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The novel acid-base magnetic recyclable catalyst prepared through carbon disulfide trapping process: Applied for green, one-pot, and efficient synthesis of 2,3-dihydroquinazolin-4 (1H) -ones and bis(indolyl)methanes in large-scale

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ABSTRACT

Herein, a nano acid-base catalyst using magnetic core and carbamodithioic acid functional group have been synthesized and characterized. Its efficiency in the synthesis of dihydroquinazolinones and bis(indolyl)methanes derivatives was investigated. This novel metal-free catalyst exhibited significant catalytic activity in both reactions under green and mild reaction conditions (the yield obtained for the first reaction products: 82–98 % and for the second one: 61–97 %). The catalyst displayed good recyclability with no significant loss of catalytic activity after eight runs (the conversion of the eighth run was found as 83 %, the fresh catalyst conversion was 95 %). The introduced approach is attractive due to its applicability in the large-scale synthesis of important medicinal compounds.

1. Introduction

Dihydroquinazolinones derivatives are extensively found in numerous pharmaceuticals and biologically active molecules [1]. Due to their significant pharmacological properties and medicinal importance, their synthesis has been considered in medicinal chemistry [2]. Some molecules included dihydroquinazolinone unit exhibited diverse biological and therapeutic properties such as antibacterial and anticancer activity [3].

A variety of synthetic methods have been reported to obtaining dihydroquinazolinones by using precursors of o-aminobenzonitrile [4], isatoic anhydride [5], o-nitrobenzamides [6], and o-aminobenzamides [7]. Condensation of alkyl, aryl, and heteroaryl ketones or aldehydes with anthranilamide in the presence of acidic catalysts is one the most well-known method for the synthesis of dihydroquinazolinones [8,9]. This reaction has been catalyzed by yttrium nitrate [10], thiamine hydrochloride [11], imidazolium Brønsted acid ionic liquid [12], organocatalyst [13] and Cu (II) supported on Fe_3O_4–DETA [14]. Moreover, application of some efficient, recoverable, and thermally stable nanocatalysts such as nickel complex immobilized on functionalized nanocomposite [15], platinum nanoparticles tagged on carbon nanotube [16] and graphene oxide [17] were also reported for the synthesis of dihydroquinazolinones. Available catalytic systems have some limitations such as long manufacturing processes, applying expensive materials, and creating metal contamination. So, designing efficient, recyclable, and novel metal-free catalysts seems an interesting strategy.

Another important class of heterocyclic compounds is indoles which their derivatives present in several natural products, pharmaceuticals, and agrochemicals [18–20]. Among them, bis(indolyl)methanes (BIMs) and their derivatives exhibit wide usages in medicinal chemistry, biochemistry, and pharmacology (Fig. 1) [21]. Due to their unique pharmacological activities, much attention has been paid to develop effective methods for the synthesis of bis(indolyl)alkanes; so, numerous synthetic approaches for their production are available [22]. However, these protocols suffer from some drawbacks such as the use of poisonous reagents and volatile organic solvents [23]. Hence, achieve the aims of green chemistry, founding new efficient, low-cost and environmentally friendly synthetic methods is valuable.

Among heterogeneous catalysts, magnetic nanoparticles (MNPs) are known as ideal core support due to their properties such as easy functionalization process, none toxicity, high stability, and simply separation [24–31]. Moreover, the preparation of heterogeneous catalysts by consumption of low-cost materials and through easy processes adds environmental and economic benefits to the catalytic system.

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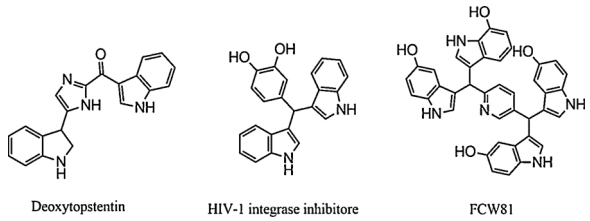


Fig. 1. Biologically active bis(indolyl)alkanes.

