SUPPORTING INFORMATION

Efficient Microwave Access to Polysubstituted Amidines from Imidoylbenzotriazoles

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General Procedure for the Preparation of *N*-Acylbenzotriazoles 4a–l. Thionyl chloride (4.00 mL, 55 mmol) and benzotriazole (18.45 g, 155 mmol) in methylene chloride (100 mL) were added dropwise to the carboxylic acid (50 mmol) in methylene chloride (100 mL). The reaction was monitored by TLC. The precipitated white solid was filtered off and the solvent was removed *in vacuo* to obtain the crude product, which was purified by recrystallization from chloroform/hexanes to obtain *N*-acylbenzotriazoles 4a–l.

1-Heptanoyl-1*H*-1,2,3-benzotriazole (4b): colorless needles (from chloroform/hexanes); mp 48–50 °C (lit.⁹ mp 50–52 °C); yield, 92% (10.63 g); ¹H NMR δ 0.91 (t, *J* = 7.0 Hz, 3H), 1.29–1.52 (m, 6H), 1.86–1.96 (m, 2H), 3.39–3.44 (m, 2H), 7.49 (td, *J* = 7.3, 0.8 Hz, 1H), 7.64 (td, *J* = 7.3, 0.8 Hz, 1H), 8.11 (d, *J* = 8.2 Hz, 1H), 8.28 (d, *J* = 8.2 Hz, 1H); ¹³C NMR δ 13.9, 22.4, 24.3, 28.7, 31.4, 35.4, 114.3, 120.0, 125.9, 130.2, 131.0, 146.1, 172.6.

1-(2-Thienylcarbonyl)-1*H*-benzotriazole (4c): colorless needles (from chloroform/hexanes); mp 172–173 °C (lit.⁷ mp 173–175 °C); yield 93% (10.65 g); ¹H NMR δ 7.30 (dd, *J* = 5.0, 4.0 Hz, 1H), 7.56 (t, *J* = 7.7 Hz, 1H), 7.71 (t, *J* = 7.6 Hz, 1H), 7.91 (dd, *J* = 5.0, 0.9 Hz, 1H), 8.18 (d, *J* = 8.2 Hz, 1H), 8.42 (d, *J* = 8.3 Hz, 1H), 8.60 (dd, *J* = 4.0, 0.9 Hz, 1H); ¹³C NMR δ 114.8, 120.2, 126.2, 128.0, 130.4, 132.1, 133.3, 137.1, 138.4, 145.7, 159.1.

1-(4-Nitrobenzoyl)-1*H*-1,2,3-benzotriazole (4d): white microcrystals (from chloroform/hexanes); mp 191–193 °C (lit.¹⁰ mp 192–193 °C); yield, 91% (12.19 g); ¹H NMR δ 7.61 (t, *J* = 7.6 Hz, 1H), 7.78 (t, *J* = 7.6 Hz, 1H), 8.21 (d, *J* = 8.2 Hz, 1H), 8.29–8.45 (m, 5H); ¹³C NMR δ 114.7, 120.5, 123.5, 126.9, 131.0, 132.0, 132.6, 136.9, 145.9, 150.5, 165.0.

1-(2-Furoyl)-1*H*-1,2,3-benzotriazole (4e): colorless needles (from chloroform/hexanes); mp 170–172 °C (lit.⁸ mp 171–173 °C); yield, 92% (9.80 g); ¹H NMR δ 6.74 (dd, *J* = 3.5, 1.3 Hz, 1H),

7.55 (t, $J = 7.7$ Hz, 1H), 7.70 (td, $J = 7.7$, 0.8 Hz, 1H), 7.88 (s, 1H), 8.15 (d, $J = 8.0$ Hz, 1H), 8.17 (s, 1H), 8.41 (d, $J = 8.2$ Hz, 1H); ^{13}C NMR δ 112.9, 114.7, 120.1, 124.7, 126.3, 130.4, 132.1, 144.5, 145.5, 148.9, 155.0.

1-(Phenylacetyl)-1*H*-1,2,3-benzotriazole (4f): colorless needles (from chloroform/hexanes); mp 63–65 °C (lit.⁸ mp 65–66 °C); yield, 89% (10.55 g); ^1H NMR δ 4.73 (s, 2H), 7.28–7.40 (m, 3H), 7.46–7.53 (m, 3H), 7.64 (td, $J = 7.7$, 0.8 Hz, 1H), 8.13 (d, $J = 8.3$ Hz, 1H), 8.27 (d, $J = 8.2$ Hz, 1H); ^{13}C NMR δ 41.9, 114.4, 120.1, 126.2, 127.6, 128.7, 129.8, 130.4, 131.1, 132.4, 146.2, 170.2.

1-(4-Methoxybenzoyl)-1*H*-1,2,3-benzotriazole (4g): colorless prisms (from chloroform/hexanes); mp 109–111 °C (lit.¹¹ mp 102–103 °C); yield, 92% (11.64 g); ^1H NMR δ 3.93 (s, 3H), 7.06 (d, $J = 8.9$ Hz, 2H), 7.51 (t, $J = 7.2$ Hz, 1H), 7.69 (t, $J = 7.1$ Hz, 1H), 8.16 (d, $J = 6.2$ Hz, 1H), 8.29 (d, $J = 8.9$ Hz, 2H), 8.37 (d, $J = 8.2$ Hz, 1H); 55.6, 103.3, 113.9, 114.8, 120.0, 126.1, 130.1, 134.4, 145.6, 164.2.

1-Benzoyl-1*H*-1,2,3-benzotriazole (4h): white needles (from chloroform/hexanes); mp 110–112 °C (lit.⁷ mp 116–117 °C); yield, 96% (10.70 g); ^1H NMR δ 7.67–7.52 (m, 3H), 7.70–7.72 (m, 2H), 8.23–8.15 (m, 3H), 8.38 (d, $J = 8.3$ Hz, 1H); ^{13}C NMR δ 114.7, 120.1, 126.3, 128.4, 130.3, 131.4, 131.7, 132.3, 133.6, 145.7, 166.6.

1-(4-Methylbenzoyl)-1*H*-1,2,3-benzotriazole (4i): colorless prisms (from chloroform/hexanes); mp 121–123 °C (lit.¹⁰ mp 123–124 °C); yield, 95% (11.20 g); ^1H NMR δ 2.47 (s, 3H), 7.37 (d, $J = 8.1$ Hz, 2H), 7.52 (t, $J = 7.6$ Hz, 1H), 7.67 (t, $J = 7.6$ Hz, 1H), 8.13 (d, $J = 8.1$ Hz, 3H), 8.14 (d, $J = 8.1$ Hz, 1H), 8.36 (d, $J = 8.2$ Hz, 1H); ^{13}C NMR δ 21.7, 114.7, 120.0, 126.1, 128.5, 129.1, 130.1, 131.8, 132.3, 144.7, 145.6, 166.4.

1-Acetyl-1*H*-1,2,3-benzotriazole (4j): colorless needles (from chloroform/hexanes); mp 50–52 °C (lit.¹³ mp 49–51 °C); yield, 96% (7.73 g); ¹H NMR δ 3.00 (s, 1H), 7.49 (t, J = 7.7 Hz, 1H), 7.63 (t, J = 7.6 Hz, 1H), 8.09 (d, J = 8.2 Hz, 1H), 8.25 (d, J = 8.2 Hz, 1H); ¹³C NMR δ 23.1, 114.2, 120.0, 126.0, 130.2, 130.8, 146.1, 169.4.

1-[(2*E*)-3-Phenylprop-2-enoyl]-1*H*-1,2,3-benzotriazole (4k): colorless needles (from chloroform/hexanes); mp 149–150 °C (lit.⁷ mp 151–152 °C); yield, 96% (11.95 g); ¹H NMR δ 7.48–7.51 (m, 3H), 7.54 (ddd, J = 7.7, 6.9, 1.0 Hz, 1H), 7.70 (ddd, J = 7.7, 6.9, 1.0 Hz, 1H), 7.76–7.79 (m, 2H), 8.13 (d, J_{AB} = 16.0 Hz, A part of AB system, 1H), 8.17 (d, J_{AB} = 16.0 Hz, B part of AB system, 1H), 8.18 (td, J = 6.5, 0.9 Hz, 1H), 8.43 (dt, J = 8.2, 1.0 Hz, 1H); ¹³C NMR δ 114.7, 116.0, 120.1, 126.1, 128.9, 129.0, 130.2, 131.4, 134.1, 146.3, 148.7, 163.8.

1-(Cyclohexylcarbonyl)-1*H*-1,2,3-benzotriazole (4l): white microcrystals (from chloroform/hexanes); mp 85–88 °C (lit.¹¹ mp 94–96 °C); yield, 85% (9.85 g); ¹H NMR δ 1.27–1.40 (m, 1H), 1.44–1.56 (m, 2H), 1.65–1.82 (m, 3H), 1.88–1.90 (m, 2H), 2.13–2.17 (m, 2H), 3.87–3.95 (m, 1H), 7.49 (t, J = 7.7 Hz, 1H), 7.63 (t, J = 7.7 Hz, 1H), 8.11 (d, J = 8.2 Hz, 1H), 8.28 (d, J = 8.2 Hz, 1H); ¹³C NMR δ 25.3, 25.6, 29.1, 43.1, 114.4, 119.9, 125.8, 130.1, 131.1, 146.0, 175.4.

