competent and inexpensive electrocatalysts. Our group recently discovered that surface modification of metallic nickel with nitrogen resulted in a superior electrocatalyst for H2 evolution under neutral condition, rivaling the performance of the state-of-the-art platinum-based catalysts. In addition, since water oxidation is more kinetically challenging while its product O2 is not of significant value, we are also interested in exploring alternative oxidation reactions which can not only provide electrons for H2 evolution at the cathode but also yield value-added organic products at the anode. Within this context, biomass-derived intermediate compounds are found to be suitable candidates for electrocatalytic upgrading, which can be integrated with H2 production under alkaline conditions. Finally, a new electrolyzer design for decoupled water splitting and organic transformation will be presented, which is able to substantially reduce voltage inputs and mitigate the H2/O2 mixing issue commonly encountered in conventional water electrolysis.