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RESEARCH ARTICLE

THREE COMPONENT HYDROTALCITE CATALYZED SYNTHESIS OF DIHYDROPYRIDINE

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ABSTRACT

Hantzsch synthesis of various substituted 1,4-dihydropyridine by Mg/Fe hydrotalcite catalyzed one-pot condensation reaction of aromatic aldehyde, α -dicarbonyl compound and ammonium carbonate at 55^oC. The products were isolated in good yields with short reaction time. Hydrotalcite are non-toxic, easy to separate and recyclable catalyst. Hence, this is process environment friendly. This work provides a simple, clean, rapid, an inexpensive and readily available catalyst and easy work up.

Key words:

Hantzsch synthesis, one-pot condensation, hydrotalcite, Aromatic aldehyde, 1,4-dihydropyridine.

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INTRODUCTION

The multicomponent one-pot condensation reaction become an important tool in the organic synthesis as they possess great ability of building up the complex and highly functionalized organic molecule with simplicity[1]. The search of cleaner, green, efficient organic synthesis process is still sustained by synthetic organic chemist. Furthermore, MCR exhibits higher atom economy and selectivity underlying such reaction, consuming less energy, time saving as well as environmental friendliness have led to significant effort to develop and improvement MCR in both industry and academia [2-5].

1,4-DHP are class of nitrogen containing heterocycles having six membered ring which are of important in biological systems As a class of useful drugs particularly as anti-oxidant. Some of the representative compound of this class possess insecticidal, bactericidal, herbicidal and acaricidal activities[6]. 1,4,-DHP compound play important role in medicinal chemistry. DHP drugs namely amlodipine, nifedipine, nicardipine and felodipine are cardiovascular agents for the treatment of hypertension [7,8.] Furthermore, DHP have received much attention in stereospecific hydrogen transfer reaction, stereospecific reduction of phenylglyoxylic and

pyruvic acid using DHP to biomimic model of lactase dehydrogenase have been reported by krechi and samrckova [9].

Moreover DHP, which are most potent calcium antagonist have been used for treatment of congestive heart failure [10]. Nowadays, various asymmetric reaction such as hydrogenation of quinolines in the synthesis of alkaloids [13], asymmetric reductive amination of aldehyde [14], Hydrogenation of α , β -unsaturated aldehyde and ketones [11,12.] are catalyzed by DHPs as organocatalyst. These unique features of DHP encourage researchers to develop new protocol for synthesis.

1,4-DHP are generally synthesized by Hantzsch reaction which involves the condensation of aldehyde, α -ketoester, and ammonia or ammonium acetate in acetic acid or refluxing alcohol for long time [15]. A number of other methods are reported for condensation, which involves Yb(OTf)₃[16], pyridine[17], silica gel/NaHSO₄[18], Molecular Iodine[19], Hy-Zeolite [20], Cynuric Chloride [21], p-TSA[22], Ionic Liquid [23], TMSCl-NaI [24], The use of microwave[25]. Conventional Bronsted acids such as sulphuric, nitric, hydrochloric and hydrofluoric acids often used in organic synthesis and industrial process. On the other hand, most of the existing methods for the synthesis of 1,4-DHP suffers from

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drawbacks such as longer reaction time, low yield, occurrence of several side reaction products, used of stoichiometric amount of reagents, high temperature, the use of expensive and toxic transition metallic reagents difficult to handle and it's tedious work up. Therefore, exploring the new catalyst system preferably in an environmentally benign method to overcome these drawbacks is a challenging task to organic synthesis.

In order to develop green protocol of synthesis, heterogeneous catalysts received much attention due to their easy recovery and subsequent reuse as well as design novel, efficient and environmentally benign process using various heterogeneous catalyst [26-28].

HT offers strong surface basicity and applied as catalyst for some selective oxidation reactions. HT and HT like material have been used for the bayer-viilliger reaction using peracids [29], oxidation of alcohols [30], epoxidation of olefins and - unsaturated ketones using hydrogen peroxides [31,32.]

In continuation of our effort to develop efficient environmentally benign protocols for the synthesis of various heterocycles. We report herein our findings with Mg/Fe HT that efficiently catalyzed the multicomponent synthesis.

