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FULL PAPER

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Design and characterization of Dendrimer of MNPs as a novel, heterogeneous and reusable nanomagnetic organometallic catalyst for one-pot synthesis of hydroxyl naphthalene-1,4-dione derivatives under solvent-free conditions

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A novel super acidic magnetic nanoparticle as catalyst was successfully synthesized. The preparation of this dendrimer sulfonic acid functionalized γ-Fe_2O_3 magnetic core-shell silica nanoparticles as a new recoverable and heterogeneous nanocatalyst was described. The new catalyst was characterized using various techniques such as scanning electron microscopy (SEM), energy dispersive spectrum (EDS), and thermo gravimetric synthesis (TGA). Moreover, we have examined the catalytic activity of the catalyst for one-pot, efficient and facile synthesis of 2-hydroxy-1,4-naphthoquinone derivatives *via* a three-component condensation reaction of 2-hydroxynaphthalene-1,4-dione, aromatic aldehydes and aniline derivatives. High yields of products, short reaction times, waste-free, mild, ambient and solvent-free reaction conditions are advantages of this protocol. Also, the catalyst can be easily recovered by an external magnetic and reused several times without significant loss of its catalytic activity.

KEYWORDS
2-Hydroxy-1,4-naphthoquinones, dendrimer magnetic nanoparticles, nanomagnetic organometallic catalyst

1 | INTRODUCTION

A class of macromolecules with specific properties and functional groups have been utilized as dendrimers.[1,2] Dendrimers are highly branched and globular macromolecules which show unique properties such as their high degree of branching, multivalency, globular architecture and well-defined molecule weight, make them promising new scaffoleds for drug delivery.[3,4] Research groups demonstrated that dendronization can significantly improve the properties of catalysts immobilized on polymeric support.[5–8] Among them, dendrimers immobilized on silica have been investigated intensively and applied for several catalytic organic transformations.[9]

The use of heterogeneous catalysts has been widely studied. These catalysts are easily removable from the reaction media by simple filtration after completion of the reaction and in many cases, they are reusable.[10–14] Magnetic core-shell nanostructures were forecast as magnetic strong catalysts in organic reactions. Within the past few years, magnetic nanoparticles emerged as a new class of semi-heterogeneous supports for catalysts.[15] Fe_3O_4 and γ-Fe_2O_3 magnetic silica nanoparticles (MSNPs) have supermagnetic properties, so that these nanoparticles can be easily recycled in the presents of an external magnetic field. Other advantages of these catalysts are mentioned to high surface area, superior stability of thermal and chemically, surface modification ability, low toxicity and cost. A silica shell does not only protect the magnetic cores, but can also prevents the direct contact of the magnetic core with additional agents linked to the silica surface thus avoiding unwanted interactions. Silica coatings have several advantages arising from their stability under aqueous conditions (at least if the pH value is sufficiently low), easy surface modification and easy control of interparticle interactions, both in solution and within structures, through variation of the shell thickness.

MSNPs can be easily synthesized at room temperature under mild conditions and the functionalization of these materials with reactive species should be made to facilitate incorporation of other molecules on the surface of the γ-Fe_2O_3/SiO_2 nanoparticles.[16–19] The existence of many hydroxyl groups on the γ-Fe_2O_3 MNPs surface leads to a reaction with alkoxysilane reagents such as tetraethyl orthosilicate (TEOS) to form Si-O bonds and provide reaction sites for further functionalization of γ-Fe_2O_3/SiO_2 MSNPs.[20–23] For the modification and functionalization of the surface of γ-Fe_2O_3/SiO_2 MSNPs, the (3-aminopropyl)triethoxysilane (APTES) and some silane reagents as potential candidates were used.[10,13,16–19]

An important class of convergent organic reactions that have an import and role in organic chemistry are multi-component reactions (MCRs).[24] In this reaction three or more starting materials react together to form a unique product. The ability of the formation of multi-bonds in a one-pot reaction with high atom efficiency and mild and environmentally benign conditions is due to special interest. Researches on MCRs have received a rapidly increasing attention in both academic and industrial researches.[25]

2-Hydroxy-1,4-napthoquinone (HNQ) has been known for the past 4000 years, is principal natural dye in the leaves of henna.[26] Today, semipermanents hair dyes containing henna as well as its pure dye ingredient HNQ are extensively used and have become increasingly popular because of their natural origin.[27] Moreover, molecules containing the hetrocyclic quinone group constitute an important class in organic chemistry because of their various biological activities (such as antitumor, antibacterial and anti-inflammatory agents), industrial applications, and potential as intermediates in the synthesis of hetrocycles.[26]

