

Silica nanoparticles as a high efficient catalyst for the one-pot synthesis of 3-oxo-3-phenylpropanamid derivatives from isocyanides, phenylacetaldehyde and secondary amines

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Reaction of an isocyanide with an iminium ion intermediate, formed by reaction between 2-oxo-2-phenylacetaldehyde and a secondary amine in the presence of silica nanoparticles, proceeds smoothly at room temperature to afford ketoamide derivatives in high yields.

Key Words: Silica nanoparticles, multicomponent reaction, isocyanide, 2-oxo-2-phenylacetaldehyde, secondary amine, iminium ion

Introduction

Recently, multicomponent reactions (MCRs) have become important tools in modern preparative synthetic chemistry because these reactions increase the efficiency by combining several operational steps without any isolation of intermediates or changes of the conditions.¹⁻⁶ Among the multicomponent reactions known to date, the most valuable reactions are those based on isocyanides. Isocyanide-based multicomponent reactions (abbreviated to IMCRs by Ugi and Dmling) by virtue of their synthetic potential, their inherent atom efficiency, convergent nature, ease of implementation, and molecular diversity have attracted much attention because of the advantages that they offer in the field of combinatorial chemistry.^{7,8}

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In recent years, nanoparticles (NPs) have attracted tremendous attention in catalysis because of their improved efficiency under mild and environmentally benign conditions in the context of ecological (green) synthesis^{9,10} Due to their enormously large and highly reactive surface area, NPs have the potential to exhibit superior catalytic activity in comparison to bulk counterparts^{11,12} The methods for the synthesis of β -keto amides have been rarely reported in the literature.^{13–15} These protocols are multi-step in nature and usually perform under harsh reaction conditions.^{13–15} In this article, we wish to report a simple and practical procedure for the preparation of ketoamide derivatives through a one-pot multicomponent condensation reaction (MCR) in the presence of silica nanoparticles (silica NPs, ca. 42 nm).

Experimental

Starting materials and solvents were obtained from Merck (Germany) and Fluka (Switzerland) and were used without further purification. TLC and NMR spectroscopy were used to follow the reactions. Melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. IR spectra were measured on a Jasco 6300 FTIR spectrometer. ¹H and ¹³CNMR spectra were measured (CDCl₃ solution) with a BRUKER DRX-250 Avance spectrometer at 250.0 and 62.5 MHz, respectively. Elemental analyses were performed using a Heraeus CHN-O-Rapid analyzer. Preparative layer chromatography (PLC) plates were prepared from Merck silica gel (F₂₅₄) powder.

General procedure for the preparation of compounds 4

Silica nanoparticles (0.1 g) were added to a mixture of 2-oxo-2-phenylacetaldehyde (**2**) (1 mmol), secondary aminederivative **1** (1 mmol), and isocyanide **3** (1 mmol) at r.t., followed by 4 h of stirring. Residue was purified by preparative layer chromatography (PLC) (silica gel (F₂₅₄) powder; petroleum ether-ethyl acetate (4:1)). The characterization data of the compounds are given below.

N-(tert-butyl)-2-(dibenzylamino)-3-oxo-3-phenylpropanamide (**4a**)

Yellow viscose oi, (yield: 92%). IR (neat): $\nu = 3341$ (NH), 2923, 2861, 1712, 1653, 1530, 1438, 1223 cm⁻¹. ¹HNMR (250 MHz, CDCl₃) δ (ppm): 1.33 (s, 9H, 3CH₃), 3.86 and 4.44 (AB_q, 4H, 2CH₂ of benzyl, $J = 14.0$ Hz), 4.91 (s, 1H, CH), 6.84 (s, 1H, NH), 7.09-8.72 (m, 15H, CH_{arom}). ¹³CNMR (62.5 MHz, CDCl₃) δ (ppm): 28.67 (3CH₃), 51.40 (C), 55.13 (2CH₂), 66.46 (CH), 127.47, 128.49, 128.51, 128.59, 128.71, 133.26, 138.58 (15CH of arom, 128.93, 129.89, 137.58 (3C of aro), 167.85 (CO of amide), 198.78 (CO of ketone). Analysis of C₂₇H₃₀N₂O₂ (414.54). (% calculation/found): C: 78.23/78.28, H: 7.29/7.24, N: 6.76/6.72.

