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FULL PAPER

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# Supramolecular $Fe_3O_4$@PEG/−diaza crown ether@Ni: a novel magnetically reusable nano catalyst for the clean synthesis of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones

Michael Aalinejad[1] | Nader Noroozi Pesyan[1] | Nosrat Heidari[2] | Hana Batmani[1] | Aria Danandeh Asl[1]

[1] Department of Organic Chemistry, Faculty of Chemistry, Urmia University, 57159 Urmia, Iran

[2] Department of Physical Chemistry, Faculty of Chemistry, Urmia University, 57159 Urmia, Iran

**Correspondence**
Nader Noroozi Pesyan, Department of Organic Chemistry, Faculty of Chemistry, Urmia University, 57159, Urmia, Iran.
Email: n.noroozi@urmia.ac.ir; nnp403@gmail.com

Ni@diaza crown ether complex supported on magnetic nanoparticle was provided by grafting technique. The catalytic activity of $Fe_3O_4$@diaza crown ether@Ni was explored through one-pot synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones and it was used as an efficient and recoverably constant nanocatalyst. FT-IR, SEM, TEM, XRD, BET, ICP, EDS, and TGA techniques were employed to specify the nanocatalyst. This heterogeneous catalyst demonstrated acceptable recyclability and could be used again several times with no considerable loss of its catalytic activity.

**KEYWORDS**
2-Aryl-2,3-dihydroquinazolin-4(1*H*)-ones, core-shell, diaza crown ether, magnetic nanoparticles

## 1 | INTRODUCTION

Shallow functionalization of nano-magnetic nanoparticles (MNPs) can be used as a well-organized system to join the hole among homogeneous and heterogeneous catalysis. Magnetite is a good-identified material which is known as ferrite and/or $Fe_3O_4$. It is worth to mention that this material is utilized to gain stable and magnetically recyclable heterogeneous catalysts for some homogeneous catalytically active metals like Cu, Ni, Co, and so on.[1] Nowadays, magnetic nanoparticles have attracted important consideration because of their various applications in catalysis and synthesis.[2,3] The superficial of MNPs could be functionalized basically through fitting surface adjustments to enable the loading of a diversity of desirable functionalities.[4] $Fe_3O_4$ magnetic nanoparticles with extensive particular high adsorption ability and surface area can be parted from fluid under external magnetic field.[5] Recently, nanocomposites of organic materials and inorganic nanoparticles have been of great concern because of their applications in rapid and high-capacity optical and magnetic information storage media.[6–14] Nanoparticles have gotten considerable attention in previous two decades due to the fascinating properties related to their size.[15] In scientific investigation, magnetite ($Fe_3O_4$) particles have been study extensively, because their synthesis is comfortable and good known.[16]

$Fe_3O_4$ particles with coated using various ligands have also been studied such as; $Fe_3O_4$–Adenine-Ni nanocatalyst for the one-pot synthesis of polyhydroquinoline derivatives, oxidation of sulfides to sulfoxides and oxidation of thiols to disulfides,[17] Immobilized $Fe_3O_4$@AMPD@Ni,[18] $Fe_3O_4$@AMPD@Cu[19] and $Fe_3O_4$-Adenin-Zn nanoparticles[20] as a green and recyclable catalyst for synthesis of 5-substituted 1*H*-tetrazoles and oxidation reactions.

Quinazolinones are a set of attached heterocycles that have received much consideration due to their possible pharmaceutical and biological activities. In all the derivatives, quinazolinones have attracted much more consideration owing to their activities for example antibacterial,[21] diuretic[22] and anticancer.[23] There are numerous approaches to prepare this stage of

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compounds.[24–29] Among them, the use of MCM-41-Biurea-Ni,[28] $Fe_3O_4$-Serin-Pd(0)[30] as a reusable nanocatalysts and straightest approach includes condensation of alkyl, aryl aldehydes with anthranilamide in the attendance of *p*-toluenesulfonic acid as a catalyst.[29,31] It should be mentioned that the usage of heterogeneous and recyclable catalysts presents one of the strongest and greenways in chemical technology. Naturally, benignant solid catalysts for example clays and zeolites rather than mineral acids are utilized for acid-catalyzed organic reactions.[32]

In this research, we introduced an interesting magnetically nanocatalyst $Fe_3O_4$@diaza crown ether@Ni, a novel magnetically reusable nanocatalyst for the clean synthesis of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones.