General Procedure for the Preparation of Amides 5a–Ab. For amides **5a–v,y**, the *N*-acylbenzotriazole (10 mmol) and amine (10.5 mmol) in chloroform (10 mL) were exposed to microwave irradiation (80 W) for 10 min at a temperature of 80 °C. The reaction mixture was diluted with chloroform (20 mL). Aqueous work-up gave a residue that was purified by recrystallization (from chloroform or methanol) or column chromatography to obtain amides **5a–v,y**. For amides **5w–x**, the *N*-acylbenzotriazole (10 mmol) and amine (21.0 mmol) in chloroform (10 mL) were exposed to microwave irradiation (80 W) for 10 min at a temperature

of 80 °C. The resulting white solid was filtered off and recrystallized from methanol to obtain amides **5w–x**. For amide **5z**, the *N*-acylbenzotriazole (21 mmol) and amine (10 mmol) in chloroform (10 mL) were exposed to microwave irradiation (80 W) for 10 min at a temperature of 80 °C. The resulting white solid was filtered off and recrystallized from methanol to obtain amide **5z**. For amides **5Aa–Ab**, the *N*-acylbenzotriazole (12 mmol) and amine (10 mmol) in chloroform (10 mL) were exposed to microwave irradiation (80 W) for 10 min at a temperature of 80 °C. The reaction mixture was diluted with chloroform (20 mL). Aqueous work-up gave a residue that was purified by recrystallization (from methanol) or column chromatography to obtain amides **5Aa–Ab**.

***N*-Pyridin-2-ylbenzamide (5b):** purified by recrystallization from methanol, white microcrystals; mp 77–78 °C (lit.¹⁵ mp 82 °C); yield, 91% (1.80 g); ¹H NMR (DMSO-*d*₆) δ 5.93 (br s, 1H), 6.42–6.48 (m, 2H), 7.33–7.38 (m, 1H), 7.50 (td, *J* = 7.4, 0.6 Hz, 2H), 7.62 (td, *J* = 7.1, 0.9 Hz, 1H), 7.89 (dd, *J* = 4.1, 0.8 Hz, 1H), 7.96 (dd, *J* = 7.5, 0.8 Hz, 2H); ¹³C NMR (DMSO-*d*₆) δ 108.1, 111.8, 128.5, 129.3, 130.9, 132.8, 137.1, 147.4, 159.7, 167.5.

***N*-Benzyl-2-phenylacetamide (5c):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 115–117 °C (lit.¹⁶ mp 117–118 °C); yield, 90% (2.03 g); ¹H NMR δ 3.56 (s, 2H), 4.36 (d, *J* = 5.9 Hz, 2H), 6.07 (br s, 1H), 7.15–7.34 (m, 10H); ¹³C NMR δ 43.4, 43.6, 127.2, 127.3, 127.4, 128.5, 128.9, 129.3, 134.8, 138.1, 170.9.

***N*-(4-Methylphenyl)-2-phenylacetamide (5d):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 133–135 °C (lit.¹⁷ mp 135–137 °C); yield, 91% (2.05 g); ¹H NMR δ 2.27 (s, 3H), 3.68 (s, 2H), 7.06 (d, *J* = 8.1 Hz, 2H), 7.24–7.39 (m, 8H); ¹³C NMR δ 20.7, 44.5, 120.1, 127.4, 129.0, 129.3, 129.4, 134.0, 134.7, 135.1, 169.2.

***N*-(4-Methylphenyl)-3-phenylpropanamide (5e):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 127–129 °C (lit.¹⁸ mp 129–130 °C); yield, 92% (2.20 g); ¹H NMR δ 2.28 (s, 3H), 2.58 (t, *J* = 7.8 Hz, 2H), 3.01 (t, *J* = 7.6 Hz, 2H), 7.07 (d, *J* = 8.1 Hz, 2H), 7.18–7.36 (m, 8H); ¹³C NMR δ 20.8, 31.6, 39.3, 120.1, 126.3, 128.3, 128.6, 129.4, 133.9, 135.2, 140.7, 170.4.

4-Chloro-*N*-(4-methylphenyl)benzamide (5f): purified by recrystallization from methanol; yellow needles; mp 206–209 °C (lit.¹⁹ mp 213–215 °C); yield, 91% (2.24 g); ¹H NMR (DMSO-*d*₆) δ 2.27 (s, 3H), 7.15 (d, *J* = 8.1 Hz, 2H), 7.59 (d, *J* = 8.2 Hz, 2H), 7.64 (d, *J* = 8.1 Hz, 2H), 7.97 (d, *J* = 8.1 Hz, 2H), 10.24 (s, 1H); ¹³C NMR (DMSO-*d*₆) δ 20.5, 120.5, 128.4, 129.0, 129.5, 132.8, 136.4, 164.2.

4-Methoxy-*N*-phenylbenzamide (5g): purified by recrystallization from chloroform/hexanes; white microcrystals; mp 128–130 °C (lit.²⁰ mp 128–129 °C); yield, 89% (2.15 g); ¹H NMR δ 3.82 (s, 3H), 4.60 (d, *J* = 4.5 Hz, 2H), 6.57 (br s, 1H), 6.89 (d, *J* = 7.4 Hz, 2H), 7.25–7.33 (m, 5H), 7.75 (d, *J* = 8.8 Hz, 2H); ¹³C NMR δ 44.0, 55.3, 113.7, 126.6, 127.4, 127.8, 128.7, 128.8, 138.4, 162.1, 167.0.

***N*-Benzyl-4-nitrobenzamide (5h):** purified by recrystallization from chloroform/hexanes; pale yellow microcrystals; mp 136–137 °C (lit.²¹ mp 141–142 °C); yield, 96% (2.46 g); ¹H NMR δ 4.64 (d, *J* = 5.6 Hz, 2H), 6.68 (s, 1H), 7.26–7.36 (m, 5H), 7.94 (d, *J* = 8.6 Hz, 2H), 8.26 (d, *J* = 8.6 Hz, 2H); ¹³C NMR δ 44.4, 123.8, 127.9, 127.9, 128.2, 128.9, 137.4, 139.9, 149.5, 165.3.

***N*-(4-Methylphenyl)-2-furamide (5i):** purified by recrystallization from chloroform/hexanes; yellow needles; mp 108–110 °C (lit.²² mp 109–110 °C), yield, 96% (1.93 g); ¹H NMR δ 2.34 (s, 3H), 6.56 (dd, *J* = 3.4, 1.8 Hz, 1H), 7.17 (d, *J* = 8.2 Hz, 2H), 7.20 (d, *J* = 3.3 Hz, 1H), 7.50 (dd, *J*

= 1.6, 0.7 Hz, 1H), 7.54 (d, J = 8.4 Hz, 2H), 8.03 (br s, 1H); ^{13}C NMR δ 20.8, 112.4, 114.9, 119.9, 129.5, 134.0, 134.7, 144.0, 147.8, 156.0.

***N*-(4-Methylphenyl)heptanamide (5j):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 77–79 °C (lit.²³ mp 78–79 °C); yield, 93% (2.04 g); ^1H NMR δ 0.88 (t, J = 6.5 Hz, 3H), 1.31–1.37 (m, 6H), 1.65–1.72 (m, 2H), 2.30 (s, 3H), 2.31–2.35 (m, 2H), 7.09 (d, J = 8.2 Hz, 2H), 7.38 (d, J = 8.2 Hz, 2H), 7.40 (br s, 1H); ^{13}C NMR δ 14.0, 20.8, 22.5, 25.6, 28.9, 31.5, 37.7, 120.0, 129.4, 133.7, 135.4, 171.5.

***N*-(2-Furylmethyl)benzamide (5k):** purified by recrystallization from chloroform/hexanes; yellow microcrystals; mp 96–98 °C (lit.²⁴ mp 99–100 °C); yield, 90% (1.81 g); ^1H NMR δ 4.61 (d, J = 5.5 Hz, 2H), 6.26–6.33 (m, 2H), 6.73 (br s, 1H), 7.35–7.42 (m, 3H), 7.46–7.50 (m, 1H), 7.79 (dd, J = 7.1, 1.5 Hz, 2H); ^{13}C NMR δ 36.9, 107.6, 110.4, 127.0, 128.5, 131.5, 134.1, 142.2, 151.2, 167.2.

***N*-Cyclohexyl-2-furamide (5l):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 106–107 °C (lit.²² mp 110–111 °C); yield, 94% (1.81 g); ^1H NMR δ 1.18–1.31 (m, 3H), 1.35–1.47 (m, 2H), 1.62–1.78 (m, 3H), 1.97–2.01 (m, 2H), 3.89–3.96 (m, 1H), 6.21 (br s, 1H), 6.49 (dd, J = 3.6, 1.8 Hz, 1H), 7.09 (dd, J = 3.4, 0.8 Hz, 1H), 7.42 (d, J = 0.9 Hz, 1H); ^{13}C NMR δ 24.8, 25.5, 33.1, 47.8, 112.0, 113.8, 143.5, 148.3, 157.4.

4-Methyl-*N*-(4-methylphenyl)benzamide (5m): purified by recrystallization from chloroform/hexanes; white plates; mp 158–160 °C (lit.¹⁹ mp 158–160 °C); yield, 91% (2.05 g); ^1H NMR δ 2.31 (s, 3H), 2.38 (s, 3H), 7.12 (d, J = 8.2 Hz, 2H), 7.21 (d, J = 8.2 Hz, 2H), 7.51 (d, J = 8.7 Hz, 2H), 7.73 (d, J = 8.7 Hz, 2H), 8.03 (s, 1H); ^{13}C NMR δ 20.8, 21.4, 120.3, 127.0, 129.2, 129.4, 132.1, 133.9, 135.4, 142.0, 165.7.

