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FULL PAPER

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Preparation of core/shell/shell $CoFe_2O_4$/OCMC/Cu (BDC) nanostructure as a magnetically heterogeneous catalyst for the synthesis of substituted xanthenes, quinazolines and acridines under ultrasonic irradiation

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Highly efficient synthesized magnetic cobalt ferrite nanoparticles supported on OCMC@Cu (BDC) was utilized in the preparation of biologically active heterocyclic compounds through one-pot three-component reactions between of aldehydes, dimedone, aryl amines/2-naphthol/urea under ultrasonic irradiation. This method has various advantages including excellent yields, little catalyst loading, simple procedure, facile catalyst separation, short reaction times, eco-friendly approach and simple purification. The catalyst was characterized by various spectroscopy methods such as fourier-transform infrared (FT-IR), energy-dispersive X-ray (EDX), scanning electron microscope (SEM), X-ray diffraction (XRD) and N_2 adsorption–desorption isotherm (BET). Furthermore, the heterocyclic compounds were characterized by spectral techniques. The nanocomposite was simply separated byusing an external magnet, and it can be recycled several times without significant loss of activity.

KEYWORDS

Acridines, $CoFe_2O_4$@OCMC@cu (BDC), metal–organic framework, Quinazolins, Xanthenes

1 | INTRODUCTION

Multi-component reactions (MCRs) are main synthetic methods for preparation of numerous complex molecules under one-pot conditions from three or more simple raw materials.[1,2] In recent years, MCRs have been considered as fast, inexpensive, and environmentally friendly methods.

Heterocyclic rings have a rich chemistry in a wide range of medicinal, pharmaceutical and industrial fields.[3] These versatile molecules extensively present in numerous natural products such as vitamins, hormones, dyes, and drugs.[4] In recent years, synthesis of heterocyclic compounds via MCRs has attracted a lot of attention due to their extensive biological effects. Among them, widespread interest in the preparation of the quinazoline compounds has been engendered using their broad biological properties such as anti-microbial, anti-diabetic, hypnotic, anti-inflammatory, and antitumoral activity.[5,6]

Xanthenes have many important biological activities including anti-tumoral, gastric secretion inhibitor, and opiaceous or cytotoxic activities.[7]

In addition, acridine derivatives have attracted considerable attention over the past few years due to their biological effects such as antimalarial, fungicidal, anti-inflammatory, anti-viral, anti-microbial, anti-cancer, anti-parasitic, and anti-tubercular activities.[8–12]

Metal ferrite nanoparticles or bimetal oxide magnetic nanoparticles (MNPs)are a class of MNPs which have

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various applications in several fields. Among these metal ferrite MNPs, cobalt ferrite ($CoFe_2O_4$) MNPs are more attractive owing to their mechanical hardness, chemical stability, high thermal, and high magnetization.[13]

Metal–organic frameworks (MOFs), which are constructed via coordinating polytopic organic ligands with clusters or metal ions, have aroused tremendous attention in the fields of catalysis,[14] gas adsorption and desorption,[15–17]drug delivery[18–20] and sensing,[21,22] due to their unique features such as various functionalities, extensive surface areas, adjustable internal surface properties, and high regular pores.[23–30]

Moreover, MOFs can be applied as precursors to take tailorable functional materials including metal oxide and carbon composites through thermal/chemical treatments.

Metal–organic frameworks have been used as an efficient catalyst in the catalytic hydrogenation of 2,3,5-trimethylbenzoquinone,[31] Mizoroki-Heck cross-coupling reaction,[32] preparation of spiro oxindole derivatives,[33] pyrano[2,3-d]pyrimidines,[34] propargylamines,[35] indolizines,[36] quinoxaline,[37] and pyrano[2,3-c]-pyrazoles.[38]

In the present work, we synthesized some quinazoline, xanthene and acridine derivatives via three-component reactions of aldehydes, dimedone and urea/anilines/2-naphthol in the presence of $CoFe_2O_4$@OCMC@Cu (BDC) as an effective, recyclable and robust catalyst (Scheme 1).

