

EXPERIMENTAL SECTION.

GO was prepared by graphite oxidation using the Hummers and Offeman method and subsequent exfoliation. Further details and GO characterization have been previously reported.

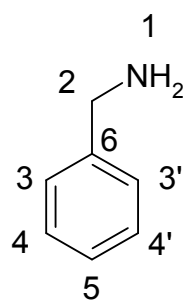
Raman spectra were collected with a Horiba Jobin Yvon – Labram HR UV–Visible–NIR Raman Microscope Spectrometer, using a 632 nm laser. The spectra were the average of 10 scans at a resolution of 2 cm⁻¹ between 1000-2000 cm⁻¹ Raman Shift. The liquid samples were analyzed using a GC-MS apparatus (Thermo Electron Trace GC Ultra/Trace DSQ) equipped with a non-polar GC column (Trace TR-1MS) operated with He as carrier gas. Temperature profile was set to 10 min dwell at 60°C, 10°/min ramp up to 240 °C followed by a 10 min dwell. The MS spectra were recorded in positive polarization and full scan mode in the range m/z=50-300.

General. ¹H and ¹³C NMR were recorded in DCCl₃ solutions using a 500 MHz Bruker Advance III spectrometer. MS were measured in a quadrupolar

Catalytic tests. Oxidation of amine (purchased from Sigma-Aldrich, purity >98%) with NaOCl sol. 12% from Olchim SA Romania in the presence or absence of molecular oxygen (Linde, Purity >99.9%) was carried out under pressure conditions and temperature, in an autoclave (reactor 316SS, HEL Group) of 16 mL capacity into which the catalyst (10% w/w), amine (0.27 mmol) and 5 mL solvent were placed. The autoclave was sealed and heated to desired temperature under vigorous magnetic stirring (600 rpm).

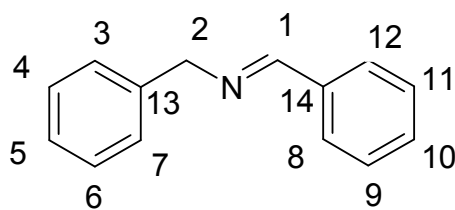
For characterization purposes, reaction products were isolated by column chromatography using mixture of hexanes dichloromethane as eluent.

Spectroscopic data of reaction products.



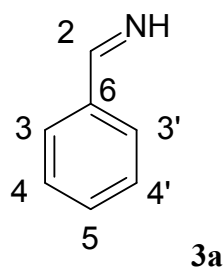
1a

¹H-NMR δ (ppm): vw 1.89 (s, 2H, H-1), 3.85 (s, 2H, H-2), 7.15-7.2 (m, H-aromatic, 3, 3', 4, 4'); ¹³C NMR δ (ppm): 46.3 (C2), 126.5 (C5), 127.1 (C3), 128.3 (C4), 143.5 (C6); MS fragment ions [m/z (relative)]: 50.93(15.50), 76.91(24.71), 77.95(13.34), 78.92(37.72), 90.91(12.98), 105.89(100), 106.94(52.42).

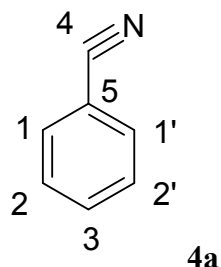


2a

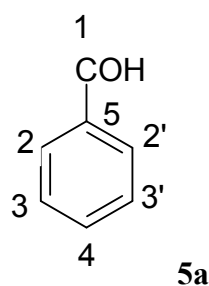
¹H-NMR δ (ppm): 4.79 (s, 2H, H-2), 7.14-7.5 (m, H-aromatic, 3, 4, 6, 7, 8, 9, 11, 12), 8.42 (s, 1H, H-1); ¹³C NMR δ (ppm): 65.01 (C2), 126.92 (C3, 5, 7), 127.25 (C4, 6), 128.5 (C8, 12), 128.3 (C9, 11), 130.7 (C10), 136.1 (C14), 139.21 (C13), 161.93 (C1); MS fragment ions [m/z (relative)]: 64.92(16.78), 88.86(11.79), 90.81(100), 91.94(26.53), 116.91(10.17), 193.88(35.27), 194.95(31.56).



