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Magnetic resorcinol—formaldehyde supported isatin-Schiff-base/Fe as a green and reusable nanocatalyst for the synthesis of pyrano[2,3-d] pyrimidines

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Herein, a novel magnetic resorcinol–formaldehyde-supported isatin-Schiff-base/Fe complex (Fe $_3O_4$ @RF-ISB/Fe) is prepared and characterized and its catalytic performance is investigated in the synthesis of pyrano [2,3-d]pyrimidines. The Fe $_3O_4$ @RF-ISB nanomaterial was prepared through the chemical immobilization of (3-aminopropyl)trimethoxysilane over the Fe $_3O_4$ @RF composite, followed by treatment with isatin. The Fe $_3O_4$ @RF-ISB was then reacted with FeCl $_3\cdot$ 6H $_2O$ to afford the Fe $_3O_4$ @RF-ISB/Fe nanocatalyst. Characterization through FT-IR, EDX, PXRD, VSM, SEM and ICP techniques confirmed that the magnetite surface was successfully modified with RF/ISB-Fe while preserving its crystalline structure. The SEM image revealed spherical particles with an average size of 44 nm for the designed nanocomposite. Various aromatic aldehydes were used as substrates in the presence of 0.01 g of Fe $_3O_4$ @RF-ISB/Fe to give the corresponding pyranopyrimidines in high yields (88–95%) within short reaction times (30–55 minutes) at RT. The designed magnetic catalyst maintained its activity for nine runs.

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Introduction

Magnetic nanocomposites play a vital role in numerous fields, such as catalysis, drug delivery, energy harvesting, plasmonics, and magnetic resonance imaging (MRI).1-6 In these nanocomposites, the high chemical reactivity of magnetic nanoparticles (MNPs) can be effectively controlled by a chemically inert shell which may consist of materials such as silica, alumina, polymers, or carbon. Recent developments in this area include Mag@mSiO₂/IL-Cu,⁷ Fe₃O₄@C-TiO₂-Ag,⁸ Fe₃O₄@-SiO₂@La₂O₃, Mo₇₂V₃₀@Fe₃O₄/C¹⁰ and Fe₃O₄/ZIF-8/TiO₂. 11 Among the different types of shells, resorcinol-formaldehyde (RF) resin has gained significant interest due to its distinctive properties. These include a hydrophobic inner framework, a surface rich in phenolic hydroxyl groups that facilitates the accumulation of reactants near the catalytic active sites, and advantages such as high biocompatibility, non-toxicity, and cost-effectiveness. 12-16 Therefore, coating magnetic nanoparticles with an RF shell not only enhances their chemical stability and prevents aggregation but also utilizes the distinctive hydrophobic and biocompatible properties of RF, thereby making these nanocomposites particularly effective for catalytic applications.

Meanwhile, Schiff-base ligands have garnered significant attention in the chemical and materials sciences due to their

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efficient complexation with a wide variety of transition metal ions, excellent solubility, and superior catalytic performance. Furthermore, Schiff-base ligands are used to improve catalytic efficiency by linking the catalytic centers to solid supports. ^{17–23} Recent studies in this area include reports such as Fe_3O_4 @-BOS@SB/In, ²⁴ Cu/SB- Fe_3O_4 , ²⁵ and $[M^{II}(L)(Cl)(H_2O)_2] \cdot H_2O$, ²⁶ BPMO@ISB/Mn(II). ²⁷

Recently, multicomponent reactions (MCRs) owing to their simple operation, atom economy and high efficiency, have become a crucial tool in organic synthesis and medicinal chemistry, attracting significant attention from chemists. 28,29 Pyrano[2,3-d]pyrimidines, prepared through multicomponent reactions, exhibit notable biological activities, including anticancer, antispasmodic, antianaphylactic, and anticoagulant properties. Therefore, the advancement of methods for their effective synthesis remains a highly significant area among chemists. Despite the establishment of various methods for synthesizing pyrano[2,3-d]pyrimidines, these techniques have significant limitations, such as high catalyst loadings, low yields, the use of expensive ligands, complex work-ups, difficulties in separating products from catalysts, and the use of hazardous solvents.30-32 Hence, there is a critical need for the development of a more environmentally friendly, efficient, straightforward, and cost-effective method for synthesizing pyrano[2,3-d]pyrimidines.

