Research on Chemical Intermediates
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# Cascade synthesis of fused polycyclic dihydropyridines by Ni–Zn–Fe hydrotalcite (HT) immobilized on silica-coated magnetite as magnetically reusable nanocatalyst

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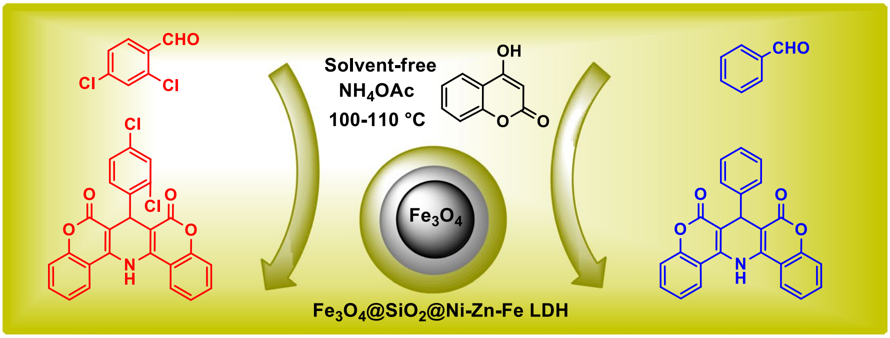
## Abstract

An efficient ecofriendly approach has been developed for one-pot multicomponent synthesis of fused polycyclic dihydropyridine (DHP) derivatives. Tandem condensation of aromatic aldehydes, 4-hydroxycoumarin, and ammonium acetate was performed using heterogeneous core–shell $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe hydrotalcite under solvent-free conditions, obtaining the corresponding products in high to excellent yield. The nanomagnetic layered double hydroxide was characterized using scanning electron microscopy coupled with energy-dispersive X-ray analysis (SEM–EDX), powder X-ray diffraction analysis, transmission electron microscopy (TEM), and the Brunauer–Emmett–Teller technique. Moreover, the recoverability of the green hydrotalcite was investigated by TEM and SEM analyses. The current research highlights the development of a green method for synthesis of DHPs without use of hazardous solvents under mild conditions.

## Graphical abstract



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**Keywords** Multicomponent reactions · Green synthesis · Nanocatalyst · $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH · Polycyclic dihydropyridines

## Introduction

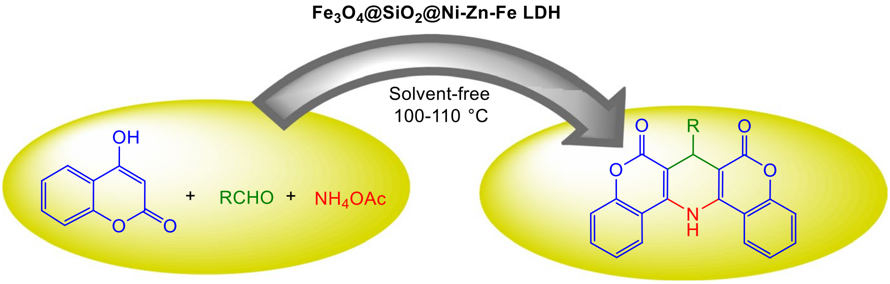
Catalytic systems have been applied in various organic transformations to improve product yields and reaction rates [1]. Homogeneous catalysts exhibit better properties in many reactions; however, the removal of the catalyst after completion of the reaction causes environmental and economic problems [2]. To overcome the problems with these types of reaction, heterogeneous catalysts can be used as efficient agents in various organic conversions [3–9]. Different heterogeneous reagents have been utilized to facilitate many reactions in organic synthesis [10–17].

Hydrotalcites (HTs) as clay minerals with anionic nature [18, 19] can be synthesized by using economic and simple procedures. However, such preparation of HTs using general methods results in powder samples with aggregated surface area, without sufficient control of morphology or particle size. The selectivity and activity of LDHs can be been improved by combining them with magnetic nanoparticles (MNPs), enabling easily separation after completion of the reaction [20–28]. Moreover, hydrotalcites offer various advantages as environmentally friendly materials, and they are simply used in many fields [29–34].

In modern processes, multicomponent reactions (MCRs) have been widely applied as a powerful tool for synthesis of organic compounds with attractive biological and pharmaceutical features [35]. These types of reaction have the ability to produce structurally complex molecules in a one-pot synthetic process without isolation of intermediates [36]. Therefore, designing new MCRs toward synthesis of a large variety of heterocycles such as polycyclic 1,4-dihydropyridine (1,4-DHP) derivatives under green conditions is essential for access to such complex heterocycle scaffolds in one pot.

