Heterocyclic Letters
Vol. 8| No.4|761-768|Aug-Oct|2018
ISSN : (print) 2231–3087 / (online) 2230-9632
CODEN: HLEEAI
http://heteroletters.org

CATALYTIC PERFORMANCE OF A PHOSPHOTUNGSTIC ACID FUNCTIONALIZED PYRAZOLIUM-BASED IONIC LIQUID IMMOBILIZED ON $CuFe_2O_4@SiO_2$ AS A MAGNETICALLY RETRIEVABLE NANOCATALYST FOR THE SYNTHESIS OF 7-AMINO-2*H*-PYRANO[2,3-*d*]PYRIMIDINE-6-CARBONITRILES

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Abstract: Under mild conditions and without any additional organic solvent, a series of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles was efficiently synthesized by one-pot three-component cyclocondensation of barbituric acid, aryl aldehydes, and malononitrile using a functionalized pyrazolium-based ionic liquid containing a phosphotungstic counter-anion $H_2PW_{12}O_{40}$ (H_2PW) immobilized on $CuFe_2O_4@SiO_2$ magnetic nanoparticles which was denoted as $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW. A wide range of aromatic aldehydes easily undergo condensation with barbituric acid and malononitrile to afford the desired products of good purity in excellent yields under solvent-free conditions. Other advantages of this new synthetic approach are short reaction times and a simple procedure with an easy work-up. Moreover, the nanomagnetic solid acid was easily recovered from the reaction mixture by simple magnetic decantation and used four runs without significant loss of activity.

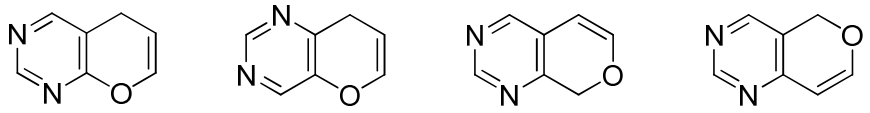
Keywords: Pyrano[2,3-*d*]pyrimidines, Pyrazolium-based ionic liquid, Magnetic nanoparticles, Solvent-free conditions

Introduction

Pyrans and pyrimidines have occupied unique positions in the design and synthesis of novel biologically-active agents due to their broad spectrum of activities. Literature reports had already established that certain pyrans exhibit significant biological properties such as antihypertensive[i], antiproliferative[ii], anti-osteosarcoma[iii], antibacterial[iv,v], antitumor[vi], and anti-inflammatory[vii] activities. A number of these compounds are useful as photoactive substances[viii]. Furthermore, polyfuctionalized pyran derivatives are common structural subunits in variety of important natural products[ix] and are employed as apoptosis inducing agents[x]. On the other hand, structures containing pyrimidine scaffold play an essential role in several biological processes of chemical and pharmacological importance. In particular, the

pyrimidine nucleus can be found in a broad variety of antibacterial[xi], antitumor[xii], antimalarial[xiii], anti-inflammatory[xiv], antiproliferative[xv], antitubercular[xvi], and antioxidant[xvii], agents. The pyrimidine skeleton is present in DNA bases[xviii] including cytosine, thymine, and uracil, and also in vitamin B1[xix]. Furthermore, pyrimidines have also been known as inhibitors of CDK4[xx], tyrosine kinase[xxi], EGFR-TK[xxii], and aurora kinase[xxiii].

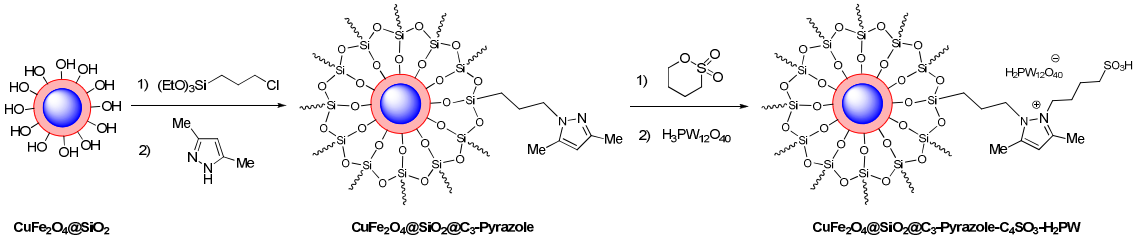
Among various pyranopyrimidine scaffolds, including pyrano[2,3-*d*]pyrimidine **I**, pyrano[3,2-*d*]pyrimidine **II**, pyrano[3,4-*d*]pyrimidine **III**, pyrano[4,3-*d*]pyrimidine **IV** (Figure 1), which are constructed from two fused pyran and pyrimidine rings, pyrano[2,3-*d*]pyrimidine ring system **I** has been a subject of increasing interest because of reported biological and pharmacological properties[xxiv-xxvii]. These compounds are generally synthesized *via* a one-pot three-component cyclocondensation of barbituric acid, aryl aldehydes, and malononitrile in the presence of several promoting agents[xxviii-xxxv]. Synthesis of these compounds using microwave irradiation has been also reported[xxxvi]. Many of these methodologies are not entirely satisfactory and suffer from limitations such as long reaction times, unsatisfactory yields, the use of relatively expensive catalysts, and using microwave irradiation for accelerated synthesis. Thus, the discovery of new methodologies using new efficient reusable catalysts for the synthesis of pyrano[2,3-*d*]pyrimidines is of certain demand.