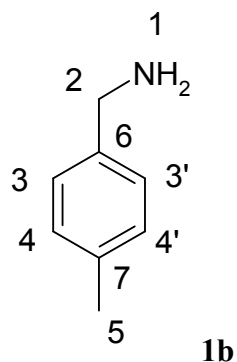
$^1\text{H-NMR}$ δ (ppm): vw 6.51 (s, 1H, H-1), 7.13-7.32 (m, H-aromatic, 3,3',4,4'), 10.15 (s, 2H, H-2); ^{13}C NMR δ (ppm): 128.14 (C3), 129 (C4), 130.95 (C5), 135.18 (C6), 150.53 (C2); MS fragment ions [m/z (relative)]: 52 (14.6), 76(14.5), 28(17.8), 50(23), 103(24.7), 78(33), 51(37.1), 77(45.8), 104(99.9), 105(85).



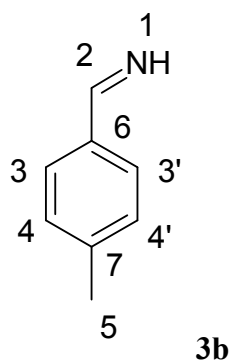
$^1\text{H-NMR}$ δ (ppm): 7.42-7.58 (m, H-1,1',2,2',3); ^{13}C NMR δ (ppm): 112.45 (C5), 118.98 (C4), 129.17 (C2), 132.08 (C1), 132.78 (C3); MS fragment ions [m/z (relative)]: 75.89(33.92), 102.88(100).



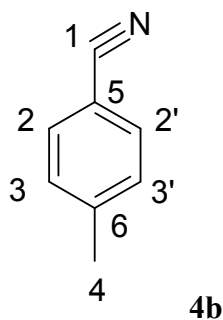
$^1\text{H-NMR}$ δ (ppm): 7.43-7.8 (m, H-aromatic, 2,2',3,3'), 9.94 (s, 1H, H-1); ^{13}C NMR δ (ppm): 129.2 (C3), 129.8 (C2), 134.2 (C4), 136.9 (C5), 192 (C1); MS fragment ions [m/z (relative)]: 50.91(42.69), 73.89(10.70), 76.88(100), 77.93(19.77), 104.91(99.19), 105.92(94.88).



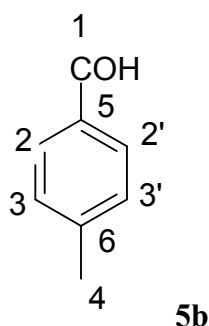
$^1\text{H-NMR}$ δ (ppm): vw 1.89 (s, 2H, H-1), 2.29 (s, 3H, H-5), 3.85 (s, 2H, H-2); 6.94 (d, J=8.1 Hz, 2H, H -3, 3'), 7.2 (d, J=8.1 Hz, 2H, H -4, 4'); ^{13}C NMR δ (ppm): 21 (C5), 46.3 (C2), 127.1 (C4, 4'), 129.1 (C3, 3'), 130.6 (C7), 140.6 (C6); MS fragment ions [m/z (relative)]: 62.92(10.04), 64.90(18.70), 76.89(28.29), 78.92(20.54), 90.93(49.82), 91.96(10.61), 92.92(39.31), 102.91(10.36), 103.92(100), 104.94(23.53), 105.93(59.61), 119.94(72.94), 120.95(32.18).



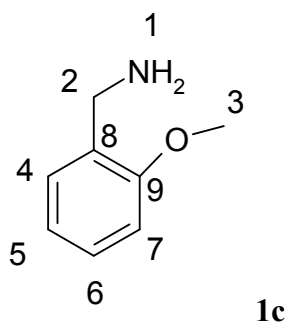
$^1\text{H-NMR}$ δ (ppm): 2.27 (s, 3H, H-5), vw 6.51 (s, 1H, H-1), 7.54 (d, J=7.9 Hz, 2H, H -3, 3'), 7.06 (d, J=7.9 Hz, 2H, H-4, 4'), 10.15 (s, 1H, H-2); ^{13}C NMR δ (ppm): 21.27 (C5), 126.75 (C3, 3'), 129.4 (C4, 4'), 133.55 (C6), 140.23 (C7), 150.53 (C2); MS fragment ions [m/z (relative)]: 76.90(60.23), 78.90(50.77), 90.96(89.39), 92.94(54.30), 103.91(100), 105(52.95), 105.95(61.78), 106.9(53.69), 116.85(56.97), 119.05(70.03).