According to our research group interest in the preparation of nano heterogeneous catalysts and investigative their catalytic activities in organic reactions [32,33]; herein, a one-step process for the preparation of magnetic-based dithionate catalyst have been developed by collecting carbon disulfide. To the best of our knowledge, this innovative way to prepare an acid/base catalyst did not report previously. Introduced catalyst display high activity in the synthesis of dihydroquinazolinones and bis(indolyl)methane derivatives. Compared to other existing methods, our presented procedure has some advantages such as low-cost, good yields, quick reaction, and easy workup. However, the most important feature of our method was its good efficiency in the large-scale synthesis of important pharmaceutical compounds.

2. Results and discussion

The synthesis procedure of the catalyst was given in Scheme 1. The silica NH_2 functional group was reacted with carbon disulfide, through a simple and mild strategy. Then, the acid-base bi-functionalized silica ligand was treated with magnetic nanoparticles to form the final solid catalyst which is named MNPs@Si-NH_2^+/S^-. Detailed procedures were given in supporting information.

The synthesis steps were checked with the relevant analysis such as Fourier-transform infrared spectroscopy (FT-IR), thermogravimetric analysis (TGA), and elemental analysis. The FT-IR spectrum of catalyst preparation steps was given in Fig. 2(1). In the spectra of pure magnetite nanoparticles (Fig. 2(1a)), the broad bands at 3400 cm^{-1} are related to O-H and the vibration mode of Fe-O appeared as a band at 560 cm^{-1}. In spectra of b, the 1090 cm^{-1} band is related to the Si-O group vibrations, these observations confirm the expected structure (Fig. 2(1b)). The absorption bands in the spectrum of the final catalyst, vibration mode of C═S appeared at 1200 cm^{-1}, these established the structure of the final catalyst (Fig. 2(1c)).

The TGA and elemental analysis were applied to investigate the number of organic functions in the catalyst. The TGA diagram was illustrated in Fig. 2(2), the catalyst was lost its weight between 30–800 °C as a function of temperature. The first step of weight loss in the cases is related to the removal of adsorbed water and the main weight loss is related to the removal of organic pieces. These observations confirmed the stability of the catalyst at high temperatures.

The phase of the catalyst was determined by the XRD spectrum. The characteristic diffraction peaks attributed to Fe_3O_4 at 21.1°, 35.1°, 41.4°, 50.6°, 63.0°, 67.4°, and 74.1° which can be assigned to diffraction of the (220), (311), (400), (422), (511), and (440) planes, respectively of spinal structured magnetite nanoparticles (JCPDS card no. 82e1533). The XRD pattern of the Fe3O4@SiO2 displays the same set of peaks in the iron oxide particle but significate reducing in the intensity of the peak were observed in comparison with iron oxide material. Some of the new peaks are attributed to silica surfaces on the magnetic material. (Fig. 2(3)) [34].

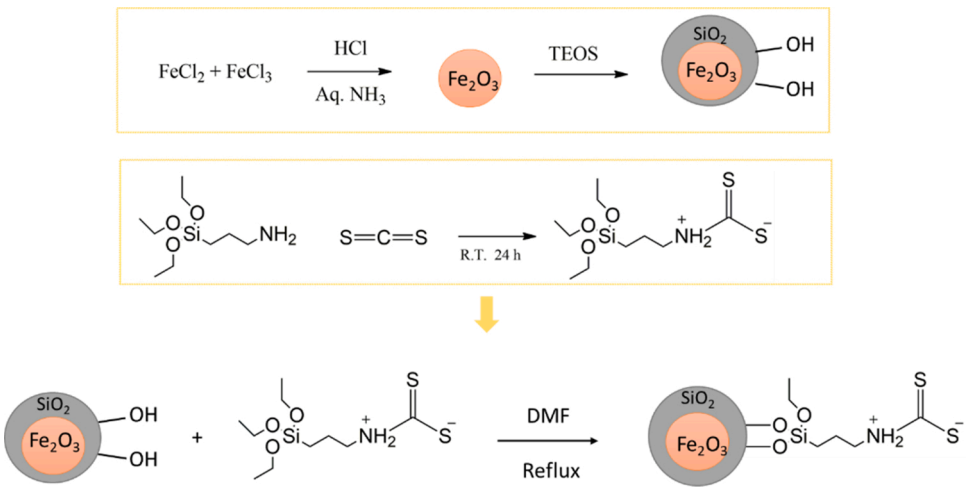
The magnetization curve of the catalyst was given in Fig. 2(4). The reduction of the magnetic behavior of the final catalyst (22.1 emu/g) in comparison with the magneticity of pure magnetic nanoparticles (69.4 emu/g), confirmed its properties and organic coating. To identified the pore structure and porous materials, the nitrogen adsorption method (BET analysis) was applied (Fig. 2(5)). A big jump in high pressure in the graph displays the microporous properties of the material. The average total pore volume is 0.4 cm^3/g, and the average surface area is calculated as 34.2 m^2/g.

