

Green preparation of tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-ones using $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ as novel and recyclable magnetic catalyst

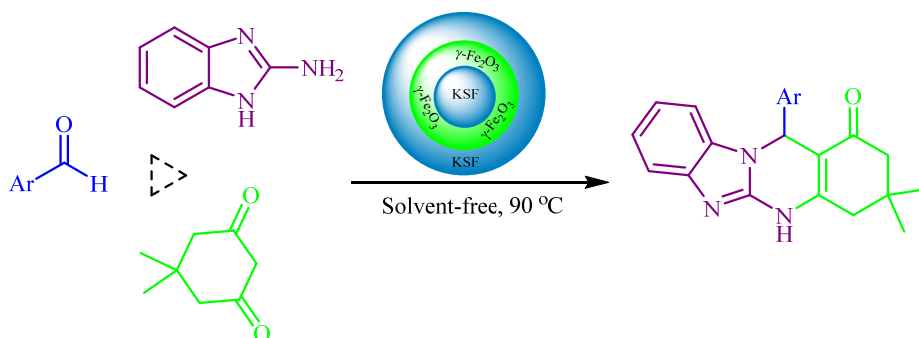
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HIGHLIGHTS

- The preparation of quinazolins by $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ -catalyzed reactions of 2-aminobenzimidazole, benzaldehydes and dimedone is reported.
- The catalyst was prepared using the successive coating of $\gamma\text{-Fe}_2\text{O}_3$ shell on KSF core.
- Using $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ offers advantages, including easy workup, high yields, reusability, no by-products and efficient product separation.

GRAPHICAL ABSTRACT



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ABSTRACT

The preparation of tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-ones via a $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ -catalyzed multicomponent coupling reaction of 2-aminobenzimidazole, benzaldehydes, and dimedone in solvent-free conditions is reported. $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ as a magnetic catalyst was prepared using the successive coating of a $\gamma\text{-Fe}_2\text{O}_3$ shell on a KSF core and was characterized by different methods including FT-IR, XRD, TGA and SEM techniques. The merits of this method include limited use of organic solvents, excellent purity of products, and an easy workup technique. The tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-ones were prepared in yields of 88-94%. The catalyst was recovered through an external magnet and reused four times without any considerable loss of its activity.

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1. Introduction

Multicomponent coupling reactions (MCRs) in environmentally friendly conditions and at room temperature are of great interest to organic chemists because they provide access to a wide number of organic molecules through a high atom economy and environmentally benign route [1-9]. Performing reactions in the presence of nanocatalysts are important, but one of the major problems of this reagents is their down activity. All nanoparticles, (NPs) known as quasi-homogeneous catalysts, are allocated between homogeneous and heterogeneous catalysts due to their extensive surface area and improved dispersal ability in most organic reactions [10]. Nanoparticles often have the desirable characteristics of both organic and inorganic catalysts [11]. Among nanoparticles, iron oxides, maghemite ($\gamma\text{-Fe}_2\text{O}_3$) and magnetite (Fe_3O_4) are excellent choices due to their biocompatibility, less toxicity than their metallic peers and super paramagnetic behaviour [12,13]. One of the unique features of nanoparticles is their ability to be separated through magnetic force [14]. Another advantage of this kind of separation is avoiding the use of dangerous solvents and additional filtration. Quinazolines are a known group of heterocyclic compounds that are used in various pharmaceutical and biological activities such as antibacterial [15], antimicrobial [16], antitumor [17], transient receptor potential A1 (TRPA1) antagonistic [18], nitric oxide synthase (NOS) inhibitive [19], and antifungal activities [20]. The quinazolinone core has also been found in some drugs and natural products (Fig. 1) [21]. In recent years, several efficient methods for the synthesis of quinazolinones have been developed. Classical synthetic methods for the preparation of quinazolinones include the utilization of anthranilic acids [22], 2-aminobenzonitriles [23], isatoic anhydrides [24] and anthranilamides [25], aza-Wittig reactions [26], transition-metal catalyzed reactions [27], and microwave-assisted dehydrative cyclization of diamides [28].