Herein, we described a method for synthesis of a novel dendrimer super acidic magnetic nanoparticles catalyst (Scheme 1). We used this dendrimer super acidic magnetic nanoparticles as catalyst for the one-pot synthesis of 2-hydroxy-1,4-naphthoquinone derivatives from 2-hydroxynaphthalene-1,4-dione, aromatic aldehydes and hetrocyclic amines under solvent free conditions (Scheme 2). The condition of Mannich reaction with this new dendrimer super acidic magnetic nano particles was very impressive. High yield of products in a few seconds time with simple work-up shows this MNPs catalyst is very suitable in the organic synthesis.

2 | EXPERIMENTAL

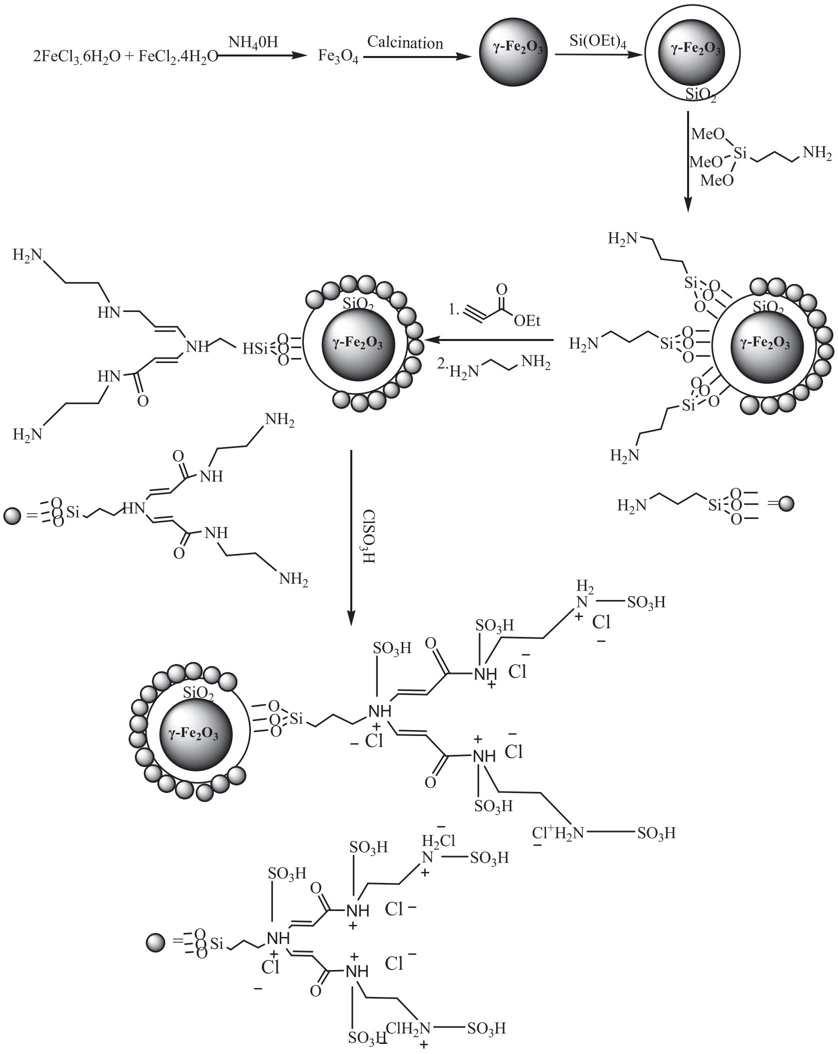
All reagents were purchased from Merck and Aldrich and used without further purification. The NMR spectra were provided on Brucker Avance 300 MHz instrument in DMSO-d_6. Infrared (IR) spectra were recorded applying a JASCO FT-IR 460 Plus spectrophotometer. Melting points were determined in open capillaries using a BUCH510 melting point apparatus. Thin layer chromatography (TLC) was performed on silica-gel Poly Gram SIL G/UV 254 plates. Elemental compositions were determined with a Leo 1450 VP scanning electron microscope equipped with an SC7620 energy dispersive spectrometer (SEM-EDS) presenting a 133 Ev resolution at 20 Kv.

