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ARTICLE

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Efficient one-pot synthesis of polyhydroquinoline derivatives through the Hantzsch condensation using IRMOF-3 as heterogeneous and reusable catalyst

Vaishali N. Rathod | Nilam D. Bansode | Premkumar B. Thombre | Machhindra K. Lande

Department of Chemistry, Dr. Babasaheb Ambedkar Marathwada University, Aurangabad, Maharashtra, India

Correspondence
Machhindra K. Lande, Department of Chemistry, Dr. Babasaheb Ambedkar Marathwada University, Aurangabad, Maharashtra 431004, India.
Email: mkl_chem@yahoo.com

Abstract

A mesoporous Zn-based 2-amino terephthalate metal organic framework (IRMOF-3) catalyst was prepared using the solvothermal method. The synthesized catalyst was characterized by powder X-ray diffraction (XRD), thermogravimetric analysis (TGA), Fourier transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), energy-dispersive spectroscopy (EDAX), and Brunauer–Emmett–Teller surface area analysis (BET). It was applied as an effective heterogeneous catalyst for the synthesis of one-pot four-component polyhydroquinoline derivatives via the Hantzsch condensation. The present method offers several advantages over other reported methods such as easy separation, mild reaction condition, and excellent yield of desired product. Furthermore, the catalyst can be reused without loss in activity.

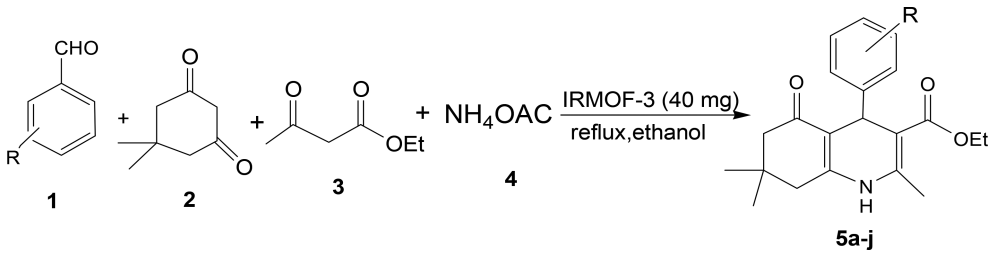
KEYWORDS

heterogeneous, metal organic framework, polyhydroquinoline, solvothermal

1 | INTRODUCTION

A metal organic framework (MOF) is a crystalline, mesoporous, designable material; these are building blocks of metal ions linked by organic linker binding with a strong bond.[1] They have extensive applications in gas storage,[2] gas separation,[3] and catalytic reaction[4] due to their tunable pore size, good thermal stability, large surface area, good chemical stability, intact catalytic properties, and multiple catalytic sites—either metal cluster, organic linker, or both.[5] Considering this view, we have successfully synthesized a zinc-based 2-amino terephthalate metal organic framework (IRMOF-3) catalyst. It was characterized by Fourier transform infrared spectroscopy (FT-IR), energy-dispersive spectroscopy (EDAX), scanning electron microscopy (SEM), Brunauer–Emmett–Teller surface area analysis (BET), and powder X-ray diffraction (XRD) and was used as a heterogeneous catalyst for the synthesis of polyhydroquinoline derivatives (Scheme 1).

In recent years, much attention has been focused on the synthesis of 1,4 dihydropyridine (1,4 DHP) because of their significant pharmacological and biological properties.[6]4-substituted 1,4 DHP compounds are well known as Ca^{+2} channel modulators[7] and have emerged as one of the most elevated classes of drugs for the treatment of cardiovascular disease, including effective treatment of hypertension.[7b,8,9] 1,4 DHP compounds are found in a variety of biological activities such as hepatoprotective, antidiabetic, geroprotective, hepatoprotective bronchodilator, vasodilator, atherosclerotic, antitumor agent, and antidiabetic activities.[8d,10,11] For this reason, the syntheses of the polyhydroquinoline compound has not only captivated attention but also represents an interesting research challenge to chemists.[12] Numerous methods have been reported for the structurally related synthesis