## 2 | RESULT AND DISCUSSION

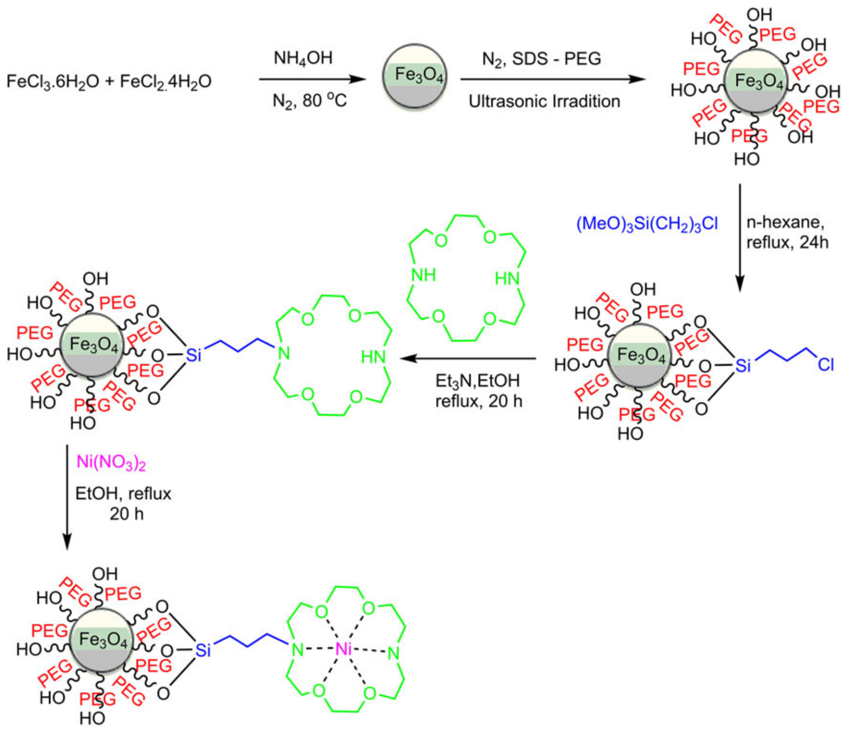
Our research on utilization of new heterogeneous and retrievable catalysts in organic transformations encouraged us to provide a new heterogeneous catalyst ($Fe_3O_4$@diaza crown ether@Ni) and to study its utilization in the synthesis of 2-aryl-2,3-dihydroquinazolin-4(1*H*)-ones. Scheme 1 represents the details of the catalyst preparation process.

### 2.1 | Catalyst characterization

Ni complex supported on nanoporous MNPs, is entirely specified by SEM, EDS, FT-IR, XRD, BET, ICP and TGA techniques. Firstly, Fourier transform infrared (FT-IR) spectra were entered discretely at different steps of catalyst preparation; see the curves (**a-c**) (Figure 1). From these curves, it can be said that the peaks between 583 and 625 $cm^{-1}$ were attributed to Fe–O bond vibration of $Fe_3O_4$. In addition, the absorption bands at 1350, 1244, 1108 $cm^{-1}$ are determinable to the symmetric and asymmetric stretching vibrations of the mesoporous structure (Si-O). Peaks at 2871.42 and 2921.52 $cm^{-1}$ were the C–H stretching vibration absorption peak of crown ether, n-propyl chain and PEG. The band at 3428 $cm^{-1}$ is assigned to the stretching vibration of the O-H groups. The peaks at around 1736–1636 $cm^{-1}$ shown in (b) and (c) demonstrated that crown ether was linked to the *n*Pr-MNPs. The absorption bands at 1106 and 1109 $cm^{-1}$ can be both specified to ether bond.

SEM images of the synthesized MNPs@diaza crown ether@Ni are depicted in Figure 2 (a-c). The SEM micrographs corroborate that samples have tidy and globular morphology. The morphological changes were checked by this technique that is shown in Figure 2 (b and c), the catalysts no remarkable changes in the plane morphology happened during plane modification. In addition, the morphology of the solid was saved. The SEM image demonstrate that the catalyst is as nano particles with the size distribution ranged from 19 to 25 nm.

To show the supporting nickel metal on MNPs surface, the EDS analysis of $Fe_3O_4$ –diaza crown ether-Ni was accomplished (Figure 3). The results show the existence of Fe, Si, Ni, O, N, and C in functionalized MNPs and also



**SCHEME 1** Synthesis of MNPs@diaza crown ether@Ni

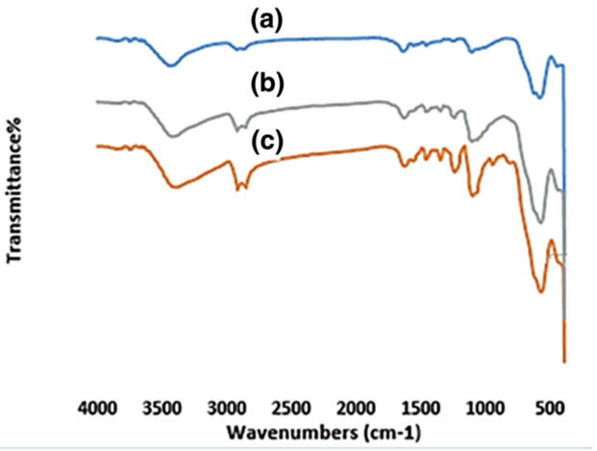


**FIGURE 1** FT-IR spectra of $Fe_3O_4$ (a), $Fe_3O_4$@*n*Pr-Cl (b), $Fe_3O_4$@ *n*Pr-aza crown ether@Ni (c)

demonstrated that the mass percents are 76.07%, 3.70%, 1.24%, 9.87%, 2.86% and 6.26%, respectively.

