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SILICA NANOPARTICLES AS A HIGHLY EFFICIENT CATALYST FOR THE ONE-POT SYNTHESIS OF STERICALLY CONGESTED 2-(DIBENZYLAMINO)-2-ARYL ACETAMIDE DERIVATIVES FROM BY PHTHALDEHYDE ISOMERS, ISOCYANIDES AND DIBENZYLAMINE

Ali Ramazani[1*], Zahra Shaghaghi[2], Hamideh Aghahosseini[1], Pegah Azimzadeh Asiabi[1] and Sang Woo Joo[3*]

[1]Department of Chemistry, University of Zanjan, P. O. Box 45195-313, Zanjan, Iran
[2]Department of Chemistry, Payame Noor University, P. O. Box 19395-3697, Tehran, Iran
[3]School of Mechanical Engineering, Yeungnam University, Gyeongsan 712-749, Republic of Korea

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ABSTRACT. A green and efficient method for the preparation of 2-(dibenzylamino)-2-aryl acetamide derivatives *via* a three-component reaction of an isocyanide, dibenzylamine and a phthalaldehyde derivative in the presence of silica nanoparticles (silica NPs, *ca.* 42 nm) as a catalyst under solvent free conditions at room temperature is described. The ease of work-up, green chemistry conditions and high yields of the products make this procedure a useful addition to modern synthetic methods. The silica nanoparticles that used in this reaction as a catalyst were prepared by thermal decomposition of rice hulls. Simple, green and cheap method for the preparation of the nanocatalyst represents a major advantage for this process. The structures of these compounds were confirmed by IR, ^{1}H and ^{13}C NMR spectroscopy.

KEY WORDS: Silica nanoparticles, Nanocatalyst, Isocyanide, Phthalaldehyde

INTRODUCTION

Recently, multicomponent condensation reactions have become one of the most powerful methods for the synthesis of small-molecule libraries, due to the fact that products are formed in a single step by simultaneous reactions of several reagents, and the molecular diversity required for such combinatorial libraries can be achieved by simply varying each component [1–6].

Recently, silica nanoparticles (SNPs) have attracted tremendous attention in catalysis because of their improved efficiency under mild and environmentally benign conditions in the context of ecological (green) preparation [7, 8]. Due to their enormously large and highly reactive surface area, silica nanoparticles (SNPs) have the potential to exhibit superior catalytic activity in comparison to bulk counterparts [9, 10].

The acetamides are useful building blocks for the preparation of biologically active natural products, especially depsipeptide compounds. They have been identified as inhibitors of methionine aminopeptidase-2 and HIV protease, potent antitumor activity, and play an important role in medicinal chemistry. Due to the significance of this core, the convenient formation of this moiety has attracted much attention in recent years. Classically, acetamides have been synthesized by condensation of lactic acid with an amine and the most direct method is the Passerini-type reaction [11].

As part of our ongoing program to develop efficient and robust methods for the preparation of organic compounds [11-48], in this paper, we wish to report an efficient green protocol for the preparation of 2-(dibenzylamino)-2-aryl acetamide derivatives **6** through reaction of an alkyl isocyanide **4**, dibenzylamine **1** and a phthaldehyde isomer **2** in the presence of silica NPs at ambient temperature.

*Corresponding author. E-mail: aliramazani@gmail.com, swjoo@yu.ac.kr

EXPERIMENTAL

Materials and solvents were obtained from Merck (Germany) and Fluka (Switzerland) and were used without further purification. Melting points were measured with an Electrothermal 9100 apparatus and are uncorrected. IR spectra were measured on a Jasco 6300 FTIR spectrometer. ^{1}H and ^{13}C NMR spectra were measured with a Bruker DRX-250 Avance spectrometer at 250.0 and 62.5 MHz, respectively. A scanning electron microscope (Philips XL-30, Amsterdam, The Netherlands) with an acceleration voltage of 17 kV was used to investigate the size of the NPs. X-Ray diffraction data of all of the NPs were recorded on a Philips X'Pert diffractometer at a scan rate of 0.5° min^{-1} with 0.02° step size in the 2θ range 20–70°.

