

Original Research Article

Synthesis of 5-arylmethylene-pyrimidine-2,4,6-trione and 2-arylidene malononitriles derivatives using a new Brønsted acid nano magnetic catalyst

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Novel magnetic nano-particles

Barbituric acid

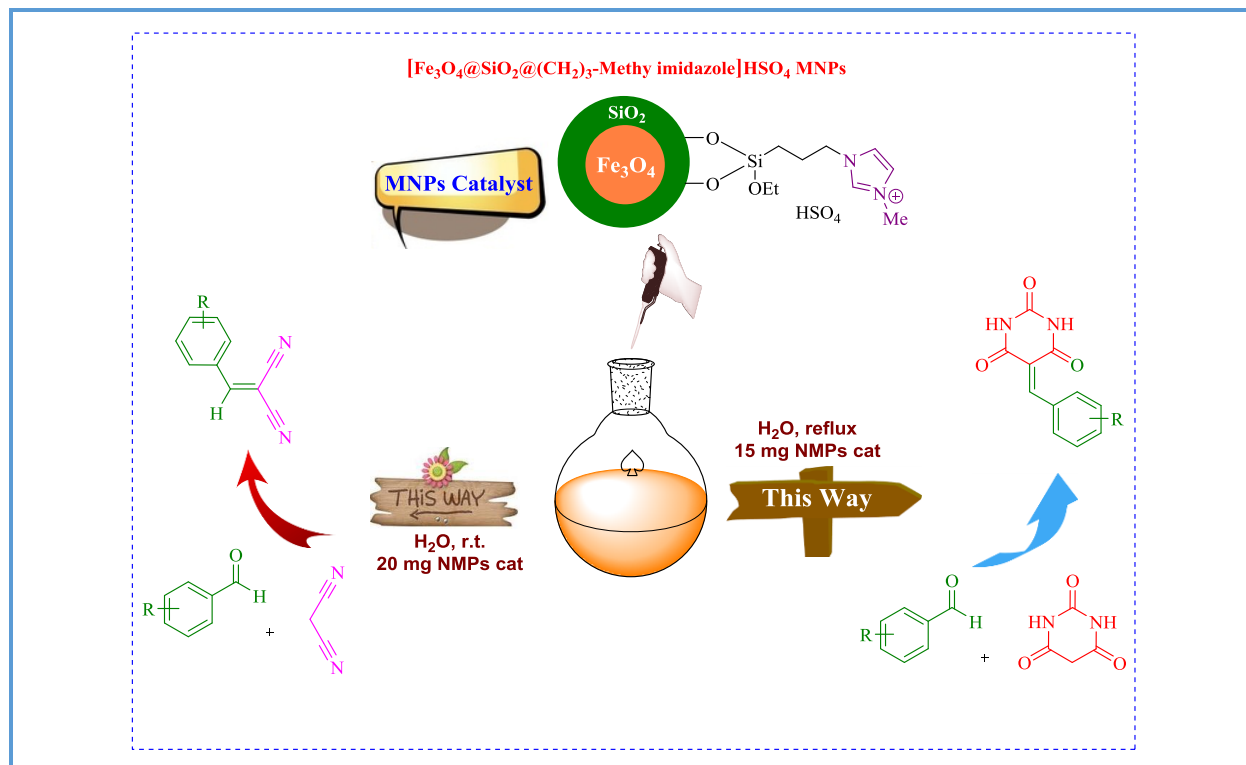
Malononitrile

ABSTRACT

In this work, 1-methyl imidazole-based ionic liquid stabilized on silica coated Fe_3O_4 magnetic nanoparticles [nano- $\text{Fe}_3\text{O}_4@ \text{SiO}_2@ (\text{CH}_2)_3$ -1-methyl imidazole] H_2SO_4 as a new, efficient, and magnetic Brønsted acid nano-magnetic catalyst was synthesized and characterized using various techniques including, Fourier transform infrared spectrometer (FT-IR), scanning electron microscopy (SEM), thermo gravimetric analysis (TGA), X-ray diffraction patterns (XRD), and vibrating sample magnetometry (VSM). Magnetic nanoparticles (MNPs) were used to synthesize the 5-arylmethylene-pyrimidine-2, 4, 6-trione derivatives by the one-pot condensation reaction between barbituric acid and various aldehydes in water at reflux conditions. Similarly, 2-arylidene malononitriles were synthesized by the one-pot of condensation reaction between various aldehydes and malononitrile in water as a green solvent at room temperature. This method has several advantages, such as short reaction times and high yields, non-using of toxic solvent, being eco-friendly benign, recyclability and reusability of the catalyst with external magnet for several time.