TEM micrograph has been a suitable tool for determining the particle shape and size distribution. As it can be seen in Figure 4, the magnetic cores of the nanoparticles were uniform in both shape and in size. It should be mentioned that the size of the MNPs@diaza crown ether@Ni nanocatalyst is around 20–30 nm.

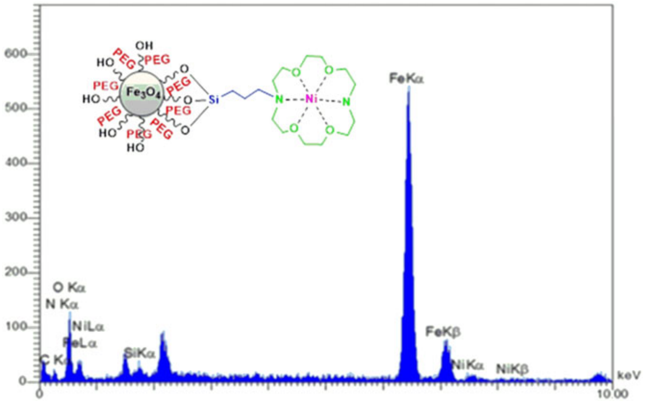


**FIGURE 3** EDS spectrum of $Fe_3O_4$@diaza crown ether@Ni nanocatalyst

Figure 5 demonstrates the low angle X-ray powder refraction pattern of samples $Fe_3O_4$, $Fe_3O_4$@diaza crown ether@Ni. For the XRD pattern of $Fe_3O_4$, the peaks at 30.2°, 35.5°, 40.1°, 57.3°, and 62.9° were respectively equivalent to the (220), (311), (400), (511), and (440) crystal planes of a pure $Fe_3O_4$ with a cubic spinal structure.[33,34] According the XRD pattern of $Fe_3O_4$@diaza crown ether@Ni, all the peaks of $Fe_3O_4$ have been observed. It should be noted that the peaks at 16.5°, 34°,