As a major category of heterocyclic materials with nitrogen atoms and intriguing molecular structures, 1,4-DHPs exhibit significant pharmacological efficiency [37–40]. These types of bioactive compound exhibit diverse activities such as antimicrobial [41], anticancer [42], antitumor [43], antimalarial [44], and antidiarrheal effects [45, 46]. Many synthetic procedures have been reported for preparation of fused dihydropyridines [47–58]. However, the reported systems suffer from some limitations, including use of expensive catalysts, or hazardous and toxic solvents, as well as severe reaction conditions. Consequently, development of new, environmentally friendly methods has been considered using efficient and recoverable catalysts under green conditions for synthesis of pharmaceutical fused polycyclic dihydropyridine derivatives.

Following previous research using magnetic LDHs in multicomponent reactions [59], we present herein an efficient approach to produce DHPs by one-pot condensation of aromatic aldehydes, 4-hydroxycoumarin, and ammonium acetate using recyclable nanomagnetic LDH (Scheme 1).



**Scheme 1** One-pot synthesis of DHPs using nanomagnetic $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH

# Results and discussion

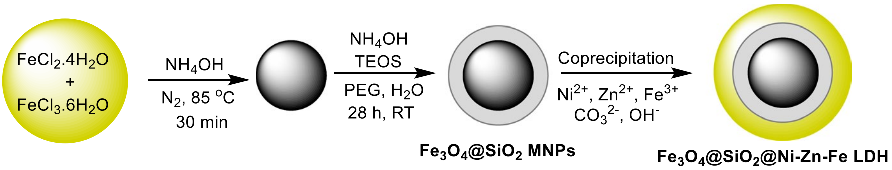
## Synthesis and characterization of catalyst

Ni–Zn–Fe LDH immobilized on silica-layered magnetite was prepared by a three-step process (Scheme 2). The green magnetic hydrotalcite was characterized by SEM, EDX, XRD, TEM, and Brunauer–Emmett–Teller (BET) analyses.

Figure 1 shows the size and morphology of the prepared $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe layered double hydroxide. The SEM images of the synthesized magnetic LDH show granules and almost spherical nanoparticles in the nanocomposite system. The images show a size distribution from 17 to 36 nm, corresponding exactly to the production of green mesoporous hydrotalcite.

Figure 2 illustrates the corresponding elements and the EDX spectrum of the prepared $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH. Energy-dispersive X-ray spectroscopy reveals the presence of Fe, O, Si, Ni, and Zn elements in the synthesized nanocatalyst.

Figure 3 shows the crystalline structure of the $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH obtained using X-ray diffraction analysis. The XRD pattern illustrates diffraction peaks at $2\theta = 11.2°$, 23.1°, 34.4°, 36.6°, 39.1°, 42.2°, 46.6°, 48.9°, 50.1°, 60.1°, and 61.2°, corresponding to (003), (006), (102), (104), (105), (0011), (0012), (109), (0013), (110), and (113) reflections of the prepared LDH with hexagonal crystal structure. The planes are in good agreement with the Inorganic Crystal Structure Database (ICSD) reference pattern 00-049-0722 for iron nickel chloride hydroxide hydrate. The characteristic features of LDH-like materials are represented by the symmetric and sharp reflections of (003), (006), (110), and (113) crystal planes [62]. The XRD pattern also shows all the diffraction peaks of $Fe_3O_4$ at $2\theta = 30.2°$, 35.5°, 43.3°, 53.7°, 57.2°, and 62.9°, related to (220), (311), (400), (422), (511),



