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ORIGINAL RESEARCH

Green synthesis of polyhydroquinolines via MCR using Fe_3O_4/SiO_2-OSO_3H nanostructure catalyst and prediction of their pharmacological and biological activities by PASS

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Abstract In this work, a library of diverse chemically and medicinally important heterocyclic polyhydroquinoline derivatives was efficiently prepared via a one-pot multi-component reaction starting from various raw materials including aromatic aldehydes, dimedone or 1,3-cyclohexandione, ethyl acetoacetate or methyl acetoacetate and ammonium acetate in the presence of Fe_3O_4/SiO_2-OSO_3H as a sulfonated silica-based magnetic nanocatalyst in high yields. Main advantages of the present practical approach are ready availability of starting materials, non-toxicity, inexpensiveness, ease of workup procedure, diversity orientation synthesis and an eco-friendly nature of the reaction. The nanocatalyst was characterized by Fourier transform infrared (FT-IR) spectra, scanning electron microscopy (SEM) images and energy-dispersive X-ray spectroscopy (EDX) spectra. The nanocatalyst was simply recovered using an external magnet and reused several times. Then, the pharmacological and biological activities of the products were theoretically examined by the prediction of activity spectra for substances (PASS) program.

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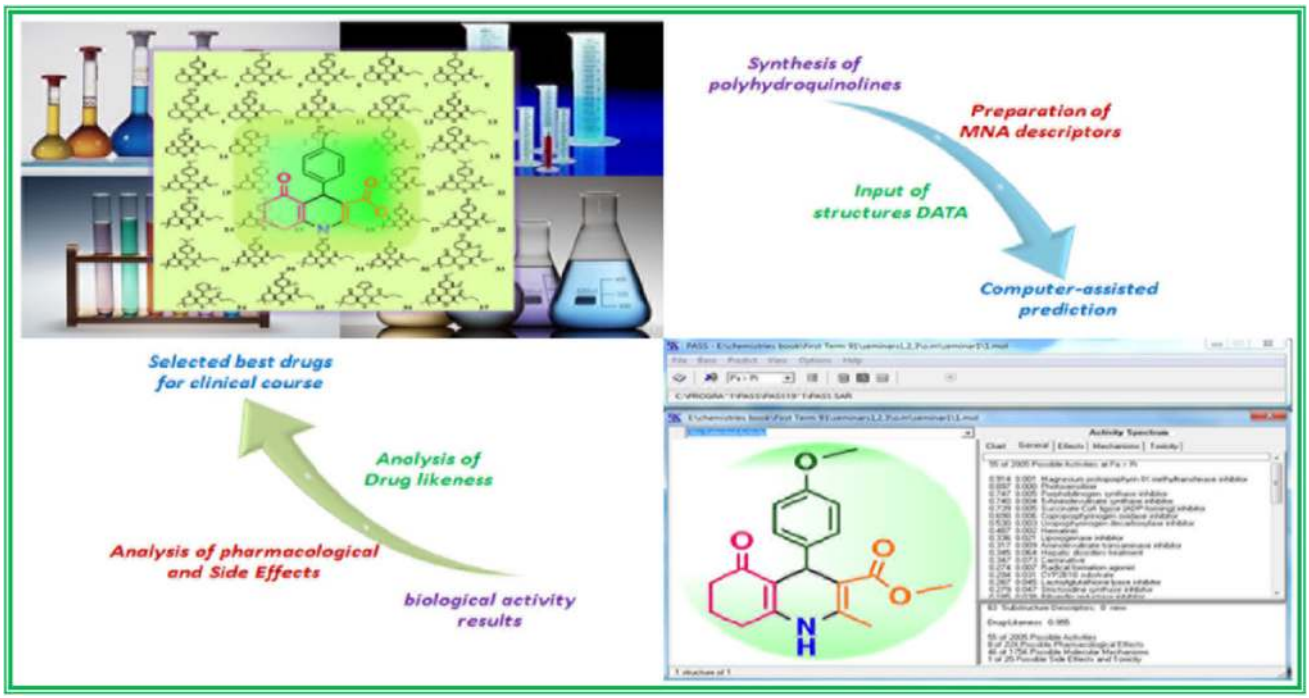
Graphical abstract



Keywords Fe_3O_4/SiO_2-OSO_3H · Nanocatalyst · PASS program · Biological activity · Polyhydroquinolines

Introduction

Multicomponent reactions (MCRs) could provide facile, efficient and practical methods in modern medicinal and combinatorial chemistry for the construction of molecular complexity and structural diversity because they have significant features such as simple design, atom-economy, environmental benignity and the possibility to construct target compounds using several assorted elements through one-pot processes and simple chemical procedures [1–5].

