DFT drives catalysis by predictions

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Amidst the widespread enthusiasm for machine learning, one often-overlooked domain is predictive catalysis. In the realm of computational chemistry for sustainability, the research group advocates the maximum utilization of predictive catalysis, employing machine learning principles. Their endeavors extend beyond identifying reaction mechanisms; once the rate-determining step (rds) is established, the focus shifts to exploring alternative catalysts, aiming for more benign reaction conditions. The computational research spans various domains, encompassing processes such as olefin metathesis using Ru-based catalysts, gold chemistry for organometallic reactions, and green chemistry strategies for CO2 avoidance or reduction, including water oxidation catalysis and alcohol transformation to aldehydes with H2 generation as an energy source [1].

DFT calculations have unveiled the mechanisms underlying the formation of N-substituted hydrazones through the coupling of alcohols and hydrazine, achieved via sequential processes of acceptorless dehydrogenation and borrowing hydrogen [2,3]. This process, facilitated by a Mn-PNN pincer-based catalyst, aligns with green chemistry principles, releasing water and H2 as environmentally friendly byproducts. [3].

The research also delves into the reductive amination of aliphatic carbonyl compounds catalyzed by a Knölker-type iron catalyst. Utilizing DFT calculations and a detailed chemical structure analysis, the team investigates the reaction mechanism [4]. Armed with insights into the mechanism, various catalyst modifications are explored with the goal of steering catalytic reactions towards milder conditions (see Figure 1).

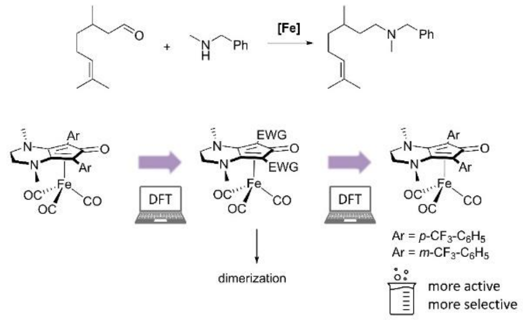

Figure 1. Two-step predictive catalysis approach for hydrogenation reactions with Knölker-type catalysts. A series of derivative Renaud catalysts with electron-withdrawing groups on the aryls of the cyclopentadienone improves the activity and selectivity.

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