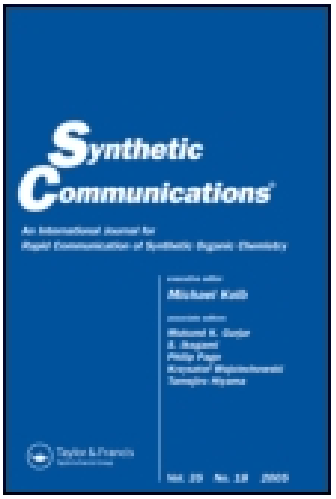

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Silica Gel Promotes Cascade Synthesis of 2-(Heteroaryl)acetamide Derivatives from Isocyanides, Dialkylamines, and Heteroarylcarbaldehydes

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SILICA GEL PROMOTES CASCADE SYNTHESIS OF 2-(HETEROARYL)ACETAMIDE DERIVATIVES FROM ISOCYANIDES, DIALKYLAMINES, AND HETEROARYLCARBALDEHYDES

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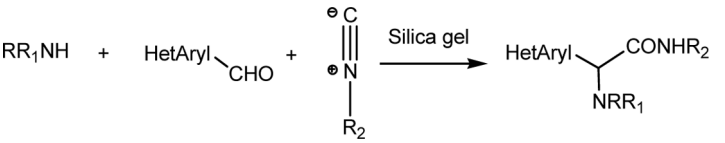
GRAPHICAL ABSTRACT



Abstract *Addition of an isocyanide to an iminium ion intermediate that forms from the reaction between a heteroarylcarbaldehyde and a secondary amine leads to formation of sterically congested 2-(heteroaryl)acetamide derivatives in the presence of silica gel at room temperature. The structures of the products were deduced from their ^{1}H NMR, ^{13}C NMR, and infrared spectra and mass spectrometry.*

Keywords Heteroarylcarbaldehyde; iminium ion; isocyanide; multicomponent reaction; secondary amine; silica gel

INTRODUCTION

Multicomponent reactions (MCRs)[1–9] have recently emerged as valuable tools in the preparation of structurally diverse chemical libraries of druglike heterocyclic compounds.[1–3] In 1921, Passerini[10] pioneered the use of isocyanides and successfully developed a three-component synthesis of α-acyloxycarboxamides by reaction of carboxylic acid, an aldehyde, and an isonitrile.[11] However, the most important breakthrough came in 1959 when Ugi[12,13] described a four-component synthesis of α -acylamino amides from an aldehyde, an amine, an acid, and an isocyanide.[12,13]

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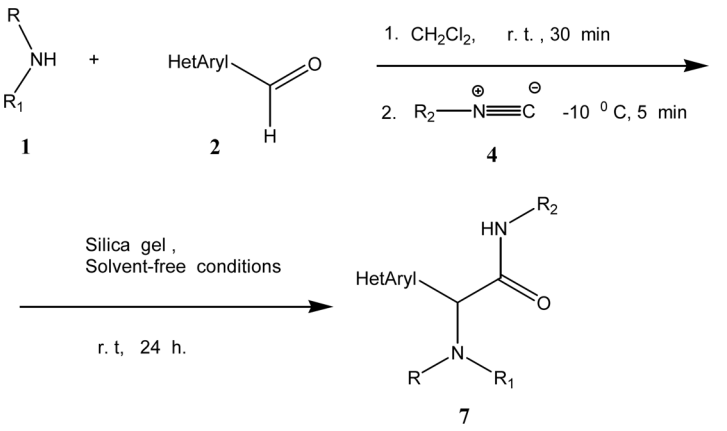


Scheme 1. Preparation of 2-(heteroaryl)acetamide derivatives **7** (see Table 1).