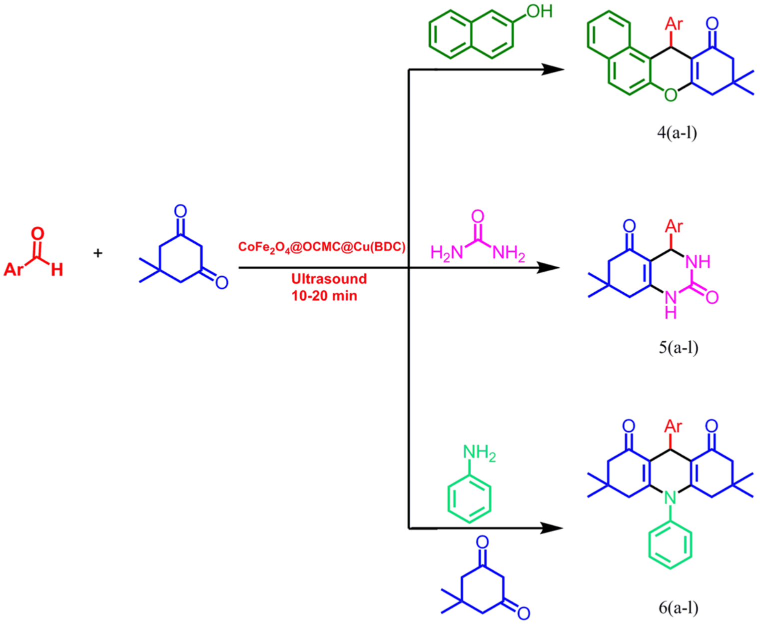
2 | EXPERIMENTAL

2.1 | Materials and instrumentation

Chemicals were purchased from the Sigma-Aldrich and Merck in high purity. All of the materials were of commercial reagent grade and were used without further purification. All melting points are uncorrected and were determined in capillary tube on Boetius melting point microscope.^{1}H NMR and ^{13}CNMR spectra were obtained on Bruker 400 MHz spectrometer with DMSO-d_6 as solvent using TMS as an internal standard. Fourier-transform infrared (FTIR) spectrum was recorded on Magna-IR, spectrometer 550. The elemental analyses (C, H, andN) were obtained from a Carlo ERBA Model EA 1108 analyzer. Powder X-ray diffraction (XRD) was carried out on a Philips diffractometer of X'pert Company with mono chromatized Cu Kα radiation (λ = 1.5406 Å). Microscopic morphology of products was visualized by scanning electron microscope (SEM) (LEO 1455VP). The mass spectra were recorded on a Joel D-30 instrument at an ionization potential of 70 eV. The compositional analysis was done by Energy-dispersive X-ray (EDX, Kevex, Delta Class I). Nitrogen adsorption–desorption isotherms were measured at 196 °C using a Belsorp mini automatic adsorption instrument after degassing the samples at 150 °C for 5 hr.

2.2 | Preparation of$CoFe_2O_4$/OCMC/cu (BDC)

$CoFe_2O_4$/OCMC/Cu (BDC)nanocomposite were synthesized according to a previously reported method by ghasemzadeh and co-workers.[39] Firstly, polyvinylpyrrolidone (PVP) (0.12 g) was dissolved in a mixture of water and ethanol (30 ml). Then, CoFe2O4/OCMC (0.06 g) was added and the reaction mixture was dispersed by ultrasonic bath for 15 min. Subsequently, a mixture of copper (II) nitrate (0.022 g) and terephthalic



SCHEME 1 Synthesis of xanthenes, quinazolines and acridines in the presence of $CoFe_2O_4$@OCMC/Cu (BDC) under ultrasonic irradiation

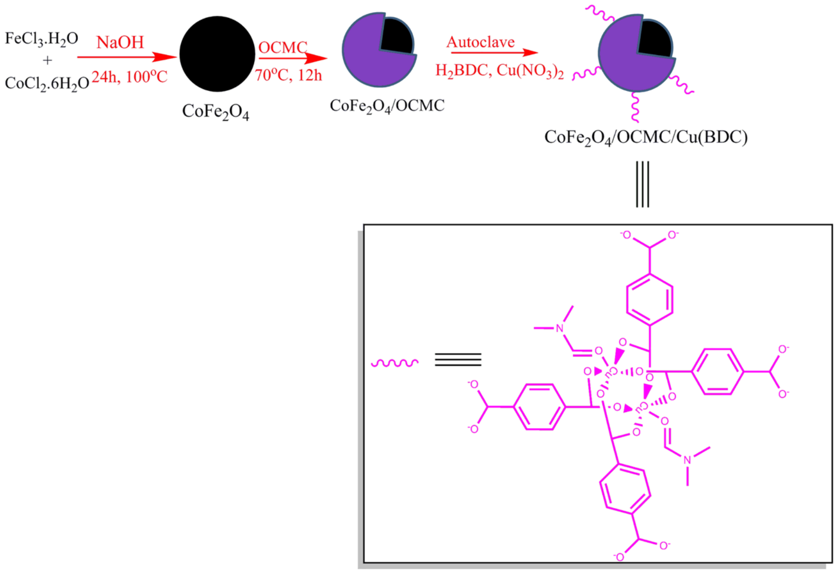
acid (0.053 g) dissolved in dimethylformamide (DMF) (2 ml) was added and the reaction mixture was dispersed for 10 min. The slurry was placed in an autoclave and heated at 100 °C for 4 hr. Finally, the resulted nanostructure was collected by an external magnet and washed with water/DMF mixture and dried in an oven at 80 °C for 24 hr (Scheme 2).

2.3 | General procedure for the synthesis oftetrahydrobenzo[a]xanthen-11-ones (4a-l) using CoFe$_2$O$_4$/OCMC/cu (BDC)

A mixture of aldehyde (1 mmol), 2-naphthol (1 mmol), dimedone (1 mmol), was added to a flask containing ethanol (2.5 ml) and water (2.5 ml). The reaction mixture was sonicated in the presence of CoFe$_2$O$_4$/OCMC/Cu (BDC) nanocomposites (0.002 g) under 30 kHz frequency at room temperature. The reaction progress was monitored by TLC using hexane/ethyl acetate (8:2). After completion of the reaction, the reaction mixture was dissolved in dichloromethane and the catalyst was separated by an external magnet. Finally, the solvent was evaporated under reduced pressure and the residue was recrystallized from ethanol to afford the pure product.

2.4 | General procedure for the synthesis of octahydroquinazolinones (5a-l) using CoFe$_2$O$_4$/OCMC/cu (BDC)

CoFe$_2$O$_4$/OCMC/Cu (BDC)nanocomposite (0.005)was added to a mixture of dimedone (1 mmol), aldehyde (1 mmol) and urea (1 mmol) in ethanol/water (1:1,10 ml). The reaction mixture was sonicated at 30 kHz frequency for 10–20 min at room temperature. The progress of the reaction was continuously monitored



SCHEME 2 The preparation steps of OCMC/Cu (BDC)-functionalized magnetic CoFe$_2$O$_4$ nanoparticles