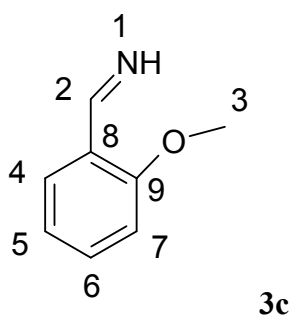
$^1\text{H-NMR}$ δ (ppm): 2.31 (s, 3H, H-4); 7.22 (d, $J=8.0$ Hz, 2H, H-3, 3'), 7.48 (d, $J=8.0$ Hz, 2H, H-2, 2'); $^{13}\text{C NMR}$ δ (ppm): 21.7 (C4), 109.51 (C5), 118.02 (C1), 130.34 (C2, 2'), 132.09 (C3, 3'), 143.99 (C6); MS fragment ions [m/z (relative)]: 62.90(15.80), 88.92(26.18), 89.92(35.09), 115.89(67.11), 116.92(100).



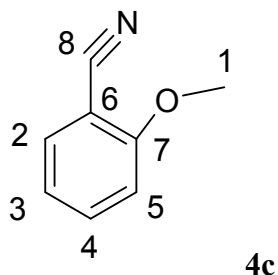
$^1\text{H-NMR}$ δ (ppm): 2.39 (s, 3H, H-4), 7.2 (d, $J=8.0$ Hz, 2H, H-3, 3'), 7.77 (d, $J=8.0$ Hz, 2H, H-2, 2'), 9.85 (s, 1H, H-1); $^{13}\text{C NMR}$ δ (ppm): 21.7 (C4), 129.9 (C2, 2', 3, 3'), 135 (C5), 145.6 (C6), 190.8 (C1); MS fragment ions [m/z (relative)]: 62.90(13.44), 64.91(23.87), 88.92(11.20), 90.89(86.19), 118.89(100), 119.92(84).



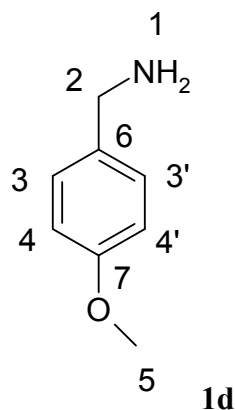
$^1\text{H-NMR}$ δ (ppm): vw 2.1 (s, 2H, H-1), 3.87 (s, 2H, H-2), 4.17 (s, 3H, H-3), 6.72 (q, $J=8.4$ Hz, $J=7.3$ Hz, H, H-6), 6.8 (d, $J=8.4$ Hz, 1H, H-7), 7.07 (q, $J=7.8$ Hz, $J=7.3$ Hz, H, H-5), 7.14 (d, $J=7.8$ Hz, 1H, H-4); $^{13}\text{C NMR}$ δ (ppm): 42.3 (C2), 55.09 (C3), 110.2 (C7), 120.86 (5), 128.3 (C4), 130.6 (C6), 132.1 (C8), 158 (C9); MS fragment ions [m/z (relative)]: 50.92(15.95), 64.91(19.43), 65.93(12.27), 76.89(34.11), 77.90(19.25), 78.93(13.77), 90.89(43.22), 92.94(16.11), 93.94(12.54), 103.89(32.56), 104.94(12.61), 105.93(52.71), 106.95(18.56), 107.94(13.23), 118.92(24.49), 119.94(35.25), 120.92(41.49), 121.95(19.49), 135.90(100), 136.94(68.44).



$^1\text{H-NMR}$ δ (ppm): 3.72 (s, 3H, H-3), vw 6.51 (s, 1H, H-1), 7.15-7.3 (m, 4H, H-4,5,6,7), 10.34 (s, 1H, H-2); $^{13}\text{C NMR}$ δ (ppm): 55.2 (C3), 111.1 (C7), 120.4 (C8), 120.8 (C5), 127.7 (C4), 133.33 (C6), 152.1 (C9), 155.62 (C2); MS fragment ions [m/z (relative)]: 76.94(15.68), 91.90(11.13), 106.9(10.99), 134.86(89), 135.21(54.52).

