

Multidentate Organocatalysis in Asymmetric Synthesis

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Counterion-mediated asymmetric induction is an effective strategy in organocatalysis or metal-based catalysis. We have initiated an investigation of this strategy in the Morita-Baylis-Hillman (MBH) reaction to address its long-standing rate/substrate scope issue. Multifunctional organocatalysts, each containing a Lewis base, a Brønsted base, and a Brønsted acid, have been rationally designed and developed to catalyse this reaction with positive cooperativity. The Brønsted base acts as the switch point to the counterion catalytic pathway, in response to the addition of a strong Brønsted acid, to furnish fast reaction rates with good enantioselectivity at ambient temperature, with the counteranion of the Brønsted acid additive controlling the sense of the asymmetric induction. Second and third generations of this new class of organocatalysts have shown improved enantioselective rates and substrate scope. Multifunctional, or multidentate, organocatalysis, with positive cooperativity and regulation, may lead to new approaches to enzyme mimicking catalysis with proficiency.