2. Experiments

2.1. Materials

All chemicals were procured from Merck and Aldrich companies. All reported yields refer to isolated

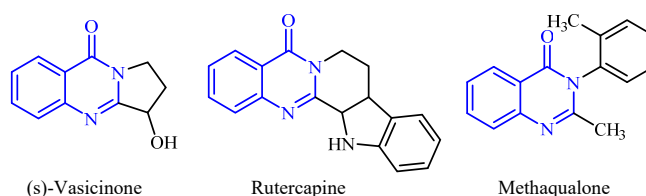


Fig. 1. Quinazolinone moiety containing pharmaceuticals and natural products.

products. The product structure of the prepared samples was confirmed by comparing their physical characteristics, FT-IR, and NMR spectroscopy with reliable samples and results reported in the literature. Monitoring the progress of the reaction and determining the purity of the compounds were accomplished by thin-layer chromatography (TLC). Melting points were measured by a Büchi B-545 apparatus, and the FT-IR spectra were recorded with a Perkin-Elmer spectrum BX series spectrophotometer. X-ray diffraction (XRD) measurements were performed by a Siemens D-4500 X-ray diffractometer (Rigaku), using $\text{Cu } K_\alpha$ radiation (1.5406 Å) at room temperature. Scanning electron microphotographs (SEM) were conducted on an SEM-Germany LEO 1430VP. Thermogravimetric analyses (TGA) were investigated by the Polymer Laboratories PL-TGA thermal analysis instrument. Catalyst was heated from 25 to 800 °C, at ramp 20 °C.min⁻¹ under N₂ atmosphere.

2.2. Synthesis of $\gamma\text{-Fe}_2\text{O}_3$ nanoparticles

The $\gamma\text{-Fe}_2\text{O}_3$ nanoparticles was prepared by the reported chemical technique [31]. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (4.89 g) and $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ (3 g) were dissolved in deionized water (50 ml) at room temperature and under Argon atmosphere. At the same temperature, NH_4OH solution (0.6 M, 330 ml) was added dropwise to the stirring mixture to reach the reaction pH to 11. The black suspension was stirred for 1 h at room temperature, and then for 1 hour under reflux conditions to produce a brown dispersion. Finally, the magnetic nanoparticles were separated by an external magnet and washed several times with deionized water. The collected sediment was heated for 5 h at 250 °C to produce a reddish-brown powder.

2.3. Synthesis of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ NPs

$\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ was prepared via the reported method

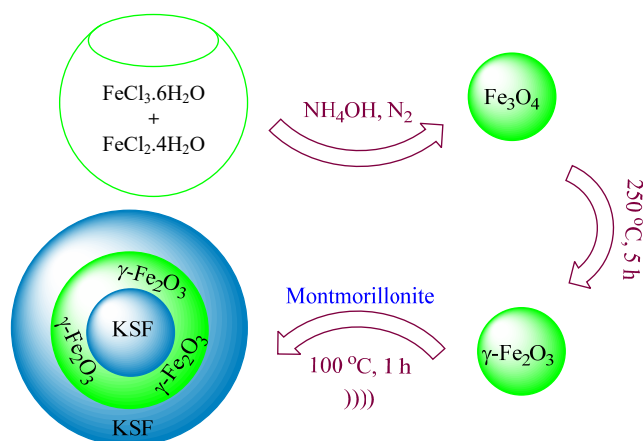
[29] with a few modifications. The synthesized $\gamma\text{-Fe}_2\text{O}_3$ (3.5 g) was added to glacial acetic acid (30 ml), then montmorillonite (1.5 g) in deionized water (100 ml) was added slowly under ultrasonic irradiation for 20 min and the mixture was kept at 100 °C for 1 h under vigorous stirring. The nanoparticles of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ were collected by an external magnet. Then, the precipitate was washed with deionized water and dried at 50 °C in vacuum for 12 h (Scheme 1).

