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Highly efficient and rapid synthesis of imines in the presence of nano-ordered MCM-41-SO₃H heterogeneous catalyst

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KEYWORDS

Imines; Carbonyl compounds; MCM-41-SO₃H; Mesoporous materials; Green chemistry. **Abstract** Nano-ordered MCM-41 anchored sulfonic acid (MCM-41-SO₃H) was used as an efficient heterogeneous catalyst for the synthesis of Schiff bases by the reaction of different aryl/alkyl aldehydes or ketones with primary amines at room temperature with high to excellent yields. This mesoporous catalyst can be reused at least four times without significant loss to its catalytic potential.

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1. Introduction

The condensation of carbonyl compounds with primary amines to produce the corresponding imines was first discovered in 1864 by Hugo Schiff. Hence, imines are often referred to as Schiff bases or azomethines [1]. The imine formation is one of the most important reactions in organic and medicinal chemistry [2]. For instance, imines are used as versatile components in the asymmetric synthesis of α -aminonitriles [3], preparation of secondary amines by hydrogenation [4], and in cycloaddition reactions [5]. In addition, imines have been discovered to have a wide range of biological activities such as lipoxygenase inhibition, anti-inflammatory, anti-cancer [6], antibacterial, and antifungal behavior [7].

Many procedures have been introduced for the preparation of imines in the literature since the pioneering work of Hugo Schiff. Among recent methods or techniques for the preparation of imines, the use of ionic liquids [8], infrared [9], microwave [6], and ultrasound [10] irradiation can be mentioned. However, the main problem which affects the

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yield of the products originates in the existence of equilibrium conditions between the reactants and the corresponding imines, along with water, as a byproduct of the reaction. To overcome this problem, various dehydrating agents such as P_2O_5/SiO_2 [11], $MgSO_4-Mg(ClO_4)_2$ [12], fuming $TiCl_4$ [13] or aromatic solvents forming azeotropic mixtures with water at high temperatures have been used. Such methodologies often suffer from complicated procedures, moisture sensitive catalysts or reagents, large quantities of toxic aromatic solvents, huge amounts of costly dehydrating agents or catalysts, high reaction temperatures, and long reaction times. In addition, some of these methods are limited only to the synthesis of aldimines and are not suitable for preparation of ketimines. Therefore, the development of more efficient and rapid protocols to produce imines remains in high demand.

On the other hand, ordered mesoporous materials are used for fine chemical synthesis as heterogeneous solid catalysts. Nano-ordered mesoporous materials offer high surface areas with high concentrations of active sites [14]. In 1992, researchers at the Mobil Corporation made a major discovery of the M41S family of silicate/aluminosilicate mesoporous molecular sieves with exceptionally large uniform pore structures. It should be taken into account that a decisive choice of surfactant, adding auxiliary organic chemicals and changing reaction parameters (e.g. temperature or composition of the reactants and templates) can play significant roles in controlling mesoporous structure [15]. Furthermore, it is likely to improve mesoporous materials by the functionalization of their surfaces by