Design of New Catalytic Reactions for "Green" Synthesis and Sustainable Energy

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We have developed a new mode of metal-ligand cooperation, based on aromatization-dearomatization of pincer-type ligands, and have designed environmentally benign catalytic reactions based on such cooperation, including (a) dehydrogenative coupling of alcohols to form esters and H₂ (b) hydrogenation of esters to alcohols under mild conditions (c) coupling of amines with alcohols to form amides with liberation of H₂ (d) selective synthesis of primary amines directly from alcohols and ammonia (e) direct formation of acetals by dehydrogenative coupling of alcohols. These reactions are efficient, proceed under neutral conditions and produce no waste. They are catalyzed by pincer-type ruthenium complexes based on pyridine and on acridine, and involve as a key mechanistic step aromatizationdearomatization of the hetero-aromatic ligand core. Metal-ligand cooperation of this type has very recently led also to a distinct approach towards water splitting, based on consecutive thermal H₂ generation and light-induced O₂ liberation.