This reaction, named after Ugi (Ugi 4CR or U-4CR) has become one of the most investigated transformations during the past decade, in conjunction with enabling technologies such as high-throughput screening and combinatorial chemistry.[14–20] The ability of an isonitrile to undergo easy R-addition with a nucleophile and an electrophile under mild conditions has made them popular reactants for the development of novel MCRs.[21] Isocyanides,[22] regarded for many years as compounds with an unpleasant odor and very few chemical and pharmaceutical applications, are now looked upon as useful synthons, attributed primarily to the renaissance of the isocyanide-based MCR[14,23] for the Passerini three-component (P-3CR)[24,25] and more importantly the Ugi four-component (U-4CR) reactions.[12,13,26]

Thiophene and pyridine derivatives are important heterocyclic compounds that are widely used as building blocks in many agrochemicals and pharmaceuticals.[27] The benzene ring of a biologically active compound may often be replaced by a thiophene without loss of activity.[28] This is seen in examples such as the nonsteroidal anti-inflammatory drug (NSAID) lornoxicam, the thiophene analog of piroxicam.[28] As part of our ongoing program to develop efficient and robust methods for the preparation of organic compounds,[29–39] we sought to develop a convenient preparation of sterically congested 2-(heteroaryl)acetamide derivatives **7** from an isocyanide, a secondary amine, and heteroarylcarbaldehydes in the presence of silica gel at room temperature in excellent yields under neutral conditions (Scheme 1).

RESULTS AND DISCUSSION

As part of our ongoing program to develop efficient and robust methods for the preparation of organic compounds,[29–39] we sought to develop an efficient route for the one-pot synthesis of sterically congested 2-(heteroaryl)acetamide derivatives[40–43] **7** from simple and readily available isocyanides **4**, secondary amines **1,** and heteroarylcarbaldehydes **2** (Scheme 1 and Table 1). The reaction occurs smoothly in the presence of silica-gel powder at ambient temperature to produce 2-(heteroaryl)acetamide derivatives **7** in 78–96% yields (Scheme 1 and Table 1). The completion of the reaction was checked by thin-layer chromatography (TLC).

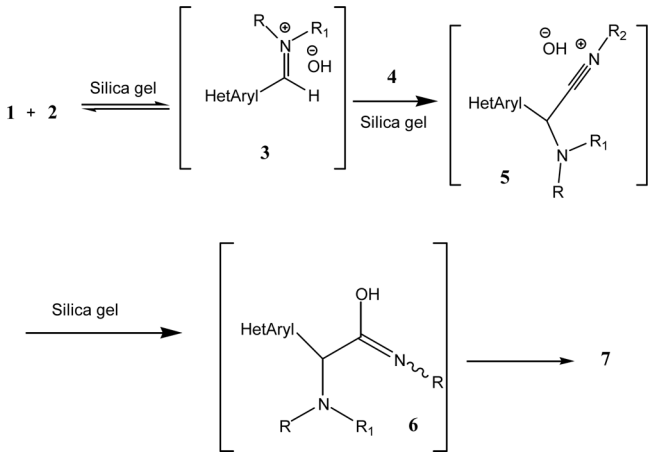
Table 1. Synthesis of 2-(heteroaryl)acetamide derivatives **7** (see Scheme 1)

7	HetAryl	R_2	R_1	R	Yield (%)
a	2-Thienyl	Cyclohexyl	Benzyl	Benzyl	90
b	2-Thienyl	*tert*-Butyl	Benzyl	Benzyl	87
c	2-Thienyl	2,6-Dimethylphenyl	Benzyl	Benzyl	85
d	2-Thienyl	Cyclohexyl	Benzyl	Methyl	78
e	2-Thienyl	*tert*-Butyl	Benzyl	Methyl	82
f	2-Pyridyl	*tert*-Butyl	Benzyl	Benzyl	93
g	2-Pyridyl	Cyclohexyl	Benzyl	Benzyl	96
h	2-Pyridyl	*tert*-Butyl	Benzyl	Methyl	87
i	2-Pyridyl	Cyclohexyl	Benzyl	Methyl	88
j	2-Pyridyl	Benzyl	Benzyl	Benzyl	92
k	2-Pyridyl	Benzyl	Benzyl	Methyl	90
l	4-Pyridyl	Cyclohexyl	Benzyl	Benzyl	80
m	4-Pyridyl	*tert*-Butyl	Benzyl	Benzyl	83

The reaction proceeds smoothly and cleanly under mild conditions, and no side reactions were observed. In the absence of silica-gel powder, the reactions did not lead to the compounds **6**, and in the all cases several products were observed (based on TLC investigations). We also used benzaldehyde in this reaction, but the yield of the corresponding product **7** was very low in the presence of silica-gel powder, and several by-products were observed. It seems that electron-poor aldehyde derivatives such as 2-thiophenecarbaldehyde, 2-pyridinecarbaldehyde, and 4-pyridinecarbaldehyde are more reactive in the reaction.