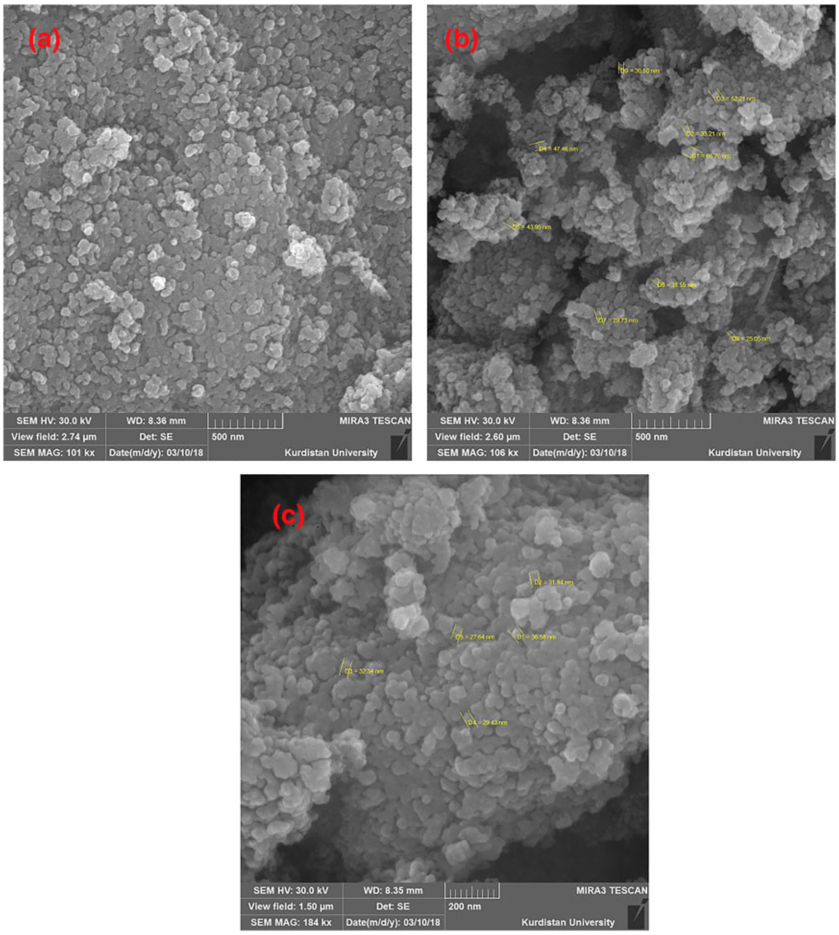


**FIGURE 2** SEM images of a) $Fe_3O_4$ and b,c) $Fe_3O_4$@diaza crown ether@Ni

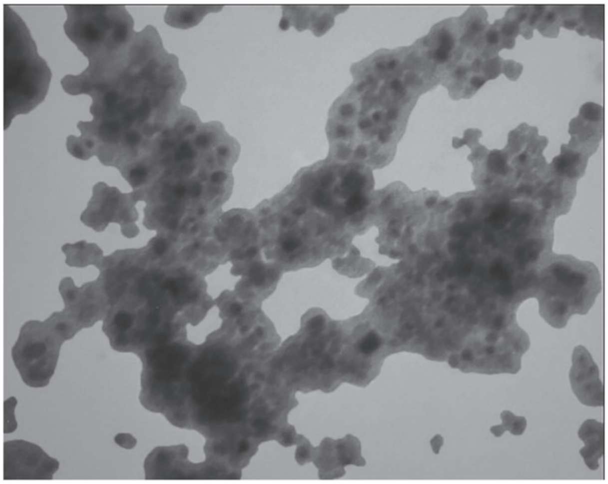


**FIGURE 4** TEM image of $Fe_3O_4$@diaza crown ether@Ni nanocatalyst

39.6°, 56.1°, 62.9°, 63.5°, 67.3° and 72.6° correspond to orthorhombic aza crown ether-Ni.

Figure 6 is indicative of the thermal gravimetric analysis (TGA) curves of (a) MNPs@diaza crown ether@Ni and (b) TGA/DTA analysis of MNPs@diaza crown ether@Ni which shows a weight loss due to the decomposition upon heating the small amount of weight loss below 200 °C is attributed removal of solvent molecules. The weight loss within range of 200–800 °C is associated with the decomposition of organic functional groups located inside framework. The TGA diagrams confirmed the formation of Ni complex.

The magnetization curves of MNPs@diaza crown ether@Ni gained from the vibration sample magnetometer (VSM), is shown in Figure 7 and determined that the severity of the saturation magnetization (Ms) of MNPs@diaza crown ether@Ni were 15.00 emu $g^{-1}$. Moreover, the magnetic particles has super paramagnetism due to the zero forcible energy and since it is free of hysteresis.