2.1 | Preparation of γ-Fe_2O_3 magnetic nanoparticles (MNPs)

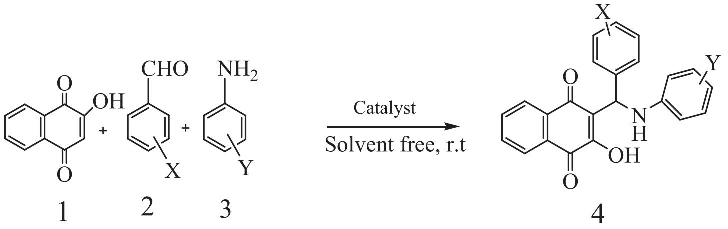
Fe_3O_4 nanoparticles were synthesized *via* the co-precipitation of Fe^{2+} and Fe^{3+} ions (molar ratio 1:2) in alkali solution. $FeCl_3.6H_2O$ (2 mmol) separately were dissolved in hydrochloric acid (4 ml) and also $FeCl_2.4H_2O$ (1 mmol) were dissolved by hydrochloric acid (1 ml). Then, two solutions of Fe^{2+} and Fe^{3+} added to each other. Resulting solution was stirred in room temperature and then was dropped a 25% NH_4OH solution to iron solution with precisely constant rate to obtain small and uniforms particles. Fe_3O_4 was collected by a permanent magnet, followed by washed several times with water and dried at 100 °C in vacuum for 24 h. In the next step, it converted to γ-Fe_2O_3 at 300 °C for 3 h.[28]

2.2 | Preparation of γ-Fe_2O_3 magnetic silica nanoparticles (MSNPs)

The γ-Fe_2O_3 was dispersed in 40 ml ethanol stirred for 1 h, and then tetraethyl orthosilicate (TEOS, 10 mL) was added to the mixture and stirred at 40 °C for 24 h. The silica-coated magnetic nanoparticles were separated by a permanent magnet, followed it washed several times with ethanol and diethyl ether and dried at 100 °C in vacuum for 12 h.



SCHEME 1 Synthesis of a novel dendrimer super acidic magnetic nanoparticles as catalyst



Catalyst: MNPs dendrimer (gamma-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H)

SCHEME 2 One-pot synthesis of hydroxyl naphthalene-1,4-dione derivatives

2.3 | Preparation of aminopropyl-modified silica-coated γ-Fe_2O_3 MNPs (γ-Fe_2O_3/SiO_2-Propyl-NH_2)

The obtained MSNPs was dispersed in 40 ml of dry toluene by sonication for 45 min, and then slowly to rise temperature to 105 °C, in this time 1 ml (3-aminopropyl)-triethoxysilane (APTES) was added to the mixture and let that the mixture was refluxed for 24 h in 105 °C. The solid product was separated with an external magnet after cooling to room temperature, washed several times with diethylether and dichloromethane and dried at 100 °C in vacuum for 12 h.[9]

2.4 | Preparation of PAMAM dendrons on the silica-coated magnetic nanoparticles (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM)

This step is the most important step in this synthesis of MNPs. Silanated particles was suspended in 50 ml of dry ethanol, and after sonication for 35 min, 0.2 ml of ethylpropiolate were added and the mixture was heated at 50 °C for at least 72 h. After cooling to the room temperature, the particles were separated magnetically, washed 3 times with dry ethanol, and dried at 100 °C under a vacuum for 24 h. Then, the nanoparticle was solved in dry ethanol and sonicated for 35 min. After that, 0.2 ml ethylenediamine dropwise was added to the mixture at room temperature. The final solution was heated at 50 °C for 72 h, Then, the magnetic nanoparticles was separated by an external magnet, washed three times with ethanol and dried at 100 °C in vacuum for 12 h.

2.5 | Preparation of PAMAM sulfonic acid functionalized γ-Fe_2O_3 MSNPs (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H)

At finally steps, result powder of nanomagnetic particles was suspended in dry dichloromethane, then chlorosolfonic acid (0.2 ml) was added dropwise at room temperature. Eventually, the mixture was separated and washed with dichloromethane and dried at 100 °C in vacuum for 12 h.

