Electronic Supporting information for

Copper-catalyzed Goldberg-type C–N coupling in *Deep Eutectic Solvents* and Water under aerobic conditions

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1. General Methods and Materials

Eutectic mixtures of solvents choline chloride-glycerol (1:2 mol/mol; 1ChCl/2Gly); choline chloride-water (1:2 mol/mol; 1ChCl/2H₂O); betaine-urea (1:1 mol/mol; 1Bet/1Urea) and choline chloride–Urea (1:2 mol/mol); 1ChCl/2Urea) were prepared by heating (up to 75 °C) under stirring the corresponding individual components until a clear solution was obtained (ca. for 10-30 min). For ¹H, and ¹³C {¹H} NMR spectra (¹H NMR 300 MHz; ¹³C NMR 75 MHz), CDCl₃ or DMSO-d₆ was used as the solvent. ¹H NMR and ¹³C NMR spectra were recorded on a Bruker 300 MHz spectrometer and chemical shifts are reported in parts per million (δ). Infrared (IR) spectra were recorded on a Diffuse Reflectance sampling cell. Conversion of the products were determined via HPLC analysis: Phenomenex® Lux Cellulose-1 column, acetonitrile (MeCN) and 0.1% trifluoroacetic acid (TFA) in water as solvents. Samples were eluted with three linear gradients from 10% to 60% MeCN during 5.70 min, followed by another from 60% to 100% MeCN during 0.5 min and a third gradient from 100% to 10% MeCN during 1.90 min, at flow rate of 2 mL/min. Detection and spectral characterization of peaks were performed at 210 nm. Analytical thin layer chromatography (TLC) was carried out on pre-coated 0.25 mm thick plates of Kieselgel 60 F254; visualization was accomplished by UV light (254 nm) or by spraying with a solution of 5 % (w/v) ammonium molybdate and 0.2 % (w/v) cerium(III) sulphate in 100 ml of aq. sulphuric acid [17.6 % (w/v)] and heating to 473 K until blue spots appeared. Chromatography was conducted by using silica gel 60 with a particle size distribution 40–63 µm and 230–400 ASTM.

4-Iodotoluene (99%, Aldrich), iodobenzene (98%, Aldrich), 3-iodoanisole (99%, Aldrich), 4iodoanisole (99%, Aldrich), 2-methylbenzamide (98%, Aldrich), 2-fluorobenzamide (98%, Aldrich), 3-methoxybenzamide (97%, Aldrich), *N*-methylbenzamide (>99%, Aldrich), propionamide (97%, Aldrich) and 4-chlorobenzamide (98%, Aldrich) were used as received without any further purification. Potassium hydroxide, ethylene diamine and copper(I) iodide were commercially available. Spectroscopic data of compounds **3a**,¹ **3b**,² **3c**,³ **3d**,⁴ **3e**,² **3f**,⁵ **3h**,⁶ **3i**,⁷ **3j**,² **3k**,⁸ and **3l**⁹ are in agreement with those previously reported in the literature. Fully characterization details, including ¹H and ¹³C{¹H} NMR spectra for the compound **3g**, are included in the following sections of this ESI.

2. Experimental procedure and characterization details

2.1 Synthesis of 2-methyl-*N*-(*p*-tolyl)benzamide (3a) in *Deep Eutectic Solvents* (*DESs*). Typical procedure.



CuI (10 mol%, 0.05 mmol, 10 mg), 4-iodotoluene (**1a**, 1 equiv., 0.5 mmol, 109 mg), 2methylbenzamide (**2a**, 1 equiv., 0.5 mmol, 67 mg), the base (KOH, 3 equiv., 1.5 mmol, 84 mg) and the ligand (ethylene diamine, 10 mol%, 0.05 mmol, 3 mg, 3 μ L), were suspended in 1 mL of 1*ChCl*/2H₂O, under air, in a vial with a Teflon screw tap under vigorous stirring at 80 °C. The reaction mixture was monitored by HPLC. After 12 h, the mixture was cooled to room temperature and 1 mL of H₂O was added. Then, the mixture was extracted with cyclopentyl methyl ether (*CPME*, 1 mL) and the organic phase was dried over anhydrous Na₂SO₄ and filtered over a celite pad. Evaporation of the solvent under reduced pressure afforded the crude that was purified by flash-chromatography on silica gel (hexane/Et₂O 5:5) to provide the desired product **3a** in 80% yield (90 mg).

