

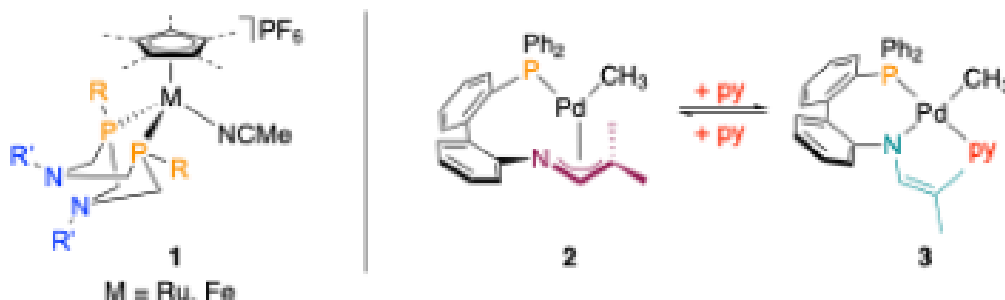
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Harnessing Ligand Dynamics for Sustainable Catalysis

Abstract:

Factors that promote sustainable catalysis include catalysts that can be used at very low catalyst loadings, and those that promote enabling transformations with minimal waste generation. These goals can be achieved with specially designed catalyst and ligand structures. We have pursued the development of catalysts with cooperative ligands that can assist catalytic turnover through reversible changes, such as proton shuttling,^[1] and changes to ligand coordination mode.^[2] Both of these ligand types will be covered in this presentation. First, we have demonstrated that $[\text{Ru}(\text{Cp})(\text{P}^{\text{R}}_2\text{N}^{\text{R}'}_2)(\text{MeCN})]\text{PF}_6$ catalysts (**1**) are active catalysts for alkyne functionalization, which depends on proton shuttling by the pendant tertiary amine groups of the $\text{P}^{\text{R}}_2\text{N}^{\text{R}'}_2$ ligand.^[3] Mechanistic insight has guided catalyst redesign strategies, as well as redeployment toward intermolecular C-O bond formation. The second portion of the presentation will cover phosphine 1-azaallyl ($\text{P}^{\text{A}}\text{AzA}$) complexes, which we designed to exploit dynamic changes in coordination mode. The $\text{P}^{\text{A}}\text{AzA}$ ligand effectively stabilizes operationally unsaturated metals via $\eta^3\text{-NCC}$ coordination of the 1-azaallyl moiety (e.g., **2**).^[4] Yet, it can permit ligand (or substrate) binding through a change in coordination to $\kappa^1\text{-N}$ (e.g., **3**). This, and other reactivity of **2**, will be discussed.



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- [3] D. E. Chapple, M. A. Hoffer, P. D. Boyle, J. M. Blacquiere, *Organometallics* 2022, 41, 1532-1542.
- [4] M. B. Kindervater, V. N. Staroverov, K. M. K. Jackman, A. A. Fogh, L. S. G. Kelley, L. Lim, S. A. Sirohey, P. D. Boyle, J. M. Blacquiere, *Dalton Trans.* 2023, 52, 10744-10750.

Bio:

Johanna Blacquiere obtained a B.Sc. (2005) from Mount Allison University, where she was a member of Steve Westcott's Wild Toads. She obtained a Ph.D. (2011) at the University of Ottawa under the supervision of Deryn Fogg, and completed an NSERC postdoctoral fellowship in the laboratory of Jim Mayer at the University of Washington. In 2013, she began her independent career at Western University and in 2019 she was promoted to Associate Professor. Her research program involves the design and mechanistic elucidation of transition metal catalysts for the low-waste synthesis of high-value organic compounds. Her expertise is in cooperative homogeneous catalysis, in which ligands undergo reversible changes in protonation state or coordination mode to enhance catalyst performance. Blacquiere's research program has been recognized with a number of awards, including the 2025 Strem Award for Pure or Applied Inorganic Chemistry, and a 2024 Western University Faculty Scholar Award.