**Scheme 2** Preparation of nanomagnetic $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH

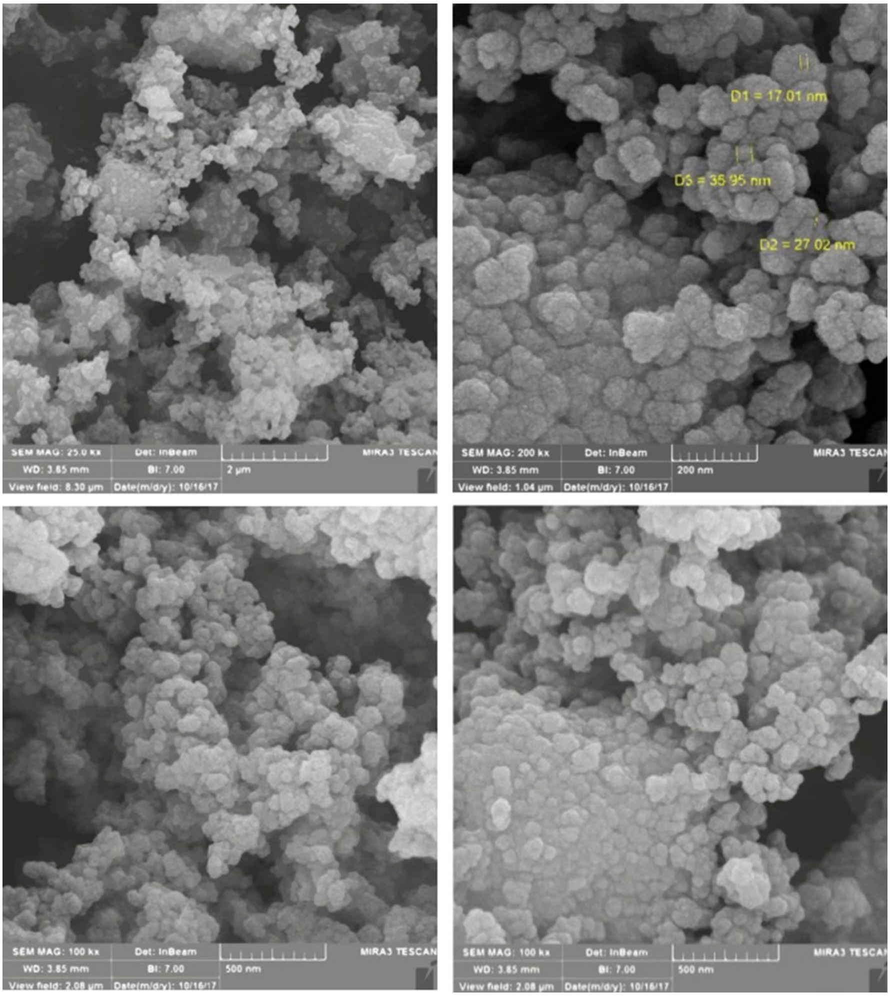


**Fig. 1** SEM images of nanomagnetic LDH

and (440) crystal planes, in agreement with the standard magnetite structure in Joint Committee on Powder Diffraction Standards (JCPDS) card no. 65-3107 [63, 64]. Furthermore, using the Debye–Scherrer equation ($D = k\lambda/\beta\cos\theta$), the mean crystalline size of $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH was calculated to be about 24.17 nm. In this equation, $\beta$ represents the full-width at half-maximum intensity (FWHM), being 0.32° (0.006 rad) at $\theta = 5.786°$.

Figure 4 presents TEM images of the prepared $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH, revealing dark nanoparticle cores surrounded by the LDH shell.

Figure 5 displays the nitrogen adsorption–desorption isotherm of the green $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH. The surface area and pore size distribution of the synthesized nanocatalyst were investigated using the BET and

| Element | Weight percent (%) | Atom percent (%) |
|---|---|---|
| **O** | 38.91 | 65.83 |
| **Si** | 11.71 | 11.29 |
| **Fe** | 20.91 | 10.14 |
| **Ni** | 20.28 | 9.35 |
| **Zn** | 8.19 | 3.39 |

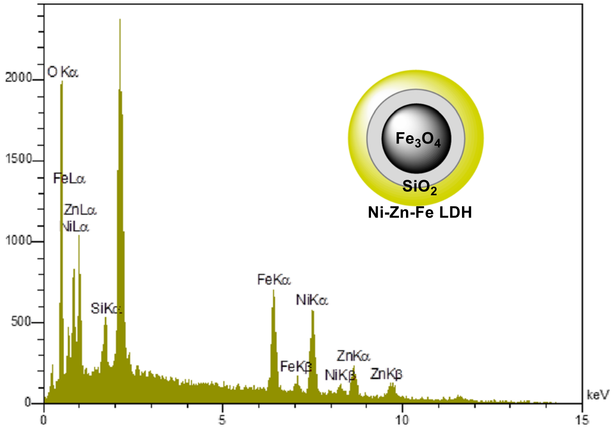


**Fig. 2** Elemental composition and EDX spectrum of nanomagnetic LDH

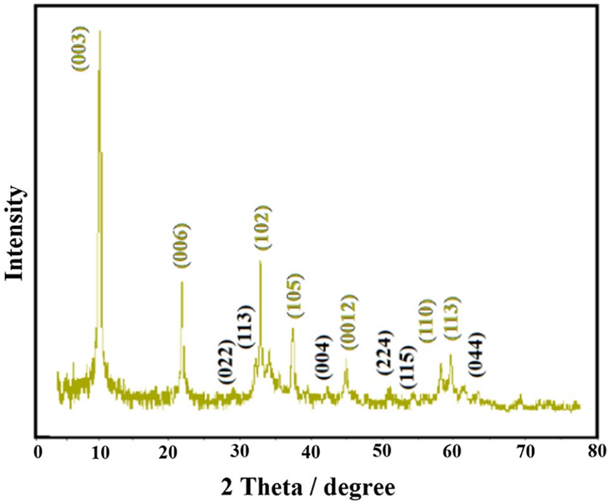


**Fig. 3** XRD pattern of nanomagnetic LDH

Barrett–Joyner–Halenda (BJH) methods. The calculated BET specific surface area ($S_{BET}$) and pore volume ($V_p$) were 90.716 $m^2\ g^{-1}$ and 0.3187 $cm^3\ g^{-1}$, respectively. The activity and selectivity of the nanocatalyst are increased due to its high surface-to-volume ratio. The average pore size of the green LDH was determined