The elemental analysis mapping was also used to investigate the existing elements in the catalytic hybrid (Fig. 3(1)). The images of field emission scanning electron microscopy (FE-SEM) and EDAX spectra exhibited surface morphologies and the existing metals of the catalyst (Fig. 3(2)).

Transmission electron microscopy (TEM) was also applied to more characterization of surface morphology, these images clearly exhibit spherical nano iron core and surrounded organic species in comparison with TEM of pure MNPs (Fig. 3(3)). The particle size distribution was also given in Fig. 3(3), the average size of nanoparticles was found around 13 nm.

For catalyst studies, the catalyst efficiency was investigated in 2,3-dihydroquinazolin-4 (1 H) -ones synthesis reaction. Initially, the reaction of 4-nitrobenzaldehyde (1 mmol) and 2-aminobenzamide (2) (1.2 mmol) was selected as a model reaction. The results for optimized reaction conditions were given in Table 1.

At first, the effect of different amounts of the catalyst was investigated in the model reaction, and 20 mg of catalyst was selected as the best amounts. The same condensation was then conducted at different temperatures. Compared with the reaction at 50 °C, no considerable improvement was achieved at 80 °C; room temperature was found to be less effective. Then, different solvents influenced was studied, from the



Scheme 1. Synthesis of catalyst.