Considering all these aspects, in this work, a novel, and environmentally friendly magnetic nanocatalyst supported isatin Schiff-base/Fe complex, denoted as Fe₃O₄@RF-ISB/Fe, is

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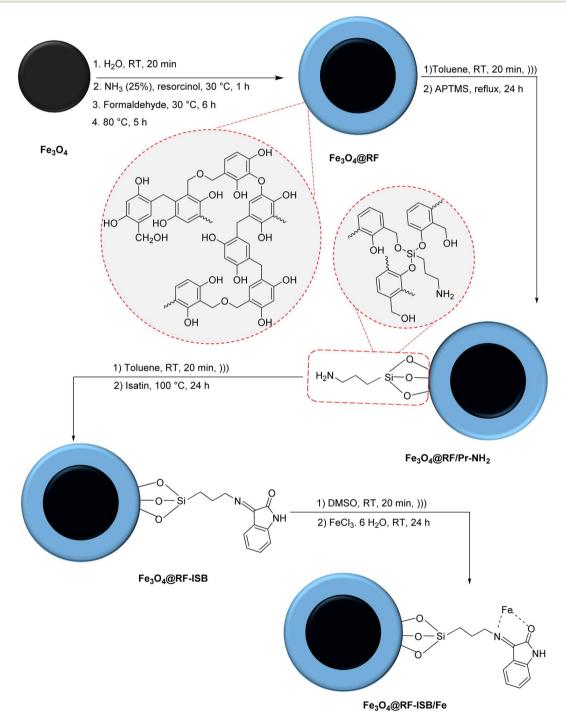
prepared, characterized and applied as a effective catalyst for the green synthesis of pyrano[2,3-d]pyrimidines.

2. **Experimental section**

Synthesis of Fe₃O₄@RF

Initially, Fe₃O₄ nanoparticles (NPs) were synthesized using a method described in our previous work.33 After that, 150 mL of deionised (DI) water was used to disperse the 1 g of Fe₃O₄ NPs.

Following 10 minutes of ultrasonic treatment, ammonia (0.3 mL, 25 wt%) and resorcinol (0.4 g) were introduced into the solution, which was then was agitated at 30 °C for 1 hour. Following a dropwise addition of 0.6 mL of formaldehyde, the mixture was constantly stirred for 6 hours. After that, the mixture was heated for five hours at 80 °C to complete the procedure. After collecting the magnetic material, it was thoroughly rinsed with EtOH and deionized (DI) water and dried at 70 °C for 6 h.34



Scheme 1 Preparation of the Fe₃O₄@RF-ISB/Fe nanocatalyst.

2.2 Synthesis of Fe₃O₄@RF/Pr-NH₂

To achieve this, 1 g of Fe₃O₄(a)RF nanoparticles was dispersed in 25 mL of toluene at 25 °C for 20 minutes. After adding 0.26 g of (3-aminopropyl)trimethoxysilane (APTMS), the mixture was refluxed at 100 °C for 24 hours. The resulting magnetic material was subsequently collected, rinsed with ethanol and DI water, and dried at 70 °C for 6 hours.

Synthesis of Fe₃O₄@RF-ISB

For this, 1 g of Fe₃O₄@RF/Pr-NH₂ was fully dispersed in 20 mL of toluene for 20 minutes. Subsequently, 0.6 g of isatin was introduced into the reaction vessel, and the mixture was agitated and refluxed for 24 hours. The resulting product was further subjected to magnetic separation, rinsed with ethanol and DI water, and dried in an oven at 60 °C for 12 hours.

2.4 Synthesis of Fe₃O₄@RF-ISB/Fe

Initially, 1 g of Fe₃O₄@RF-ISB was fully dispersed in 20 mL of DMSO for 20 minutes. Then, the reaction vessel was agitated at room temperature for 24 hours after adding 0.2 g of FeCl₃-·6H₂O. After being separated using a magnet, the final product was washed with ethanol and DI water and then dried for 12 hours at 60 °C. The ICP analysis was performed on both Fe₃O₄@RF-ISB and Fe₃O₄@RF-ISB/Fe. The results indicated that the Fe loading onto the material framework was 0.14 mmol Fe per g.

2.5 Procedure for the synthesis of pyrano[2,3-d]pyrimidines using the Fe₃O₄@RF-ISB/Fe nanocatalyst

To carry out the reaction, aldehyde (1 mmol), malononitrile (1 mmol), barbituric acid (1 mmol), and 0.01 g of Fe₃O₄@RF-ISB/ Fe were added to a reaction vessel containing EtOH (10 mL) and stirred at RT. Upon completion, monitored by TLC, the Fe₃O₄@RF-ISB/Fe catalyst was removed using an external magnet. The pure products were obtained by recrystallization from EtOH.

3. Results and discussion

Characterization of Fe₃O₄@RF-ISB/Fe

Scheme 1 illustrates the synthesis pathway for the Fe₃O₄@RF-ISB/Fe nanocomposite. Initially, Fe₃O₄@RF NPs were synthesized via the interface polymerization of resorcinol and formaldehyde on Fe₃O₄ NPs under alkaline conditions. The RF shell was then chemically modified with (3-aminopropyl)trimethoxysilane (APTMS), resulting in Fe₃O₄@RF/Pr-NH₂. This nanomaterial was subsequently reacted with isatin to afford Fe₃O₄@RF-ISB. Finally, the Fe₃O₄@RF-ISB support was treated with FeCl₃·6H₂O to yield the Fe₃O₄@RF-ISB/Fe nanocomposite.

