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Three-component Passerini-Smiles coupling reaction of aldehydes and isocyanides with tropolone catalyzed by silica nanoparticles

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The use of Smiles rearrangement in Passerini-type couplings with tropolone allows very straightforward multicomponent formation of α-aryloxy amide derivatives. The 3-component addition of isocyanide to tropolone (2-hydroxy-2,4,6-cycloheptatrien-1-one) and aldehyde in the presence of silica nanoparticles (silica NP, ca. 70 nm) proceeds easily in methanol to form the title compounds in a new Passerini-Smiles-type reaction.

Key Words: Smiles rearrangement, Passerini-type coupling, tropolone, isocyanide, silica nanoparticles, Passerini-Smiles-type reaction

Introduction

Multicomponent reactions allow more than 2 facile and flexible building blocks to be combined in practical, timesaving 1-pot operations. Due to their valued features such as atom economy, various starting materials and products, inherent simple experimental procedures, and their 1-pot character, they are perfectly suited for automated synthesis.[1] Therefore, MCRs have attracted much attention because of their exceptional synthetic applications.[2−7] Since all the organic reactants employed are used and moved toward the target compound the purification of products resulting from MCRs is simple.[8,9] Isocyanide-based multicomponent reactions

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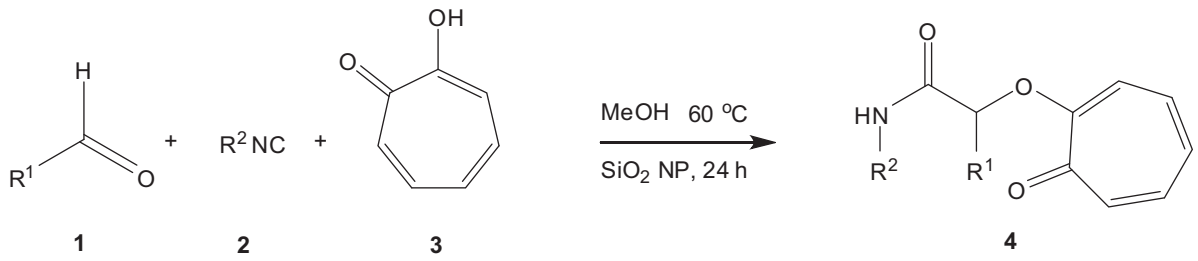
(abbreviated to IMCRs by Ugi and Dömling) have an interesting position. The special features of IMCRs, including unique synthetic potential, convergent nature, high atom economy, ease of implementation, and the generation of molecular diversity, are considered as acceptable factors in the relative efficiency of the reactions.[9−17]

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.[18,19] Due to their enormously large and highly reactive surface area, NPs have potential to exhibit superior catalytic activity in comparison to their bulk counterparts.[20,21]

Recently, we have established a 1-pot method for the preparation of organic compounds.[22−25] As part of our ongoing program to develop efficient and robust methods for the synthesis of heteroatom-containing compounds,[26−34] we report the preparation of a new class of α-aryloxy amide derivatives **4a-g** by a novel 3-component condensation reaction of aldehydes **1**, isocyanides **2**, and tropolone **3** in the presence of silica NPs in excellent yields (Scheme 1).



Scheme 1. Three-component synthesis of α-aryloxy amide derivatives **4** in the presence of silica nanoparticles (see Table 1).

Table 1. SiO_2 nanoparticle (NP)-promoted synthesis of α-aryloxy amide derivatives **4**.[a]

4	R^1	R^2	Yield (%)[b]
a	H	Cyclohexyl	82
b	H	*tert*-Butyl	79
c	*n*-Butyl	Cyclohexyl	81
d	Ethyl	*tert*-Butyl	78
e	3-Nitrophenyl	Cyclohexyl	8
f	3-Formylphenyl	Cyclohexyl	78
g	Ethyl	Cyclohexyl	8

[a] See Scheme 1; 0.3 g SiO_2 NP/mmol reactants were applied. [b] Yield of isolated **4.**

Experimental

Starting materials and solvents were obtained from Merck (Germany) and Fluka (Switzerland) and were used without further purification. The methods were used to follow the reactions were TLC and NMR. TLC and NMR

indicated that there was no side product. IR spectra were measured on a Jasco 6300 FTIR spectrometer. ^{1}H- and ^{13}C-NMR spectra ($CDCl_3$) were recorded on a BRUKER DRX-250 AVANCE spectrometer at 400.22 and 100.63 MHz, respectively. Elemental analyses were performed using a Heraeus CHN-O-Rapid analyzer. Mass spectra were recorded on a FINNIGAN-MATT 8430 mass spectrometer operating at an ionization potential of 70 eV. Preparative thin layer chromatography was prepared from Merck silica gel powder.