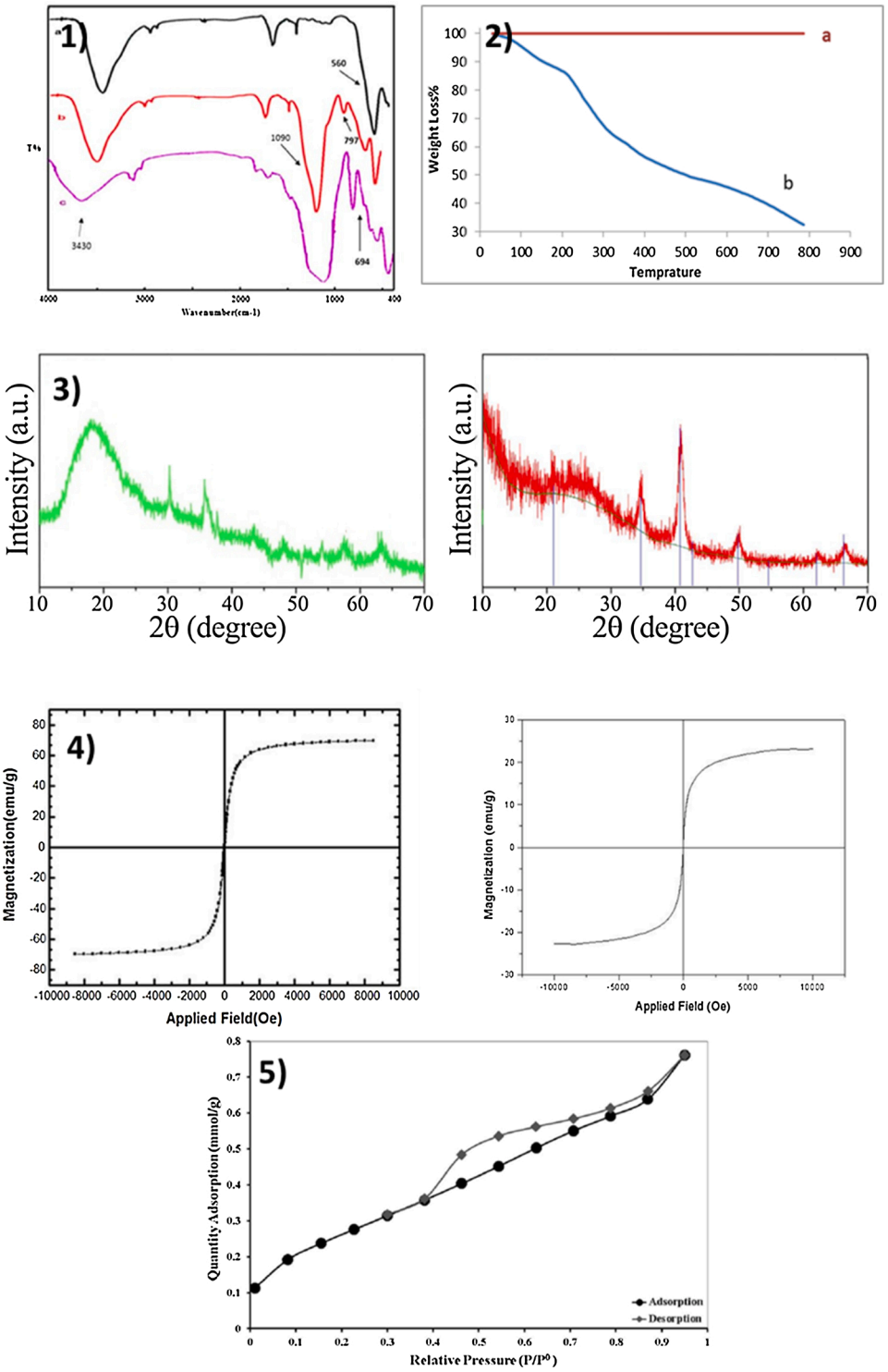


Fig. 2. 1) FT-IR spectra: (a) magnetite nanoparticles (MNPs); (b) silica-coated magnetite nanoparticles (MNPs@Si); (c) the catalyst. 2) TGA thermogram of the MNPs (a) and the final catalyst (b). 3) XRD pattern of MNPs (left) final catalyst (right). 4) Room temperature magnetization curve of MNPs (left) and final catalyst (right). 5) Nitrogen adsorption and desorption isotherms (at 77 K) for catalyst.

perspective of green chemistry, solvent-free was chosen as ideal reaction medium conditions.

After optimization of the reaction conditions, the scope of the catalyst was examined, the results were given in Table 2. The reaction of 4-nitrobenzaldehyde (1.0 mmol) with 2-aminobenzamide (1.2 mmol) was performed as a model reaction in presented magnetic nanoparticles without any ligands and no products were achieved (Table 2, entry 1). The functionalized magnetic nanoparticle with just an amino group has also no significant effect in this process (Table 2, entry 2). The catalyst with oxy-onion instead of thio-anion was synthesis to comparison, the results indicated the more efficiency of our catalyst (Table 2, entry 3). The magnetic-free catalyst was also prepared and its efficiency was examined, no significant difference between the operation of this catalyst, and our main catalyst exhibited an inert effect of the iron element on improvement of the reaction (Table 3).