2.4. General method for the synthesis of tetrahydrobenzimidazo[2,1-b]quinazolin-1(2H)-one derivatives

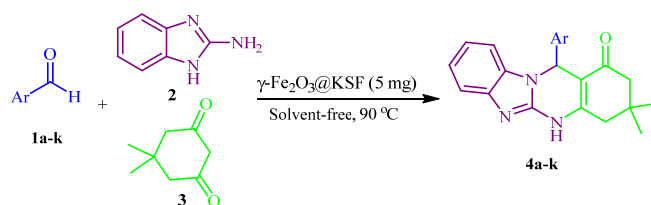
A mixture of 2-aminobenzimidazole (1 mmol), aromatic aldehyde (1 mmol), dimedone (1 mmol), and $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ (5 mg) was heated at 90 °C under solvent-free conditions for the appropriate time. After completion of the reaction [characterized using TLC (*n*-Hexane: EtOAc, 4:1), it was cooled to room temperature and 3 ml of hot EtOH was added to the mixture. The nanocatalyst was removed in the presence of an external magnet, and the desired product was purified by recrystallization in hot EtOH (Scheme 2).

3. Results and discussion

As part of our broad interest in the synthesis of heterocyclic compounds [31–39], we report the synthesis of tetrahydrobenzimidazo[2,1-b]quinazolin-1(2H)-ones **4a–k** by $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ -catalyzed multi-component coupling reaction of benzaldehydes **1a–k**, 2-aminobenzimidazole **2**, and dimedone **3** in solvent-free conditions.



Scheme 1. Preparation of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ NPs.



Scheme 2. Preparation of tetrahydrobenzimidazo[2,1-b]quinazolin-1(2H)-ones **4a–k**.

3.1. Catalyst characterization

3.1.1. FT-IR analysis

Fig. 2 displays the FT-IR spectra of $\gamma\text{-Fe}_2\text{O}_3$ and $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$. Significant change in the wide band ranging from 480 to 1600 cm^{-1} is detected in the FT-IR spectra of the $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$, which can be allocated to the overlapping peak between the KSF bridging stretching modes and Fe–O vibrations. The O–H bending bond modes observed peaks at about 1432 and 1520 cm^{-1} . The broad peak at around 3400 and 1630 cm^{-1} corresponds to the bending H–O–H bond vibration of water molecules adsorb on the surface of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ [30].

3.1.2. Powder X-ray diffraction

The XRD chart of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ is shown in Fig. 3. In this chart diffraction peaks from the $\gamma\text{-Fe}_2\text{O}_3$ can be clearly seen at $2\theta = 30.1^\circ, 35.5^\circ, 43.1^\circ, 53.5^\circ, 57.0^\circ$, and 62.6° and marked by their indices (2 2 0), (3 1 1), (4 0 0), (4 2 2), (5 1 1), and (4 4 0). Moreover, five diffraction peaks appeared at around $2\theta = 9.3^\circ, 16.8^\circ, 18.6^\circ, 20.1^\circ$ and 27.3° , which can be attributed to the diffraction peaks of KSF. These results indicate that $\gamma\text{-Fe}_2\text{O}_3$ nanoparticles are successfully installed on the surface of KSF [33].

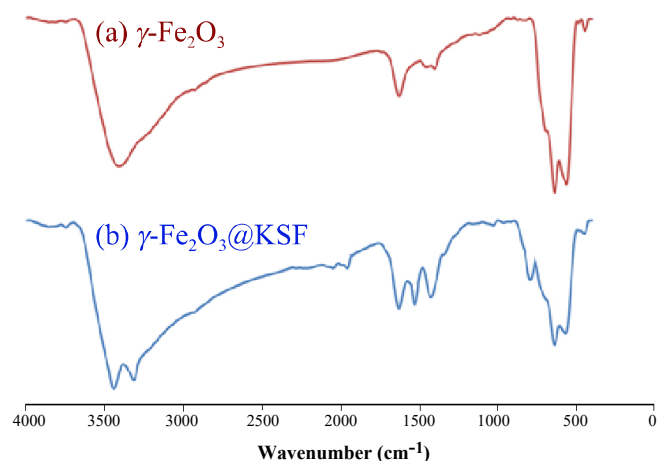


Fig. 2. FT-IR spectra of (a) $\gamma\text{-Fe}_2\text{O}_3$ and (b) $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$.