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Graphical Abstract



Introduction

Heterogeneous catalyst has lower catalytic activities compared with that of the homogeneous catalysts. This is due to the fact that homogeneous catalysts have better solubility in the reaction media. However, these catalysts suffer from one of the following disadvantages: i) recovery and reusability of the homogeneous catalyst is one of the disadvantages of these catalysts, because of the fact that they are tedious and time consuming. ii) When these catalysts are used, the product pollution was observed [1]. Other advantages of the heterogeneous catalyst can be their recovery, reusability and simple and convenient separation after completion of the reaction [2, 3].

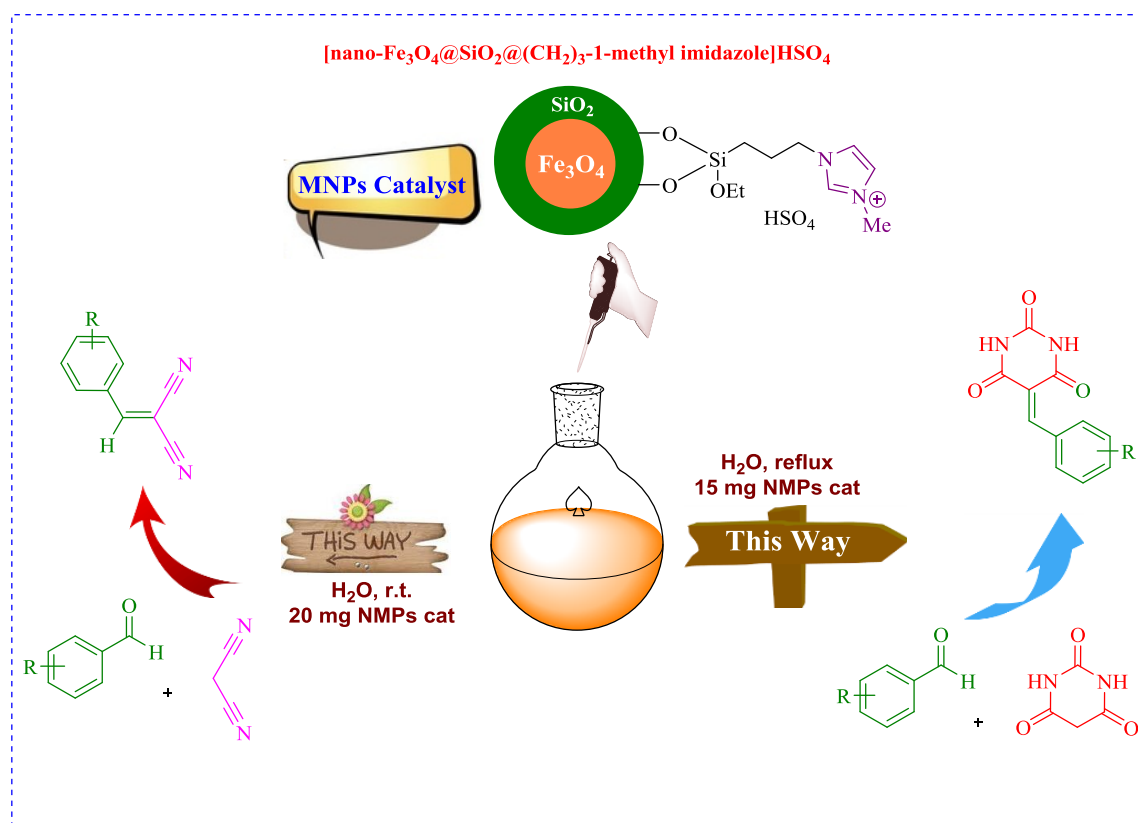
MNPs play a very important and vital role in a wide range of organic synthesis. MNPs are efficient, easily prepared and functionalized.

Moreover, they have low toxicity, large surface, high stability and available. These characteristics have made MNPs a suitable and efficient alternatives to other catalyst supports [4]. In addition, MNPs, rather than other catalysts (heterogeneous and homogeneous catalyst), can be separated from the reaction mixture by an external magnet without using filtration or centrifugation after completion of the reaction. In separation by an external magnet which leads to decrease environmental contamination as well as, MNPs have been widely reported in different fields such as drug delivery [5, 6], waste-water treatment [7], sonochemical approach [8], electrochemical storage of hydrogen [9], magnetic resonance imaging [10], biomolecular sensors [11], bioseparations [12], data storage [13], and environmental remediation [14].