I **II** **III** **IV**

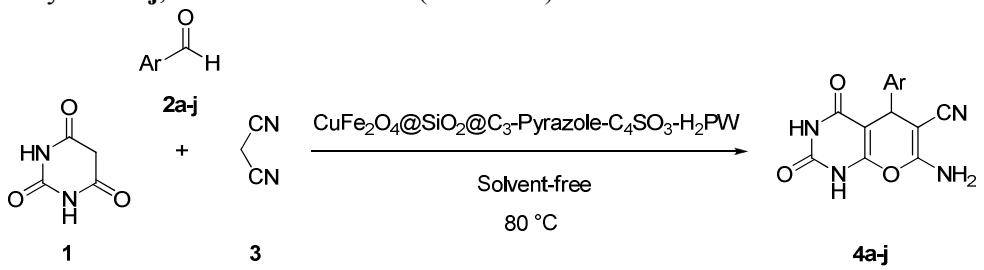
Figure 1. Structures of pyranopyrimidines

The immobilization of liquid catalysts on a solid support such as magnetic nanoparticles (MNPs) is one of the important routes for developing novel heterogeneous catalysts with easier recovery and reusability over homogeneous systems. These materials with high thermal and mechanical stability can be easily separated and recycled from the reaction medium using an external permanent magnet and enhance performance of the process, preventing the loss of catalyst[xxxvii,xxxviii]. Functionalized MNPs have been widely applied in biochemical and biomedical fields including in enzyme immobilization, protein purification, hyperthermia and bacterial detection, and also as heterogeneous catalysts in organic transformations[xxxix,xl]. Very recently, we have synthesized and characterized novel $CuFe_2O_4$@SiO_2@C_3-Pyrazole-C_4SO_3-H_2PW MNPs by immobilizing of a new functionalized pyrazolium-based ionic liquid containing a phosphotungstic counter-anion $H_2PW_{12}O_{40}^-$ (H_2PW) on $CuFe_2O_4$@SiO_2 MNPs (Scheme 1) which successfully applied as highly efficient catalyst in the synthesis of polyhydroquinolines[xli].



Scheme 1. Preparation of $CuFe_2O_4$@SiO_2@C_3-Pyrazole-C_4SO_3-H_2PW MNPs

Taking all these facts into consideration, and in conjunction with our earlier studies of synthesis of heterocyclic compounds[xlii-xlvii], and our interest in catalysis[xlviii-xlxv], we report here another application of $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW as catalyst in the synthesis of a series of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles **4a-j** by one-pot three-component reaction of barbituric acid **1**, aryl aldehydes **2a-j**, and malononitrile **3** (Scheme 2).



Scheme 2. Synthesis of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles catalyzed by $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW MNPs

Experimental

All chemicals were purchased from Merck and Aldrich and used without purification. Ultrasonication was performed by Soltec sonicator (Italy, 2200ETH S3) at a frequency of 40 kHz and a nominal power of 260 W. Melting points were measured on a Stuart SMP3 melting point apparatus. The ^{1}H NMR spectra were measured on a Bruker 300 spectrometer using tetramethyl silane (TMS) as internal standard. IR spectra were recorded on a Tensor 27 Bruker spectrophotometer in KBr disks.

Preparation of $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW MNPs.

