Ferrite-Nb_2O_5 Nanocatalyst: An Efficient Magnetically Recoverable Catalyst for Biginelli Reaction

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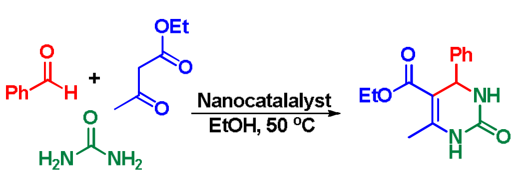
INTRODUCTION

Nanocatalysis has arisen in the last few years as a bridge between heterogeneous and homogenous catalysis.[1] Metals deposited on the surface of magnetite nanomaterials have been used with the aim to increase the contact between the reagents and catalyst, mimicking homogeneous catalysts while presenting the advantages of heterogeneous catalysis such as easy separation from the reaction mixture, and recyclability.[1] Lewis acid supported on magnetite, constitute a very important synthetic tool for the multi-component approaches, e.g. Biginelli reaction. This transformation still attracts researchers attention because 3,4-dihydropyrimidin-2(1*H*)-ones obtained from this multicomponent reaction showed a wide range of biological activities.[2] Attempting to disclose further developments on sustainable protocols, heterogeneous catalysis and nanomaterials,[3] herein, we report the multicomponent synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones using a new, efficient, recyclable and reusable ferrite-Nb_2O_5 nanocatalyst.

RESULTS AND DISCUSSION

Initially, we focused our attention on the influence of different nanocatalyst supported on magnetite. To investigate this, we chose benzaldehyde, ethyl acetoacetate and urea as model substrates (Scheme 1). We have found that the nature and the loading of nanocatalyst have a strong influence in the yields of Biginelli compound.



Scheme 1. Biginelli reaction.

Niobium pentoxide supported on magnetite nanocatalyst showed better results when compared with the others (Table 1), affording the Biginelli product in 99 % of yield when 1 mol% or only 0,1 mol% of the nanocatalyst were used, (entries 8 and 9).

Table 1. Biginelli reaction – evaluation of nanocatalyst.

Entry [a]	Nanocatalyst	Cat. loading (mol%) [b]	Yield (%) [c]
1	Fe_3O_4-NiO	10	99
2	Fe_3O_4-NiO	1.0	95
3	Fe_3O_4-NiO	0.1	70
4	Fe_3O_4-CuO	10	99
5	Fe_3O_4-CuO	1.0	99
6	Fe_3O_4-CuO	0.1	72
7	Fe_3O_4-Nb_2O_5	10	0
8	Fe_3O_4-Nb_2O_5	1.0	99
9	Fe_3O_4-Nb_2O_5	0.1	99
10	Fe_3O_4	40	50
11	Fe_3O_4	4.0	67
12	Fe_3O_4	0.4	35

[a] Reactions were performed in the presence of benzaldehyde (1.0 equiv.), ethyl acetoacetate (1.0 equiv.) and urea (1.5 equiv.) in EtOH at 50ºC in a closed vial for 12 h. [b] Calculated relating load of metal impregnated (about 10% m/m) over magnetite support. [c] Isolated yields.

CONCLUSION

The new ferrite-Nb_2O_5 nanocatalyst afforded the Biginelli product in better yields when compared to others ferrite modified metal catalysts. The methodology allowed mild reaction conditions and small amount of nanocatalyst. The scope and mechanism of the reaction is now under investigation in our laboratory.

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