Various catalysts have been used for the synthesis of 5-arylmethylene-pyrimidine-2, 4,

6-triones and 2-arylidene malononitriles; such as nickel nanoparticles [15], succinimidinium *N*-sulfonic acid hydrogen sulfate [16], $KAl(SO_4)_2 \cdot 12H_2O$ [17], copper oxide nanoparticles [18], tetrabutylammonium bromide [19], polyethylene glycol-bis(*N*-methylimidazolium) dihydroxide [20], zinc chloride [21], calcite and fluorite [22], solvent-free microwave enhanced [23], silica gel [24], diammonium hydrogen phosphate [25], and MgO/ZrO_2 [26]. Despite the large number of available and effective methods, some of these methods suffer from significant disadvantages including, long reaction times,

tedious work-up, low yield of products, harsh reaction conditions, expensive reagent, and toxic solvent, and the requirement of excess amounts of catalysts. Therefore, there is a requirement to develop novel catalysts which can overcome these problems. Herein we describe a simple and convenient method for the synthesis of 5-arylmethylene-pyrimidine-2, 4, 6-triones and 2-arylidene malononitriles using $[nano-Fe_3O_4@SiO_2@((CH_2)_3-1\text{-methyl imidazole})]HSO_4$, as a new, efficient, and magnetic Brønsted acid nanocatalyst (Scheme 1).



Scheme 1. Synthesis of 5-arylmethylene-pyrimidine-2, 4, 6-trione and 2-arylidene malononitriles using novel $[nano-Fe_3O_4@SiO_2@((CH_2)_3-1\text{-methyl imidazole})]HSO_4$

Experimental

Materials and methods

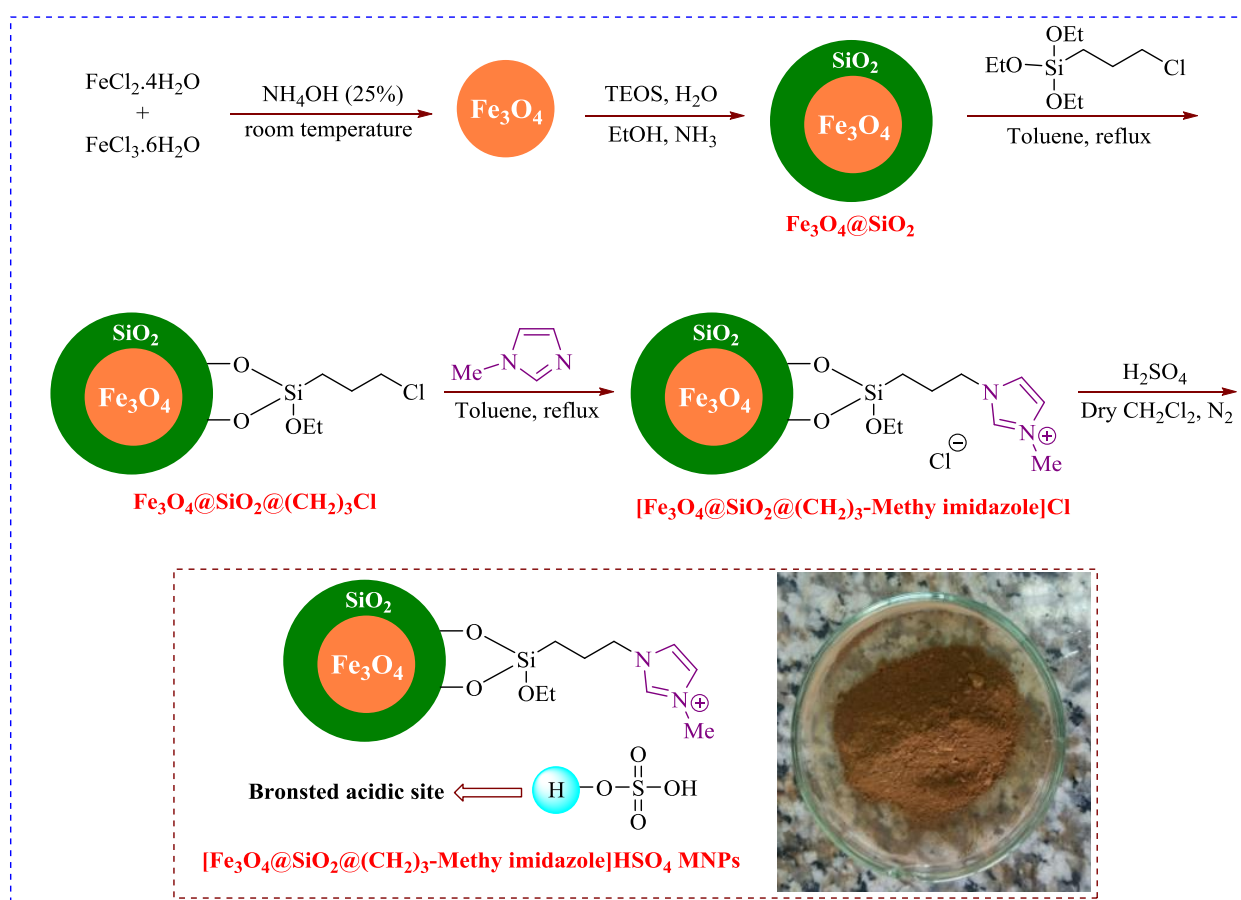
All the chemicals were purchased from the Merck or Fluka Chemical Companies. All the known compounds were identified by comparing their melting points and spectral