Experimental

Melting points of all synthesized compounds were measured on electro thermal apparatus using open capillary tubes and are uncorrected. TLC for purity of compounds were performed on silica gel coated aluminum plate as adsorbent and which are analysed with U.V. light as visualizing agent. FT-IR Spectra were recorded on Bruker Spectrometer in the region of 400-4000 cm^{-1} . ^1H and ^{13}C NMR spectra were recorded on Varian 500 MHz NMR spectrophotometer using TMS as an internal standard and $\text{CDCl}_3/\text{DMSO-d}_6$ as solvent (chemical shifts in ppm). Powder X-ray diffraction pattern were collected with monochromatic Cu K radiation ($\lambda = 1.54059 \text{ \AA}$) at 40 kV and 15 mA using Shimadzu 7000S diffractometer. Thermogravimetric analysis were performed with a RIGAKU Thermo Plus TG 8120 thermo balance with a heating rate of $10^\circ\text{C}/\text{min}$ from 25 to 900°C . The morphological information gathered using scanning electron microscope ZEISS Ultra FESEM.

RESULTS AND DISCUSSION

In continuation of our programmes on the chemistry of 1,4,-DHP. Herein we report an efficient and environmental benign one pot procedure for the catalytic synthesis of 1,4-DHP from aromatic aldehyde, ethylacetacetate and ammonium carbonate using Mg/Fe Hydrotalcite, a heterogeneous base catalyst. The HT's are synthetic clays which contain brucite layer structure. Mg/Fe Hydrotalcite of Mg/Fe mole ratio 2:1,3:1,4:1,5:1. were prepared by procedure reported in literature[33] and were characterized by XRD, FTIR and TGA.

From XRD spectrum shown in fig 1, it was seen that calcined hydrotalcite with Mg/Fe=3:1 molar ratio (fig 1) shows peaks at $2\theta = 43.08^\circ, 62.59^\circ$ which are corresponding to MgO and at $2\theta = 30.37^\circ, 35.50^\circ, 43.08^\circ$ and 62.59° which can be attributed to MgFe_2O_4 spinel structure (JCPDS 17-0465) those peak have been observed in literature.[34,35.]

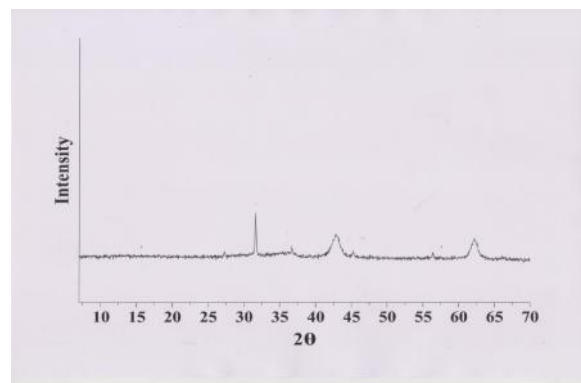


Fig 1 XRD spectrum for hydrotalcite with Mg/Fe=3:1 calcined at 500°C .

The FTIR spectra of LDH with Mg/Fe=3:1 molar ratio as shown in (fig 2&3.) are typical of LDH and which are in agreement well with the typical results previously report[34-40.]. The broad band observed at 3442cm^{-1} is attributed to interlayer water molecules, this band become weaker and is shifted to 3409cm^{-1} when hydrotalcite calcined. The strong band at 1355cm^{-1} is due to the mode ν_3 of the interlayer carbonate species as reported in the literature. The bands in the range of $500-750\text{cm}^{-1}$ are attributed to metal-oxygen-metal bond stretching.

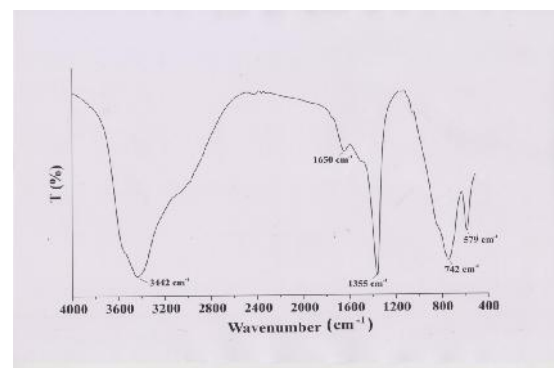


Fig 2 FTIR spectra of LDH with uncalcined Mg/Fe=3:1.