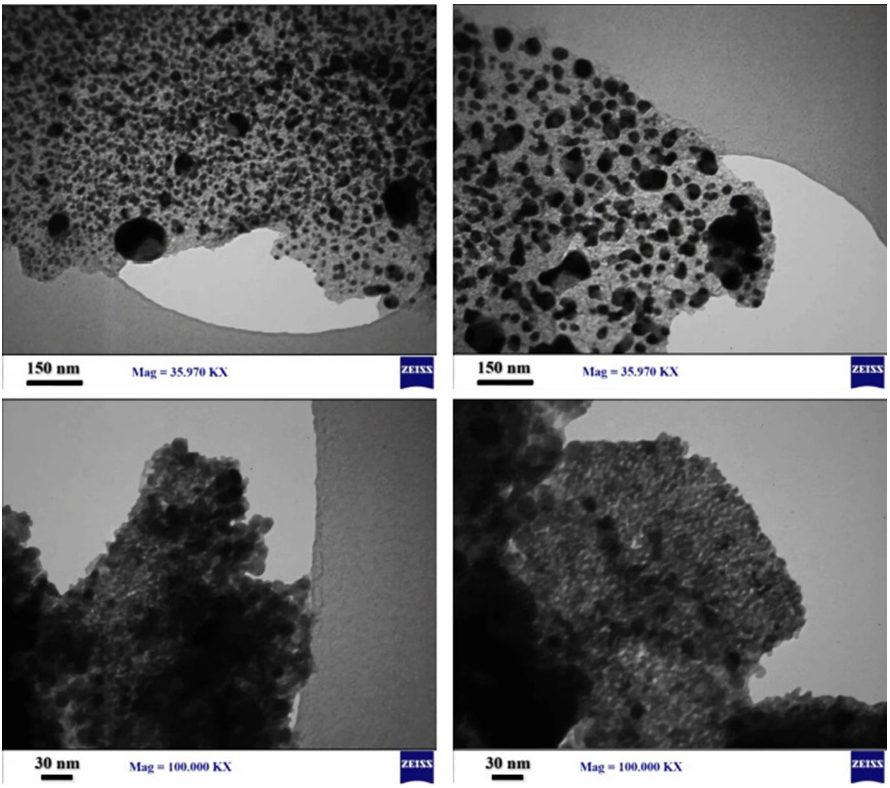
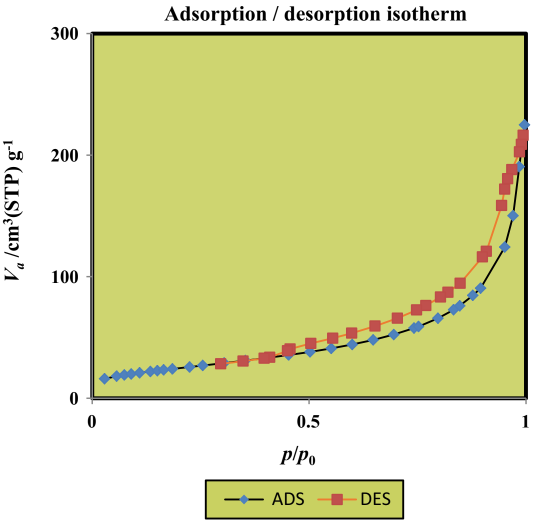


**Fig. 4** TEM images of nanomagnetic LDH

**Fig. 5** Nitrogen adsorption–desorption isotherm of nanomagnetic LDH



to be 14.054 nm using the Barrett–Joyner–Halenda (BJH) method, corresponding to the size range of mesoporous materials.

## Synthesis of fused polycyclic dihydropyridines

Polycyclic dihydropyridine derivatives have great pharmaceutical properties and exhibit a wide variety of biological activities. In the present research, an efficient green method was developed to prepare DHPs in accordance with the importance of this category of compounds. Domino condensation of benzaldehyde (0.5 mmol), 4-hydroxycoumarin (1 mmol), and ammonium acetate (3 mmol) was selected as model reaction. At the beginning of the research, the model reaction was heated without catalyst; after 2 h, only trace amount of product was formed. In the next step, the effect of $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH on the model reaction was studied; the desired product formed quickly when using the magnetic nanocatalyst. The effects of diverse temperatures, solvents, and amounts of catalyst were evaluated to optimize the conditions. The effect of temperature was also investigated using the model reaction at gradually increasing temperatures from room temperature to 110 °C. The results indicated that increase of the reaction temperature enhanced the performance of the reaction, with the maximum yield being obtained in the range of 100–110 °C (Fig. 6). The effect of different solvents on the model reaction was then studied. The findings confirmed that the best result was obtained without solvent (Fig. 7). Also, the effect of the amount of catalyst on the model reaction was studied to determine the exact concentration of catalyst (Fig. 8). The amount of catalyst was increased from 0 to 50 mg, with continuous growth in yield being observed. However, increasing the catalyst content to 60 mg did not show any further improvement in the performance of the reaction. Therefore, 50 mg of catalyst was determined to be optimum amount with the best activity. The results showed that use of 50 mg of $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH was sufficient to achieve condensation between 4-hydroxycoumarin (1 mmol), benzaldehyde (0.5 mmol), and ammonium acetate (3 mmol) under solvent-free conditions (100–110 °C) within 10 min (Table 1, entry 1).