N-Benzyl-3-phenylpropanamide (5n): purified by recrystallization from chloroform/hexanes; white microcrystals; mp 80–82 °C (lit.²⁵ mp 82–83 °C); yield, 93% (2.22 g); ¹H NMR δ 2.57 (d, *J* = 7.6 Hz, 2H), 3.04 (t, *J* = 7.5 Hz, 2H), 4.43 (d, *J* = 5.8 Hz, 2H), 5.99 (br s, 1H), 7.18–7.38 (m, 10H); ¹³C NMR δ 31.6, 38.3, 43.4, 126.2, 127.3, 127.6, 128.3, 128.5, 138.1, 140.7, 172.0.

N-(4-Methoxyphenyl)benzamide (5o): purified by recrystallization from chloroform/hexanes; white microcrystals; mp 156–157 °C (lit.²² mp 157–158 °C); yield, 88% (2.01 g); ¹H NMR δ 3.81 (s, 3H), 6.90 (d, *J* = 9.2 Hz, 2H), 7.44–7.56 (m, 5H), 7.84–7.87 (m, 3H); ¹³C NMR δ 55.5, 114.2, 122.1, 127.0, 128.7, 130.9, 131.7, 135.0, 156.6, 164.9.

4-Nitro-N-phenylbenzamide (5p): purified by recrystallization from methanol; yellow needles; mp 206–208 °C (lit.²⁶ mp 211 °C); yield; 95% (2.30 g); ¹H NMR (DMSO-*d*₆) δ 7.58 (t, *J* = 7.3 Hz, 2H), 7.66 (t, *J* = 7.2 Hz, 1H), 8.02 (dd, *J* = 7.0, 1.5 Hz, 2H), 8.11 (d, *J* = 9.2 Hz, 2H), 8.28 (d, *J* = 9.2 Hz, 2H), 10.8 (s, 1H); ¹³C NMR (DMSO-*d*₆) δ 119.8, 124.8, 128.0, 128.5, 132.2, 134.3, 142.4, 145.6, 166.3.

N-Butyl-4-methylbenzamide (5q): purified by recrystallization from chloroform/hexanes; white microcrystals; mp 52–54 °C (lit.²⁷ mp 55–57 °C); yield, 92% (1.76 g); ¹H NMR δ 0.93 (t, *J* = 7.3 Hz, 3H), 1.34–1.41 (m, 2H), 1.52–1.60 (m, 2H), 2.37 (s, 3H), 3.38–3.45 (m, 2H), 6.51 (br s, 1H), 7.19 (d, *J* = 8.1 Hz, 2H), 7.68 (d, *J* = 8.1 Hz, 2H); ¹³C NMR δ 13.7, 20.1, 21.3, 31.6, 39.6, 126.8, 129.0, 131.9, 141.5, 167.4.

N-(4-Methylphenyl)thiophene-2-carboxamide (5r): purified by recrystallization from chloroform/hexanes; white microcrystals; mp 103–105 °C (lit.²⁸ mp 104–105 °C); yield, 89% (1.97 g); ¹H NMR δ 2.34 (s, 3H), 7.12 (td, *J* = 4.4, 1.2 Hz, 1H), 7.17 (d, *J* = 8.2 Hz, 2H), 7.49 (d,

$J = 8.4$ Hz, 2H), 7.54 (dd, $J = 4.9, 1.2$ Hz, 1H), 7.61 (dd, $J = 3.7, 1.2$ Hz, 1H), 7.64 (br s, 1H); ^{13}C NMR δ 20.9, 120.3, 127.8, 128.3, 129.6, 130.6, 134.3, 135.0, 139.3, 159.9.

***N*-Benzylthiophene-2-carboxamide (5s):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 116–118 °C (lit.²⁹ mp 119–120 °C); yield, 92% (2.00 g); ^1H NMR (DMSO- d_6) 4.48 (d, $J = 5.9$ Hz, 2H), 7.16 (t, $J = 4.0$ Hz, 1H), 7.23–7.34 (m, 5H), 7.76 (d, $J = 4.9$ Hz, 1H), 7.85 (d, $J = 3.6$ Hz, 1H), 9.08 (t, $J = 5.8$ Hz, 1H); ^{13}C NMR δ (DMSO- d_6) 42.5, 126.8, 127.2, 127.9, 128.1, 128.3, 130.8, 139.5, 139.9, 161.1.

***N*-(2-Furanylmethyl)-2-thiophenecarboxamide (5t):** purified by recrystallization from chloroform/hexanes; yellow needles; mp 100–102 °C; yield, 92% (1.90 g); ^1H NMR δ 4.62 (d, $J = 5.5$ Hz, 2H), 6.30–6.35 (m, 3H), 7.07 (td, $J = 4.4, 1.1$ Hz, 1H), 7.29 (s, 1H), 7.38 (d, $J = 0.8$ Hz, 1H), 7.48 (dd, $J = 4.9, 1.0$ Hz, 1H); ^{13}C NMR δ 36.6, 107.4, 110.3, 127.5, 128.3, 130.1, 138.6, 141.9, 151.1, 161.9. Anal. Calcd for $\text{C}_{10}\text{H}_9\text{NO}_2\text{S}$: C, 57.95; H, 4.38; N, 6.76. Found: C, 57.93; H, 4.28; N, 6.84.

***N*-Benzyl-1*H*-indole-2-carboxamide (5u):** purified by recrystallization from methanol; white microcrystals; mp 224–226 °C (lit.²⁹ mp 230 °C); yield, 88% (2.20 g); ^1H NMR (DMSO- d_6) δ 4.52 (d, $J = 6.0$ Hz, 2H), 7.03 (t, $J = 7.4$ Hz, 1H), 7.15–7.28 (m, 3H), 7.34–7.35 (m, 4H), 7.43 (d, $J = 8.2$ Hz, 1H), 7.61 (d, $J = 8.0$ Hz, 1H), 9.05 (t, $J = 5.9$ Hz, 1H), 11.62 (s, 1H); ^{13}C NMR (DMSO- d_6) δ 42.1, 102.6, 112.3, 119.7, 121.5, 123.3, 126.8, 127.1, 127.2, 128.3, 131.6, 136.5, 139.6, 161.1.

***N*-Benzylbenzamide (5v):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 101–103 °C (lit.³⁰ mp 105–106 °C); yield, 92% (1.94 g); ^1H NMR δ 4.63 (d, J

= 5.8 Hz, 2H), 6.57 (br s, 1H), 7.25–7.51 (m, 8H), 7.79 (dd, $J = 7.7, 1.4$ Hz, 2H); ^{13}C NMR δ 44.1, 126.9, 127.5, 127.9, 128.5, 128.7, 131.5, 134.3, 138.2, 167.3.

***N,N'*-Dibenzylterephthalamide (5w):** purified by recrystallization from methanol; white microcrystals; mp 262–264 °C (lit.²¹ mp 264–266 °C); yield, 92% (3.16 g); ^1H NMR (DMSO- d_6) δ 0.91 (t, $J = 7.3$ Hz, 6H), 1.29–1.39 (m, 4H), 1.47–1.56 (m, 4H), 3.24–3.30 (m, 4H), 7.90 (s, 4H), 8.56 (s, 2H); ^{13}C NMR (DMSO- d_6) δ 13.7, 19.7, 31.2, 127.1, 136.8, 165.4.

***N,N'*-Dibutylterephthalamide (5x):** purified by recrystallization from methanol; white microcrystals; mp 230–232 °C (lit.³¹ mp 233–234 °C); yield, 96% (2.65 g); ^1H NMR (DMSO- d_6) δ 0.91 (t, $J = 7.3$ Hz, 6H), 1.29–1.39 (m, 4H), 1.47–1.56 (m, 4H), 3.24–3.30 (m, 4H), 7.90 (s, 4H), 8.56 (s, 2H); ^{13}C NMR (DMSO- d_6) δ 13.7, 19.7, 31.2, 39.2, 127.1, 136.8, 165.4.

***N*-(4-Methylphenyl)acetamide (5y):** purified by recrystallization from chloroform/hexanes; pale yellow microcrystals; mp 150–152 °C (lit.³² mp 153 °C); yield, 94 % (1.40 g); ^1H NMR δ 2.14 (s, 3H), 2.30 (s, 3H), 7.10 (d, $J = 8.1$ Hz, 2H), 7.37 (d, $J = 8.4$ Hz, 2H), 7.57 (br s, 1H); ^{13}C NMR δ 20.8, 24.4, 120.1, 129.4, 133.9, 135.3, 168.5.

***N,N'*-Dibenzoylthylenediamine (5z):** purified by recrystallization from methanol; white microcrystals; mp 250–252 °C (lit.³³ mp 254 °C); yield, 94% (2.52 g); ^1H NMR (DMSO- d_6) δ 3.45–3.46 (m, 4H), 7.44 - 7.55 (m, 6H), 7.85 (d, $J = 7.0$ Hz, 4H), 8.62 (s, 2H); ^{13}C NMR (DMSO- d_6) δ 40.1, 127.2, 128.3, 131.1, 134.5, 166.6.

***N*-(4-Methylphenyl)-3-phenyl-2-propenamide (5Aa):** purified by recrystallization from methanol; white microcrystals; mp 156–158 °C (lit.³⁴ mp 159 °C); yield, 87% (2.06 g); ^1H NMR (DMSO- d_6) δ 2.27 (s, 3H), 6.84 (d, $J = 15.7$ Hz, 1H), 7.15 (d, $J = 8.4$ Hz, 2H), 7.41–7.48 (m,

4H), 7.56–7.61 (m, 3H), 7.62 (d, $J = 15.6$ Hz, 1H), 7.92 (br s, 1H); ^{13}C NMR (DMSO- d_6) δ 20.5, 119.2, 122.4, 127.7, 129.0, 129.2, 129.7, 132.3, 134.8, 136.8, 139.9, 163.3.