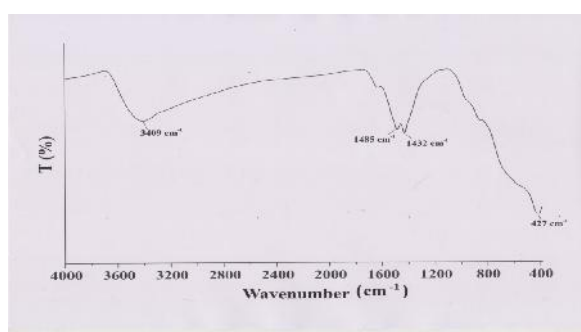


Fig 3 FTIR spectra of LDH with Mg/Fe=3:1 calcined at 500°C .

The TGA Plot of LDH having Mg/Fe molar ratio 3:1 shows three distinct phase loss in the range $50-200^\circ\text{C}$, $200-400^\circ\text{C}$ and $460-750^\circ\text{C}$ (fig 4). The first weight loss in temp range of $50-200^\circ\text{C}$ which was about 13%. This weight loss of hydrotalcite mainly due to interlayer and physisorbed water. Further weight loss of 21% which occurs between $200-460^\circ\text{C}$ which is

related to removal of carbonate ions from the interlayer of hydrotalcite and first step dehydroxylation. final the third mass loss, that occurs further than 460°C can due to continuous dehydroxylation and decarbonation and formation of oxide metals as MgO which are detected in X-ray differ action of calcined Idh and possibly MgFe₂O₄. as reported in the literature[34,38.]. Beyond 600°C temp, there was no significant mass loss was observed.

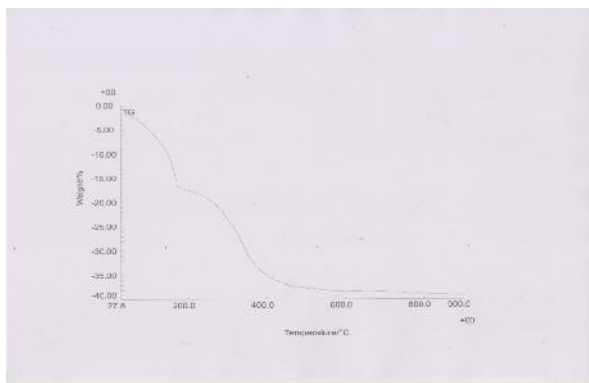


Fig 4 TGA Plot of hydrotalcite with Mg/Fe=3:1.

Catalyst morphologies as indicated by the SEM images of Mg-Fe-Ht-3 shown in fig 5, showed the materials to be clearly point out the homogeneity in shapes for the sample and high crystallinity.

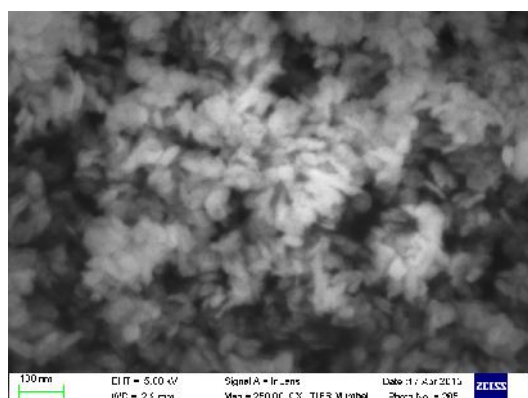
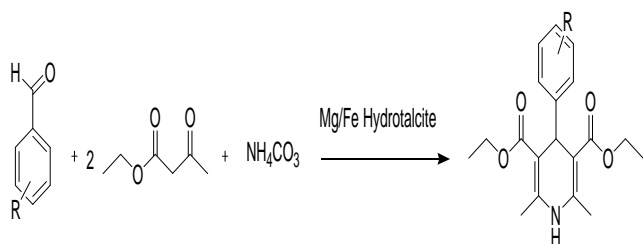


Fig 5 SEM images of hydrotalcite with Mg/Fe=3:1 calcined at 500°C.