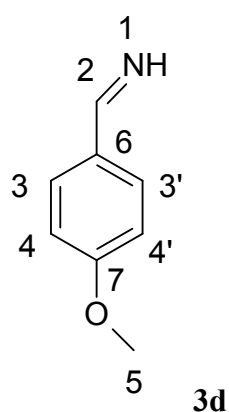


$^1\text{H-NMR}$ δ (ppm): 4 (s, 3H, H-1), 6.86 (t, $J=7.3$ Hz, $J=7.8$ Hz, 1H, H-3), 7.23 (d, $J=8.4$ Hz, 1H, H-5), 7.42 (d, $J=7.8$ Hz, 1H, H-2), 7.52 (t, $J=7.3$ Hz, $J=8.4$ Hz, 1H, H-4); $^{13}\text{C NMR}$ δ (ppm): 56.84 (C1), 99.42 (C6), 107.79 (C5), 116.1 (C8), 121.8 (C3), 128.37 (C4), 134. (C2), 163.61 (C7); MS fragment ions [m/z (relative)]: 61.91(10.77), 62.90(30.57), 63.90(25.92), 74.87(13.16), 75.89(17.16), 76.93(12.67), 77.93(14.84),

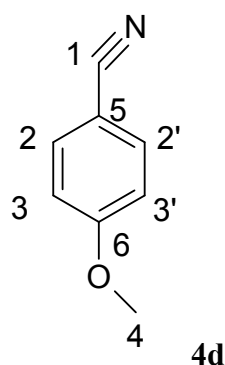


89.88(48.14), 102.93(28.36), 103.92(92.74), 104.94(46), 131.84(29.40), 132.89(100).

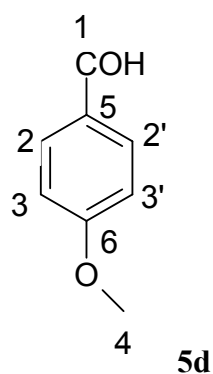
$^1\text{H-NMR}$ δ (ppm): vw 1.89 (s, 2H, H-1), 3.68 (s, 2H, H-2), 3.85 (s, 3H, H-5), 7.23 (d, $J=8.5$ Hz, 2H, H-3, 3'), 7.28 (d, $J=8.5$ Hz, 2H, H-4, 4'); ^{13}C NMR δ (ppm): 46.3 (C2), 55.3 (C5), 113.9 (C4), 128 (C3), 135.85 (C6), 158.79 (C7); MS fragment ions [m/z (relative)]: 28(21.3), 30(28), 77(26.5), 78(19.3), 94(18.1), 106(42.2), 109(20), 121(29), 136(100), 137(48).



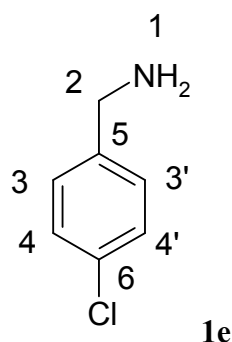
$^1\text{H-NMR}$ δ (ppm): 3.62 (s, 3H, H-5), vw 6.51 (s, 1H, H-1), 6.75 (d, $J=8.5$ Hz, 2H, H-4, 4'), 7.1 (d, $J=8.5$ Hz, 2H, H-3, 3'), 10.15 (s, 1H, H-2); ^{13}C NMR δ (ppm): 54.3 (C5), 114.1 (C4, 4'), 125.6 (C3, 3'), 129.65 (C6), 150.53 (C2), 161.36 (C7); MS fragment ions [m/z (relative)]: 62.90(10.35), 76.90(26.12), 91.8(12.30), 106(10.01), 134.86(98).



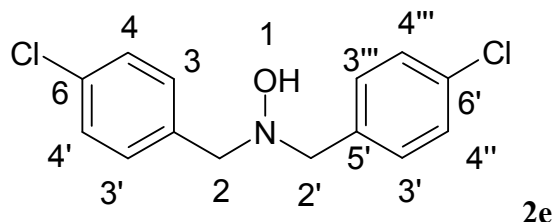
$^1\text{H-NMR}$ δ (ppm): 3.74 (s, 3H, H-4), 6.86 (d, $J=8.8$ Hz, 2H, H-3, 3'), 7.43 (d, $J=8.8$ Hz, 2H, H-2, 2'); ^{13}C NMR δ (ppm): 55 (C4), 105.76 (C5), 115.1 (C3,3'), 118.49 (C1), 134.1 (C2, 2'), 164.25 (C6); MS fragment ions [m/z (relative)]: 62.92(17), 63.91(15.06), 89.90(44.36), 102.92(37.69), 103.94(11.7), 132.90(100).



