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# An efficient and green one-pot synthesis of indazolo [1,2-*b*]-phthalazinetriones via three-component reaction of aldehydes, dimedone, and phthalhydrazide using $Fe_3O_4@SiO_2$ core–shell nanoparticles

**Boshra Mirhosseini-Eshkevari · Mohammad Ali Ghasemzadeh · Javad Safaei-Ghomi**

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**Abstract** An efficient and eco-friendly procedure for the one-pot synthesis of biologically active heterocyclic compounds including indazolo[1,2-*b*]-phthalazinetriones has been developed in the presence of $Fe_3O_4@SiO_2$ nanoparticles. The multicomponent reactions of aldehydes, dimedone, and phthalhydrazide were carried out under solvent-free conditions to obtain some indazolo[1,2-*b*]-phthalazinetrione derivatives. The present approach provides several advantages such as excellent yields, a simple workup, low reaction times, easy catalyst separation, and little catalyst loading.

**Keywords** Indazolo[1,2-*b*]-phthalazinetrione · Multicomponent reaction · Solvent-free · $Fe_3O_4@SiO_2$ · Core–shell

## Introduction

Multicomponent reactions (MCRs) are defined as one-pot processes in which three or more substrates merge either simultaneously (so-called tandem or domino reactions), or through a successive addition procedure that does not need any change of solvent. MCRs are acquiring increasing importance, particularly in the total synthesis of natural products and medicinal heterocyclic compounds because of their simplicity, high yield of products, and low reaction times [1, 2]. Phthalazine derivatives have been given considerable attention in recent years because of their wide range of pharmaceutical and biological activities such as antimicrobial [3], anticonvulsant [4], antifungal [5], anticancer [6], and anti-inflammatory [7]. Moreover, these compounds display good agreement as new luminescent materials

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B. Mirhosseini-Eshkevari · M. A. Ghasemzadeh (✉) · J. Safaei-Ghomi
Department of Chemistry, Qom Branch, Islamic Azad University, Qom, Islamic Republic of Iran
e-mail: ghasemzadeh@qom-iau.ac.ir

or fluorescence probes [8]. In recent years, the development of new methods for the synthesis of heterocyclic rings containing phthalazine is an interesting challenge and therefore a number of procedures have been reported for the synthesis of phthalazine derivatives [9–12]. The first synthesis of indazolo[1,2-*b*]-phthalazinetriones was reported by Bazgir et al. [13] using *p*-toluene sulfonic acid (*p*-TSA) as a catalyst.

In addition, three-component reactions of dimedone, aldehydes, and phthalhydrazide/urazole have been carried out in the presence of various catalysts such as silica sulfuric acid [14], $H_2SO_4$ in water–ethanol or an ionic liquid [15], silica-supported poly phosphoric acid [16], $Mg(HSO_4)_2$ [17], heteropoly acids [18], N-halo sulfonamides [19], sulfonated poly(ethylene glycol) [20], wet cyanuric chloride [21], molecular iodine [22], and nano-silica sulfuric acid [23]. Thus, great attention has been paid to providing new approaches for the synthesis of phthalazinetriones.

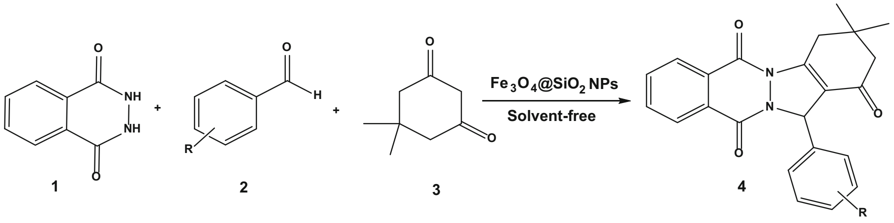
In recent years, nanoscience and nanotechnology are wonderful growing research areas that include the preparation, characterization, and application of nanostructures with diverse shapes and sizes in many chemical transformations. Core–shell nanoparticles have recently been subject to wide research for the combined functionalities of cores and shells, which endow them with many possible application potentials in various fields [24].

Nanomaterials, particular metal nanoparticles (MNPs) and supported magnetic metal nanoparticles (S-MMNPs), have appeared as new classes of nanocatalysts. Some important features of these catalysts are simple separation using an external magnet without the need for filtration, high catalytic activity, low toxicity, and a high degree of chemical stability in various organic solvents [25–28]. Iron has a great deal to offer on the nanoscale, including very potent catalytic properties [29]. $Fe_3O_4$ coated with silica was commonly applied as the support of metal and non-metal catalysts [30–34].