General scheme



Scheme 1 Synthesis of 1, 4-DHP.

The reaction pathway for this reaction is mechanistically complex. There could be two possible reaction routes to obtain DHP

1. The reaction may go through condensation of two molecules of ethyl acetoactate with ammonium carbonate forming imine which subsequently undergoes condensation reaction with benzaldehyde to produce DHP.

2. The reaction may go through aldol condensation of ethyl acetoacetate with benzaldehyde and subsequent reaction of aldol with enamine which is obtained from condensation of one molecule of ethyl acetoacetate with ammonium carbonate.
3. In order to find out correct pathway, we have carried out independent experiments and unravel the following observation.
 1. Reaction between equivalent of benzaldehyde with ethyl acetoacetate did not results in aldol condensation product under our reported reaction condition.
 2. Reaction of ethyl acetoacetate with ammonium carbonate under our reported reaction conditions resulted in an intermediate product and addition of benzaldehyde to this intermediate resulted in spontaneous formation of DHP.

Based on above experiment, a probable reaction pathway for the DHP synthesis over Mg/Fe hydrotalcite is given in (scheme 1). Initially, proton abstraction from active methylene group of EAA followed by addition to protonated carbonyl group subsequent dehydration results in ethyl-3-aminobut-2-enoate. This moiety undergo condensation with another molecule of EAA to form imine. This imine will form an enamine on base catalyzed tautomerisation with benzaldehyde results in DHP.

A systematic study was carried out to optimize the reaction condition including the quantity of catalyst, reaction medium and nature of catalyst. The reaction between aromatic aldehyde, ethyl acetoacetate and ammonium carbonate a three component reaction as a model reaction.

Initially, this three component reaction was carried out in water as a medium in presence of catalytic amount of Mg/Fe Hydrotalcite at room temperature. But reaction could not proceed for long time. In ordered to run the reaction, we raised the temperature gradually upto 60°C. It was found that from the TLC. Observation, the intermediate form converted into desired product. On increasing temperature beyond 60°C the yield of products remains same.

The reaction of benzaldehyde, ammonium Carbonate, and ethyl acetoacetate in the presence of C-Mg-Fe HT-3 in different solvents at 55°C temperature.

Table 1 Solvent Selection

Entry	Solvent	Yield of product (%)
1	H ₂ O	53
2	EtOH(2 drop of H ₂ O)	76
3	MeOH(2 drop of H ₂ O)	62
4	DMF(2 drop of H ₂ O)	38
5	DMSO(2 drop of H ₂ O)	40
6	Without solvent	Trace

Reaction conditions: Substituted Benzaldehyde (3mmol), Ammonium Carbonate (3mmol), ethyl acetoacetate(6mmol), solvent (5ml), C-Mg-Fe HT-3 (0.3g), temperature (55°C).

Then we examined the effect of solvents along with 2 drop of water to dissolved the ammonium carbonate in reaction mixture (table 1). The results indicate that solvents affected the

efficiency of reaction. Yields were poor in DMF, DMSO and Water. The best results were obtained in ethanol. However, when the reaction run under solvent free condition traces of product was obtained.

The reaction was carried out using different calcined HT's (Mg/Fe-2:1, 3:1, 4:1, 5:1) at 55°C (table 2). The calcined Mg-Fe-HT-3 give good yields of the product. On calcinations at 500°C for 5 hours, the Lewis basicity of HT increases while the Brønsted basicity decreases. When the reaction was carried out in absence of catalyst reported yield was 60% and there was recovery of starting reacting materials. The catalytic activity of calcined Mg-Fe-HT-3 was found to be significantly better.

The reaction of benzaldehyde, ammonium carbonate, and ethyl acetoacetate in the presence of different C-Mg-Fe HTs in EtOH (2 drop of H₂O) at 55°C temperature.

Table 2 Catalyst Selection

Entry	Hydrotalcite	Yield of product (%)
1	C-Mg-Fe HT-2	45
2	C-Mg-Fe HT-3	76
3	C-Mg-Fe HT-4	33
4	C-Mg-Fe HT-5	58
5	Without HT	60

Reaction conditions: Substituted Benzaldehyde (3mmol), ammonium carbonate (3mmol), ethyl acetoacetate (6mmol), EtOH (2 drop of H₂O) (5ml), catalyst (0.3g), temperature (55°C).

