

New magnetic periodic mesoporous organosilica functionalized 3-propyl amine (Fe₃O₄@PMO-ICS-PrNH₂) as highly efficient and recyclable catalysts in one-pot synthesis of imidazopyrimidine derivatives

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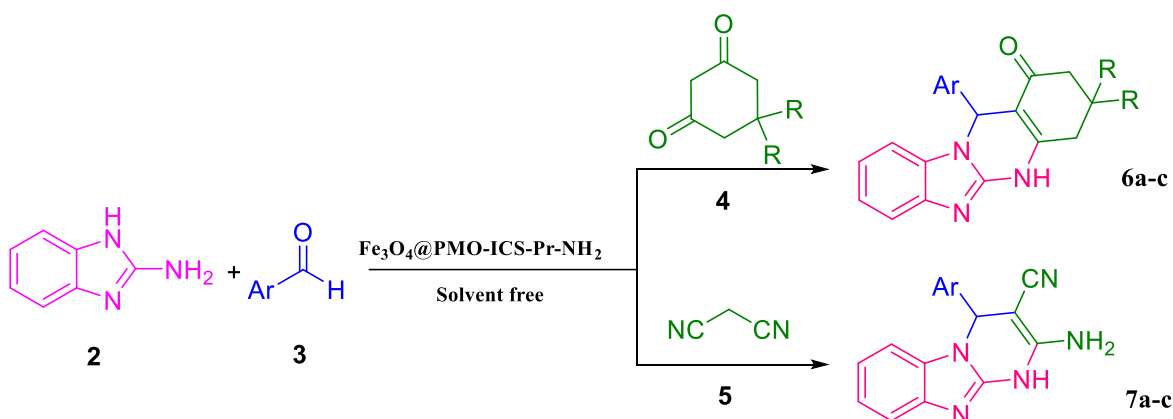
Abstract: The catalytic application of a novel magnetic periodic mesoporous organosilica functionalized 3-propylamine($\text{Fe}_3\text{O}_4@\text{PMO-ICS-PrNH}_2$) in the synthesis of imidazopyrimidine derivatives was investigated. At first, the $\text{Fe}_3\text{O}_4@\text{PMO-ICS-PrNH}_2$ nanocatalyst was prepared and characterized by N_2 adsorption-desorption isotherms techniques, field emission scanning electron microscopy (FESEM), thermogravimetric analysis (TGA), energy-dispersive X-ray (EDX) spectroscopy, Fourier transform infrared (FTIR) spectroscopy and vibrating sample magnetometer (VSM). Then, the catalyst was shown to be a efficient recyclable and highly active catalyst and used in the three-component synthesis of 2-aminobenzaldehyde with aromatic aldehydes various and dimedone/malononitrile under H_2O solvent conditions. Also, imidazopyrimidine products were obtained in mild reaction conditions, good to excellent yields and at short reaction times. Moreover, the catalyst was recovered and successfully reused at least five times without considerable reduction in its activity.

Keywords Multi-component reactions (MCRs); Magnetic periodic mesoporous organosilicas (PMOs); Imidazopyrimidine derivatives.

1. Introduction

In recent years, the use of porous compounds has been widely considered by researchers. In recent years, the use of porous compounds has been widely considered by researchers. Since the first discovery of ordered periodic mesoporous organosilica by Inagaki et al., this compounds mesopores have been extensively explored for applications that range from adsorbents, chromatography, sensor technology, separation, drug delivery, solid phase extraction and catalysis, to uses. These materials has a high specific surface area, high mechanical and hydrothermal stability, narrow pore size distribution and available pore volume. PMOs normally be achieved through co-condensation of bridged organosilane precursors $[(R'O)_3Si-R-Si(OR')_3]$; R: organic bridging group, R': methyl or ethyl to produce organofunctionalized ordered periodic mesoporous organosilica materials [1, 2].

Multi-component reaction (MCR) is one of the most making source for designing and production heterocyclic compounds having biological and pharmaceutical activities. Multi-component reaction in terms of atomic economics, operational simplicity and complexity of products are important. between these types of compounds the synthesis of imidazopyrimidine derivatives is attracted the attention of many organic chemists to their important pharmaceutical activities, therapeutic [3]. These imidazopyrimidine derivatives have broad spectrum of pharmacological properties such as antimalarial, anti-tubercular herbicidal activity, antibiotic, anticancer activities, anti-tumour, antidiabetics etc. Recently, considering the catalysis applications of porous compounds, we were encouraged to use PMO-ICS-PrNH₂ as a catalyst for the facile and efficient multicomponent synthesis of imidazopyrimidine derivatives. In recent years, the use of porous compounds has been widely considered by researchers. In recent years, the use of porous compounds has been widely considered by researchers. Since the first discovery of ordered periodic mesoporous organosilica by Inagaki et al., this compounds mesopores have been extensively explored for applications that range from adsorbents, chromatography, sensor technology, separation, drug delivery, solid phase extraction and catalysis, to uses. These materials has a high specific surface area, high mechanical and hydrothermal stability, narrow pore size distribution and available pore volume. PMOs normally be achieved through co-condensation of bridged organosilane precursors $[(R'O)_3Si-R-Si(OR')_3]$; R: organic bridging group, R': methyl or ethyl to produce organofunctionalized ordered periodic mesoporous organosilica materials [4, 5]