After founding the best catalyst, its generality and versatility in the

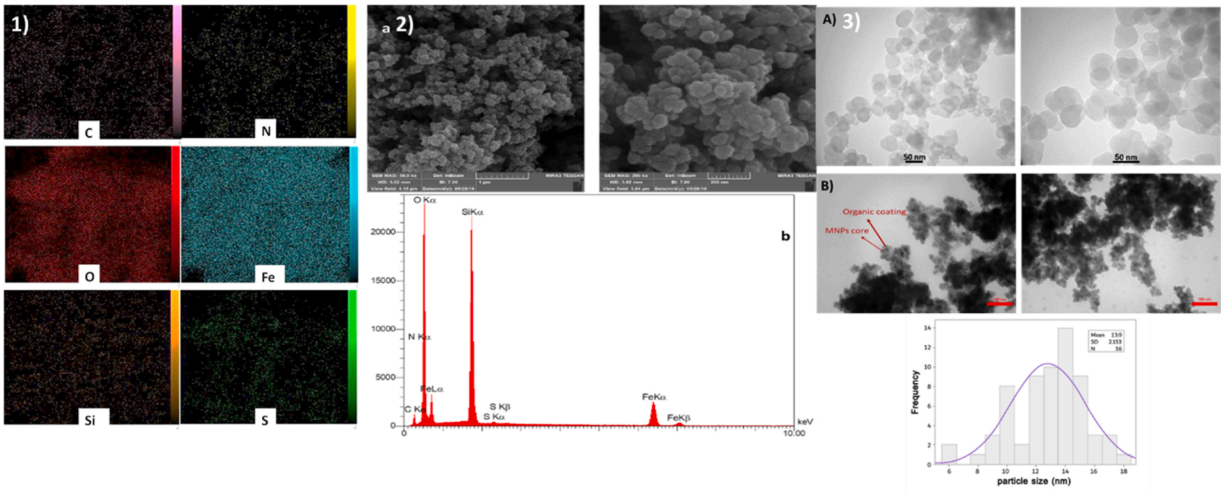


Fig. 3. 1) Analysis mapping of catalyst. 2) The FE-SEM images and (b) SEM–EDX spectrum of the catalyst. 3) TEM images of MNPs (A) and final catalyst (B) and sized distribution.

Table 1
Optimization of reaction conditions for 2,3-dihydroquinazolin-4 (1 H) -ones synthesis of 4-nitrobenzaldehyde with 2-aminobenzamide in the presence of the catalyst.[a].

Entry	Solvent	Temp (° C)	Time(min`)	cat. (mg)	Yield[b] (%)
1	EtOH	80	45	40	80
2	EtOH	r.t	45	40	70
3	EtOH	50	45	40	88
4	EtOH	50	45	40	80
5	EtOH	50	45	40	86
6	EtOH	50	45	40	73
7	H_2O	50	45	40	76
8	Solvent-free	50	45	40	95
9	Solvent-free	50	45	20	93
10	Solvent-free	50	45	50	97
11	Solvent-free	50	40	20	96
12	Solvent-free	50	30	20	95
13	Solvent-free	50	15	20	95

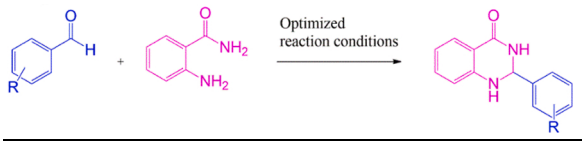
[a] Reaction carried out with 4-nitrobenzaldehyde (1.0 mmol), 2-aminobenzamide (1.2 mmol).
[b] Isolated.

Table 2
Screening of the catalysts.[a].

Entry	Catalyst	Time (h)	Yield[b](%)
1	SiO_2 Fe_2O_3	5	–
2	SiO_2 Fe_2O_3	5	32
3	SiO_2 Fe_2O_3	1	67
4	SiO_2 Fe_2O_3	0.25	95
5	SiO_2	0.25	94

[a] Reaction carried out with 4-nitrobenzaldehyde (1.0 mmol), 2-aminobenzamide (1.2 mmol), 20 mg catalyst, at 50 ℃, in solvent-free conditions.
[b] GC yield.

Table 3
Scope of dihydroquinazolinones derivatives synthesis reaction.[a].