The structures of compounds **7a–m** were deduced from their ^{1}H NMR, ^{13}C NMR, infrared (IR), and mass spectra. For example, the IR spectrum of **7a** showed strong adsorption at 3346 cm^{-1} indicating the presence of amide, and sharp bands at 1669 and 1500 cm^{-1}, were assigned to the amide carbonyl and the aromatic rings, respectively. The ^{1}H NMR spectrum of **7a** consisted of two multiplets for the cyclohexyl ring $\delta = 0.86$–2.01 (10H, m, Cy) and $\delta = 3.91$–3.93 (1H, m, CH-N of Cy), two methylenes ($\delta = 3.46$ and 3.81, $^2J_{HH} = 14.0$ Hz, $2CH_2$ of two benzyls), a methyne ($\delta = 4.73$ ppm), and an amide hydrogen atom ($\delta = 6.94$ ppm), exchangeable by D_2O. The presence of diastereotopic group (PhCH_AH_B) in the benzyl moieties resulted from the existence of stereogenic center (aliphatic CH) in the molecule of **7a**. The phenyl and thiophene rings gave rise to characteristic signals in the aromatic region. The ^{1}H decoupled ^{13}C NMR spectrum of **7a** showed 17 resonances; for example, the benzylic carbon, the aliphatic carbon (CH), and the amidic carbon were observed at $\delta = 54.6$, 62.7, and 169.1 ppm respectively. The ^{1}H and ^{13}C NMR spectra of compounds **7b–m** were similar to those of **7a**, except for the aromatic and heteroaromatic 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 isocyanides **4**, secondary amines **1**, and heteroarylcarbaldehyde **2** in the presence of the silica gel in an experimental manner, a plausible reaction sequence that accounts for the formation of the product **7** is shown in Scheme 2. Thus, condensation of a heteroarylcarbaldehyde **2** and a secondary amine **1** gave the iminium ion intermediate **3**, which reacted with the alkyl isocyanides **4** to afford the



Scheme 2. Proposed mechanism for the formation of 2-(heteroaryl)acetamide derivatives **7**.

intermediate **5**. The ionic intermediate **5** was converted to intermediate **6**, and the intermediate **6** was unstable and quickly converted to compound **7** (Scheme 2).

CONCLUSIONS

In summary, we develop an efficient route for the one-pot synthesis of sterically congested 2-(heteroaryl)acetamide derivatives **7** from simple and readily available alkyl isocyanides **4**, secondary amine **1,** and heteroarylcarbaldehyde **2** in the presence of silica gel (Scheme 1 and Table 1). Its ease of workup and good yields makes it a useful addition to modern synthetic methodologies.[44–55]

EXPERIMENTAL

Starting materials and solvents were obtained from Merck (Germany) and Fluka (Switzerland) and were used without further purification. The methods used to follow the reactions were TLC and NMR. TLC and NMR indicated that there was no side product. Melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. IR spectra were measured on a Shimadzu IR-460 spectrometer. ^{1}H and ^{13}C NMR spectra were measured ($CDCl_3$ 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. Mass spectra were recorded on a Finnigan-MAT 8430 mass spectrometer operating at an ionization potential of 20 eV. Flash chromatography columns were prepared from Merck silica-gel powder.

General Procedure for the Preparation of Compounds 7a–e, Exemplified by 7a

A mixture of dibenzylamine (0.19 ml, 1 mmol) and 2-thiophenecarbaldehyde (0.09 ml, 1 mmol) in dry CH_2Cl_2 (5 mL) was stirred at room temperature for 0.5 h.

A solution of cyclohexylisocyanide (0.13 ml, 1 mmol) in 2 mL dry CH_2Cl_2 at $-10\,^{\circ}C$ was added rapidly to this mixture, and the mixture stirred for 5 min. at $-10\,^{\circ}C$. Silica-gel powder (Merck, 1 g) was quickly poured into the reaction mixture, and the mixture was allowed to warm up to room temperature. The solvent was removed under reduced pressure, and the residue was allowed to remain for 24 h in solvent-free conditions at room temperature and then 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, and the products were obtained.

