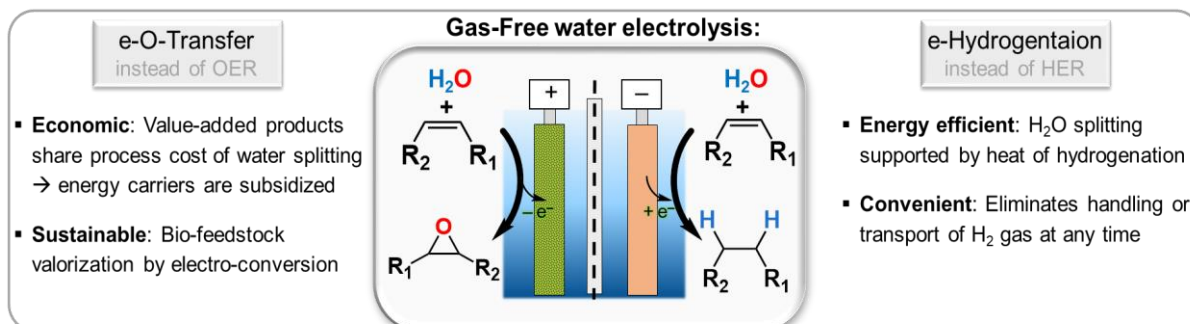


Transition Metal Catalyzed Electro-Organic Synthesis with Water & Ex Situ Electrolysis for Unique Selectivity

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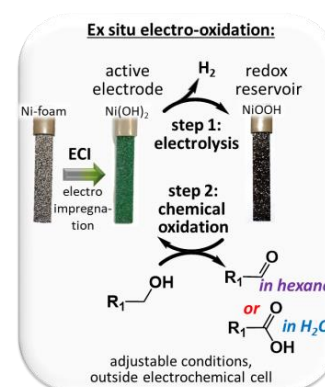


H₂ production by water electrolysis is proposed to store excess renewable electricity in chemical energy carriers. O₂ evolution is the typically paired anode reaction in standard electrolyzers, but the unsalable side product O₂ cannot support process costs.

We develop **electrocatalytic oxygen atom transfer (e-OAT)**, to replace O₂ evolution by anodic value-added product formation together with water electrolysis-based energy carrier production. A specific target reaction is the electro-epoxidation of alkenes with H₂O.^[1]

Besides cost, H₂ gas storage is another challenge, for which liquid organic hydrogen carriers (LOHCs) emerged as a potent solution. In this context, we develop **direct electro-hydrogenation of alkenes,^[2] organic carbonyls, and aromatic systems with H₂O in one step**, mediated by transition metal pincer complexes and porphyrins. Mild overpotentials (energy efficiency) and suppression of typically dominant parasitic HER (current efficiency), is achieved by structure-reactivity tuning of catalysts and the design of unique mechanisms, including H₂O activation instead of H⁺ reduction.

Beyond classic *in situ* electro-synthesis (substrates added into electrolyzer), we develop **ex situ electro-synthesis—a new concept to separate chemical and electrochemical reaction steps in time and space for unrestricted flexibility in reaction design.**^[3] Exemplarily, we use the reversible Ni(OH)₂ / NiOOH couple for short term storage of redox equivalents. The bulk electro-conversion of Ni(OH)₂ to NiOOH is performed together with energy carrier (H₂) production in an electrolyzer. The chemical oxidation of organic substrates to value-added products is then performed outside the electrolyzer under freely choosable reaction conditions (solvent, temperature, pressure), which allows orthogonal selectivity for products that are inaccessible to traditional electro-synthesis.



References:

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