Recently, design, synthesis and application of supported nanoparticles as heterogeneous nanostructured catalysts have attracted much attention in chemical reactions and especially to apply them in known important organic transformations. They can be isolated from the reaction mixture by simple filtration and reused several times in subsequent reactions. Additionally, core/shell, hybrid and composite nanoparticles are very useful nanostructure materials to provide new heterogeneous catalysts because of their high specific surface area that would promote enhanced activities. Additionally, metal-based nanocatalysts such as supported magnetic nanoparticles have been widely studied in various academic and industrial research groups. Readily availability and ease of separation have made them strong alternatives to classic bulk materials [6, 7].

Due to diverse chemical, biological and medicinal properties, polyhydroquinolines are attractive and important heterocyclic compounds. Therefore, several synthetic methods have been reported in the literature for their synthesis [8–13]. Although these various methods have some advantages for the synthesis of polyhydroquinolines, they have some drawbacks such as harsh reaction conditions, use of expensive catalysts or reagents, tedious work-up procedures, sensitivity to water and toxicity. Therefore, design and development of new synthetic protocols are of prime importance. Besides these points, catalysis is an important principal of clean protocol in green chemistry, and one of the fundamental challenges chemists face now is design and development of eco-friendly catalysts. The cutting-edge research works in this field are focusing on stable and green catalysts that deserve high catalytic efficiency, ease of preparation, high selectivity, reusability and environmentally compatible properties.

The prediction of activity spectra for substances (PASS) software predicts the probability of biological properties of chemicals. The predictive accuracy percentage of thousands of chemical compounds by PASS could be about 85% [14]. The results of PASS prediction for a compound are demonstrated as a list of activity names and probability activity (P_a) values. In this case, the P_a values can be interpreted as $P_a > 0.7$, $0.5 < P_a < 0.7$ and $P_a < 0.5$. As a

result, the possibility of finding this activity in the experiments will be high, less and least, respectively [15].

Our group previous works have been on preparation and application of nanostructure heterogeneous catalysts in chemical reactions, especially MCRs [16–23]; in this article, we want to describe the practical synthesis and theoretical prediction of the pharmacological and biological activities of polyhydroquinoline derivatives **4–37**. In this purpose, sulfonated silica-based magnetic nanocatalyst (Fe_3O_4/SiO_2-OSO_3H) was used as a green heterogeneous nanocatalyst in a one-pot MCR starting from diverse aromatic and heteroaromatic aldehyde **1**, dimedone/cyclohexandione **2**, ethyl- or methyl-acetoacetate **3** and NH_4OAc.

The present MCR protocol includes various outcomes and advantages such as using green reaction media, mild reaction conditions, simple preparation and purification, high yields, simple workup procedure and reusability of the magnetic nanocatalyst. Furthermore, for the first time, the pharmacological and biological activities of the various polyhydroquinoline derivatives were computationally examined using the PASS program.

Experimental

General

The chemicals, reagents and solvents were used as received, without further purification. Melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. SEM images were taken with Zeiss-DSM 960A microscope with an attached camera. FT-IR spectra were recorded on PerkinElmer spectrometer using KBr pellets. ^{1}H NMR spectra were recorded on Bruker DRX-300 Avance spectrometer at 300.13 MHz. Elemental analysis was performed by an Elementar Analysensysteme GmbH VarioEL. EDX analysis was recorded on Numerix DXP–X10P. The magnetic property was measured on VSM-AGFM of Meghnatis Daghigh Kavir Co., Iran.