General procedure for the preparation of compounds 4

Silica NPs (0.3 g) were added to a mixture of aldehyde (1 mmol), tropolone (1 mmol), and isocyanide (1 mmol) in 7 mL of CH_3OH at 60 °C, followed by reflux for 24 h. Silica NPs were isolated from the reaction mixture by simple filtration and washed with methanol (3 mL). The solvent was removed under reduced pressure and the viscous residue was purified by preparative thin layer chromatography (silica gel; petroleum ether–ethyl acetate (2:1)). The solvent was removed under reduced pressure and the products **(4a-g)** were obtained.

N^{1}-cyclohexyl-2-[(7-oxo-1,3,5-cycloheptatrienyl) oxy]acetamide (4a)

White crystal, (yield: 82%), R_f = 0.20 (petroleum ether/ethyl acetate 2:1), mp 96-98 °C. IR (neat): v = 3479, 2939, 1635, 1620, 1595, 1289, 1194, 1091, 773 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 1.10-2.20 (m, 10H, 5 CH_2 of cyclohexyl), 3.80-4.00 (m, 1H, CH–N), 4.54 (s, 2H, OCH_2), 6.80 (d, 1H, CH of tropone, J = 9.6 Hz), 6.98 (t, 1H, CH of tropone, J = 9.0 Hz), 7.10 (t, 1H, CH of tropone, J = 10.2 Hz), 7.25-7.50 (m, 3H, CH of tropone and NH). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 24.75, 25.44, 32.75 (3 CH_2 of cyclohexyl), 48.04 (CH of cyclohexyl), 68.63 (OCH_2), 116.27, 129.78, 132.55, 137.15, 137.91 (5 CH of tropone), 163.57 (C of tropone), 165.96 (C=O of amide), 180.61 (C=O of tropone). Analysis of $C_{15}H_{19}NO_3$ (261.32). (% calculation/found): C: 68.94/68.91, H: 7.33/7.38, N: 5.36/5.32. MS, m/z(%): 261(2), 163 (46), 140 (9), 136 (100), 135 (95), 123 (15), 107 (44), 106 (28), 105 (38), 90 (10), 83 (28), 77 (44), 65 (15), 58 (20), 55 (31), 41 (27).

N^{1}-(tert-butyl)-2-[(7-oxo-1,3,5-cycloheptatrienyl) oxy]acetamide (4b)

White crystal, (yield: 79%). R_f = 0.23 (petroleum ether/ethyl acetate 2:1), mp 75-76 °C. IR (neat): v = 3420, 2982, 1670, 1621, 1594, 1282, 1198, 1089, 775 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 1.44 (s, 9H, CMe_3), 4.45 (s, 2H, OCH_2), 6.79 (d, 1H, CH of tropone, J = 9.6 Hz), 6.98 (t, 1H, CH of tropone, J = 9.0 Hz), 7.10 (t, 1H, CH of tropone, J = 10.2 Hz), 7.20-7.40 (m, 3H, CH of tropone and NH). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 28.68 (CMe_3), 51.41 (CMe_3), 68.73 (CH_2), 116.06, 129.65, 132.51, 137.07, 137.86 (5 CH of tropone), 163.53 (C of tropone), 166.03 (C=O of amide), 180.54 (C=O of tropone). Analysis of $C_{13}H_{17}NO_3$ (235.28). (% calculation/found): C: 66.36/66.39, H: 7.28/7.26, N: 5.95/5.94. MS, m/z(%): 235 (4), 163 (62), 136 (100), 135 (91), 123 (21), 113 (15) 107 (55), 106 (29), 105 (48), 90 (15), 77 (46), 65 (20), 58 (49), 57 (57), 41 (37).

