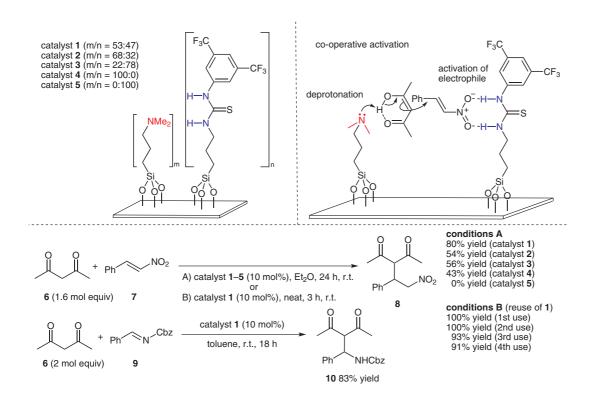
A. PUGLISI,* R. ANNUNZIATA, M. BENAGLIA,* F. COZZI, A. GERVASINI, V. BERTACCHE, M. C. SALA (UNIVERSITÀ DEGLI STUDI DI MILANO, ITALY)

Hybrid Inorganic-Organic Materials Carrying Tertiary Amine and Thiourea Residues Tethered on Mesoporous Silica Nanoparticles: Synthesis, Characterization, and Co-Operative Catalysis

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Michael Additions Catalyzed by Bifunctional Mesoporous Silica



Significance: The Michael reaction mediated by mesoporous silica-supported amine-urea bifunctional catalysts is reported in which the tertiary amine and the urea moieties work as a base and an acid, respectively. The reaction of acetylacetone (6) with 2-nitrostyrene (7) was carried out with bifunctional catalyst 1 (amine/urea = 53:47) under conditions A to give 3-[1-(1-phenyl-2-nitroethyl)]-2,4-pentadione (8) in 80% yield. When catalysts 2-5 were used, product 8 was obtained in 0-56% yield. Catalyst 1 was reused three times under conditions B to afford product 8 in high yields.

Comment: The reaction of Cbz-benzaldimine (9) and 6 with catalyst 1 in toluene was also performed to afford product 10 in 83% yield. Catalysts 1-5 were prepared in accordance to the previously reported procedure (V. S.-Y. Lin and coworkers Angew. Chem. Int. Ed. 2005, 44, 1826). Enantioselective Michael addition of malonates to nitroolefins using bifunctional organocatalysts was reported by Takemoto and co-workers (J. Am. Chem. Soc. 2003, 125, 12672).

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thiourea residues

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