2.6 | General procedure for synthesis of 2-hydroxyl naphthalene-1,4-dione derivatives under ambient and solvent free conditions

The mixture of the aldehydes (1 mmol), anilines (1 mmol), 2-hydroxynaphthalene-1,4-dione (1 mmol), and 0.05 g super acidic magnetic nanoparticles as catalyst was stirred under ambient and solvent free conditions for the specified time. Completion of the reaction was monitored using TLC. For separation of catalyst, ethanol (5 ml) was added to the reaction mixture and heated to solve the desired product. Then, the catalyst was separated with an external magnet. Ethanolic solution was remained to cold, the precipitated crude product was filtered. The obtained crude was dried and recrystallized with EtOH to produce the pure product. The recovered catalyst was dried and reused in subsequent runs. All the desired products were characterized by comparison of their physical data with those of known compounds.

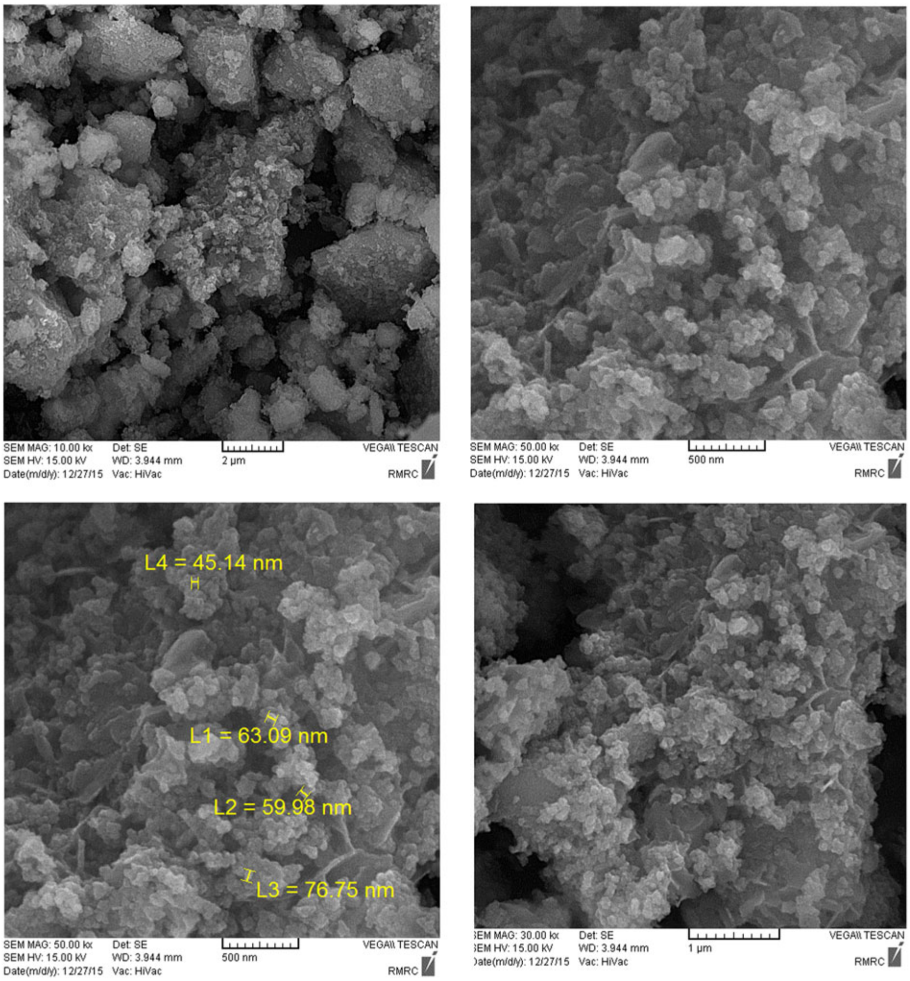


FIGURE 1 SEM images of MNPs dendrimer (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H) as catalyst

2.6.1 | Selected spectra for two Known products are given below

2-hydeoxy-3-[phenyl(phenylamino)methyl]naphthalene-1,4-dione (Table 2, Entry 1): mp 142-145 °C; IR (KBr) (ν_{max}/cm^{-1}): 3431 (OH), 3319 (NH), 1660 (CO), 1639 (CO). ^{1}H NMR (300 MHZ, DMSO-d_6): δ= 6.00 (IH, s, CH), 7.26-8.09 (14H, m, Ar and NH), 9.27 (1H, s,OH). ^{13}C NMR (75 MHz, DMSO- d_6): δ=37.4, 113.5, 123.9, 126.0, 126.6, 128.5, 129.7, 130.4, 132.8, 133.2, 133.7, 135.1, 138.1, 146.2, 156.9, 157.7, 181.9, 184.2.