Preparation of silica nanoparticles

Dry rice hulls from Tarom region in the Zanjan province of Iran were treated with 20 vol% hydrochloric acid solution for 24 h, followed by a 20 vol% sulfuric acid solution for 24 h. Both treatments were done by boiling the solution under atmospheric pressure, followed by washing with distilled water. The washed hulls were then heated in a furnace at 700 ^{o}C for 2.0 h and silica nanoparticles (*ca.* 20% of the amount of starting dry rice hulls) were obtained [7].

*General procedure for the preparation of compounds **6a-f**, exemplified on **6a***

A mixture of dibenzylamine (0.19 mL, 1 mmol) and *m*-phthalaldehyde (0.134 g, 1 mmol) in dry CH_2Cl_2 (2 mL) was stirred at room temperature for 30 min. To this mixture, a solution of cyclohexyl isocyanide (0.13 mL, 1 mmol) was added rapidly and the mixture was stirred for 5 min. Silica NPs (0.5 g) was quickly poured to the reaction mixture and was stirred for 5 min at room temperature. The solvent was removed under reduced pressure and the residue was allowed to remain (without stirring) for 12 h in solvent-free conditions at room temperature. Then residue was placed over a flash column of silica gel powder (5 g). Flash column chromatography was washed using petroleum ether-diethyl ether (10:1) as eluent. The solvent was removed under reduced pressure to afford pure compound **6a** as white crystals.

*N-Cyclohexyl-2-(dibenzylamino)-2-(3-formylphenyl) acetamide (**6a**)*. White crystals, yield (90%). m.p.: 65 ^{o}C. IR (KBr) (ν_{max}, cm^{-1}): 3295 (NH), 2935, 2859, 1644, 1553, 699. ^{1}H NMR (250 MHz, $CDCl_3$) δ_H: 1.34-1.97 (10H, m, Cy), 3.31 and 3.86 (4H, 2d, J_{AB} = 12.50 Hz, $2CH_2$ of benzyl group), 3.89-3.98 (1H, m, CH-N of Cy), 4.47 (1H, s, CH), 7.26 (1H, s, NH, exchanges by D_2O addition), 7.28-7.36 (10H, m, $2C_6H_5$), 7.56 (1H, d, C_6H_4), 7.58 (1H, s, C_6H_4), 7.85 (1H, t, C_6H_4), 10.03 (1H, s, CHO). ^{13}C NMR (62.5 MHz, $CDCl_3$) δ_c: 24.65 and 24.72 (2 $CH_{2,\beta}$ of Cy), 25.50 (1 $CH_{2,\gamma}$ of Cy), 32.99 and 33.34 (2 $CH_{2,\alpha}$ of Cy), 48.02 (CHNH of Cy), 54.73 (2 CH_2 of benzyl groups), 67.20 (CH), 127.57, 128.61, 128.72, 131.59 (4C of C_6H_4), 128.85, 129.14, 129.37 (10C of $2C_6H_5$), 130.30 ($C_{ipso(C=C-CH)}$ of C_6H_4), 136.31 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 136.35 ($C_{ipso(C=C-CHO)}$ of C_6H_4), 169.45 (CONH), 192.14 (CHO).

*N-Cyclohexyl-2-(dibenzylamino)-2-(4-formylphenyl) acetamide (**6b**)*. Yellow crystals, yield (85%). m.p.: 132-136 ^{o}C. IR (KBr) (ν_{max}, cm^{-1}): 3288 (NH), 2930, 2853, 2364, 1712, 1644, 1209, 700. ^{1}H NMR (250 MHz, $CDCl_3$) δ_H: 1.19-1.42 (10H, m, Cy), 3.31 and 3.85 (4H, 2d, J_{AB} = 12.50 Hz, $2CH_2$ of benzyl group), 3.83-3.87 (1H, m, CH-N of Cy), 4.43 (1H, s, CH), 7.13 (1H, s, NH, exchanges by D_2O addition), 7.25-7.39 (10H, m, $2C_6H_5$), 7.48 (2H, d, C_6H_4), 7.91 (2H, d, C_6H_4), 10.04 (1H, s, CHO). ^{13}C NMR (62.5 MHz, $CDCl_3$) δ_c: 24.67 and 29.73 ($2CH_{2,\beta}$ of Cy), 25.52 ($1CH_{2,\gamma}$ of Cy), 33.03 and 33.38 ($2CH_{2,\alpha}$ of Cy), 48.01 (CHNH of Cy), 54.66 ($2CH_2$ of benzyl groups), 67.29 (CH), 128.60, 127.64, 128.58 (10CH of $2C_6H_5$), 127.56, 128.72 (4C of

C_6H_4), 130.91 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 129.45($C_{ipso(C=C)}$ of C_6H_4), 138.25 ($C_{ipso(CHO)}$ of C_6H_4), 168.90 (CONH), 191.96 (CHO).