SCHEME 1 IRMOF-3 catalyzed unsymmetrical Hantzschreaction

of polyhydroquinoline derivatives and of the biologically importance associated with these compound, and there are many classical methods, as well as various catalysts, such as silica per chloric acid ($HClO_4$-SiO_2)[13], ionic liquid[14], heteropoly acid,[15] iron (II) trifluoro acetate[16], trimethyl silyl chloride (TMSCl),[17] HY-zeolite,[18] $Sc(OTf)_3$, $Yb(OTf)_3$[19] montmorillonite K-10,[20] and silica-supported sulfuric acid (SSA); each of the methods offers different advantages for the Hantzsch condensation but, at the same time, suffer from certain drawbacks such as longer reaction time, unsatisfactory yields, toxic catalysts, expensive catalysts, harsh reaction conditions, difficult workup, and nonreusable catalysts. Hence, in order to overcome these limitations, it is necessary to develop an efficient, cost-effective, and eco-friendly protocol synthesis of polyquinoline derivatives.

Here, we report a facile nonsymmetric Hantzsch condensation in the presence of an IRMOF-3 heterogeneous catalyst under the reflux condition using substituted aldehyde (1), 5,5- dimethyl-1,3-cyclohexanedione (2), ethyl acetoacetate (3), and ammonium acetate (4) to frame polyhydroquinoline derivatives (5a–j) in excellent yield.

2 | CHEMICAL AND INSTRUMENTATION

All solvents and chemicals were of analytical grade and were purchased from Merck, Spectrochem, HPLC, and absolute and were used as such. FT-IR analysis was recorded by using attenuated total reflection (ATR) on a Bruker instrument. Powder XRD patterns were recorded by a D_8 advance Bruker X-ray diffractometer using monochromatic Cu-K_α radiation ($\lambda = 1.5405$ A^o). ^{1}H and ^{13}C NMR spectra were recorded on a 400 MH_Z FT-NMR spectrometer with dimethyl sulfoxide (DMSO) as the solvent.

2.1 | Synthesis of IRMOF-3

Zinc nitrate hexahydrate (3.75 g) and 2-amino terephthalic acid (H_2ATA) (0.75 g) were dissolved separately in 30 ml of dimethylformamide (DMF) and 10 ml of ethanol with mild stirring. The clear solution of zinc nitrate and 2-amino terephthalic acid was mixed drop wise with constant stirring. The resulting mixture was transferred in a Teflon-lined stainless steel 100-ml autoclave, treated solvothermally under a static condition and autogenous pressure at 130°C for 24 hr, and then, the solvothermal reaction product was cooled naturally. The obtained compound was dried under vacuum in vacuum desiccators. The synthesized IRMOF-3 heterogeneous catalyst was characterized and catalyzed for synthesis of polyhydroquinoline derivatives (5a–j).

2.2 | General procedure for preparation of polyhydroquinoline

A mixture of aldehyde (1 mmol), 5,5-dimethyl-1,3-cyclohexanedione (1 mmol),ethyl acetoacetate (1 mmol), and ammonium acetate (1 mmol) with a catalyst IRMOF-3 (40 mg) was refluxed in ethanol with constant stirring for 2–3 hr, and the reaction was monitored by using thin layer chromatography (TLC) (pet ether: ethyl acetate (8:2) as the solvent system. After completion of the reaction, the mixture was poured into crushed ice. The precipitate reaction mass was filtered and was purified by recrystallization from ethanol to afford the desired compound in pure form (5a–j).