2.2 Synthesis of 2-methyl-*N*-(*p*-tolyl)benzamide (3a) in *water*. Typical procedure.



CuI (10 mol%, 0.05 mmol, 10 mg), 4-iodotoluene (**1a**, 1 equiv., 0.5 mmol, 109 mg), 2methylbenzamide (**2a**, 1 equiv., 0.5 mmol, 67 mg), the base (KOH, 3 equiv., 1.5 mmol, 84 mg) and the ligand (ethylene diamine, 10 mol%, 0.05 mmol, 3 mg, 3 μ L), were suspended in 1 mL of H₂O, under air, in a vial with a Teflon screw tap under vigorous stirring at 80 °C. The reaction mixture was monitored by HPLC. After 12 h, the mixture was cooled to room temperature and 1 mL of H₂O was added. Then, the mixture was extracted with cyclopentyl methyl ether (*CPME*, 1 mL) and the organic phase was dried over anhydrous Na_2SO_4 and filtered over a celite pad. Evaporation of the solvent under reduced pressure afforded the crude that was purified by flash-chromatography on silica gel (hexane/Et₂O 5:5) to provide the desired product **3a** in 82% yield (96 mg).

2.3 Scaled-up synthesis of 2-methyl-*N*-(*p*-tolyl)benzamide (3a)



CuI (10 mol%, 0.25 mmol, 50 mg), 4-iodotoluene (**1a**, 1 equiv., 2.5 mmol, 545 mg), 2methylbenzamide (**2a**, 1 equiv., 2.5 mmol, 335 mg), the base (KOH, 3 equiv., 7.5 mmol, 420 mg) and the ligand (ethylene diamine, 10 mol%, 0.25 mmol, 15 mg, 15 μ L), were suspended in 5 mL of *DES* 1*ChCl*/2H₂O, under air, in a vial with a Teflon screw tap under vigorous stirring at 80 °C. The reaction mixture was monitored by HPLC. After 12 h, the mixture was cooled to room temperature and 5 mL of H₂O were added. Then, the mixture was extracted with cyclopentyl methyl ether (*CPME*, 5 mL) and the organic phase was dried over anhydrous Na₂SO₄ and filtered over a celite pad. Evaporation of the solvent under reduced pressure afforded the crude that was purified by flash-chromatography on silica gel (hexane/Et₂O 5:5) to provide the desired product **3a** in 90% yield (504 mg).

2.4 Experimental procedure for the recyclability studies



The recyclability of our catalytic system was investigated using the CuI-catalyzed Goldberg-type coupling between iodobenzene (**1b**) and 2-methylbenzamide (**2a**) as a model reaction. Thus, CuI (10 mol%, 0.05 mmol, 10 mg), iodobenzene (**1b**, 1 equiv., 0.5 mmol, 102 mg), 2-methylbenzamide (**2a**, 1 equiv., 0.5 mmol, 67 mg), the base (KOH, 3 equiv., 1.5 mmol, 84 mg) and the ligand (ethylene diamine, 10 mol%, 0.05 mmol, 3 mg, 3 μ L), were suspended in 1 mL of *DES*

 $1ChCl/2H_2O$, under air, in a vial with a Teflon screw tap under vigorous stirring at 80 °C. The reaction mixture was monitored by HPLC. The catalytic system recovery was carried out by liquid-liquid extraction of the organic products with 1 mL of cyclopentyl methyl ether (*CPME*) and subsequent addition of iodobenzene (**1b**, 1 equiv., 0.5 mmol, 102 mg) and 2-methylbenzamide (**2a**, 1 equiv., 0.5 mmol, 67 mg) to the reaction media. The mixture was then stirred for 12 h. This procedure was repeated up to three consecutive times.