In the context of our interest in sustainable approaches in the preparation of heterocyclic compounds and in continuing our attempts towards the advancement of effective and environmentally friendly nanocatalysts in organic synthesis [35–41], herein we report a green and mild method for the preparation of indazolo[1,2-*b*]-phthalazinetriones in high yields and short reaction times using $Fe_3O_4@SiO_2$ core–shell nanoparticles as an effective, green, and reusable catalyst (Scheme 1).

## Experimental

Chemicals were purchased from 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 tubes with a Boetius melting point microscope. 1H NMR and 13C NMR spectra were obtained on a Bruker 400-MHz spectrometer with $CDCl_3$ as solvent using TMS as an internal standard. FT-IR spectrum was recorded on Magna-IR spectrometer 550. The elemental analyses (C, H, N) were obtained from a Carlo ERBA Model EA 1108 analyzer. Powder X-ray diffraction (XRD) was carried out

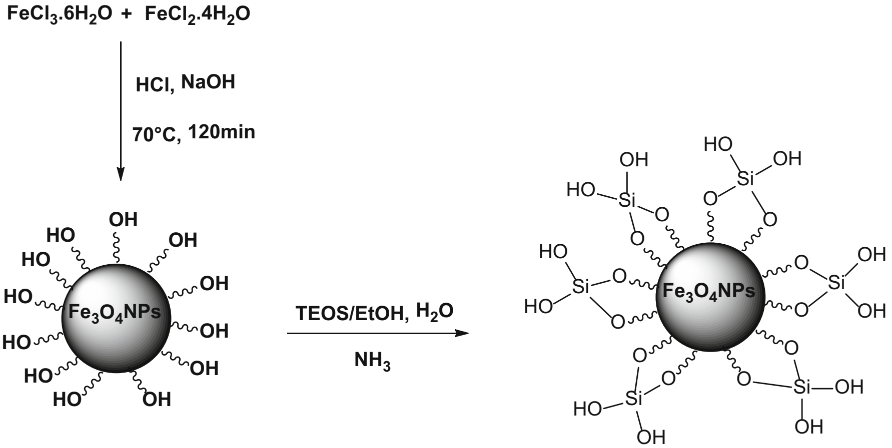


**Scheme 1** $Fe_3O_4@SiO_2$ nanoparticle-catalyzed one-pot synthesis of indazolo[1,2-*b*]-phthalazinetriones derivatives

on a Philips X'pert diffractometer with mono-chromatized Cu-Kα radiation ($\lambda = 1.5406$ Å). Microscopic morphology of products was visualized by SEM (LEO 1455VP). The mass spectra were recorded on a Joel D-30 instrument at an ionization potential of 70 eV. Magnetic properties were obtained on a BHV-55 vibrating sample magnetometer (VSM). The compositional analysis was done by energy-dispersive analysis of X-ray.

## Preparation of $Fe_3O_4$ nanoparticles

$Fe_3O_4$ MNPs were prepared according to a previously reported procedure by Hu et al. [42] using the chemical co-precipitation method. Typically, $FeCl_3$–$6H_2O$ (2.7 g) and $FeCl_2$–$4H_2O$ (1 g) were dissolved in 100 ml of 1.2 mmol $l^{-1}$ aqueous HCl followed by ultrasonic bath for 30 min. Then, 1.25 mol $l^{-1}$ aqueous NaOH (150 ml) was added under vigorous stirring and a black precipitate was immediately formed. The resulting transparent solution was heated to 80 °C with rapid mechanical stirring under $N_2$ atmosphere (Scheme 2). After vigorous stirring for 2 h, the black products were centrifuged, filtered out, and washed with deionized water and alcohol several times, and finally dried at 60 °C for 12 h.



