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Abstract:

3d metals serve as excellent alternatives to heavy metals in catalysis due to lower toxicity, cost- effectiveness, and greater abundance of the former. However, first-row transition metals typically undergo single electron processes. To replicate the two-electron chemistry of heavy metals, redox-active ligands are often an attractive choice as they act as a redox reservoir or sink. Ideally, the metal-ligand covalency developed between the base metal and the redox active ligand facilitates the redox process in tandem. Our keen interest in base-metal catalysis, where ligands play a pivotal role in reversible electron transfer, led us to combine base metals with redox-active ligands to explore their catalytic potential. In this lecture, we present a compelling case involving pincer and formazan ligands, showcasing their redox non-innocence in various homogeneous catalytic transformations. In these instances, redox-active ligands predominantly regulate substrate activation. The study involves a series of control reactions, isolation of critical intermediates, spectroscopic analysis, and DFT calculations, providing substantial evidence for ligand-based electron transfer that facilitates the reductive cleavage of strong carbon-halide bonds in the substrate. This method successfully achieved several challenging catalytic transformations, underscoring the significant role played by ligands in base metal catalysis. We further hope that these examples will inspire future exploration of the untapped potential within various redox-active ligand frameworks that might be applied to important chemical transformations.

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