Selected Data

***N*-Cyclohexyl-2-(dibenzylamino)-2-(2-thienyl)acetamide (7a).** Orange oil; yield 90%. IR (KBr): $\upsilon = 3346$ (NH), 3030, 2930, 1669, 1500, 1453, 1369 and 1253 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 0.86$–2.01 (m, 10H, Cy), 3.46 and 3.81 (AB quartet, 4H, $^2J_{HH} = 13.8$ Hz, $2CH_2$ of benzyl groups), 3.91–3.93 (m, 1H, CH-N of Cy), 4.73 (s, 1H, CH), 6.94 (bs, 1H, NH, exchanged by D_2O addition), 7.01–7.06 (m, 3H, thiophene), 7.26–7.68 (m, 10H, $2C_6H_5$).^{13}C NMR ($CDCl_3$): $\delta = 24.6$ and 24.7 ($2CH_2$, β of Cy), 25.5 ($1CH_2$, γ of Cy), 33.0 and 33.2 ($2CH_2$, α of Cy), 48.0 (CHNH of Cy), 54.6 ($2CH_2$ of benzyl groups), 62.7 (CH), 125.8, 126.3, 127.4 (3CH of thiophene), 138.6 (C_{ipso} of thiophene), 128.4, 128.6, 128.7 (10 CH), 136.7 [2 C_{ipso}, (C=C) of $2C_6H_5$]; 169.1 (CONH). MS: *m/z* (%); 418 (M+, 2), 294 (10), 292 (100), 196 (20), and 91 (100). $C_{26}H_{30}N_2OS$ (418.21): C, 74.60; H, 7.22; N, 6.69; S, 7.66%. Found: C, 74.30; H, 7.19; N, 6.57; S, 7.51%.

***N*-(*tert*-butyl)-2-(dibenzylamino)-2-(2-thienyl)acetamide (7b).** Colorless oil; ylide: 87%. IR (KBr): $\upsilon = 3361$ (NH), 3030, 2969, 1684, 1515, 1453, 1369, and 1230 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.40$ (s, 9H, CMe_3), 3.44 and 3.82 (AB quartet, 4H, $^2J_{HH} = 13.8$ Hz, $2CH_2$ of benzyl groups), 4.57 (s, 1H, CH), 6.99 (bs, 1H, NH, exchanged by D_2O addition), 7.04–7.07 (m, 3H, thiophene), 7.26–7.38 (m, 10H, 2 C_6H_5). ^{13}C NMR ($CDCl_3$): $\delta = 28.8$ ($3CH_3$ of C*Me*$_3$), 51.1 (C of *C*Me$_3$NH), 54.6 ($2CH_2$ of benzyl groups), 63.2 (CH), 125.9, 126.3, 127.4 (3CH of thiophene), 138.7 (C_{ipso} of thiophene), 128.6, 128.7 (10 CH), 136.2 ($2C_{ipso,}$(C=C) of $2C_6H_5$), 169.5 (CONH). MS: *m/z* (%): 393 (M+, 4), 294 (10), 292 (100), 279 (5), 216 (5), 196 (20), 181 (15), 111 (10) and 91 (100). Anal. calcd. for $C_{24}H_{28}N_2OS$ (392.19): C, 73.43; H, 7.19; N, 7.14%. Found: C, 72.90; H, 7.17; N, 7.12%.