**Scheme 2** Preparation steps for fabricating $Fe_3O_4@SiO_2$

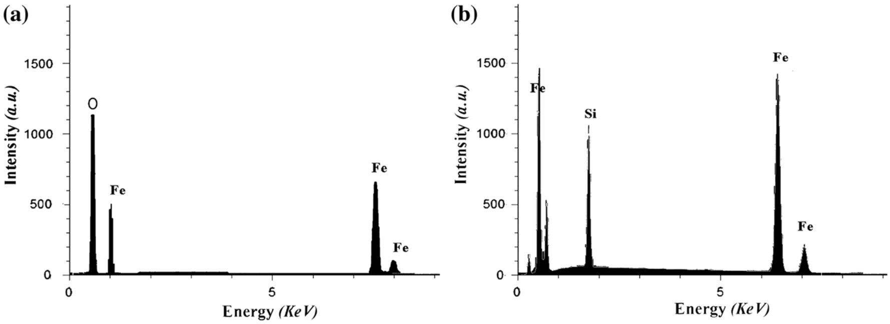


**Fig. 1** The EDX spectra of $Fe_3O_4$ (**a**) and $Fe_3O_4$@$SiO_2$ NPs (**b**)

### Preparation of $Fe_3O_4$@$SiO_2$ nanoparticles

$Fe_3O_4$@$SiO_2$ core–shell particles were prepared via a modified Stöber sol–gel process [43]. Thirty milligrams as-prepared $Fe_3O_4$ submicrospheres were ultrasonically dispersed in a solution containing 160 ml of ethanol, 40 ml of water, and 10 ml of concentrated ammonia (28 wt%). Then, 0.4 ml of TEOS was added dropwise to the solution under sonication, followed by mechanical stirring for 3 h at room temperature. Subsequently, the resulting particles were separated using a magnet and washed with deionized water and ethanol. The step was repeated several times before drying at 60 °C for 12 h.

The chemical purity of the samples as well as their stoichiometry was tested by EDAX studies. The EDAX spectra given in Fig. 1a shows the presence of Fe and O as the only elementary components of $Fe_3O_4$ NPs. EDAX spectrum $Fe_3O_4$@$SiO_2$ (Fig. 1b) shows the elemental compositions are (Fe, Si and O) of core–shell nanoparticles.

In order to study the morphology and particle size of $Fe_3O_4$ nanoparticles, an SEM image of $Fe_3O_4$ nanoparticles is presented in Fig. 2a. These results show that spherical $Fe_3O_4$ NPs were obtained with an average diameter of about 20–30 nm as confirmed by X-ray line broadening analysis. Figure 2b shows that $Fe_3O_4$@$SiO_2$ nanoparticles still keep the morphological properties of $Fe_3O_4$ except for a slightly larger particle size and smoother surface, where silica are uniform coated on the $Fe_3O_4$ particles to form a silica shell in compared to the $Fe_3O_4$@$SiO_2$.

The X-ray diffraction patterns of $Fe_3O_4$ and $Fe_3O_4$@$SiO_2$ are shown in Fig. 3. The position and relative intensities of all peaks conform well with the standard XRD pattern of $Fe_3O_4$ with P63mc group (JCDPS No. 75-0449), indicating retention of the crystalline cubic spinel during functionalization of MNPs. The characteristic peak of $SiO_2$ in core shell structure has been hidden under a weak peak of $Fe_3O_4$ at 2 h = 30. The average MNPs core diameter was calculated to be 25 nm from the XRD results by Scherrer's equation, $D = k\lambda/b\cos h$ where k is a constant (generally considered as 0.94), λ is the wavelength of Cu-Ka (1.54 Å), b is the corrected diffraction line full-width at half-maximum (FWHM), and h is Bragg's angle.

