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Solvent-free synthesis of 4-substituted coumarins using sulfonated magnetic-nanoparticle catalyst: A useful complement to the Pechmann reaction

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ABSTRACT

Sulfonic acid supported carbon coated magnetic nanoparticles (Fe_3O_4@C@SO_3H), was prepared by using low cost precursors and a facile immobilization technique. The final catalyst, which was characterized by FT-IR, vibrating, and SEM techniques, was found to be an efficient and environmentally benign solid acid for the Pechmann condensation of substituted phenols with ethyl acetoacetate leading to the formation of coumarin derivatives. This reaction was catalyzed by Fe_3O_4@C@SO_3H under solvent-free condition at 120°C, to give the corresponding products in excellent yields. The catalyst is easily separated from the reaction condition and can be reused for several times with consistence in the activity. After the reaction, the catalyst could be effortlessly separated by external magnet. The catalytic system presented offers a useful strategy for the efficient synthesis of coumarin, simplicity in operation, and a green reaction profile by avoiding toxic conventional catalysts and solvents.

Keywords: Heterogeneous catalyst, Fe_3O_4@C@SO_3H, Coumarin, Pechmann condensation, Magnetic nanoparticles

1. INTRODUCTION

There are widespread interests in the preparation of heterogeneous acid catalyst for application in acid catalyzed organic transformations [1]. In recent years, magnetic nanoparticles (MNPs) have been used as an efficient support for synthesis of heterogeneous catalyst. This material is an abundantly available and highly stable substrate, mainly characterized by the fact that organic groups can be linked to its surface with strong bonding to generate catalytic sites [2, 3]. Development of magnetic reusable catalysts for organic synthesis based on solid acids has become an environmental chemical procedure for academic and industrial applications [4]. Several types of magnetic nanoparticle sulfonic acids have been prepared and applied in chemical transformations in recent years [5-7]. All of these magnetic nanoparticles based on sulfonic acid catalyst are suitable for some acid catalyzed organic transformations, but such preparation often suffers from some deficiency such as expensive starting reagents and requires more than two steps for the preparation [8].

In this study, we wished to evaluate the catalytic performance of this new catalyst in the ‘‘Pechmann reaction’’ as an acid catalyzed process. Although various methods have been reported for this reaction, they suffer from disadvantages such as long reaction time, using expensive and/or nonstable catalyst and using heterogeneous catalysts [9]. There is widespread interest in the synthesis of new coumarin derivatives, because of their applications in organic synthesis, biology, medicine, perfumes and cosmetics fields [10, 11]. Also some of the coumarin derivatives are known as antioxidant compounds, due to their radical scavenging properties. In material science, they have been used as fluorescence dyes in composite with other heterocyclic moieties. Furthermore, for DNA and RNA labeling, some fluorescent materials containing a coumarin moiety have been reported. Due to these