



NiFe₂O₄@SiO₂/SO₃H AS A NOVEL AND ENVIRONMENTALLY BENIGN CATALYST FOR THE SYNTHESIS OF SUBSTITUTED IMIDAZOLE SCAFFOLDS.

K. R. Kadam¹, A. S. Waghmare^{1*}, S. N. Shringare², D. S. Wankhede^{3*}

¹Department of Chemistry and Research Laboratory, Padmashri Vikhe Patil College, Pravaranagar, Ahilyanagar-413713, (Maharashtra) India.

²School of Chemical Sciences, Punyashlok Ahilyadevi Holkar Solapur University, Solapur-413255, (Maharashtra) India.

³School of Chemical Sciences, Swami Ramanand Teerth Marathwada University, nanded-431605, (Maharashtra), India

*Corresponding authors: ✉ A. S. Waghmare (asw6807@gmail.com) and ✉ D. S. Wankhede (dswchem@yahoo.co.in)

Introduction

The development of efficient, sustainable, and environmentally benign catalytic systems remains a central objective in modern synthetic organic chemistry [1,2]. In recent years, increasing attention has been devoted to heterogeneous catalysts derived from nanostructured materials due to their high surface area, tunable surface properties, excellent catalytic efficiency, and ease of recovery [3–5]. Among these, magnetically separable nanocatalysts have emerged as particularly attractive candidates, as they combine superior catalytic performance with straightforward recyclability, thereby minimizing waste generation and reducing the overall environmental footprint of chemical processes [6,7].

Silica-supported nanomaterials have gained prominence as robust catalyst platforms owing to the chemical inertness, thermal stability, high surface area, and facile surface functionalization of silica matrices [8,9]. Functionalization of silica with acidic groups, especially sulfonic acid (–SO₃H) moieties, imparts strong Brønsted acidity and enhances catalytic activity for a wide range of organic transformations [10–12]. The incorporation of –SO₃H groups onto silica frameworks not only improves the dispersion of active sites but also prevents leaching, thereby ensuring catalyst durability and reusability [13]. When combined with magnetic nanostructures, such functionalized silica systems offer a synergistic platform that merges catalytic efficiency with operational simplicity [14,15].

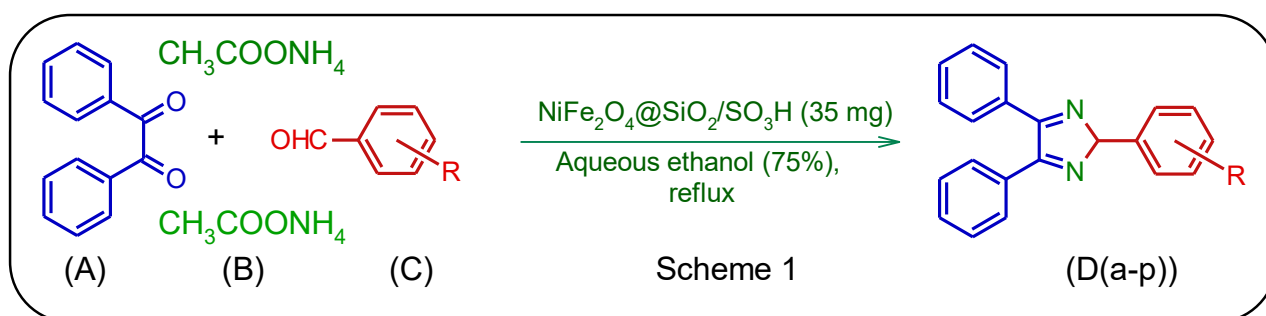
Spinel ferrite nanoparticles, particularly nickel ferrite (NiFe₂O₄), have attracted considerable interest due to their unique magnetic properties, chemical stability, and low toxicity [16,17]. NiFe₂O₄ nanoparticles exhibit strong magnetic responsiveness, enabling rapid separation from reaction media using an external magnetic field [18]. Moreover, their compatibility with silica coatings allows the formation of core–shell or embedded nanostructures that enhance structural stability while preserving magnetic functionality [19]. Embedding NiFe₂O₄ within an –SO₃H functionalized silica matrix therefore represents a rational strategy for designing multifunctional nanocatalysts with high activity, stability, and recyclability [20].