2-(Dibenzylamino)-*N*-(2,6-dimethylphenyl)-2-(2-thienyl)acetamide (7c). Colorless oil; ylide: 85%. IR (KBr): $\upsilon = 3423$ (NH), 3030, 2923, 1646, 1500, 1453, 1392, and 1230 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.84$ and 1.91 (s, 6H, $2CH_3$ of C_6H_3), 3.56 and 4.38 (AB quartet, 2H, $^2J_{HH} = 14$ Hz, CH_2 of benzyl), 4.16 and 4.27 (AB quartet, 2H, $^2J_{HH} = 16.3$ Hz, CH_2 of benzyl), 5.24 (s, 1H, CH), 6.99 (bs, 1H, NH, exchanged by D_2O addition), 6.53–6.85 (m, 3H, C_6H_3), 6.96–7.03 (m, 3H, thiophene), 7.16–7.69 (m, 10H, $2C_6H_5$). ^{13}C NMR ($CDCl_3$): $\delta = 19.0$ and 19.4 ($2CH_3$ of C_6H_3), 52.0 and 54.7 ($2CH_2$ of benzyl groups), 60.8 (CH), 125.1, 126.5, 127.3 (3CH of thiophene), 126.9, 127.0, 127.5 (3CH of C_6H_3), 139.6 (C_{ipso} of thiophene), 128.2, 129.1 (10CH of $2C_6H_5$), 137.8 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 139.4 ($2C_{ipso(C=C)}$ of C_6H_3), 166.1 (CONH). MS: *m/z* (%); 440 (M+, 9), 294 (10), 292

(100), 279 (5), 216 (5), 196 (20), 181 (15), 111 (45), and 91 (100). Anal. calcd. for $C_{28}H_{2\ 8}N_2OS$ (440.19): C, 76.33; H, 6.41; N, 6.36%. Found: C, 76.27; H, 6.32; N, 6.25%.

2-[Benzyl(methyl)amino]-*N*-cyclohexyl-2-(2-thienl)acetamide (7d). Colorless oil; yield: 78%. IR (KBr): $\upsilon = 3301$ (NH), 2931, 1650, 1519, 1450, 1388 and 1249 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.18$–1.94 (m, 10H, Cy), 2.13 (s, 3H, NCH_3), 3.43 and 3.61 (AB quartet, 2H, $^2J_{HH} = 13.3$ Hz, CH_2 of benzyl), 3.12 (m, 1H, CH-N of Cy), 4.36 (s, 1H, CH), 7.18 (bs, 1H, NH, exchanged by D_2O addition), 6.97–7.05 (m, 3H, thiophene) 7.26–7.69 (m, 5H, C_6H_5). ^{13}C NMR ($CDCl_3$): $\delta = 24.7$ ($2CH_2$, β of Cy), 25.5 ($1CH_2$, γ of Cy), 33.0 ($2CH_2$, α of Cy), 39.4 (NCH_3), 47.8 (CH-N of Cy), 59.3 (CH_2 of benzyl), 69.3 (CH), 125.8, 126.5, 127.4 (3CH of thiophene), 138.3 (C_{ipso} of thiophene), 128.3, 128.5, 128.7 (5CH of C_6H_5), ($2C_{ipso(C=C)}$ of C_6H_5), 169.6 (CONH). MS: m/z (%); 342 (M+, 2), 279 (9), 211 (16), 196 (18), 167 (14), 149 (23), 123 (20), 106 (83), 91 (100), 78 (15), 65 (22), 51 (19), and 41 (7). Anal. calcd. for $C_{20}H_{26}N_2OS$ (342.18): C, 70.14; H, 7.65; N, 8.18%. Found: C, 70.12; H, 7.62; N, 8.15%.

2-[Benzyl(methyl)amino]-*N*-(*tert*-butyl)-2-(2-thienyl)acetamide (7e). White crystals; yield: 82%. IR (KBr): $\upsilon = 3292$ (NH), 2923, 1669, 1515, 1461, 1392, and 1230 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.39$ (s, 9H, CMe_3), 2.12 (s, 3H, NCH_3), 3.43 and 3.58 (AB quartet, 2H, $^2J_{HH} = 13.5$ Hz, CH_2 of benzyl), 4.28 (s, 1H, CH), 7.19 (bs, 1H, NH, exchanged by D_2O addition), 6.98–7.03 (m, 3H, thiophene), 7.26–7.33 (m, 5H, C_6H_5). ^{13}C NMR ($CDCl_3$): $\delta = 28.6$ ($3CH_3$ of C*Me*$_3$), 39.4 (NCH_3), 50.8 (1C of *C*Me_3NH), 59.3 (CH_2 of benzyl), 70.0 (CH), 125.7, 126.4, 127.3 (3CH of thiophene), 138.5 (C_{ipso} of thiophene), 128.3, 128.5 (5CH of C_6H_5), 138.2 ($C_{ipso(C=C)}$ of C_6H_5), 169.8 (CONH). MS: m/z (%); 316 (M+, 5), 297 (5), 287 (75), 211 (9), 196 (13), 181 (4), 106 (18), 91 (100), 65 (12), 51 (5), and 41 (5). Anal. calcd. for $C_{18}H_{24}N_2OS$ (316.16): C, 68.32; H, 7.64; N, 8.85%. Found: C, 68.22; H, 7.62; N, 8.55%.