2-[benzyl(ethyl)amino]-N-(tert-butyl)-3-oxo-3-phenylpropanamide (**4b**)

Yellow viscose oi, (yield: 93%). IR (neat): $\nu = 3456$, 3069, 2927, 1718, 1638, 1449, 1269 cm⁻¹. ¹HNMR (250 MHz, CDCl₃) δ (ppm): 1.06 (t, 3H, CH₃), 1.31 (s, 9H, 3CH₃), 2.82 (q, 2H, CH₂), 3.86 and 3.99 (AB_q, 2H, CH₂ of benzyl, $J = 14.0$ Hz), 4.96 (s, 1H, CH), 6.96 (s, 1 H, NH), 7.13-8.91 (m, 10H, CH_{arom}). ¹³CNMR (62.5 MHz, CDCl₃) δ (ppm): 23.00 (CH₃), 28.57 (3CH₃), 45.50, 48.00 (2CH₂), 55.00 (C), 68.56 (CH), 127.47,

128.49, 128.51, 128.59, 128.71, 138.55 (10CH of arom, 129.89, 137.58 (2C of aro), 161.89 (CO of amide), 198.75 (CO of ketone). Analysis of C₂₂H₂₈N₂O₂ (352.22). (% calculation/found): C: 74.97/74.93, H: 8.01/8.06, N: 7.95/7.90.

2-[benzyl(methyl)amino]-N-(tert-butyl)-3-oxo-3-phenylpropanamide (4c)

Yellow viscose oi, (yield: 92%). IR (neat): $\nu = 3356, 3059, 2967, 1719, 1648, 1434, 1243 \text{ cm}^{-1}$. ¹HNMR (250 MHz, CDCl₃) δ (ppm): 1.33 (s, 9H, 3CH₃), 2.35 (s, 3H, CH₃), 3.74 (s, 2H, br, CH₂ of benzyl), 4.76 (s, 1H, CH), 7.00 (s, 1H, NH), 7.32-8.03 (m, 10H, CH_{arom}). ¹³CNMR (62.5 MHz, CDCl₃) δ (ppm): 28.53 (3CH₃), 39.37 (CH₃), 51.10 (C), 59.35 (CH₂), 73.46 (CH), 128.50, 128.64, 128.70, 128.87, 133.60 (10CH of arom), 128.19, 137.12 (2C of aro), 169.85 (CO of amide), 195.68 (CO of ketone). Analysis of C₂₁H₂₆N₂O₂ (338.20) (% calculation/found): C: 74.52/74.57, H: 7.74/7.69, N: 8.28/8.33

2-(dibenzylamino)- N-(2,6-dimethylphenyl)-3-oxo-3-phenylpropanamid (4d)

Yellow viscose oi, (yield: 95%). IR (neat): $\nu = 3332, 2989, 2927, 1712, 1656, 1460, 1233 \text{ cm}^{-1}$. ¹HNMR (250 MHz, CDCl₃) δ (ppm): 2.25 (s, 6H, 2CH₃), 3.86 and 4.15 (AB_q, 4H, 2CH₂ of benzyl, $J = 13.7 \text{ Hz}$), 5.20 (s, 1H, CH), 7.10-7.77 (m, 18H, CH_{arom}), 8.49 (s, 1H, NH). ¹³CNMR (62.5 MHz, CDCl₃) δ (ppm): 18.88 (2CH₃), 55.03 (2CH₂), 65.97(CH), 127.66, 128.31, 128.58, 128.65, 128.88, 129.02, 135.20, 137.99 (18CH of arom), 127.32, 133.57, 133.64, 136.46, 137.27 (6C of aro), 168.50 (CO of amide), 198.20 (CO of ketone). Analysis of C₃₁H₃₀N₂O₂ (462.58). (% calculation/found): C: 80.49/80.44, H: 6.54/6.50, N: 6.06/6.11