data with those reported in the literature. Progress of the reactions was monitored by thin layer chromatography (TLC) using silica gel SIL G/UV 254 plates. Infrared (IR) spectra were recorded on a Frontier FT-IR (Perkin Elmer) spectrometer using a potassium bromide (KBr) disk. The phases present in the magnetic materials were assessed using XRD (model X0 Pert with X'Pert; Philips, The Netherlands) with Cu K α 1 radiation ($k = 1.5401 \text{ \AA}$), operating the X-ray generator at 40 kV and 30 mA. Diffraction

patterns were collected in the 2 h range of 20–80°.

General procedure for the preparation of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄

1-methyl imidazole-based ionic liquid-stabilized on silica-coated Fe₃O₄ magnetic nano particles [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as a brown solid was synthesized according early report (Scheme 2) [27].



Scheme 2. The preparation of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as MNPs

General procedure for preparation of 5-arylmethylene-pyrimidine-2, 4, 6-trione derivatives

A mixture of compounds containing various aldehydes (1 mmol) and barbituric acid (1

mmol) in a 25 mL round-bottomed flask was added [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as MNPs (15 mg) in water (5 mL). Then the mixture was stirred under reflux conditions. After completion of the reaction, as monitored by TLC (ethyl acetate and *n*-hexane

1:3), the catalyst was magnetically recovered by adding 5 mL ethanol and, then, the product was purified *via* recrystallization from ethanol, which resulted in precipitation of the desired 5-arylmethylene-pyrimidine-2, 4, 6-trionederivatives.

General procedure for preparation of 2-arylidene malonitriles derivatives

A mixture of compounds consisting of various aldehydes (1 mmol) and malonitrile acid (1 mmol) in a 25 mL round-bottomed flask was added [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as MNPs (20 mg) in water (5 mL), and then the resulting mixture was stirred at room temperature. After completion of the reaction, as monitored by TLC (ethyl acetate and *n*-hexane 1:3), the catalyst was magnetically recovered by adding 5 mL ethanol and the product was purified *via* recrystallization from ethanol, which resulted in precipitation of the desired 2-arylidene malonitriles derivatives.

Results and Discussion

Characterization of novel 1-methyl imidazole-based ionic liquid-stabilized on silica coated Fe₃O₄ magnetic nano particles [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ catalyst

Recently, Zolfigol and coworkers reported novel ionic liquid supported on magnetic nanoparticles for the synthesis of 1, 8-dioxo-octahydroxanthene and dihydropyrano[2, 3-c]pyrazole derivatives [28], and also Gu and *et al.* succeed to report of Brønsted acid ILs consist of sulfonyl group for the synthesis of Friedel Crafts alkylation of indoles and cyclic ketones and also Gu and *et al.* succeed to report of Brønsted acid ILs consist of sulfonyl for the synthesis of Friedel Crafts alkylation of indoles

and cyclic ketones [29-30]. In this research study, we report a new 1-methyl imidazole-based ionic liquid-stabilized on silica-coated Fe₃O₄ magnetic nano particles [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄. 1-methyl imidazole-based ionic liquid-stabilized on silica-coated Fe₃O₄ magnetic nano particles [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as a new, magnetic Brønsted acid nanocatalyst which was synthesized and characterized by FT-IR, SEM, TGA, XRD, and VSM [27].