2-hydroxy-3-[(4-methoxyphenyl)(p-tolyamino) methyl)]naphthalene-1,4-dione (Table 2, Entry 5): mp 144-147∘ C; IR (KBr)(ν_{max}/cm^{-1}): 3452 (OH),3324(NH), 1665 (CO), 1631 (CO).; ^{1}H NMR (300 MHz, DMSO-d_6): δ=2.31 (3H, s, CH_3),3.69 (3H, s, OCH_3),6.03 (1H, s,CH), 7.22-8.06 (12H, m, H-Ar), 9.19 (1H, s,OH).^{13}C NMR (75 MHz, DMSO-d_6): δ= 21.2, 37.3, 55.3, 113.4, 123.9, 125.9, 126.4, 128.5, 129.6, 130.3, 132.6, 133.1, 135.0,138.1, 146,156.8,157.6, 181.8,184.

3 | RESULTS AND DISCUSSION

New dendrimer MNPs as a novel catalyst was prepared based on a chemical co-precipitating method. The magnetic nanocatalyst was characterized using different techniques such as SEM, EDS and TGA. Figure 1 exhibits SEM images of MNPs dendrimer. As it is shown, the nanoparticles have uniformity spherical morphology. The average size of particles was determined using histogram curve, and it is about 65 nm (Figure 2). The energy dispersive spectrum (EDS) indicates the presence of Fe, O, Si, C, N, Cl, S atoms in the catalyst (Figure 3); which it verified the success of silica-coating and stabilization of NH-AMAM-SO_3H on chloro-functionalized γ-Fe_2O_3@SiO_2 substrate. TGA analysis of the catalyst was depicted in Figure 4. As it was observed, γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H is stable up to 300 °C and keeps its thermal stability in reaction. The lost weight at about 100 °C, corresponds to water desorption of surface catalyst. The second weights loss can be related to the decomposition of the immobilized organic part. The total weight loss was calculated to be 12.6% and the amount of organic part loaded on γ-Fe_2O_3/SiO_2 was equal to 10.8 mmol/gr.

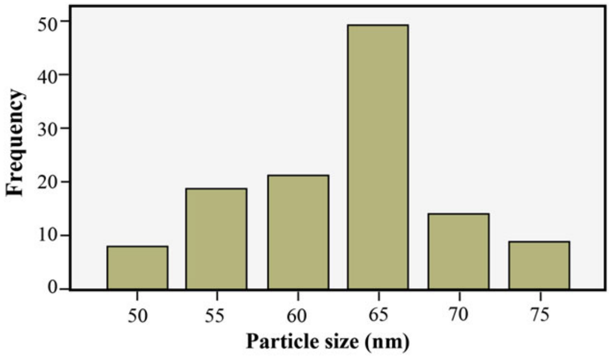


FIGURE 2 Particle size distribution histogram of MNPs dendrimer (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H) as catalyst

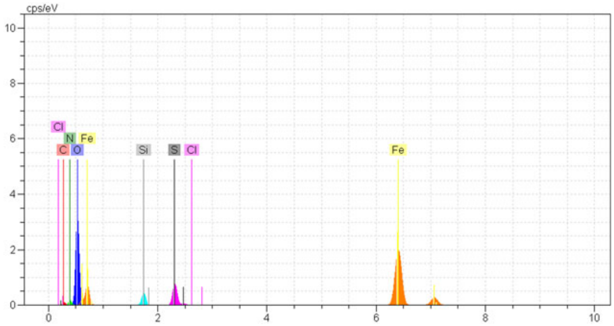


FIGURE 3 EDS spectrum of MNPs dendrimer (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H) as catalyst

