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Tin exchanged zeolite as catalyst for direct synthesis of α -amino nitriles under solvent-free conditions

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ABSTRACT

Sn exchanged HBeta zeolite was prepared and characterized by PXRD, surface area, TPD and TEM analysis. The Sn exchanged zeolite was found to be highly efficient catalyst for the direct synthesis of α -amino nitrile from various ketones and aldehydes with amine and trimethyl silylcyanide (TMSCN) under solvent-free condition. Excellent yield of α -amino nitrile (up to 96%) was achieved within 10–120 min at room temperature. The Sn exchanged HBeta zeolite was recovered and reused several times without the loss of its catalytic performance.

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1. Introduction

There are intentionally different approaches among the chemists towards the carbon-carbon bond forming reactions. Among these a very elegant and versatile solution is provided through the addition of cyanide to an imine (Modified Strecker reaction) which also represents one of the intrigued methods for preparing α -amino acids [1]. α -Amino nitrile plays a significant role in organic chemistry because of its bifunctional nature, an amino and a nitrile group, thus it offers various synthetic approaches to different classes of natural and bioactive compounds [2]. α -Amino nitrile is also a valuable prebiotic precursor to porphyrins, corrins, nicotinic acid, nucleic acids, safromycin A (anti-tumor potency) and phthalascidin [3]. The versatile synthon α -amino nitriles are prepared by the reaction of aldehydes or ketones [4] with amines in the presence of various cyanide sources such as HCN [3,5], KCN [6,7], TMSCN [8] and Et₂AlCN [9]. Among these TMSCN has emerged as a promising, effective and relatively safer cyanide source [10] for the Strecker reaction. These catalytic systems performed well with aldehydes [11–49] but were not so effective in the case of ketone in most cases. Nevertheless, there are few catalysts reported for the Strecker reaction using less reactive electrophilic ketones [11,21,43-53]. Significant among these are Lewis acid/Brønsted acid catalysts like Fe(cp)₂ PF₆ [11], PEG [27], sulfamic acidfunctionalized magnetic Fe₃O₄ nanoparticles [43], sulfonate group anchored on mesoporous silica [44], tin ion-exchanged montmorillonite [45], nanocrystalline MgO [46], NHC-amidate Pd(II) complex [47], Li-trifluoroboronate [48], mesoporous aluminosilicate [49], TMSOTf [50], xanthan sulfuric acid [51], gallium(III)triflate [52], under ambient or high pressure [21,53] and solvent/solvent-free conditions. Although some outstanding catalytic systems have been reported, still there is a plenty of room for improvement, particularly in terms of catalyst recyclability, high catalyst activity at lower catalyst loading and conducting the reaction under solvent-free condition in lesser time. Onaka et al. [45] showed that individually Na-Mont, Sn(OH)₄, SnO₂, Sn-MCM-41 and Al-MCM-41 do not catalyze Strecker reaction. However, low to moderate product yields were obtained when SnO₂/silica (7.2%), SnO₂/MCM-41 (22%) and $SnO_2/SiO_2-Al_2O_3$ (37%) were used as catalyst. The best results were however obtained when Sn-Mont (84% in 3h) was used as catalyst. Exploring the utility of Sn exchanged zeolite as catalyst for this reaction was conspicuously absent despite the fact that zeolites have been extensively used as solid catalyst in various organic transformations [54] and is industrially valuable [55,56] because of its easy synthesis, porous structure with tunable ion exchange capacity. In view of the above and our ongoing interest in developing environment friendly and solvent free protocol for various organic transformations [11,57–59], here we are reporting Sn

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