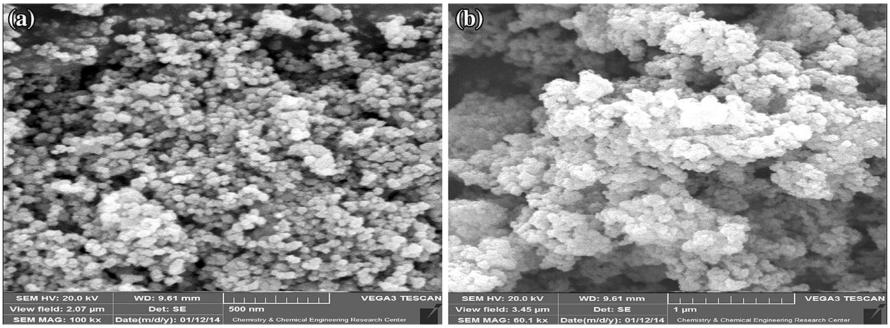


**Fig. 2** SEM images of $Fe_3O_4$ (**a**) and $Fe_3O_4$@$SiO_2$ NPs (**b**)

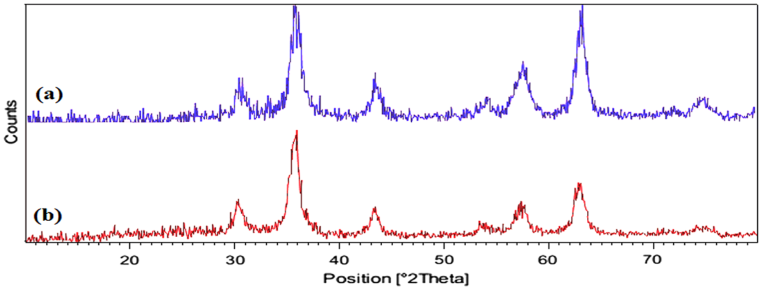


**Fig. 3** XRD patterns of $Fe_3O_4$ (*a*) and $Fe_3O_4$@$SiO_2$ NPs (*b*)

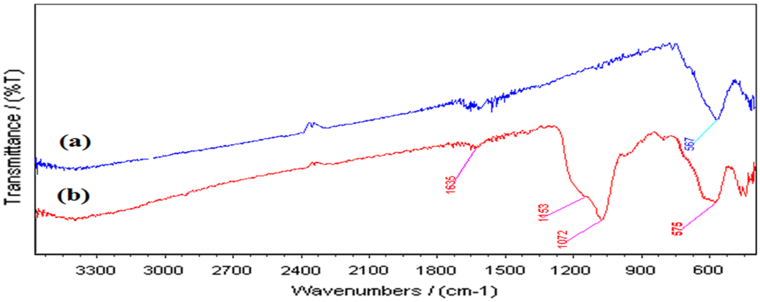


**Fig. 4** The comparative FT-IR spectra of $Fe_3O_4$ (*a*) and $Fe_3O_4$@$SiO_2$ NPs (*b*)

Figure 4 shows the FT-IR spectrum for the samples of $Fe_3O_4$ NPs and $Fe_3O_4$@$SiO_2$ microspheres catalysts. For the bare magnetic nanoparticle (Fig. 3a), the vibration band at 575 $cm^{-1}$ is the typical IR absorbance induced by structure Fe–O vibration. In the case of $Fe_3O_4$@$SiO_2$ nanoparticles (Fig. 3b), the band at 1,072 $cm^{-1}$ corresponds to Si–O–Si antisymmetric stretching vibrations, being indicative of the existence of $SiO_2$ in the nanoparticles.

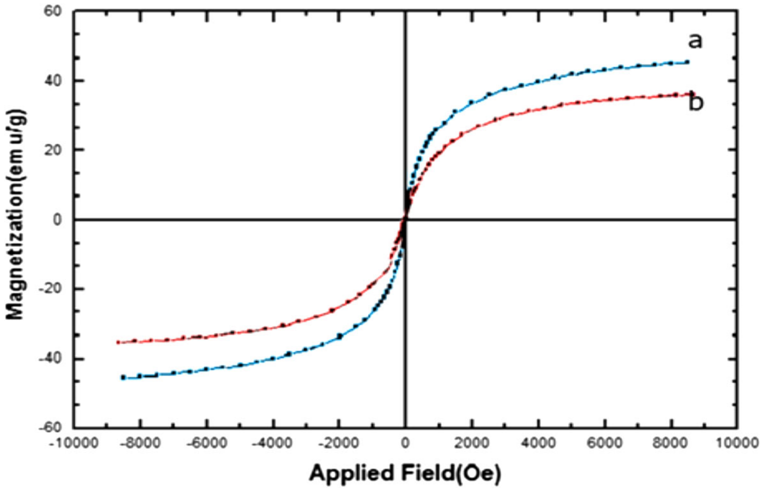


**Fig. 5** Magnetization curves for the prepared of $Fe_3O_4$ (*a*) and $Fe_3O_4@SiO_2$ NPs (*b*)

The magnetic properties of the samples containing a magnetite component were studied by a vibrating sample magnetometer (VSM) at 300 K. Figure 5 shows the absence of hysteresis phenomenon and indicates that all of the products have superparamagnetism at room temperature. The saturation magnetization values for $Fe_3O_4$ (a) and $Fe_3O_4@SiO_2$ (b) were 48.12 and 38.16 emu/g, respectively. These results indicate that the magnetization of $Fe_3O_4$ decreased considerably with the increase of $SiO_2$.