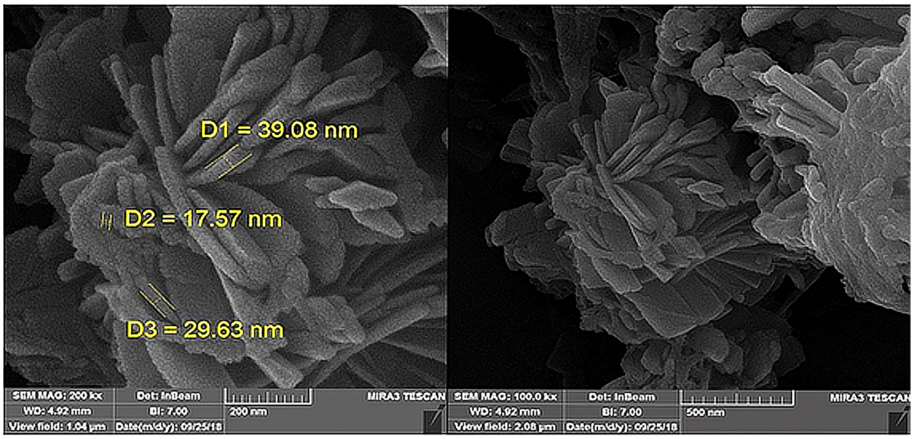


FIGURE 1 The SEM images of CoFe$_2$O$_4$@OCMC@Cu (BDC)

by TLC. After completion of the reaction, the reaction mixture was dissolved in dichloromethane and the catalyst was separated by an external magnet. The product was obtained after evaporation of the solvent and recrystallization from ethanol.

2.5 | General procedure for the synthesis of 1,8-dioxo-decahydroacridines(6a-6 l) using $CoFe_2O_4$/OCMC/cu (BDC)

A mixture of anilines (1 mmol), dimedone (2 mmol), aldehyde (1 mmol) and nanocatalyst (0.002 g) was added to a flask containing ethanol (2.5 ml) and water (2.5 ml). The reaction progress was continuously monitored by TLC. Upon completion, the reaction mixture was dissolved in dichloromethane and the catalyst was separated by an external magnet. The product was afforded by evaporation of the solvent without any purification.

Spectral data of some products are given in Supplementary Information.

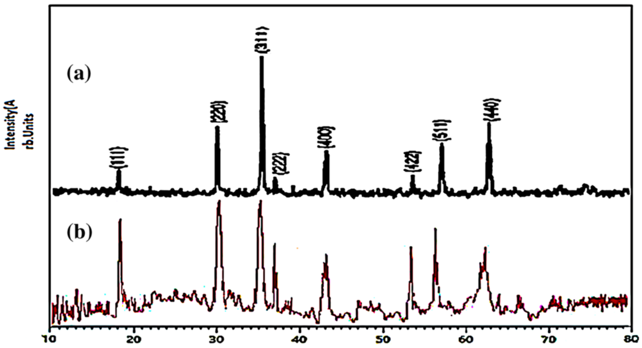


FIGURE 2 XRD patterns of $CoFe_2O_4$ (a) and $CoFe_2O_4$@OCMC@Cu (BDC) (b)

3 | RESULTS AND DISCUSSION

3.1 | Investigation of catalytic activity of cu (BDC)-MOF fanctionalized $CoFe_2O_4$@OCMC in the synthesis of 1,8-dioxo-decahydroacridines, octahydroquinazolinones and tetrahydrobenzo[a]xanthen-11-ones

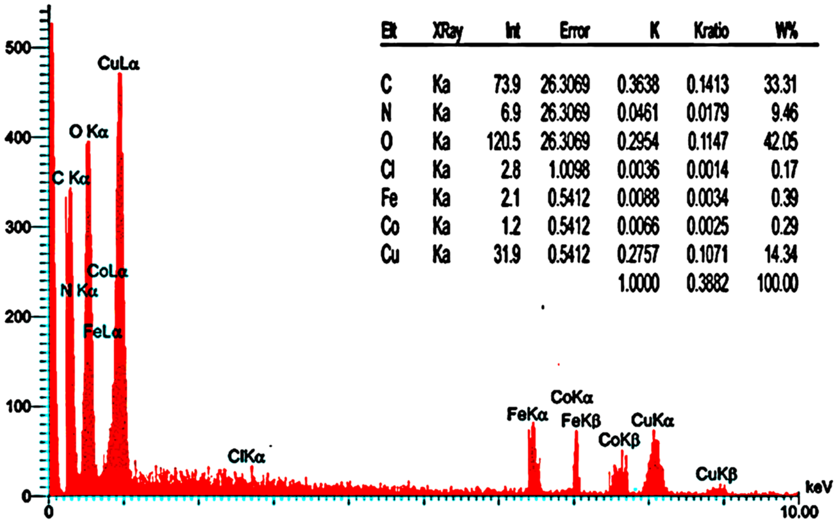
$CoFe_2O_4$/OCMC/Cu (BDC) nanostructure was initially synthesized and evaluated by FE-SEM, EDX, FT-IR, XRD and BET analysis.

The surface morphology of the synthesized $CoFe_2O_4$/OCMC/Cu (BDC)-MOF was investigated by FE-SEM analysis (Figure 1). It is clear that the magnetic $CoFe_2O_4$/OCMC/Cu (BDC) nanocomposite has a flower shape with diameter about 20–40 nm.

The structure and crystalline phase of the catalyst was analyzed by XRD spectroscopy (Figure 2). In the XRD pattern of $CoFe_2O_4$@OCMC@Cu (BDC) (Figure 3B), the diffraction peaks at 2θvalues of 18.29°, 30°, 35.44°, 37.06°, 42°, 53°, 56.98° and 62.59° were observed indicating the maintenance of the crystalline structure of $CoFe_2O_4$ during the functionalization.

The chemical purity of the $CoFe_2O_4$/OCMC/Cu (BDC) nanocatalyst was investigated by energy-dispersive X-ray spectroscopy (EDX), which is given in Figure 3. The EDX spectrum shows the presence of Co, Cu, Fe, C, O and N as the elements of the nanocomposites.

