

ORGANIC SEMINAR

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Access to “Inaccessible” Arynes and Redox Chemistry Using Transition Metals



COURTNEY ROBERTS

Assistant Professor
University of Minnesota

Abstract:

Research in the Roberts group involves looking at unsolved problems in organic synthesis through the perspective of organometallic/inorganic chemistry. One main area of interest for the group is the synthesis of heterocycles through aryne intermediates. Despite their useful reactivity, a number of challenges still remain in the use of arynes including problems with regioselectivity and the synthesis of N-heterocyclic arynes. Using fundamental principles of Ni chemistry, our group is the first to be able to access previously “inaccessible” 5-membered heterocyclic arynes for the first time since they were hypothesized to exist 120 years ago. We are also the first group to demonstrate catalyst controlled regioselectivity in arynes, where all previous examples operated under substrate control. Another challenge in organic synthesis lies in alkyl–alkyl cross-coupling. This is due to challenges with oxidative addition and off cycle pathways such as beta-hydride elimination. Our group has pioneered the use of Group 3 metal catalysts supported by redox-active ligands to overcome some of these challenges. Using 10 mol% of a Sc, Y, or Lu tris(amido) catalyst, coupling partners that both have beta-hydrogens can be successfully cross-coupled for the first time using early transition metals. These improvements related to organic synthesis can only be accessed using inorganic/organometallic chemistry.



Department of Chemistry