화학과 세미나

이윤미 교수
광운대학교 화학과

Catalytic Regiodivergent Hydrofunctionalization of Allenes using Diisobutylaluminum Hydride

Allenes, featuring orthogonal π-systems within a cumulated diene framework, exhibit unique reactivity and serve as versatile building blocks in organic synthesis. However, their selective hydrofunctionalization remains challenging due to intrinsic difficulties in controlling regio- and stereoselectivity. To address these limitations, we have developed a catalytic regiodivergent hydrofunctionalization platform for di- and trisubstituted allenes using diisobutylaluminum hydride (DIBAL-H) as a key hydride source. First, a Cu-catalyzed hydroalumination of allenes was established, enabling highly regioselective formation of allylaluminum intermediates. These organoaluminum species serve as versatile synthetic handles and undergo diverse C–C and C–heteroatom bond-forming reactions with electrophiles such as CO2, isocyanides, cyanating agents, aldehydes, and iminium salts. This strategy provides rapid access to structurally diverse molecules, including products bearing quaternary carbon centers, with excellent yields and selectivity. Furthermore, we developed a regiodivergent hydroarylation platform that enables selective α-, β-, and γ-arylation of unactivated allenes through catalyst control. For α- and γ-selective hydroarylation, a cooperative Cu/Pd dual catalytic system employing DIBAL-H was developed, where ligand-controlled transmetallation governs regioselectivity. In contrast, a complementary palladium-catalyzed β-selective hydroarylation provides efficient access to tetra-substituted alkenes with high stereoselectivity. This operationally simple, modular, and regioselective protocol offers a general platform for allene hydrofunctionalization and significantly expands the synthetic utility of Cu–H, Pd–H catalysis and organoaluminum intermediates in modern allene chemistry.

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