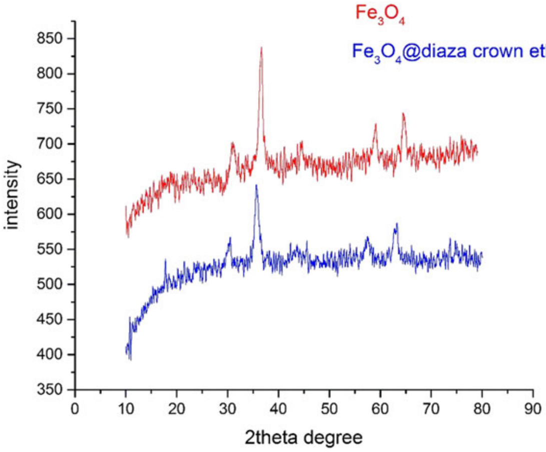


**FIGURE 5** The XRD patterns of a) $Fe_3O_4$ nanoparticles b) $Fe_3O_4$@diaza crown ether@Ni

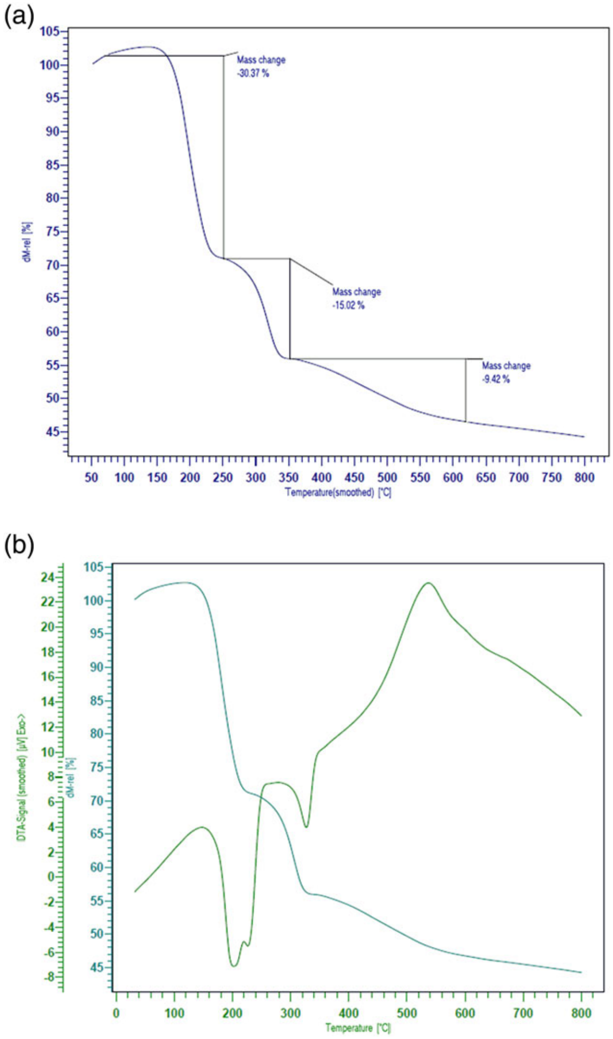


**FIGURE 6** (A) TGA thermograms, (B) TGA/DTA analysis of MNPs@diaza crown ether@Ni nanocatalyst

The hole properties of the made microspheres have been specified by nitrogen absorption. The specific plane areas computed using the Brunauer–Emmett–Teller (BET) technique for the synthesized MNPs@diaza crown ether@Ni catalyst and it was 7.573 $m^2.g^{-1}$. The hole

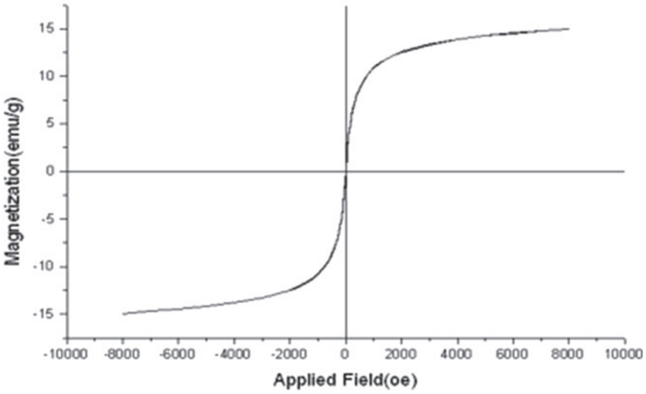


**FIGURE 7** VSM curve of MNPs@diaza crown ether@Ni nanocatalyst

volume was 4.838 $cm^3.g^{-1}$ (Figure 8). It should be mentioned that the corresponding hole size distributions of the catalyst was specified as 2.555 nm which has been done using the Barrett–Joyner–Halenda (BJH) technique. Note that MNPs@diaza crown ether@Ni catalyst is mesoporous ($2 < Dv < 50$ nm).

## 2.2 | Synthesis of quinazolines compounds by catalytic amounts of MNPs@diaza crown ether@Ni

We checked catalytic activity of $Fe_3O_4$@diaza crown ether@Ni as a new heterogeneous catalyst in the one-pot synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones. Towards this end, the reaction of 2-aminobenzamide and benzaldehyde was explored to establish the ideal conditions. Optimization of the catalyst for the synthesis of the desired quinazoline in attendance various catalytic amount of $Fe_3O_4$@diaza crown ether@Ni is shown in Table 1. The best results were gained with benzaldehyde (1.0 mmol) and 2-aminobenzamide (1.0 mmol) in the presence of MNPs@diaza crown ether@Ni (8 mg) and PEG (2 ml) at room temperature (Table 1, entry 2). It should be noted that the use of lower amounts of catalyst (6 mg) does not have any improvement on the outcome (Table 2, entry 3). The reaction of different aldehydes with 2-aminobenzamide were done just after successfully synthesizing of 2,3-dihydroquinazolin-4(1*H*)-ones. The obtained results have been reported in Table 2. From this Table, it can be concluded that a broad limit of aldehydes can react with 2-aminobenzamide and as a result, compounds **3** are obtained (For more information, see Supporting information).

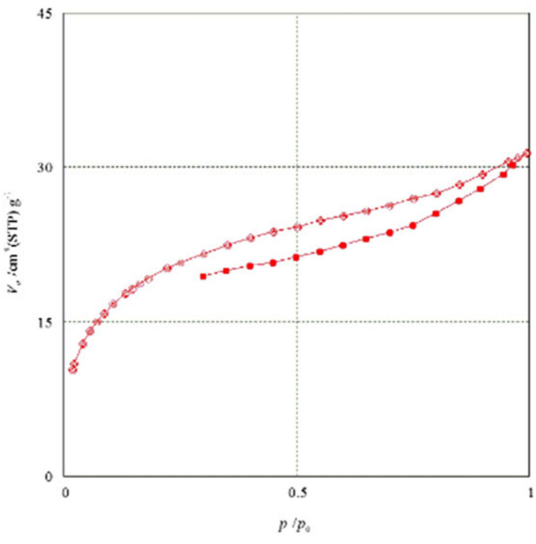
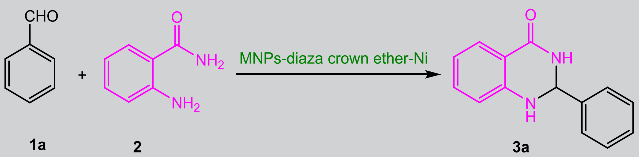