2-[benzyl(ethyl)amino]- N-(2,6-dimethylphenyl)-3-oxo-3-phenylpropanamide (4e)

Yellow viscose oi, (yield: 90%). IR (neat): $\nu = 3346, 3082, 2978, 1711, 1642, 1434, 1220 \text{ cm}^{-1}$. ¹HNMR (250 MHz, CDCl₃) δ (ppm): 1.25 (t, 3H, CH₃), 2.29 (s, 6H, 2CH₃), 2.77 (q, 2H, CH₂), 3.91 (s, br, 2H, CH₂ of benzyl), 4.27 (s, 1H, CH), 7.14(s, 1H, NH), 7.31-7.99 (m, 13H, CH_{arom}). ¹³CNMR (62.5 MHz, CDCl₃) δ (ppm): 21.08 (2CH₃), 24.23 (CH₃), 49.45, 56.13 (2CH₂), 63.57(CH), 127.66, 127.84, 128.52, 128.87, 129.02, 129.46, 133.27, 137.89 (13CH of arom, 127.02, 135.07, 137.06, 141.27 (4C of aro), 159.22 (CO of amide), 194.68 (CO of ketone). Analysis of C₂₆H₂₈N₂O₂ (400.22). (% calculation/found): C: 77.97/77.92, H: 7.05/7.10, N: 6.99/6.94.

2-[3a,6-dihydro-1H-benzo[de]isoquinolin-2(3H)-yl]-3-oxo-3-phenyl-N-(1,1,3,3-tetramethylbutyl) propanamide (4f)

Yellow viscose oi, (yield: 90%). IR (neat): $\nu = 3361, 2959, 2917, 1717, 1679, 1446, 1218 \text{ cm}^{-1}$. ¹HNMR (250 MHz, CDCl₃) δ (ppm): 0.96 (s, 9H, 3CH₃), 1.02 (s, 6H, 2CH₃), 1.41 (s, 2H, CH₂), 3.84 and 4.02 (AB_q, 4H, 2CH₂ of benzyl, $J = 14.3 \text{ Hz}$), 4.92 (s, 1H, CH), 6.92 (s, 1H, NH), 7.13-7.71 (m, 15H, CH_{arom}). ¹³CNMR (62.5 MHz, CDCl₃) δ (ppm): 28.67, 31.45 (5CH₃), 29.11, 31.57 (2C), 53.65, 55.13 (2CH₂), 65.50 (CH), 127.47, 128.37, 128.50, 128.56, 128.68, 128.79, 128.93, 130.10 (9CH of arom), 127.72, 133.35, 138.50(3C of aro), 167.52 (CO of amide), 199.28 (CO of ketone). Analysis of C₃₁H₃₈N₂O₂ (470.29). (% calculation/found): C: 79.11/79.16, H: 8.14/8.09, N: 5.95/5.90

2-[benzyl(methyl)amino]-N-cyclohexyl-3-oxo-3-phenylpropanamide (4g)

Yellow viscose oi, (yield: 92%). IR (neat): $\nu = 3416, 3069, 2925, 1723, 1642, 1434, 1243 \text{ cm}^{-1}$. $^1\text{HNMR}$ (250 MHz, CDCl_3) δ (ppm): 1.25-1.83 (m, 10H, C_{H_2} of cyclohexyl), 2.95 (s, 3H, CH_3), 3.73 (s, br, 2H, CH_2 of benzyl), 4.29 (m, 1H, CH of cyclohexyl), 4.35(s, 1H, CH), 7.04 (s, 1H, NH), 7.33-8.01 (m, 10H, CH_{arom}). $^{13}\text{CNMR}$ (62.5 MHz, CDCl_3) δ (ppm): 24.90, 25.00, 26.20 (5CH_2 of cyclohexyl), 33.15 (CH_3), 48.05 (CH of cyclohexyl), 57.12 (CH_2), 63.50(CH), 128.00, 128.57, 128.60, 128.64, 128.69, 129.15 (10CH of arom, 129.72, 131.75(2C of aro), 175.32 (CO of amide), 196.68 (CO of ketone). Analysis of $\text{C}_{23}\text{H}_{28}\text{N}_2\text{O}_2$ (364.22). (% calculation/found): C: 75.79/75.73, H: 7.74/7.79, N: 7.69/7.64.