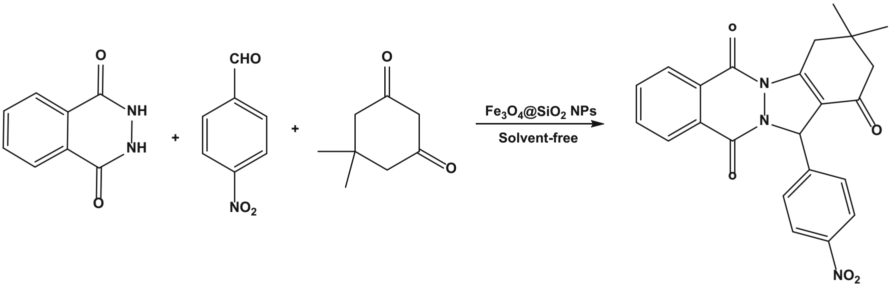
## General procedure for the preparation of indazolo[1,2-*b*]-phthalazinetriones

A mixture of dimedone (0.14 g, 1 mmol), phthalhydrazide (0.16 g, 1 mmol), aldehyde (1 mmol), and $Fe_3O_4$ @ $SiO_2$ (0.0 23 gr, 0.8 mmol, 8 mol%) was heated at 120 °C for 10–20 min. After completion of the reaction as indicated by TLC, the reaction mixture was cooled to room temperature and the solid obtained was dissolved in dichloromethane; the catalyst was insoluble in $CH_2Cl_2$ and separated by using an external magnet. The solvent was evaporated and the residue was recrystallized from ethanol to afford the pure indazolo[1,2-*b*]-phthalazinetriones.

All the products were identified with m.p., 1H NMR, 13C NMR, and FT-IR spectroscopy techniques. Spectral data of the new products are given below.

### *3,4-Dihydro-3,3-dimethyl-13-(4-cyano-phenyl): 2H-indazolo[2,1-b]-phthalazine-1,6,11-(13H)-trione (4p)*

Yellow powder; m.p. = 261–263 °C; $^1$H NMR (400 MHz, $CDCl_3$) 1.21 (3H, s), 1.24 (3H, s), 2.12 (2H, s), 3.26–3.30 (2H, m), 6.42 (1H, s), 7.32–8.41 (8H, m). $^{13}$C NMR (100 MHz, $CDCl_3$): 27.3, 28.3, 35.1, 39.2, 51.1, 65.3, 112.4, 119.1, 126.3, 127.7, 128.4, 128.6, 129.3, 130.1, 132.5, 137.4, 137.9, 150.9, 155.2, 157.1, 193.2.



**Scheme 3** The model reaction for the synthesis of indazolo[1,2-*b*]-phthalazinetriones in the presence of $Fe_3O_4@SiO_2$ NPs

FT-IR (KBr, $cm^{-1}$): 2,952(C–H), 1,655 (C=O), 1,631 (C=C), 1,463 (C=C, Ar), 2,180 (C≡N), Anal. Calcd. For $C_{24}H_{19}N_3O_3$: C 72.53, H 4.82. N 10.57. Found C 72.63, H 4.74. N 10.49.; MS (EI) (*m/z*): 397.14 ($M^+$).

*3,4-Dihydro-3,3-dimethyl-13-(4-formyl-phenyl): 2H-indazolo[2,1-b]-phthalazine-1,6,11-(13H)-trione(4q)*

Yellow powder; m.p. = 265–267 °C; $^1H$ NMR (400 MHz, $CDCl_3$): 1.22 (3H, s), 1.23 (3H, s), 2.32 (2H, s), 3.24–3.32 (2H, m), 6.43(1H, s), 7.25–8.29 (8H, m). $^{13}C$ NMR (100 MHz, $CDCl_3$): 27.9, 28.2, 33.2, 38.6, 51.2, 63.5, 119.1, 121.8, 126.6, 127.9, 128.3, 128.5, 129.4, 130.9, 132.4, 133.7, 136.2, 152.1, 155.6, 157.3, 193.4, 197.3. FT-IR (KBr, $cm^{-1}$): 2,957 (C–H), 1,702(C=O), 1,662 (C=O), 1,632 (C=C), 1,552 (C=C, Ar); Anal. Calcd. For $C_{24}H_{20}N_2O_4$: C 71.99, H 5.03, N 7.00. Found C 71.92, H 5.08. N 7.08.; MS (EI) (*m/z*): 400.14 ($M^+$).

## Results and discussion

In this research, in order to determine the optimized reaction conditions, we selected the reaction of 4-nitrobenzaldehyde, phthalhydrazide, and dimedone as a model study in the presence of $Fe_3O_4@SiO_2$ core–shell nanoparticles (Scheme 3).