Fourier transform infrared (FT-IR) spectroscopy was utilized to confirm the catalyst structure (Figure 4). For the pure magnetic nanoparticles (Figure 4a), the strong peak at 579 cm^{-1}is related to the Fe-O vibration of the nanocatalyst. The other significant peaks at 3410 cm^{-1}, 1628 cm^{-1}attribute to O – H stretching and bending vibrations of the absorbed water molecules on the surface



Elt	XRay	Int	Error	K	Kratio	W%
C	Ka	73.9	26.3069	0.3638	0.1413	33.31
N	Ka	6.9	26.3069	0.0461	0.0179	9.46
O	Ka	120.5	26.3069	0.2954	0.1147	42.05
Cl	Ka	2.8	1.0098	0.0036	0.0014	0.17
Fe	Ka	2.1	0.5412	0.0088	0.0034	0.39
Co	Ka	1.2	0.5412	0.0066	0.0025	0.29
Cu	Ka	31.9	0.5412	0.2757	0.1071	14.34
				1.0000	0.3882	100.00

FIGURE 3 The EDX spectrum of $CoFe_2O_4$@OCMC@Cu (BDC)

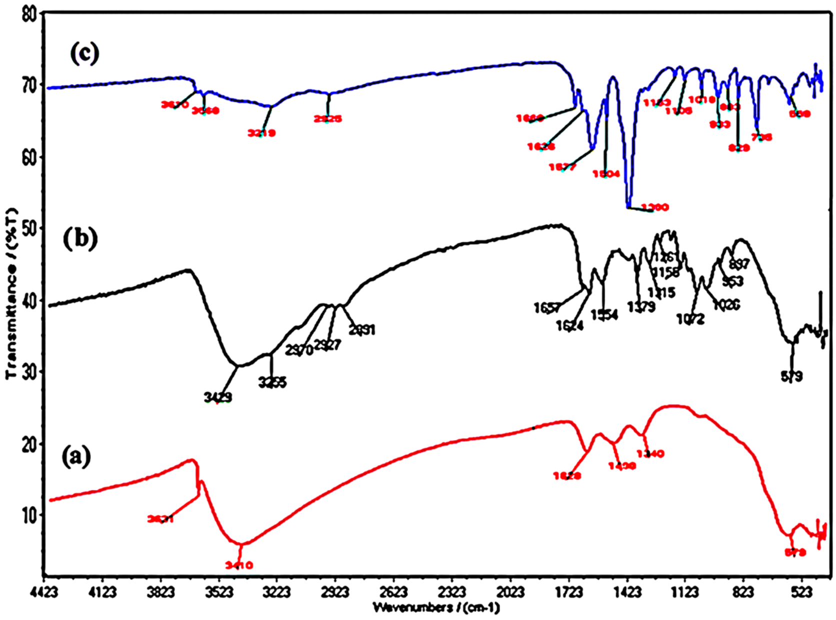


FIGURE 4 FT-IR spectra of $CoFe_2O_4$ NPs (a), $CoFe_2O_4$@OCMC(b), $CoFe_2O_4$ @OCMC@Cu (BDC) (c)

of cobalt ferrite nanoparticles. In the case of $CoFe_2O_4$@OCMCnanocomposite, the broad peak at 3429 cm^{-1} corresponds to stretching vibrations of amine and hydroxyl groups of chitosan (Figure 4b). The peaks at 1657 cm^{-1}and 1379 cm^{-1}are also related to the stretching vibrations of carbonyl group and N-H bending vibration of chitosan. In terms of the $CoFe_2O_4$@OCMC@Cu (BDC) (Figure 4c), the band at2925cm^{-1}attributes to aliphatic C-H asymmetric stretching vibrations of DMF, and the characteristic peak at 1504 cm^{-1} is related to C=C vibrations of aromatic ring moiety.

The strong peaks at 1390 and 1577 cm^{-1} are due to symmetric and asymmetric vibrations of carboxylate anions in the MOF structure. It is important to say that the bond observed at 1666 cm^{-1} corresponds to C=O vibration of DMF.

The surface area, total pore volumes and average pore diameter of the catalyst were investigated by N_2 adsorption–desorption isotherm spectrum (BET) (Figure 5).[39] It is clear that the specific surface area, the average pore diameter, and the average pore diameter of $CoFe_2O_4$@OCMC@Cu (BDC) are 64.933 m^2/g, 11.46 nm, and 0.186 cm^3/g, respectively. It is also important to mention that the surface area of $CoFe_2O_4$@OCMC@Cu (BDC) decreases gradually due to the presence of the OCMC polymer and the heavy atoms of Cu (BDC) clusters in the catalyst framework.

Afterwards, the catalytic influence of OCMC/Cu (BDC) coated $CoFe_2O_4$ NP was evaluated for the synthesis of acridine, quinazolinone and xanthene derivatives through the reaction of dimedone, 4-chlorobenzaldehyde, urea/2-naphthol/aniline as model reactions (Table 1).