Preparation of Fe_3O_4/SiO_2-OSO_3H nanocomposite

Fe_3O_4/SiO_2-OSO_3H nanocomposite was prepared according to our previous reports using $FeCl_2$ and $FeCl_3$, tetraethyl orthosilicate (TEOS) as main starting materials, and then sulfonation by $ClSO_3H$ in an ice bath, and then, stirring at room temperature [18].

General procedure for the synthesis of polyhydroquinolines 4–37

Initially, to a mixture of 1.0 mmol of each of an aldehyde (aromatic or heteroaromatic), CH-acid (1,3-cyclohexanedione or dimedone), β-keto-ester (ethyl- or methyl-acetoacetate) and NH_4OAc in 5 mL of EtOH, 0.03 g of Fe_3O_4/SiO_2-OSO_3H was added to stir at room temperature. After the reaction was completed, as indicated by TLC, the magnetic nanostructure catalyst was isolated from the reaction mixture by an external magnet, washed with EtOH, dried and reused for the neat fresh reactions. Evaporation of the filtrate's solvent and recrystallization by ethanol (96%) gave the desired pure polyhydroquinolines **4–37**.

Characterization data of ethyl 2,7,7-trimethyl-5-oxo-4-(thiophen-2-yl)-1,4,5,6,7,8-hexahydroquinoline-3-carboxylate 36

FT-IR (KBr) (υ_{max}, cm^{-1}) = 3275, 3205, 2961, 1701, 1647, 1604, 1492, 1379. ^{1}H NMR (300.13 MHz, $CDCl_3$): δ_H (ppm) = 1.04 (3H, s, CH_3), 1.09 (3H, s, CH_3), 1.25 (3H, t, J = 7.1 Hz, CH_3), 2.20–2.35 (4H, m, $2CH_2$), 2.37 (3H, s, CH_3), 4.16 (2H, q, J = 7.1 Hz, OCH_2), 5.42 (1H, s, CH), 6.19 (1H, br s, NH), 6.84 (2H, d, J = 1.8 Hz, H_{arom}), 7.03 (1H, t, J = 2.1 Hz, H_{arom}). Anal. Calcd for $C_{19}H_{23}NO_3S$: C, 66.06; H, 6.71; N, 4.05. Found: C, 65.78; H, 6.93; N, 3.98.

Results and discussion

The Fe_3O_4/SiO_2-OSO_3H nanocatalyst was first prepared by a sol–gel method modified in our previously reported work [18]. The nanocatalyst was then characterized by SEM analysis. The particle size was studied by SEM and the identification of Fe_3O_4/SiO_2-OSO_3H morphology was based on the analysis of SEM images. As can be seen in Fig. 1, the obtained SEM image of the prepared catalyst showed proper dispersion of the composite nanostructure

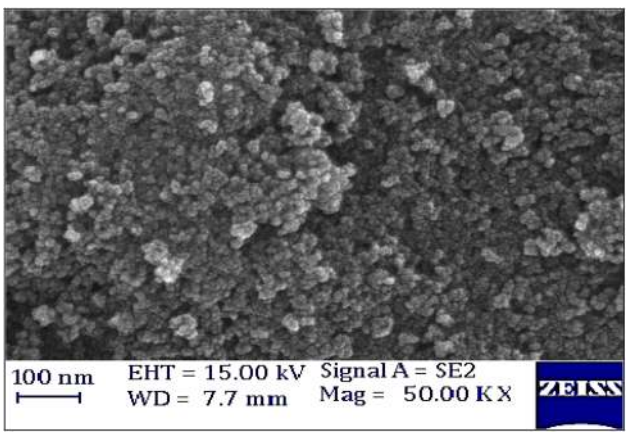
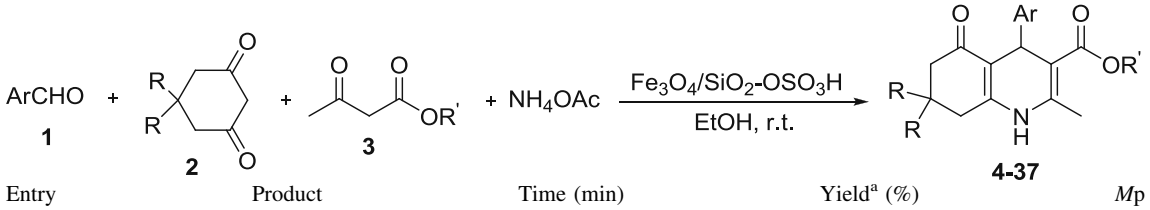