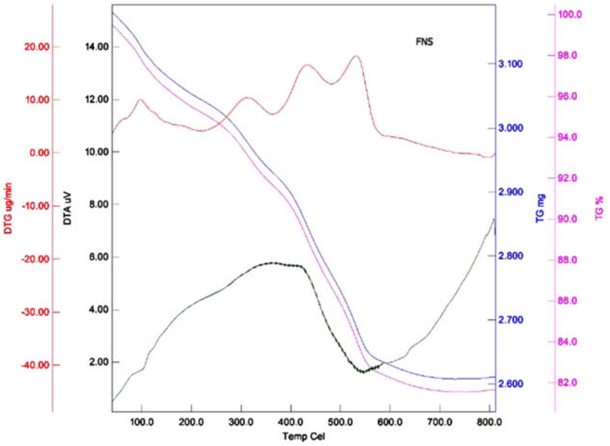


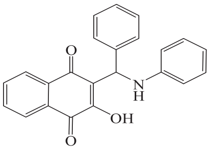
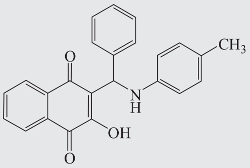
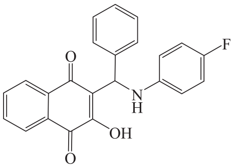
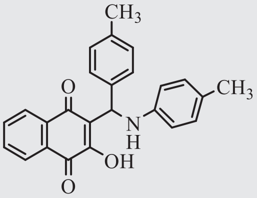
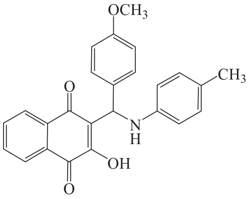
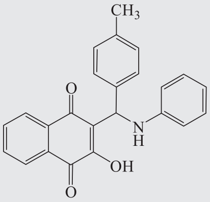
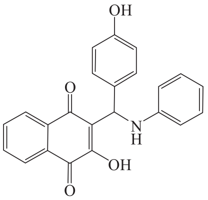
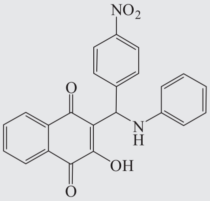
FIGURE 4 Thermogravimetric (TGA) and differential thermogravimetric (DTG) of MNPs dendrimer (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H) as catalyst

To optimize reaction conditions, the solvent-free reaction of 2-hydroxynaphthalene-1,4-dione, benzaldehyde and aniline in the presence of different amount of

TABLE 1 Optimization of the amount of γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H as catalyst in three-component synthesis of 2-hydroxy-1,4-naphthoquinone from the reaction of benzaldehyde, aniline and 2-hydroxynapthalene-1,4-dione under solvent free and ambient conditions

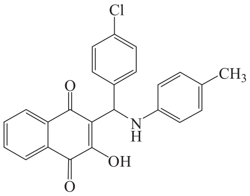
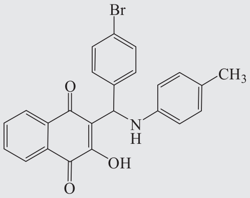
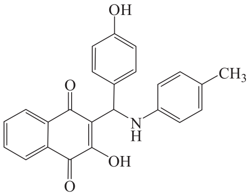
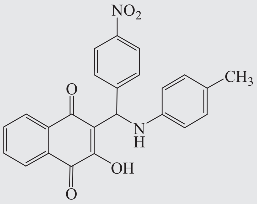
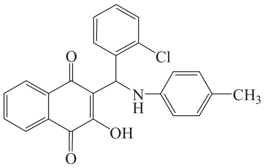
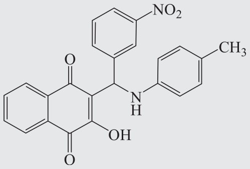
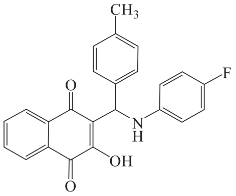
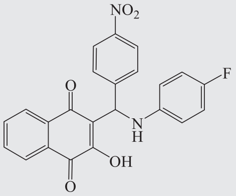
Entry	Catalyst (gr)	Time (min)	Yield (%)
1	0.02	3:30	80
2	0.05	2:45	93
3	0.1	2:00	92

TABLE 2 Three-Component synthesis of hydroxyl naphthalene-1,4-dione derivatives from the reaction of 2-hydroxynapthalene-1,4-dione, anilines and aldehydes in presence of dendrimer super magnetic nanoparticles (0.05 gr) as catalysts under solvent-free and ambient conditions

Entry	X	Y	Product	Time(min)	Yield^a	Mp: Obsd (Lit)	Ref.
1	H	H		2:45	93	143-146 (144-146)	23
2	H	4-CH_3		2:45	92	145-148 (147-150)	32
3	H	4-F		3:30	92	152-155 (150-153)	29
4	4-CH_3	4-CH_3		3:00	91	147-150 (146-149)	32
5	4-OCH_3	4-CH_3		3:15	93	142-144 (144-147)	32
6	4-CH_3	H		2:15	92	144-147 (145-148)	29
7	4-OH	H		2:15	90	188-192 (185-190)	29
8	4-NO_2	H		2:30	90	140-144 (145-148)	29