**FIGURE 8** Nitrogen adsorption–desorption isotherm of MNPs@diaza crown ether@Ni nanocatalyst

**TABLE 1** Effects of various parameters on the synthesis of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one



| Entry | Cat. (mg) | Solvent | Temp (°C) | Time (min) | Yield (%) |
|---|---|---|---|---|---|
| 1 | 0 | PEG | rt | 200 | Trace |
| 2 | 8 | PEG | rt | 50 | 95 |
| 3 | 6 | PEG | rt | 60 | 90 |
| 4 | 8 | PEG | 60 | 30 | 87 |
| 5 | 8 | EtOH | 70 | 40 | 91 |
| 6 | 8 | EtOAC | 70 | 100 | 80 |
| 7 | 8 | $CH_2Cl_2$ | 30 | 120 | 85 |

Reaction conditions: Aldehyde (1.0 mmol), 2-aminobenzamide (1.0 mmol).

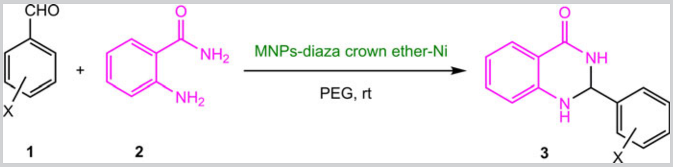
The productivity and capability of MNPs@diaza crown ether@Ni system in synthesis of quinazolines were highlighted by comparison of the gained result with other reported systems (Table 3). As shown in this table, The MNPs@diaza crown ether@Ni is the best catalyst for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives in green solvent under gentle reaction conditions. This catalyst has important characteristics such as eco-friendly, commercially available, cheap and chemically stable materials and also the low cost.

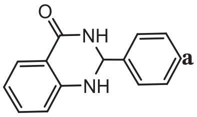
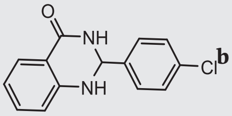
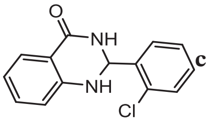
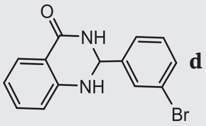
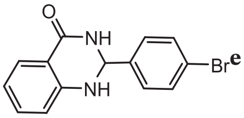
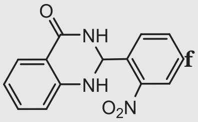
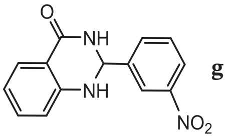
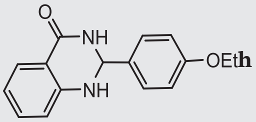
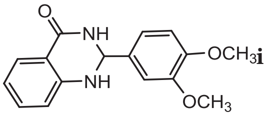
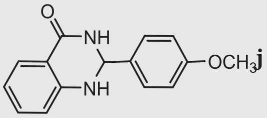
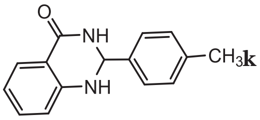
## 2.3 | Recyclability of the catalyst

From a practical point of view, the reusability of catalysts is a significant privilege. To consider the optimal reaction situation (Table 2, entry 1), we checked the recycling of the catalyst. When the reaction was done, the catalyst was trapped by an external magnet, washed with $Et_2O$, and used again 6 runs with no remarkable reduction in activity (Figure 9). The mediocre isolated yield for 6 runs was 93%, which shows the applied reusability of this catalyst. Reproducible catalytic activity and remarkably low leaching of Ni from MNPs functionalized with diaza crown ether were seen. Based on ICP-OES measurement, the measured Ni has been specified for unreacted catalyst (2.5%) and after sixth run (2%).

To test the leaching of nickel in the reaction mixture and the heterogeneity of the catalyst, we accomplished hot filtration for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones with 2-aminobenzamide and various derivatives of aldehydes. In this test, we got the yield of product in first twenty minutes of the reaction of 32%. Then, the

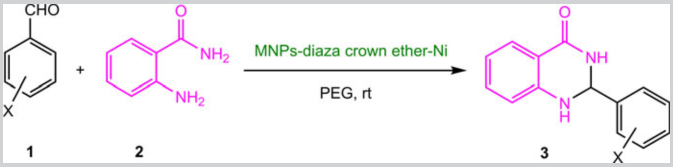
**TABLE 2** MNPs@diaza crown ether@Ni catalyzed the one-pot synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones in PEG