*N-tert-butyl-2-(dibenzylamino)-2-(3-formylphenyl)acetamide (**6c**)*. Orange viscous oil, yield (80%). IR (neat) (ν_{max}, cm^{-1}): 3384 (NH), 2933, 1679, 1509, 1454, 1365, 700. ^{1}H NMR ($CDCl_3$) δ_H: 1.42 (9H, s, $C(CH_3)_3$), 3.34 and 3.85 (4H, 2d, J_{AB} = 11.99 Hz, $2CH_2$ of benzyl group), 4.39 (1H, s, CH), 7.26 (1H, s, NH, exchanges by D_2O addition), 7.26-7.42 (10H, m, $2C_6H_5$), 7.42-7.44 (2H, m, C_6H_4), 7.78 (1H, s, C_6H_4), 7.85 (1H, m, C_6H_4), 10.03 (1H, s, CHO). ^{13}C NMR (62.5 MHz, $CDCl_3$) δ_c: 28.83 (3C of $C(CH_3)_3$), 51.19 (C of $C(CH_3)_3NH$), 54.77 ($2CH_2$ of benzyl groups), 67.70 (CH), 127.54, 128.57, 128.72, 129.56 (4C of C_6H_4), 128.80, 128.83, 129.07 (10CH of $2C_6H_5$), 131.68 ($C_{ipso(C=C-CH)}$ of C_6H_4), 135.52 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 136.48 ($C_{ipso(C=C-CHO)}$ of C_6H_4), 169.91 (CONH), 192.17 (CHO).

*N-tert-butyl-2-(dibenzylamino)-2-(4-formylphenyl)acetamide (**6d**)*. Yellow viscous oil, yield (75%). IR (neat) (ν_{max}, cm^{-1}): 3375 (NH), 2930, 1683, 1510, 1455, 1365, 1213, 749, 700. ^{1}H NMR (250 MHz, $CDCl_3$) δ_H: 1.41 (9H, s, $C(CH_3)_3$), 3.33 and 3.9 (4H, 2d, J_{AB} = 11.99 Hz, $2CH_2$ of benzyl group), 4.39(1H, s, CH), 7.14 (1H, s, NH, exchanges by D_2O addition), 7.29-7.39 (10H, m, $2C_6H_5$), 7.49 (2H, d, C_6H_4), 7.89 (2H, d, C_6H_4), 10.03(1H, s, CHO). ^{13}C NMR (62.5 MHz, $CDCl_3$) δ_c: 28.83 (3C of $C(CH_3)_3$), 49.08 (C of $C(CH_3)_3NH$), 52.99 ($2CH_2$ of benzyl groups), 67.85 (CH), 127.66, 129.45 (4CH of C_6H_4),128.63, 128.79, 129.10 (10C of $2C_6H_5$), 131.06 ($C_{ipso(C=C-CH)}$ of C_6H_4), 130.39 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 135.83 ($C_{ipso(C=C-CHO)}$ of C_6H_4), 169.91 (CONH), 192.17 (CHO).