2.5 E-factor calculations

According to its original definition,¹⁰ the Sheldon E-factor value (total mass of waste/mass of product) takes into account only the mass of waste generated in a process, and its calculation is performed by simply dividing the sum of the molecular weight of all substances produced by molecular weight of the desired products, with reference to the stoichiometric equation. Thus, the amount of silica gel, the celite pad, the drying agents, and the mass of eluent solvent used for chromatography are usually not included in the calculation. We have followed this general equation in our own calculations. Note: CPME 0.860 g/mL, at 25 °C.



Total amount of reactants: $1 g + (4 \times 0.067 g) + (4 \times 0.102 g) + 0.010 g + 0.003 g + 0.084 g + (4 \times 0.860 g) = 5.215 g$

Amount of final product: 0.329 g

Amount of waste: 5.215 g – 0.329 g = 4.886 g

E-Factor = amount of waste/amount of product = 4.886 g/0.329 g = 15



2-Methyl-*N***-(***p***-tolyl)benzamide (3a):** white solid (yield: 82%) m.p. 146–149 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.53 (bs, 1H), 7.48 (d, *J* = 8.4 Hz, 2H), 7.43 (d, *J* = 7.6 Hz, 1H), 7.34-7.30 (m, 1H), 7.24-7.18 (m, 2H), 7.15 (d, *J* =

8.0 Hz, 2H), 2.46 (s, 3H), 2.32 (s, 3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 167.9, 136.5, 136.3, 135.3, 134.1, 131.1, 130.1, 129.5, 126.6, 125.8, 119.9, 20.9, 19.7. FT-IR (KBr, cm⁻¹) = 3225, 1650, 1601, 1514, 1323. GC/MS (70 eV) *m/z* (%): 225 (M⁺, 30), 119 (100), 106 (2), 77 (5), 65 (11). HRMS (ESI) *m/z* calcd. for [C₁₅H₁₅NO + H]⁺ = 226.1226; found: 226.1228. Spectroscopic data are in good agreement with those reported in: *Adv. Synth. Catal.*, **2018**, *360*, 4784-4789.



2-Methyl-*N***-phenylbenzamide (3b)** white solid (yield: 90%) m.p. 125–128 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.76 (d, *J* = 7.6 Hz, 2H), 7.73 (s, 1H), 7.46 (d, *J* = 7.4 Hz, 1H), 7.36 (t, *J* = 8.0 Hz, 3H), 7.27 (dd, *J* = 11.6, 7.9 Hz, 2H), 7.11

(t, J = 7.4 Hz, 1H), 2.38 (s, 3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 167.9, 139.3, 137.3, 135.2, 130.5, 129.6, 127.2, 125.6, 123.5, 119.6, 19.3. FT-IR (KBr, cm⁻¹) = 3218, 1652, 1597, 1533, 1439, 1321, 1260, 889, 757. HRMS (ESI) *m*/*z* calcd. for [C₁₄H₁₄NO + H]⁺ = 212.1070; found: 212.1078. Spectroscopic data are in good agreement with those reported in: *Tetrahedron Lett.*, **2014**, *55*, 124-127.