Fig. 1 SEM image of the Fe_3O_4/SiO_2-OSO_3H nanostructure

Table 1 Fe_3O_4/SiO_2-OSO_3H—catalyzed synthesis of polyhydroquinolines **4–37** via MCR strategy



Entry	Product	Time (min)	Yield[a] (%)	*M*p (°C)
1	**4**	45	97	207–208
2	**5**	60	92	243–244
3	**6**	35	85	235–236
4	**7**	60	94	258–260
5	**8**	60	95	257–259
6	**9**	40	96	255–256
7	**10**	30	84	257–258
8	**11**	50	92	245–246
9	**12**	50	96	233–235
10	**13**	50	95	175–176
11	**14**	60	95	212–214
12	**15**	45	85	196–198
13	**16**	50	96	245–248
14	**17**	50	97	235–237
15	**18**	45	94	249–250
16	**19**	55	95	236–237
17	**20**	30	90	236–237
18	**21**	50	87	186–188
19	**22**	60	94	189–191
20	**23**	40	90	292–293
21	**24**	60	96	256–258
22	**25**	45	97	272–273
23	**26**	40	89	254–256
24	**27**	50	89	263–265
25	**28**	30	92	248–250
26	**29**	50	88	256–258
27	**30**	50	90	221–223
28	**31**	40	85	215–217
29	**32**	55	96	233–234
30	**33**	45	95	250–252
31	**34**	60	94	250–252
32	**35**	40	90	215–217
33	**36**	55	85	208–211
34	**37**	45	96	237–239

[a] Isolated yield

and also almost uniform sizes and good spherical morphology of the nanoparticles.

Furthermore, EDX analysis of the magnetic nanocatalyst indicated the presence of Si, S, O and Fe elements in the nanocomposite and the amount of iron loading in Fe_3O_4/SiO_2-OSO_3H catalyst was about enough to act in the magnetic field.

To investigate the catalytic application of the present composite nanostructure, an important organic reaction was used. A pilot experiment was started from 1 mmol of