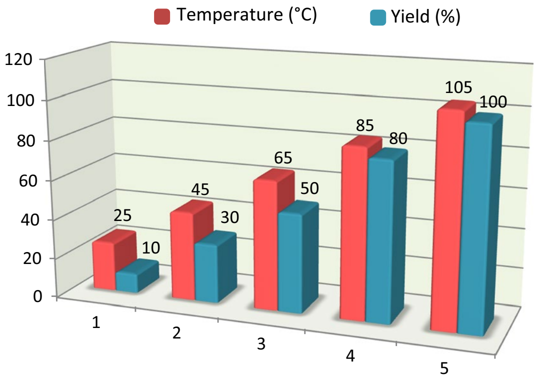


**Fig. 6** Effect of temperature on model reaction

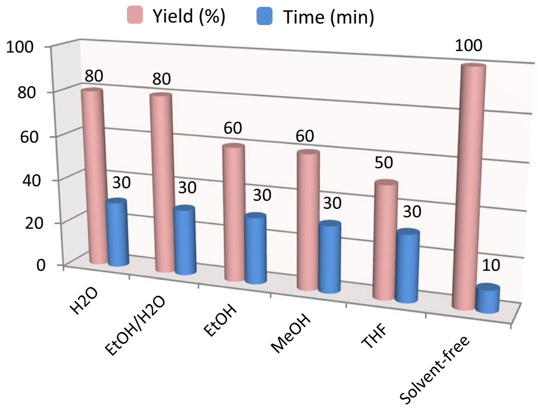


**Fig. 7** Effect of solvent on model reaction

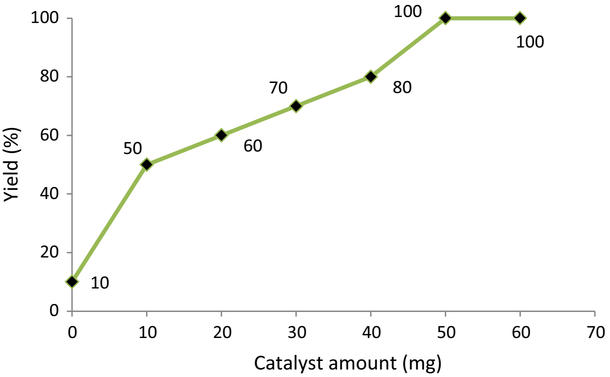


**Fig. 8** Effect of amount of catalyst on model reaction

To investigate the scope and generality of the green $Fe_3O_4@SiO_2@Ni–Zn–Fe$ LDH, the multicomponent condensation was further studied in the reaction of structurally diverse aromatic aldehydes, 4-hydroxycoumarin, and ammonium acetate to produce corresponding polycyclic dihydropyridine derivatives under the optimized conditions. The summarized results in Table 1 show that all the condensations were carried out successfully within 10–40 min to afford the products in yield of 75–88 %. The results in this table also show that the products are similar to those reported in literature in terms of their spectral and physical data.

Although the precise mechanism of this synthetic method is not clear, we propose the following plausible mechanistic pathway for formation of fused polycyclic dihydropyridines by multicomponent condensation of aromatic aldehydes, ammonium acetate, and 4-hydroxycoumarin (Scheme 3). First, all the substrates are activated by active sites of the nanocatalyst. Second, aromatic aldehyde is attacked by activated methylene group to form C–C bond, then dehydration of intermediate **1** occurs instantly to obtain the arylidene derivative. On the other hand, an additional molecule of 4-hydroxycoumarin reacts with ammonium acetate as a source of ammonia

