Expert Opin Environ Biol 2018, Volume 7
DOI: 10.4172/2325-9655-C3-027

A sustainable catalyst for dicarbofunctionalization of unactivated olefins and its application to the synthesis of lignan natural products

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The dicarbofunctionalization of unactivated olefins using a sustainable catalyst via cyclization/cross-coupling could afford a straightforward synthetic route to complex carbon scaffolds relevant to natural products, bioactive molecules, and pharmaceuticals. However, interception of C(sp3)-M species, which are generated as intermediates in catalytic reactions, is a formidable challenge due to their high propensity to undergo β-H elimination that leads to the formation of Heck products. Herein, we report a Ni/terpyridine catalyst that overcomes this challenge and enable to develop environmentally improved routes and processes to important products. This reaction protocol provides rapid access to (arylmethyl)carbo- and heterocyclic scaffolds, which occur widely as structural cores in various natural products and bioactive molecules. We applied this new method for the concise synthesis of six lignan natural products containing three different structural frameworks in gram-scale quantities. Mechanistic studies with radical probes and product selectivities show that the current reaction proceeds via a single electron transfer (SET) process.

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