2-[3a,6-dihydro-1H-benzo[de]isoquinolin-2(3H)-yl]-N-cyclohexyl-3-oxo-3-phenylpropanamide (4h)

Yellow viscose oi, (yield: 91%). IR (neat): $\nu = 3372, 3084, 2976, 1713, 1636, 1461, 1271 \text{ cm}^{-1}$. $^1\text{HNMR}$ (250 MHz, CDCl_3) δ (ppm): 0.83-1.63 (m, 10H, C_{H_2} of cyclohexyl), 3.84 and 4.07 (AB_q , 2H, CH_2 of benzyl, $J = 14.3 \text{ Hz}$), 4.50 (m, 1H, CH of cyclohexyl), 4.92(s, 1 H, CH), 6.92 (s, 1H, NH), 7.13-7.71 (m, 15H, CH_{arom}). $^{13}\text{CNMR}$ (62.5 MHz, CDCl_3) δ (ppm): 30.30, 31.02, 31.46 (5CH_2 of cyclohexyl), 52.18 (CH of cyclohexyl), 55.11 (2CH_2), 65.48(CH), 127.50, 128.19, 128.45, 128.61, 129.84, 138.50 (10CH of arom, 127.30, 129.25(3C of aro), 167.82 (CO of amide), 199.38 (CO of ketone). Analysis of $\text{C}_{29}\text{H}_{32}\text{N}_2\text{O}_2$ (440.25). (% calculation/found): C: 79.06/79.11, H: 7.32/7.38, N: 6.36/6.42.

2-[benzyl(methyl)amino]-3-oxo-3-phenyl-N-(1,1,3,3-tetramethylbutyl)propanamide (4i)

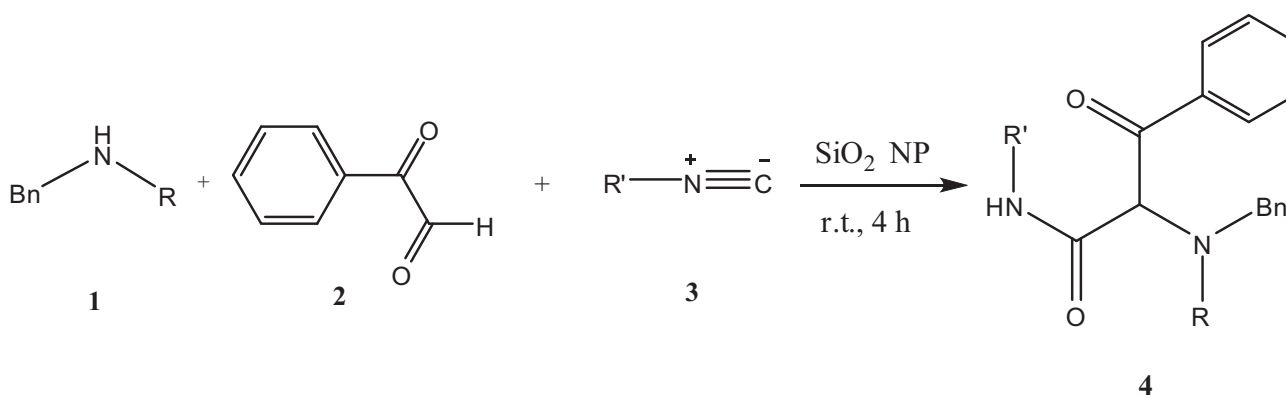
Yellow viscose oi, (yield: 89%). IR (neat): $\nu = 3365, 3058, 2968, 1718, 1668, 1457, 1263 \text{ cm}^{-1}$. $^1\text{HNMR}$ (250 MHz, CDCl_3) δ (ppm): 1.24 (s, 9H, 3CH_3), 1.58 (s, 6H, 2CH_3), 2.84 (s, 2H, CH_2), 3.06 (s, 3H, CH_3), 4.20 and 4.40 (AB_q , 2H, CH_2 of benzyl), 4.74 (s, 1H, CH), 6.92(s, 1H, NH), 7.31-7.98 (m, 15H, CH_{arom}). $^{13}\text{CNMR}$ (62.5 MHz, CDCl_3) δ (ppm): 26.47, 30.22 (5CH_3), 22.11, 31.46 (2C), 40.06 (CH_3), 53.35, 55.01 (2CH_2), 67.50(CH), 127.50, 128.19, 128.45, 128.61, 129.84, 138.50 (10CH of arom, 127.30, 129.25(2C of aro), 165.52 (CO of amide), 198.28 (CO of ketone). Analysis of $\text{C}_{25}\text{H}_{34}\text{N}_2\text{O}_2$ (394.26). (% calculation/found): C: 76.10/76.05, H: 8.69/8.74 N: 7.10/7.15