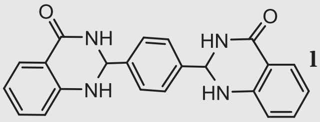


| Entry | Product (3) | Time (min) | Yield(%) | MP (°C) [ref] |
|---|---|---|---|---|
| 1 |  | 25 | 92 | 222–225[30,35] |
| 2 |  | 25 | 88 | 200–202[30,36] |
| 3 |  | 10 | 73 | 230–231[30,37] |
| 4 |  | 15 | 92 | 185–187[30,38] |
| 5 |  | 15 | 95 | 197–199[30,35] |
| 6 |  | 10 | 87 | 190–192[30,37] |
| 7 |  | 5 | 92 | 179–182[37] |
| 8 |  | 50 | 77 | 168–170[38] |
| 9 |  | 35 | 91 | 209–211[38] |
| 10 |  | 40 | 80 | 189–191[30,35] |
| 11 |  | 30 | 95 | 225–226[30,35] |

(Continues)

**TABLE 2** (Continued)



| Entry | Product (3) | Time (min) | Yield(%) | MP (°C) [ref] |
|---|---|---|---|---|
| 12 |  l | 50 | 93 | 241–243[38] |

The products were specified by comparing their spectral and physical data with the valid samples.

catalyst was separated and the filtrate permitted to react further. After this hot filtration, it was concluded that no further reaction was seen. The yield of reaction in this stage was 32%, corroborating the leaching of nickel was insignificant (Figure 10).

# 3 | EXPERIMENTAL

## 3.1 | Materials and instruments

All substrates and reagents were obtained from Merck and Aldrich chemical companies in high purity and used with no further purification. As it has been mentioned earlier, all the products were characterized by valuation their spectral data and physical with valid samples. KBr pellets using a Nexus 670 FT-IR spectrometer (Urmia University, Urmia, Iran) was employed to perform FT-IR spectra. TLC was also used to monitor the reaction over silica gel 60 $F_{254}$ aluminum sheet. The X-ray diffraction (XRD) measurements were collected by a X'PertPro with wavelength 1.54 Å while some diffraction patterns were entered in the 2θ range (10–80°). The EDX of the MNPs were done utilizing a FESEM TESCAN MIRA3 (University of Kurdistan, Sanandaj, Iran). The particle morphology was checked by SEM using FESEM-TESCAN MIRA3 (University of Kurdistan, Sanandaj, Iran). Bruker Avance spectrometers, 300 for $^{1}$H and 75 MHz for $^{13}$C was employed in order to obtain the $^{1}$H, $^{13}$CNMR spectra (Urmia University, Urmia, Iran). The Barrett–Joyner-Halanda (BJH) method was utilized and the pore volume and pore measure distribution were resulted from the desorption profiles of the isotherms. TGA curves were entered using a Shimadzu DTG- 60 instrument (University of Kurdistan, Sanandaj, Iran). In addition, the Brunauer–Emmett–Teller (BET) technique was used to measure the specific surface area of instance.

**TABLE 3** Comparison of the synthesis of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one with MNPs@diaza crown ether@Ni and other reported systems

| Entry | Catalyst | Temp °C | Time (min) | Yield (%) | Lit. |
|---|---|---|---|---|---|
| 1 | MNPs–diaza crown ether-Ni | rt | 50 | 95 | This work |
| 2 | Sc (OTf)3 | 70 | 25 | 92 | [39] |
| 3 | PPA-SiO2 | 70 | 90 | 91 | [40] |
| 4 | Ga (OTf)3 | 70 | 55 | 87 | [41] |
| 5 | TFE | 78 | 90 | 90 | [42] |
| 6 | TABA | 100 | 90 | 82 | [43] |
| 7 | NaHSO4 | 70 | 30 | 94 | [44] |

## 3.2 | Preparation of nanoparticle $Fe_3O_4$@PEG (MNPs)

In a typical procedure, in a two-neck flask, $Fe_3O_4$ nanoparticles were synthesized by $FeCl_3.6H_2O$ and $FeCl_2.4H_2O$ according to the reported method.[45] Under ultrasonic irradiation, $Fe_3O_4$@PEG magnetic nanoparticles were prepared. In a 250 ml, three-necked flask $Fe_3O_4$ nanoparticles (1.0 g), deionized water (50 ml) and a solution of 25 ml sodium dodecyl sulfate (20 mol%)