N-(3-Methoxyphenyl)-2-methylbenzamide (3c) white solid (yield: 74%) m.p. 143–146 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.53 (bs, 1H), 7.48-7.41 (m, 2H), 7.34 (td, J = 7.6, 1.3 Hz, 1H), 7.30-7.23 (m, 3H), 7.21 (d, J = 7.6 Hz, 1H), 7.15 (dd, J = 8.3, 2.0 Hz, 1H), 3.84 (s, 3H), 2.46 (s, 3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 168.0, 160.3, 139.3, 136.5, 136.3, 131.1, 130.1, 129.5, 126.6, 125.8, 112.0, 110.4, 105.6, 55.9, 19.8. FT-IR (KBr, cm⁻¹) = 3286, 1681, 1523, 1491, 1391, 1266. GC/MS (70 eV) m/z (%): 241 (M⁺, 35), 224 (4), 119 (100), 91 (36), 65 (10). HRMS (ESI) m/z calcd. for [C₁₅H₁₅NO₂ + H]⁺ = 242.1176; found: 212.1084. Spectroscopic data are in good agreement with those reported in: *Chem. Comm*, **2015**, *51*, 1371-1374.



3-Methoxy-*N***-(p-tolyl)benzamide (3d)** white solid (yield: 83%) m.p. 128–131 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.91 (s, 1H), 7.53 (d, *J* = 8.2 Hz, 2H), 7.42 (s, 1H), 7.38 (q, *J* = 7.9 Hz, 2H), 7.17 (d, *J* = 8.2 Hz, 2H), 7.07 (d, *J* = 7.6

Hz, 1H), 3.84 (s, 3H), 2.34 (s, 3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 165.5, 159.9, 136.5, 135.3, 134.2, 129.7, 129.5, 120.3, 118.7, 117.9, 112.4, 55.4, 20.9. FT-IR (neat, cm⁻¹) = 3296, 2922, 1648, 1594, 1517. GC/MS (70 eV) *m/z* (%) = 241 (M). HRMS (ESI) *m/z* calcd. for [C₁₅H₁₆NO₂ + H]⁺ = 242.1176; found: 242.1183. Spectroscopic data are in good agreement with those reported in: *Org. Lett.*, **2019**, *21*, *12*, 4878.



3-Methoxy-*N***-phenylbenzamide (3e)** white solid (yield: 95%) m.p. 112–115 °C;

¹H NMR (CDCl₃) δ (ppm) = 8.16 (s, 1H), 7.64 (d, *J* = 7.6 Hz, 2H), 7.42 (d, *J* = 1.9 Hz, 1H), 7.39-7.33 (m, 4H), 7.32-7.14 (m, 1H), 7.05-7.03 (m, 1H), 3.80 (s, 1H), 7.10 - 100 (m, 1H), 7.10 (m, 1H), 7.10

3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 167.9, 160.3, 139.1, 135.4, 130.1, 129.5, 126.6, 125.8, 112.0, 110.4, 105.6, 56.9. FT-IR (KBr, cm⁻¹) = 3314, 2936, 2831, 1648, 1549, 1487, 1434, 1319, 1271, 1243, 1222, 1180, 1076, 1035, 901, 876, 861, 804. GC/MS (70 eV) *m/z* (%) = 227 (M⁺, 65), 105 (100). HRMS (ESI) *m/z* calcd. for [C₁₄H₁₄NO₂ + H]⁺ = 228.1019; found: 228.1025. Spectroscopic data are in good agreement with those reported in: *Tetrahedron Lett.*, **2014**, *55*, 124-127.



Hz, 2H), 7.42-7.36 (m, 2H), 7.33-7.16 (m, 3H). ${}^{13}C{}^{1}H$ NMR (CDCl₃) δ (ppm) = 161.6 (d, J_{CF} = 3.5 Hz), 160.4 (d, J_{CF} = 245 Hz), 137.7, 133.8 (d, J_{CF} = 3 Hz), 129.0 (d, J_{CF} = 9 Hz), 125.0, 120.6, 116.3 (d, J_{CF} = 22 Hz). FT-IR (KBr, cm⁻¹): 3347, 3082, 2919, 1654, 1587, 1524. HRMS (ESI) m/z calcd. for [C₁₃H₁₁FNO + H]⁺ = 216.0819; found: 216.0825. Spectroscopic data are in good agreement with those reported in: *Green Chem.*, **2019**, *21*, 3675-3681.