Application of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as a magnetic Brønsted acid nanocatalyst for the synthesis of 5-arylmethylene-pyrimidine-2,4,6-trione derivatives

At first, the reaction between the benzaldehyde (1 mmol) and barbituric acid (1 mmol) was investigated in different amounts of catalyst as a magnetic Brønsted acid nanocatalyst under reflux conditions. The results are listed in Table 1. As seen Table 1, the reaction was carried out in various conditions. The best results were achieved when the reaction was carried out in the 15 mg of MNP catalyst in water at reflux conditions (Table 1, entry 14). Entry 1 shows that the reaction in catalyst-free conditions was without product, even after 120 minute. Furthermore, to compare the result of the reflux conditions versus solvent-free or room temperature, a mixture of benzaldehyde and barbituric acid by 15 mg of MNPs catalyst was investigated. As can be seen in Table 1, when the reaction is carried out in the solvent-free conditions, the reaction time was long and the product low (Table 1, entry 2-8). When the reaction is carried out in various solvents at room temperature, the reaction time was fairly long and the product was between 68-77% (Table 1, entry 9-12).

Table 1. Result of amount of the catalyst, solvent and temperature in the synthesis of 5-arylmethylene-pyrimidine-2, 4, 6-trione derivatives

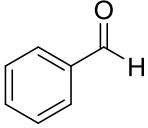
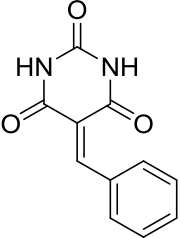
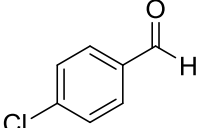
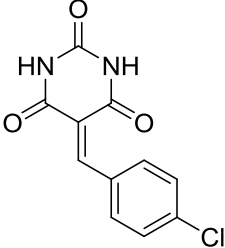
Entry	Catalyst loading (mg)	Temperature (° C)	Solvent (5mL)	Time (min)	Yield (%) ^a
1	Catalyst-free	r.t.	Solvent-free	120	N.R
2	1	r.t.	Solvent-free	12 h	12
3	5	r.t.	Solvent-free	12 h	15
4	7	r.t.	Solvent-free	10 h	42
5	10	r.t.	Solvent-free	10 h	65
6	15	r.t.	Solvent-free	6 h	68
7	20	r.t.	Solvent-free	6 h	55
8	25	r.t.	Solvent-free	6 h	55
9	15	r.t.	EtOH	2 h	87
10	15	r.t.	H ₂ O	2 h	87
11	15	r.t.	CH ₂ Cl ₂	1.5 h	68
12	15	r.t.	<i>n</i> -Hexane	3 h	73
13	15	Reflux	EtOH	25	92
14	15	Reflux	H ₂ O	25	96
15	15	Reflux	CH ₂ Cl ₂	60	83
16	15	Reflux	<i>n</i> -Hexane	60	85
17	20	Reflux	H ₂ O	60	86
18	25	Reflux	H ₂ O	60	85

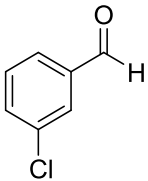
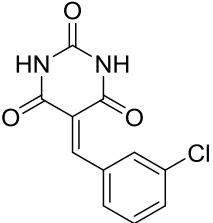
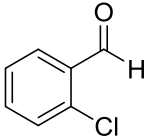
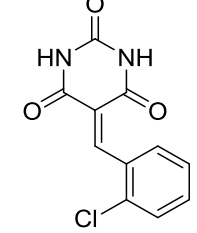
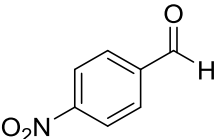
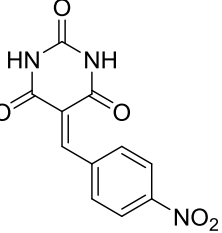
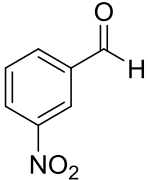
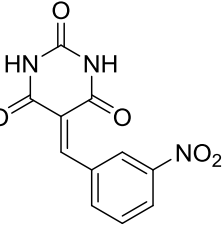
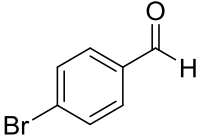
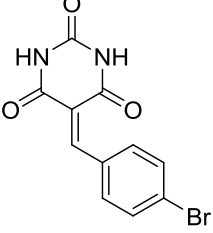
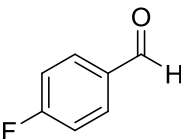
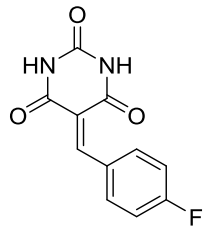
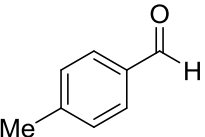
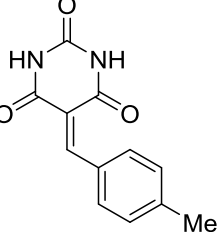
^a Isolated yield