N^{1}-cyclohexyl-2-[(7-oxo-1,3,5-cycloheptatrienyl) oxy]hexanamide (4c)

Yellow oil, (yield: 81%). R_f = 0.26 (petroleum ether/ethyl acetate 2:1), IR (neat): v = 3480, 2935, 1657, 1639, 1595, 1283, 1192, 1080, 778 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 0.92 (t, 3H, CH_3, J = 7.2 Hz),

1.10-2.20 (m, 16H, 5 CH_2 of cyclohexyl and 3 CH_2 of acyclic), 3.70-3.80 (m,1H, CH–N), 4.61 (t, 1H, H-C, J = 5.8 Hz), 6.81 (d, 1H, CH of tropone, J = 9.6 Hz), 6.88 (t, 1H, CH of tropone, J = 10.0 Hz), 7.06 (t, 1H, CH of tropone, J = 10.0 Hz), 6.90-7.60 (m, 3H, CH of tropone and NH). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 13.94 (CH_3), 22.48 (CH_2 of acyclic), 24.77, 25.42, 27.82, 32.60, 32.84 (5 CH_2 of cyclohexyl and acyclic), 47.98 (CH of cyclohexyl), 82.39 (CH); 118.28, 130.03, 132.62, 137.09, 138.03 (5CH of tropone), 164.47 (C of tropone), 169.50 (C=O of amide), 181.42 (C=O of tropone). Analysis of $C_{19}H_{27}NO_3$ (317.42). (% calculation/found): C: 71.89/71.84, H: 8.57/8.59, N: 4.41/4.44. MS, m/z(%): 318 (100), 317 (13), 219 (94), 196 (52), 191 (36), 122 (31), 114 (27), 107 (44), 106 (30), 105 (20), 94 (23), 83 (37), 77 (18), 65 (18), 55 (66), 41 (53).

N^1-(tert-butyl)-2-[(7-oxo-1,3,5-cycloheptatrienyl)oxy]butanamide (4d)

Yellow oil, (yield: 78%). R_f = 0.27 (petroleum ether/ethyl acetate 2:1), IR (neat): v = 3483, 2937, 1673, 1648, 1594, 1281, 1194, 1083, 780 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 1.08 (t, 3H, CH_3, J = 7.2 Hz), 1.37 (s, 9H, CMe_3), 2.00-2.20 (m, 2H, CH_2), 4.40 (t, 1H, H-C, J = 5.4 Hz), 6.88 (d, 1H, CH of tropone, J = 9.6 Hz), 7.08 (t, 1H, CH of tropone, J = 10.0 Hz), 6.94-7.50 (m, 4H, CH of tropone and NH). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 9.26 (CH_3), 25.77 (CH_2); 28.61 (CMe_3), 51.17 (CMe_3), 82.98 (CH), 117.90, 129.86, 132.57, 136.99, 137.96 (5 CH of tropone), 164.37 (C of tropone), 169.41 (C=O of amide), 181.30 (C=O of tropone). Analysis of $C_{15}H_{21}NO_3$ (263.33). (% calculation/found): C: 68.42/68.40, H: 8.04/8.01, N: 5.32/5.37.

N^1-cyclohexyl-2-(3-nitrophenyl)-2-[(7-oxo-1,3,5-cycloheptatrienyl) oxy]acetamide (4e)

White crystal, (yield: 80 %). R_f = 0.26 (petroleum ether/ethyl acetate 2:1), mp 125-126 °C. IR (neat): v = 3265, 2927, 1660, 1657, 1627, 1595, 1592, 1530, 1284, 1193, 1082, 774 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 1.20-2.10 (m, 10H, 5 CH_2 of cyclohexyl), 3.70-3.80 (m, 1H, CH–N), 5.58 (s, 1H, OCH), 6.78 (d, 1H, CH of tropone, J = 10.0 Hz), 6.94-7.35 (m, 4H, CH of tropone), 7.60 (t, 1H, CH of arom, J = 8.0 Hz), 7.93-8.30 (m, 3H, CH of arom and NH), 8.45 (s, 1H, CH of arom). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 24.65, 25.43, 32.67 (3 CH_2 of cyclohexyl), 48.44 (CH of cyclohexyl), 81.08 (CH), 119.43, 121.74, 123.75, 129.77, 130.93, 132.25, 132.74, 137.28, 138.61 (9 CH of tropone and arom), 138.00 (C, arom), 148.43 (C, C-NO_2), 163.10 (C of tropone), 166.65 (C=O of amide), 181.03 (C=O of tropone). Analysis of $C_{21}H_{22}N_2O_5$ (382.41). (% calculation/found): C: 65.96/65.91, H: 5.80/5.82, N: 7.33/7.36.