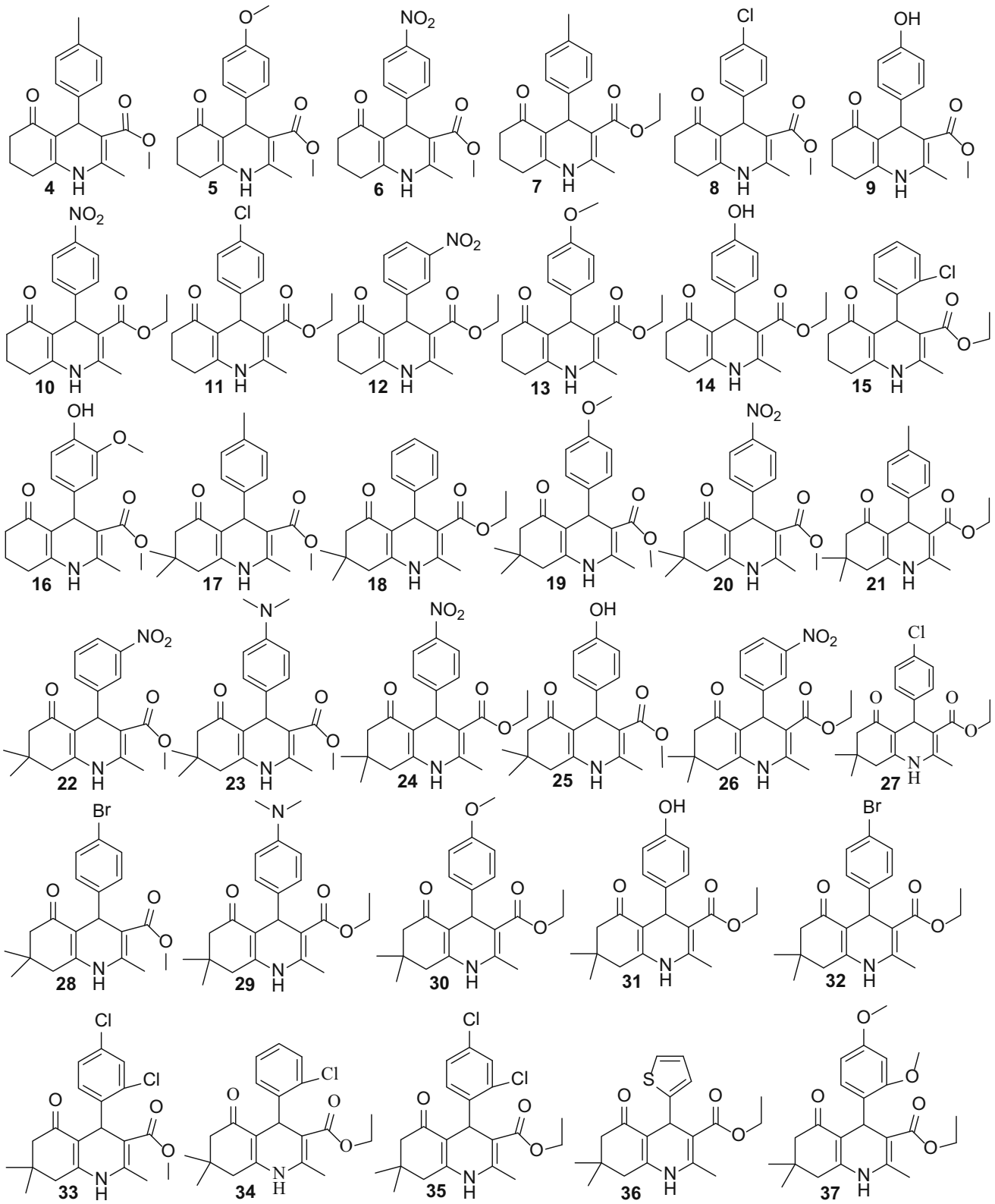


Fig. 2 The chemical structures of the products **4–37**

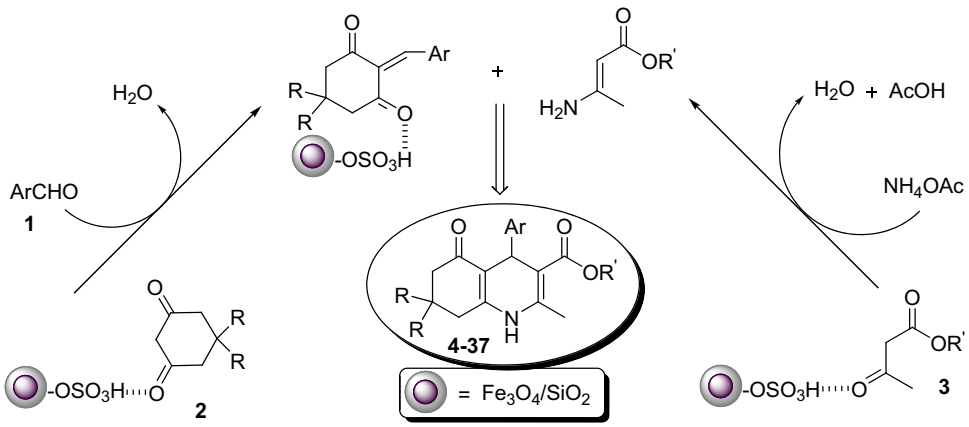
Table 2 The prediction of biological activities of **4–37**