2.3 | Spectral data of representative sample

2.3.1 | Ethyl-1, 4, 7, 8-tetrahydro-2, 7, trimetyl-4-(phenyl)-5(6H)-oxoquinolin-3 carboxylate (5a)

^{1}HNMR (DMSO-d_6): 0.84 δ (s, 3H CH_3), 1.01 δ (s, 3H CH_3), 1.11–1.14 δ (t, 3H CH_3), 2.27–2.31 δ (m, 4H 2xCH_2), 2.40 δ (s, 3H CH_3), 3.95–4.00 δ (q, 2H CH_2), 4.86 δ (s, 1H CH), 9.08 δ (s, 1H NH), 7.05–7.08 δ (m, 1H Ar-H), 7.15–7.20 δ (m, 4H Ar-H). **^{13}C NMR (DMSO-d_6):** 14.11, 18.26, 26.40, 29.11, 32.10, 50.19, 59.00, 103.56, 109.93, 125.65, 144.65, 166.81, 194.24. **FT-IR (ATR, ν cm^{-1}):** 3,280, 1,693, 1,605, 1,202. **HRMS:** $m/z = 340.19$.

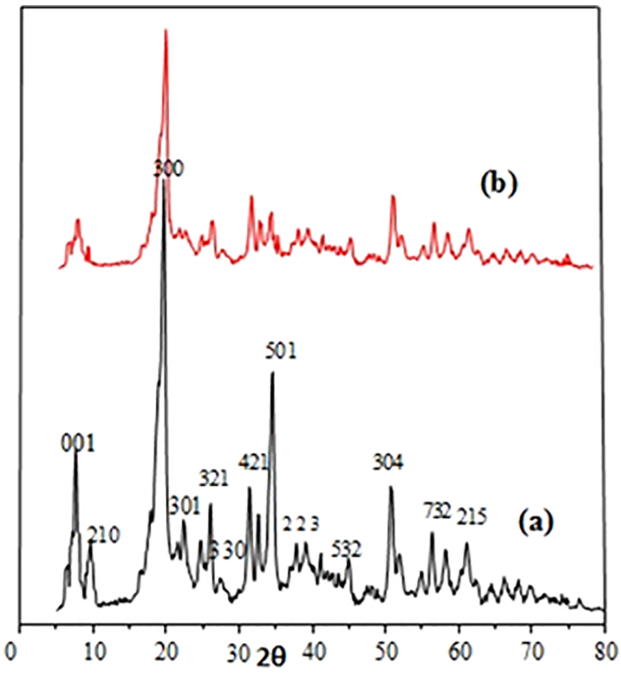


FIGURE 1 XRD pattern of IRMOF-3. (a) Newly prepared (black). (b) Used four times for 5b (red)

TABLE 1 Selected FT-IR frequencies (cm^{-1}) of the ligand and IRMOF-3

Ligand/MOF	$\nu_{(C=O)}$	$\nu_{(C=N)}$	$\nu_{(C=C)}$
Ligand	1,713	1,315	1,553
IRMOF-3	1,586	1,311	1,538

2.3.2 | Ethyl-1, 4, 7, 8-tetrahydro-2, 7, trimetyl-4-(4 Chloro phenyl)-5(6H)-oxoquinolin-3 carboxylate (5b)

^{1}HNMR (DMSO-d_6): 0.83 δ (s, 3H CH_3), 1.01 δ (s, 3H CH_3), 1.10–1.14 δ (t, 3H CH_3), 2.26–2.29 δ (m, 4H 2xCH_2), 2.40 δ (s, 3H CH_3), 3.94–3.99 δ (q, 2H CH_2), 4.84 δ (s, 1H CH), 9.13 δ (s, 1H NH), 7.14–7.17 δ (m, 2H Ar-H), 7.23–7.26 δ (m, 2H Ar-H). **^{13}C NMR (DMSO-d_6):** 14.09, 18.27, 26.38, 32.11, 35.55, 40.08, 50.10, 59.09, 103.05, 109.61, 127.67, 129.29, 145.40, 166.61, 194.25. **FT-IR (ATR, ν cm^{-1}):** 3,270, 1,700, 1,641, 1,204. **HRMS:** m/z = 374.15.