Fig. 1 displays the FTIR spectra of the synthesized materials, including Fe₃O₄@RF (a), Fe₃O₄@RF/Pr-NH₂ (b), Fe₃O₄@RF-ISB (c), and Fe₃O₄@RF-ISB/Fe (d). For all samples, the FTIR analvsis revealed a characteristic absorption peak for the Fe-O bond at 588 cm⁻¹ and a stretching vibration of the O-H bond at 3419 cm⁻¹. The absorption bonds appeared at 2857–2932 and

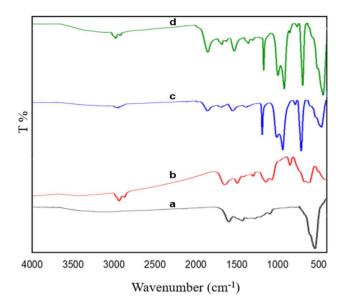


Fig. 1 The FTIR spectra of Fe₃O₄@RF (a), Fe₃O₄@RF/Pr-NH₂ (b), Fe_3O_4 @RF-ISB (c), and Fe_3O_4 @RF-ISB/Fe (d).

1450 cm⁻¹, corresponding to the aliphatic C-H stretching and bending vibrations of the propyl amines and RF resin, respectively. Additionally, the signals at 1021 and 1110 cm⁻¹ are attributed to the C-O-C methylene ether bridges between resorcinol units.35,36 The presence of the Si-O-Si signals at 806 and 1029 cm⁻¹ supported the successful surface modification of Fe₃O₄@RF with (3-aminopropyl)trimethoxysilane (Fig. 1b-d). Furthermore, the peak at 1621 cm $^{-1}$ is attributed to the C=N bond, indicating that the isatin-Schiff-base (ISB) ligand was successfully formed on the material surface (Fig. 1c and d).37

Powder X-ray diffraction (PXRD) analysis was conducted to examine the crystallinity of the Fe₃O₄@RF-ISB/Fe nanocomposite. As shown in Fig. 2, the wide-angle PXRD pattern reveals six peaks at 2θ values of 30.4°, 35.82°, 43.03°, 54.22°, 57.36°, and 62.98°, corresponding to the (220), (311), (400), (422), (511), and (440) crystal planes, respectively. These observations are consistent with the PXRD pattern of Fe₃O₄,

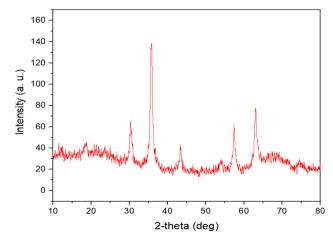


Fig. 2 The PXRD pattern of the Fe₃O₄@RF-ISB/Fe.

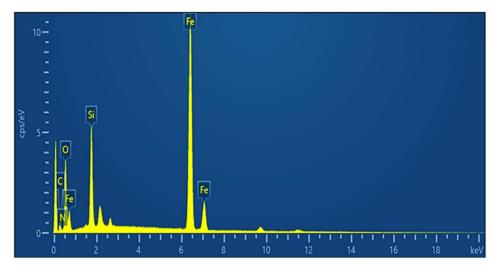


Fig. 3 The EDX spectrum of Fe₃O₄@RF-ISB/Fe.

indicating that these NPs retain high stability throughout the modification processes.^{38,39}

Fig. 3 shows the EDX spectrum of the $Fe_3O_4@RF\text{-}ISB/Fe$ catalyst. The EDX spectrum confirms the presence of C, N, O, Si, and Fe in the sample. This result is in accordance with the FTIR analysis, confirming the effective incorporation and immobilization of the expected species in the $Fe_3O_4@RF\text{-}ISB/Fe$ nanocomposite.

The magnetic properties of Fe₃O₄@RF and Fe₃O₄@RF-ISB/Fe nanomaterials were investigated using a vibrating sample magnetometer (VSM, Fig. 4). The saturation magnetization values obtained were 50 and 39.8 emu g⁻¹, respectively, for Fe₃O₄@RF and Fe₃O₄@RF-ISB/Fe. The observed decrease in magnetic properties after modification is attributed to the successful immobilization of the isatin-Schiff-base/Fe complex on the Fe₃O₄@RF NPs.

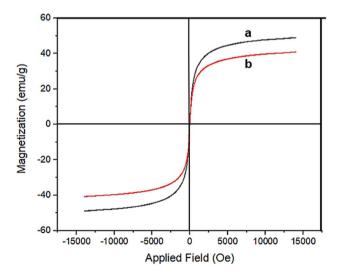


Fig. 4 The VSM diagrams of Fe $_3O_4$ @RF (a) and Fe $_3O_4$ @RF-ISB/Fe (b) nanocomposites.

The SEM analysis was conducted to examine the morphology of the Fe₃O₄@RF-ISB/Fe nanocomposite (Fig. 5). The analysis revealed that the catalyst exhibits a spherical morphology with an average particle size of approximately 44 nm.