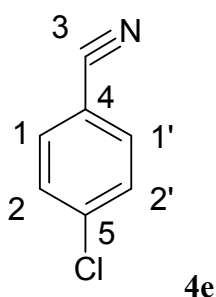
$^1\text{H-NMR}$ δ (ppm): 3.77 (s, 3H, H-4), 6.96 (d, $J=8.5$ Hz, 2H, H-3, 3'), 7.8 (d, $J=8.5$ Hz, 2H, H-2, 2'), 9.88 (s, 1H, H-1); ^{13}C NMR δ (ppm): 55.4 (C4), 114.3 (C3), 130.3 (C3, 3'), 131.6 (C2, 2'), 164.7 (C6), 190.4 (C5); MS fragment ions [m/z (relative)]: 62.92(11.49), 76.91(25.68), 91.88(15.51), 106.88(12.89), 134.86(100), 135.91(68.52).



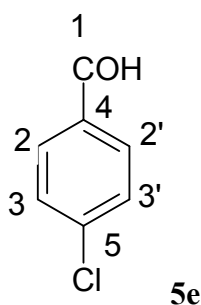
$^1\text{H-NMR}$ δ (ppm): vw 1.89 (s, 2H, H-1), 3.85 (s, 2H, H-2), 7.17 (d, $J=8.4$ Hz, 2H, H-4,4'), 7.24 (d, $J=8.4$ Hz, 2H, H-3,3'); $^{13}\text{C NMR}$ δ (ppm): 46.3 (C2), 128.4 (C3, 3'), 12.8.86 (C4,4'), 131.71 (C6), 140.18 (C7); MS fragment ions [m/z (relative)]: 50.94(12.09), 74.90(10.79), 76.90(28.98), 78.93(11.52), 88.85(10.80), 105.88(100), 124.85(10.44), 139.85(45.26), 140.93(11.97), 141.90(14.17).



$^1\text{H-NMR}$ δ (ppm): 3.85 (s, 4H, H-2,2'), vw 5.1 (s, 1H, H-1), 7.17 (d, $J=8.4$ Hz, 4H, H-4,4',4'',4'''), 7.25 (d, $J=8.4$ Hz, 4H, H-3,3',3'',3'''); $^{13}\text{C NMR}$ δ (ppm): 54.7 (C2,2'), 127.86 (C4), 129.02(C3), 131.03 (C6), 135.61(C5); MS fragment ions [m/z (relative)]: 124.81(100), 125.92(12.18), 126.87(30.35), 262.84(18.13), 264.85(11.20), 281 (5), 283 (3).



$^1\text{H-NMR}$ δ (ppm): 7.38 (d, $J=8.4$ Hz, 2H, H-2,2'), 7.64 (d, $J=8.4$ Hz, 2H, H-1,1'); $^{13}\text{C NMR}$ δ (ppm): 108 (C4), 114 (C3), 127.1 (C2), 131.15 (C1), 140.8 (C5); MS fragment ions [m/z (relative)]: 74.89(13.82), 101.90(26.23), 136.83(100), 138.86(31.45).



$^1\text{H-NMR}$ δ (ppm): 7.45 (d, $J=8.3$ Hz, 2H, H-3,3'), 7.75 (d, $J=8.3$ Hz, 2H, H-2,2'), 9.8 (s, 1H, H-1); $^{13}\text{C NMR}$ δ (ppm): 129.4 (C3), 130.8 (C2), 134.9 (C4), 139.23 (C5), 191.06 (C1); MS fragment ions [m/z (relative)]: 50.93(12.15), 73.91(14.60), 74.91(24.68), 76.91(13.87), 110.86(46.02), 112.87(16.19), 138.86(100), 139.89(71.49), 140.89(35.80), 141.87(22.27).

