

Development of Novel C–H Functionalization Methodologies and Beyond

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ABSTRACT

We have developed a set of new transition metal-catalyzed C-H functionalization methodologies employing various tether strategies. These methods feature: (a) use of silyl group as a tether between a substrate and a reagent, thus transforming intermolecular reaction into an intramolecular reaction; (b) employment of a silicon-tethered directing group, which is traceless or easily convertible into valuable functionalities; (c) use of reactive silyl-tethered hydrosilane group; and (d) introduction of new N/Si-chelation concept that allows for a remote activation of aliphatic C-H bonds.

We uncovered new reactivity of hybrid Pd-radical species, generated at room temperature under visible light without use of exogenous photosensitizers. This led to the development of novel transformations, including new types of Heck reaction, aliphatic C–H functionalization methods, as well as new cascade transformations. A set of both directed and direct functionalization methods have been developed.

The scope of these transformations will be demonstrated, and the mechanisms will be discussed.