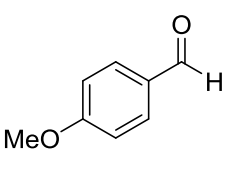
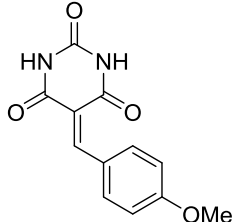
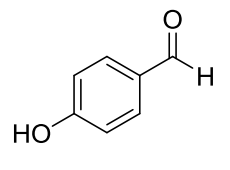
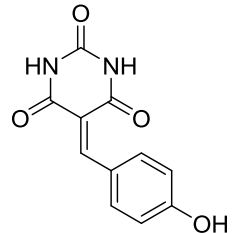
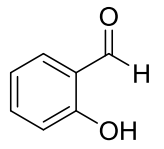
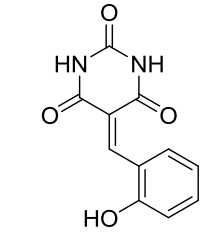
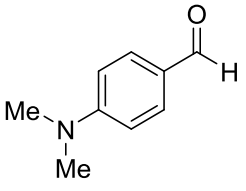
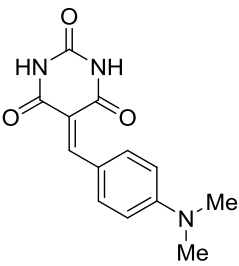
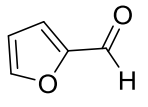
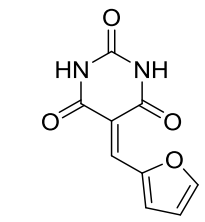
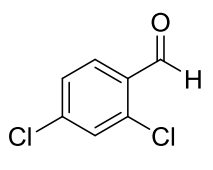
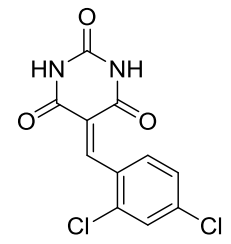
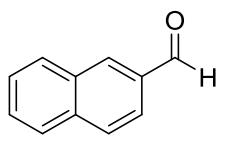
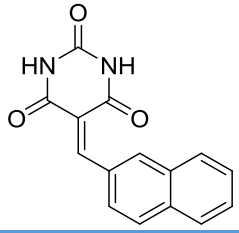
After optimization of the reaction conditions, in order to study the efficiency of [nano-Fe₃O₄@SiO₂-(CH₂)₃-1-methyl imidazole]HSO₄ in this reaction, various aldehydes were capably reacted with barbituric acid under the same

conditions (Table 2). As can be seen in Table 2, it seems that, all aldehydes (electron-donating and electron-withdrawing groups) reacted to furnish the corresponding products in excellent yields and short reaction times.

Table 2. Synthesis of 5-arylmethylene-pyrimidine-2, 4, 6-trione derivatives using 15 mg of [nano-Fe₃O₄@SiO₂-(CH₂)₃-1-methyl imidazole]HSO₄

Entry	Aldehyde	Product	Time (min)	Yield (%) ^a	TON	TOF
1			30	95	118.7	3.95
2			25	96	120	4.8

3			25	93	116.2	4.6
4			28	94	117.5	4.1
5			25	96	120	4.8
6			30	95	118.7	3.9
7			22	94	117.5	5.3
8			25	91	113.7	4.5
9			22	93	116.2	5.2

10			16	95	118.7	7.4
11			28	91	113.7	4.0
12			35	93	116.2	3.3
13			16	90	112.5	7.0
14			20	89	111.2	5.5
15			27	92	115	4.2
16			25	95	118.7	4.7

^a Isolated yield

To improve the efficiency of the catalyst, the turnover number (TON) and turnover frequency (TOF) for the [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄ were calculated (Table 2 and 4). For this purpose, the acidic content of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄ was characterized by determining the H⁺ of the catalyst and titration with NaOH (0.01 mol/L). For the calculation of mol% of catalyst, [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄ (15 mg) was titrated by 0.8 mL of NaOH (0.01 mol/L). We calculated TOF and TON for the synthesis of 5-arylmethylene-pyrimidine-2, 4, 6-trione and 2-arylidene malononitriles derivatives (Table 2 and 4).

Application of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄ as a magnetic Brønsted acid nanocatalyst for the synthesis of 2-arylidene malononitriles derivatives

After the successful synthesis of 5-arylmethylene-pyrimidine-2, 4, 6-trione derivatives using 15 mg of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄, we decided to use MNPs in the synthesis of the 2-arylidene malononitriles derivatives *via* the reaction of aldehyde and malononitrile. At first, the reaction between benzaldehyde (1 mmol) and malononitrile (1 mmol) was assessed in different amounts of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄

as a magnetic Brønsted acid nanocatalyst under room temperature. The results are presented in Table 3. As can be seen in Table 3, the reaction was carried out in various conditions. The best results are achieved when the reaction is carried out in the 20 mg of MNP catalyst in water at room temperature (Table 3, entry 10).