3-Phenyl-*N*-(phenylmethyl)-2-propenamide (5Ab): purified by column chromatography on basic alumina with ethyl acetate/hexanes (5:1) as eluent; white microcrystals (from chloroform/hexanes); mp 107–109 °C (lit.³⁵ mp 107–110 °C); yield, 92% (2.18 g); ^1H NMR δ 4.58 (d, $J = 5.8$ Hz, 2H), 5.95 (br s, 1H), 6.42 (d, $J = 15.5$ Hz, 1H), 7.29–7.37 (m, 8H), 7.48–7.51 (m, 2H), 7.68 (d, $J = 15.5$ Hz, 1H); ^{13}C NMR δ 43.9, 120.3, 127.6, 127.8, 127.9, 128.8, 128.8, 129.7, 134.7, 138.1, 141.5, 165.7.

General Procedure for the Preparation of Imidoylbenzotriazoles 6a–w. (a) For amides with R^1 =aliphatic group: to a solution of amide (5.0 mmol) in methylene chloride (20 mL), pyridine (0.45 mL, 5.5 mmol) followed by oxalyl chloride (0.48 mL, 5.5 mmol) in methylene chloride (20 mL) was added dropwise at 0 °C. Gas evolution was observed during the process. After the addition, the reaction was continued for 15 min, then benzotriazole (1.25 g, 10.5 mmol) was added in one portion to the reaction flask. The ice bath was removed to allow the reaction to continue at room temperature and the reaction was monitored by TLC. The precipitated white solid was filtered off and sodium bicarbonate solution (saturated) was added to dilute the reaction mixture. Aqueous work-up gave a crude product which was purified by column chromatography on basic alumina using hexanes/EtOAc (8:1 to 5:1) as eluent. (b) For amides with R^1 =aryl group: Thionyl chloride (0.77 mL, 10.5 mmol) and benzotriazole (2.44 g, 20.5 mmol) were dissolved in chloroform (10 mL) in a 50 mL round-bottomed flask, and then amide (5 mmol) was added to the flask. The reaction mixture was exposed to microwave irradiation for 10 min at 80 °C and 80 W. The precipitated solid was filtered off and aqueous workup gave a crude product which was purified by either recrystallization from chloroform/hexanes or column chromatography on basic alumina using hexanes/EtOAc (8:1 to 5:1) as eluent.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(4-methylphenyl)methylidene]-4-methylaniline (6c):**

purified by recrystallization from chloroform/hexanes; yellow needles; mp 138–140 °C; yield, 82% (1.34 g); ¹H NMR δ 2.29 (s, 3H), 2.37 (s, 3H), 6.76 (d, *J* = 8.3 Hz, 2H), 7.04 (d, *J* = 8.1 Hz, 2H), 7.17 (d, *J* = 8.0 Hz, 2H), 7.25 (d, *J* = 8.1 Hz, 2H), 7.49 (t, *J* = 8.0 Hz, 1H), 7.61 (t, *J* = 7.7 Hz, 1H), 8.15 (d, *J* = 8.2 Hz, 1H), 8.44 (d, *J* = 8.2 Hz, 1H); ¹³C NMR δ 20.9, 21.6, 115.3, 119.9, 121.5, 125.4, 127.5, 129.0, 129.1, 129.4, 130.2, 132.1, 133.7, 140.7, 144.5, 146.4, 153.5. Anal. Calcd for C₂₁H₁₈N₄: C, 77.27; H, 5.57; N, 17.17. Found: C, 77.33; H, 5.59; N, 17.29.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(2-furyl)methylidene]-4-methylaniline (6d):** purified by recrystallization from chloroform/hexanes; yellow microcrystals; mp 120–123 °C; yield, 84% (1.27 g); ¹H NMR δ 2.38 (s, 3H), 6.72–6.74 (m, 1H), 7.21 (d, *J* = 8.2 Hz, 2H), 7.54 (t, *J* = 7.3 Hz, 1H), 7.70 (t, *J* = 7.7 Hz, 1H), 7.78 (d, *J* = 8.3 Hz, 2H), 7.88 (s, 1H), 8.16 (d, *J* = 6.2 Hz, 1H), 8.18 (s, 1H), 8.42 (d, *J* = 8.4 Hz, 1H); ¹³C NMR δ 21.7, 113.0, 114.7, 120.2, 124.8, 126.3, 127.2, 129.8, 130.5, 132.1, 140.7, 141.4, 144.5, 145.5, 148.9, 155.0. Anal. Calcd for C₁₈H₁₄N₄O: C, 71.51; H, 4.68; N, 18.54. Found: C, 71.13; H, 4.49; N, 18.65.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(phenyl)methylidene]-4-methoxyaniline (6e):** purified by recrystallization from chloroform/hexanes; yellow plates; mp 100–103 °C (lit.^{4c} mp 104–105 °C); yield, 82% (1.34 g); ¹H NMR δ 3.76 (s, 3H), 6.77–6.82 (m, 4H), 7.35–7.55 (m, 6H), 7.60–7.67 (m, 1H), 8.15 (d, *J* = 8.2 Hz, 1H), 8.47 (d, *J* = 8.4 Hz, 1H); ¹³C NMR δ 55.3, 114.1, 115.3, 119.9, 123.1, 125.4, 128.4, 129.1, 130.1, 130.3, 132.0, 139.8, 146.4, 153.1, 156.6.

1-[1-[(4-Methylphenyl)imino]ethyl]-1*H*-benzotriazole (6f): purified by column chromatography on basic alumina; white microcrystals (from chloroform/hexanes); mp 113–115 °C (lit.^{4a} mp 115 °C); yield, 65% (0.81 g); ¹H NMR δ 2.39 (s, 3H), 2.75 (s, 3H), 6.86 (d, *J* = 8.1 Hz, 2H), 7.23 (d, *J* = 8.1 Hz, 2H), 7.44–7.50 (m, 1H), 7.56–7.62 (m, 1H), 8.12 (d, *J* = 8.2 Hz,

1H), 8.54 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 16.1, 20.8, 115.7, 119.6, 120.2, 125.2, 129.1, 129.7, 131.3, 133.8, 144.7, 146.5, 153.9.

***N*-[1-(Benzotriazole-1-yl)-2-phenylethylidene]-4-methylaniline (6g):** purified by column chromatography on basic alumina; pale yellow microcrystals (from chloroform/hexanes); mp 124–126 °C (lit.^{4d} mp 123–125 °C); yield, 62% (1.01 g); ^1H NMR δ 2.38 (s, 3H), 4.61 (s, 2H), 6.88 (d, $J = 8.1$ Hz, 2H), 7.14–7.22 (m, 7H), 7.45 (t, $J = 7.3$ Hz, 1H), 7.58 (t, $J = 7.1$ Hz, 1H), 8.09 (d, $J = 8.2$ Hz, 1H), 8.53 (d, $J = 8.3$ Hz, 1H); ^{13}C NMR δ 21.0, 34.8, 115.7, 119.8, 120.2, 125.5, 126.8, 128.6, 128.8, 129.3, 129.9, 131.6, 134.1, 135.4, 144.5, 146.6, 154.7.

***N*-[(1*E*)-1-(1*H*-1,2,3-Benzotriazol-1-yl)-2-phenylethylidene]-*N*-benzylamine (6h):**^{4d} purified by column chromatography on basic alumina; colorless oil; yield, 56% (0.92 g); ^1H NMR δ 4.76 (s, 2H), 4.95 (s, 2H), 7.16–7.46 (m, 11H), 7.51 (t, $J = 7.6$ Hz, 1H), 8.06 (d, $J = 8.2$ Hz, 1H), 8.51 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 33.9, 53.8, 115.7, 119.6, 125.1, 126.9, 127.0, 127.5, 128.4, 128.5, 128.8, 128.9, 131.5, 134.4, 139.2, 146.6, 155.4.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(phenyl)methylene]-*N*-benzylamine (6i):** purified by column chromatography on basic alumina; colorless needles (from chloroform/hexanes); mp 108–110 °C (lit.^{4b} mp 108 °C); yield, 93% (1.45 g); ^1H NMR δ 4.76 (s, 2H), 7.36–7.39 (m, 4H), 7.43–7.48 (m, 4H), 7.54–7.61 (m, 4H), 8.11 (d, $J = 8.2$ Hz, 1H), 8.52 (d, $J = 8.2$ Hz, 1H); ^{13}C NMR δ 55.3, 115.4, 119.8, 125.2, 126.9, 127.5, 127.7, 128.5, 128.7, 129.0, 130.3, 130.5, 131.8, 139.5, 146.3, 155.7.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(4-chlorophenyl)methylene]-*N*-(4-methylphenyl)amine (6j):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 118–120 °C; yield, 90% (1.56 g); ^1H NMR δ 2.29 (s, 3H), 6.73 (d, $J = 8.1$ Hz, 2H), 7.05 (d, $J = 8.1$ Hz, 2H),

7.29–7.37 (m, 4H), 7.51 (t, $J = 7.3$ Hz, 1H), 7.63 (t, $J = 7.7$ Hz, 1H), 8.15 (d, $J = 8.2$ Hz, 1H), 8.48 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 20.9, 115.3, 120.0, 121.4, 125.7, 128.6, 128.8, 129.3, 129.6, 131.6, 131.9, 134.1, 136.5, 143.9, 146.4, 152.3. Anal. Calcd for $\text{C}_{20}\text{H}_{15}\text{ClN}_4$: C, 69.26; H, 4.37; N, 16.16. Found: C, 69.34; H, 4.25; N, 16.13.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(4-methoxyphenyl)methylene]-*N*-benzylamine (6k):** purified by column chromatography on basic alumina; white microcrystals (from chloroform/hexanes); mp 115–118 °C; yield, 78% (1.34 g); ^1H NMR (mixture of two isomers) δ 3.80 (s, 0.7H), 3.88 (s, 3H), 4.56 (s, 0.5H), 4.82 (s, 2H), 6.86 (d, $J = 8.9$ Hz, 0.5H), 7.06 (d, $J = 8.8$ Hz, 2H), 7.23–7.47 (m, 10H), 7.56 (t, $J = 7.6$ Hz, 1H), 8.10 (d, $J = 8.2$ Hz, 1H), 8.47 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 55.3, 110.5, 114.0, 115.3, 119.7, 120.2, 122.4, 124.6, 125.1, 126.3, 126.9, 127.4, 127.7, 128.4, 128.5, 128.9, 130.1, 130.4, 131.9, 138.8, 139.7, 146.3, 155.5, 160.9. Anal. Calcd for $\text{C}_{21}\text{H}_{18}\text{N}_4\text{O}$: C, 73.66; H, 5.31; N, 16.37. Found: C, 73.66; H, 5.32; N, 16.54.