3.2 Catalytic activity of the Fe_3O_4 @RF-ISB/Fe nanocomposite

After characterization, the performance of the Fe₃O₄@RF-ISB/ Fe catalyst was investigated in the synthesis of pyrano[2,3-d] pyrimidines. The condensation between barbituric acid, benzaldehyde and malononitrile at room temperature (RT) was selected as a test model to optimize the reaction conditions. By evaluating the effect of several parameters, such as catalyst amount and solvent, the optimal conditions were determined (Table 1). Initially, the effect of solvent-free conditions, as well as the use of H₂O and EtOH solvents, was investigated (Table 1, entries 1-3). The results demonstrated that the highest yield of 95% was observed with EtOH (Table 1, entry 3). The reaction progress was also affected by the amount of catalyst, and 0.01 g of Fe₃O₄@RF-ISB/Fe provided the best result (Table 1, entry 3). Interestingly, there was no noticeable difference in the product yield when the catalyst amount increased to 0.015 g (Table 1, entry 5). It should be noted that no product was observed in the absence of the catalyst (Table 1, entry 6). To demonstrate the role of supported-Fe centers in the catalytic process, the reaction was performed in the presence of Fe-free Fe₃O₄@RF and Fe₃O₄@RF-ISB materials under identical reaction conditions (Table 1, entries 7 and 8). Interestingly, only a low yield of the desired product was obtained in the latter cases. These findings indicate that the supported-Fe species play a major role in catalyzing the reaction. In addition, the catalytic activity of Fe₃O₄@RF-ISB/Fe was compared with that of FeCl₃·6H₂O salt (Table 1, entry 3 vs. entry 9). The result indicates that the reaction achieves a satisfactory conversion using FeCl₃·6H₂O. This result confirms that the catalytic activity of Fe₃O₄(a)RF-ISB/ Fe is comparable to that of the homogeneous FeCl₃·6H₂O catalyst, as both systems achieve similar yields. This finding

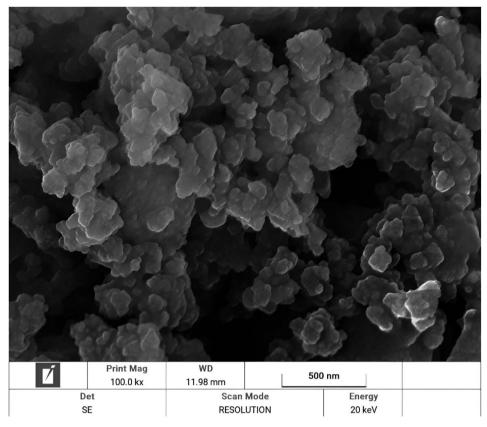


Fig. 5 The SEM image of the $Fe_3O_4@RF$ -ISB/Fe nanocomposite.

Table 1 Effect of different parameters in the synthesis of pyranopyrimidines

Entry	Solvent	Catalyst	Catalyst loading (g)	Yield (%)	
1 —		Fe ₃ O ₄ @RF-ISB/Fe	0.01	86	
2	H_2O	Fe ₃ O ₄ @RF-ISB/Fe	0.01	76	
3	EtOH	Fe ₃ O ₄ (a)RF-ISB/Fe	0.01	95	
4	EtOH	Fe ₃ O ₄ @RF-ISB/Fe	0.005	75	
5	EtOH	Fe ₃ O ₄ @RF-ISB/Fe	0.015	96	
6	EtOH	_	_	_	
7	EtOH	Fe ₃ O ₄ @RF	0.01	15	
8	EtOH	Fe ₃ O ₄ @RF-ISB	0.01	14	
9	EtOH	$FeCl_3 \cdot 6H_2O$	0.004	96	
9	EtOH	$FeCl_3 \cdot 6H_2O$	0.004	96	

clearly demonstrates that the supported Fe(III) species exhibit high efficiency, closely resembling the catalytic behavior of free Fe(III) ions, while also providing significant advantages in terms of recoverability and reusability. Accordingly, the following

optimal parameters were chosen: room temperature, EtOH as the solvent, and 0.01 g of the $Fe_3O_4@RF$ -ISB/Fe catalyst.

Following condition optimization (Table 1, entry 3), various aldehyde derivatives were employed as substrates. As illustrated in Table 2, the corresponding pyranopyrimidines were formed

