Title:	Borrowing Hydrogen-Mediated N-Alkylation Reactions by a Well-Defined Homogeneous Nickel Catalyst
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Abstract:	We report herein a well-defined and bench-stable azo-phenolate ligand-coordinated nickel catalyst which can efficiently execute N-alkylation of a variety of anilines by alcohol. We demonstrate that the redox-active azo ligand can store hydrogen generated during alcohol oxidation and redelivers the same to an in-situ-generated imine bond to result in N-alkylation of amines. The reaction has wide scope, and a large array of alcohols can directly couple to a variety of anilines. Mechanistic studies including deuterium labeling to the substrate establishes the borrowing hydrogen method from alcohols and pinpoints the crucial role of the redox-active azo moiety present on the ligand backbone. Isolation of the ketyl intermediate in its trapped form with a radical quencher and higher kH/kD for the alcohol oxidation step suggest altogether a hydrogen-atom transfer (HAT) to the reduced azo backbone to pave alcohol oxidation as opposed to the conventional metal–ligand bifunctional mechanism. This example clearly demonstrates that an inexpensive base metal catalyst can accomplish an important coupling reaction with the help of a redox-active ligand backbone.
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