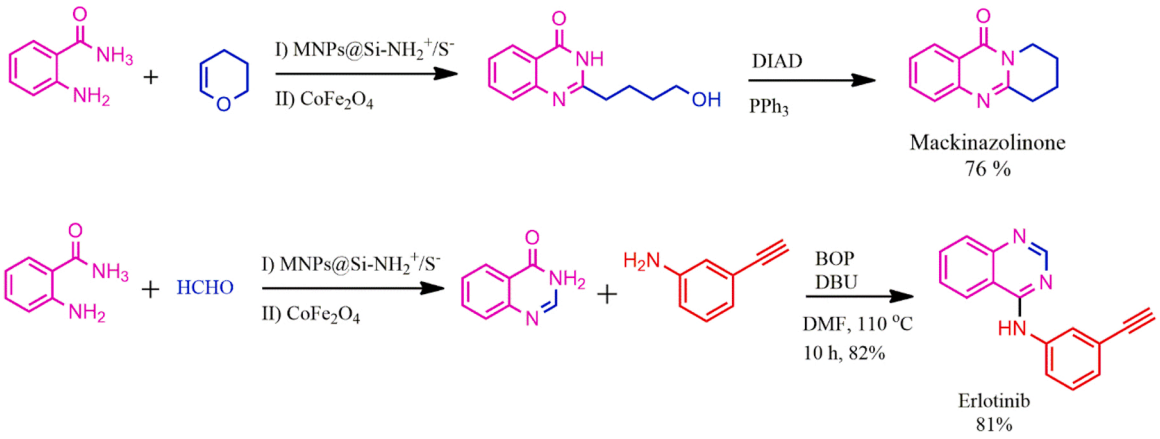


Entry	Benzaldehydes	Yield[b] (%)
1	4-Bromobenzaldehyde	90
2	4-Chlorobenzaldehyde	98
3	4-Hydroxybenzaldehyde	87
4	4-Methylbenzaldehyde	82
5	4-Methoxybenzaldehyde	90
6	Benzaldehyde	95
7	4-Nitrobenzaldehyde	96
8	Furan-2-carbaldehyde	84
9	Thiophene-2-carbaldehyde	93
10	Cyclohexanecarbaldehyde	94
11	Butyraldehyde	91
12	Acetaldehyde	88
13	Formaldehyde	82
14	Cinnamaldehyde	86

[a] Reaction carried out with benzaldehyde (1.0 mmol), 2-aminobenzamide (1.2 mmol), in solvent-free conditions, 20 mg of catalyst at 50 °C for 15 min.
[b] Isolated yield.

synthesis of 2,3-dihydroquinazolin-4 (1 H) -ones derivatives was examined. Using optimized reaction conditions, benzaldehydes contain substituents with different electronic properties reacted efficiently with 2-aminobenzamide. High yields of products were reached at a short reaction time besides safe and green reaction conditions. According to the results, it seems that the electronic properties of the precursors had no significant influence on the reaction outcomes.

To confirm the application of our method to prepare important compounds, the known antitumor agents of Mackinazolinone and Erlotinib (truncated) were synthesized on a large-scale, their structure and synthesis steps were given in Scheme 2. To achieve intermediate, an oxidizer was needed; so, considering one-pot reaction conditions, it was preferable to use a magnetic heterogeneous oxidizer. Traditionally, oxides of noble metals have been active catalysts, but their scarceness and cost promoted us to use abundant metal oxidizer. Herein, the green, low-cost and efficient nanoparticles of cobalt ferrite catalyst without any additives were applied as oxidizers. The success of this catalyst as a green oxidizer and efficient catalyst has already been confirmed [35, 36]. In addition to the mentioned benefits, it separated easily alongside



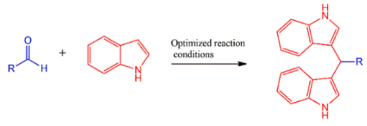
Scheme 2. Synthetic applications of catalyst for the production of important derivatives of dihydroquinazolinones.

Table 4
Optimization of 3,3′-(phenylmethylene)bis(1H-indole) synthesis reaction using benzaldehyde and 1H-indole.[a].