2-[benzyl(methyl)amino]-N-(2,6-dimethylphenyl)-3-oxo-3-phenylpropanamide (4j)

Yellow viscose oi, (yield: 88%). IR (neat): $\nu = 3374, 3092, 2977, 1719, 1638, 1444, 1225 \text{ cm}^{-1}$. $^1\text{HNMR}$ (250 MHz, CDCl_3) δ (ppm): 2.30 (s, 6H, 2CH_3), 2.63 (s, 3H, CH_3), 3.09 (s, br, 2H, CH_2 of benzyl), 4.92 (s, 1H, CH), 7.14 (s, 1H, NH), 7.30-7.99 (m, 10H, CH_{arom}) $^{13}\text{CNMR}$ (62.5 MHz, CDCl_3) δ (ppm): 20.06 (2CH_3), 39.11(CH_3), 54.23 (CH_2), 67.94 (CH), 124.67, 127.06, 128.21, 128.40, 128.65, 128.98, 129.00, 132.20 (13CH of arom), 127.35, 134.62, 136.43, 138.22 (4C of aro), 165.50 (CO of amide), 196.27 (CO of ketone). Analysis of $\text{C}_{25}\text{H}_{26}\text{N}_2\text{O}_2$ (386.20). (% calculation/found): C: 77.69/77.75, H: 6.78/6.83, N: 7.25/7.20.

Results and discussion

As part of our ongoing program to develop efficient and robust methods for the preparation of organic compounds,¹⁶⁻⁴⁴ we sought to develop an efficient route for the one-pot synthesis of sterically congested ketoamide derivatives **4** from simple and readily available alkyl isocyanides **3**, secondary amine **1**, and 2-oxo-2-phenylacetaldehyde (**2**) (Scheme 1 and Table 1). The reaction occurs smoothly in the presence of silica NPs at ambient temperature, to produce ketoamide derivatives **4** in excellent yields (Table 1). In the absence of silica NPs, the reactions were not conducted in the compounds **4** and in all cases several products were observed (based on TLC investigations).



Scheme 1. Three-component synthesis of ketoamide derivatives **4** in the presence of silica nanoparticles.