2.3.3 | Ethyl-1, 4, 7, 8-tetrahydro-2, 7, trimetyl-4-(3 nitro phenyl)-5(6H)-oxoquinolin-3 carboxylate (5c)

^{1}HNMR (DMSO-d_6): 0.83 δ (s, 3H CH_3), 1.05 δ (s, 3H CH_3), 1.09–1.13 δ (t, 3H CH_3), 2.29–2.35 δ (m, 4H 2xCH_2), 2.43 δ (s, 3H CH_3), 3.93–3.99 δ (q, 2H CH_2), 4.97 δ (s, 1H CH), 9.28 δ (s, 1H NH),7.50–7.56 δ (m, 2H Ar-H), 7.96–7.99 δ (m, 2H Ar-H). **^{13}C NMR (DMSO-d_6):** 13.98, 18.33, 26.27, 32.17, 49.95, 102.58, 109.17, 129.41, 146.13, 166.35, 194.32. **FT-IR (ATR, ν cm^{-1}):** 3,204, 1,699, 1,602, 1,527, 1,348.

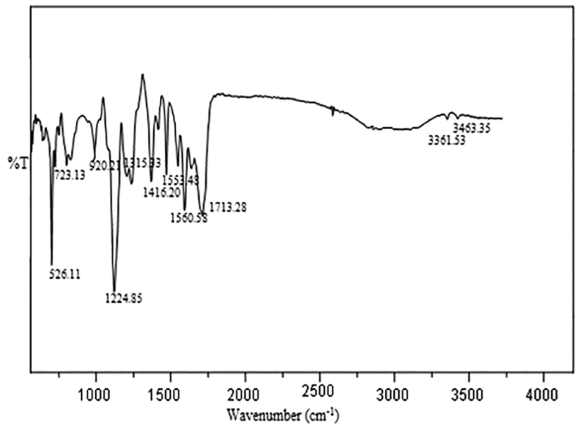


FIGURE 2 IR Spectrum of ligand

FIGURE 3 FT-IR spectrum of IRMOF-3 catalyst

2.3.4 | Ethyl-1, 4, 7, 8-tetrahydro-2, 7, trimetyl-4-(4 hydroxy phenyl)-5(6H)-oxoquinolin-3 carboxylate (5d)

^{1}HNMR (DMSO-d_6): 0.85 δ (s, 3H CH_3), 1.06 δ (s,3H CH_3), 1.12–1.15 δ (t,3H CH_3), 2.25–2.29 δ (m, 4H 2xCH_2), 2.38 δ (s, 3H CH_3), 3.96–3.98 δ (q, 2H CH_2), 4.74 δ (s, 1H CH), 9.06 δ (s, 1H NH), 6.54–6.57 δ (m, 2H Ar-H), 6.92–6.94 δ (m, 2H Ar-H), 8.99 δ (s, 1H Ar-OH). **^{13}C NMR (DMSO-d_6):** 14.15, 18.23, 29.15, 32.10, 50.25, 58.42, 104.06, 110.28, 114.39, 138.38, 144.37, 166.98. **FT-IR (ATR, ν cm^{-1}):** 3,273, 1,680, 1,605, 1,213. **HRMS:** m/z = 356.18.

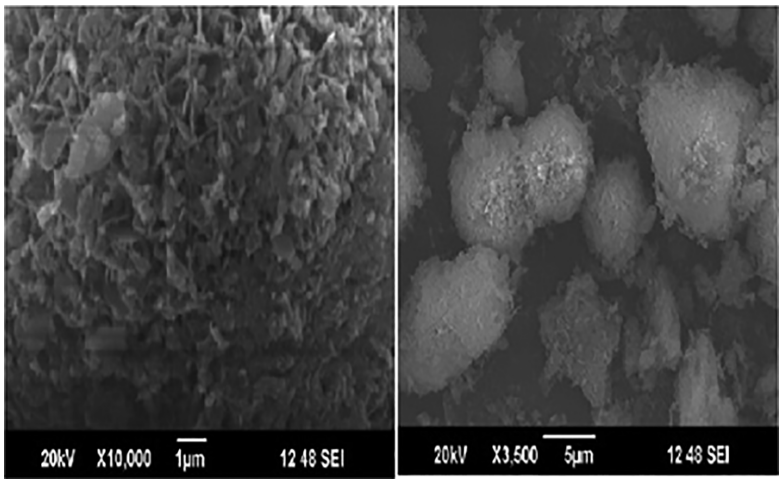


FIGURE 4 SEM image of IRMOF-3 catalyst

2.3.5 | Ethyl-1, 4, 7, 8-tetrahydro-2, 7, trimetyl-4-(4 fluoro phenyl)-5(6H)-oxoquinolin-3 carboxylate (5j)