Imidazole and its substituted derivatives constitute a vital class of nitrogen-containing heterocycles that are widely distributed in biologically active molecules, pharmaceuticals, agrochemicals, and functional materials [21,22]. These scaffolds display a broad spectrum of pharmacological properties,

including antifungal, antibacterial, anti-inflammatory, anticancer, and antiviral activities [23]. Additionally, substituted imidazoles serve as key structural motifs in ionic liquids, corrosion inhibitors, ligands, and advanced materials [24]. Consequently, the development of efficient and sustainable synthetic protocols for imidazole derivatives continues to be of significant interest to both academic and industrial researchers [25].

Conventional methods for the synthesis of substituted imidazoles often suffer from drawbacks such as harsh reaction conditions, prolonged reaction times, use of toxic solvents, low selectivity, and difficulties in catalyst recovery [26,27]. In this context, the application of heterogeneous nanocatalysts under mild and green reaction conditions offers a promising alternative [28]. Acid-functionalized magnetic nanocatalysts, in particular, provide an effective platform for promoting multicomponent reactions leading to imidazole frameworks, while simultaneously addressing issues related to sustainability, catalyst separation, and reuse [29,30].

In light of these considerations, the present scientific communication reports the design and application of an SO₃H functionalized silica embedded NiFe₂O₄ nanomaterial as a novel and environmentally benign heterogeneous catalyst for the synthesis of substituted imidazole scaffolds (**Scheme 1**). In the present study, we reported an environmentally benign synthesis of highly substituted imidazole scaffolds (D(a-p)) from a multi component reaction of benzil ((A), 1 mmol), ammonium acetate ((B), 2.2 mmol) and substituted aldehyde ((C), 1 mmol) in presence of catalytic amounts of newly synthesized NiFe₂O₄@SiO₂/SO₃H. The reaction mixture was swirled at 80°C in aqueous ethanol for the durations reported in **Table 3** to obtain the desired imidazole scaffolds.



Materials and Methods

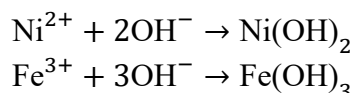
Materials and experimental:

Benzil, Ammonium acetate, substituted aldehydes, tetra-ethoxy-silicate, ethanol and rest of the chemicals used were purchased from the Fisher Scientific, Loba, or Merck chemical companies. Solvents are purified by suitable methods before their use and rest of chemicals were used as received, without further purification. Majority of chemicals used are high purity “AR” grade. The ¹³C-NMR (75 MHz) and ¹H-NMR (300 MHz) spectra were recorded on Bruker Avance (300 MHz) NMR spectrometer using TMS as the internal standard and CDCl₃ or DMSO as solvents. The FT-IR, TGA-DTA and EDX studies of the catalyst material NiFe₂O₄@SiO₂/SO₃H were carried out on the Perkin Elmer RX-I FT-IR spectrometer, Shimadzu TGA-50H thermo-gravimetric analyzer and JEOL JSM 6100 FESEM instrument respectively. The MASS analysis of the synthesized compounds was performed on MALDI SYNAPT XS HD Mass Spectrometer. Melting points of the synthesized compounds were carried out in open capillary tubes by gradual heating using a paraffin bath. Reactions were monitored using TLC technique, Silica coated aluminum foils were used as stationary phase and n-hexane, pet ether and ethyl acetate mixtures were used as mobile phase.

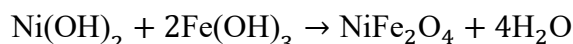
Synthesis of NiFe₂O₄ nanoscale materials.

NiFe₂O₄ nanoparticles were synthesized by a chemical co-precipitation method, as described in the literature [31]. Aqueous solutions of nickel (II) nitrate (2.5 mmol) and iron (III) nitrate (5 mmol) were prepared separately by dissolving the stoichiometric amounts in 100 ml deionized water each. The

two solutions were then mixed together under continuous magnetic stirring to obtain a homogeneous mixed metal nitrate solution containing Ni²⁺ and Fe³⁺ ions in a 1:2 molar ratio. The mixed metal nitrate solution was heated to 60–80 °C under constant stirring. A freshly prepared aqueous solution of NH₄OH was added dropwise to the reaction mixture. The pH of the solution was carefully monitored and maintained at about 10–11. During the addition of the base, a dark brown/black precipitate formed, indicating the simultaneous precipitation of nickel and iron hydroxides according to the following reactions:



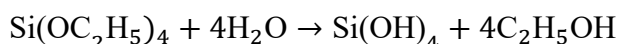
The reaction mixture was stirred continuously at the same temperature for 1–2 hours to ensure complete precipitation and uniform nucleation. After completion of the precipitation process, the reaction mixture was allowed to age for 12–24 hours at room temperature. Aging promotes crystal growth and improves homogeneity of the nanoparticles. The precipitate was separated by centrifugation or filtration and washed repeatedly with deionized water (and finally with ethanol, if required) until the pH of the washings became neutral. This step removes residual nitrate ions and excess alkali. The washed precipitate was dried in a hot air oven at 80–100°C for 10–12 hours, resulting in a dry precursor powder containing mixed metal hydroxides. The dried powder was finely ground and calcined in a muffle furnace at 500–800 °C for 3–5 hours in air. During calcination, the metal hydroxides decomposed and reacted to form crystalline NiFe₂O₄ spinel ferrite nanoparticles according to:



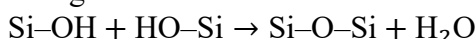
After calcination, the furnace was allowed to cool naturally to room temperature, and the obtained powder was gently ground to obtain fine NiFe₂O₄ nanoparticles. The synthesized NiFe₂O₄ nanomaterial appears as a black magnetic powder, suitable for structural, morphological, magnetic, and catalytic studies.

Synthesis of NiFe₂O₄@SiO₂ nanoscale materials

The silica coating of NiFe₂O₄ nanoparticles was carried out using a modified Stöber sol–gel process involving ammonia-catalyzed hydrolysis and condensation of TEOS in an ethanol–water medium, as widely reported in the literature [32]. Pre-synthesized NiFe₂O₄ nanoparticles (1 g) was dispersed in a 100 ml mixture of absolute ethanol and deionized water (typically ethanol:water = 4:1 v/v). The suspension was ultrasonicated for 30–45 minutes to ensure uniform dispersion and to prevent agglomeration of the magnetic nanoparticles. In a separate beaker, tetraethoxysilicate, TEOS (4 ml) was dissolved in absolute ethanol (20 ml) under continuous stirring to obtain a clear and homogeneous solution. This solution acts as the silica precursor. The TEOS solution was added slowly and dropwise to the NiFe₂O₄ suspension under vigorous magnetic stirring at room temperature. Subsequently, ammonium hydroxide solution was added dropwise to the reaction mixture to adjust the pH to ~9–11, which catalyzes the hydrolysis and condensation reactions. The hydrolysis of TEOS proceeds as follows:



The hydrolyzed silanol groups undergo condensation reactions:



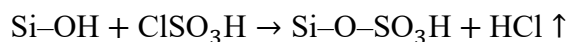
During this process, the silica network forms and encapsulates or embeds the NiFe₂O₄ nanoparticles, yielding a SiO₂ embedded NiFe₂O₄ nanocomposite, which is designated as NiFe₂O₄@SiO₂. The reaction mixture was stirred continuously for 6–12 hours at room temperature to ensure complete hydrolysis and uniform silica coating.

Synthesis of NiFe₂O₄@SiO₂ / SO₃H nanoscale materials

Sulfonic acid functionalization of silica-coated magnetic nanoparticles was achieved via reaction of surface silanol groups with chlorosulfonic acid, generating covalently anchored –SO₃H Brønsted acid

sites, as widely reported in the literature [33]. The surface of NiFe₂O₄@SiO₂ nanoparticles contains abundant surface silanol (–Si–OH) groups. Chlorosulfonic acid reacts with these hydroxyl groups through electrophilic substitution, forming covalently anchored sulfonic acid (–SO₃H) groups on the silica surface. This functionalization introduces strong Brønsted acidity, rendering the material suitable for acid catalysis.