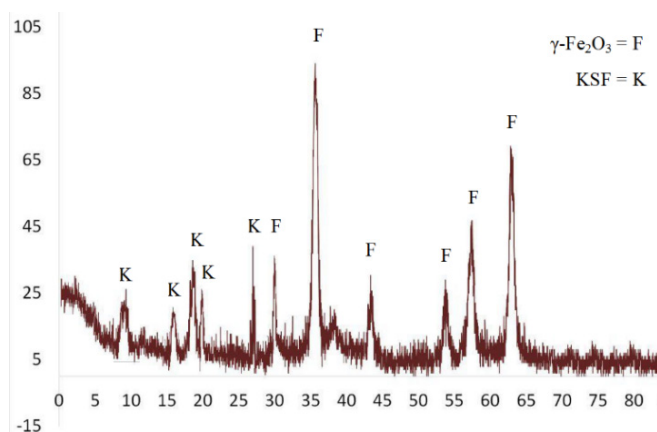


Fig. 3. XRD patterns of the $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ catalyst.

3.1.3. SEM Analysis

Fig. 4 show the SEM images of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ prepared with the montmorillonite precursor at 100°C. The image shows that the newly formed $\gamma\text{-Fe}_2\text{O}_3$ magnetic layers are uniformly and porous coated onto the KSF core. As shown in Fig. 4(a), the external surface of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ is rough. Moreover, from Fig. 4(b), it was found that the size of the $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ catalyst was around 38–44 nm.

3.1.4. TGA Analysis

Fig. 5 shows the TGA of the $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ sample. The loss of about 5% of its weight at 130 °C can be related to the loss of surface absorbed water, and the reduction of about 12% of its weight up to 430 °C is ascribed to the decomposition of part from the surface bond.

To optimize reaction conditions, including the amount of the catalyst, temperature, various solvents, and solvent-free conditions in the synthesis of tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-ones, the coupling one-pot reaction of benzaldehyde (1 mmol), 2-aminobenzimidazole (1 mmol), and dimedone (1 mmol) was studied in the presence of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ as a model reaction and the results are summarized in Table 1. On the basis of this information, it can be concluded that the best results can be obtained using 5 mg of the catalyst at 90 °C under solvent-free conditions (Table 1, entry 6). Using higher amounts of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ did not affect the reaction yields.

In order to explore the public applicability of this catalyst, a diverse collection of tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-ones were prepared

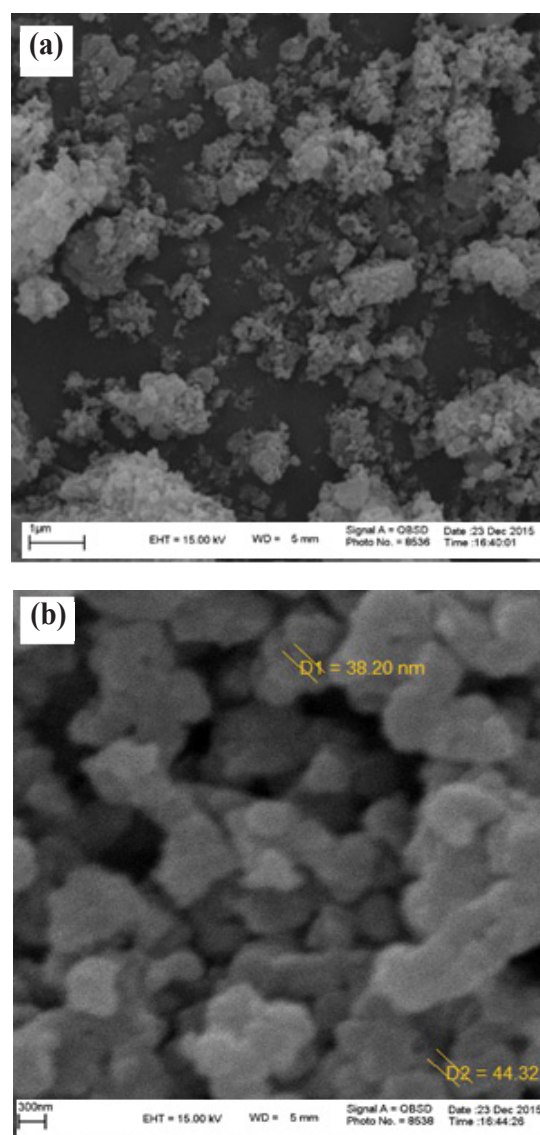


Fig. 4. SEM images of the $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ catalyst.