*N-(1,1,3,3-Tetramethylbutyl)-2-(dibenzylamino)-2-(3-formylphenyl)acetamide (**6e**)*. Red viscous oil, yield (70%). IR (neat) (ν_{max}, cm^{-1}): 3460 (NH), 2934, 1677, 1510, 1455, 1366, 700. ^{1}H NMR (250 MHz, $CDCl_3$) δ_H: 0.96 (9H, s, $C(CH_3)_3$), 1.46 (3H, s, $C(CH_3)_2NH$), 1.51 (3H, s, $C(CH_3)_2NH$), 1.65 (1H, d, $^2J_{HH}$ = 15 Hz, $CH_2C(CH_3)_3$), 1.96 (1H, d, $^2J_{HH}$ = 15 Hz, $CH_2C(CH_3)_3$), 3.18 and 3.87 (4H, 2d, J_{AB} = 14.99 Hz, 2CH2 of benzyl group), 3.83 (1H, s, CH), 7.27 (1H, s, NH, exchanges by D_2O addition), 7.32-7.36 (10H, m, $2C_6H_5$), 7.56-7.58 (2H, m, C_6H_4), 7.78 (1H, s, C_6H_4), 7.80 (1H, m, C_6H_4), 10.05(1H, s, CHO). ^{13}C NMR (62.5 MHz, $CDCl_3$) δ_c: 29.30 and 29.44 (2C of $C(CH_3)_2NH$, diastereotopic), 31.59 (3C of $C(CH_3)_3$), 31.45 (C of $C(CH_3)_3$), 51.39 (CH_2 of $CH_2C(CH_3)_3$), 54.70 ($2CH_2$ of benzyl groups), 55.16 (C of $C(CH_3)_2NH$), 67.75(CH), 127.48, 127.55, 128.14, 131.96 (4CH of C_6H_4), 127.59, 128.69, 128.79 (10CH of $2C_6H_5$), 129.06 ($C_{ipso(C=C-CH)}$ of C_6H_4), 136.75 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 138.12 ($C_{ipso(C=C-CHO)}$ of C_6H_4), 169.57 (CONH), 192.19 (CHO).

*N-(1,1,3,3-Tetramethylbutyl)-2-(dibenzylamino)-2-(4-formylphenyl)acetamide (**6f**)*. Orange viscous oil, yield (68%). IR (neat) (ν_{max}, cm^{-1}): 3457 (NH), 2930, 2864, 1678, 1510, 1455, 1366, 1214. ^{1}H NMR (250 MHz, $CDCl_3$): δ_H: 0.98 (9H, s, $C(CH_3)_3$), 1.96 (3H, s, $C(CH_3)_2NH$), 1.60 (3H, s, $C(CH_3)_2NH$), 1.65 (1H, d, J_{AB} = 15Hz, $CH_2C(CH_3)_3$), 1.93 (1H, d, $^2J_{HH}$ = 15 Hz, $CH_2C(CH_3)_3$), 3.19 and 3.84 (4H, 2d, $^2J_{HH}$ = 14.99 Hz, 2CH2 of benzyl group), 3.84 (1H, s, CH), 7.21 (1H, s, NH, exchanges by D_2O addition), 7.30-7.45 (10H, m, $2C_6H_5$), 7.50 (2H, d, C6H4), 7.91 (2H, d, C6H4), 10.04(1H, s, CHO). ^{13}C NMR (62.5 MHz, $CDCl_3$): δ_c: 29.25 and 29.41 (2CH3 of $C(CH_3)_2NH$, diastereotopic), 31.44 (3C of $C(CH_3)_2$), 31.54 (C of $C(CH_3)_3$), 51.39 (CH_2 of $CH_2C(CH_3)_3$), 54.69 ($2CH_2$ of benzyl groups), 67.80 (CH), 127.96, 128.74,128.68 (10CH of $2C_6H_5$), 127.61, 128.72(4C of C_6H_4), 131.06 ($C_{ipso(C=C-CH)}$ of C_6H_4), 135.73 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 138.08 ($C_{ipso(C=C-CHO)}$ of C_6H_4), 169.41 (CONH), 191.96 (CHO).

RESULTS AND DISCUSSION

Silica NPs were prepared by thermal decomposition of rice hulls based on previously reported method [31]. Figure 1 shows XRD pattern of the prepared silica NPs. X-Ray diffraction pattern of the prepared silica NPs, shows a broad peak around 2θ = 22° that confirm its amorphous structure [31]. The morphology and grain size of the silica NP was investigated by scanning electron microscopy (SEM) (Figure 2). The sample showed nearly smooth spherical and uniform nanoparticles for surface of the prepared silica NPs. The uniformity of the silica NPs may be resulted from highly molecular and cellular ordered of the natural rice hulls.