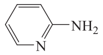
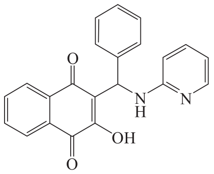
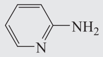
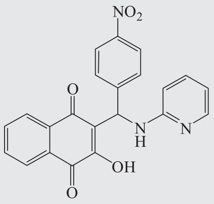
(Continues)

TABLE 2 (Continued)

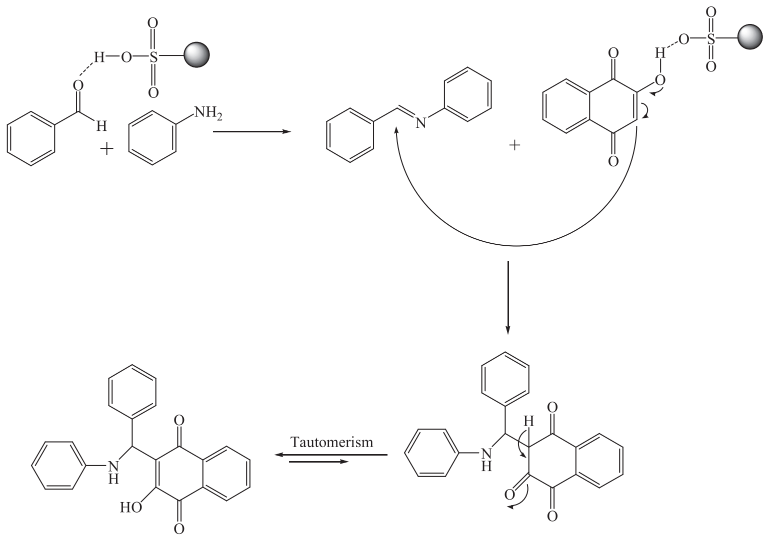
Entry	X	Y	Product	Time(min)	Yield[a]	Mp: Obsd (Lit)	Ref.
9	4-Cl	4-CH_3		2:00	93	146-149(146-149)	32
10	4-Br	4-CH_3		3:40	88	146-147 (147-150)	32
11	4-OH	4-CH_3		3:10	84	150-151 (149-152)	29
12	4-NO_2	4-CH_3		3:20	82	142-143 (141-144)	29
13	2-Cl	4-CH_3		3:15	80	147-149 (149-152)	29
14	3-NO_2	4-CH_3		3:40	80	144-143 (144-147)	32
15	4-CH_3	4-F		2:55	87	148-150 (149-152)	29
16	4-NO_2	4-F		3:10	83	146-150 (149-152)	29

(Continues)

TABLE 2 (Continued)

Entry	X	Y	Product	Time(min)	Yield[a]	Mp: Obsd (Lit)	Ref.
17	H			2:30	86	144-146 (145-148)	32
18	4-NO_2			2:50	82	146-149 (148-151)	32

[a]Yields refer to isolated yields after purifications



SCHEME 3 The suggest mechanism for preparation of hydroxyl naphthalene-1,4-dione derivatives using MNPs dendrimer (γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H) as catalyst

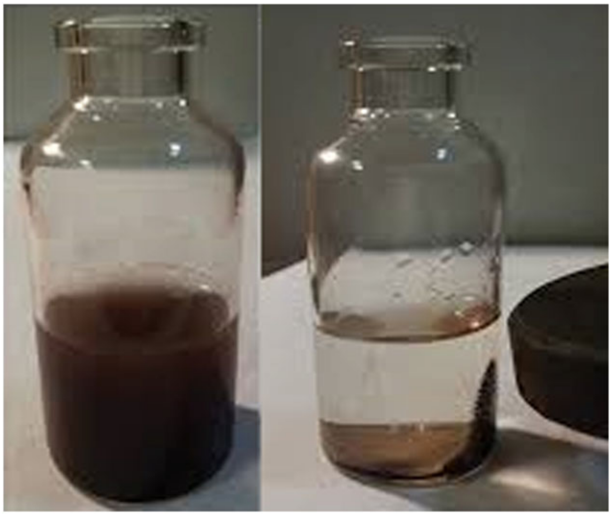


FIGURE 5 The recovery of MNPs dendrimer (catalyst) by an external magnet

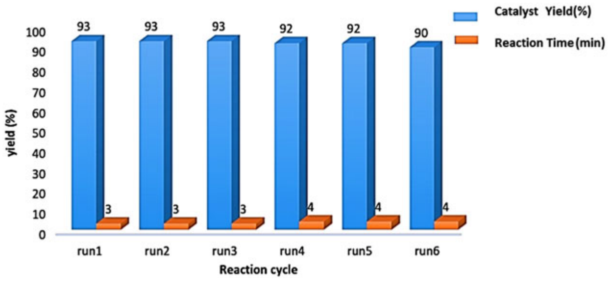


FIGURE 6 The investigation of the recyclability of the catalyst in the reaction of 2-hydroxynapthalene-1,4-dione, aniline and benzaldehyde (model reaction)