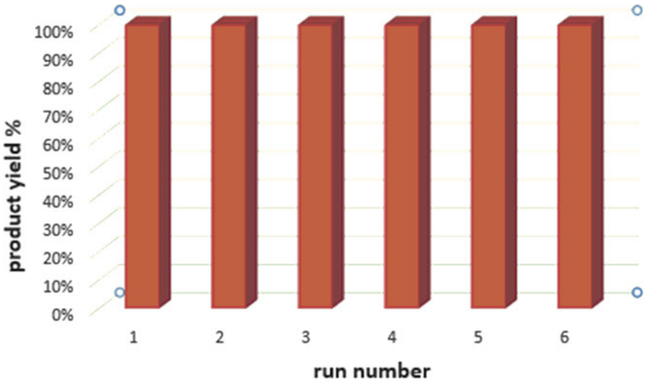


**FIGURE 9** Recycling of MNPs@diaza crown ether@Ni in the synthesis of 2-phenyl-2,3 dihydroquinazolin 4(1*H*)-one

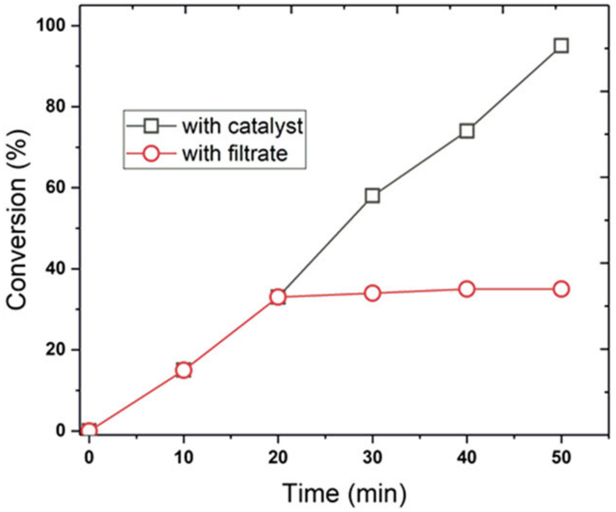


**FIGURE 10** Hot filtration diagram for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones

were mixed under nitrogen climate. For half an hour, the obtained mixture was kept under ultrasonic vibration with 95 W. Next, for an hour, the PEG-400 solution (25 ml) was added to the suspension again under ultrasonic vibration. Then, the obtained products were purified by magnetic field separation. Finally, for nine hrs, the products were dried under vacuum at 50 °C.[46]

### 3.3 | Preparation of MNPs@$(CH_2)_3Cl$

The obtained MNPs powder (4.8 g) with of 3-chloropropyltrimethoxysilane (CPTMS) (5.0 g) was added to *n*-hexane (90 ml) and the combination under refluxing and nitrogen atmosphere was stirred for 24 hrs. An external magnet was employed to collect the resulting solid where it was washed with *n*-hexane. At the last step, the solid was dried under vacuum in order to obtain MNPs@$(CH_2)_3$-Cl.

### 3.4 | Preparation of $Fe_3O_4$@PEG -diaza crown ether

In a 100 ml round bottom flask a mixture of MNPs@$(CH_2)_3Cl$ (1 g), aza crown ether (1 g) and $Et_3N$ (2 ml) in EtOH (50 ml) under reflux was stirred for 20 hrs. Subsequently, the product was washed for many times with EtOH. Finally, for 12 hrs, it was dried in vacuum oven at 70 °C.

### 3.5 | Preparation of $Fe_3O_4$@PEG -diaza crown ether@Ni

The functionalized MNPs@diaza crown ether (3 g) was blended with Ni $(NO_3)_2.6H_2O$ (1.5 g) in EtOH (50 ml) and stirring under reflux for 20 hrs. Then, the resulting solid was collected by an external magnet. Next, in order to remove any unanchored metal ion, it was washed for many times with EtOH and dried in vacuum at 70 °C for 12 hrs. Finally, the catalyst gained is designated as MNPs@diaza crown ether@Ni.

### 3.6 | General procedure for the preparation of 2,3-dihydroquinazolin-4 (1*H*)-ones compounds

In a 25 ml round bottom flask equipped by a magnetically stirrer, dissolved aldehydes (1 mmol), 2-aminobenzamide (1 mmol), MNPs@diaza crown ether@Ni (5 mg) in 2 ml PEG. The blend was mixed at the room temperature for a suitable reaction period of time (Table 2). TLC was employed to check the advance of the reaction. After the termination of the reaction, the catalyst was separated by an external magnet and washed with ethanol, vacuum dried and stockpiled for other runs. The combination was extracted with $H_2O$ and Ethyl acetate. Finally, the solid residue was recrystallized in minimum hot EtOH, afforded 2,3-dihydroquinazolin-4(1*H*)-ones in good yields. Then, by FT-IR, $^1H$ and $^{13}C$ NMR spectroscopy data reaction products **3** were specified (See Supporting Information).

## 4 | CONCLUSION

In closing, $Fe_3O_4$@diaza crown ether@Ni nanocomposite was synthesized by means of core-shell method. The catalytic activity of $Fe_3O_4$@diaza crown ether@Ni was explored through one-pot synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones. This catalyst had high conversion amount and could be easily regained and reused many times with no considerable loss of its catalytic activity. To sum up, the proposed catalyst is useful and adsorbent for synthesis of these series of compounds for economic accessibility and more selectivity.

## ORCID

*Nader Noroozi Pesyan* https://orcid.org/0000-0002-4257-434X

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## SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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