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Nano-ordered B-MCM-41: An efficient and recoverable solid acid catalyst for three-component Strecker reaction of carbonyl compounds, amines and TMSCN

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Abstract The three-component Strecker reaction of a wide range of aldehydes or ketones and different amines with trimethylsilyl cyanide (TMSCN) was efficiently catalyzed by the nano-ordered mesoporous borosilicate (B-MCM-41) at room temperature to afford the corresponding α-amino nitriles in high to quantitative yields in short reaction time. B-MCM-41 showed higher catalytic activity than Fe(III)-modified MCM-41 (Fe-MCM-41). Furthermore, the use of EtOH increased the rate of reaction remarkably compared to CH_2Cl_2 or CH_3CN. The B-MCM-41 catalyst can be recovered at least 4 times without significant loss of its catalytic activity.

1. Introduction

Multi-component Condensation Reactions (MCRs) have recently received great interest, due to the formation of multi-bonds in a one pot reaction, high atom efficiency, mild and simplified conditions and environmentally benign friendliness [1]. The Strecker reaction, which was reported initially in 1850, is the oldest known MCR and the most straightforward route for the synthesis of α-amino acids on both lab and technical scales. This reaction comprises a condensation of an aldehyde, ammonia and cyanide source, followed by subsequent hydrolysis of the resulting α-amino nitrile. Since the first synthesis of cyanohydrins by Winkler in 1832 and α-amino nitriles by Strecker in 1850, α-functionalized nitriles have attracted considerable attention of organic chemists, due to the rich chemistry of the cyano group (CN) [2]. In particular, α-amino nitriles are very important intermediates in the preparation of α-amino acids, 1,2-diamines and a variety of nitrogen-containing heterocycles, such as imidazoles or thiadiazoles [3]. The classical Strecker reaction employed KCN as a cyanide source in aqueous solution, which does have several drawbacks, such as the high toxicity of KCN and production of an alkaline reaction mixture [2]. The latter can severely affect the sensitive substrates and thus limit the scope of this valuable reaction. Due to these limitations, a variety of cyanide sources and methods have been investigated; some of these are Bu_3SnCN, Et_2AlCN, $(Et)_2P(O)CN$, $CH_3(OH)(CN)CH_3$, $K_2[Fe(CN)_6]$ [4] and $(CH_3)_3SiCN$ (TMSCN) [5]. Among these non-alkali metal cyanide variants, TMSCN has emerged as a very effective, relatively safe and easy-to-handle cyanide source for a nucleophilic addition to imines and carbonyl double bonds [2,5b–g].

Protocols based on TMSCN often require Bronsted or Lewis acid catalytic systems. Therefore, many homogeneous catalysts, such as $BiCl_3$, $NiCl_2$, $RuCl_3$, RhI_3, $CeCl_3$, InI_3, $ThCl_3.4H_2O$, $GdCl_3.6H_2O$, $La(NO_3)_3.6H_2O$, $Fe(Cp)_2PF_6$, $Cu(OTf)_2$, $Ga(OTf)_3$, $Pr(OTf)_3$ and I_2, have been developed in recent years [6]. To overcome problems associated with homogeneous catalysts, some simple heterogeneous catalysts, including $Zr(HSO_4)_4$ [7a], guanidine HCl [7b] and $LiClO_4$ [7c], have also been introduced. However, supported heterogeneous catalysts demonstrate several intrinsic advantages over their homogeneous counterparts: additional reactants activation by support, high surface area, ease of product separation and catalyst reuse, lower

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