Table 2 Synthesis of pyranopyrimidines using Fe₃O₄@RF-ISB/Fe

Aldehyde	Time (min)	Yield (%)	Mp found (°C)	Mp reported (°C)
PhCHO	40	95	219-221	222-224 (ref. 40)
4-Cl-PhCHO	35	91	240-242	242-244 (ref. 41)
4-Me-PhCHO	50	88	227-229	226-227 (ref. 42)
4-MeO-PhCHO	55	90	268-270	268–271 (ref. 40)
3-NO ₂ -PhCHO	35	94	265-267	265 (ref. 41)
4-Br–PhCHO	30	93	230-232	235-236 (ref. 42)
-	PhCHO 4-Cl-PhCHO 4-Me-PhCHO 4-MeO-PhCHO 3-NO ₂ -PhCHO	PhCHO 40 4-Cl-PhCHO 35 4-Me-PhCHO 50 4-MeO-PhCHO 55 3-NO ₂ -PhCHO 35	PhCHO 40 95 4-Cl-PhCHO 35 91 4-Me-PhCHO 50 88 4-MeO-PhCHO 55 90 3-NO ₂ -PhCHO 35 94	PhCHO 40 95 219-221 4-Cl-PhCHO 35 91 240-242 4-Me-PhCHO 50 88 227-229 4-MeO-PhCHO 55 90 268-270 3-NO ₂ -PhCHO 35 94 265-267

in high yields from all types of aldehydes. The remarkable performance of the Fe₃O₄@RF-ISB/Fe catalyst in the synthesis of various derivatives of pharmacologically active pyranopyrimidines was confirmed by the fact that electronic characteristics and the substituent positions had a little effect on this process.

In the following investigation, the recoverability and reusability of the Fe₃O₄@RF-ISB/Fe catalyst were evaluated. After completing the reaction, the Fe₃O₄@RF-ISB/Fe catalyst was magnetically separated and reused under the same conditions as the initial run. The results indicated that the Fe₃O₄@RF-ISB/ Fe catalyst retained its effectiveness for at least nine cycles, highlighting its remarkable durability under the optimized conditions (Fig. 6).

A leaching test was performed to investigate the nature of the Fe₃O₄@RF-ISB/Fe catalyst under the applied conditions. During the test, the catalyst was removed from the reaction mixture after reaching a 50% conversion. The reaction of the residue

was then allowed to continue for 60 min. The result demonstrated no significant increase in conversion, confirming the heterogeneous nature of the designed catalyst. Additionally, the ICP analysis proved the absence of Fe species in the reaction vessel confirming no leaching of the active Fe catalyst under the applied conditions. These results demonstrate the successful immobilization and high stability of the active Fe species, which is attributed to the Schiff-base ligand, preventing Fe-leaching during the reaction.

The PXRD analysis of the recovered Fe₃O₄@RF-ISB/Fe revealed six peaks at $2\theta = 30.48^{\circ}$, 35.75°, 43.2°, 54.2°, 57.32°, and 63.05° (Fig. 7), which aligns well with the PXRD pattern of the fresh nanocatalyst. This confirms the exceptional stability of the crystalline structure of magnetite nanoparticles even after nine cycles of recovery and reuse.

A proposed mechanism for the synthesis of pyrano[3,2-d] pyrimidine derivatives catalyzed by Fe₃O₄@RF-ISB/Fe is presented in Scheme 2. The initial step involves forming intermediate 1 through a Knoevenagel condensation between malononitrile and the Fe-activated aldehyde. Subsequently, as

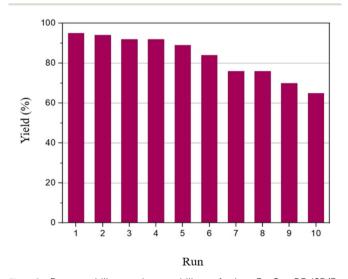


Fig. 6 Recoverability and reusability of the Fe₃O₄@RF-ISB/Fe nanocatalyst.

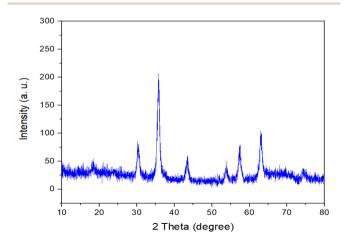


Fig. 7 The PXRD pattern of the recovered Fe₃O₄@RF-ISB/Fe nanocatalyst.

CN Fe₃O₄@RF-ISB/Fe -H₂O

Scheme 2 A plausible mechanism for the synthesis of pyrano[2,3-d]pyrimidines using Fe₃O₄@RF-ISB/Fe.

Table 3 Comparative study of the efficiency of Fe₃O₄@RF-ISB/Fe with the former catalysts in the synthesis of pyrano[2,3-d]pyrimidines

Entry	Catalyst	Conditions	Time	Recovery times	Ref.
1	$[\mathrm{BMIm}]\mathrm{BF_4}$	Cat. (1.5 g), solvent-free, 90 °C	180 min	4	43
2	Fe ₃ O ₄ @cellulose	Cat. (5 mg), H ₂ O, 25 °C	35 min	5	30
3	L-Proline	Cat. (5 mol%), EtOH, 25 °C	40 min	_	43
4	Fe ₃ O ₄ @RF-ISB/Fe	Cat. (0.01 g), EtOH, 25 °C	40 min	9	This work

a Michael acceptor, this intermediate reacts with the enol form of barbituric acid to produce intermediate 2. Next, intermediate 2 undergoes an intramolecular cyclization to form intermediate 3. Finally, the desired product 4 is obtained through a tautomerization process.