using the $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$ catalyst under optimized reaction conditions. As it is clear from Table 2, various aldehydes with different electron-donating and electron-

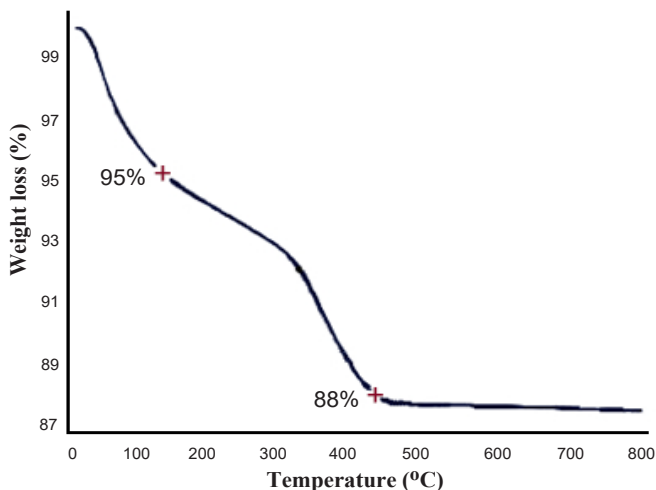


Fig. 5. BTGA curve of $\gamma\text{-Fe}_2\text{O}_3\text{@KSF}$.

Table 1. The effect of various conditions on the synthesis of quinazolinone **4a**.

Entry	Catalyst (mg)	Temperature (°C)	Solvent	Time (min)	Yield (%) ^a
1	5	Reflux	CH ₃ CN	60	...
2	5	Reflux	MeOH	60	75
3	5	Reflux	EtOH	60	80
4	5	Reflux	H ₂ O	60	55
5	5	80	Solvent-free	20	90
6	5	90	Solvent-free	20	92
7	5	100	Solvent-free	20	89
8	10	90	Solvent-free	20	92
9	15	90	Solvent-free	20	90

^a Isolated yields.**Table 2.** Data for the preparation of tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one derivatives using Fe₂O₃@KSF catalyst.

Entry	Products	Aldehyde	Time (min)	Yield (%) ^a	Mp (°C)	
					Found	Reported [ref.]
1	4a	C ₆ H ₅ CHO	10	92	355–357	359–363 [40]
2	4b	2-ClC ₆ H ₄ CHO	12	88	343–345	>300 [41]
3	4c	3-ClC ₆ H ₄ CHO	10	92	340–342	>300 [41]
4	4d	4-ClC ₆ H ₄ CHO	10	94	356–358	>300 [40]
5	4e	3-NO ₂ C ₆ H ₄ CHO	12	90	335–336	>300 [41]
6	4f	4-NO ₂ C ₆ H ₄ CHO	10	93	345–346	>300 [40]
7	4g	3-MeOC ₆ H ₄ CHO	12	90	337–339	>300 [41]
8	4h	4-MeOC ₆ H ₄ CHO	15	88	345–346	>300 [40]
9	4i	4-MeC ₆ H ₄ CHO	12	92	339–341	>300 [42]
10	4j	4-OHC ₆ H ₄ CHO	10	94	345–347	>300 [40]
11	4k	4-BrC ₆ H ₄ CHO	15	90	358–360	>300 [40]

^a Isolated yields.

withdrawing groups were well tolerated under the selected conditions, and this conversion can also occur with high yields in short times.