***N*-(*tert*-butyl)-2-(dibenzylamino)-2-(2-pyridyl)acetamide (7f).** Green viscous oil; yield: 93%. IR (KBr): $\upsilon = 3343$ (NH), 3035, 2959, 1699, 1524, 1479, 1367, and 1258 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.38$ (s, 9H, CMe_3), 3.50 and 3.94 (AB quartet, 4H, $^2J_{HH} = 13.75$ Hz, $2CH_2$ of benzyl groups), 4.49 (s, 1H, CH), 7.21 (bs, 1H, NH, exchanged by D_2O addition), 7.26–8.66 (m, 14H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 28.8$ ($3CH_3$ of C*Me*$_3$), 51.0 (C of *C*Me_3NH), 55.0 ($2CH_2$ of benzyl groups), 68.9 (CH), 127.6, 128.5, 128.9 (10CH of $2C_6H_5$), 138.7 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 122.5, 125.4, 136.1, 148.9 (4CH of pyridine), 156.1 (C_{ipso} of pyridine), 169.7 (CONH). Anal. calcd. for $C_{25}H_{29}N_3O$ (387.52): C, 77.48; H, 7.54; N, 10.84%. Found: C, 77.41; H, 7.49; N, 7.50%.

***N*-Cyclohexyl-2-(dibenzylamino)-2-(2-pyridyl)acetamide (7g).** Colorless oil; yield: 96%. IR (KBr): $\upsilon = 3342$ (NH), 3029, 2934, 1678, 1509, 1453, 1372, and 1252 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.25$–1.92 (m, 10H, Cy), 3.50 and 3.93 (AB quartet, 4H, $^2J_{HH} = 13.75$ Hz, $2CH_2$ of benzyl groups), 3.70–3.85 (m, 1H, CH-N of Cy), 4.54 (s, 1H, CH), 7.21 (bs, 1H, NH, exchanged by D_2O addition), 7.24–8.65 (m, 14H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 24.6$ and 24.7 ($2CH_2$, β of Cy), 25.6 ($1CH_2$, γ of Cy), 33.0 and 33.1 ($2CH_2$, α of Cy), 47.8 (CHNH of Cy), 54.8 ($2CH_2$ of benzyl groups),

68.2 (CH), 127.3, 128.5, 128.9 (10CH of $2C_6H_5$), 138.6 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 122.5, 125.3, 136.2, 148.9 (4CH of pyridine), 156.2 (C_{ipso} of pyridine), 169.2 (CONH). MS: *m/z* (%); 414 (M+, 2), 287 (80), 218 (35), 197 (12), 196 (25), 180 (10), 119 (7) and 91 (100). Anal. calcd. for $C_{27}H_{31}N_3O$ (413.55): C, 78.42; H, 7.56; N, 10.16%. Found: C, 78.40; H, 7.53; N, 10.11%.

2-[Benzyl(methyl)amino]-*N*-(*tert*-butyl)-2-(2-pyridyl)acetamide (7h). Red viscous oil; yield: 87%; IR (KBr): $\upsilon = 3365$ (NH), 3031, 2964, 1684, 1520, 1435, 1364, and 1227 cm^{-1}. ^1H NMR ($CDCl_3$) $\delta = 1.37$ (s, 9H, CMe_3), 2.11 (s, 3H, NCH_3), 3.54 and 3.67 (AB quartet, 2H, $^2J_{HH} = 13.25$ Hz, CH_2 of benzyl), 4.16 (s, 1H, CH), 7.19 (bs, 1H, NH, exchanged by D_2O addition), 7.22–8.64 (m, 9H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 28.7$ ($3CH_3$ of CMe_3), 39.3 (NCH_3), 50.9 (1C of CMe_3NH), 59.7 (CH_2 of benzyl), 77.22 (CH), 127.3, 128.4, 128.8 (5CH of C_6H_5), 138.9 ($C_{ipso(C=C)}$ of C_6H_5), 122.7, 124.5, 136.4, 149.4 (4CH of pyridine), 162.2 (C_{ipso} of pyridine), 169.6 (CONH). MS: *m/z* (%); 312 (M+, 20), 287 (2), 211 (100), 196 (2), 192 (7), 121 (10), 107 (3), and 91 (30). Anal. calcd. for $C_{19}H_{25}N_3O$ (311.42): C, 73.28; H, 8.09; N, 13.49%. Found: C, 73.25; H, 8.05; N, 13.43%.

