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| Title: | Nickel-Catalyzed Selective Synthesis of α-Alkylated Ketones via Dehydrogenative Cross-Coupling of Primary and Secondary Alcohols |
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| Abstract: | Herein, we describe an isolable, air-stable, homogeneous, nickel catalyst that performs dehydrogenative cross-coupling reaction between secondary and primary alcohols to result α-alkylated ketone products selectively. The sequence of steps involve in this one-pot reaction is dehydrogenation of both alcohols, condensation between the ketone and the aldehyde, and hydrogenation of the in situ-generated α,β-unsaturated ketone. Preliminary mechanistic investigation hints a radical mechanism following borrowing hydrogen reaction. The construction of C−C bond with structural diversity and complexity is quintessential in organic synthesis.1 Traditional approaches toward their fabrication include nucleophilic attack by an appropriate C-nucleophile to the alkyl halide or similar alkylating agent.2 This approach often requires cryogenic conditions, employs toxic or mutagenic alkyl halides and generates a copious amount of inorganic waste. Henceforth, alternative methods are intensely sought after that can be environmentally benign, atom- and process-efficient by utilizing cheap, nontoxic starting materials. In this regard, borrowing hydrogen (BH) approach provides a very promising solution, where biomass-derived, abundantly available alcohols can be the starting feedstocks.3 In BH method, hydrogen is extracted from the alcohol, kept stored in the catalyst and at the final step the hydrogen is redelivered to an in situ-generated bond to accomplish efficient forging of C−C bond, giving water as the sole by-product. |
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