^{1}HNMR (DMSO-d_6): 0.83δ(s, 3H CH_3), 1.00 δ (s, 3H CH_3), 1.10–1.13 δ (t,3H CH_3), 2.26–2.30 δ (m, 4H 2xCH_2), 2.39 δ (s, 3H CH_3), 3.94–3.99 δ (q, 2H CH_2), 4.84 δ (s, 1H CH), 9.11 δ (s, 1H NH), 6.98–7.02 δ (m, 2H Ar-H), 7.14–7.18 δ (m, 2H Ar-H). **^{13}C NMR (DMSO-d_6):**14.09, 18.26, 26.38, 32.11, 35.24, 40.07, 50.12, 59.04, 103.37, 109.86, 114.43, 129.15, 143.81, 166.69, 194.27. **FT-IR (ATR, ν cm^{-1}):** 3,270, 1,702, 1,641, 1,212.

3 | RESULT AND DISCUSSION

3.1 | Powder X-ray diffraction

Powder XRD patterns of IRMOF-3 are given in Figure 1. According to the calculation (JCPDS Id No. 47-2088 PDF# 472088, wavelength = 1.5418, volume [CD]- 1,467.47), which indicates an intense peak at 2θ^{o} = 19.436, 34.680, 31.433, and 50.723 with corresponding planes of (300), (501), (421), and (304), respectively, the presence of planes is found in the tetragonal phase of IRMOF-3 material. The XRD patterns of IRMOF-3 show that the crystallinity and integrity were intact after the organic transformations, as shown Figure 1a,b. Therefore, IRMOF-3 is a thermally and chemically stable heterogeneous catalyst.

3.2 | Infrared spectroscopy analysis

The infrared spectroscopy (IR Spectroscopy) spectrum indicated valuable information concerning the nature of the function group of the ligand coordinated to the metal ions. The IR spectrum of IRMOF-3 shows two peaks in 3,453 and 3,340 cm^{-1} due to the existence of the amino group of the NH_2-terephthalic acid ligand.[20] The selected spectrum data of the 2-amino terephthalic acid and IRMOF-

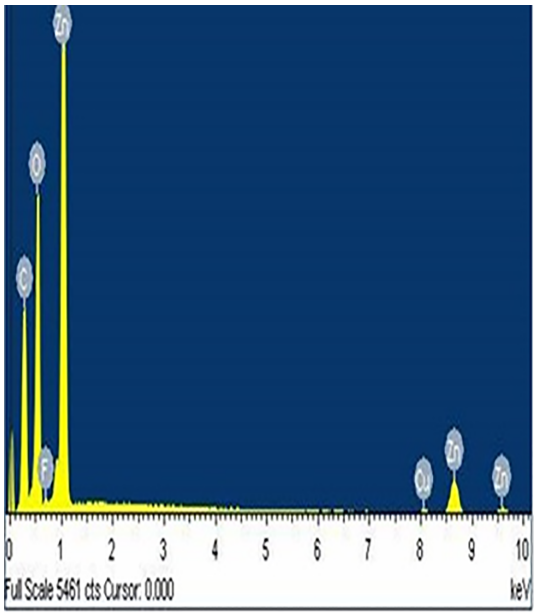


FIGURE 5 EDAX spectrum of IRMOF-3 catalyst

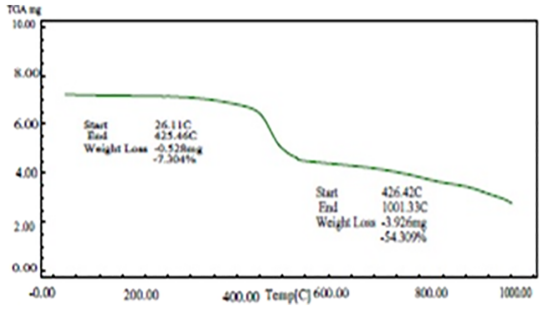


FIGURE 6 TGA analysis of IRMOF-3 catalyst

TABLE 2 Optimization of IRMOF-3 catalyst and solvent effect on the synthesis of 5b