In order to display the suitability of this nano catalyst, the efficiency of γ -Fe₂O₃@KSF with the other catalysts in the synthesis of 3,3-dimethyl-12-phenyl-3,4,5,12-tetrahydrobenzo[4,5]imidazo[2,1-*b*]quinazolin-1(2*H*)-one **4a** are compared in Table 3. As shown in this table, the newly developed method avoids some of the disadvantages associated with the other procedures such as harsh reaction conditions, use of toxic or moisture sensitive catalyst, high temperature, use of higher amounts of the catalysts, and recoverability of the catalyst. On the basis of these results, we can state that γ -Fe₂O₃@KSF is more efficient than the other methods.

Recycling of the catalyst is a key function in

laboratory processes. Hence, to check the reusability of catalyst after completion of the reaction, the nano particles were recovered by an external magnet, washed with EtOH, and dried at 80 °C for 1 h. The recovered catalyst successfully catalyzed the reaction between benzaldehyde, 2-aminobenzimidazole, and dimedone under the optimized reaction conditions. This catalyst was used for at least four runs without considerable change in the reaction time and yield (Fig. 6).

4. Conclusion

In summary, we have introduced γ -Fe₂O₃@KSF, prepared by the successive coating of γ -Fe₂O₃ shell on KSF core using a mixed solvent procedure, as a stable magnetic catalyst. This reagent was applied as a

Table 3. Comparison of the results obtained from the synthesis of quinazolinone **4a** in the presence of $\text{Fe}_2\text{O}_3@\text{KSF}$ with other catalysts.

Entry	Catalyst [ref.]	Conditions	Time (min)	Yield (%) ^a
1	p-TSA (15 mol %) [43]	CH_3CN , 50 °C	25	95
2	SiO_2 (10 mol %) [44]	EtOH/MW	3	95
3	I_2 (10 mol%) [45]	CH_3CN , reflux	10	84
4	SBA-Pr- SO_3H (10 mol%) [46]	Solvent-free, 100 °C	10	90
5	$\text{NH}_2\text{SO}_3\text{H}$ (50 mol%) [47]	DMF, reflux	15	94
6	$\text{H}_6\text{P}_2\text{W}_{18}\text{O}_{62} \cdot 18\text{H}_2\text{O}$ (1 mol%) [40]	$\text{CH}_3\text{CN}/\text{reflux}$	15	96
7	— [41]	Solvent-free, 110 °C	8 (h)	79
8	— [42]	DMF, MW	8	64
9	$\gamma\text{-Fe}_2\text{O}_3@\text{KSF}$ (5 mg) ^b	Solvent-free, 90 °C	10	93

^a Isolated yields. ^b This work.

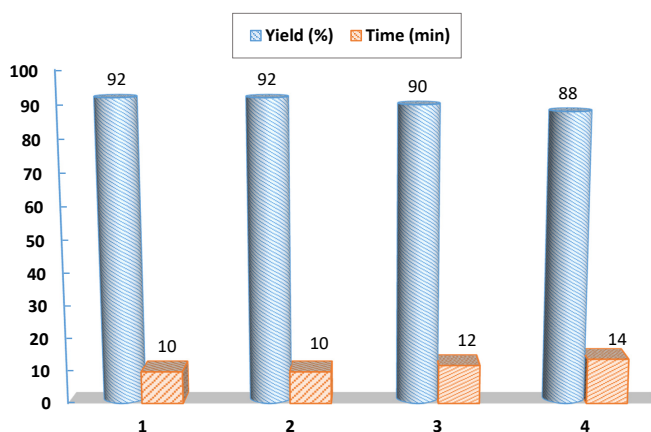
magnetically heterogeneous catalyst for the synthesis of tetrahydrobenzimidazo [2,1-*b*] quinazolin-1(2*H*)-one derivatives from a one-pot reaction of available aldehydes, 2-aminobenzimidazole and dimedone in solvent-free conditions. The practical method shows some advantages including easy workup, high yields, no by-products, product separation, and easy and simple catalyst recycling with the assistance of an external magnet.

Acknowledgments

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**Fig. 6.** EReusability of the $\gamma\text{-Fe}_2\text{O}_3@\text{KSF}$ catalyst in the synthesis of quinazolinone **4a**.

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