**Table 1** One-pot synthesis of fused polycyclic dihydropyridines using nanomagnetic $Fe_3O_4@SiO_2@Ni–Zn–Fe$ LDH[a]

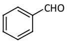
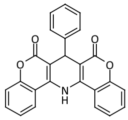
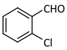
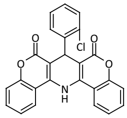
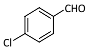
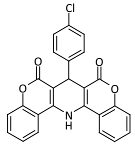
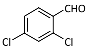
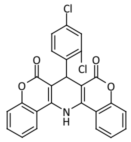
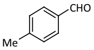
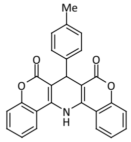
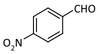
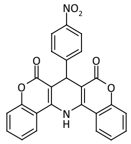
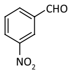
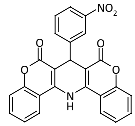
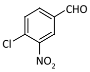
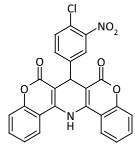
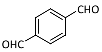
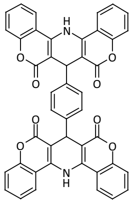
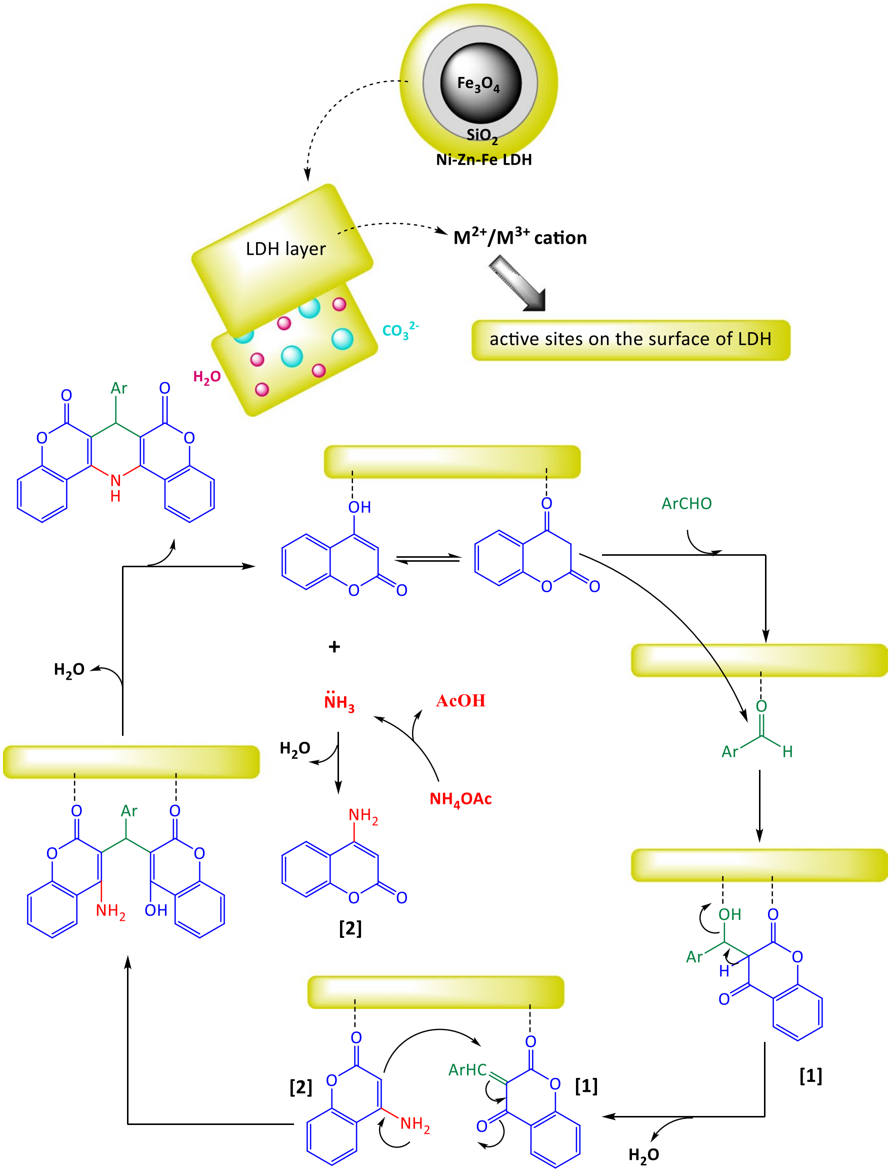
| Entry | Substrate | Product | Time (min) | Yield (%)[b] | Mp (°C) | |
|---|---|---|---|---|---|---|
| | | | | | Found | Reported [Ref.] |
| 1 |  |  | 10 | 85 | 208-210 | 209 [58] |
| 2 |  |  | 13 | 85 | 275-277 | 275-278 [57] |
| 3 |  |  | 15 | 87 | 274-276 | 275-276 [56] |
| 4 |  |  | 10 | 88 | 221-223 | - |
| 5 |  |  | 20 | 87 | 257-259 | 258 [56] |
| 6 |  |  | 18 | 81 | 240-242 | 242-244 [56] |
| 7 |  |  | 10 | 88 | 217-219 | 216-218 [37] |
| 8 |  |  | 15 | 75 | 203-205 | - |
| 9 |  |  | 40 | 75 | >300 | - |

**Table 1** (continued)

[a] All reactions carried out with 4-hydroxycoumarin (1 mmol), aromatic aldehyde (0.5 mmol), ammonium acetate (3 mmol), and $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH (50 mg) under solvent-free conditions in oil bath (100–110 °C)

[b] Yields refer to isolated pure products



**Scheme 3** Plausible mechanism for synthesis of fused polycyclic dihydropyridines using nano-$Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH

to produce β-aminobenzopyran-2-one **2**. Subsequently, the two intermediates **1** and **2** react via Michael-type addition. Finally, the fused polycyclic dihydropyridine derivative is obtained after dehydration of the product.