acidic dendrimer nanomagnetic particles as catalyst under ambient and solvent-free conditions was selected as a model (Table 1).

TABLE 3 Comparison the results of dendrimer super magnetic nanocatalyst with other catalysts in the literature for preparation of 2-hydroxy-3-(phenyl(phenylamino)methyl)naphthalene-1,4-dione

Entry	Catalyst	Conditions	Time	Yield (%)	Ref.
1	PPA	H_2O/ambient temperature	6 h	87	[30]
2	$InCl_3$(25 mol%)	H_2O (reflux)	6 h	91	[29]
3	ZrOCl	H_2O (Neat) 100 °C	7 h	88	[33]
4	MCM-41	EtOH/Room temperature	3 h	91	[32]
5	Nano Copper(II)oxide	Solvent-free/Room temperature	11 min	91	[12]
6	SBPPSA	Solvent-free/Room temperature	8 min	90	[23]
7	3-HPAA	Solvent-free/Room temperature	8 min	90	[31]
8	$[Et_3NH][HSO_4]$	Solvent-free/Room temperature	6 min	91	[13]
9	Dendrimer nanoparticle	Solvent-free/Room temperature	2:45 min	93	Present work

As it was shown in Table 1, the effective amount of the catalyst was investigated. Generally, the reaction rate increased with the amount of catalyst. It was found that 0.05 g of the catalyst was the appropriate amount in basis of time and yield for the preparation of the desired product. Hence, the optimal amount of catalyst was chosen to be 0.05 gr in the subsequent reactions.

Next, three-component condensation reaction of aromatic aldehydes, anilines, and 2-hydroxynapthalene-1,4-dione under optimized conditions for preparation of hydroxyl naphthalene-1,4-diones was investigated. Interestingly, a variety of aromatic aldehydes and anilines including, *ortho*, *meta*, *para*-substituted were used and corresponding products were synthesized in excellent yields at short reaction times (Table 2). In accordance with Table 2, both aromatic aldehydes and anilines carrying electron-donating or electron-withdrawing substituent act well under these reaction conditions.

We suggest mechanism according to the literature.[29] At first, aromatic aldehyde was activated by the acidic group of dendrimer super magnetic nano catalyst. Then, aniline was condensed with the activated aldehyde to afford imine. Subsequently, 2-hydroxynaphthalene-1,4-dione was added to the imine. Tautomerization converts intermediate to the desired product (Scheme 3).

The recyclability of the catalyst in the model reaction of 2-hydroxynapthalene-1,4-dione, aniline and benzaldehyde in the presence of γ-Fe_2O_3/SiO_2-Propyl-NH-AMAM-SO_3H nanoparticles was checked (Table 2, Entry 1). The catalyst can be recovered by an external magnet (Experimental section, Figure 5), washing with hot ethanol and drying at 100 °C. The catalyst was recovered in high yields and the catalyst was used in the mentioned reaction for six times. It showed the same activity as fresh catalyst without any loss of significant in its activity (Figure 6).

In order to show the accessibility of the present work in comparison with the other reported results in the literature, we have summarized some of the results for the preparation of 2-hydroxy-3-[phenyl(phenylamino) methyl]naphthalene-1,4-dione (Table 3, Entry 1). The results show that dendrimer super magnetic nanoparticle is the most efficient catalyst with respect to the reaction time and yields of the product relative to the other catalysts.

4 | CONCLUSIONS

In this research, a new synthesized dendrimer supermagnetic nano catalyst was used for preparation of 2-hydroxy-1,4-naphthoquinone derivatives under solvent-free and ambient conditions for the first time. The attractive features of this protocol are the simple procedure, cleaner reaction, and use of recyclable nanocatalyst. High yields of products, shorter reaction time toward other catalysts and easy separation of the catalyst from the reaction media without any noticeable loss of its activity after at least six times makes this green method suitable for applications in organic synthesis.

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