Silica NPs were prepared by thermal decomposition of rice hulls.⁴⁵ The results from X-ray diffraction (XRD) showed that the sample was silica NP as indicated by broadened peaks around $2\theta = 22^\circ$ (Figure 1). The morphology and grain size of the silica NP was investigated by scanning electron microscopy (SEM) (Figure 2).^{43,44}

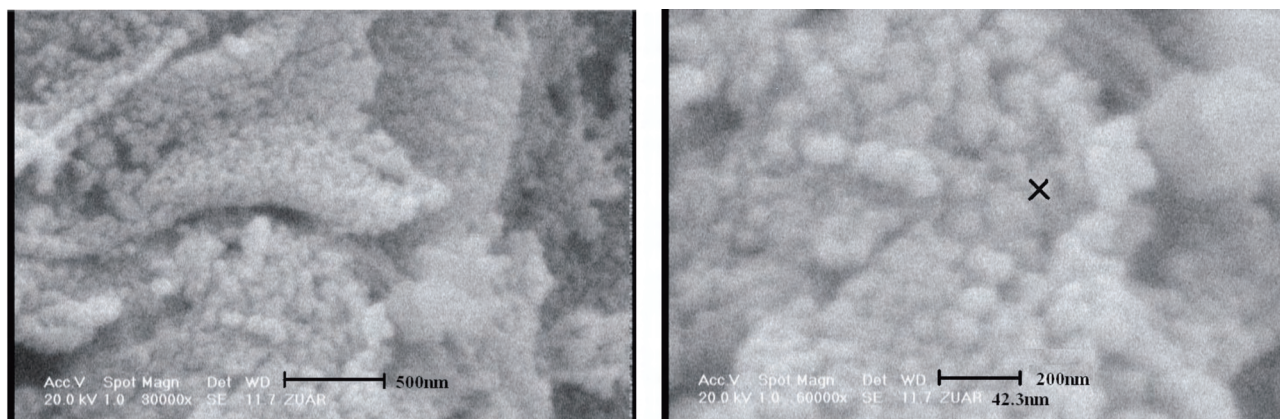


Figure 1. X-ray diffraction pattern of the synthesized silica nanoparticles.

Table. Synthesis of ketoamide derivatives **4a-j** in the presence of silica nanoparticles.

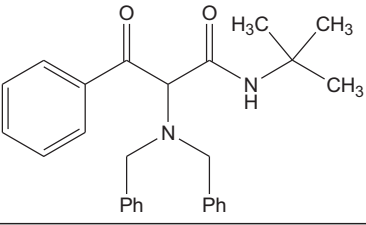
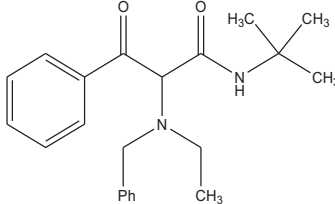
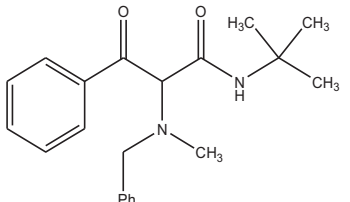
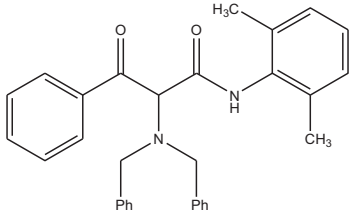
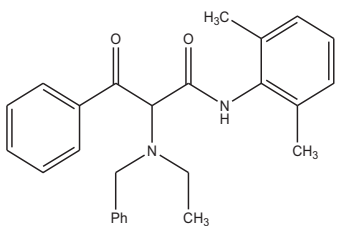
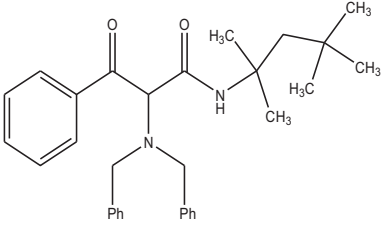
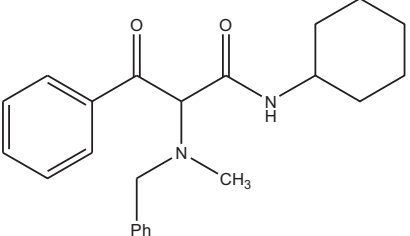
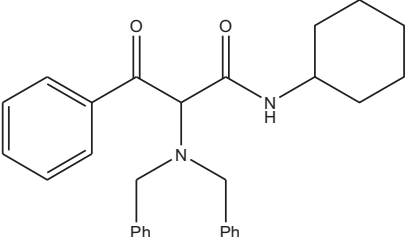
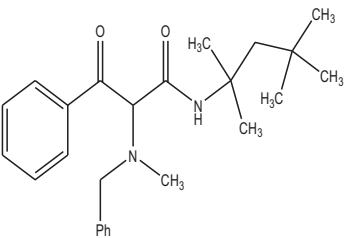
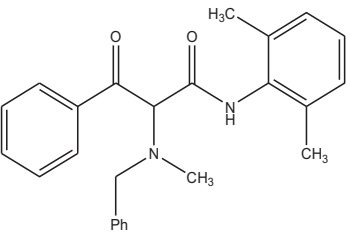
4	R	R'	Product	Yield (%)
a	Benzyl	<i>tert</i> -Butyl		92
b	Et	<i>tert</i> -Butyl		93
c	Me	<i>tert</i> -Butyl		92
d	Benzyl	2,6-Dimethylphenyl		95
e	Et	2,6-Dimethylphenyl		90
f	Benzyl	1,1,3,3-Tetramethylbutyl		90