Entry	Solvent	T (°C)	Time (h)	Cat. (mg)	Yield[b] (%)
1	H_2O	100	10	40	83
2	H_2O	80	10	40	72
3	H_2O	60	10	40	69
4	H_2O	50	10	40	62
5	H_2O	r.t.	10	40	60
6	CH_3CN	50	10	40	49
7	MeOH	50	10	40	79
8	EtOH	50	10	40	96
9	Solvent-free	50	10	40	63
10	EtOH	50	10	20	96
11	EtOH	50	8	20	67
12	EtOH	50	5	20	95
13	EtOH	50	3	20	95
14	EtOH	r.t.	3	20	94

[a] Reaction carried out with benzaldehyde (1.0 mmol), 1H-indole (2.2 mmol).
[b] GC yield.

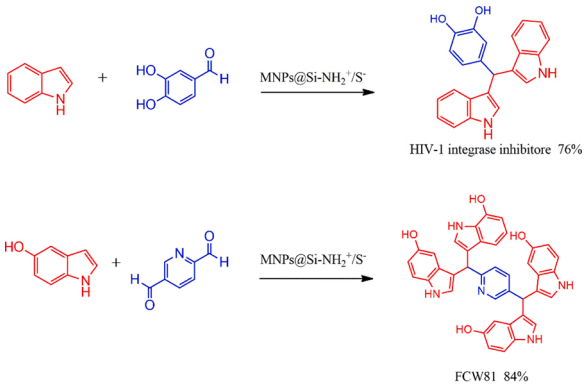
Table 5
Scope of 3,3′-(phenylmethylene)bis(1H-indole) derivatives synthesis.[a].



Entry	Aldehydes	Yield[b](%)
1	4-Bromobenzaldehyde	88
2	4-Chlorobenzaldehyde	91
3	4-Methoxybenzaldehyde	87
4	Benzaldehyde	94
5	4-Nitrobenzaldehyde	96
6	Thiophene-2-carbaldehyde	97
7	Furan-2-carbaldehyde	92
8	4-(Dimethylamino)benzaldehyde	83
9	Picolinaldehyde	95
10	2-Phenylacetaldehyde	71
11	Anthracene-9-carbaldehyde	53
12	Formaldehyde	61
13	Acetaldehyde	69

[a] Reaction carried out with benzaldehydes (1.0 mmol), 1H-indole (2.2 mmol), in 2.0 mL of EtOH, 20 mg of catalyst at r.t. for 3 h.
[b] Isolated yield.

the first catalyst by an external magnet bar from reaction media. Accordingly, the introduced method was successfully applied to the synthesis of Mackinazolinone in 76 % overall yield.



Scheme 3. Synthetic applications of catalyst for the production of important derivatives of 3,3′-(phenylmethylene)bis(1H-indole).

Furthermore, N-(3-ethynylphenyl) quinazolin-4-amine, which is a main part of Erlotinib, was obtained in 61 % yield through two steps (Scheme 2). The synthesis procedures of catalyst and products along with their characterizations were given in supplementally data file.

The excellent results obtained in the dihydroquinazolinones derivatives synthesis reaction excited us to explore the potential activity of the catalyst in 3,3′-(phenylmethylene)bis(1H-indole) synthesis reaction. To reach the optimum reaction conditions, the reaction of benzaldehyde and 1H-indole was selected as a model reaction. Different reaction conditions were examined such as type of solvent, catalyst amount, and different temperatures, results were given in Table 4.

Various aldehydes were reacted with 1H-indole to cover the scope of catalytic performance in this reaction in founding optimized reaction conditions (Table 5). The catalyst displayed higher activity for benzaldehyde in comparison with alkyl aldehydes; moreover, the electronic properties of substituted benzaldehyde had no significate effect on the reaction.

The experimental procedures for the above reactions are remarkably facile without the need to use toxic or expensive metal catalysts or reagents along with very good results; these results encouraged us to investigate the scale-up synthesis of functional derivatives of 3,3′-(phenylmethylene)bis(1H-indole) which known as anti-HIV (HIV-1 integrase) and anticancer (FCW81) (Scheme 3 and Fig. 1). With reaction stoichiometry intact, the scale of the reaction was increased to 10.0 mmol. The reaction proceeded successfully and products were obtained in 76 % and 84 %, respectively.

