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One Pot Synthesis of Imines from Aromatic Nitro Compounds with a Novel Ni/SiO_2 Magnetic Catalyst

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Abstract We have successfully prepared a novel Ni/SiO_2 magnetic catalyst, and put it into the process of one pot synthesis of imines directly from aromatic nitro compounds and aldehydes for the first time. The catalyst has been characterized by XRD, IR, TEM, SEM, TPR, H_2-TPD and XPS. The prominent merits of the Ni/SiO_2 magnetic catalyst passivated with a gas mixture are that it can be stored safely in air below 423 K and needs no activation before use. In the catalytic test, most conversion and selectivity is almost up to 100%, and it is found that the catalyst is highly efficient, stable, and reusable for the synthesis of imines.

Keywords One pot synthesis · Imines ·
Novel Ni/SiO_2 magnetic catalyst ·
Nitroaromatic compounds · Surface passivated

1 Introduction

Imines and their derivatives are versatile intermediates in organic synthesis [1], in particular for the preparation of heterocycles [2], anti-inflammatory agents [3], non-natural aminoacids [4], liquid crystalline materials [5], and anti-cancer agents [6].

Since the first preparation of imines was reported by Schiff more than a century ago [7], a variety of methods/systems for the synthesis of imines have been described, such as $ZnCl_2$ [8], $TiCl_4$ [9], $MgSO_4$–PPTS [10], alumina [11], $Ti(OR)_4$ [12], $Er(OTf)_3$ [13], $MgSO_4$ [14], $MgClO_4$ [15], P_2O_5/Al_2O_3 [16], $CuSO_4$ [17], and $NaHCO_3$ [18], in which metal species act as Lewis acids to activate the carbonyl group as well as facilitating the removal of water. In the past few years, with the development of experimental techniques, some innovations were reported, including solid-state synthesis [19], solvent-free/clay/microwave irradiation [20], water suspension medium [21], solvent/reflux [22], infrared irradiation/no solvent [23], K-10/microwave [24], silica/ultrasound irradiation [25], $NaHSO_4 \cdot SiO_2$/microwave/solvent-free [26], dirhodium Caprolactamate [27], [bmim]BF_4/molecular sieves [28] and et al. The methods/systems aforementioned showed some disadvantages such as the requirements of high reaction temperatures, prolonged reaction periods, moisture sensitive catalysts, large quantities of aromatic solvents, costly dehydrating reagents/catalysts and special instruments [25].

As an alternative, one pot system for imine synthesis has been under investigations, such as $Rh_6(CO)_{16}$ [29], $PdCl_2(PPh_3)_2/SnCl_2$/CO [30], Pd/C/H_2 [31], and $Ru_3(CO)_{12}$/CO [32]. However, these protocols required transition-metal catalysts, elevated pressures, high temperatures, which made them far from ideal for laboratory-scale synthetic chemistry.

Recently, there have been some examples of heterogeneous bifunctional catalysts for one pot synthesis [33–35]. The discovery and utilization of a single catalyst to promote more than one transformation in a selective manner is a promising research area. Such direct synthetic routes avoid side product formation and loss of starting material as well as reducing capital investment and operation costs.

In the one pot synthesis of imines, a few new reports appeared in recent years. Sithambaram reported direct catalytic synthesis of imines from alcohols using manganese octahedral molecular sieves [35], Taylor described a tandem oxidation-imine formation process from alcohols

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with active manganese oxide [36, 37], Korich developed a facile, one pot procedure to afford diarylimines [38], Macho reported one stage preparation of Schiff's bases from nitroarenes, aldehydes and carbon monoxide in the presence of water [39], tandem nitroarene reduction and intramolecular Schiff base condensation to provide heteroarenes has also been reported [40, 41].

All the facts discussed above plus the increased interest in environmental protection issues inspired us to develop an efficient, mild and practical one pot synthesis of imines from aromatic nitro compounds with a novel Ni/SiO_2 magnetic catalyst. Based upon literature, Ni compounds are excellent catalysts for the reduction of nitroaromatics [42, 43] and SiO_2 is a commonly-utilized catalyst for imine formation [25]. It seemed promising that Ni/SiO_2 can be an effective bifunctional catalyst for one pot synthesis imines from nitroaromatics.

In this paper we reported an efficient tandem catalytic process to furnish imines directly from aromatic nitro compounds using Ni/SiO_2 as the catalyst. Ni/SiO_2 acts as a bifunctional catalyst in this process to reduce the nitroaromatics to amines and subsequently to form imines. These two distinct steps are catalyzed by Ni/SiO_2 in a single reaction vessel under the same conditions. The process does not require any additives for water removal. In addition, the novel Ni/SiO_2 magnetic catalyst passivated with a gas mixture does not require activation prior to use.