Entry	Solvent	Catalyst (mg)	Time (hr)	Yield (%)[a]
1	THF	40	2	72
2	Toluene	40	1.5	70
3	DCM	40	2	71
4	Chloroform	40	1.5	70
5	Acetonitrile	30	2	69
6	Ethanol	—	8	40
7	Ethanol	10	2	65
8	Ethanol	20	2	80
9	Ethanol	40	2	95

[a]Isolated yield.

3-based MOF are given in Table 1.The absorption band is in the range of 1,713–1,590 cm^{-1} due to the presence of C=O group in the 2-amino terephthalic acid ligand (Figure 2). The absorption band of IRMOF-3 was found to be in the range of 1,586–1,538 cm^{-1}, the results of which confirmed the coordination of the C=O group of 2-amino terephthalic acid ligand with Zn metal salts (Figure 2). C=N and C=C stretching frequency was observed in the range of 1,315–1,224 and 1,553–1,416 cm^{-1}, respectively, but it decreased in IRMOF—1,311–1,221 and 1,520–1,412 cm^{-1}. Thus, this result confirms that the organic acid coordinated to the metal ion to form the target compound (Figure 3).

3.3 | Scanning electron microscopy

SEM used to study the surface morphology of IRMOF-3 is shown in Figure 4. The SEM image observed crystalline and irregular duab shape particle morpholog.

The element compositions of the IRMOF-3 catalyst are given in Figure 5, which includes elements of zinc (14.24%), carbon (46.12%), and oxygen (26.28%) (F(0.80%), Cu (0.65%) trace amount analyzed impurity). The EDS study found that the elements zinc, carbon and oxygen are present, which confirms the successful synthesis of IRMOF-3.

3.4 | BET surface area and porosity

The BET surface area, average pore diameter, and pore volume of IRMOF-3 were found to be 130.69 m^2/g, 48.53 A°, and 0.436 cm^2/g, respectively, calculated by the N_2 absorption desorption barrett-joyner-halenda (BJH) isotherm.

3.5 | Thermogravimetric analysis of IRMOF-3 catalyst

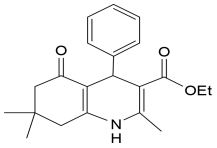
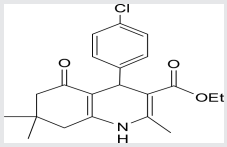
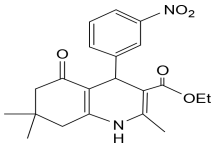
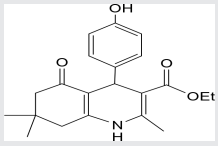
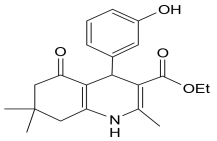
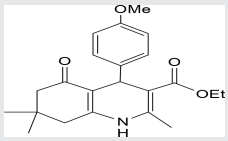
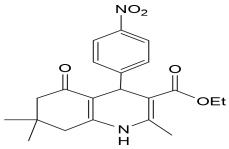
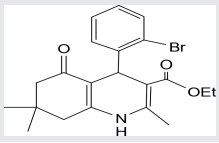
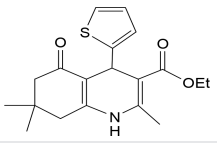
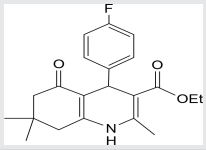
Thermo gravimetric analysis (TGA) was performed to determine the thermal stability of synthesized IRMOF-3. The TGA curve shows two weight losses. The first weight loss (7.304%) was found in the range between 80 and 200°C; it indicates the loss of moisture. The second weight loss (54.309%) started in the range 450–800°C, temperatures at which the structure of the IRMOF-3 collapsed and the 2-amino terephthalate was fractured. It found that the synthesized IRMOF-3 was stable up to 450°C (*Figure* 6).