The efficiency of the prepared catalyst for the synthesis of bis (indolyl)methanes was compared with other similar catalyst used for this reaction. These results which are presented in Table 6 indicate good

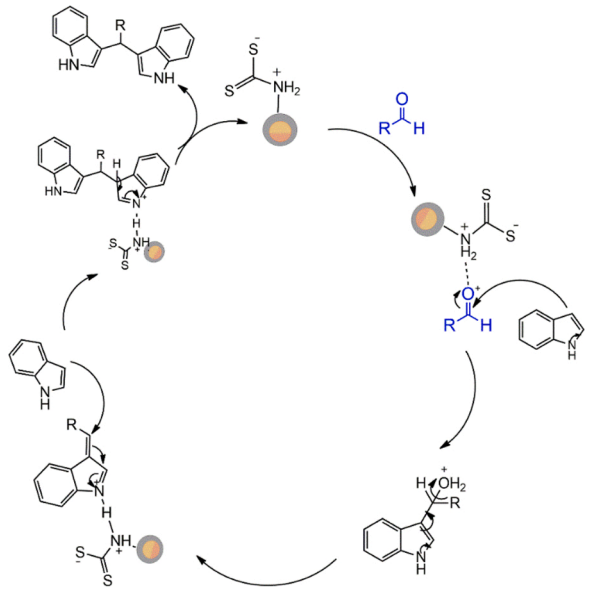
Table 6
Comparison of recent literature catalysts and this work for the synthesis of 3,3′-(phenylmethylene)bis(1*H*-indole).

Entry	Catalyst (mol%)	T (°C)	Time (h)	Yield (%)	Ref.
1	[Msim][$FeCl_4$] (5)	r.t.	1.25	98	[39]
2	MOF-891 (1) (sonication)	r.t.	1.5	92	[40]
3	[DABCO-H][HSO_4] (10)	90	2	91	[41]
4	Cu/MWCNTs-GAA@Fe2O3	70	1.3	88	[42]
5	Cu-base-Fe_2O_3 (0.5)	80	2	81	[43]
6	GO	40	3	76	[44]
7	Meglumine	r.t.	4	65	[45]
8	Our catalyst (20)	r.t.	3	94	This work

Table 7
Recyclability of catalyst in the model reaction of dihydroquinazolinones synthesis.[a].

Run	Yield[a] (%)	Run	Yield[a] (%)
1	95	5	90
2	93	6	89
3	92	7	83
4	90	8	–

[a] Isolated yield.



Scheme 4. A proposed mechanism for the synthesis of bis(indolyl)methanes.

performance of our heterogeneous catalyst in the synthesis of bis (indolyl)methanes.

One of the serious problems for homogenous catalysts as a conventional promoter in these types of synthesis is difficult separation and product contaminations [37,38]. The recyclability of the catalyst was studied using the model reaction of dihydroquinazolinones synthesis. After the complect of the reaction, the catalyst was easily separated by a magnetic bar and washed with hot ethanol, then dried and applied in the next catalytic cycle.

Until the eighth reuse, no important reduction in activity was detected. The results were given in Table 7. The reused catalyst was characterized by FT-IR analysis, the spectra are given in supplementally data.

The preparation bis(indolyl)methanes plausible mechanism was given in Scheme 4.

3. Conclusion

Herein, a green method using a recyclable catalyst has successfully been advanced for the synthesis of 2,3-dihydroquinazolin-4 (1 H) -ones and bis(indolyl)methanes in mild reaction conditions. A variety of derivatives of both reactions have been readily synthesized with excellent product yields at a short reaction time (15 min). More importantly, large-scale synthesis of four biologically active molecules using our introduced procedure have been reported with excellent yields. In general, the applied procedure was facile, mild, and environmentally friendly without any metal pollution. Finally, the heterogeneous property of the catalyst was confirmed.

CRediT authorship contribution statement

Fatemeh Mohammadi Metkazini: Design and performed the experiments. **Zahra Khorsandi:** Co-design and performed experiments, analyzed data, and writing original draft preparation. **Akbar Heydari:** Supervised the research.

Declaration of Competing Interest

The authors report no declarations of interest.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.mcat.2021.111532.

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