N^1-cyclohexyl-2-(3-formylphenyl)-2-[(7-oxo-1,3,5-cycloheptatrienyl)oxy]acetamide (4f)

Yellow oil, (yield: 78%). R_f = 0.36 (petroleum ether/ethyl acetate 2:1), IR (neat): v = 3267, 2930, 2851, 1710, 1661, 1650,1621, 1594, 1282, 1191, 1080, 777 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 1.20-2.10 (m, 10H, 5 CH_2 of cyclohexyl), 3.70-3.80 (m, 1H, CH–N of cyclohexyl), 5.57 (s, 1H, OCH), 6.76 (d, 1H, CH of tropone, J = 10.4 Hz), 7.76-7.35 (m, 4H, CH of tropone), 7.60 (t, 1H, CH of arom, J = 7.60 Hz), 7.85-7.97 (m, 3H, CH of arom and NH), 8.04 (s, 1H, CH of arom), 10.06 (s, 1H, CH of ArCOH). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 24.66, 25.46, 32.65 (3 CH_2 of cyclohexyl), 48.32 (CH of cyclohexyl), 81.38 (CH); 118.73, 128.13, 129.57, 129.76, 130.47, 132.33, 132.51, 137.17, 138.39 (9 CH of tropone and arom), 136.77 (C, arom),

137.03 (C, C-NO_2), 163.17 (C of tropone), 167.06 (C=O of amide), 181.01 (C=O of tropone), 191.90 (C=O of ArCOH). Analysis of $C_{22}H_{23}NO_4$ (365.42). (% calculation/found): C: 72.31/72.35, H: 6.34/6.36, N: 3.83/3.85.

N^1-cyclohexyl-2-[(7-oxo-1,3,5-cycloheptatrienyl)oxy]butanamide (4g)

White crystal, (yield: 80%), R_f = 0.37 (petroleum ether/ethyl acetate 2:1), mp 94-95 °C. IR (neat): v = 3491, 2933, 1655, 1642, 1597, 1280, 1193, 1082, 767 cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$) δ (ppm): 9.41 (t, 3H, CH_3, J = 7.4 Hz), 1.10-2.20 (m, 12H, 5 CH_2 of cyclohexyl and CH_2 of acyclic), 3.70-3.80 (m, 1H, CH–N); 4.59 (t, 1H, H-C, J = 5.4 Hz), 6.88 (d, 1H, CH of tropone, J = 9.6 Hz), 7.06 (t, 1H, CH of tropone, J = 10.0 Hz), 6.90-7.60 (m, 4H, CH of tropone and NH). ^{13}C-NMR (100 MHz, $CDCl_3$) δ (ppm): 9.41 (CH_3), 24.75, 25.43, 25.99, 32.64, (4 CH_2 of cyclohexyl and acyclic), 47.98 (CH of cyclohexyl), 83.12 (CH), 118.33, 130.03, 132.57, 137.04, 138.06 (5 CH of tropone), 164.48 (C of tropone), 169.25 (C=O of amide), 181.42 (C=O of tropone). Analysis of $C_{17}H_{23}NO_3$ (289.37). (% calculation/found): C: 70.56/70.53, H: 8.01/8.02, N: 4.84/4.86.

Results and discussion

The aldehyde **1** was trapped by the isocyanide **2** and tropolone **3** in the presence of silica NPs in methanol at 60 °C over 24 h, leading to the formation of α-aryloxy amide derivatives **4** (Scheme 1 and Table 1). The reaction proceeded smoothly and cleanly under mild and neutral conditions and no side reactions were observed.