2 Experimental

2.1 The Preparation of Ni/SiO_2 Catalyst

The Ni/SiO_2 catalyst was prepared according to the literatures with slight modification [44–48]: Some commercial diatomite [obtained from Jilin Linjiang; BET = 60 m^2/g; treated with acid before usage] is added to an aqueous solution of $Ni(NO_3)_2 \cdot 6H_2O$ (1.0 mol/L) and stirred for 1 h at 323 K. 10 mL monodisperse silica sol is added into the above mixture and stirred for 1 h. Then, an aqueous solution of $(NH_4)_2CO_3$ (1.0 mol/L) is dropped. After stirring for 20 h, the resulting solid is filtered, washed, extruded forming, dried at 333 K for 24 h and crushed to powder. Finally, the sample is reduced in H_2 for 4 h at 773 K, and then passivated (forming a thin protective oxide covering) with a gas mixture (3% N_2 in air) until the catalyst is cooled to room temperature. The Ni loading is about 55 wt% and the BET is 180 m^2/g.

2.2 Characterization of the Catalyst

The content of nickel in Ni/SiO_2 was determined by atomic absorption spectroscopic analysis on a varian-AA240 spectrometer. Total surface area determination was performed in a Micromeritics ASAP 2010. Powder X-ray diffraction (XRD) for samples were measured with XRD-6000 diffractometer made in Japan, using Cu Kα radiation ($\lambda = 0.15418$ nm). Fourier transform infrared (FT-IR) spectra were recorded with a Nicolet NEXUS 670 FT-IR instrument with a DTGS detector. TEM measurements were taken on a Hitachi-600 electron microscope, and samples were suspended in ethanol and ultrasonically dispersed. Scanning electronmicroscopic (SEM) images of selected samples were taken on a JSM-6701F scanning electron microscope. The temperature programmed reductions (TPR) of samples were carried out on Micromeritics Chemisorb 2750 instrument. TPR were carried out from room temperature to 800 °C in 10% H_2 in Ar at a flow rate of 25 mL/min. Temperature programmed desorption of hydrogen (H_2-TPD) were performed on a Micromeritics Chemisorb 2750 instrument equipped with a thermal conductivity detector (TCD). The chemical states of the elements in the catalysts were determined using a PHI-5702 multifunctional X-ray photoelectron spectroscope (XPS). Magnetic properties of the samples were measured by a vibrating sample magnetometers. (TOEI, VSM-5S-15) with a maximal applied field of 16 kOe. The samples were measured under a magnetic field of 10 kOe at room temperature, and the amount of the sample for the measurement was 0.022 g.

2.3 General Procedure for the Catalytic Test

The catalytic reactions were carried out in a 100 mL stainless steel reactor equipped with a magnetic stirrer. In a typical reaction procedure, 1.0 mmol nitroarenes, 1.3 mmol aldehydes and 15 mL ethanol (solvent) were mixed with 15 mg Ni/SiO_2 catalyst. Biphenyl was used as internal standard. The reactor was flushed three times with 0.5 MPa H_2 and pressurized to the desired pressure. Then, it was heated to the desired temperature in an oil bath with stirring. After reaction, the reactor was cooled in an ice-water bath and then slowly depressurized. Finally, the catalyst was separated, and the reactants and products were analyzed by GC (P.E. AutoSystem XL) or GC–MS (Agilent 6,890N/5,973N).

3 Results and Discussion

3.1 Characterization

3.1.1 XRD and IR

Figure 1 illustrates the wide-angle powder XRD patterns (in the 2θ range of 10–90°) of the fresh Ni/SiO_2 (Fig. 1a)

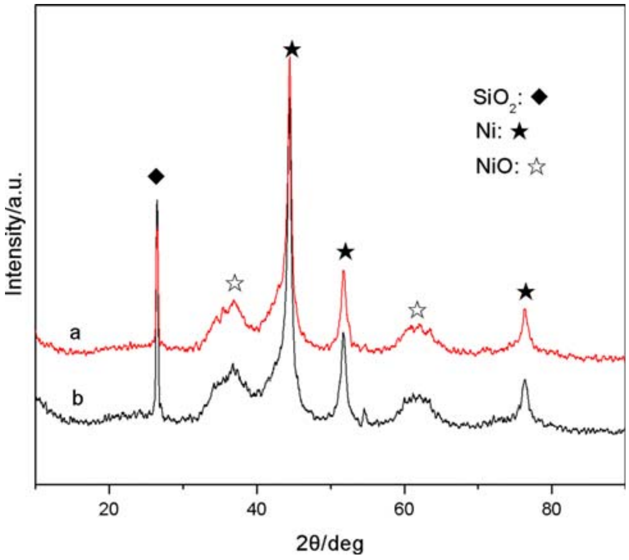


Fig. 1 XRD patterns of Ni/SiO_2 catalysts. **a** Fresh catalyst; **b** regenerated catalyst (after four cycles)