## Recycling of $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH

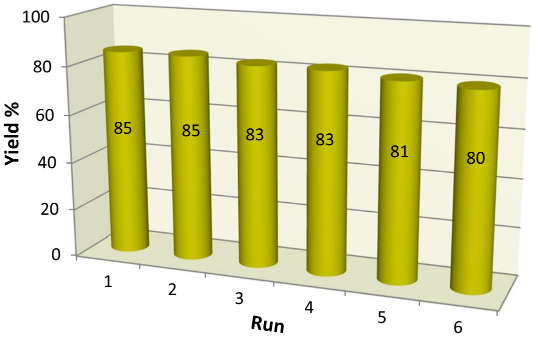
The economical attractiveness of this approach is the ability to recover the green nanomagnetic $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH at the end of the reaction. The reusability of LDH was thus investigated in the condensation of benzaldehyde, ammonium acetate, and 4-hydroxycoumarin at the optimized conditions. After reaction completion, hot ethanol was added to the mixture, and the catalyst was separated magnetically, washed with diethyl ether, and dried for reuse in the next run. The results indicated that the nanocatalyst could be recovered several times without significant loss of activity (Fig. 9).

Further studies on the recycled nanocatalyst were performed by SEM (Fig. 10) and TEM (Fig. 11) analyses. The images revealed that the structure of the recovered catalyst was not altered but remained intact.

## Experimental

Chemicals and materials were purchased from Merck and Fluka Companies. The FT-IR spectrum was obtained using a Thermo-Nicolet-Nexus 670 spectrophotometer. $^1$H nuclear magnetic resonance (NMR) spectra were measured on a Bruker Avance spectrometer (300 MHz). Reaction progress was determined using thin-layer chromatography (TLC). The morphology of the nanoparticles was observed by field-emission scanning electron microscopy (FESEM, TESCAN). X-ray diffraction (XRD) patterns were obtained using a PANalytical X'PertPRO diffractometer with Cu $K_\alpha$ radiation in the scan range between 5° and 80°. Transmission electron microscopy (TEM) images were captured on a Zeiss EM10C microscope at accelerating voltage of 100 kV in each case. Nitrogen adsorption–desorption isotherms were measured using a BELsorp-mini (BEL, Japan). The specific surface area, pore volume, and pore size distribution were obtained using the Brunauer–Emmett–Teller (BET) and Barrett–Joyner–Halenda (BJH) methods.