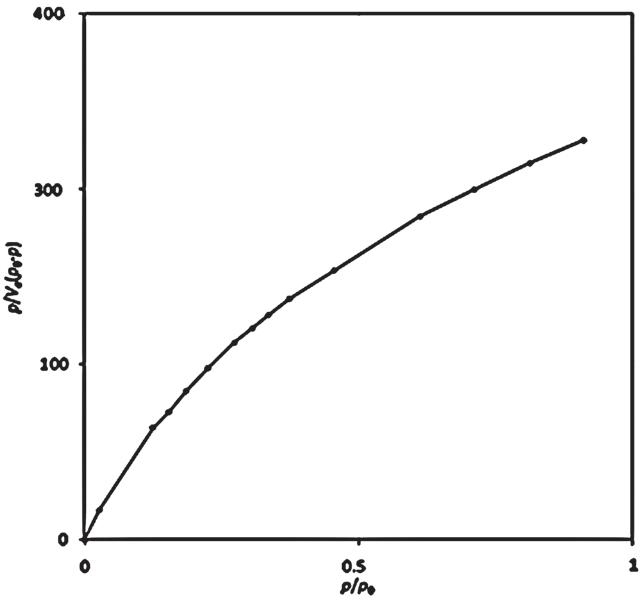
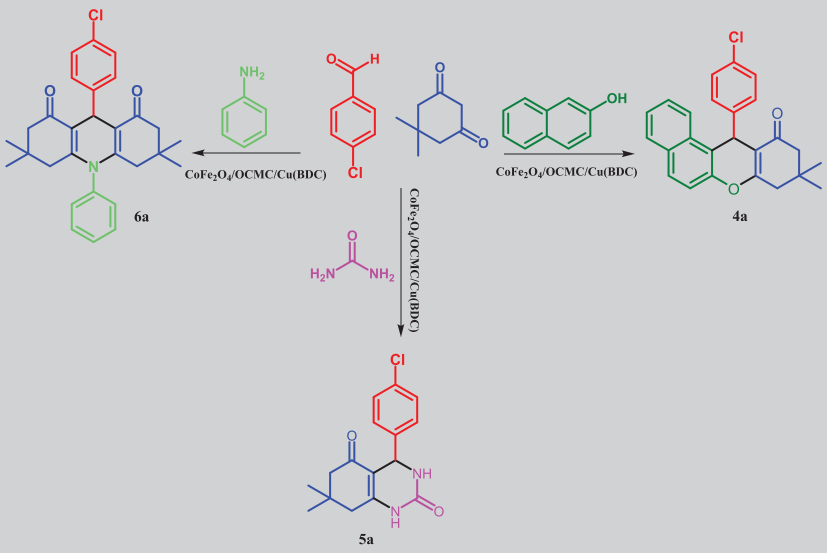


FIGURE 5 N_2 adsorption/desorption isotherms of $CoFe_2O_4$@OCMC@Cu (BDC)

The reaction was optimized under various conditions such as temperature, catalyst, and solvent.

Initially, the preparation of 4a, 5a, 6a (as examples of three types of the prepared xanthenes, quinazolines and acridines) was studied using different solvents such as ethanol, dichloromethane, water, and acetonitrile under catalyst-free conditions. The results showed that no product was formed under ultrasonic irradiations. Subsequently, the reaction was carried out for the preparation of 4a, 5a, 6a compounds in the presence of various solvents and $CoFe_2O_4$/OCMC/Cu (BDC) (0.01 g) as catalyst.

TABLE 1 The effect of different solvents and catalysts in the synthesis of 4a, 5a, 6a derivatives



Entry	Compound	Solvent	Catalyst	T (°C)	Time (min)	Yield%
1	4a	CH_3CN	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	25	56
2	4a	H_2O	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	20	60
3	**4a**	**C_2H_5OH**	**$CoFe_2O_4$/OCMC/Cu (BDC)**	**u.s.**	**10**	**96**
4	4a	CH_2Cl_2	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	60	56
5	5a	H_2O	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s	48	64
6	5a	**C_2H_5OH**	**$CoFe_2O_4$/OCMC/Cu (BDC)**	**u.s.**	**10**	**94**
7	5a	CH_3CN	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	45	61
8	5a	CH_2Cl_2	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	32	79
9	**6a**	**C_2H_5OH**	**$CoFe_2O_4$/OCMC/Cu (BDC)**	**u.s.**	**10**	**97**
10	6a	CH_2Cl_2	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	30	84
11	6a	H_2O	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	18	90
12	6a	CH_3CN	$CoFe_2O_4$/OCMC/Cu (BDC)	u.s.	15	83
13	4a	C_2H_5OH	ZnO NPs	u.s.	40	82
14	4a	C_2H_5OH	Fe_3O_4NPs	u.s.	35	80
15	4a	C_2H_5OH	$CoFe_2O_4$NPs	u.s.	25	85
16	5a	C_2H_5OH	ZnONPs	u.s.	32	78
17	5a	C_2H_5OH	Fe_3O_4 NPs	u.s.	25	84
18	5a	C_2H_5OH	$CoFe_2O_4$NPs	u.s.	30	81
19	6a	C_2H_5OH	ZnO NPs	u.s.	35	85
20	6a	C_2H_5OH	Fe_3O_4NPs	u.s.	25	80
21	6a	C_2H_5OH	$CoFe_2O_4$NPs	u.s.	22	95

The results are summarized in Table 1. As shown in this Table, the best results were obtained using the ethanol as solvent (entries 3, 6, and 9).

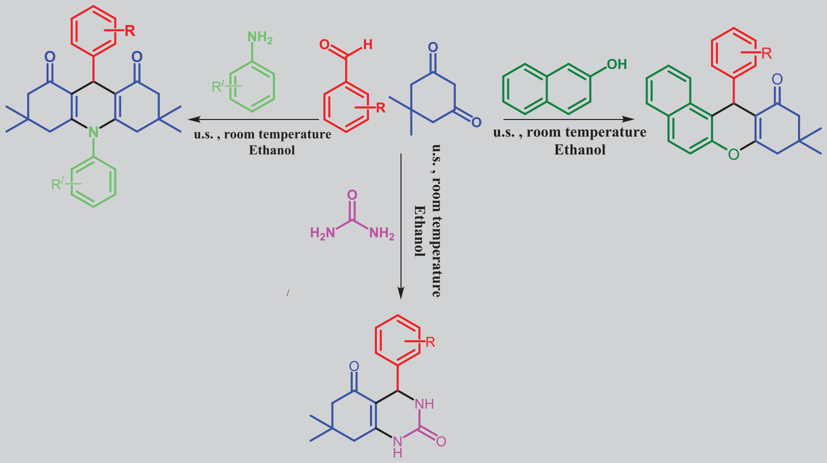
In order to investigate the catalytic activity, we also carried out the model reactions in the presence of several catalysts in ethanol. The best results were observed when the $CoFe_2O_4$/OCMC/Cu (BDC) nanostructure was used as catalyst.