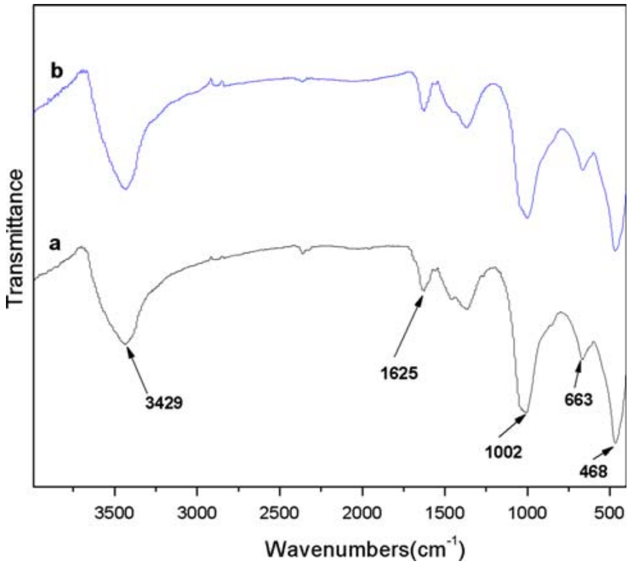


Fig. 2 FT-IR spectra of Ni/SiO_2 catalysts. **a** Fresh catalyst; **b** regenerated catalyst (after four cycles)

and the regenerated one (Fig. 1b). The fresh Ni/SiO_2 shows three diffraction peaks in the 2θ of 44.3, 51.7, and 76.3, which correspond to nickel metal. The peaks attributed to nickel oxide are observed at 36.7 and 61.2 in 2θ, and the peak of SiO_2 is found at 26.7 in 2θ. The result is consistent with the related reports in characterizations [49–51]. In addition, there exist no other peaks, indicating the very weak interaction between the Ni and carrier SiO_2. By comparing the XRD pattern of the regenerated Ni/SiO_2 with that of the fresh one, the catalyst structure does not change significantly after multiple catalytic recycles.

Figure 2 shows the IR spectra of the fresh and regenerated Ni/SiO_2 catalysts in the wave length range from 4,000 to 400 cm^{-1}. In the spectrum of the fresh Ni/SiO_2, the maximum absorptions at 1,002, 663 and 468 cm^{-1} are attributed to an anti-symmetric stretch, symmetric stretch and bending mode of Si–O–Si, respectively. The strong and wide absorption band at 3,429 cm^{-1} of the catalyst samples indicates that there are plenty of –OH groups on the surface of the Ni/SiO_2 catalyst. The presence of Si–OH shows that the sample does not have a completely condensed network. The band at 1,625 cm^{-1} is attributed to absorbed water, similar to the related reports [44, 52, 53]. By comparing the spectrum of the regenerated Ni/SiO_2 with that of the fresh one, there are not noticeable changes in Fig. 2a and b, which agrees with the above XRD analysis.

3.1.2 TEM and SEM

The representative electron micrographs (TEM) of the Ni/SiO_2 catalysts samples are presented in Fig. 3. The catalyst is mainly in amorphous phase, agglomerates partially, and there exists a large amount of interspersed particles, including the interspersed Ni and NiO particles. The diameter of the Ni/SiO_2 powder is about 40 nm. Moreover, after comparing the TEM of the regenerated Ni/SiO_2 with that of the fresh one, we find that no remarkable change occurs in the bulk structure of the catalyst due to the repeated uses. This result suggests that the catalyst is stable during the process.

Usually, SEM is used to determine the particle size and particle morphology of synthesized samples. The representative scanning electron microscopy (SEM) of the Ni/SiO_2 catalyst samples were presented in Fig. 4. Most of the catalyst particles are in good dispersibility. Based on SEM, the diameters of the main spherical particles are about 40 nm. No remarkable change of the catalyst occurs.

3.1.3 H_2-TPR and H_2-TPD

TPR is a useful technique for the characterization of interactions between metal and support. Figure 5 shows the TPR profiles of the nickel catalysts. For fresh Ni/SiO_2 catalyst, besides a reduction peak at 276 °C ascribed to the reduction of NiO particles, another broad reduction peak at higher temperature about 440 °C appeared, which could be attributed to the reduction of NiO nearly contacted with the diatomite support [54, 55]. For regenerated catalyst, the main reduce peak at 264 °C, which is somewhat lower than fresh catalyst. The TPR peak shift to lower temperature is due to the catalyst particle growth after being used.

H_2-TPD is one of effective methods to characterize active surface of a catalyst. Classical supported nickel