Silica NPs were prepared from silica gel HF_{254} residues. The morphology and grain size of the silica NPs were investigated by SEM (Figure 1).[33]

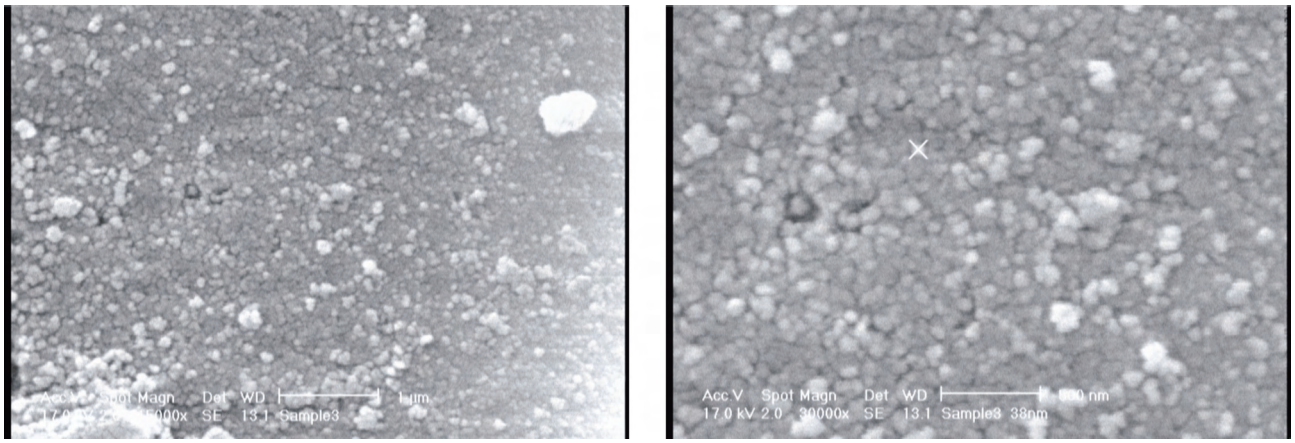


Figure 1. SEM of the synthesized silica nanoparticles.

Silica NPs were found to catalyze the synthesis of α-aryloxy amide derivatives from aldehydes **1**, isocyanides **2**, and tropolone **3** in methanol at 60 °C with high efficiency (Scheme 1 and Table 1). We also used silica gel powder (Merck) instead of silica NPs in this reaction, but increasing reaction times and decreasing α-aryloxy amide yields were observed (Scheme 1 and Table 1). The use of just 0.3 g of silica NPs per mmol of reactants is sufficient to push the reaction forward. Higher amounts of silica NP (0.4 g) did not improve the result to a great extent (Table 2, entries 4-7).

Table 2. Synthesis of α-aryloxy amide **4a** from the reaction of formaldehyde, cyclohexylisocyanide, and tropolone under various conditions.

Entry	Catalyst[a] or solvent	Temp (°C)	Time (h)	Yield (%)[b]
1	MeOH	60	72	35
2	H_2O	90	72	27
3	neat	90	72	25
4	MeOH/Silica gel powder (Merck) (1 g)	60	48	67
5	MeOH/SiO_2 NP (0.1 g)	60	24	70
6	MeOH/SiO_2 NP (0.3 g)	60	24	82
7	MeOH/SiO_2 NP (0.4 g)	60	24	82

[a] Amount of SiO_2 catalyst per mmol of reactants. [b] Yields of isolated **4a**.

