Special Organic Colloquium

Presents:

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“Development of Direct C-H Amination Reactions:
Inner- versus Outer-Sphere Pathways”

Abstract: The direct conversion of hydrocarbons to valuable heterocyclic scaffolds is a dream reaction, because it will open the door to many new chemical applications in synthesis. Although tremendous progress has been made, our current ability to prepare arylamines and heterocycles via direct C–H functionalization is limited. A novel methodology is developed that employs transition metal-based catalysts and oxazolone substrates to access short-lived metal-nitrenoid complexes, which are key intermediates in the efficient construction of thus far elusive γ-lactams through direct C–H bond amidation. Stoichiometric studies with robust carbonylnitrene precursor, 1,4,2-dioxazol-5-ones, suggest that the insertion of C–H into metal-nitrenoid moiety is possible and mechanistic clues from the initial proof-of-concept studies further enabled the design of efficient and versatile catalysts that allow for the straightforward amidation of various sp\(^3\) and sp\(^2\) C–H bonds with exceptional selectivity leading to lactam products. The power of this new method is demonstrated in the successful late-stage functionalization of bio-active molecules with amino acid derivatives to produce molecules that are highly sought after for pharmaceutical and other applications in synthesis. We have also developed direct amidation and amination reactions of hydrocarbons using organic azides as the amino source releasing molecular nitrogen as the single by-product. The reaction is catalyzed by a cationic group 9 metal complexes under external oxidant-free conditions, and a broad range of chelate group-containing substrates are selectively aminated with excellent functional group tolerance, thereby opening a new avenue to environmentally benign carbon-nitrogen (C-N) bond formation.

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