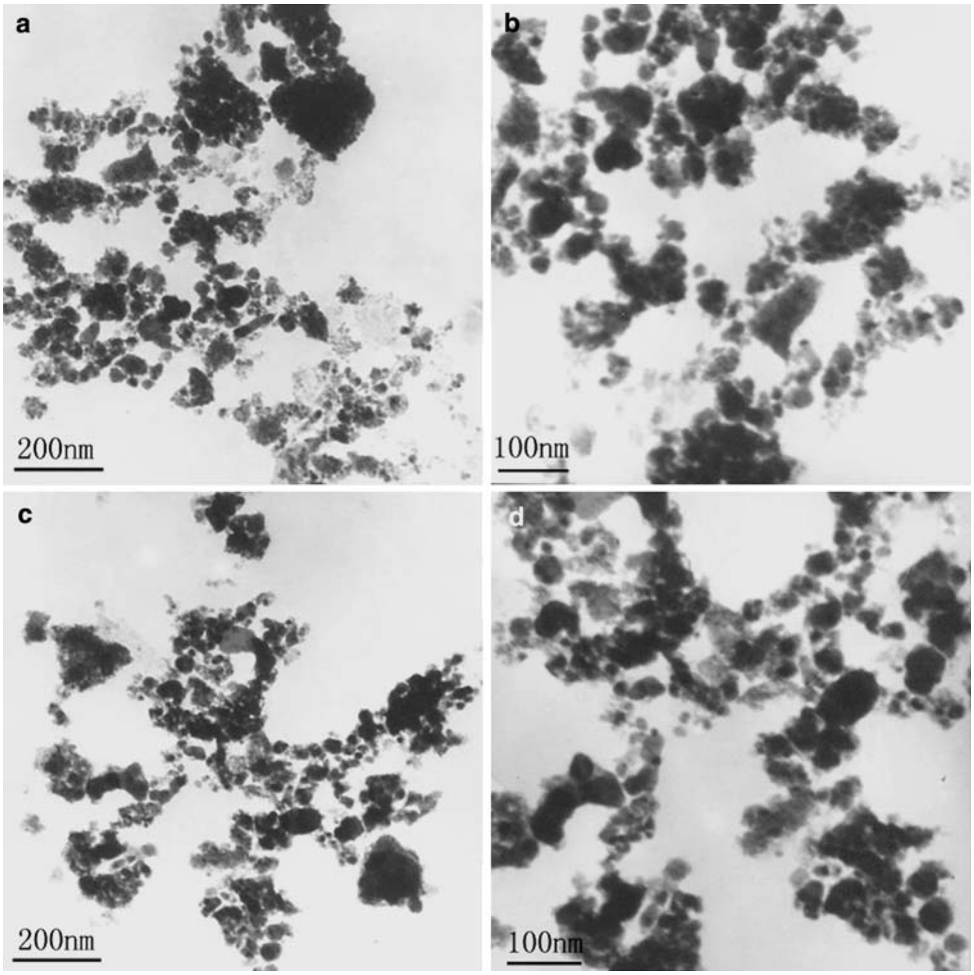


Fig. 3 TEM images of Ni/SiO_2 catalysts. **a, b** Fresh catalyst; **c, d** regenerated catalyst (after four cycles)

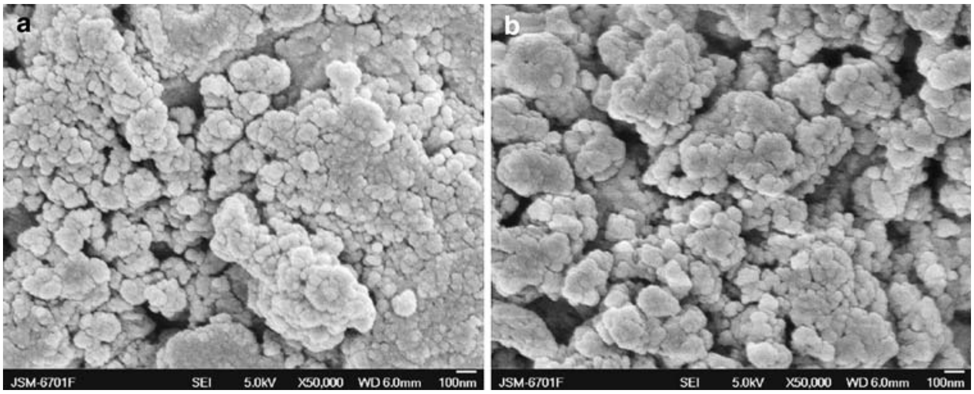


Fig. 4 SEM images of Ni/SiO_2 catalysts. **a** Fresh catalyst; **b** regenerated catalyst (after four cycles)

catalysts reduced by gaseous hydrogen give rise to H_2-TPD profiles comprising two or more temperature peaks as a result of the formation of several active sites [56]. These catalysts, like other transition metal catalysts, are also good H_2 reservoirs, capable of adsorbing and storing large amounts of H_2 [56–58].

H_2-TPD profiles (Fig. 6) showed that hydrogen desorption comprised two domains of temperature denoted type I (around 106 °C) and type II (around 409 °C). The first one was ascribed to hydrogen linked to nickel active sites. The second one was attributed to hydrogen much more bonded to the catalyst surface, probably that of the support or at the nickel-support interface boundary. The broad width of TPD peaks suggested the presence of metal particles [59, 60]. The similar results are found for the regenerated catalyst.