2-[Benzyl(methyl)amino]-*N*-cyclohexyl-2-(2-pyridyl)acetamide (7i). Colorless oil; yield: 88%; IR (KBr): $\upsilon = 3332$ (NH), 3031, 2939, 1677, 1530, 1455, 1372, and 1251 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.18$–1.94 (m, 10H, Cy), 2.10 (s, 3H, NCH_3), 3.53 and 3.66 (AB quartet, 2H, $^2J_{HH} = 13.25$ Hz, CH_2 of benzyl), 3.74–3.82 (m, 1H, CH-N of Cy), 4.21 (s, 1H, CH), 7.20 (bs, 1H, NH, exchanged by D_2O addition), 7.22–8.63 (m, 9H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 24.7$ ($2CH_2$, β of Cy), 25.5 ($1CH_{2,\gamma}$ of Cy), 32.9 and 33.0 ($2CH_2$, α of Cy), 39.3 (NCH_3), 47.7 (CH-N of Cy), 59.8 (CH_2 of benzyl), 75.7 (CH), 127.2, 128.4, 129.0 (5CH of C_6H_5), 138.2 ($C_{ipso(C=C)}$ of C_6H_5), 122.8, 124.5, 136.4, 149.4 (4CH of pyridine), 156.4 (C_{ipso} of pyridine), 169.5 (CONH). MS: *m/z* (%); 338 (M+, 2), 211 (100), 182 (3), 170 (4), 121 (10), 105 (4) and 91 (55). Anal. calcd. for $C_{21}H_{27}N_3O$ (337.46) C, 74.74; H, 8.06; N, 12.45%. Found: C, 74.70; H, 8.01; N, 12.39%.

***N*-Benzyl-2-(dibenzylamino)-2-(2-pyridyl)acetamide (7j).** Yellow viscous oil; yield: 92%. IR (KBr): $\upsilon = 3339$ (NH), 3028, 2930, 1677, 1515, 1455, 1366, and 1250 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 3.49$ and 3.96 (AB quartet, 4H, $^2J_{HH} = 13.75$ Hz, $2CH_2$ of benzyl groups), 4.55 (m, 2H, CH_2-NH), 4.64 (s, 1H, CH), 6.98 (bs, 1H, NH, exchanged by D_2O addition), 7.36 (m, 19H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 43.4$ (CH_2-NH), 54.9 ($2CH_2$ of benzyl groups), 68.0 (CH), 127.3, 127.4, 127.7, 128.5, 128.7, 128.9 (15CH of $3C_6H_5$), 138.3 and 138.5 ($3C_{ipso(C=C)}$ of $3C_6H_5$), 122.6, 125.4, 136.3, 148.9 (4CH of pyridine), 155.9 (C_{ipso} of pyridine), 170.4 (CONH). MS: *m/z* (%); 422 (M+, 30), 287 (95), 226 (45), 211 (5), 197 (23), 195 (40), 180 (10), 106 (13), and 91 (100). Anal. calcd. for $C_{28}H_{27}N_3O$ (421.22): C, 79.78; H, 6.46; N, 9.97%. Found: C, 79.75; H, 6.40; N, 9.80%.