To dispersed ultrasonically $CuFe_2O_4@SiO_2$ (2.0 g) in dry toluene (5 ml), (3-chloropropyl)triethoxysilane (2.0 ml) was added and the mixture was stirred at room temperature for 15 min and then refluxed for 24 h. After cooling to room temperature, the obtained solid was isolated by a magnet and repeatedly washed with toluene and dried under vacuum at 80 °C for 7 h. The resulting MNPs were ultrasonically dispersed in dry toluene (5 ml) for 15 min at 60 °C and then 3,5-dimethyl-1*H*-pyrazole (8 mmol) was added and the mixture was heated under reflux for 24 h. After cooling to room temperature, the new MNPs were collected and repeatedly washed with toluene and dried under vacuum at 80 °C for 5 h to form $CuFe_2O_4@SiO_2@C_3$-Pyrazole MNPs. The later MNPs were sonicated in dry toluene (7 ml) for 20 min at 60 °C and then 1,4-butane sultone (12 mmol) was added dropwise during 20 min and the mixture was refluxed for 6 h. After cooling to room temperature, the solid was collected using a permanent magnet and repeatedly washed with dry toluene and dried under vacuum at 70 °C for 3 h to form $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3 MNPs. After ultrasonically dispersion of these MNPs (2.0 g) in dry THF (8 ml) for 20 min at 60 °C, $H_3PW_{12}O_{40}$ (3 mmol) was added and sonication continued for another 1 h. The resulting MNPs were isolated by magnetic decantation and washed with dry THF and dried under vacuum at 60 °C for 24 h to form final $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW MNPs[xli].

General procedure for the synthesis of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles catalyzed by $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW

A mixture of barbituric acid **1** (1 mmol), an aryl aldehyde **2a-j** (1 mmol), malononitrile **3** (1 mmol), and $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW (0.04 g) was heated in the oil bath at 80 °C for 3-10 min. During the procedure, the reaction was monitored by TLC. Upon completion of the transformation, the reaction mixture was cooled to room temperature and

hot ethanol was added. The catalyst was readily recycled by simple magnetic decantation, washed with dry THF, dried under vacuum at 60 °C for 2 h, and then used in the next run. The solvent was evaporated in *vacuo* and the residue crude product was recrystallized from ethanol to give compounds **4a-j** in high yields.

7-Amino-5-(3-chlorophenyl)-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (4c): ^{1}H NMR (300 MHz, DMSO-d_6, δ, ppm): 4.32 (s, 1H, pyran CH), 7.18-7.38 (m, 6H, arom-H and NH_2), 11.12 (s, 1H, NH), 12.14 (s br., 1H, NH); FT-IR (KBr disk, *υ*, cm^{-1}): 3416, 3323, 3199, 3099, 2193, 1710, 1663, 1529, 1402, 1278, 1182, 799.

7-Amino-5-(4-chlorophenyl)-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (4d): ^{1}H NMR (300 MHz, DMSO-d_6, δ, ppm): 4.28 (s, 1H, pyran CH), 7.16 (s br., 2H, NH_2), 7.27 (d, 2H, *J* = 8.3 Hz, arom-H), 7.36 (d, 2H, *J* = 8.3 Hz, arom-H), 11.09 (s, 1H, NH), 12.10 (s br., 1H, NH); FT-IR (KBr disk, *υ*, cm^{-1}): 3393, 3194, 3075, 2198, 1684, 1639, 1521, 1403, 1346, 1277, 1188, 775.

7-Amino-5-(4-methylphenyl)-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (4f): ^{1}H NMR (300 MHz, DMSO-d_6, δ, ppm): 2.23 (s, 3H, CH_3), 4.27 (s, 1H, pyran CH), 7.00-7.20 (m, 6H, arom-H and NH_2), 11.13 (s, 1H, NH), 12.16 (s br., 1H, NH); FT-IR (KBr disk, *υ*, cm^{-1}): 3366, 3189, 3072, 2848, 2198, 1690, 1521, 1402, 1347, 1277, 1188, 1097, 844, 777.

7-Amino-5-(4-nitrophenyl)-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitrile (4j): ^{1}H NMR (300 MHz, DMSO-d_6, δ, ppm): 4.45 (s, 1H, pyran CH), 7.29 (s br., 2H, NH_2), 7.55 (d, 2H, *J* = 8.8 Hz, arom-H), 8.18 (d, 2H, *J* = 8.8 Hz, arom-H), 11.14 (s, 1H, NH), 12.20 (s br., 1H, NH); FT-IR (KBr disk, *υ*, cm^{-1}): 3392, 3191, 2197, 1717, 1639, 1523, 1407, 1348, 1280, 1188, 1102, 822.