3.6 | Optimization of reaction condition

We aimed to find the optimal condition by examining the mixture of 4-chloro benzaldehyde (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol), and ammonium acetate (1 mmol) as a model reaction with different amounts of the heterogeneous catalyst IRMOF-3 under the reflux condition for the synthesis of polyhydroquinoline derivatives in high yields. We also studied the efficiency of model reaction carried out under the different polar and non polar solvent chooses as the medium shown Table 2. We found that the reaction proceeded faster in a polar solvent, such as ethanol and acetonitrile, and these solvents were much better than nonpolar solvents.

We then continued to optimize the conditions electronically to test diverse different aromatic aldehydes, and results are summarized in Table 3. Aldehydes having an electron-withdrawing group such as 4-Cl, 4-NO_2, 3-NO_2, and 4-F were found to be highly reactive and gave maximum yield of the product compared to aldehydes with an electron-releasing group such as 4-OH, 3-OH, and 4-Ome. After optimizing the reaction conditions, efforts were made toward the recovery and reusability of the IRMOF-3 catalyst. The catalyst was removed after the completion of the reaction by dissolving the reaction mixture in hot ethanol and filtration. The recovered catalyst was washed four to five times with DMF and then immersed in methanol for 24 hr and dried at 130°C for

TABLE 3 IRMOF-3-catalyzed synthesis of polyhydroquinoline derivatives

Sr.no.	Product	Entry	Yield (%)[a]	Melting point (°C)	
				Observed	Reported
1		5a	91	203–204	202–204[19]
2		5b	95	244–245	245–246[21]
3		5c	92	176–177	177–178[22]
4		5d	91	233–234	232–234[19]
5		5e	89	225–230	—
6		5f	88	255–256	256–257[19]
7		5g	91	241–243	242–244[21]
8		5h	86	254–256	253–255[19]
9		5i	87	239–241	238–240[22]
10		5j	92	184–186	184–186[19]

Note: Reaction condition: aldehyde (1 mmol), dimedone (1 mmol), ethyl acetoacetate (1 mmol) and ammonium acetate (1 mmol), Catalyst (40 mg) and ethanol (10 ml).
[a]Isolated yield.

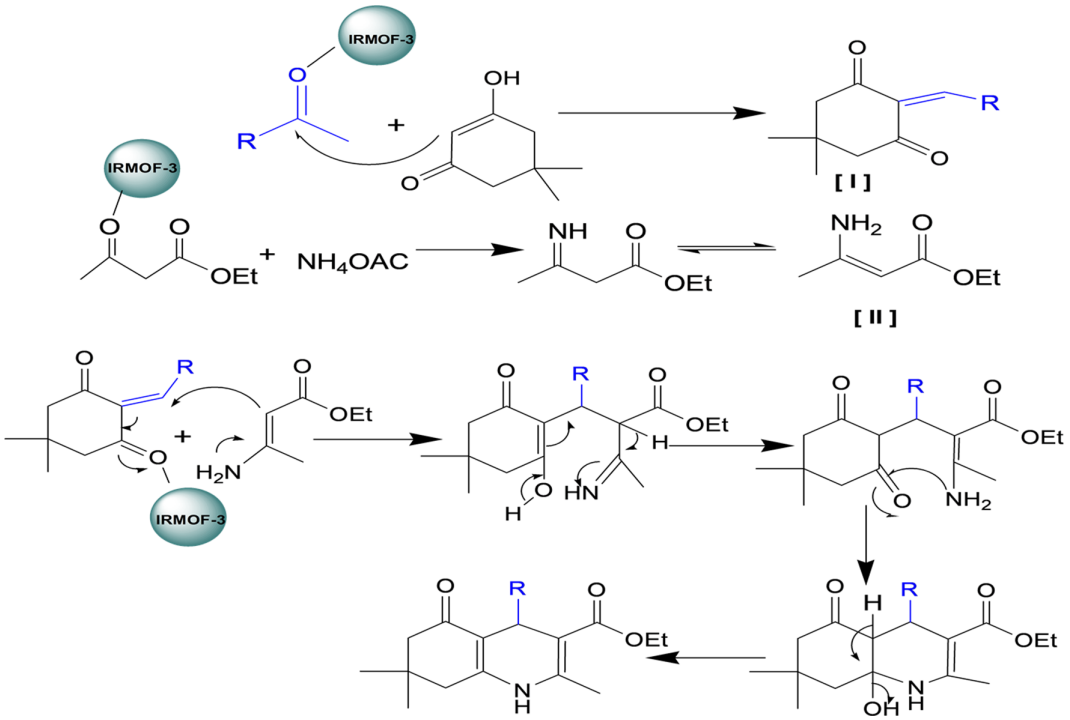
TABLE 4 Recyclability studies of IRMOF-3 catalyst for the synthesis of 5b