The NiFe₂O₄@SiO₂ nanoparticles were dried under vacuum at 100–120 °C for 4–6 hours to remove physically adsorbed moisture. This step is crucial to ensure effective reaction between surface –OH groups and chlorosulfonic acid. Dried NiFe₂O₄@SiO₂ nanoparticles (typically 1.0 g) was dispersed in dry dichloromethane (20–30 mL) under magnetic stirring. The suspension was stirred for 30 minutes to obtain a uniform dispersion. The reaction flask was placed in an ice bath (0–5 °C) to control the exothermic nature of the reaction. A measured amount of chlorosulfonic acid (typically 0.5–1.5 mL, depending on desired acid density) was added dropwise to the nanoparticle suspension under continuous stirring. During addition, hydrogen chloride (HCl) gas evolved, indicating successful sulfonation of the silica surface. The surface reaction can be represented as:



After complete addition of chlorosulfonic acid, the reaction mixture was allowed to warm to room temperature and stirred continuously for 4–6 hours to ensure complete functionalization of surface silanol groups. Upon completion of the reaction, the solid material was separated using a magnet or centrifugation. The obtained solid was carefully washed several times with anhydrous dichloromethane, followed by ethanol, to remove unreacted chlorosulfonic acid and adsorbed by-products. The SO₃H-functionalized NiFe₂O₄@SiO₂ nanoparticles were dried under vacuum or in a hot air oven at 80–100 °C for 8–10 hours. The final product is a dark, magnetically separable powder, designated as NiFe₂O₄@SiO₂/SO₃H, possessing strong Brønsted acid sites uniformly anchored on the silica surface.

Formation and properties of SO₃H-functionalized NiFe₂O₄@SiO₂ nanoparticles were studied using adequate analytical techniques.

Synthesis of 2,4,5-trisubstituted imidazole scaffolds

The synthesis was carried out following a one-pot multicomponent strategy reported for Brønsted acid catalyzed imidazole formation with minor modification [34]. In a round-bottom flask equipped with an arrangement of stirring and heating, benzil (1.0 mmol), substituted aldehyde (1.0 mmol), ammonium acetate (2.2 mmol) and NiFe₂O₄@SiO₂/SO₃H (35 mg) were refluxed in aqueous ethanol (75% v/v) for the period mentioned in table 1. Progress of the reaction was monitored using TLC technique with silica coated aluminum foil as stationary phase and n-hexane, pet-ether and ethyl acetate as mobile phase. As indicated by TLC, after completion of reaction, the precipitated product and heterogeneous catalyst were separated from solvent by filtration at pump. The isolated product was dried and crude yield was measured. The catalyst NiFe₂O₄@SiO₂/SO₃H was recovered magnetically from the reaction product by dissolving it in proper solvent for recrystallization purpose. The recrystallized product was further purified by column chromatography if needed. The purified product was characterized by adequate analytical techniques.

2,4,5-Triphenyl-1H-imidazole (a)

White solid; mp: 274–276 °C.

FTIR (neat, cm^{–1}): 1589 (C=C), 1627 (C=N), 3057 (C–H, Ar), 3215 (N–H).

¹H NMR (400 MHz, DMSO-d₆): δ 7.57–7.23 (m, 13H, Ar–H), 8.10 (d, 2H, Ar–H), 12.70 (s, 1H, N–H).

¹³C NMR (100 MHz, DMSO-d₆): δ 125.66, 127.00, 127.54, 128.77, 129.17, 130.81, 131.55, 135.62, 137.55, 145.97.

Mass (m/z): Calcd for C₂₁H₁₆N₂: 296.13; Found: 294.07.

Separated catalyst was washed thrice with absolute ethanol and employed further for next catalytic cycle.

Result and Discussion

In the initial phase of the study, a three-component condensation reaction involving benzil (1 mmol), ammonium acetate (2 mmol), and benzaldehyde (1 mmol) was selected as a model system to investigate and optimize the reaction parameters of the proposed protocol. The catalytic efficacy of NiFe₂O₄@SiO₂/SO₃H in this condensation reaction was evaluated by conducting two parallel experiments: one in the presence of 50 mg of the NiFe₂O₄@SiO₂/SO₃H catalyst and the other under identical conditions in the absence of any catalyst, both performed under reflux in ethanol. The catalyzed reaction afforded the desired product with 82% yield within 135 minutes, whereas the uncatalyzed reaction required more than 24 hours and produced only trace amounts of the product. These results unequivocally demonstrate the significant catalytic role of NiFe₂O₄@SiO₂/SO₃H in promoting the condensation reaction.