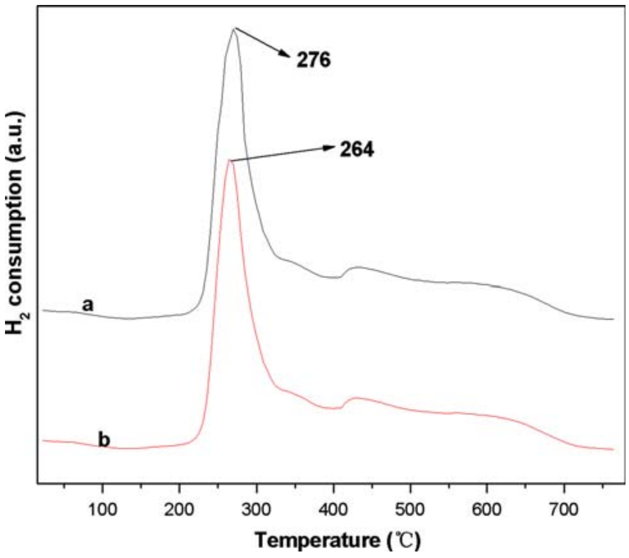


Fig. 5 H_2-TPR profiles of Ni/SiO_2 catalysts. **a** Fresh catalyst; **b** regenerated catalyst (after four cycles)

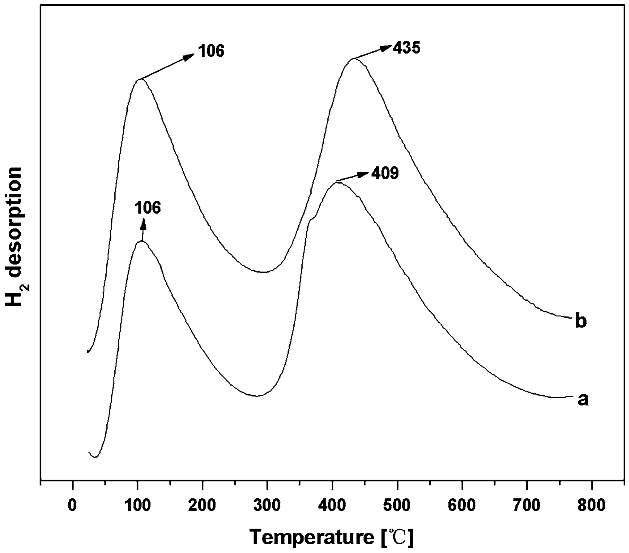


Fig. 6 H_2-TPD profiles of Ni/SiO_2 catalysts. **a** Fresh catalyst; **b** regenerated catalyst (after four cycles)

3.1.4 *XPS*

X-ray photoelectron spectroscopy is a convenient method of studying supported nickel catalysts. The XPS spectra of the Ni/SiO_2 samples are presented in Fig. 7. The dispersion of the supported phase may be judged by the intensity of the Ni 2p3/2 peak, while the peak position is informative with respect to the oxidation state of nickel. The spectrum of Fig. 7a exhibits a Ni 2p3/2 band at 856.3 ev, which is typical of NiO [61, 62]. In addition, the peak at about 852.9 eV, that is characteristic of metallic nickel [61, 62]. In Fig. 7b, the peak about 852.9 eV is lower than that of Fig. 7a, these results show part of the metallic nickel has been oxidized after the continuous using of catalyst [63].

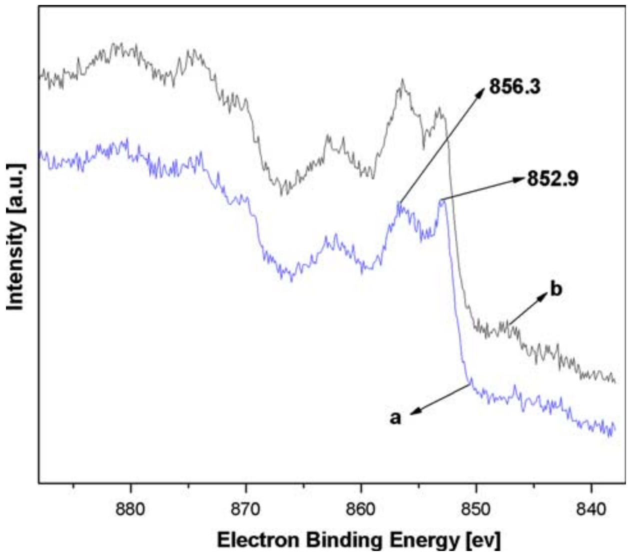


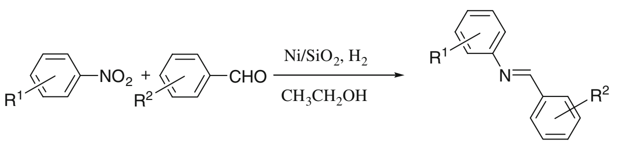
Fig. 7 XPS spectra of Ni/SiO_2 catalysts. **a** Fresh catalyst; **b** regenerated catalyst (after four cycles)

3.2 One Pot Synthesis Imines Over the Novel Ni/SiO_2 Magnetic Catalysts

The one pot synthesis of imines with the novel Ni/SiO_2 magnetic catalysts directly from nitroaromatic compounds and aldehydes (Scheme 1) may contain two major steps: reduction of nitro group to amine and subsequent condensation with aldehydes with water removal facilitated by the catalyst [20, 64]. In our experiments, only ethanol was chosen as solvent based on requirements of "green chemistry", since ethanol can easily be generated from renewable sources and available all over the world [25].