The performance of the Fe_3O_4 @RF-ISB/Fe catalyst was compared with that of various catalytic systems previously reported for the synthesis of pyrano[2,3-d]pyrimidines (Table 3).

The study showed that the present catalyst outperforms previously reported catalytic systems in terms of mild reaction conditions, reaction rates, and recovery times.

4. Conclusion

In summary, a novel magnetic RF modified with an isatin-Schiff-base/Fe (Fe₃O₄@RF-ISB/Fe) complex was successfully

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designed, prepared, characterized and applied as a powerful catalyst. The FT-IR and EDX analyses clearly indicated that the RF moieties were successfully coated/immobilized on the magnetic NPs. The PXRD analysis further confirmed that the crystalline structure of the Fe₃O₄ NPs remains highly stable throughout the modification process. Additionally, the SEM image of Fe₃O₄@RF-ISB/Fe revealed well-defined spherical particles of the nanomaterial. The Fe₃O₄@RF-ISB/Fe nanocomposite was effectively employed as a robust and highly recoverable catalyst for the synthesis of pyrano[2,3-d]pyrimidines, providing the desired products in high yields and short reaction times under mild conditions. Due to the advantages of the Fe₃O₄@RF-ISB/Fe nanocatalyst, including simple preparation, high stability, and ease of recovery and separation, this catalyst exhibits significant potential for use in various important organic reactions, such as other multicomponent processes, C-H activation, nucleophilic substitution of benzylic C-X bonds, C-N bond formation, and so on.

Data availability

All data and materials are included in the manuscript.

Conflicts of interest

There are no conflicts to declare.

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References

- 1 P. D. Tomke and V. K. Rathod, Facile fabrication of silver on magnetic nanocomposite (Fe₃O₄@chitosan-AgNP nanocomposite) for catalytic reduction of anthropogenic pollutant and agricultural pathogens, *Int. J. Biol. Macromol.*, 2020, **149**, 989-999.
- 2 Y. Huang, L.-c. Nengzi, X. Zhang, J. Gou, Y. Gao, G. Zhu, Q. Cheng and X. Cheng, Catalytic degradation of ciprofloxacin by magnetic CuS/Fe₂O₃/Mn₂O₃ nanocomposite activated peroxymonosulfate: influence factors, degradation pathways and reaction mechanism, *Chem. Eng. I.*, 2020, 388, 124274.
- 3 M. Abd El-Aal, A. E.-A. A. Said, M. N. Goda, E. F. A. Zeid and S. M. Ibrahim, Fe₃O₄@CMC-Cu magnetic nanocomposite as an efficient catalyst for reduction of toxic pollutants in water, *J. Mol. Liq.*, 2023, **385**, 122317.
- 4 S. Naghash-Hamed, N. Arsalani and S. B. Mousavi, The catalytic performance of CuFe₂O₄@CQD nanocomposite as a high-perform heterogeneous nanocatalyst in nitroaniline group reduction, *Sci. Rep.*, 2023, **13**, 3329.
- 5 A. Adam and D. Mertz, Iron oxide@mesoporous silica coreshell nanoparticles as multimodal platforms for magnetic resonance imaging, magnetic hyperthermia, near-infrared

light photothermia, and drug delivery, *Nanomaterials*, 2023, 13, 1342.