To identify the most appropriate solvent for the proposed protocol, a series of solvents were systematically evaluated under reflux conditions for 135 minutes using NiFe₂O₄@SiO₂/SO₃H (50 mg) as the catalyst. The results obtained (**Table 1**) revealed that methanol, ethanol, aqueous ethanol (75%), DMF, and DMSO afforded comparable product yields under identical set reaction conditions. Among the solvents examined, aqueous ethanol (75%) was selected as the solvent of choice owing to its relatively low toxicity, environmental compatibility, and safer handling characteristics.

Table 1. Selection of appropriate solvent for condensation reaction of benzil (1 mmol), ammonium acetate (2 mmol), and benzaldehyde (1 mmol) under reflux for 135 minutes using NiFe₂O₄@SiO₂/SO₃H (50 mg) as a catalyst.

Sr. No.	Solvent	Yield of product (%)
1.	Deionized Water	trace
2.	Methanol	82
3.	Ethanol	80
4.	Aqueous ethanol (25%)	33
5.	Aqueous ethanol (50%)	74
6.	Aqueous ethanol (75%)	82
7.	Acetone	54
8.	Acetonitrile	77
9.	Chloroform	63
10.	Dioxane	54
11.	DMF	80
12.	DMSO	78

The catalytic efficiency of NiFe₂O₄@SiO₂/SO₃H in the present protocol was evaluated using a model reaction for 135 minutes reflux in aqueous ethanol (75%) by varying the catalyst loading from 5 to 50 mg. The results, summarized in **Table 2**, clearly indicate that a catalyst loading of 35 mg per mole of the 1,2-diketocompound is optimal, ensuring efficient and smooth progression of the reaction.

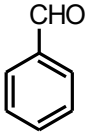
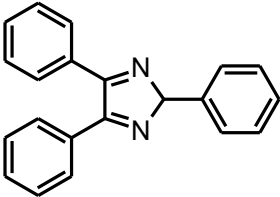
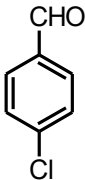
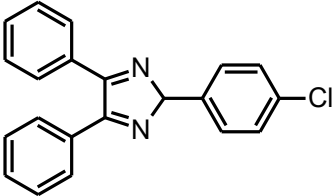
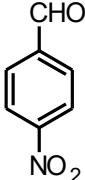
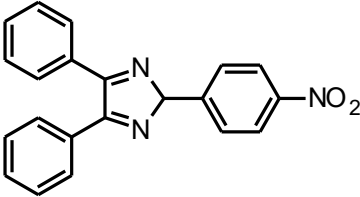
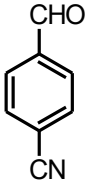
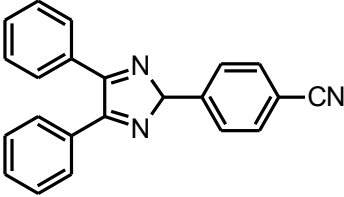
Table 2. Optimization catalyst amount for the model reaction between benzil (1 mmol), ammonium acetate (2 mmol), and benzaldehyde (1 mmol) under reflux in aqueous ethanol for 135 minutes using NiFe₂O₄@SiO₂/SO₃H (from 5 mg to 50 mg) as a catalyst.