The amounts of catalyst loading was examined by using the various amounts of $CoFe_2O_4$@OCMC@Cu (BDC) for the preparation of xanthene(4a), quinazolinone (5a) and acridine (6a) (Table 2). 0.005 g

TABLE 2 The optimization of model reaction by using various amounts of the catalyst

Entry	Compound	Catalyst (g)	Time (min)	Yield%
1	4a	0.001	25	82
2	**4a**	**0.002**	**10**	**96**
3	4a	0.005	10	96
4	5a	0.001	25	82
5	5a	0.002	18	95
6	5a	0.003	15	90
7	**5a**	**0.005**	**10**	**94**
8	5a	0.007	10	94
7	6a	0.001	30	80
8	**6a**	**0.002**	**10**	**97**
9	6a	0.003	10	96

TABLE 3 Preparation of acridine, quinazolinone and xanthene derivatives using CoFe$_2$O$_4$/OCMC/Cu (BDC) as the catalyst[a]



		Product 4 (a-l)		Product 5 (a-l)		Product 6 (a-l)		
Entry	R	Time (min)	Yield%[b]	Time (min)	Yield%	R′	Time (min)	Yield%[b]
1	*p*-Cl	10	96	10	94	4-F	10	98
2	H	10	94	10	92	H	10	97
3	3,4-Cl$_2$	10	90	10	91	*p*-CH$_3$O	10	93
4	*p*-Me	10	92	10	89	*p*-OMe	10	91
5	2-F	15	87	10	85	*p*-Cl-2-F	13	86
6	*p*-OH	15	84	20	84	*p*-CH$_3$O	13	90
7	*p*-CN	10	91	10	90	*p*-OMe	10	93
8	3-NO$_2$	10	93	10	88	H	10	91
9	2-Me	15	86	20	83	H	13	92
10	*p*-Br	10	91	10	93	3-Cl-*p*-CH$_3$	10	94
11	2-Me	15	83	20	86	H	13	91
12	*p*-OMe	10	92	10	90	H	10	92

[a] Isolated yield.

[b] Reaction conditions: dimedone (1 mmol), aryl amines/2-naphthol/urea(1 mmol) and aldehydes (1 mmol) in the.
Presence of CoFe$_2$O4@OCMC/Cu (BDC) in ethanol under ultrasonic irradiation

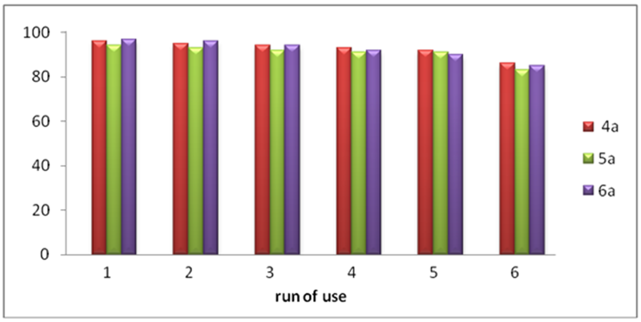
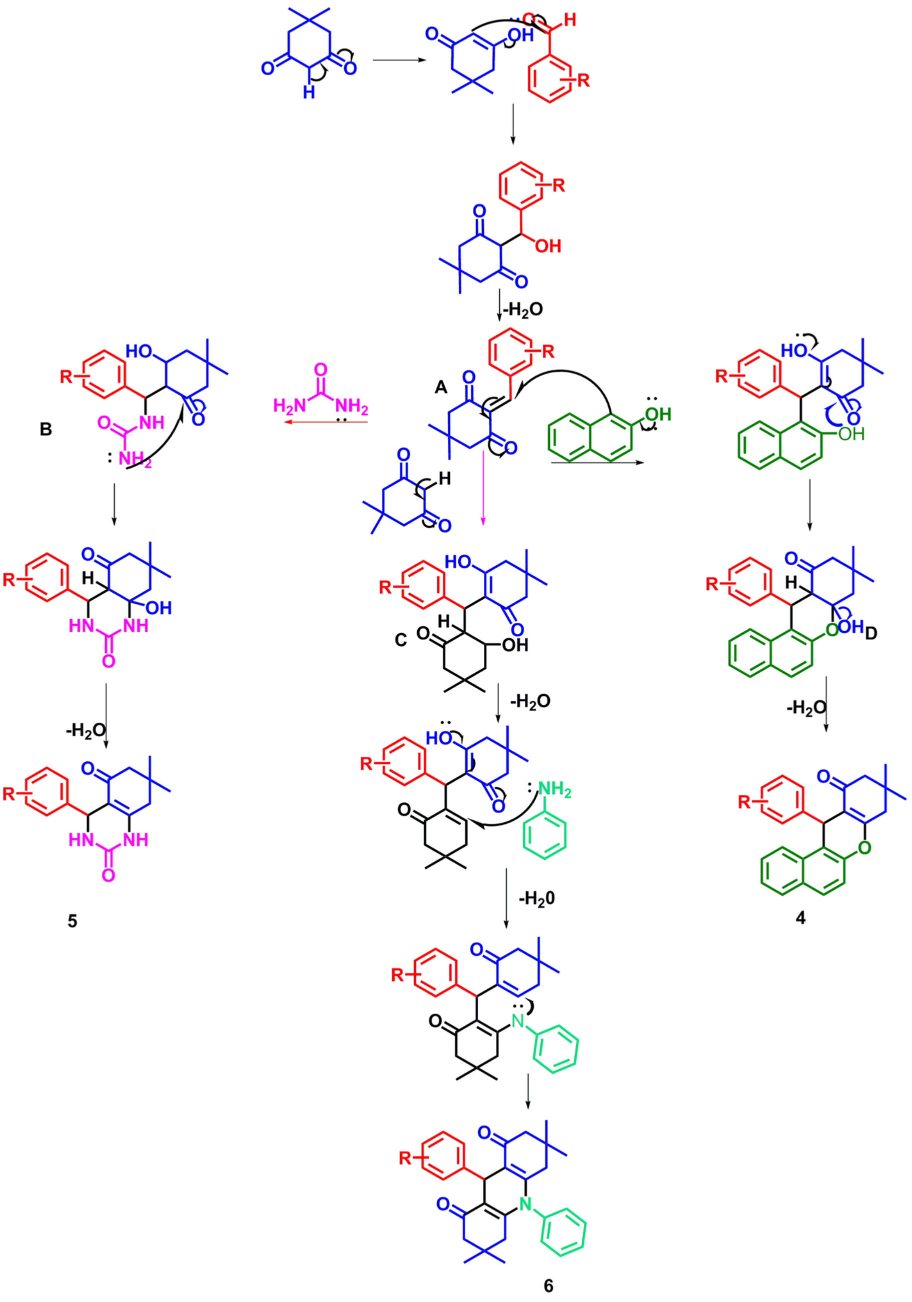