- 6 A. F. Oliveira and E. M. B. de Sousa, Synthesis and characterization of MSN/Fe₃O₄/Gd₂O₃ nanocomposite as theranostic systems, *J. Nanopart. Res.*, 2023, 25, 115.
- 7 F. Mousavi, D. Elhamifar and S. Kargar, Copper/IL-containing magnetic nanoporous MCM-41: a powerful and highly stable nanocatalyst, *Surf. Interfaces*, 2021, **25**, 101225.
- 8 A. N. Chishti, Z. Ma, Y. Liu, M. Chen, J. Gautam, F. Guo, L. Ni and G. Diao, Synthesis of highly efficient and magnetically separable Fe₃O₄@C-TiO₂-Ag catalyst for the reduction of organic dyes and 4-nitrophenol, *Colloids Surf.*, A, 2021, 631, 127694.
- 9 J. Song, N. Ma, W. Chen, J. Chen and Q. Dai, Insights into mechanism of catalytic ozonation of cinnamyl alcohol over core–shell Fe₃O₄@SiO₂@La₂O₃ catalyst, *Sep. Purif. Technol.*, 2022, **282**, 119969.
- 10 M. Gong, X.-Y. Wang, M.-Q. Li, W.-X. Mu, Y.-D. Cao, H. Liu, Y.-G. Lv, X.-H. Qi and G.-G. Gao, High-efficient and recoverable Mo₇₂V₃₀@Fe₃O₄/C catalyst for oxidation of hydroxyfurfural, *Fuel*, 2023, 332, 126050.
- 11 H. Kargar, M. Ghahramaninezhad, M. N. Shahrak and S. S. Balula, An Effective Magnetic Catalyst for Oxidative Desulfurization of Model and Real Fuels: Fe₃O₄/ZIF-8/TiO2, *Microporous Mesoporous Mater.*, 2021, 317, 110992.
- 12 L. Yu, P. Pan, Y. Zhang, Y. Zhang, L. Wan, X. Cheng and Y. Deng, Nonsacrificial self-template synthesis of colloidal magnetic yolk–shell mesoporous organosilicas for efficient oil/water interface catalysis, *Small*, 2019, 15, 1805465.
- 13 Q. Yue, J. Li, Y. Zhang, X. Cheng, X. Chen, P. Pan, J. Su, A. A. Elzatahry, A. Alghamdi and Y. Deng, Plasmolysis-inspired nanoengineering of functional yolk-shell microspheres with magnetic core and mesoporous silica shell, *J. Am. Chem. Soc.*, 2017, 139, 15486–15493.
- 14 M. Wang, Y. Ni and A. Liu, Fe $_3O_4$ @resorcinol-formaldehyde resin/Cu $_2O$ composite microstructures: solution-phase construction, magnetic performance, and applications in antibacterial and catalytic fields, *ACS Omega*, 2017, 2, 1505–1512.
- 15 C. Shi, C. Lin, X. Huang, Q. Wu, H. Ge and Y. Yang, Facile synthesis of magnetic resorcinol-formaldehyde resin Fe₃O₄@RF-Au composites for enhanced tetracycline photodegradation with simultaneous H₂O₂ production, *J. Mater. Sci.: Mater. Electron.*, 2024, 35, 1–12.
- 16 Y. Zhao, C. Wang, S. Wang, C. Wang, Y. Liu, A. A. Al-Khalaf, W. N. Hozzein, L. Duan, W. Li and D. Zhao, Magnetic mesoporous TiO₂ microspheres for sustainable arsenate removal from acidic environments, *Inorg. Chem. Front.*, 2018, 5, 2132–2139.
- 17 M. Nikoorazm, A. Ghorbani-Choghamarani and N. Noori, Oxo-vanadium(IV) Schiff base complex supported on modified MCM-41: a reusable and efficient catalyst for the oxidation of sulfides and oxidative S–S coupling of thiols, *Appl. Organomet. Chem.*, 2015, **29**, 328–333.
- 18 N. Amirmahani, N. O. Mahmoodi, M. Malakootian, A. Pardakhty and N. Seyedi, Pd nanoparticles supported on Fe₃O₄@SiO₂-Schiff base as an efficient magnetically

- recoverable nanocatalyst for Suzuki-Miyaura coupling reaction, *Res. Chem. Intermed.*, 2020, **46**, 4595–4609.
- 19 V. K. Juyal, A. Pathak, M. Panwar, S. C. Thakuri, O. Prakash, A. Agrwal and V. Nand, Schiff base metal complexes as a versatile catalyst: a review, *J. Organomet. Chem.*, 2023, 122825.
- 20 A. El-Sonbati, W. Mahmoud, G. G. Mohamed, M. Diab, S. M. Morgan and S. Abbas, Synthesis, characterization of Schiff base metal complexes and their biological investigation, *Appl. Organomet. Chem.*, 2019, 33, e5048.
- 21 G.-l. Liu, S.-f. He, S. Zhang and H. Li, In situ ligand and complex transformation of an iron(III) Schiff base complex: structural evidence and theoretical calculations, *Dalton Trans.*, 2012, **41**, 6256–6262.
- 22 L. Pogány, B. Brachňaková, P. Masárová, J. Moncol, J. Pavlik, M. Gál, M. Mazúr, R. Herchel, I. Nemec and I. Šalitroš, Impact of the Schiff base ligand substituents on the solid state and solution properties of eleven iron(III) complexes, New J. Chem., 2019, 43, 13916–13928.
- 23 D. Elhamifar, D. Elhamifar and F. Shojaeipoor, Synthesis, characterization and catalytic application of a novel polyethylene-supported Fe/ionic liquid complex, *J. Mol. Catal. A: Chem.*, 2017, 426, 198–204.
- 24 R. Mirbagheri and D. Elhamifar, Magnetic ethyl-based organosilica supported Schiff-base/indium: a very efficient and highly durable nanocatalyst, *J. Alloys Compd.*, 2019, **790**, 783–791.
- 25 D. Elhamifar, P. Mofatehnia and M. Faal, Magnetic nanoparticles supported Schiff-base/copper complex: an efficient nanocatalyst for preparation of biologically active 3,4-dihydropyrimidinones, *J. Colloid Interface Sci.*, 2017, **504**, 268–275.
- 26 V. Arun, N. Sridevi, P. P. Robinson, S. Manju and K. K. M. Yusuff, Ni(II) and Ru(II) Schiff base complexes as catalysts for the reduction of benzene, *J. Mol. Catal. A: Chem.*, 2009, **304**, 191–198.
- 27 M. Norouzi and D. Elhamifar, Phenylene and Isatin Based Bifunctional Mesoporous Organosilica Supported Schiff-Base/Manganese Complex: An Efficient and Recoverable Nanocatalyst, Catal. Lett., 2019, 149, 619–628.
- 28 J. Mondal, A. Modak, M. Nandi, H. Uyama and A. Bhaumik, Triazine functionalized ordered mesoporous organosilica as a novel organocatalyst for the facile one-pot synthesis of 2amino-4*H*-chromenes under solvent-free conditions, *RSC* Adv., 2012, 2, 11306–11317.
- 29 S. K. Kundu and A. Bhaumik, A triazine-based porous organic polymer: a novel heterogeneous basic organocatalyst for facile one-pot synthesis of 2-amino-4*H*-chromenes, *RSC Adv.*, 2015, 5, 32730–32739.
- 30 A. Maleki, A. A. Jafari and S. Yousefi, Green cellulose-based nanocomposite catalyst: design and facile performance in aqueous synthesis of pyranopyrimidines and pyrazolopyranopyrimidines, *Carbohydr. Polym.*, 2017, 175, 409–416.
- 31 S. Beheshti, V. Safarifard and A. Morsali, Isoreticular interpenetrated pillared-layer microporous metal-organic