***N*-Benzyl-2-[benzyl(methyl)amino]-2-(2-pyridyl)acetamide (7k).** Yellow viscous oil; yield: 90%. IR (KBr): $\upsilon = 3339$ (NH), 3030, 2932, 1680, 1519, 1454, 1362, and 1240 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 2.13$ (s, 3H, NCH_3), 3.56 and 3.63 (AB quartet, 2H, $^2J_{HH} = 13.5$ Hz, CH_2 of benzyl), 4.33 (1H, s, CH), 4.53 (m, 2H, CH_2-NH), 7.22 (bs, 1H, NH, exchanged by D_2O addition), 7.27–8.65 (m, 14H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 39.5$ (NCH_3), 43.3 (CH_2-NH), 59.8 (CH_2 of benzyl),

75.2 (CH), 127.2, 127.3, 127.7, 128.3, 128.6, 129.0 (10CH of C_6H_5), 138.1 and 138.3 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 122.9, 124.8, 136.5, 149.5 (4CH of pyridine), 156.0 (C_{ipso} of pyridine), 170.6 (CONH). MS: *m/z* (%); 345 (M+, 4), 226 (7), 211 (25), 120 (20), 106 (15), 91 (50), 58 (28), and 43 (100). Anal. calcd. for $C_{22}H_{23}N_3O$ (345.44): C, 76.49; H, 6.71; N, 12.16%. Found: C, 76.40; H, 6.68; N, 12.13%.

***N*-Cyclohexyl-2-(dibenzylamino)-2-(4-pyridyl)acetamide (7l).** Colorless oil; yield: 80%. IR (KBr): $\upsilon = 3324$ (NH), 3033, 2924, 1666, 1500, 1453, 1375, and 1258 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.25$–2.03 (m, 10H, Cy), 3.32 and 3.83 (AB quartet, 4H, $^2J_{HH} = 13.75$ Hz, $2CH_2$ of benzyl groups), 3.86–3.99 (m, 1H, CH-N of Cy), 4.34 (s, 1H, CH), 7.12 (bs, 1H, NH, exchanged by D_2O addition), 7.15–8.64 (m, 14H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 24.6$ and 24.7 ($2CH_2$, β of Cy), 25.5 ($1CH_2$, γ of Cy), 33.0 and 33.4 ($2CH_2$, α of Cy), 48.0 (CHNH of Cy), 54.6 ($2CH_2$ of benzyl groups), 66.3 (CH), 127.6, 128.5, 128.8 (10CH of $2C_6H_5$), 138.1 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 125.3, 149.6 (4CH of pyridine), 149.8 (C_{ipso} of pyridine), 168.8 (CONH). MS: *m/z* (%); 413 (M+, 2), 279 (5), 211 (13), 196 (15), 106 (80), and 91 (100). Anal. calcd. for $C_{27}H_{31}N_3O$ (413.55): C, 78.42; H, 7.56; N, 10.16%. Found: C, 78.39; H, 7.50; N, 10.11%.

***N*-(*tert*-Butyl)-2-(dibenzylamino)-2-(4-pyridyl)acetamide (7m).** Colorless oil; yield: 83%. IR (KBr): $\upsilon = 3334$ (NH), 3032, 2964, 1696, 1520, 1481, 1365, and 1258 cm^{-1}. ^1H NMR ($CDCl_3$): $\delta = 1.42$ (s, 9H, CMe_3), 3.29 and 3.85 (AB quartet, 4H, $^2J_{HH} = 13.75$ Hz, $2CH_2$ of benzyl groups), 4.29 (s, 1H, CH), 7.10 (bs, 1H, NH, exchanged by D_2O addition), 7.22–8.86 (m, 14H, arom). ^{13}C NMR ($CDCl_3$): $\delta = 28.8$ ($3CH_3$ of C*Me*$_3$), 51.3 (C of *C*Me$_3$NH), 54.7 ($2CH_2$ of benzyl groups), 66.8 (CH), 127.6, 128.5, 128.8 (10CH of $2C_6H_5$), 139.7 ($2C_{ipso(C=C)}$ of $2C_6H_5$), 125.4, 149.6 (4CH of pyridine), 150.3 (C_{ipso} of pyridine), 169.2 (CONH). MS: *m/z* (%); 388 (M+, 2), 297 (4), 287 (75), 196 (12), 106 (19), and 91 (100). Anal. calcd. for $C_{25}H_{29}N_3O$ (387.52): C, 77.48; H, 7.54; N, 10.84%. Found: C, 77.40; H, 7.50; N, 10.73%.

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