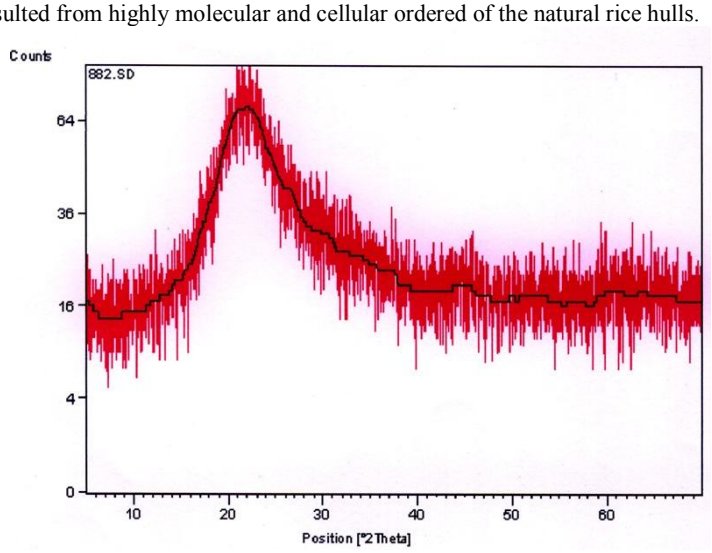


Figure 1. X-ray powder diffraction (XRD) pattern of the prepared silica nanoparticles shows a broad peak around 2θ = 22° that confirm its amorphous structure.

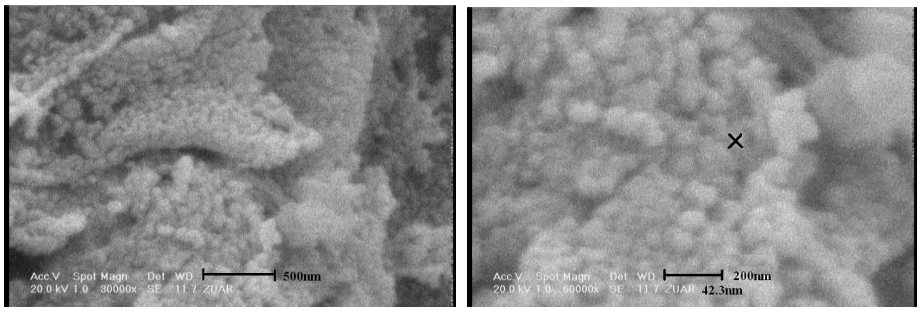
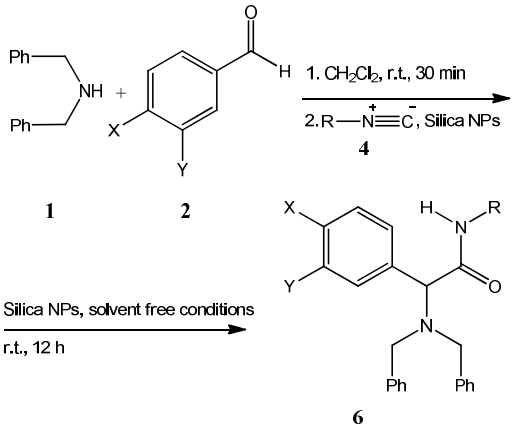


Figure 2. SEM image of the prepared silica nanoparticles.

In the present study, we sought to develop an efficient route for the synthesis of 2-(dibenzylamino)-2-aryl acetamide derivatives from a phthaldehyde isomer, an isocyanide and dibenzylamine. The reaction occurs smoothly in the presence of silica NPs at room temperature,

to produce 2-(dibenzylamino)-2-aryl acetamide derivatives **6** in fairly medium to good yields (Scheme 1 and Table 1).



Scheme 1. Preparation of 2-(dibenzylamino)-2-aryl acetamide derivatives **6** from dibenzylamine **1**, phthalaldehyde isomer **2** and isocyanide **4** in the presence of silica NPs.

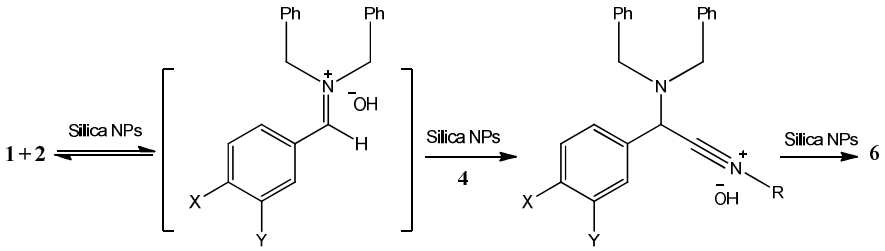
Table 1. Synthesis of 2-(dibenzylamino)-2-aryl acetamide derivatives **6** (Scheme 1).

