

New Chemistry beyond Metathesis Catalyzed by Ruthenium Alkylidenes

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The main utility of Grubbs-type ruthenium alkylidenes involves metathesis chemistry. These ruthenium complexes, however, have proven their potential as a precatalyst for many non-metathetic transformations such as hydrogenation, dihydroxylation, and cyclopropanation. In light of expanding the scope of enyne metathesis, we streamlined enyne metathesis and metallotropic [1,3]-shift such that various enyne-based functional groups could be constructed by the tandem catalysis of ruthenium alkylidenes. While exploring these tandem reactions of bis-1,3-diyne substrates, unprecedented hydrovinylation reaction was discovered when ethylene was employed as an alkene counterpart. More interestingly, these bis-1,3-diynes could undergo hexadehydro Diels-Alder reaction to form aryne intermediates, which were hydrohalogenated by ruthenium alkylidenes complexes in the presence of halohydrocarbons. In this presentation, the scope and utility of these powerful and novel transformations will be discussed in the context of their discovery and application to natural product synthesis.