2-Fluoro-N-phenylbenzamide (3g) white solid (yield: 57%) m. p. 155-157 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.53 (bs, 1H), 7.48 (d, J = 8.4 Hz, 2H), 7.46 (d, J = 7.6 Hz, 1H), 7.34-7.26 (m, 1H), 7.17-7.14 (m, 2 H), 6.99 (d, J = 8.0 Hz, 2H), 2.32 (s, 3H). ${}^{13}C{}^{1}H, {}^{19}F{} NMR (CDCl_3) \delta (ppm) = 165.5, 159.0, 136.5, 134.2, 129.6, 129.5, 120.3, 118.6, 136.5$ 117.9, 112.4, 20.9. FT-IR (film, cm⁻¹): 3379, 3051, 2821, 1620, 1505, 1313. GC/MS (70 eV) m/z (%): 229 (M⁺, 46), 123 (100), 95 (21), 75 (6). HRMS (ESI) m/z calcd. for $[C_{14}H_{13}FNO + H]^+$: 230.0976; found: 230.0981.



N-Methyl-*N*-phenylbenzamide (3h) white solid (yield: 65%) m.p. 62–64 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.30-7.27 (m, 2H), 7.23-7.19 (m, 3H), 7.16-7.10 (m, 3H), 7.04-7.02 (m, 2H), 3.49 (s, 3H). ${}^{13}C{}^{1}H{}$ NMR (CDCl₃) δ (ppm) = 170.5, 144.7, 135.8, 129.5, 129.0, 128.6, 127.6, 126.8, 126.3, 38.3. FT-IR (KBr, cm^{-1}) = 3056, 2921, 2850, 1644, 1594, 1486, 1360, 1302, 1105, 704, 684, 590. GC/MS (70 eV) m/z (%) = 211 (M+, 30), 105 (100). HRMS (ESI) m/z calcd. for $[C_{14}H_{14}NO + H]^+ = 212.1070$; found: 212.1075. Spectroscopic data are in good agreement with those reported in: Green Chem., 2018, 20, 3457-3462.



N-(*p*-Tolyl)propionamide (3i) white solid (yield: 92%) m.p. 119–121 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.48 (s, 1H), 7.42 (d, *J* = 8.1 Hz, 2H), 7.12 (d, J = 8.0 Hz, 2H), 2.39 (q, J = 7.5 Hz, 2H), 2.32 (s, 3H), 1.25 (t, J = 7.5 Hz,

3H). ${}^{13}C{}^{1}H{}$ NMR (CDCl₃) δ (ppm) = 172.2, 135.5, 133.8, 129.4, 120.0, 30.7, 20.9, 9.8. FT-IR (KBr, cm⁻¹): 3303, 1664. GC/MS (70 eV) m/z (%) = 163 (M+, 100). HRMS (ESI) m/z calcd. for $[C_{10}H_{14}NO + H]^+ = 164.1070$; found: 164.1078. Spectroscopic data are in good agreement with those reported in: Eur. J. Org. Chem., 2019, 30, 4911-4915.

N-Phenylpropionamide (3j) white solid (yield: 60%) m.p. 107–110 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.62 (d, J = 7.9 Hz, 2H), 7.51 (s, 1H), 7.27 (t, J = 7.7 Hz, 2H), 7.09 (t, J = 7.3 Hz, 1H), 2.38 (q, J = 7.5 Hz, 2H), 1.23 (t, J = 7.5

Hz, 3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 172.1, 138.1, 129.1, 124.3, 120.0, 30.9, 9.8. FT-IR

(KBr, cm⁻¹) = 3256, 1665. HRMS (ESI) m/z calcd. for [C₉H₁₂NO + H]⁺: 150.0913; found: 150.0919. Spectroscopic data are in good agreement with those reported in: *Tetrahedron Lett.*, **2014**, *55*, 124-127.