Results and discussion

Our efforts to develop an efficient and environmentally benign methodology for the synthesis of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles focused initially on the synthesis of compound **4d** as a model reaction. Thus, a catalytic amount of $CuFe_2O_4$@SiO_2@C_3-Pyrazole-C_4SO_3-H_2PW was added to a mixture of barbituric acid **1** (1 mmol), 4-chlorobenzaldehyde **2d** (1 mmol), and malononitrile **3** (1 mmol) in different solvents and under solvent-free conditions. Pleasingly, we discovered that the reaction was efficiently catalyzed by $CuFe_2O_4$@SiO_2@C_3-Pyrazole-C_4SO_3-H_2PW under solvent-free conditions, providing a high yield of product **4d**. The reaction conditions were then optimized by conducting the reaction at different temperatures and employing different loadings of the catalyst. The results are summarized in Table 1. Trace amounts of the product **4d** were formed in the absence of the catalyst in refluxing H_2O or EtOH and also under solvent-free conditions at high temperature (Entries 1-3) indicating that the catalyst is necessary for the reaction. As can be seen, among the tested solvents such as H_2O, MeOH, EtOH, $CHCl_3$, MeCN, and also solvent-free conditions and various amounts of the catalyst, the reaction was more facile and proceeded to give the highest yield, using 0.04 g of $CuFe_2O_4$@SiO_2@C_3-Pyrazole-C_4SO_3-H_2PW under solvent-free conditions at 80 °C (Entry 13). The higher amounts of the catalyst or temperature had no significant effect on the yield and reaction time. All subsequent reactions were carried out under these optimized conditions.

Table 1. Screening of reaction parameters for the formation of compound **4d** catalyzed by $CuFe_2O_4$@SiO_2@C_3-Pyrazole-C_4SO_3-H_2PW MNPs[a]

Entry	Catalyst (g)	Solvent	T (°C)	Time (min)	Isolated Yield (%)

1	-----	H_2O	Reflux	120	Trace
2	-----	EtOH	Reflux	120	Trace
3	-----	-----	120	120	Trace
4	0.01	-----	70	10	71
5	0.01	-----	80	10	82
6	0.02	-----	60	7	84
7	0.02	-----	70	7	85
8	0.02	-----	80	6	89
9	0.02	-----	90	5	88
10	0.03	-----	80	5	90
11	0.03	-----	90	5	92
12	0.04	-----	70	5	88
13	0.04	-----	80	4	97
14	0.04	-----	90	5	97
15	0.05	-----	80	5	96
16	0.04	H_2O	Reflux	8	82
17	0.04	MeOH	Reflux	9	75
18	0.04	EtOH	Reflux	6	76
19	0.04	$CHCl_3$	Reflux	10	89
20	0.04	MeCN	Reflux	5	63

[a]Reaction conditions: barbituric acid **1** (1 mmol), 4-chlorobenzaldehyde **2d** (1 mmol), and malononitrile **3** (1 mmol).

After optimizing the conditions, we next examined the generality of these conditions to other substrates using a wide range of aromatic aldehydes bearing either electron-donating or electron-withdrawing substituents. The results are summarized in Table 2. It could be seen that $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW catalyzed the condensation of **1**, **2a-j**, and **3** at 80 °C. As indicated in Table 2, in all cases the reaction gives the products in high yields under solvent-free conditions and prevents problems which many associate with solvent use such as cost, handling, safety and pollution.

Table 2. Synthesis of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles **4a-j** catalyzed by $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW MNPs[a]