Entry 1 demonstrates that the reaction in catalyst and solvent-free conditions was without product, even after 120 min. Furthermore, in order to compare the result of the solution at room temperature versus solution at reflux conditions, a mixture of benzaldehyde and malononitrile by 20 mg of MNPs catalyst was investigated. As can be seen in Table 3, when the reaction is carried out in the solvent-free conditions, the reaction time was long and the product was low (Table 1, entry 2-8). When the reaction is carried out in various solvents at reflux conditions, the reaction time was fairly long and the product was between 80-89% (Table 1, entry 13-16).

In order to study the efficiency of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methylimidazole]HSO₄ in this reaction, various aldehydes were capably reacted with malononitrile under the same conditions (Table 4). As seen in Table 4, it seems that all aldehydes (electron-donating and electron-withdrawing groups) reacted to furnish the corresponding products in excellent yields and short reaction times.

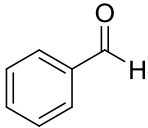
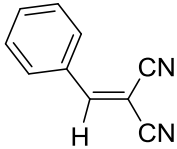
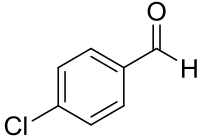
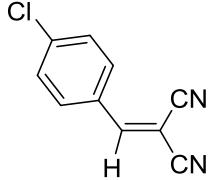
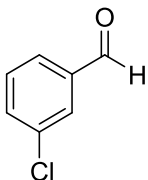
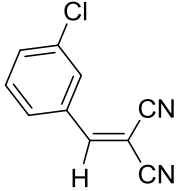
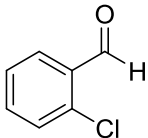
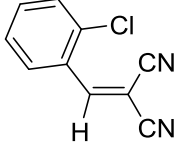
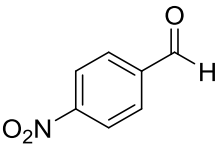
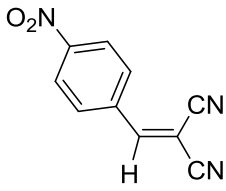
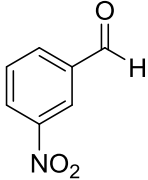
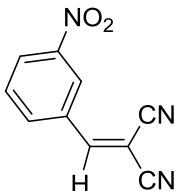
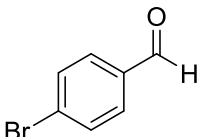
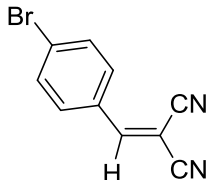
Table 3. Result of amount of the catalyst, solvent and temperature in the synthesis of 2-arylidene malononitriles derivatives

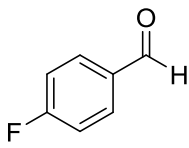
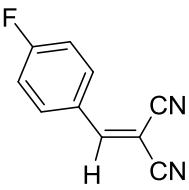
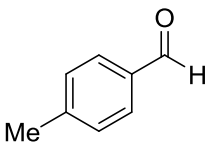
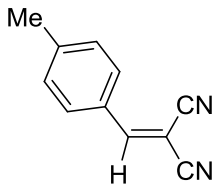
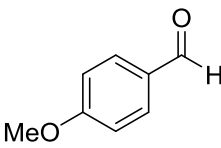
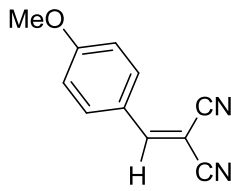
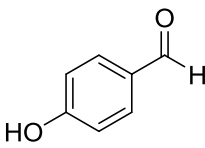
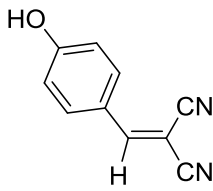
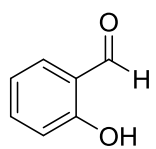
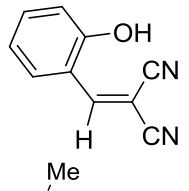
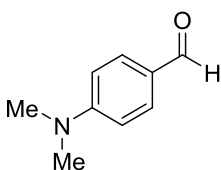
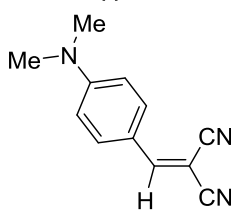
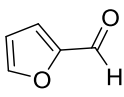
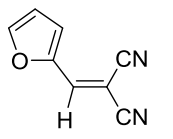
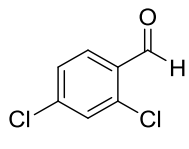
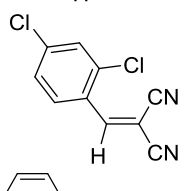
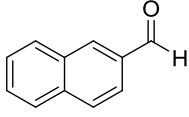
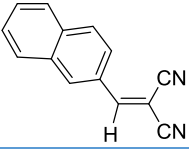
Entry	Catalyst loading (mg)	Temperature (°C)	Solvent (5 mL)	Time (min)	Yield (%) ^a
1	Catalyst-free	r.t.	Solvent-free	120	N.R
2	1	r.t.	Solvent-free	8 h	30
3	5	r.t.	Solvent-free	8 h	45
4	7	r.t.	Solvent-free	8 h	50
5	10	r.t.	Solvent-free	6 h	65
6	15	r.t.	Solvent-free	5 h	71
7	20	r.t.	Solvent-free	5 h	75
8	25	r.t.	Solvent-free	5 h	60
9	20	r.t.	EtOH	32	91