FIGURE 6 The reusability of $CoFe_2O_4$/OCMC/Cu (BDC) as a catalyst in the synthesis of xanthene (4a), quinazolinone (5a), acridine (6a) in model reaction

and 0.002 g were chosen as the optimal amounts of the catalyst for the preparation of 5a, 4a and 6a respectively.

With the optimized reaction conditions in our hand, evaluation of the catalyst efficiency was investigated for the reaction of a variety of aryl aldehydes, dimedone and urea/2-naphthol/amines to produce of acridine, xanthene and quinazolinone derivatives with good to excellent yields (Table 3). Table 3 shows that the aldehydes bearing electron withdrawing groups lead to products with higher yields with shorter reaction time. On the other hand, the aldehydes with electron donating groups exhibit lower efficiency with longer reaction time.



SCHEME 3 The reasonable mechanism for the synthesis of xanthenes (4), quinazolinones (5), and acridines (6) in the presence of $CoFe_2O_4$/OCMC/Cu (BDC) catalyst

3.2 | Recyclability of catalyst

Eventually, the reusability of theCoFe$_2$O$_4$/OCMC/Cu (BDC)-MOF was studied under the optimized reaction conditions. After completion of the reaction, the catalyst was easily separated from the reaction mixture by an external magnet and was reused in the three model reactions shown in Figure 6. It is clear that the catalyst can be used six times without a significant loss in activity performance (Figure 6).

A plausible mechanism for the preparation of octahydroquinazolinones, 1,8-dioxo decahydroacridines, and tetrahydrobenzo[a]xanthen-11-ones using CoFe$_2$O$_4$/OCMC/Cu (BDC) is shown in Scheme 3. It is presumed that CoFe$_2$O$_4$ and Cu (BDC) acts as Lewis acids which increase the electrophilicity of the carbonyl groups of the dimedone and aldehyde through a strong coordination bond.[40,41] The first step is assumed to be a Knoevenagel condensation between the aldehyde and dimedone to generate the adduct A, which can act as a Michael acceptor. Then urea/amine and 2-naphthol attack to intermediate A in a Michael-type reaction to produce an open chain intermediate B,C,D. Finally, the intermediates undergo intramolecular cyclization using the nucleophilic attack followed by dehydration to form the products.

4 | CONCLUSIONS

In conclusion, we have demonstrated that CoFe$_2$O$_4$/OCMC/Cu (BDC) nanocatalyst is an effective catalyst for the MCRs of xanthenes, quinazolines and acridines using aromatic aldehydes, dimedone, aryl amines/2-naphthol/urea at room temperature under ultrasonic irradiation conditions. The heterocyclic compounds including tetrahydrobenzo[a]xanthen-11-ones, octahydroquinazolinones and 1,8-dioxo-decahydroacridines were obtained in high yields. The catalyst can be recovered and reused up to six times without substantial loss of its reactivity for the synthesis of the corresponding products. The one-pot nature and the use of eco-friendly heterogeneous catalyst could make this reported methodology an interesting alternative for various multi-step approaches.

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REFERENCES

[1] A. Maleki, *Ultrason. Sonochem.* **2018**, *40*, 460.

[2] A. Maleki, *Tetrahedron* **2012**, *68*, 7827.

[3] C. Cabrele, O. Reiser, *J. Org. Chem.* **2016**, *81*, 10109.

[4] A. Hollis, Z. Ahmed, *New Engl. J. Med.* **2013**, *369*, 2474.

[5] (a)A. Witt, J. Bergman, *Curr. Org. Chem.* **2003**, *7*, 659. (b) S. B. Mhaske, N. P. Argade, *Tetrahedron* **2006**, *62*, 9787.

[6] (a)D. J. Connolly, D. Cusack, T. P. O'Sullivan, P. J. Guiry, *Tetrahedron* **2005**, *61*, 10153. (b)Z. Ma, Y. Hano, T. Nomura, *Heterocycles* **2005**, *65*, 2203.

[7] M. C. Fuente, D. Domínguez, *Tetrahedron* **2004**, *60*, 10019.

[8] Y. L. Chen, C. M. Lu, I. L. Chen, L. T. Tsao, J. P. Wang, *J. Med. Chem.* **2002**, *53*, 4689.

[9] G. Alang, G. Kaur, R. Kaur, A. Singh, R. Tiwari, *J. Young Pharm.* **2010**, *2*, 394.