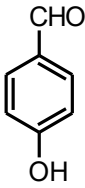
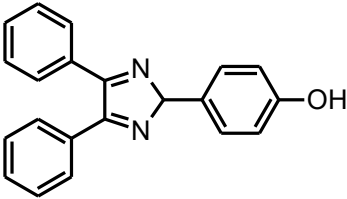
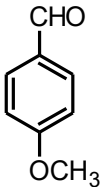
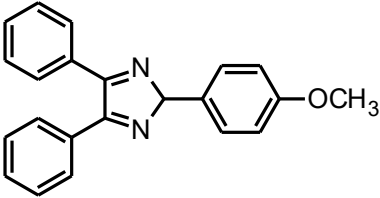
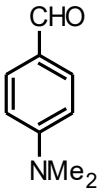
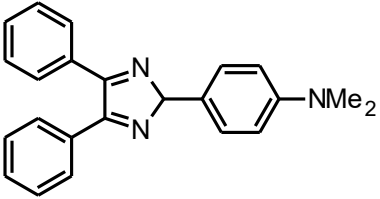
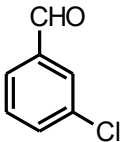
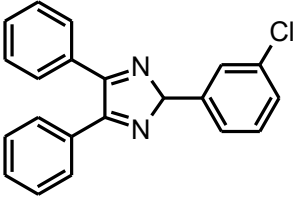
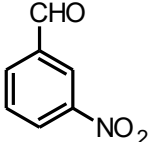
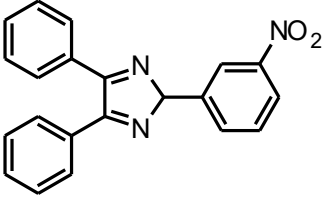
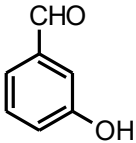
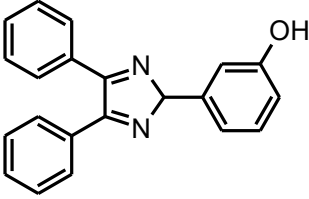
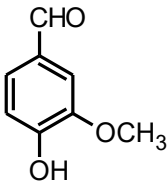
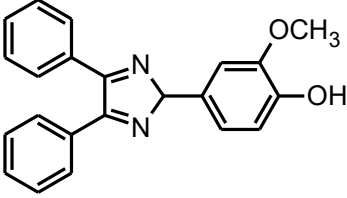
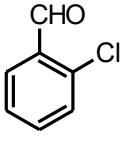
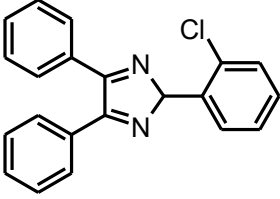
Sr. No.	Amount of NiFe ₂ O ₄ @SiO ₂ -SO ₃ H (mg)	Yield of product (%)
1.	00	trace
2.	05	08
3.	10	19
4.	15	29
5.	20	43

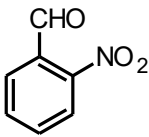
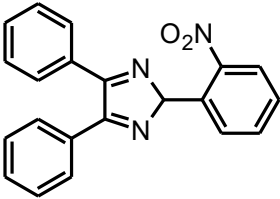
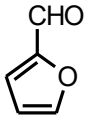
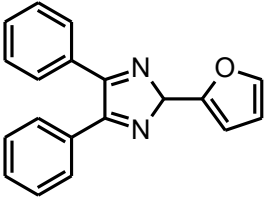
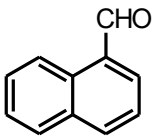
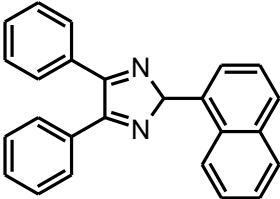
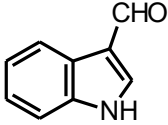
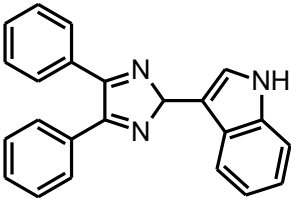
6.	25	57
7.	30	73
8.	35	82
9.	40	82
10.	45	81
11.	50	81

The practical applicability of the proposed protocol was demonstrated through a model reaction employing a diverse set of aldehydes under reflux conditions in aqueous ethanol (75%) for 135 minutes, using NiFe₂O₄@SiO₂/SO₃H (35 mg) as the catalyst. The results obtained are summarized in **Table 3**. The study clearly indicates that the designed methodology proceeds smoothly across a wide range of aldehydes. As anticipated, both electronic and steric effects of substituents on the aromatic ring significantly influence the reaction outcomes. Aldehydes bearing electron-donating groups required comparatively longer reaction times (**Table 3**, entries e, f and k), whereas sterically hindered aldehydes exhibited prolonged reaction times along with reduced product yields (**Table 3**, entries l, m, o and p). Reactions of aldehydes with electron withdrawing groups, as expected completed in shorter reaction times with better yields (**Table 3**, entries c, d and i).

Table 3. Synthesis of 2,4,5-trisubstituted imidazoles from 1,2 diketocompounds (1mmol), ammonium acetate (2.2 mmol), substituted aldehydes (1 mmol) and NiFe₂O₄@SiO₂/SO₃H (35 mg) under reflux in aqueous ethanol (75%).