**Fig. 9** Catalyst reusability in the model reaction



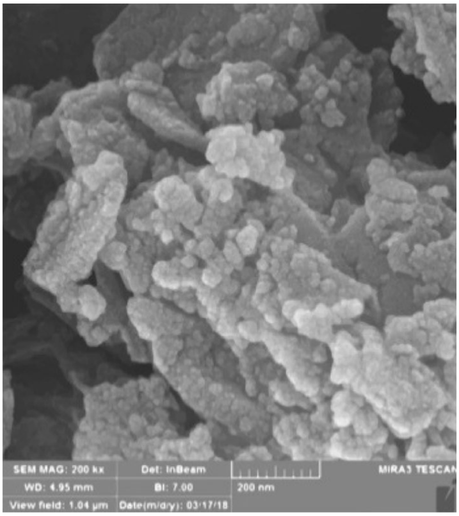


**Fig. 10** SEM image of nanomagnetic LDH after second recovery

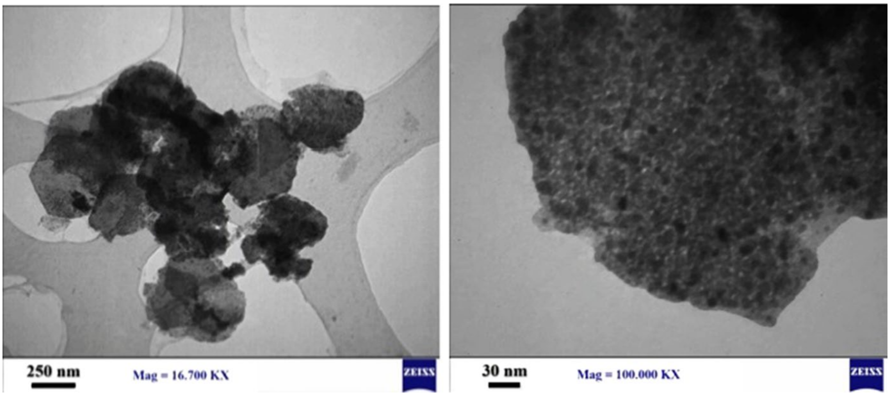


**Fig. 11** TEM images of nanomagnetic LDH after second recovery

### Synthesis of magnetite ($Fe_3O_4$) MNPs

Nanomagnetite was prepared by chemical coprecipitation of $Fe^{3+}$ and $Fe^{2+}$ chloride salts [60]. Generally, $FeCl_3{\cdot}6H_2O$ (5.838 g, 0.0216 mol) and $FeCl_2{\cdot}4H_2O$ (2.147 g, 0.0108 mol) were added to $H_2O$ (100 mL). The reaction content was mixed magnetically in $N_2$ atmosphere at 85 °C. After stirring for about 10 min, 10 mL aqueous ammonia (25 wt%) was added, and black precipitate formed immediately. The mixture was kept at 85 °C under stirring for 30 min in $N_2$ atmosphere, then the resulting mixture was placed at ambient temperature. The obtained $Fe_3O_4$ was cooled down, separated, and washed three times using 0.02 M NaCl solution and distilled water.

### Preparation of silica-layered magnetite ($Fe_3O_4$@$SiO_2$) MNPs

$Fe_3O_4$@$SiO_2$ was synthesized using a previously reported method [61]. Distilled water (20 mL) was poured into 1.5 g magnetite, and 2-propanol (200 mL) was added. Next, the mixture was homogenized using ultrasound for about 30 min, then 5.36 g polyethylene glycol (PEG), 20 mL $H_2O$, 10 mL $NH_4OH$ (28 wt%), and 2 mL tetraethylorthosilicate (TEOS) were added to the suspension. The mixture was kept at ambient temperature under stirring for 28 h. The prepared $Fe_3O_4$@$SiO_2$ was magnetically separated and washed with water and ethanol.

### Preparation of Ni–Zn–Fe LDH immobilized on silica-layered magnetite ($Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH)

$Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH was prepared as follows: 0.25 g $Fe_3O_4$@$SiO_2$, 1.060 g $Na_2CO_3$ (0.01 mol), and 0.160 g NaOH (0.004 mol) were added to $H_2O$ (30 mL) (solution A). Solution B was prepared from 1.352 g $FeCl_3{\cdot}6H_2O$ (0.005 mol), 2.617 g $Ni(NO_3)_2{\cdot}6H_2O$ (0.009 mol), and 1.785 g $Zn(NO_3)_2{\cdot}6H_2O$ (0.006 mol) in $H_2O$ (30 mL). Solution A and B were exposed to ultrasound for 30 min. The solutions were then added simultaneously to distilled water (30 mL) under electromagnetic stirring, and the pH value was adjusted to 10 by addition of HCl and NaOH solutions. After addition, the slurry was fiercely stirred at ambient temperature (30 min), then the suspension was left for 20 h at 80 °C. Finally, the resulting mixture was filtrated and the precipitate was dried (150 °C) to obtain nanomagnetic $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH.

### General procedure for multicomponent condensation of benzaldehyde, ammonium acetate, and 4-hydroxycoumarin using $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH

In a test tube equipped with a magnetic stirrer, a mixture of benzaldehyde (0.053 g, 0.5 mmol), $NH_4OAc$ (0.231 g, 3 mmol), and 4-hydroxycoumarin (0.162 g, 1 mmol) was prepared. Magnetic $Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH (50 mg) was added, and the resulting mixture was stirred at 100–110 °C for 10 min under solvent-free conditions. Reaction progress was monitored by TLC (*n*-hexane:EtOAc at 4:2 as eluent). On reaction completion, hot ethanol (5 mL) was poured into the mixture, and the nanocatalyst was separated magnetically. The precipitate was collected after evaporation of solvent, and the pure product was obtained by recrystallization from ethanol (entry 1, Table 1, 85 %).

## Conclusions

An extremely efficient method for tandem synthesis of fused polycyclic dihydropyridine derivatives via multicomponent condensation of aromatic aldehydes, ammonium acetate, and 4-hydroxycoumarin using nano-Ni–Zn–Fe LDH immobilized on silica-layered magnetite ($Fe_3O_4$@$SiO_2$@Ni–Zn–Fe LDH) was developed. All reactions were performed under solvent-free condition within 10–40 min. The prepared nanomagnetic LDH was characterized using SEM, EDX, XRD, TEM, and BET analyses. The attractive features of the current protocol include short reaction time, absence of hazardous solvents, high catalytic efficiency, and the ability to recover the green nanomagnetic LDH.

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