6	R	X	Y	Yield (%)
a	cyclohexyl	H	CHO	90
b	cyclohexyl	CHO	H	85
c	*tert*-butyl	H	CHO	80
d	*tert*-butyl	CHO	H	75
e	1,1,3,3-tetarmethylbutyl	H	CHO	70
f	1,1,3,3-tetramethylbutyl	CHO	H	68

The silica gel powder has been used instead of silica NPs in this reaction, but increasing of reaction times and decreasing of 2-(dibenzylamino)-2-aryl acetamide derivatives yields were observed. The use of just 0.5 g of silica NPs for 1 mmol of reactants is sufficient to push the reaction forward. The larger surface area of the silica NPs in comparison with the silica gel powder could be plausible factor in the rate acceleration of the reaction in the presence of silica NPs. Silica NPs was quickly poured to the reaction mixture and was stirred for 5 min at room temperature. The solvent was removed under reduced pressure and the residue was allowed to remain (without stirring) for 12 h in solvent-free conditions at room temperature. Then residue was placed over a flash column of silica gel powder (5 g). Flash column chromatography was washed using petroleum ether-diethyl ether (10:1) as eluent. The solvent was removed under reduced pressure to afford pure products **6a-f**.

The structures of compounds **6a–f** were deduced from their IR, and ^{1}H- and ^{13}C-NMR spectra. The IR spectrum of **6a** showed strong absorption at 3295 cm^{-1} indicating the presence of amide group (NH), and sharp bands at 1644 and 1553 cm^{-1} were assigned to the amide carbonyl group and the aromatic rings, respectively. The ^{1}H-NMR spectrum of **6a** consisted of three multiplets for the cyclohexyl ring (δ = 1.34-1.97ppm and 3.89-3.98 ppm), two methylene (δ = 3.31 and 3.86, J_{AB} = 12.50 Hz, $2CH_2$ of two benzyls), a methyne (δ = 4.47 ppm), and an amide hydrogen atom exchangeable by D_2O (δ = 7.26 ppm). Presence of diastereotopic group ($PhCH_AH_B$) in the benzyl moieties resulted from existence of stereogenic center (aliphatic CH) in the molecule of **6a**. The phenyl and phenylene rings gave rise to characteristic signals in the

aromatic region. The ^{1}H-decoupled ^{13}C-NMR spectrum of **6a** showed 20 distinct resonances, partial assignment of these resonances is given in the experimental section. The ^{1}H- and ^{13}C-NMR spectra of compounds **6b–f** were similar to those of **6a**, except for the aromatic moieties, and the alkyl groups, which exhibited characteristic signals with appropriate chemical shifts. Although we have not established the mechanism of the reaction between the alkyl isocyanide **4**, dibenzylamine **1** and a phthaldehyde isomer **2** in the presence of silica NPs in an experimental manner, a plausible reaction sequence that accounts for the formation of the product **6** is shown in Scheme 2. Thus, condensation of a phthaldehyde isomer **2** and dibenzylamine **1** gave the iminium ion intermediate **3**, which reacted with the alkyl isocyanide **4** to afford the intermediate **5**. The ionic intermediate **5** was unstable and quickly converted to a compound **6** (Scheme 2).



Scheme 2. Proposed mechanism for the formation of 2-(dibenzylamino)-2-aryl acetamide derivatives **6**.

CONCLUSION

In this paper, an efficient route for the one-pot synthesis of 2-(dibenzylamino)-2-aryl acetamide derivatives **6** *via* a three-component reaction of an isocyanide, dibenzylamine and a phthalaldehyde derivative in the presence of silica nanoparticles (silica NPs, *ca.* 42 nm) as a catalyst under solvent free conditions at room temperature is developed. The ease of work-up, green chemistry conditions and fairly medium to good yields of the products makes this procedure a useful addition to modern synthetic methods [7, 17]. The silica nanoparticles that used in this reaction as a catalyst were prepared by thermal decomposition of rice hulls [31]. Simple, green and cheap method for the preparation of the nanocatalyst represents a major advantage for this process.

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