Table. Continued.

4	R	R'	Product	Yield (%)
g	Me	Cyclohexyl		92
h	Benzyl	Cyclohexyl		91
i	Me	1,1,3,3 - Tetramethylbutyl		89
j	Me	2,6-Dimethylphenyl		88

^aYield of isolated **4**

The structures of the products were elucidated from their IR, ¹H-NMR, ¹³C-NMR, and elemental analyses. For example, the ¹H-NMR spectrum of **4a** consisted of a singlet for 3CH₃ of *tert*-butyl ($\delta = 1.33$ ppm), and an AB quartet for the H-atoms of CH₂Ph ($\delta = 3.86$ and 4.44 ppm, $J = 14.0$ Hz), a singlet for CH ($\delta = 4.91$), and a singlet for NH ($\delta = 6.84$). The aryl groups exhibited characteristic signals in the aromatic region of the spectrum. The ¹H-decoupled ¹³C-NMR spectrum of **4a** showed 16 distinct signals; partial assignment of these signals is given in the experimental section. The ¹H- and ¹³C-NMR spectra of compounds **4b-j** were similar to those of **4a**, except for the aromatic moiety, and the alkyl groups, which exhibited characteristic signals with appropriate chemical shifts.

Although we have not established the mechanism of the reaction in an experimental manner, a plausible reaction sequence that accounts for the formation of **4** is shown in Scheme 2. Thus condensation of 2-oxo-

2-phenylacetaldehyde (**2**) and secondary amine **1** would give the iminium ion intermediate **5**, which would react with the alkyl isocyanide **3** to afford intermediate **6**. The ionic intermediate **6** was unstable and quickly converted to a compound **4** (Scheme 2).