N-(3-Methoxyphenyl)propionamide (3k) white solid (yield: 42%) m.p. 76–78 °C;

¹H NMR (CDCl₃) δ (ppm) = 7.57 (b, 1H), 7.34 (s, 1H), 7.16 (t, *J* = 8.0 Hz, 1H), 7.86 (d, *J* = 7.6 Hz, 1H), 6.64 (d, *J* = 8.4 Hz, 1H), 3.77 (s, 3H), 2.37 (q, *J* = 7.6 Hz, 2H), 1.22 (t, *J* = 7.6 Hz, 3H). ¹³C{¹H} NMR (CDCl₃) δ (ppm) = 172.3, 160.3, 139.4, 129.7, 112.0, 110.0, 105.0, 55.4, 31.8, 9.8. FT-IR (neat, cm⁻¹) = 3304, 2977, 1661, 1596, 1541, 1491, 1451, 1284, 1218, 1154. HRMS (ESI) *m/z* calcd. for [C₁₀H₁₄NO₂ + H]⁺ = 180.1019; found: 180.1025. Spectroscopic data are in good agreement with those reported in: *Org. Process. Res. Dev.*, **2016**, *209*, 1662-1667.



4-Chloro-*N***-(4-methoxyphenyl)benzamide (31)** pale-brown solid (yield: 87%); m. p. 208–210 °C;

¹H NMR (DMSO-d₆) δ (ppm) = 10.20 (bs, 1H), 7.98 (d, *J* = 9.0 Hz, 2H), 7.67 (d, *J* = 9.0 Hz, 2H), 7.61 (d, *J* = 9.0 Hz, 2H), 6.94

(d, J = 9.0 Hz, 2H), 3.75 (s, 3H).¹³C{¹H} NMR (DMSO-d₆) δ (ppm) = 164.4, 156.1, 136.7, 134.2, 132.5, 130.0, 128.9, 122.5, 114.2. FT-IR (KBr, cm⁻¹) = 3345, 2929, 1647, 1601, 1532, 1514, 1232, 1028, 831, 821, 754, 524. Spectroscopic data are in good agreement with those reported in: *Tetrahedron Lett.*, **2008**, *49*, 1636-1640.

3. ¹H and ¹³C NMR spectra

¹H NMR 300 MHz, CDCl₃ (3a)



¹³C NMR 75.4 MHz, CDCl₃ (3a)



¹H NMR 300 MHz, CDCl₃ (**3b**)



¹³C NMR 75.4 MHz, CDCl₃ (**3b**)



¹H NMR 300 MHz, CDCl₃ (3c)



¹³C NMR 75.4 MHz, CDCl₃ (3c)



¹H NMR 300 MHz, CDCl₃ (**3d**)



¹³C NMR 75.4 MHz, CDCl₃ (**3d**)



¹H NMR 300 MHz, CDCl₃ (3e)



¹³C NMR 75.4 MHz, CDCl₃ (**3e**)



¹H NMR 300 MHz, CDCl₃ (**3f**)



¹³C NMR 75.4 MHz, CDCl₃ (**3f**)



¹H NMR 300 MHz, CDCl₃ (**3**g)



¹³C NMR 75.4 MHz, CDCl₃ (**3g**)



¹H NMR 300 MHz, CDCl₃ (**3h**)



¹³C NMR 75.4 MHz, CDCl₃ (**3h**)



¹H NMR 300 MHz, CDCl₃ (**3i**)



¹³C NMR 75.4 MHz, CDCl₃ (**3i**)



¹H NMR 300 MHz, CDCl₃ (**3j**)



¹³C NMR 75.4 MHz, CDCl₃ (**3j**)



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¹H NMR 300 MHz, CDCl₃ (3k)



¹³C NMR 75.4 MHz, CDCl₃ (3k)



¹H NMR 300 MHz, DMSO-d₆ (**3l**)



¹³C NMR 75.4 MHz, DMSO-d₆ (3l)



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