- framework as a highly effective catalyst for three-component synthesis of pyrano[2,3-d]pyrimidines, *Inorg. Chem. Commun.*, 2018, **94**, 80–84.
- 32 T. Farahmand, S. Hashemian and A. Sheibani, Efficient one-pot synthesis of pyrano[2,3-d]pyrimidinone and pyrido[2,3-d]pyrimidine derivatives by using of Mn-ZIF-8@ZnTiO₃ nanocatalyst, *J. Mol. Struct.*, 2020, **1206**, 127667.
- 33 J. H. Bang and K. S. Suslick, Applications of ultrasound to the synthesis of nanostructured materials, *Adv. Mater.*, 2010, 22, 1039–1059.
- 34 X.-B. Zhang, H.-W. Tong, S.-M. Liu, G.-P. Yong and Y.-F. Guan, An improved Stöber method towards uniform and monodisperse Fe₃O₄@C nanospheres, *J. Mater. Chem. A*, 2013, **1**, 7488–7493.
- 35 B. S. Heidari, V.-S. Cheraghchi, S. Motahari, G. H. Motlagh and S. M. Davachi, Optimized mercapto-modified resorcinol formaldehyde xerogel for adsorption of lead and copper ions from aqueous solutions, *J. Sol-Gel Sci. Technol.*, 2018, **88**, 236–248.
- 36 B. Tahmasbi and A. Ghorbani-Choghamarani, Magnetic MCM-41 nanoparticles as a support for the immobilization of a palladium organometallic catalyst and its application in C-C coupling reactions, *New J. Chem.*, 2019, 43, 14485–14501.
- 37 M. Norouzi and D. Elhamifar, Phenylene and isatin based bifunctional mesoporous organosilica supported Schiffbase/manganese complex: an efficient and recoverable nanocatalyst, *Catal. Lett.*, 2019, **149**, 619–628.
- 38 Y. Zhang, W. Ma, D. Li, M. Yu, J. Guo and C. Wang, Benzoboroxole-Functionalized Magnetic Core/Shell Microspheres for Highly Specific Enrichment of Glycoproteins under Physiological Conditions, *Small*, 2014, **10**, 1379–1386.
- 39 G. Liu, D. Wang, F. Zhou and W. Liu, Electrostatic Self-Assembly of Au Nanoparticles onto Thermosensitive Magnetic Core-Shell Microgels for Thermally Tunable and Magnetically Recyclable Catalysis, *Small*, 2015, 11, 2807–2816.
- 40 S. Abaeezadeh, D. Elhamifar, M. Norouzi and M. Shaker, Magnetic nanoporous MCM-41 supported ionic liquid/ palladium complex: an efficient nanocatalyst with high recoverability, *Appl. Organomet. Chem.*, 2019, 33, e4862.
- 41 M. M. Heravi, A. Ghods, K. Bakhtiari and F. Derikvand, Zn[(L) proline]2: an efficient catalyst for the synthesis of biologically active pyrano[2,3-d]pyrimidine derivatives, *Synth. Commun.*, 2010, **40**, 1927–1931.
- 42 G. M. Ziarani, S. Faramarzi, S. Asadi, A. Badiei, R. Bazl and M. Amanlou, Three-component synthesis of pyrano[2,3-*d*]-pyrimidine dione derivatives facilitated by sulfonic acid nanoporous silica (SBA-Pr-SO₃H) and their docking and urease inhibitory activity, *Daru, J. Pharm. Sci.*, 2013, **21**, 1–13.
- 43 M. Bararjanian, S. Balalaie, B. Movassag and A. Amani, One-pot synthesis of pyrano[2,3-*d*]pyrimidinone derivatives catalyzed by L-proline in aqueous media, *J. Iran. Chem. Soc.*, 2009, **6**, 436–442.