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ORIENTAL JOURNAL OF CHEMISTRY
An International Open Free Access, Peer Reviewed Research Journal

www.orientjchem.org

ISSN: 0970-020 X
CODEN: OJCHEG
2012, Vol. 28, No. (1):
Pg. 141-143

A Green, Reusable and Highly Efficient Heterogeneous Catalyst for the Synthesis of Arylpyrazoles using Nano-Fe_2O_3

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(Received: January 01, 2012; Accepted: February 03, 2012)

ABSTRACT

Series of some new arylpyrazole derivatives have been synthesized in good yields via one-pot condensation reaction using nano-Fe_2O_3 as heterogeneous catalyst.

Keywords: Arylpyrazole, Nano-Fe_2O_3, solvent-free, Baylis-Hillman.

INTRODUCTION

Recently, organic reactions catalyzed by nanoparticles (NPs) have attracted much attention. Consequently, there is a great demand for development of novel catalysts with higher catalytic activities, lower prices, good recyclability and less pollute to the environment in their catalytic systems. Many organic reactions occur more efficiently in the solid-state than in solution and in many cases even more selectively, because molecules in the crystals are arranged tightly and regularly. Nanoparticles have emerged as sustainable alternatives to conventional materials, as robust high–surface-area heterogeneous catalyst supports[1]. A magnetic nanoparticle catalyst was readily prepared from inexpensive starting materials which catalyzed the Hantzsch reaction. High catalytic activity and ease of recovery from the reaction mixture using an external magnet, and several reuse times without significant losses in performance are additional eco-friendly attributes of this catalytic system[2].

Arylpyrazole is an important class of organic compounds, which received a considerable attention due to their wide range of biological activities and widely used as pharmaceuticals, agrochemicals, anti-inflammatory, antiviral, antibacterial[3-8]. Although heterocycle compounds are valuable compounds and many applications have been reported[9].

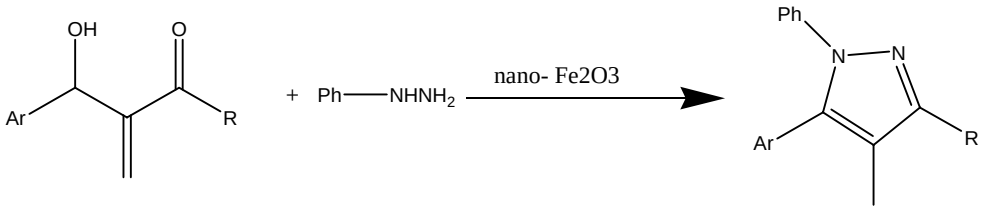
Previously, we have synthesized a number of heterocyclic compounds[10-13]. In our ongoing research prompted by our interest in multiple component reactions and as part of programs in the area of heterocyclic compounds containing nitrogen[14], and due to the resultant pharmacological interest in compounds which

belong to the diarylpyrazoles, although this reaction done previously in other conditions[14-16], herein we report in a different condition one pot reaction with high yields and easy separation of product for the construction of some 1, 5-diarylpyrazole derivatives, via condensation of Baylis-Hillman adduct and phenyl hydrazine Using nano- Fe_2O_3 (Scheme 1).

Baylis-Hillman adducts were prepared by the reaction of ethyl vinyl ketone, arylaldehydes[17]. For synthesis of 1, 5-diarylpyrazole derivatives, the reaction of Baylis-Hillman adduct (1), phenylhydrazine hydrochloride in 1,2-dicloroethane Using nano-Fe2O3 was used (Scheme 1). Therefore preparation of all the 1, 5-diarylpyrazoles described in this paper, the reaction was complete within 30-40 min on solid support nano-Fe_2O_3 in excellent yields (78-88%) to afford **4a-e** (Table 1).

RESULTS AND DISCUSSION

Herein, we report synthesis of 1, 5-diarylpyrazole derivatives, the reaction of Baylis-



1 **4a-d**

Scheme 1

Table 1: Three-component synthesis of some 1, 5-diarylpyrazoles.

Entry	R	Ar	Yields (%)	Time (Min)	IR $\nu_{C=N}$ cm^{-1}	^{1}H-NMR ($CDCl_3$)δ, ppm
4a	-Me	phenyl	78	30	1610	1.36 (t, J = 7.6 Hz, 3H), 1.94 (s, 3H), 2.75 (q, J = 7.6 Hz, 2H),7.21-7.26 (m, 5H), 7.34 (dd, J = 7.6, 1.4 Hz, 1H), 7.55 (dt, J = 8.1, 1.4 Hz, 1H), 7.62 (dt, J = 7.5, 1.30 Hz, 1H), 7.98 (dd, J = 8.1, 1.2 Hz, 1H)
4b	-Et	o-nitrophenyl	82	40	1615	1.38 (t, *J* = 7.5 Hz, 3H), 1.95 (s, 3H), 2.7 (q, *J* = 7.5 Hz, 2H), 7.20-7.26 (m, 5H), 7.33 (dd, *J* = 7.5, 1.5 Hz, 1H), 7.58 (dt, *J* = 8.5, 1.5 Hz, 1H), 7.65 (dt, *J* = 7.5, 1.30 Hz, 1H), 8.0 (dd, *J* = 8.5, 1.3 Hz, 1H)
4c	-Et	p-nitrophenyl	83	32	1600	1.34 (t, *J* = 7.2 Hz, 3H), 2.12 (s, 3H), 2.79 (q, *J* = 7.2 Hz, 2H), 7.20 (dd, *J* = 8.5, 1.3 Hz, 2H), 7.30-7.38 (m, 3H), 7.40 (d, *J* = 8.5 Hz, 2H), 8.30 (d, *J* = 8.5 Hz, 2H)
4d	-Et	m-Chlorophenyl	85	35	1605	1.41 (t, J = 7.5 Hz, 3H), 2.4 (s, 3H), 2.9 (q, J = 7.5 Hz, 2H), 7.3 (dt, J = 7.5, 1.2 Hz, 1H), 7.25-7.35 (m, 8H)
4e	-Et	p-Chlorophenyl	88	36	1609	1.38 (t, J = 7.5 Hz, 3H), 2.13 (s, 3H), 2.81 (q, J = 7.5 Hz, 2H), 7.14 (d, J = 7.0 Hz, 2H), 7.20-7.26 (m, 3H), 7.31 (m, 2H)

Hillman adduct (1), phenylhydrazine hydrochloride in 1,2-dicloroethane Using nano-Fe2O3 was used (Scheme 1), which could provide an efficient and simple route for the synthesis of diarylpyrazoles and also the required compound obtained this product in a very high yield.

ACKNOWLEDGEMENTS

We gratefully acknowledge the financial support from the Research Council of Tonekabon Branch Islamic Azad University.

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