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Mesoporous Silica with Site-Isolated Amine and Phosphotungstic Acid Groups: A Solid Catalyst with Tunable Antagonistic Functions for One-Pot Tandem Reactions

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One-Pot Tandem Reactions with a Bifunctional Solid Catalyst

Preparation of mesoporous silica catalyst **3**:

Reaction pathway:

Selected results:

Significance: A mesoporous silica catalyst **3** bearing both Brønsted base and acid groups was prepared by immobilization of 3-aminopropyltriethoxysilane [(EtO)₃Si(CH₂)₃NH₂] and phosphotungstic acid (H₃PW₁₂O₄₀) to a mesoporous silica. Catalyst **3** drove the tandem deacetalization–Henry reaction and deacetalization–Knoevenagel reaction of **4** with nitromethane and malononitrile to give *trans*-1-nitro-2-phenylethylene **5** and benzylidene malononitrile **6** in 92% and 91% yield, respectively.

Comment: The acid/base properties were controlled by the ratio and surface concentration of an amino group and phosphotungstate. Catalyst **3** was characterized by XANES (X-ray absorption near-edge structure spectrum), BET, BJH, ³¹P NMR, ²⁹Si and ¹³C CP-MAS NMR spectroscopy and GC-MS. Catalyst **3** was recovered by filtration and reused several times without significant loss of catalytic activity (4th use: **5** in 90% isolated yield, **6** in 91% isolated yield).

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