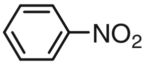
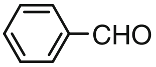
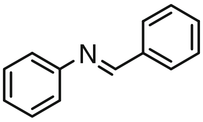
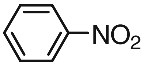
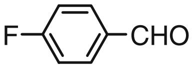
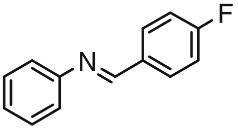
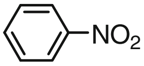
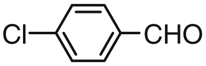
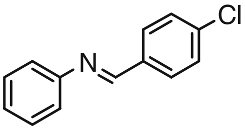
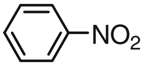
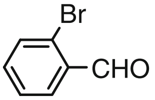
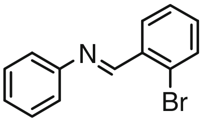
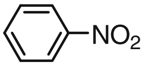
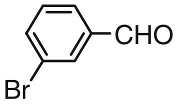
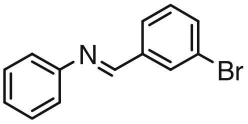
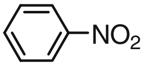
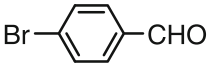
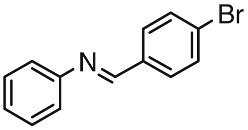
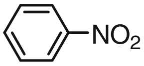
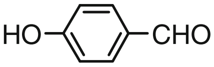
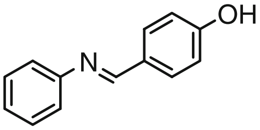
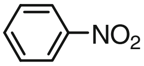
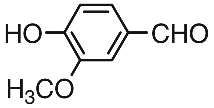
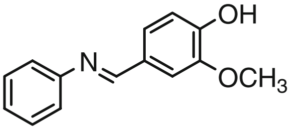
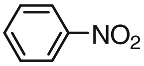
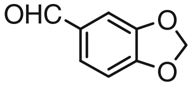
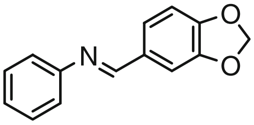
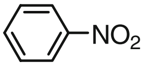
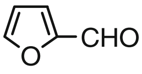
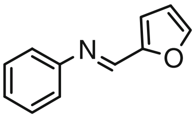
A wide range of aromatic aldehydes and aromatic nitro-containing compounds were tested with the Ni/SiO_2 magnetic catalyst under the optimized conditions and almost all desired transformations were achieved in excellent conversion and with satisfactory selectivity (Tables 1, 3, entries 1–21). Further investigations (Tables 1, 3) revealed more detailed information for this reaction.

In Table 1, imine formation from nitrobenzene and a variety of aldehydes has been investigated. A series of



Scheme 1 One pot synthesis of imines over the novel Ni/SiO_2 magnetic catalysts directly from aromatic nitro compounds and aldehydes

Table 1 Imines formation from nitrobenzene and different aldehydes

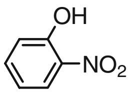
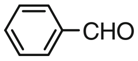
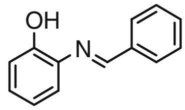
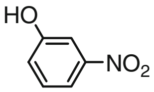
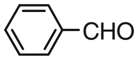
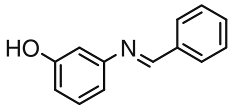
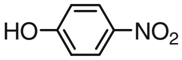
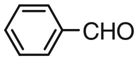
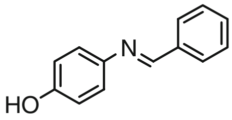
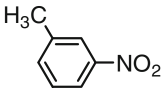
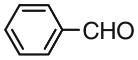
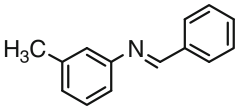
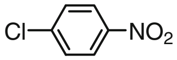
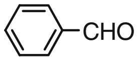
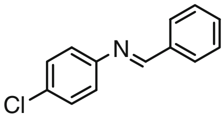
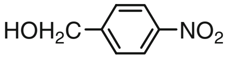
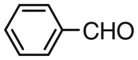
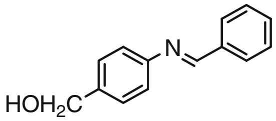
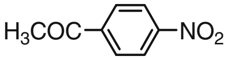
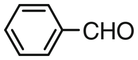
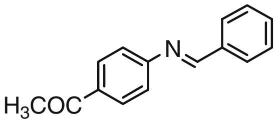
Entry	$PhNO_2$	Ar^2CHO	Product	Conversion (%)[a]	Selectivity (%)[b]
1				98.54 95.65^{c}	99.30^{c}
2				98.68	98.93
3				100	96.10
4				100	96.42
5				100	97.94
6				100	100
7				100	66.25
8				98.09	96.09
9				100	98.30
10				92.84	99.24