Initially, to show the merits of the present work, a comparative study of the catalytic efficiency of $Fe_3O_4@SiO_2$ NPs with other nanocatalysts was examined in the model study. As shown in Table 1, it was observed that $Fe_3O_4@SiO_2$ exhibited high activity in comparison with other nanocatalysts and the corresponding product 4b was performed in excellent yield and short reaction time. It seems that a high surface area and better dispersion of nanoparticles in the reaction mixture are reasons for better activities of $Fe_3O_4@$ $SiO_2$ NPs.

Afterwards, the optimization of catalyst amounts was carried out in the model study using different amounts of the $Fe_3O_4@SiO_2$ NPs. As shown in Table 1, 8 mol% of the magnetic $Fe_3O_4@SiO_2$ NPs and solvent-free conditions are the best operative experimental conditions. Increasing the amount of catalyst does not improve the yield of the product any further, whereas decreasing the amount of

**Table 1** The model study catalyzed in the presence of various catalysts

| Entry | Catalyst (mol%) | Catalyst loading (mol%) | Time (min) | Yield (%)[a] |
|---|---|---|---|---|
| 1 | None | – | 120 | None |
| 2 | ZnO NPs | 15 | 45 | 60 |
| 3 | CuI NPs | 15 | 50 | 65 |
| 4 | MgO NPs | 15 | 120 | 40 |
| 5 | $Fe_3O_4$ NPs | 15 | 40 | 70 |
| 6 | AgI NPs | 15 | 50 | 60 |
| 7 | CaO NPs | 15 | 140 | 35 |
| 8 | *MCM-41* | 15 | 25 | 78 |
| 9 | $Fe_3O_4$–$SiO_2$ | 15 | 12 | 96 |
| 10 | $Fe_3O_4$–$SiO_2$ | 10 | 12 | 96 |
| **11** | **$Fe_3O_4$–$SiO_2$** | **8** | **12** | **96** |
| 12 | $Fe_3O_4$–$SiO_2$ | 5 | 20 | 90 |
| 13 | $Fe_3O_4$–$SiO_2$ | 2 | 35 | 60 |

Reaction conditions: phthalhydrazide (1 mmol), dimedone (1 mmol) and 4-nitro-benzaldehyde (1 mmol) under solvent-free conditions at 120 °C

[a] Isolated yields

Bold value indicates best result

catalyst leads to a decrease in the product yield. Hence, the optimum concentration of $Fe_3O_4@SiO_2$ was chosen as 8 mol% in the model reaction.

In continuation of this research, the model study was performed several times in order to provide the optimum ratio of the reactants. We observed that using aldehyde, dimedone, and phthalhydrazide in a 1:1:1 mol ratio afforded the product in excellent yields and short reaction times.

To study the effect of temperature in the reaction of 4-nitro-benzaldehyde, phthalhydrazide, and dimedone, the model study was carried out under various temperatures. The resulting Table 2 shows that by increasing temperatures from room temperature to 140 °C, the yield of formation of product was increased. Among different temperatures, 120 °C was chosen as the optimum temperature. A further increase in temperature from 120 to 140 °C in the mentioned reaction had no significant effect on the time and product yield.

In another effort to investigate the effect of solvent and as well as solvent-free conditions, we carried out a test reaction in the presence of various solvents under reflux conditions and the results are presented in Table 3. It is observed that excellent results were obtained under solvent-free conditions at 120 °C using $Fe_3O_4@SiO_2$ as a catalyst.

The significant results of the above-mentioned experiments prompted us to investigate the catalytic activity of $Fe_3O_4@SiO_2$ in the synthesis of indazolo[1,2-*b*]-phthalazinetrione derivatives. Thus, we used optimized reaction conditions in the presence of $Fe_3O_4@SiO_2$ NPs to produce various indazolo[1,2-*b*]-phthalazinetriones.

A series of experiments were carried out and as a result of these we prepared a number of indazolo[1,2-*b*]-phthalazinetriones in high yields and short reaction

**Table 2** Effect of temperature for the preparation of indazolo[1,2-*b*]-phthalazinetriones using $Fe_3O_4@SiO_2$

| Entry | T (°C) | Time (min) | Yield (%)[a] |
|---|---|---|---|
| 1 | r.t | 120 | 30 |
| 2 | 50 | 80 | 50 |
| 3 | 80 | 45 | 60 |
| 4 | 100 | 25 | 70 |
| **5** | **120** | **12** | **96** |
| 6 | 140 | 12 | 96 |

Reaction conditions: phthalhydrazide (1 mmol), dimedone (1 mmol) and 4-nitro-benzaldehyde (1 mmol), $Fe_3O_4@SiO_2$ 8 mol% under solvent-free conditions at 120 °C

[a] Isolated yields

Bold value indicates best result

times. We observed that various aryl aldehydes could be introduced in high efficiency and produce high yields of products in high purity.