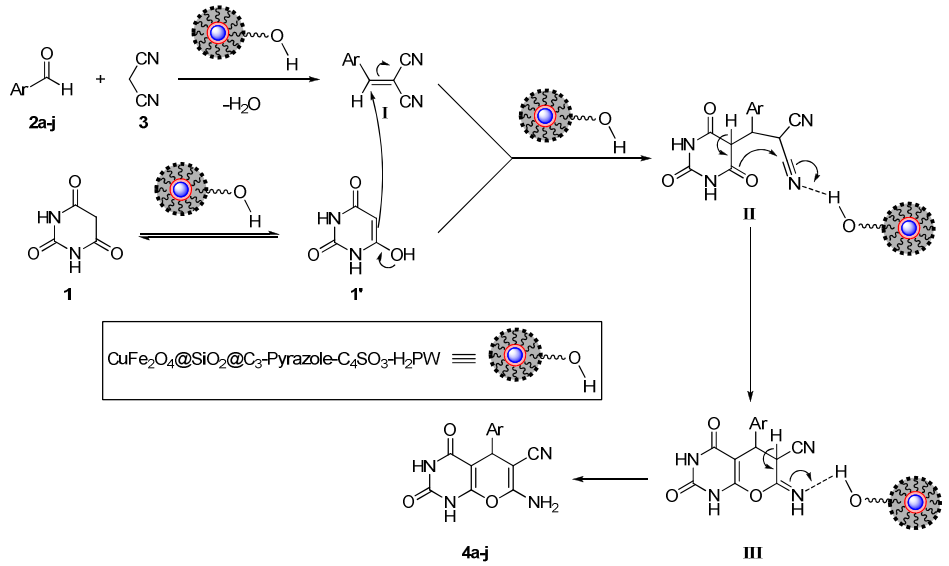
Entry	Ar	Product	Time (min)	Isolated Yield (%)	m.p. (ºC)	
					Found	Reported
1	C_6H_5	**4a**	3	94	217-220	215-217[xxviii]
2	2-ClC_6H_4	**4b**	4	90	214-217	211-212[xxviii]
3	3-ClC_6H_4	**4c**	5	92	238-240	240-241[xxviii]
4	4-ClC_6H_4	**4d**	4	97	234-236	232-234[xxix]
5	3-BrC_6H_4	**4e**	10	75	249-251	254-256[xxix]
6	4-MeC_6H_4	**4f**	7	91	218-220	222-224[xxix]
7	4-$MeOC_6H_4$	**4g**	5	92	280-283	279-281[xxix]
8	4-HOC_6H_4	**4h**	3	82	>350	>300[xxx]
9	3-$O_2NC_6H_4$	**4i**	5	86	270-272	272-274[xxxi]
10	4-$O_2NC_6H_4$	**4j**	7	87	238-240	237-240[xxxi]

[a]Reaction conditions: barbituric acid **1** (1 mmol), aromatic aldehyde **2a-j** (1 mmol), malononitrile **3** (1 mmol), $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW (0.04 g), 80 ºC, solvent-free.

The principle advantage of the use of MNPs as catalyst in organic transformations is their reusability. Hence, we decide to study the catalytic activity of recycled $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW in the synthesis of compound **4d**. $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW being insoluble in organic solvents, upon completion of the reaction, the reaction mixture was cooled to room temperature, and hot ethanol was added. The catalyst

was recovered by simple magnetic decantation, washed with dry THF, dried at 60 °C in vacuo for 2 h and reused in a similar reaction. The catalyst could be used at least four times with only slight reduction in the catalytic activity (97% for 1st use; 96% for 2nd use; 94% for 3rd use, and 93% for 4th use).

At the end, we believe that $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW can act as Brønsted acid and therefore promotes the reactions by increasing the electrophilic character of carbonyl, cyano, and imino groups in starting materials and also in the intermediates. A plausible mechanism may proceed as depicted in Scheme 3. The reaction occurs *via* initial formation of the dicyano olefin [**I**], prepared by Knoevenagel condensation of aryl aldehydes **2a-j** with malononitrile **3**, which reacts with **1'**, the enolic form of barbituric acid, to give the intermediate [**II**] which subsequently cyclizes to afford the desired compounds **4a-j** via the intermediate [**III**]. Attempts to isolate the proposed intermediates failed even after careful monitoring of the reactions.



Scheme 3. Plausible mechanism for the formation of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles catalyzed by $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW MNPs

Conclusion

In summary, newly synthesized $CuFe_2O_4@SiO_2@C_3$-Pyrazole-C_4SO_3-H_2PW was used as catalyst in the synthesis of 7-amino-5-aryl-2,4-dioxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles by one-pot three-component cyclocondensation of barbituric acid, aryl aldehydes, and malononitrile, giving high yields of the products within short reaction times. Furthermore, the catalyst is readily recovered by simple magnetic decantation, and can be reused for subsequent reactions with no significant loss of its activity.

Acknowledgement

This work was supported by Islamic Azad University, Mashhad Branch, Iran.

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Received on September 20, 2018,