Formation of C-C Bonds via Catalytic Hydrogenation and Transfer Hydrogenation

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Abstract: Carbon-carbon bond formation lies at the heart of chemical synthesis. Research in the Krische laboratory reveals reductive C-C bond formation can be achieved under the conditions of catalytic hydrogenation. This concept is extended further via “C-C bond forming transfer hydrogenations”, wherein hydrogen exchange between alcohols and π-unsaturated reactants triggers generation of aldehyde-organometal pairs that combine to give products of carbonyl addition. Direct alcohol CH-functionalization via redox-triggered carbonyl addition enhances efficiency by merging alcohol-to-aldehyde redox reactions with C-C bond construction events. This new pattern of reactivity enables the direct conversion of lower alcohols to furnish higher alcohols in the absence of premetallated reagents. These methods have been applied to the construction of diverse polyketide natural products. In each case, the most concise route to any member of the respective natural product family was achieved.