As seen from Table 4, aromatic aldehydes with electron-withdrawing groups such as $NO_2$ and Cl reacted faster than those with electron-releasing groups such as isopropyl and methoxy as expected; also sterically hindered aromatic aldehydes required longer reaction times.

To study the scope of this procedure, we next used a diversity of aldehydes to investigate three component reactions under the optimized conditions. $Fe_3O_4@SiO_2$ NPs was thus shown to be an effective catalytic system, which gave the desired products in excellent yields.

The reusability of the catalyst was examined by repeating the model reaction using $Fe_3O_4@SiO_2$ nanoparticles under optimized reactions. The results of these experiments showed that the catalytic activity of nano $Fe_3O_4@SiO_2$ did not decrease significantly even after five catalytic cycles (Table 5).

## Proposed mechanism

A plausible mechanism for the synthesis of 2*H*-indazolo[2,1-*b*]-phthalazine-1,6,11-trione(13H)-trione using $Fe_3O_4@SiO_2$ NPs is shown in Scheme 4. On the basis of our experimental results together with some literature [22, 44, 45], it is likely that $Fe_3O_4@SiO_2$, as a Lewis acid, increases the electrophilicity of the carbonyl groups on the aldehyde and dimedone by means of a strong coordinate bond.

The first step is believed to be the Lewis acid-catalyzed Knoevenagel condensation between the aldehyde and dimedone to generate adduct **A**, which acts as a Michael acceptor. The phthalhydrazide attacks adduct **A** in a Michael-type fashion to produce an open-chain intermediate **B**. Intermediate **B** undergoes intramolecular cyclization by the reaction of nucleophilic amino function to carbonyl group followed by dehydration to form 2*H*-indazolo[2,1-*b*]-phthalazine-1,6,11-trione(13H)-trione.

**Table 3** Effect of solvents on the model reaction in the presence of $Fe_3O_4@SiO_2$ NPs

| Entry | Solvent | Time (min) | Yield (%)[a] |
|---|---|---|---|
| 1 | EtOH | 110 | 40 |
| 2 | DMF | 120 | 50 |
| 3 | $H_2O$ | 130 | 35 |
| 4 | $CH_3CN$ | 120 | 45 |
| 5 | Toluene | 240 | 25 |
| **6** | **Solvent-free** | **12** | **96** |

Reaction conditions: phthalhydrazide (1 mmol), dimedone (1 mmol) and 4-nitro-benzaldehyde (1 mmol) using 8 mol% of $Fe_3O_4@SiO_2$

[a] Isolated yields

Bold value indicates best result

**Table 4** Preparation of indazolo[1,2-*b*]-phthalazinetriones using $Fe_3O_4@SiO_2$ NPs under solvent-free conditions

| Entry | R | R′ | Product | Time (min) | Yield (%)[a] | M.p. (°C) | Lit. M.p. (°C) |
|---|---|---|---|---|---|---|---|
| 1 | H | $CH_3$ | 4a | 16 | 91 | 208–210 | 207–209 [10] |
| 2 | 4-$NO_2$ | $CH_3$ | 4b | 12 | 96 | 220–221 | 223–225 [10] |
| 3 | 4-Cl | $CH_3$ | 4c | 10 | 95 | 262–264 | 262–264 [10] |
| 4 | 4-Br | $CH_3$ | 4d | 14 | 93 | 258–260 | 265–267 [10] |
| 5 | 3-$NO_2$ | $CH_3$ | 4e | 17 | 92 | 269–272 | 270–272 [10] |
| 6 | 4-OH | $CH_3$ | 4f | 20 | 88 | 258–260 | 265–266 [15] |
| 7 | 4-$OCH_3$ | $CH_3$ | 4g | 18 | 85 | 218–220 | 218–220 [12] |
| 8 | 4-Isopropyl | $CH_3$ | 4h | 20 | 85 | 196–197 | 195–197 [46] |
| 9 | 3-$CH_3$ | $CH_3$ | 4i | 17 | 87 | 232 | 232–233 [15] |
| 10 | 3-$CF_3$ | $CH_3$ | 4j | 12 | 94 | 214 | 213–215 [15] |
| 11 | 2,4-$Cl_2$ | $CH_3$ | 4k | 15 | 92 | 220 | 219–221 [13] |
| 12 | 4-$(CH_3)_2N$ | $CH_3$ | 4l | 16 | 88 | 256 | 255–257 [12] |
| 13 | 3-OH | $CH_3$ | 4m | 20 | 85 | 267 | 266–268 [47] |
| 14 | 4-F | $CH_3$ | 4n | 15 | 94 | 224–226 | 217–219 [10] |
| 15 | 4-$CH_3$ | $CH_3$ | 4o | 17 | 91 | 227–228 | 227–229 [10] |
| 16 | 4-CN | $CH_3$ | 4p | 14 | 94 | 213–215 | –[b] |
| 17 | 4-CHO | $CH_3$ | 4q | 15 | 92 | 233–234 | –[b] |