Entry	Products	% PASS activities ($P_a > P_i$)							% side effects and toxicity ($P_a > P_i$)			% drug likeness ($P_a > P_i$)
		A_1	A_2	A_3	A_4	A_5	A_6	A_7	T_1	T_2	T_3	
1	**4**	77.2	61.0	61.1	55.0	60.9	47.3	47.8	–	19.3	24.6[a]	70.5
2	**5**	76.5	62.5	61.0	54.8	62.6	48.4	50.0	–	–	31.9[a]	76.4
3	**6**	76.3	69.7	60.9	56.2	65.0	51.3	56.1	–	24.3	28.3[b]	49.9
4	**7**	76.2	58.1	58.3	53.7	59.0	45.0	43.9	17.3	24.1	21.1[a]	67.7
5	**8**	76.0	59.4	56.6	53.5	57.9	46.3	46.3	–	–	29.5[a]	61.9
6	**9**	75.4	56.8	55.6	52.3	58.2	46.5	43.3	–	–	24.3[a]	84.1
7	**10**	75.3	64.6	58.3	55.0	63.0	48.4	51.0	22.7	29.6	30.7[b]	47.8
8	**11**	75.0	57.1	54.5	52.3	55.1	44.3	42.6	19.1	22.3	29.4[c]	59.2
9	**12**	74.8	65.0	58.5	54.4	62.9	48.6	51.6	22.5	29.9	31.4[b]	45.2
10	**13**	74.5	58.6	57.4	53.1	61.1	45.8	46.1	–	21.5	29.0[a]	73.3
11	**14**	74.3	55.2	53.5	51.0	55.6	44.5	39.8	–	21.5	21.0[a]	81.7
12	**15**	72.0	60.4	59.1	55.2	60.1	45.3	45.0	20.4	22.7	33.5[c]	64.2
13	**16**	71.9	55.4	55.0	50.9	59.2	44.5	42.2	–	–	31.5[a]	86.5
14	**17**	70.9	57.2	58.8	52.5	54.5	44.8	44.0	–	–	22.8[a]	51.0
15	**18**	70.2	57.1	59.0	51.9	55.1	44.5	43.4	21.0	19.9	21.2[a]	55.3
16	**19**	69.6	58.1	58.8	52.3	57.9	45.7	46.1	–	–	29.7[a]	58.7
17	**20**	69.6	63.1	58.8	54.0	60.7	48.1	51.1	18.8	21.7	25.6[b]	32.6
18	**21**	69.3	55.5	56.5	51.2	52.1	42.9	40.4	22.5	21.5	19.8[a]	48.7
19	**22**	69.0	63.5	58.9	53.4	60.7	48.3	51.7	18.8	22.1	26.4[b]	30.6
20	**23**	68.7	55.9	52.9	49.0	46.0	43.7	41.6	–	–	–	55.3
21	**24**	68.1	59.9	56.6	52.8	58.9	45.9	47.4	28.6	26.1	27.7[b]	31.4
22	**25**	67.8	54.7	53.9	49.7	51.5	44.3	39.9	–	–	22.7[a]	69.8
23	**26**	67.5	60.2	56.8	52.2	59.0	46.2	48.0	28.2	26.5	28.6[b]	29.5
24	**27**	67.2	54.9	52.9	49.7	48.8	42.3	39.3	24.9	19.9	23.8[a]	41.7
25	**28**	67.2	54.8	52.4	49.4	45.8	42.8	40.4	–	–	–	36.4
26	**29**	67.1	54.6	51.0	47.7	44.2	42.1	38.4	20.3	–	20.8[b]	53.0
27	**30**	66.6	56.0	55.9	55.7	50.7	43.7	42.7	19.7	19.1	27.1[a]	56.6
28	**31**	66.2	53.6	51.9	48.4	49.3	42.6	36.8	21.0	19.1	19.8[a]	67.2
29	**32**	65.5	53.6	50.4	48.1	43.9	41.1	37.3	22.5	22.1	–	34.9
30	**33**	62.9	57.5	55.3	53.1	51.3	43.6	42.7	19.1	–	27.6[c]	43.0
31	**34**	62.2	57.1	57.2	53.0	54.0	43.3	41.6	26.4	20.4	24.4[c]	46.5
32	**35**	61.2	55.8	53.3	51.9	49.2	42.0	39.4	28.7	19.3	32.1[c]	41.4
33	**36**	60.6	55.4	46.7	39.0	46.1	39.0	32.9	–	–	22.6[a]	27.7
34	**37**	60.5	55.7	56.9	53.0	53.6	42.9	41.3	17.0	18.8	19.2[a]	62.9

A_1 urinary incontinence treatment; A_2 calcium channel antagonist; A_3 antihypertensive; A_4 calcium channel agonist; A_5 vasodilator; A_6 calcium regulator; A_7 calcium antagonist; T_1 skin irritation, weak; T_2 eye irritation, weak; T_3: *a* ulcerogenic; *b* carcinogenic, female rats; *c* embryotoxic