10	20	r.t.	H ₂ O	25	95
11	20	r.t.	CH ₂ Cl ₂	25	73
12	20	r.t.	<i>n</i> -Hexane	60	81
13	20	Reflux	EtOH	70	89
14	20	Reflux	H ₂ O	70	89
15	20	Reflux	CH ₂ Cl ₂	70	83
16	20	Reflux	<i>n</i> -Hexane	70	80

^a Isolated yield

Table 4. Synthesis of 2-arylidene malononitriles derivatives using 20 mg of [nano-Fe₃O₄@SiO₂-(CH₂)₃-1-methyl imidazole]H₂SO₄

Entry	Aldehyde	Product	Time (min)	Yield (%) ^a	TON	TOF
1			25	95	118.7	4.7
2			18	97	121.2	6.7
3			20	94	117.5	5.8
4			20	94	117.5	5.8
5			22	96	120	5.4
6			25	93	116.2	4.6
7			15	92	115	7.6

8			18	90	112.5	6.2
9			12	92	115	9.5
10			10	94	117.5	11.7
11			25	93	116.2	4.6
12			30	89	111.2	3.7
13			20	92	115	5.7
14			22	90	112.5	5.1
15			30	93	116.2	3.87
16			25	94	117.5	4.7

^a Isolated yield

Finally, we evaluated the recovery and reusability of the [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-

methyl imidazole]HSO₄ for the synthesis of 5-(4-chlorobenzylidene) pyrimidine-2,4,6(1H,

3H, 5H)-trione by a condensation reaction between 4-chlorobenzaldehyde and barbituric acid and the synthesis of 2-(4-nitrobenzylidene)malononitrile by a condensation reaction between 4-nitrobenzaldehyde and malononitrile. The results are summarized in Table 5 and 6. After the separation of the product with an external

magnet, the catalyst was washed with EtOH (2×5 mL), dried, and stored for another consecutive reaction run. A very small decrease is visible for the 5-(4-chlorobenzylidene)pyrimidine-2,4,6(1H,3H,5H)-trione from 96% to 90%, also, a very small decrease is visible for the 2-(4-nitrobenzylidene)malononitrile from 96% to 89%.

Table 5. Reusability of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ for the synthesis of 5-(4-chlorobenzylidene)pyrimidine-2,4,6(1H,3H,5H)-trione

Run	1	2	3	4	5	6	7
Isolated yield (%)	96	95	94	93	93	91	90
Reaction time (min)	25	25	27	29	29	31	33

Table 6. Reusability of [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ for the synthesis of 2-(4-nitrobenzylidene)malononitrile

Run	1	2	3	4	5	6	7
Isolated yield (%)	96	95	95	93	92	90	89
Reaction time (min)	22	22	23	25	25	27	30

Conclusions

In this research study [nano-Fe₃O₄@SiO₂@(CH₂)₃-1-methyl imidazole]HSO₄ as a novel, efficient, and green nanocatalyst was synthesized and characterized by FT-IR, SEM, TGA, XRD, and VSM analysis. We have developed an eco-friendly and green procedure for the synthesis of 5-arylmethylene-pyrimidine-2,4,6-trione derivatives and 2-arylidene malononitriles derivatives in water at ambient temperature. The notable advantages of this method are green and simple experimental procedure, good yields of products, short reaction time, operational simplicity, mild reaction conditions, and recovery and reusability of the catalyst with external magnet.

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