In order to optimize the quantity of the catalyst, the reaction was attempted using different quantity of catalyst (table 3). Calcined Mg-Fe-HT-3, 0.2 gm of quantity was found to be optimal quantity. Increasing the quantity of calcined Mg-Fe-HT-3 beyond 0.2gm did not show further increase in the yield. However, in order to reveal the utility of the catalyst, its reusability was tested using the same catalyst. It was easily separated from the reaction mixture by simple filtration. The catalyst could be reused there was no significant change in the yield of products observed.

The reaction of benzaldehyde, ammonium carbonate, and ethyl acetoacetate using different quantity of C-Mg-Fe-HT-3 in EtOH at ambient temperature.

Table 3 Catalyst quantity

Entry	Catalyst quantity (g)	Yield of product (%)
1	0.10	51
2	0.15	63
3	0.20	76
4	0.25	76
5	0.30	76

Reaction conditions Substituted Benzaldehyde (3mmol), ammonium carbonate (3mmol), ethyl acetoacetate (6mmol), EtOH (2 drop of H₂O) (5ml), temperature (55°C). Under the above optimized condition, different substituted aromatic aldehyde, ethyl acetoacetate or malononitrile and ammonium carbonate are reacted to obtain 1,4-DHP's in excellent yields.

The aromatic aldehyde containing electron withdrawing group at ortho, meta and para position reacted as expected and gave the expected product in excellent yields as compared to the aromatic aldehyde containing electron donating group (Table 4).

Catalyst preparation

Mg-Fe-HTs with different Mg/Fe molar ratio (Mg/Fe = 2:1, 3:1, 4:1 and 5:1) were synthesized by co-precipitation method [38]. An aqueous solution of Mg(NO₃)₂·6H₂O and Fe(NO₃)₂·9H₂O were prepared and mixed aqueous solution of Mg(NO₃)₂·6H₂O and Fe(NO₃)₂·9H₂O was added drop wise using addition funnel to an aqueous solution containing NaOH and Na₂CO₃ under vigorous stirring. After complete addition the solution was heated at 80°C for 18hr and maintain pH of solution in range of 10-11 during stirring. After complete stirring, the solution was allowed to cool at room temp and filtered. The obtained residue was washed with hot deionised water several times till filtrate was neutral. The solid was dried in an oven at 60°C in air. Dry solid then calcined at 500°C for 5hrs.

Experimental procedures

In a 50 ml round bottom flask, ethyl acetoacetate (6mmol), ammonium carbonate (3mmol) and benzaldehyde (3mmol) were stirred in presence of Mg/Fe Hydrotalcite (0.2g) in solvent ethanol along with 2-3 drops of water in order to dissolve ammonium carbonate at 55°C for required time. The progress of reaction was monitored by TLC. After completion of reaction, the reaction mixture was heated in order to dissolve solid product form and filtered. Catalyst wash with hot ethanol. Filtrate concentrate and solid product obtained was filtered and recrystallised from hot ethanol. Purified product characterized by M.P., N.M.R. and I.R.