Entry	Aldehyde	Imidazole derivatives	Time (min.)	*Yield (%)
a.			135	82
b.			120	89
c.			90	94
d.			90	89

e.			140	78
f.			140	80
g.			150	80
h.			135	84
i.			120	88
j.			145	80
k.			155	82
l.			175	72

m.			162	75
n.			140	79
o.			190	74
p.			165	78
*Isolated yields				

Reusability strength of catalyst NiFe₂O₄@SiO₂/SO₃H was tested with the model reaction of benzaldehyde, benzil, ammonium acetate under reflux in aqueous ethanol (75%). A catalytic material made to undergo to six catalytic re-cycles, after every cycle catalyst was quantitatively recovered magnetically. Results of examined catalytic recycles are summarized in the form of a bar chart given in **Figure 1**.

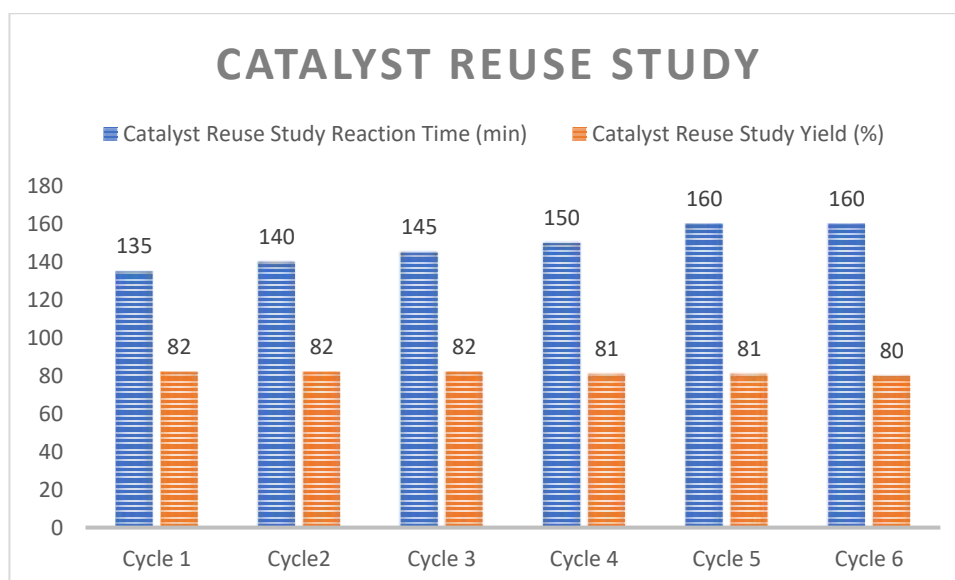


Figure 1: Catalyst reuse study with the model reaction under reflux in aqueous ethanol.

It was observed that the catalyst is enough potential to promote the model reaction even after five cycles of its reuse. A slight decrement in reaction time was observed this is may due to slight poisoning of catalyst due to ammonium acetate.

Conclusion

In summary, an SO₃H-functionalized silica-embedded NiFe₂O₄ nanocatalyst (NiFe₂O₄@SiO₂/SO₃H) has been successfully designed and demonstrated as an efficient, magnetically recoverable, and environmentally benign heterogeneous catalyst for the synthesis of 2,4,5-trisubstituted imidazole scaffolds. The developed protocol offers several notable advantages, including mild reaction conditions, use of aqueous ethanol as a green solvent, high product yields, broad substrate scope, and straightforward magnetic separation of the catalyst. The catalyst exhibited excellent recyclability, retaining substantial catalytic activity over multiple reaction cycles with only a marginal decline in performance, highlighting its structural stability and durability. The strong Brønsted acidity imparted by surface-anchored –SO₃H groups, combined with the magnetic core–shell architecture, plays a crucial role in enhancing catalytic efficiency and operational simplicity. Overall, this study underscores the potential of NiFe₂O₄@SiO₂/SO₃H as a sustainable and practical nano-catalyst for multicomponent heterocyclic syntheses and contributes meaningfully to the advancement of green and recyclable catalytic systems in organic synthesis.

Disclosure Statement

The authors declare no conflicts of interest regarding this manuscript.

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