***N*-[1-(1*H*-1,2,3-benzotriazol-1-yl)heptylidene]-*N*-(2-furylmethyl)amine (6l):** purified by column chromatography on basic alumina; colorless oil; yield, 84% (1.27 g); ^1H NMR δ 4.72 (s, 2H), 6.27–6.28 (m, 1H), 6.36–6.38 (m, 1H), 7.34–7.51 (m, 4H), 7.56–7.62 (m, 4H), 8.10 (d, $J = 8.2$ Hz, 1H), 8.48 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 48.7, 106.7, 110.3, 110.6, 115.4, 119.8, 125.3, 128.6, 128.8, 129.1, 130.1, 130.4, 131.8, 142.0, 146.3, 152.6. Anal. Calcd for $\text{C}_{18}\text{H}_{14}\text{N}_4\text{O}$: C, 71.51; H, 4.68; N, 18.54. Found: C, 71.15; H, 4.74; N, 18.55.

***N*-[1-(1*H*-1,2,3-benzotriazol-1-yl)heptylidene]-*N*-(4-methylphenyl)amine (6m):** purified by column chromatography on basic alumina; colorless oil; yield, 57% (0.91 g); ^1H NMR δ 0.82 (t, $J = 6.8$ Hz, 3H), 1.16–1.36 (m, 6H), 1.71–1.81 (m, 2H), 2.38 (s, 3H), 3.10–3.15 (m, 2H), 6.83 (d, $J = 8.1$ Hz, 2H), 7.21 (d, $J = 8.0$ Hz, 2H), 7.45 (dd, $J = 8.2, 0.8$ Hz, 1H), 7.56 (dd, $J = 7.8, 0.9$ Hz, 1H), 8.11 (d, $J = 8.2$ Hz, 1H), 8.51 (d, $J = 8.2$ Hz, 1H); ^{13}C NMR δ (1 signal is hidden) 13.9,

20.8, 22.3, 27.8, 29.1, 31.1, 115.7, 119.6, 119.8, 125.2, 129.0, 129.7, 131.4, 133.5, 144.8, 146.4, 157.6. Anal. Calcd for $C_{20}H_{24}N_4$: C, 74.97; H, 7.57; N, 17.49. Found: C, 74.80; H, 7.67; N, 17.65.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(2-furyl)methylene]-*N*-cyclohexylamine (6n):** purified by column chromatography on basic alumina; colorless oil; yield, 95% (1.40 g); 1H NMR δ 1.00–1.96 (m, 15H), 2.98–3.06 (m, 0.5H), 3.94–4.04 (m, 1H), 6.18 (d, J = 3.4 Hz, 0.5 H), 6.40–6.42 (m, 0.5H), 6.60–6.62 (m, 1H), 6.88 (d, J = 3.6 Hz, 1H), 7.35–7.60 (m, 4H), 7.66 (d, J = 1.0 Hz, 1H), 8.09 (dd, J = 8.2, 0.6 Hz, 1H), 8.16–8.21 (m, 1.5H); ^{13}C NMR δ 24.0, 24.1, 25.0, 25.4, 33.6, 34.2, 59.5, 60.0, 109.7, 111.1, 111.8, 114.1, 115.7, 116.7, 119.5, 120.0, 124.5, 124.8, 128.4, 128.6, 132.2, 132.9, 136.3, 141.5, 142.7, 144.5, 145.9, 146.0, 147.7. Anal. Calcd for $C_{17}H_{18}N_4O$: C, 69.35; H, 6.18; N, 19.04. Found: C, 69.40; H, 6.30; N, 18.99.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(4-nitrophenyl)methylene]-*N*-benzylamine (6o):** purified by column chromatography on basic alumina; pale yellow microcrystals (from chloroform/hexanes); mp 117–119 °C; yield, 86% (1.54 g); 1H NMR δ 4.73 (s, 2H), 7.30–8.57 (m, 13H); ^{13}C NMR δ 55.2, 115.3, 119.9, 124.0, 125.7, 127.2, 127.3, 128.7, 129.5, 129.8, 131.5, 136.5, 138.7, 146.3, 148.7, 153.4. Anal. Calcd for $C_{20}H_{15}N_5O_2$: C, 67.21; H, 4.24; N, 19.60. Found: C, 67.32; H, 4.15; N, 19.37.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(4-nitrophenyl)methylene]-*N*-phenylamine (6p):** purified by recrystallization from chloroform/hexanes; pale yellow microcrystals; mp 183–185 °C; yield, 88% (1.50 g); 1H NMR δ 6.95 (d, J = 8.6 Hz, 2H), 7.38–7.57 (m, 6H), 7.68 (t, J = 7.4 Hz, 1H), 8.11 (d, J = 8.5 Hz, 2H), 8.17 (d, J = 8.2 Hz, 1H), 8.43 (d, J = 7.8 Hz, 1H); ^{13}C NMR δ 115.2, 119.6, 120.3, 121.9, 124.9, 126.1, 128.6, 129.8, 130.1, 131.1, 131.9, 144.1, 146.5, 153.2, 155.1. Anal. Calcd for $C_{19}H_{13}N_5O_2$: C, 66.47; H, 3.82; N, 20.40. Found: C, 66.16; H, 3.70; N, 20.16.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(4-methylphenyl)methylene]-*N*-butylamine (6q):** purified by column chromatography on basic alumina; colorless oil; yield, 84% (1.23 g); ^1H NMR (mixture of two isomers) δ 0.85 (t, J = 7.4 Hz, 0.7H), 0.94 (t, J = 7.4 Hz, 3H), 1.31–1.40 (m, 0.5H), 1.42–1.52 (m, 2H), 1.60–1.67 (m, 0.5H), 1.70–1.79 (m, 2H), 2.36 (s, 0.9H), 2.44 (s, 3H), 3.34 (t, J = 7.0 Hz, 0.5H), 3.57 (t, J = 6.9 Hz, 2H), 7.14–7.19 (m, 0.9H), 7.28–7.39 (m, 5H), 7.42–7.47 (m, 1.5H), 7.59 (td, J = 7.7, 0.7 Hz, 1H), 8.16–8.19 (m, 0.2H), 8.45 (d, J = 8.4 Hz, 1H); ^{13}C NMR δ 13.7, 13.8, 20.4, 20.5, 21.4, 21.5, 32.7, 33.3, 51.2, 51.7, 109.2, 110.3, 115.2, 119.6, 120.0, 124.4, 125.0, 127.7, 128.0, 128.4, 128.5, 128.7, 129.2, 129.3, 131.2, 131.9, 140.2, 142.1, 144.7, 146.2, 146.6, 154.5. Anal. Calcd for $\text{C}_{18}\text{H}_{20}\text{N}_4$: C, 73.93; H, 6.91; N, 19.17. Found: C, 74.17; H, 7.07; N, 19.38.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(thien-2-yl)methylene]-*N*-(4-methylphenyl)amine (6r):** purified by recrystallization from chloroform/hexanes; yellow needles; mp 133–135 °C; yield, 91% (1.43 g); ^1H NMR δ (mixture of two isomers) 2.12 (s, 1.8H), 2.35 (s, 3H), 6.58 (d, J = 8.1 Hz, 1.2H), 6.83 (d, J = 8.1 Hz, 3.3H), 7.00–7.16 (m, 5H), 7.29–7.33 (m, 2.2H), 7.46–7.53 (m, 2H), 7.57–7.62 (m, 1.7H), 8.03–8.06 (m, 0.6H), 8.16 (d, J = 8.2 Hz, 1H), 8.32 (d, J = 8.2 Hz, 1H); ^{13}C NMR δ 20.8, 20.9, 110.5, 114.7, 119.9, 120.3, 121.0, 124.4, 125.4, 126.5, 128.0, 128.4, 129.1, 129.4, 129.8, 131.3, 131.9, 132.3, 132.6, 134.1, 134.6, 135.4, 139.5, 141.2, 143.2, 144.7, 144.9, 146.3, 146.7. Anal. Calcd for $\text{C}_{18}\text{H}_{14}\text{N}_4\text{S}$: C, 67.90; H, 4.44; N, 17.60. Found: C, 67.65; H, 4.35; N, 17.73.

***N*-[1-(1*H*-1,2,3-Benzotriazol-1-yl)-3-phenylpropylidene]-*N*-benzylamine (6s):** purified by column chromatography on basic alumina; white needles (from chloroform/hexanes); mp 77–79 °C; yield, 57 % (0.97 g); ^1H NMR δ (mixture of two isomers) 3.08 (t, J = 7.8 Hz, 2H), 3.23 (t, J = 7.6 Hz, 0.3H), 3.60 (t, J = 7.8 Hz, 2H), 3.76 (t, J = 7.7 Hz, 0.3H), 4.62 (s, 2H), 7.19–7.38 (m,

11H), 7.40–7.49 (m, 1.3H), 7.51–7.54 (m, 1.3H), 7.63 (t, $J = 7.3$ Hz, 0.3H), 8.11 (d, $J = 8.2$ Hz, 1H), 8.29 (d, $J = 8.2$ Hz, 0.3H), 8.45 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 30.3, 32.9, 53.4, 115.7, 119.5, 125.1, 126.6, 126.9, 127.5, 128.4, 128.5, 128.6, 129.0, 131.4, 139.4, 140.0, 146.5, 156.7. Anal. Calcd for $\text{C}_{22}\text{H}_{20}\text{N}_4$: C, 77.62; H, 5.93; N, 16.46. Found: C, 77.41; H, 5.97; N, 16.48.