[10] A. S. Gamage, A. J. Spicer, J. G. Atwell, J. G. Finlay, C. B. Baguley, A. W. Denny, *J. Med. Chem.* **1999**, *42*, 2383.

[11] N. Srividya, P. Ramamurthy, P. Shanmugasundaram, V. T. Ramakrishnan, *J. Org. Chem.* **1996**, *61*, 5083.

[12] M. Gensicka-Kowalewska, G. Cholewinski, K. Dzierzbicka, *RSC Adv.* **2017**, *7*, 157.

[13] C. Pereira, A. M. Pereira, C. Fernandes, M. Rocha, R. Mendes, M. P. Fernández-García, A. Guedes, P. B. Tavares, J.-M. Grenèche, J. O. P. Araújo, *Chem. Mater.* **2012**, *24*, 1496.

[14] M. -L. Hu, V. Safarifard, E. Doustkhah, S. Rostamnia, A. Morsali, N. Nouruzi, S. Beheshti, K. Akhbari, *Micropor. Mesopor. Mater.* **2018**, *256*, 111.

[15] Y. Li, R. T. Yang, *Langmuir* **2007**, *23*, 12937.

[16] B. Li, H.-M. Wen, W. Zhou, B. Chen, *J. Phys. Chem. Lett.* **2014**, *5*, 3468.

[17] K. Tan, S. Zuluaga, E. Fuentes, E. C. Mattson, J.-F. Veyan, H. Wang, J. Li, T. Thonhauser, Y. J. Chabal, *Nat. Commun.* **2016**, *7*, 13871.

[18] V. Pillai, D. Shah, *J. Magn. Magn. Mater.* **1996**, *163*, 243.

[19] P. Horcajada, T. Chalati, C. Serre, B. Gillet, C. Sebrie, T. Baati, J. F. Eubank, D. Heurtaux, P. Clayette, C. Kreuz, *Nat. Mater.* **2010**, *9*, 172.

[20] H. Zheng, Y. Zhang, L. Liu, W. Wan, P. Guo, A. M. Nyström, X. Zou, *J. Am. Chem. Soc.* **2016**, *138*, 962.

[21] G. Lu, J. T. Hupp, *J. Am. Chem. Soc.* **2010**, *132*, 7832.

[22] I. Stassen, B. Bueken, H. Reinsch, J. Oudenhoven, D. Wouters, J. Hajek, V. Van Speybroeck, N. Stock, P. Vereecken, R. VanSchaijk, *Chem. Sci.* **2016**, *7*, 5827.

[23] S. Rostamnia, F. Mohsenzad, *Mol. Catal.* **2018**, *445*, 12.

[24] B. Salahshournia, H. Hamadi, V. Nobakht, *Appl. Organomet. Chem.* **2018**, *32*, e4416.

[25] A. Rossin, G. Tuci, L. Luconi, G. Giambastiani, *ACS Catal.* **2017**, *7*, 5035.

[26] X. Zhang, Z. Zhang, J. Boissonnault, S. M. Cohen, *Chem. Commun.* **2016**, *52*, 8585.

[27] S. Rostamnia, H. Alamgholiloo, M. Jafari, R. Rookhosh, A. R. Abbasi, *Appl. Organomet. Chem.* **2016**, *30*, 954.

[28] S. Rostamnia, H. Alamgholiloo, X. Liu, *J. Colloid Interface Sci.* **2016**, *469*, 310.

[29] Z. Zhang, J. Chen, Z. Bao, G. Chang, H. Xing, Q. Ren, *RSC Adv.* **2015**, *5*, 79355.

[30] A. Dhakshinamoorthy, M. Alvaro, H. Chevreau, P. Horcajada, T. Devic, C. Serre, H. Garcia, *Catal. Sci. Technol.* **2012**, *2*, 324.

[31] S. Li, J. Pan, X. Wu, Y. Fu, Q. Xiao, F. Zhang, W. Zhu, *Appl. Organomet. Chem.* **2019**, *33*, e5233. https://doi.org/10.1002/aoc.5233

[32] H. Alamgholiloo, S. Rostamnia, A. Hassankhani, J. Khalafy, M. M. Baradarani, G. Mahmoudi, X. Liu, *Appl. Organomet. Chem.* **2018**, *32*, e4539.

[33] B. Mirhosseini-Eshkevari, M. A. Ghasemzadeh, M. B. Esnaashari, *Appl. Organomet. Chem.* **2019**, *33*, e5027.

[34] Z. Mahmoudi, M. A. Ghasemzadeh, H. Kabiri-Fard, *J. Mol. Struct.* **2019**, *1194*, 1.

[35] I. Luz, F. X. Llabrés i Xamena, A. Corma, *J. Catal.* **2012**, *285*, 285.

[36] G. H. Dang, Y. T. H. Vu, Q. A. Dong, D. T. Le, T. Truong, N. T. S. Phan, *Appl. Catal. A-Gen.* **2015**, *491*, 189.

[37] G. H. Dang, H. Q. Lam, A. T. Nguyen, D. T. Le, T. Truong, N. T. S. Phan, *J. Catal.* **2016**, *337*, 167.

[38] M. A. Ghasemzadeh, B. Mirhosseini-Eshkevari, M. H. Abdollahi-Basir, *Appl. Organomet. Chem.* **2019**, *33*, e4679.

[39] F. Ghaffarian, M. A. Ghasemzadeh, S. S. Aghaei, *J. Mol. Struct.* **2019**, *1186*, 204.

[40] D. Chanda, J. Hnat, M. Paidar, K. Bouzek, *Int. J. Hydrogen Energy* **2014**, *39*, 5713.

[41] L. M. Aguirre-Díaz, F. Gándara, M. Iglesias, N. Snejko, E. Gutiérrez-Puebla, M. Á. Monge, *J. Am. Chem. Soc.* **2015**, *137*, 6132.

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