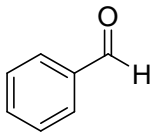
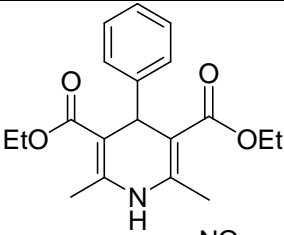
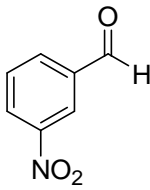
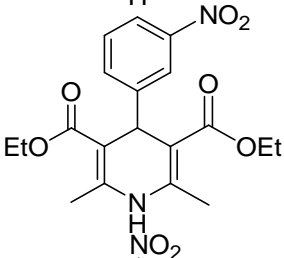
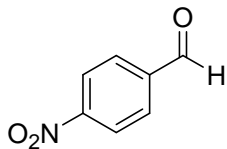
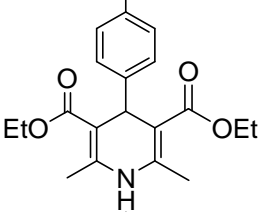
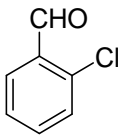
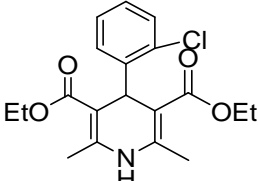
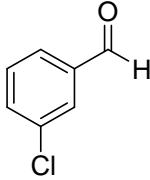
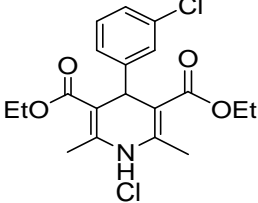
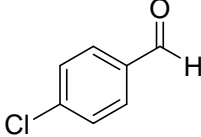
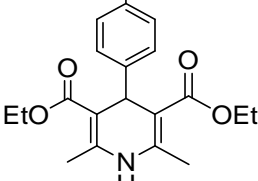
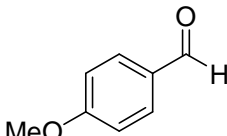
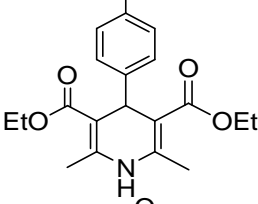
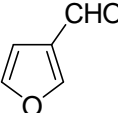
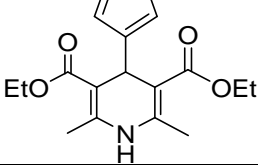
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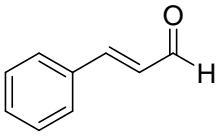
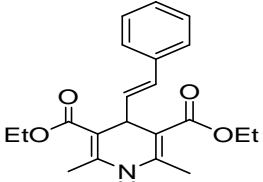
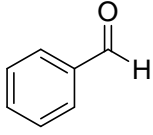
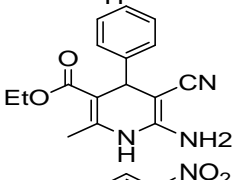
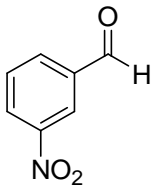
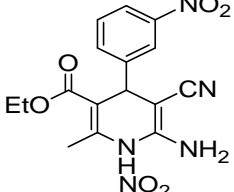
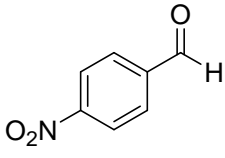
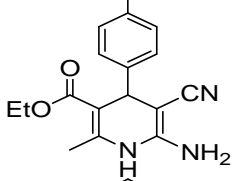
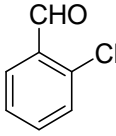
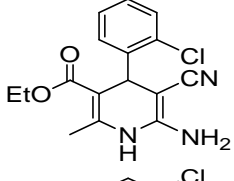
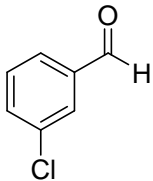
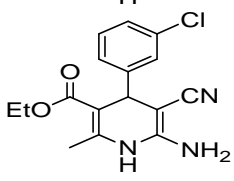
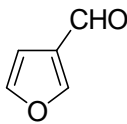
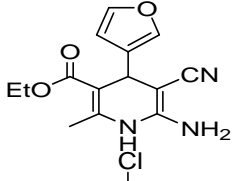
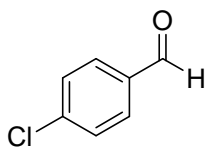
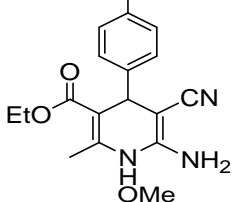
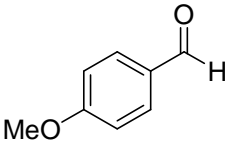
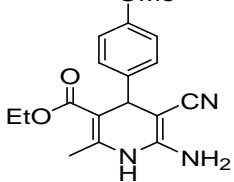
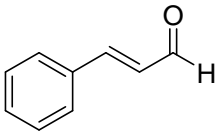
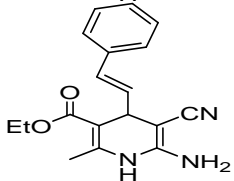
Yellow colored crystalline solid, IR (KBr, cm⁻¹): 3348 (N-H), 3038 (Ar-H), 2957 (CH₃), 1749 (C=O, ester), ¹H-NMR (500 MHz, DMSO-d₆, / ppm): 1.333 (t, 6H, 2xCH₃), 2.509 (s, 6H, 2xCH₃), 3.990 (q, 4H, 2xCH₂), 4.862 (s, 1H, CH), 7.086-7.216 (m, 5H, Ph), 8.794 (s, 1H, NH). ¹³C-NMR (500MHz, DMSO-d₆, ppm): 14.589 (CH₃), 18.636 (CH₃), 39.690 (CH₂), 59.384 (CH), 102.282-148.595 (2xC=C, ArC), 167.372 (C=O). MS m/z: 329 (M+).