Reaction condition: 378 K, 1 mmol $PhNO_2$, 1.3 mmol Ar^2CHO, 1.4 MPa, 15 mg Ni/SiO_2 as catalyst, 15 mL ethanol as solvent, 8 h

[a] Conversion of nitrobenzene

[b] Selectivity to imine

Table 2 Imines formation from benzaldehyde and different nitroarenes

Entry	Ar^1NO_2	PhCHO	Product	Conversion (%)[a]	Selectivity (%)[b]
11				100	79.66
12				100	93.32
13				100	100
14				99.28	99.20
15				100	98.81
16				100	100
17				100	63.33

Reaction condition: 378 K, 1 mmol Ar^1NO_2, 1.3 mmol PhCHO, 1.4 MPa, 15 mg Ni/SiO_2 as catalyst, 15 mL ethanol as solvent, 8 h

[a] Conversion of Ar^1NO_2

[b] Selectivity to imine

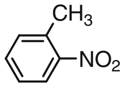
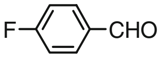
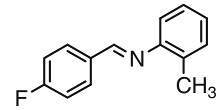
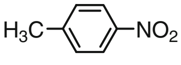
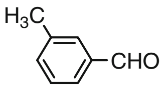
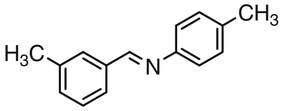
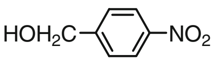
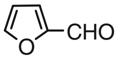
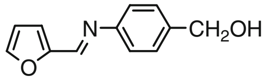
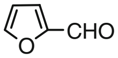
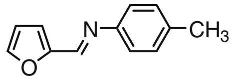
aromatic aldehydes were utilized and all imines were obtained in excellent conversion and selectivity (Table 1, entries 1–10) except entry 7, which demonstrated it is a mild method that tolerates both electron-withdrawing and electron-donating substituents. In addition, the selectivity of the product formation (entries 4–6) largely depends on the steric effect.

In Table 2, imine formation from benzaldehyde and different nitroarenes has been studied (entries 11–17). Generally the selectivities are promising, except for entry 17, the selectivity drops to 63.33%; in part due to the harsh experiment conditions leading to over reduction of the ketone, which is in agreement with our previous report about nitro hydrogenation [65]. From the results of these substrates (entries 11–13), the steric effect affection was also found.

Table 3 showed imines formation from different nitroarenes and different aldehydes (entries 18–21). The electron demand of the substituents has no effect on reaction efficiency; all results are excellent although the entries (nitroarenes and aldehydes) have many substituents, such as –CH_2OH, –CH_3 and –F.

(Table 4) when Pd/C was used (number 6–9), no desired imine was obtained (only benzyl alcohol, aniline and

Table 3 Imines formation from different nitroarenes and different aldehydes

Entry	Ar^1NO_2	Ar^2CHO	Product	Conversion (%)[a]	Selectivity (%)[b]
18				100	96.99
19				96.61	94.87
20				100	78.99
21				100	88.72

Reaction condition: 378 K, 1 mmol Ar^1NO_2, 1.3 mmol aldehydes, 1.4 MPa, 15 mg Ni/SiO_2 as catalyst, 15 mL ethanol as solvent, 8 h

[a] Conversion of Ar^1NO_2

[b] Selectivity to imine

Table 4 Different catalyst systems for one pot synthesis

Number	Catalyst	By-catalyst	Condition	Conversion (%)[a]	Selectivity (%)[b]
1	Ni/SiO_2 (15 mg)		105 °C, 1.4 MPa, 8 h	98.54	99.47
2	Ru/C (15 mg)		25 °C, 1 atm, 8 h	0	0
3	Ru/C (15 mg)		50 °C, 1.4 MPa, 8 h	0	0
4	Ru/C (15 mg)		105 °C, 1.4 MPa, 8 h	24.75	78.45
5	Ru/C (15 mg)	SiO_2 (9 mg)	105 °C, 1.4 MPa, 8 h	32.94	78.01
6	Pd/C (15 mg)	SiO_2 (9 mg)	105 °C, 1.4 MPa, 8 h	100	0
7	Pd/C (15 mg)		105 °C, 1.4 MPa, 8 h	100	0
8	Pd/C (15 mg)	SiO_2 (9 mg)	25 °C, 1.4 MPa, 8 h	100	0
9	Pd/C (15 mg)		25 °C, 1 atm, 6 h	100	0.22

Reaction condition: 1 mmol nitrobenzene, 1.3 mmol benzaldehyde, 15 mL ethanol

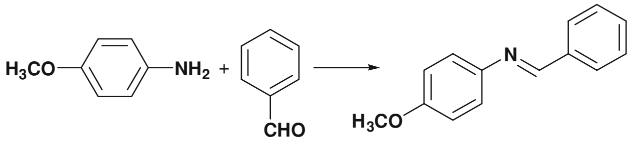
[a] Conversion of nitrobenzene

[b] Selectivity to imine

N-benzylaniline were observed), in part due to the strong reactivity of Pd/C, which is basically consistent with the previous reports [38]. In the condition (number 4–5), SiO_2 is a good promoter, which plays an important role in imine formation (number 4–5) [25]. All the results in Table 4 show that Ni/SiO_2 is the best catalyst for one pot synthesis of imine.