Scheme 1. Synthesis of imidazopyrimidine derivatives catalyzed by $(\text{Fe}_3\text{O}_4@PMO\text{-ICS-PrNH}_2$ (1)

2. Experimental section

2.1. General

Reagents and Apparatus

High-purity chemical reagents were purchased from Merck. All reactions and the purity of the products were monitored by thin-layer chromatography (TLC) using aluminum plates coated with silica gel F254 plates (Merck) using ethyl acetate and n-hexane as eluents. The spots were detected either under UV light or by placing in an iodine chamber. Melting points were determined in open capillaries using an Electrothermal 9100.

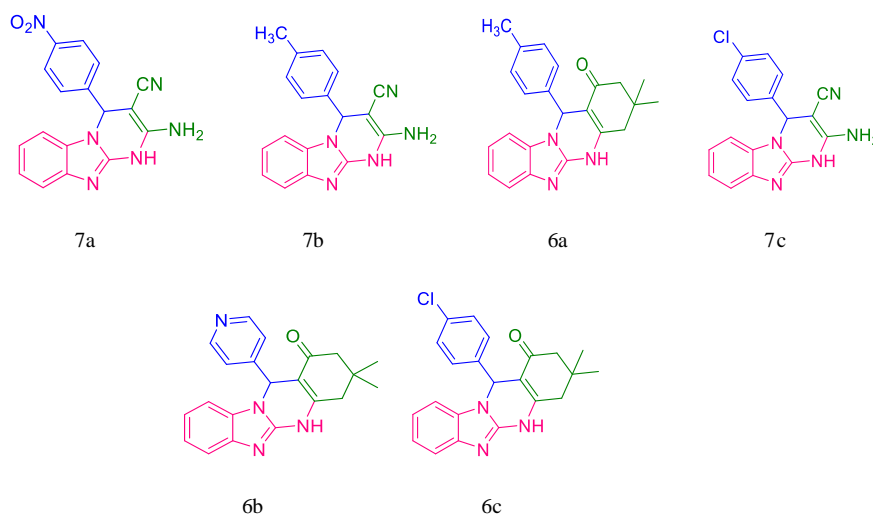
General procedure for the synthesis of imidazopyrimidine derivatives (6a-c/7a-c)

$\text{Fe}_3\text{O}_4@PMO\text{-ICS-Pr-NH}_2$ (1, 5 mg) was added to a mixture of 2-aminobenzimidazole (1 mmol, 0.133 mg), dimedone/malononitrile (1 mmol) and the corresponding aromatic aldehydes (1 mmol). The obtained reaction mixture was stirred under solvent-free conditions at 80 °C for the proper times indicated in Table 2. The progress of the reaction was monitored by TLC (EtOAc: n-hexane, 1:3). After completion of the reaction, DMF (2 mL) was added and the reaction mixture was heated to dissolve organic materials. The magnetic nanocatalyst **1** was then collected by an external magnet. After that, distilled water (5 mL) was added to the DMF solution and the obtained

precipitate was filtered off and washed using n-hexane (2 mL) to afford pure products. The obtained powders were then dried in an oven at 80 °C for 1 h.

3. Results and discussion

In conclusion, we have reported an environmentally benign and highly efficient one-pot methodology for the synthesis of a wide range of imidazopyrimidine derivatives in the presence of a low loading of Fe₃O₄@PMO-ICS-PrNH₂ as a new and readily recoverable nanoporous catalyst. In addition, the present protocol not only offers substantial improvements in the reaction rates and yields but also avoids the use of heavy metals, corrosive reagents, or toxic solvents.



Scheme 2. Scope of 3-indole derivatives (**6a-c/7a-c**)

4. Conclusions

A highly efficient and facile method for the synthesis of 3,3-dimethyl-12-phenyl-3,4,5,12-tetrahydrobenzo[4,5]imidazo[1,2-*b*]quinazolin-1(2*H*)-one and 2-amino-4-phenyl-1,4-dihydrobenzo[4,5]imidazo[1,2-*a*]pyrimidine-3-carbonitril derivatives using a one-pot, three-component protocol through condensation of aldehydes,

dimedone/malononitrile, and 2-aminobenzimidazole in the presence of $\text{Fe}_3\text{O}_4@\text{PMO-ICS-PrSO}_3\text{H}$ under solvent-free conditions were developed. This methodology offers outstanding advantages such as: (i) high efficiency, Shorter reaction time with high to excellent yields, (ii) Simple work-up procedure, low catalyst loading, Simple and fast separation of the catalyst, catalyst recyclability, and low cost.

Acknowledgments

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