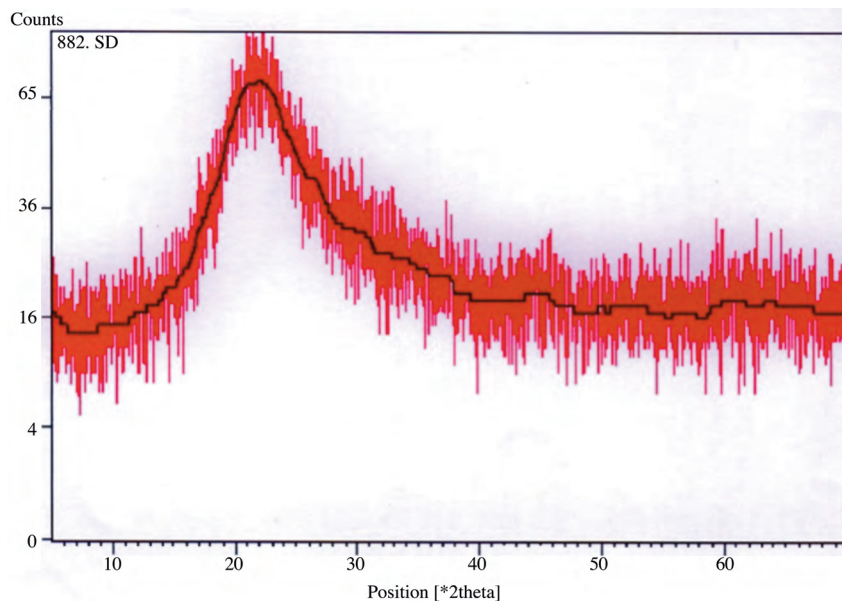
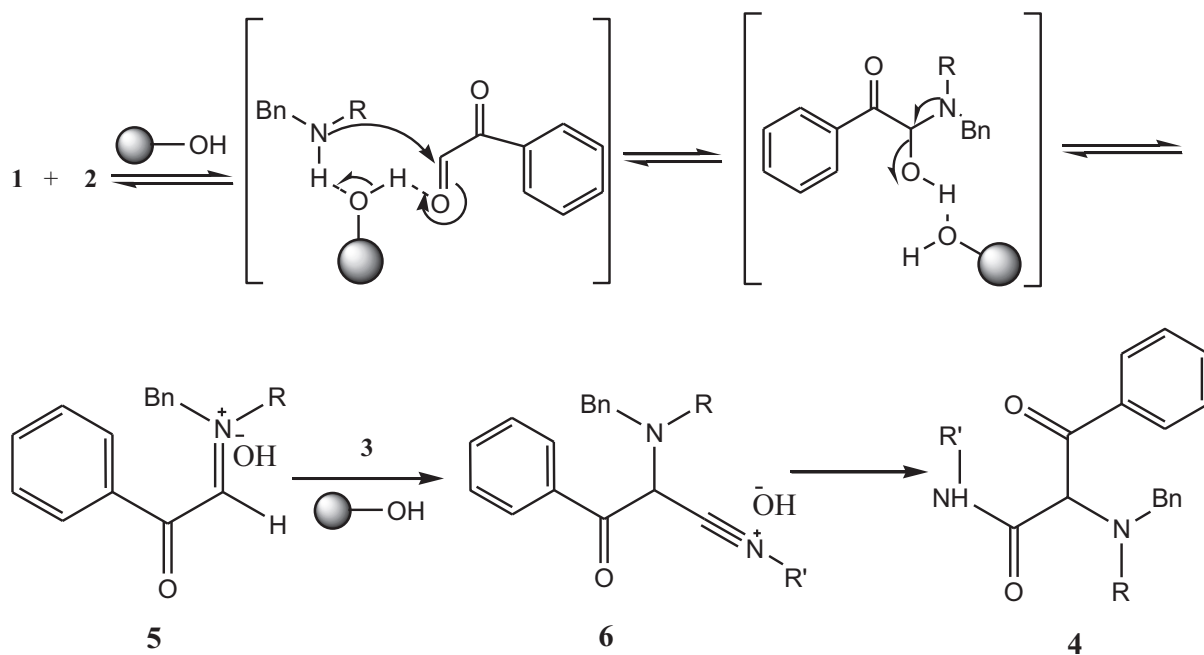


Figure 2. SEM of the synthesized of silica nanoparticles.



Scheme 2. Proposed mechanism for the formation of ketoamide derivatives **4** in the presence of silica nanoparticles.

The silica NPs could be recycled and reused by separating them from the mixture through centrifugation, and frequent washing with EtOH and then drying under vacuum to remove the residual solvent. The results show that the yield of products was only slightly reduced after 5 runs.

Conclusions

We developed an efficient route for the one-pot synthesis of ketoamide derivatives **4** from simple and readily available isocyanides **3**, secondary amines **1**, and 2-oxo-2-phenylacetaldehyde (**2**) in the presence of silicaNPs. The ease of work-up and high yields of products make this procedure a useful addition to modern synthetic methods.

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