***N*-[1-(1*H*-1,2,3-Benzotriazol-1-yl)-3-phenylpropylidene]-*N*-(4-methylphenyl)amine (6t):** purified by column chromatography on basic alumina; white needles (from chloroform/hexanes); mp 116–118 °C; yield, 64 % (2.10 g); ^1H NMR δ 2.37 (s, 3H), 3.09 (t, $J = 8.4$ Hz, 2H), 3.42 (t, $J = 7.8$ Hz, 2H), 6.64 (d, $J = 7.8$ Hz, 2H), 7.03 (d, $J = 6.0$ Hz, 2H), 7.14–7.22 (m, 5H), 7.48 (t, $J = 7.1$ Hz, 1H), 7.59 (t, $J = 8.4$ Hz, 1H), 8.14 (d, $J = 8.2$ Hz, 1H), 8.48 (d, $J = 8.1$ Hz, 1H); ^{13}C NMR δ 20.8, 31.4, 33.8, 115.6, 119.6, 119.7, 125.4, 126.4, 128.4, 128.5, 129.2, 129.7, 131.4, 133.6, 140.0, 144.5, 146.5, 156.2. Anal. Calcd for $\text{C}_{22}\text{H}_{20}\text{N}_4$: C, 77.62; H, 5.93; N, 16.46. Found: C, 77.36; H, 5.94; N, 16.35.

***N*-[1*H*-1,2,3-Benzotriazol-1-yl(2-thienyl)methylidene](phenyl)methanamine (6u):** purified by column chromatography on basic alumina; yellow oil; yield, 83% (2.64 g); ^1H NMR (mixture of two isomers) δ 4.52 (s, 1.8H), 4.95 (s, 2H), 6.73 (s, $J = 3.7$ Hz, 0.9H), 6.96 (t, $J = 4.4$ Hz, 0.9H), 7.19–7.59 (m, 18.3H), 7.66 (d, $J = 5.1$ Hz, 1H), 8.11 (d, $J = 8.2$ Hz, 1H), 8.19 (d, $J = 7.7$ Hz, 0.9H), 8.36 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 55.1, 55.9, 110.3, 115.0, 119.8, 120.3, 124.7, 125.2, 126.9, 127.00, 127.04, 127.5, 127.6, 128.4, 128.6, 128.8, 129.0, 129.5, 131.0, 131.6, 131.7, 132.0, 132.7, 138.2, 139.1, 139.3, 143.0, 144.8, 146.4, 149.0. Anal. Calcd for $\text{C}_{18}\text{H}_{14}\text{N}_4\text{S}$: C, 67.90; H, 4.43; N, 17.60. Found: C, 67.97; H, 4.40; N, 17.51.

***N*-[1*H*-1,2,3-benzotriazol-1-yl(2-thienyl)methylidene](2-furyl)methanamine (6v):** purified by column chromatography on basic alumina; orange oil; yield, 81% (2.50 g); ^1H NMR (mixture of two isomers) δ 4.51 (s, 1.5 H), 4.91 (s, 2.2 H), 6.19 (d, $J = 3.3$ Hz, 0.8 H), 6.27–6.32 (m, 1.9

H), 6.38–6.39 (m, 1.9 H), 6.74 (d, $J = 3.7$ Hz, 0.7 H), 6.94–6.97 (m, 0.8 H), 7.18–7.26 (m, 1.9 H), 7.31 (d, $J = 1.0$ Hz, 0.8 H), 7.37–7.60 (m, 7.6 H), 7.68 (dd, $J = 5.1, 0.8$ Hz, 1.2 H), 7.77 (td, $J = 5.4$ Hz, 1.0 Hz, 0.8 H), 8.10 (d, $J = 8.2$ Hz, 1.1 H), 8.19 (d, $J = 8.1$ Hz, 0.7H), 8.33 (d, $J = 8.2$ Hz, 1H); ^{13}C NMR δ 48.5, 49.3, 106.9, 107.3, 110.3, 110.4, 115.0, 119.8, 120.3, 124.8, 125.3, 126.9, 127.7, 128.2, 128.5, 128.8, 129.0, 129.8, 131.3, 131.9, 132.0, 132.7, 135.1, 142.0, 142.1, 143.9, 144.8, 146.3, 149.9, 151.2, 152.4. Anal. Calcd for $\text{C}_{16}\text{H}_{12}\text{N}_4\text{OS}$: C, 62.32; H, 3.92; N, 18.17. Found: C, 61.96; H, 3.83; N, 17.78.

***N*–[(*E*,2*E*)-1-(1*H*-1,2,3-benzotriazol-1-yl)-3-phenyl-2-propenylidene]-4-methylaniline**

(6w): purified by column chromatography on basic alumina; yellow oil; yield, 65% (1.10 g); ^1H NMR δ 2.41 (s, 3H), 6.99 (d, $J = 8.2$ Hz, 2H), 7.09 (d, $J = 16.6$ Hz, 1H), 7.24 (d, $J = 8.1$ Hz, 2H), 7.34–7.40 (m, 5H), 7.40 (d, $J = 16.3$ Hz, 1H), 7.43 (m, 6H), 7.49 (t, $J = 7.6$ Hz, 1H), 7.60 (t, $J = 7.6$ Hz, 1H), 8.16 (d, $J = 8.2$ Hz, 1H), 8.29 (d, $J = 8.4$ Hz, 1H); ^{13}C NMR δ 21.0, 105.3, 114.6, 115.2, 119.8, 120.9, 125.3, 127.8, 128.8, 129.0, 129.8, 130.1, 134.4, 135.1, 144.2, 144.7, 146.1, 151.1. Anal. Calcd for $\text{C}_{22}\text{H}_{18}\text{N}_4$: C, 78.08; H, 5.36; N, 16.56. Found: C, 77.69; H, 5.42; N, 16.59.

General Procedure for the Preparation of Amidines (7a–Aa). (i) Imidoylbenzotriazole (0.5 mmol), a primary or secondary amine (1 mmol), and acetic acid (1 mL) were placed in a 10 mL microwave reaction tube. The tube containing the reaction mixture was sealed with an aluminum crimp cap fitted with a silicon septum and then exposed to microwave irradiation (120 W) for 10 min at 120 °C. The reaction mixture was diluted with chloroform (20 mL). Aqueous work-up gave a residue that was purified by column chromatography on basic alumina using methylene chloride/methanol (20:1 to 10:1). (ii) Imidoylbenzotriazole (0.5 mmol) was dissolved in chloroform (1 mL) in a 10 mL microwave reaction tube. Aluminum chloride (catalytic amount)

was added to the tube followed by amine (1 mmol). The tube containing the reaction mixture was sealed with an aluminum crimp cap fitted with a silicon septum and then exposed to microwave irradiation (80 W) for 10 min at 80 °C. The reaction mixture was diluted with chloroform (20 mL). Aqueous work-up gave a residue that was purified by recrystallization from chloroform or column chromatography on basic alumina using hexanes/EtOAc (10:1 to 5:1) as eluent. (iii) Imidoylbenzotriazole (0.5 mmol), amine hydrochloride (1.0 mmol) and chloroform (1 mL) were placed in a 10 mL microwave reaction tube. The tube containing the reaction mixture was sealed with an aluminum crimp cap fitted with a silicon septum and then exposed to microwave irradiation (80 W) for 10 min at 80 °C. The reaction mixture was diluted with chloroform (20 mL) and the insoluble portion was filtered off. Aqueous work-up gave a residue that was purified by recrystallization from chloroform or column chromatography on basic alumina using hexanes/EtOAc (10:1 to 5:1) as eluent.

***N*-[*(1E)*-1-morpholin-4-ylethylidene]aniline (**7a**):** purified by column chromatography on basic alumina; colorless needles (from chloroform/hexanes); mp 80–82 °C (lit.³⁷ mp 82 °C); yield, 76% (0.080 g); ¹H NMR δ 1.85 (s, 3H), 3.48 – 3.51 (m, 4H), 3.74 – 3.77 (m, 4H), 6.70 (dd, *J* = 7.3, 0.8 Hz, 2H), 6.96 (t, *J* = 7.4 Hz, 1H), 7.24 (t, *J* = 7.6 Hz, 2H); ¹³C NMR δ 14.5, 45.2, 66.7, 121.7, 121.9, 128.8, 151.5, 156.9.

***(1E)*-*N,N*-Diethyl-*N'*-phenylethanimidamide acetate (**7b**):** purified by column chromatography on basic alumina; colorless needles (from chloroform/hexanes); mp 102–104 °C; yield, 88% (0.110 g); ¹H NMR δ 1.11 (t, *J* = 7.0 Hz, 3H), 1.18 (t, *J* = 7.1 Hz, 3H), 2.08 (s, 3H), 2.14 (s, 3H), 3.26–3.41 (m, 4H), 7.06 (t, *J* = 7.4 Hz, 1H), 7.29 (t, *J* = 3.8 Hz, 2H), 7.55 (d, *J* = 7.7 Hz, 1H), 8.71 (s, 1H); ¹³C NMR δ 12.9, 14.0, 21.3, 24.2, 39.9, 42.8, 119.9, 123.8, 128.6,

138.3, 169.0, 169.7. Anal. Calcd for $C_{14}H_{22}N_2O_2$: C, 67.17; H, 8.86; N, 11.19. Found: C, 67.18; H, 8.89; N, 10.92.

