

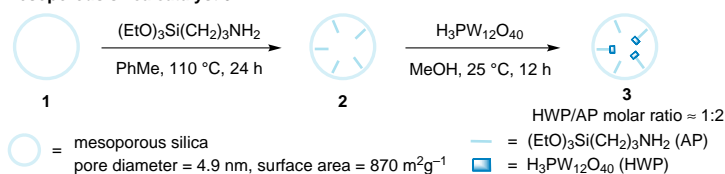
N. R. SHIJU,* A. H. ALBERTS, S. KHALID, D. R. BROWN, G. ROTHENBERG* (UNIVERSITY OF AMSTERDAM, THE NETHERLANDS; UNIVERSITY OF HUDDERSFIELD, UK AND BROOKHAVEN NATIONAL LABORATORY, UPTON, USA)

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

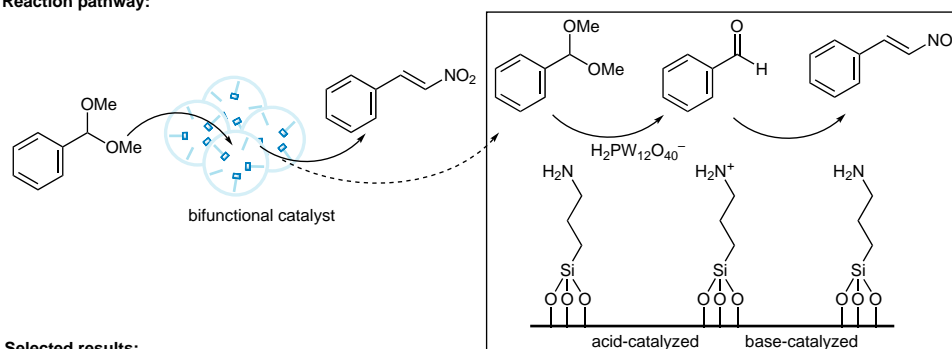
Angew. Chem. Int. Ed. 2011, 50, 9615–9619.

One-Pot Tandem Reactions with a Bifunctional Solid Catalyst

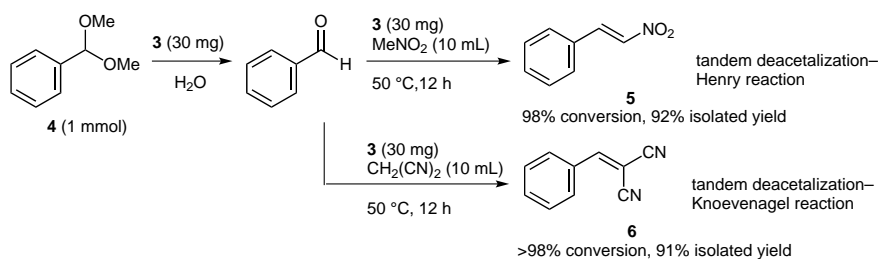
Preparation of mesoporous silica catalyst 3:



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).

SYNFACTS Contributors: Yasuhiro Uozumi, Yoichi M. A. Yamada, Heeyoel Baek

Synfacts 2012, 8(1), 0103 Published online: 19.12.2011

DOI: 10.1055/s-0031-1289491; Reg-No.: Y14511SF