2,6-Dimethyl-4-(3-nitro-phenyl)-1,4-dihydro-pyridine-3,5-dicarboxylic acid diethyl ester (Sr.No:2)

Yellow colored crystalline solid, IR (KBr, cm⁻¹): 3343 (N-H), 3098 (Ar-H), 2985 (CH₃), 1703 (C=O, ester), 1523 and 1346 (C-NO₂), ¹H-NMR (500 MHz, DMSO-d₆, / ppm): 1.135 (t, 6H, 2xCH₃), 2.510 (s, 6H, 2xCH₃), 3.990 (q, 4H, 2xCH₂), 4.848 (s, 1H, CH), 7.153-7.227 (m, 4H, Ph), 8.848 (s, 1H, NH). ¹³C-NMR (500MHz, DMSO-d₆, ppm): 14.147 (CH₃), 18.228 (CH₃), 39.089 (CH₂), 59.195 (CH), 101.049-147.393 (2xC=C, ArC), 166.487 (C=O).

Table 4 Synthesis of Dihydropyrimidine via multicomponent reaction using C-Mg-Fe HT-3 at 55°C.

Sr No	Substituted benzaldehyde	Product	Yield (%)	Time (min)	Melting Point (°C)	Melting point Reported (°C)
1			76	30	158-160	156-158 ⁴¹
2			77	25	162-164	161-163 ⁴¹
3			80	20	129-131	128-129 ⁴¹
4			75	35	126-127	126-127 ⁴¹
5			72	40	138-140	-
6			77	30	145-147	143-145 ⁴¹
7			73	45	160-162	160-162 ⁴¹
8			72	40	160-162	161-163 ⁴¹

9			70	45	145-147	-
10			80	25	188-190	188-190 ⁴²
11			81	20	178-180	-
12			83	20	173-175	175-176 ⁴²
13			80	32	160-162	-
14			77	35	158-160	-
15			80	42	203-204	204-205 ⁴²
16			81	30	198-200	202-204 ⁴³
17			78	40	189-190	188-190 ⁴³
18			75	45	158-160	-

Diethyl 4-(4-chlorophenyl)-2,6-dimethyl-1,4-dihydropyridine-3,5-dicarboxylate(Sr.No:6)

Yellow colored crystalline solid, IR (KBr, cm^{-1}): 3334 (N-H), 3084 (Ar-H), 2959 (CH_3), 1648 (C=O, ester), 617 (C-Cl). $^1\text{H-NMR}$ (500 MHz, DMSO- d_6 , / ppm): 1.135 (t, 6H, $2 \times \text{CH}_3$), 2.510 (s, 6H, $2 \times \text{CH}_3$), 3.990 (q, 4H, $2 \times \text{CH}_2$), 4.848 (s, 1H, CH), 7.153–7.264 (m, 4H, Ph), 8.848 (s, 1H, NH). $^{13}\text{CNMR}$ (500MHz, DMSO- d_6 , ppm): 14.115(CH_3), 18.162(CH_3), 39.626(CH_2), 59.005(CH), 101.160-147.077($2 \times \text{C}=\text{C}$, ArC), 166.708(C=O).

CONCLUSION

In summary, we have developed one- pot methodologies for the synthesis of 1,4,-DHP, catalyzed with Mg/Fe=3 hydrotalcite, a heterogeneous base catalyst. Compare with other procedures, this method has the advantages of easy operation with short reaction time, mild, non- hazardous reaction conditions and good yields of potentially active compounds.

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