Entry	Cycle	Yield (%)[a]
1	Fresh	95
2	First	94
3	Second	91
4	Third	90

[a]Isolated yield.

3 hr before the next catalytic run, which could be subsequently performed several times,as shown in Table 4, for compound 5b. Reusability of the catalyst was investigated four times, and it was found that the catalyst has intact, almost consistent activity.

The plausible reaction mechanism shows the preparation of the polyhydroquinoline derivatives. The IRMOF-3 catalyst enhances the electrophilicity of the carbonyl of aldehyde, and then, the dimedone reacts with the



SCHEME 2 Proposed reaction mechanism for IRMOF-3-catalyzed polyhydroquinoline synthesis

TABLE 5 Comparison of the catalytic performance of different reported catalysts used in synthesis of 5b

Entry	Catalyst	Temp	Time (hr)	Yield (%)[a]	Literature
1	$Yb(DTF)_3$	25	5	90	[23]
2	$AlCl_3$	25	24	48	[23]
3	$ZnCl_2$	25	24	42	[23]
4	I_2	25	1.5	93	[24]
5	ZnO	80	1	92	[25]
6	$Sc(OTf)_3$	60	5	93	[23]
7	Hy-zeolite	25	2	93	[18]
8	L-proline	RT	6	92	[26]
9	Nano-γ-Fe_2O_3-SO_3H	60	1.5	93	[27]
10	SO_4^{2-}/TiO_2NPs	Reflux	1	93	[29]
11	IRMOF-3	Reflux	1–2	95	Our results

[a]Isolated yield.

aldehyde to form intermediate (I) and with ethyl acetoacetate and ammonium acetate to form intermediate (II). Intermediates I and II undergo cyclocondensation to obtain the final desired product (Scheme 2).

To specify the advantage of the defined methodology, the results of different reported methods are compared with our results and summarized in Table 5. It was observed from the tabulated results that the IRMOF-3 catalyst promoted the reaction more effectively than other reported methods.

4 | CONCLUSIONS

In summary, we have synthesized a mesoporous IRMOF-3 MOF catalyst using the solvothermal method. Efficient catalytic activity has been developed for the synthesis of polyhydroquinoline derivatives via the Hantzsch condensation reaction. In the IRMOF-3 catalyst, there is organization between the Lewis acidic site and rigid NH_2-benzenedicarboxylate linking in the zinc-based MOF cluster. XRD, IR, BET, SEM analysis confirm the tetragonal phase, greater porediameter, morphology observed crystalline and irregular duab shape and it has no significance loss of catalytic activity when used in organic transformation for three to four run. This method offers remarkable benefits over other reported methods, for example, easy to separate, reusable catalytic characters, nontoxic, short reaction time, large pore size, and provided excellent yield of polyhydroquinoline derivatives (5a–j).

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