In order to determine the role of Ni/SiO_2 in the formation of the imine in the last step, we screened a variety of promoters to the imines synthesis (Table 5) using benzaldehyde and p-methoxyaniline in ethanol as a standard reaction. As shown in Table 5, the best result was obtained with Ni/SiO_2 (entry 2) in the yield of 91.05%. When no promoter (entry 1) was added, the reaction yield decreased to 86.09%. The silica (entry 3) was also employed as a promoter, and the yield is 88.90%, similar trend in results

Table 5 Promoters to imine synthesis



Entry	Catalyst	Yield[a] (GC) (%)
1	Absence	86.09
2	Ni/SiO_2	91.05
3	SiO_2	88.90

Reaction condition: 1 mmol benzaldehyde, 1 mmol p-methoxyaniline, 15 mg catalyst, 10 mL ethanol, reflux 1 h

[a] Yield to p-methoxyaniline

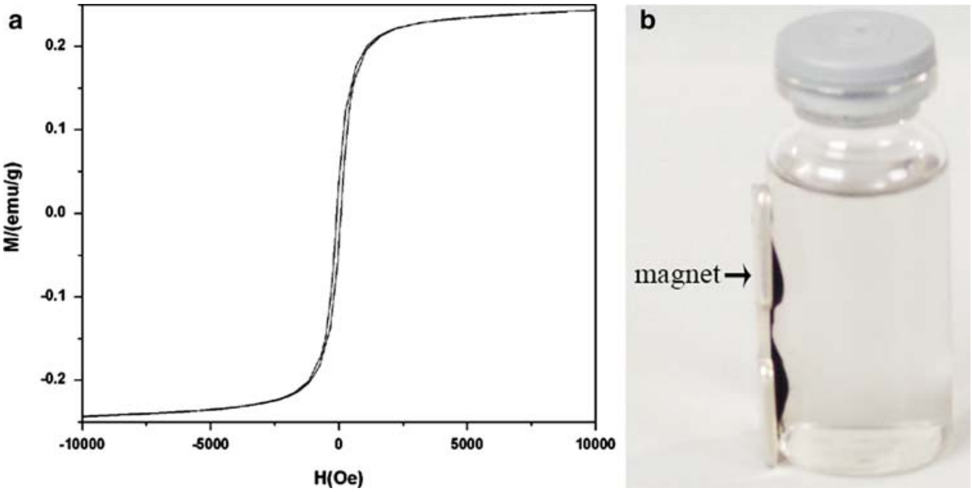


Fig. 8 Magnetic hysteresis cycle for Ni/SiO$_2$ catalyst (**a**); separation of Ni/SiO$_2$ catalyst by a magnet (**b**)

was observed in the related reports [25]. All the results in Table 5 show that Ni/SiO$_2$ plays as an important promoter in the imine synthesis.

3.3 Catalyst Regeneration and Stability

The magnetic properties of the Ni/SiO$_2$ catalyst were measured in fields between ±10 kOe at room temperature. Figure 8a showed hysteresis curves collected at 300 K. Ni/SiO$_2$ catalyst showed super paramagnetic behavior and no remanence at room temperature. These data indicated Ni/SiO$_2$ can be used as an easily recovered catalyst. From Fig. 8b, the Ni/SiO$_2$ catalyst shows obvious magnetism. It can easily be separated from reaction mixture in a relatively low magnetic field with a small laboratory magnet, recovered quantitatively by simple filtration, and regenerated by washing with ethanol. Moreover, after magnetic recovery, the catalyst can be dispersed ready for further use. The catalyst recycling experiment demonstrates that the conversion and selectivity are almost unchanged (Table 1, entry 1^{c}) after four turnovers. The Ni loading of the regenerated catalyst (after four cycles) is about 53.8 wt% (slight lower than the fresh catalyst), no leaching of the catalyst is occurring under our reaction condition.

4 Conclusion

In summary, we firstly reported an efficient, mild and practical one pot method for the synthesis of imines with the novel Ni/SiO$_2$ magnetic catalysts. This passivated Ni/SiO$_2$ catalyst showed excellent conversion and selectivity for the synthesis of imines without any extra additives or promoters. It is anticipated that this simple, inexpensive catalytic system could be employed for preparing different substituted imines on a large scale.

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