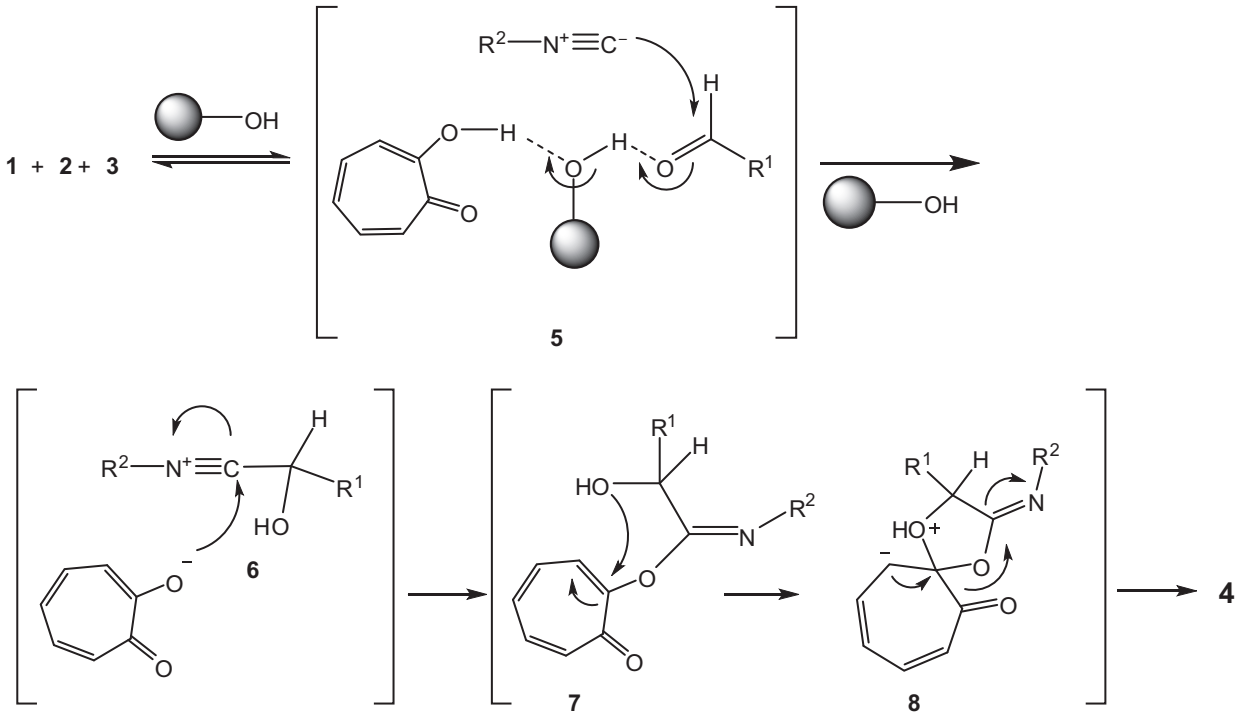
In order to investigate the effects of other reaction media in this reaction, we carried out the described condensation in MeOH, H_2O, and solvent-free (neat) systems (Table 2, entries 1-3).

The structures of the products were deduced from their IR, ^{1}H-NMR, and ^{13}C-NMR spectra, and mass spectrometry. The characterization data of the compounds are given below.

The ^{1}H-NMR spectrum of **4a** consisted of several multiplets for the cyclohexyl (δ = 1.10-2.20 ppm) moieties, a multiplet for the NCH (δ = 3.80-4.00 ppm) of cyclohexyl moiety, a singlet for the OCH_2 (δ = 4.54 ppm) of the acyclic part, a doublet for the tropolone hydrogen (δ = 6.80 ppm, J = 9.6 Hz), a triplet for the tropolone hydrogen (δ = 6.98 ppm, J = 9.0 Hz), a triplet for the tropolone hydrogen (δ = 7.10 ppm, J = 10.2 Hz), and a multiplet for a tropolone hydrogen and NH (δ = 7.25-7.50 ppm). The ^{1}H-decoupled ^{13}C-NMR spectrum of **4a** showed 13 distinct signals. Partial assignment of these signals is given in the experimental section. The ^{1}H- and ^{13}C-NMR spectra of compounds **4b-g** were similar to those of **4a**, except for the aromatic or aliphatic moieties, which exhibited characteristic signals with appropriate chemical shifts.

A mechanistic pathway for this reaction is provided in Scheme 2. On the basis of the chemistry of isocyanides, it is reasonable to assume that the first step may involve nucleophilic addition of the isocyanide **2** to aldehyde **1**, which is facilitated by its protonation with the tropolone **3**, leading to nitrilium intermediate **6**. This intermediate may be attacked by the conjugate base of the tropolone **3** to form 1:1:1 adduct **7**. The intermediate **7** may undergo Smiles rearrangement to afford the isolated α-aryloxy amide derivatives **4** via intermediate **8**.

It may be speculated that the polar amphoteric surface (hydroxyl groups of the silica NP) facilitates the interaction of adsorbed weak acidic and basic components due to stabilization of the corresponding transition states and intermediates by H-bonding. This interaction with the neighboring silanol groups is shown in Scheme 2 for the first reaction step. Participation of 2 proximate silanol groups (one as a H-bond donor and the other as a H-bond acceptor) in the reaction mechanism also seems plausible.



Scheme 2. Proposed mechanism for the formation of α-aryloxy amide derivatives **4** in the presence of silica nanoparticles.

Conclusions

The reported method offers a mild, simple, and efficient route for the preparation of α-aryloxy amide derivatives **4** from aldehydes **1**, isocyanides **2**, and tropolone **3** in the presence of silica NPs. Its ease of work-up, high yields, and fairly mild reaction conditions make it a useful addition to modern synthetic methodologies. Other aspects of this process are under investigation.

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