Denver

CHEMISTRY

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Seminar Series

11am-12pm
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Zoom

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"Towards Mechanistically Guided Design of Transition Metal Catalysts for C-H Bond and Olefin Functionalization"

Due to limited mechanistic insights and the complexity of catalyst-substrate interactions, catalyst design and optimization in transition metal catalysis are often based on chemical intuition and experimental trial-and-error. I will discuss our recent efforts towards a streamlined computational approach to understand and predict the reactivity and selectivity of various transition metal catalyzed reactions, including the functionalization of C-H/C-C bonds and olefins, as well as olefin polymerization reactions.

We use computational tools to investigate reaction mechanisms and steric, electronic, dispersion, and strain effects of the transition metal catalyst. We utilized the energy decomposition analysis (EDA) methods to quantitatively analyze different types of through-bond and through-space interactions between the transition metal catalyst and the substrate. These studies provided a straightforward way to identify the dominant factor controlling reactivity and selectivity. We demonstrated these theoretical insights can facilitate catalyst discovery. In collaborations with synthetic organic chemistry groups, we employed a mechanistically-guided catalyst design approach to develop new catalysts for the Cu-catalyzed olefin hydroamination and the Rh-catalyzed asymmetric allenic Pauson-Khand reactions. We also showed that the in-depth analysis of catalyst-substrate interactions in transition states can aid the development of multivariate mathematical equations for the rapid prediction of catalyst and initiator reactivities in the Cu-catalyzed atom transfer radical polymerization (Cu-ATRP). Recently, we have applied these computational approaches to mechanistically more complex reactions, including multi-component coupling reactions with open-shell first-row transition metal catalysts and conformationally flexible ligands, and glycosylation reactions involving strong interactions with solvent molecules.