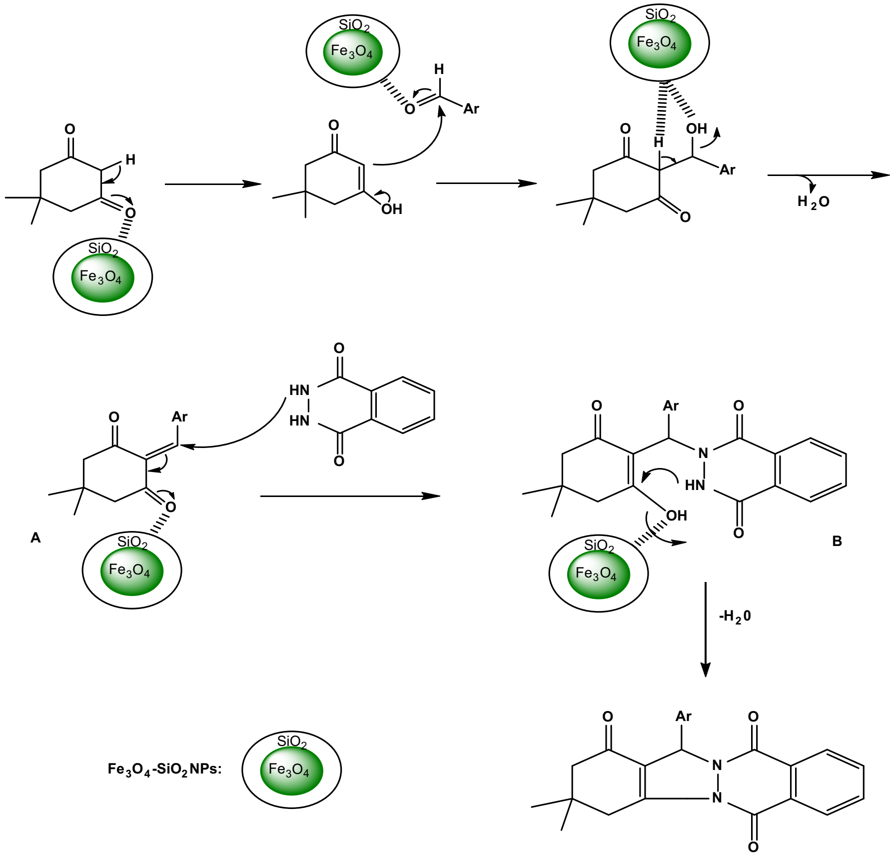
[a] Isolated yield

[b] New products

**Table 5** The catalyst reusability for the synthesis of indazolo[1,2-*b*]-phthalazinetriones

| Cycle | First | Second | Third | Fourth | Fifth |
|---|---|---|---|---|---|
| Yield (%)[a] | 96 | 94 | 93 | 89 | 87 |

[a] Yields refer to the isolated pure product



**Scheme 4** Proposed mechanism for the synthesis of indazolo[1,2-*b*]-phthalazinetriones catalyzed by $Fe_3O_4$@$SiO_2$ NPs

## Conclusions

In summary, an efficient protocol for the one-pot preparation of 2*H*-indazolo[2,1-*b*]-phthalazine-1,6,11(13*H*)-trione derivatives from the three-component condensation reaction of phthalhydrazide, dimedone, and aromatic aldehydes using $Fe_3O_4$@$SiO_2$ nanoparticles as a commercially available, environmental friendly and reusable catalyst was described. The reactions were carried out under thermal solvent-free conditions in short reaction times and produced the corresponding products in good to excellent yields. The catalyst could also be successfully recovered and recycled at least for five runs without any significant loss in activity. The one-pot nature and the use of heterogeneous solid acid as an eco-friendly catalyst make it an interesting alternative to multi-step approaches.

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