1,3-cyclohexadione, 1 mmol of methyl acetoacetate, 1 mmol of *p*-methylbenzaldehyde and 1 mmol of ammonium acetate using various amounts of Fe_3O_4/SiO_2-OSO_3H at room temperature in 5 mL of EtOH. The best amount of Fe_3O_4/SiO_2-OSO_3H catalyst was 0.03 g to produce compound **4** in 97% yield after 45 min. Increasing the amounts of the catalyst did not improve the yield of the reaction. In addition, the effect of various solvents was studied in the pilot test. As a result, EtOH was the best vs different solvents with diverse range of polarities such as MeOH, H_2O, CH_2Cl_2, Et_2O and *n*-hexane.

To investigate the scope and limitations of this protocol, various raw materials were tested for the synthesis of polyhydroquinoline derivatives. As can be seen in Table 1, various aromatic and heteroaromatic aldehydes including both electron-donating and electron-accepting moieties gave efficiently the desired products in high yields. The structures of the prepared products **4–37** are shown in Fig. 2.



Scheme 1 A proposed mechanism of the synthesis of polyhydroquinolines **4–37**

In this study, evaluation of computer system for the prediction of biological activity on the set of well-known polyhydroquinoline derivatives using MNA descriptor was studied via PASS. Seven important activities extracted from PASS software include three classes: pharmacological effects, molecular mechanisms, and side effects/toxicity (Table 2). In the first column of the table, biological activities of the synthesized compounds have been sorted according to the percentage of urinary incontinence treatment activity with values of 60.5–77.2% that represent the highest average activity among the seven activities are listed below in Table 2.

The compounds **4** and **5**, with the urinary incontinence treatment values of 76.5% and 77.2%, respectively, showed the highest amount of deals as a scientific achievement. Therefore, this new finding is very important for patients suffering from urinary incontinence. The second column of Table 2 shows the toxicity and side effects of synthetic drugs. This column shows that of the 34 compounds synthesized, compounds **23** and **28** did not show toxicity and side effects. These two drugs considered for completing the clinical course of further investigation can be marketed.

The percentage of similarity of the prepared products to the commercial drugs in mentioned pharmaceutical composition were 55.3 and 63.4, respectively. As a result, in this way, use of this optimal biological properties will be easy. In general, due to less toxicity and side effects of compound **23** and also its similarity to drugs and biological activity of more than 50%, it can be used in the treatment of urinary incontinence, as calcium channel antagonist and antihypertensive.

A plausible reaction mechanism in the presence of Fe_3O_4/SiO_2-OSO_3H includes a Knoevenagel reaction between aldehydes and dimedone and a parallel Michael addition of an enamine intermediate to give polyhydroquinolines **4–37** (Scheme 1) [13].

The recoverability of the Fe_3O_4/SiO_2-OSO_3H nanocatalyst was one of the most important advantages of the heterogeneous catalysts in both industrial and academic applications. The present nanostructure catalyst can easily be separated from the reaction pot using an external magnet, washed with a solvent like ethanol or acetone, dried at room temperature, and reused at least six times. The reusability was examined in the model reaction for the synthesis of product **4**. The isolated yields were 97, 95, 94, 92, 91 and 90% for the fresh catalyst and five subsequent recycled runs, respectively. As a result, the catalytic performance and recyclability of Fe_3O_4/SiO_2-OSO_3H were better than catalysts previously reported in the literature [8–12] and also our earlier works [13, 20].

Conclusions

In summary, Fe_3O_4/SiO_2-OSO_3H was synthesized, characterized and efficiently used for the synthesis of biologically important polyhydroquinolines. In addition, this method has enough ability to be used in the synthesis of various other important heterocycles via MCRs or even traditional and classic parallel syntheses. The present nanocatalyst could easily be recycled from the reaction mixture and reused for at least six runs. Furthermore, for the first time, the pharmacological and biological activities of the various synthesized polyhydroquinoline derivatives were examined by PASS program (MNA descriptor). Therefore, with further chemical development, molecule **23** could potentially serve as medication for urinary incontinence treatment, calcium channel antagonist and antihypertensive.

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