(1E)-N-Benzyl-N'-phenylethanimidamide acetate (7c): purified by column chromatography on basic alumina; colorless needles (from chloroform/hexanes); mp 88–90 °C; yield, 77% (0.110 g); 1H NMR δ 1.97 (s, 3H), 2.08 (s, 3H), 4.37 (d, J = 5.8 Hz, 2H), 6.47 (s, 1H), 7.06 (t, J = 7.3 Hz, 1H), 7.25–7.33 (m, 7 H), 7.52 (d, J = 8.0 Hz, 2H), 8.51 (br s, 1H); ^{13}C NMR δ 23.0, 24.2, 43.5, 119.9, 123.9, 127.3, 127.6, 128.5, 28.7, 138.1, 138.2, 168.9, 170.2. Anal. Calcd for $C_{17}H_{20}N_2O_2$: C, 71.79; H, 7.10; N, 9.85. Found: C, 71.50; H, 7.19; N, 9.85.

(1E)-N-(4-Methylphenyl)-N'-phenylethanimidamide (7d): purified by column chromatography on basic alumina; white microcrystals (from chloroform/hexanes); mp 87–89 °C (lit.³⁸ mp 90 °C); yield, 89% (0.100 g); 1H NMR (DMSO- d_6) δ 1.88 (s, 3H), 2.24 (s, 3H), 6.64–6.74 (m, 2H), 6.92 (t, J = 7.3 Hz, 1H), 7.05 (d, J = 8.1 Hz, 2H), 7.24 (t, J = 7.7 Hz, 2H), 7.65–7.76 (m, 2H), 8.82 (br s, 1H); ^{13}C NMR (DMSO- d_6) δ 17.7, 20.4, 119.0, 121.3, 128.6, 130.0, 153.0.

N-[(1E)-Morpholin-4-yl(phenyl)methylene]aniline (7f): purified by recrystallization from chloroform/hexanes; colorless needles; mp 85–87 °C (lit.³⁷ mp 85 °C); yield, 90 % (0.120 g); 1H NMR δ 3.42–3.45 (m, 4H), 3.75 (m, 4H), 6.56 (d, J = 7.4 Hz, 2H), 6.75 (t, J = 7.3 Hz, 1H), 7.00 (t, J = 8.0 Hz, 2H), 7.10–7.13 (m, 2H), 7.21–7.24 (m, 3H); ^{13}C NMR δ 46.6, 66.8, 121.2, 122.6, 128.1, 128.7, 129.0, 133.0, 150.7, 160.6.

N-(4-Methylphenyl)-N'-phenylbenzenecarboximidamide (7g): purified by recrystallization from chloroform/hexanes; pale yellow microcrystals; mp 130–132 °C (lit.⁴⁰ mp 133–135 °C);

yield, 91% (0.130 g); ^1H NMR δ 2.26 (s, 3H), 6.40–7.62 (m, 14H); ^{13}C NMR δ 20.8, 121.4, 122.0, 123.4, 128.3, 128.6, 129.2, 131.7, 135.1, 154.3.

***N*-[*(1Z)*-(4-Methoxyphenyl)(morpholin-4-yl)methylene]-1-phenylmethanamine (7i):** purified by column chromatography on basic alumina; colorless oil; yield, 88% (0.140 g); ^1H NMR δ 3.20 (s, 4H), 3.59 (t, J = 4.8 Hz, 4H), 3.74 (s, 3H), 4.21 (s, 2H), 6.85 (d, J = 8.8 Hz, 2H), 7.01–7.21 (m, 7H); ^{13}C NMR δ 46.6, 54.3, 55.2, 66.8, 114.0, 125.6, 125.9, 127.0, 128.0, 129.2, 142.4, 169.7, 162.7. Anal. Calcd for $\text{C}_{19}\text{H}_{22}\text{N}_2\text{O}_2$: C, 73.52; H, 7.14; N, 9.02. Found: C, 73.12; H, 7.13; N, 8.69.

***(1Z)*-*N,N'*-Bis(4-methylphenyl)-2-phenylethanimidamide (7j):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 89–91 °C; yield, 94 % (0.150 g); ^1H NMR δ 2.29 (s, 6H), 3.66 (s, 2H), 7.06–7.34 (m, 13H); ^{13}C NMR δ (4 signals are hidden) 20.8, 37.7, 120.6, 127.1, 129.1, 129.4, 131.8. Anal. Calcd for $\text{C}_{22}\text{H}_{22}\text{N}_2$: C, 84.04; H, 7.05; N, 8.91. Found: C, 84.38; H, 7.32; N, 8.92.

***N*-(4-Methylphenyl)-*N*-[*(1E)*-1-morpholin-4-yl-2-phenylethylidene]amine (7k):** purified by recrystallization from chloroform/hexanes; colorless needles; mp 104–106 °C; yield, 88% (0.130 g); ^1H NMR δ 2.25 (s, 3H), 3.42–3.45 (m, 4H), 3.56–3.59 (m, 4H), 3.70 (s, 2H), 6.65 (d, J = 8.1 Hz, 2H), 6.99 (d, J = 8.0 Hz, 2H), 7.14–7.23 (m, 3H), 7.27–7.32 (m, 2H); ^{13}C NMR δ 20.7, 33.5, 45.2, 66.7, 121.5, 126.4, 127.7, 128.7, 129.4, 131.1, 136.3, 148.4, 157.1. Anal. Calcd for $\text{C}_{19}\text{H}_{22}\text{N}_2\text{O}$: C, 77.51; H, 7.55; N, 9.52. Found: C, 77.21; H, 7.72; N, 9.69.

***N,N'*-bis(4-Methylphenyl)thiophene-2-carboximidamide (7l):** purified by recrystallization from chloroform/hexanes; yellow microcrystals; mp 121–123 °C; yield, 88% (0.135 g); ^1H NMR δ 2.28 (s, 6H), 6.32 (s, 1H), 6.78–7.29 (m, 11H); ^{13}C NMR δ 20.8, 120.8, 121.4, 123.3, 126.7,

127.0, 128.0, 128.5, 129.3, 129.8, 130.1, 131.9, 132.9, 133.9, 134.8, 138.1, 139.0, 145.8, 147.2, 149.5. Anal. Calcd for $C_{19}H_{18}N_2S$: C, 74.47; H, 5.93; N, 9.14. Found: C, 74.32; H, 5.99; N, 9.05.

***N*-(4-Methylphenyl)-*N*-[(1*E*)-morpholin-4-yl(thien-2-yl)methylene]amine (7m):** purified by recrystallization from chloroform/hexanes; pale yellow needles; mp 109–111 °C; yield, 92% (0.130 g); 1H NMR δ 2.20 (s, 3H), 3.42 (t, J = 4.8 Hz, 4H), 3.76 (t, J = 4.8 Hz, 4H), 6.52 (d, J = 8.2 Hz, 2H), 6.81 (d, J = 3.4 Hz, 1H), 6.86–6.90 (m, 3H), 7.29 (d, J = 4.9 Hz, 1H); ^{13}C NMR δ 20.7, 47.3, 66.7, 121.7, 126.8, 127.5, 128.9, 129.9, 131.0, 132.7, 148.1, 154.4. Anal. Calcd for $C_{16}H_{18}N_2OS$: C, 67.10; H, 6.33; N, 9.78. Found: C, 67.24; H, 6.44; N, 9.79.

***N,N'*-Bis(4-methylphenyl)furan-2-carboximidamide (7n):** purified by recrystallization from chloroform/hexanes; yellow needles; mp 120–122 °C; yield, 90% (0.130 g); 1H NMR δ 2.31 (s, 6H), 5.73 (s, 1H), 6.27 (s, 1H), 7.11–7.35 (m, 10H); ^{13}C NMR δ (3 signals are hidden) 20.8, 111.8, 115.3, 120.2, 129.5, 131.9, 141.6. Anal. Calcd for $C_{19}H_{18}N_2O$: C, 78.59; H, 6.29; N, 9.66. Found: C, 78.20; H, 6.29; N, 9.66.

***N*-[(1*E*)-2-Furyl(morpholin-4-yl)methylene]-*N*-(4-methylphenyl)amine (7o):** purified by recrystallization from chloroform/hexanes; colorless needles; mp, 94–96 °C; yield, 89% (0.120 g); 1H NMR δ 2.23 (s, 3H), 3.39 (t, J = 4.8 Hz, 4H), 3.78 (t, J = 4.8 Hz, 4H), 6.06 (d, J = 3.3 Hz, 1H), 6.25–6.27 (m, 1H), 6.51 (d, J = 8.1 Hz, 2H), 6.92 (d, J = 8.1 Hz, 2H), 7.36 (d, J = 1.2 Hz, 1H); ^{13}C NMR δ 20.7, 46.8, 66.7, 110.8, 114.2, 121.3, 129.0, 131.1, 142.5, 144.3, 148.3, 151.3. Anal. Calcd for $C_{16}H_{18}N_2O_2$: C, 71.09; H, 6.73; N, 10.37. Found: C, 71.13; H, 6.92; N, 10.23.

