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Transition Metal-Catalyzed Amination and Amidation Reactions

Research focuses on the development of transition metal-catalyzed reactions to access important organic products in a single step from easily accessible functionalities, ideally, without the generation of any stoichiometric byproducts.

Abstract
Carbon–nitrogen bonds are ubiquitous in pharmaceuticals, organic materials, and natural products. These C–N bonds are often incorporated as amines or amides. Despite their prominence, they are often formed in poor atom and step economy. One of the primary goals of the Hull Group is to develop alternative syntheses of this two important functionalities.

Hydroamination, the addition of an amine across an alkene or alkyne, couples two readily available functional groups to form new C–N and C–H bonds with 100% atom economy. Our group has demonstrated that Lewis basic groups proximal to the olefin can coordinate to a cationic rhodium catalyst and promote a regio-, chemo-, and stereoselective hydroamination reaction for the synthesis of 1,2- and 1,4-diamines. The reactions proceed selectively through five-membered metallacyclic intermediates, which is the key to the high selectivity observed.

Oxidative amidation reactions are a promising approach to the synthesis of amides; in this approach alcohols and amines are coupled directly through dehydrogenation or transfer hydrogenation processes. The Hull Group has demonstrated that allylic alcohols or aldehydes undergo a selective oxidative coupling reaction for the synthesis of amides. The reactions are highly chemoselective for reacting with the allylic alcohol or aldehyde, as other alcohols and alkenes are well tolerated under the reaction conditions. Further, primary and secondary amines as well as anilines are readily coupled to afford the corresponding amide.

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