***N'*-Benzyl-*N*-(4-methylphenyl)-4-nitrobenzenecarboximidamide (7p):** purified by recrystallization from chloroform/hexanes; yellow microcrystals; mp 121–123 °C; yield, 92%

(0.160 g); ^1H NMR (mixture of two isomers) δ 2.19 (s, 3H), 2.23 (s, 1.2H), 4.68 (s, 2H), 6.51 (d, $J = 8.0$ Hz, 2H), 6.58 (d, $J = 8.2$ Hz, 0.8H), 6.87 (d, $J = 8.0$ Hz, 2H), 6.95 (d, $J = 8.1$ Hz, 0.8H), 7.28–7.44 (m, 8.4H), 7.77 (d, $J = 10.6$ Hz, 1H), 8.05 (d, $J = 8.5$ Hz, 2H), 8.23 (d, $J = 8.8$ Hz, 0.8H); ^{13}C NMR δ 20.7, 46.2, 115.2, 122.5, 123.4, 127.6, 128.1, 128.7, 129.2, 131.3, 138.4, 141.5, 147.7, 154.8. Anal. Calcd for $\text{C}_{21}\text{H}_{19}\text{N}_3\text{O}_2$: C, 73.03; H, 5.54; N, 12.17. Found: C, 73.03; H, 5.57; N, 12.15.

(1*E*)-*N,N'*-Bis(4-methylphenyl)ethanimidamide (7q): purified by recrystallization from chloroform/hexanes; white microcrystals; mp 118–120 °C (lit.⁴¹ mp 119–120 °C); yield, 91 % (0.110 g); ^1H NMR δ 1.96 (s, 3H), 2.30 (s, 6H), 7.04–7.11 (m, 8H); ^{13}C NMR δ 20.8, 121.3, 129.5, 153.6.

***N'*-Benzyl-*N*-(4-methylphenyl)benzenecarboximidamide (7r):** purified by recrystallization from chloroform/hexanes; white microcrystals; mp 114–116 °C (lit.⁴² 127–127.5 °C); yield, 87% (0.130 g); ^1H NMR δ 2.19 (s, 3H), 4.68 (s, 2H), 6.57 (d, $J = 7.6$ Hz, 2H), 6.86 (d, $J = 7.6$ Hz, 2H), 7.23–7.44 (m, 10H); ^{13}C NMR δ (6 signals are hidden) 20.7, 46.1, 122.8, 127.4, 128.2, 128.5, 128.6, 128.9, 139.0.

***N*-Cyclohexyl-*N'*-(4-methylphenyl)furan-2-carboximidamide (7s):** purified by column chromatography on basic alumina; colorless oil; yield, 85% (0.120 g); ^1H NMR δ 1.23–1.75 (m, 8H), 2.16 (d, $J = 8.6$ Hz, 2H), 2.31 (s, 3H), 3.97 (br s, 1H), 5.17 (br s, 1H), 5.60 (s, 1H), 6.20 (s, 1H), 6.68 (d, $J = 7.7$ Hz, 2H), 7.05 (d, $J = 7.8$ Hz, 2H), 7.27 (s, 1H); ^{13}C NMR δ 20.8, 24.9, 25.9, 33.1, 48.8, 111.5, 114.3, 121.3, 129.6, 131.0, 141.2, 144.7, 145.0, 149.4. Anal. Calcd for $\text{C}_{18}\text{H}_{22}\text{N}_2\text{O}$: C, 76.56; H, 7.85; N, 9.92. Found: C, 76.41; H, 7.81; N, 9.56.

***N*-Methyl-*N'*-(4-methylphenyl)-*N'*,3-diphenylpropanimidamide (7t):** purified by recrystallization from chloroform/hexanes; colorless microcrystals; mp 89–91 °C; yield, 91% (0.150 g); ^1H NMR δ 2.31 (s, 3H), 2.40–2.51 (m, 4H), 3.36 (s, 3H), 6.56–6.58 (m, 2H), 6.70 (d, J = 8.2 Hz, 2H), 7.03–7.11 (m, 5H), 7.23–7.32 (m, 3H), 7.38–7.43 (m, 2H); ^{13}C NMR δ 20.8, 30.5, 33.4, 39.8, 121.7, 125.9, 126.6, 127.8, 128.0, 128.1, 129.4, 130.9, 140.8, 146.2, 148.5, 159.5. Anal. Calcd for $\text{C}_{23}\text{H}_{24}\text{N}_2$: C, 84.11; H, 7.36; N, 8.53. Found: C, 84.09; H, 7.51; N, 8.56.

4-Chloro-*N*-methyl-*N'*-(4-methylphenyl)-*N*-phenylbenzenecarboximidamide (7u): purified by recrystallization from chloroform/hexanes; yellow needles; mp 123–125 °C, yield, 90% (0.150 g); ^1H NMR δ 2.19 (s, 3H), 3.54 (s, 3H), 6.49 (d, J = 8.1 Hz, 2H), 6.86 (d, J = 8.1 Hz, 2H), 6.90–7.00 (m, 7H), 7.13 (t, J = 7.7 Hz, 2H); ^{13}C NMR δ 20.7, 40.2, 122.3, 125.1, 126.9, 127.8, 128.8, 129.0, 130.6, 131.5, 132.1, 133.8, 146.0, 147.8, 158.1. Anal. Calcd for $\text{C}_{21}\text{H}_{19}\text{ClN}_2$: C, 75.33; H, 5.72; N, 8.37. Found: C, 75.17; H, 5.80; N, 8.22.

***N*-Butyl-*N'*-phenylbenzenecarboximidamide (7w):** purified by recrystallization from chloroform/hexanes; white microcrystals; 95–97 °C, yield, 87% (0.110 g); ^1H NMR δ 0.98 (t, J = 7.1 Hz, 3H), 1.43–1.50 (m, 2H), 1.61–1.70 (m, 2H), 6.63 (d, J = 7.4 Hz, 2H), 6.78 (t, J = 7.0 Hz, 1H), 7.03 (t, J = 7.3 Hz, 2H), 7.21–7.29 (m, 5H); ^{13}C NMR δ 13.9, 20.3, 31.4, 41.6, 121.0, 123.1, 128.2, 128.5, 129.0, 135.5, 151.0, 157.5. Anal. Calcd for $\text{C}_{17}\text{H}_{20}\text{N}_2$: C, 80.91; H, 7.99; N, 11.10. Found: C, 80.90; H, 7.98; N, 11.06.

Methyl 3-phenyl-2-[(1*E*)-*N*-phenylethanimidoyl]amino}propanoate acetate (7x): purified by recrystallization from chloroform/hexanes; colorless microcrystals; yield, 90% (0.160 g); mp 55–57 °C; ^1H NMR δ 1.97 (s, 1H), 2.13 (s, 3H), 3.01–3.17 (m, 2H), 3.70 (s, 3H), 4.86 (q, J = 6.7 Hz, 1H), 6.36 (d, J = 7.5 Hz, 1H), 7.05–7.32 (m, 8H), 7.53 (d, J = 7.8 Hz, 2H), 8.47 (s, 1H); ^{13}C NMR δ 22.9, 24.2, 37.6, 52.2, 119.9, 123.9, 127.0, 128.4, 128.7, 129.0, 135.7, 138.1, 169.0,

170.0, 172.0. Anal. Calcd for $C_{20}H_{24}N_2O_4$: C, 67.40; H, 6.79; N, 7.86. Found: C, 67.14; H, 7.05; N, 8.40.

***N*-(4-Methylphenyl)-*N*-[(1*E*)-1-morpholin-4-ylethylidene]amine (7y):** purified by recrystallization from chloroform/hexanes; white microcrystals; yield, 92% (0.100 g); mp 67–68 °C; ^1H NMR δ 1.84 (s, 3H), 2.29 (s, 3H), 3.47 (t, $J = 4.9$ Hz, 4H), 3.74 (t, $J = 4.9$ Hz, 4H), 6.60 (d, $J = 8.1$ Hz, 2H), 7.05 (d, $J = 8.1$ Hz, 2H); ^{13}C NMR δ 14.4, 20.7, 45.2, 66.7, 121.6, 129.3, 130.9, 148.7, 157.1. Anal. Calcd for $C_{13}H_{18}N_2O$: C, 71.53; H, 8.31; N, 12.83. Found: C, 71.28; H, 8.78; N, 12.62.

***N*-Cyclohexyl-4-nitro-*N'*-phenylbenzenecarboximidamide (7z):** purified by recrystallization from chloroform/hexanes; yellow microcrystals; mp 142–144 °C; 90% (0.145 g); ^1H NMR δ ^1H NMR δ 1.18–1.51 (m, 6H), 1.74–1.79 (m, 2H), 2.16 (d, $J = 11.1$ Hz, 2H), 3.99 (br s, 1H), 4.75 (d, $J = 6.6$ Hz, 2H), 6.65 (d, $J = 8.9$ Hz, 2H), 7.19–7.36 (m, 5H), 7.94 (d, $J = 8.9$ Hz, 2H); ^{13}C NMR δ 24.7, 25.6, 32.8, 50.1, 122.9, 124.6, 128.3, 128.6, 129.8, 134.4, 141.4, 157.4, 158.5. Anal. Calcd for $C_{19}H_{21}N_3O_2$: C, 70.57; H, 6.55; N, 12.99. Found: C, 71.07; H, 6.96; N, 12.40.

***N*-(4-Methylphenyl)-4-nitro-*N'*-phenylbenzenecarboximidamide (7Aa):** purified by recrystallization from chloroform/hexanes; yellow microcrystals; mp 127–129 °C (lit.⁴³ mp 138 °C); 91% (0.150 g); ^1H NMR δ 2.31 (s, 3H), 6.88 (br s, 2H), 7.11 (d, $J = 7.7$ Hz, 2H), 7.31 - 7.38 (m, 7H), 8.02 (d, $J = 8.7$ Hz, 2H); ^{13}C NMR δ (3 signals are hidden) 20.8, 120.6, 122.1, 124.8, 128.5, 129.5, 130.3, 134.1, 142.2, 154.9.