Mixture: 1a, 2a, 3a, 4a, 5a

$^1\text{H-NMR}$ δ (ppm): vw 1.89 (s, 2H, **1a**), 3.85 (s, 2H, **1a**), 4.79 (s, 2H, **2a**), vw 6.51 (s, H, **3a**), 8.42 (s, H, **2a**), 9.94 (s, H, **5a**), 10.15 (s, 2H, **3a**), 7.15-7.58 (m, H-aromatics); $^{13}\text{C NMR}$ (500mHz, CDCl_3 , δ ppm, J Hz): 46.3 (CH_2 -**1a**), 65.01 (CH_2 -**2a**), 161.93 (CH -**2a**), 150.53 (CH - **3a**), 192 (CH - **5a**), 118.98 (Cq - **4a**), 126.5, 126.92, 127.1, 127.25, 128.15, 128.3, 128.5, 129, 129.2, 129.8, 130.95, 130.7, 132.1, 132.78, 134.2 (CH- aromatics, **1a**, **4a**, **5a**, **3a**, **2a**), 112.45, 135.18, 136.1, 136.9, 139.2, 143.5 (Cq- aromatics **1a**, **4a**, **5a**, **3a**, **2a**)

Mixture: 1b, 3b, 4b, 5b

$^1\text{H-NMR}$ δ (ppm):

vw 1.89 (s, 2H, **1b**), 2.27 (s, 3H, **3b**), 2.29 (s, 3H, **1b**), 2.31 (s, 3H, **4b**), 2.39 (s, 3H, **5b**), vw 6.51 (s, H, **3b**), 7.77 ; 7.2 (d, d, 4H, **5b**), 7.54; 7.06 (d, d, 4H, **3b**), 7.5; 7.2 (d, d, 4H, **4b**), 7.2; 6.96 (d, d, 4H, **1b**), 9.89 (s, H, **5b**), 10.15 (s, H, **3b**); ^{13}C NMR δ (ppm): 21, 21.27, 21.7 ($\text{CH}_3 - \mathbf{1b, 3b, 4b, 5b}$), 46.3 ($\text{CH}_2 - \mathbf{1b}$), 118.02 ($\text{Cq} - \mathbf{4b}$), 109.51, 126.75, 127.1, 129.1, 129.4, 129.9, 130.09, 132.09 ($\text{Cq} - \text{aromatics, } \mathbf{1b, 3b, 4b, 5b}$), 150.53 ($\text{CH}_2 - \mathbf{3b}$), 190.8 ($\text{CH}_2, \mathbf{5b}$)

Mixture: 1c, 3c, 4c

^1H -NMR δ (ppm):
vw 2.1 (s, 2H, **1c**), 3.72 (s, 3H, **3c**), 4.00 (s, 3H, **4c**), 4.17 (s, 3H, **1c**), 3.87 (s, 2H, **1c**), vw 6.51 (s, H, **3c**), 7.55-6.7 (m, **1c, 3c, 4c**), 10.34 (s, H, **3c**); ^{13}C NMR δ (ppm): 42.3 ($\text{CH}_2 - \mathbf{1c}$), 55.09, 55.2, 56.84, ($\text{CH}_3 - \mathbf{1c, 3c, 4c}$), 116.1 ($\text{Cq} - \mathbf{4c}$), 99.42, 120.4, 132.1, 152.1, 157.99, 163.61 ($\text{Cq} - \text{aromatics, } \mathbf{1c, 3c, 4c}$), 107.79, 110.2, 111.1, 120.77, 120.8, 121.8, 127, 128.3, 128.37, 130.8, 133.33, 134 ($\text{CH- aromatics, } \mathbf{1c, 3c, 4c}$), 155.62 ($\text{CH-}\mathbf{3c}$)

Mixture: 1d, 3d, 4d, 5d

^1H -NMR δ (ppm):
vw 1.89 (s, 2H, **1d**), 3.63 (s, 3H, **3d**), 3.68 (s, 3H, **1d**), 3.74 (s, 3H, **4d**), 3.77 (s, 3H, **5d**), 3.85 (s, 2H, **1d**), vw 6.51 (s, H, **3d**), 7.8; 6.96 (d,d, 4H, **5d**) 7.43, 6.86 (d, d, 4H, **4d**), 7.28, 7.23 (d, d, 4H, **1d**) 7.1; 6.75 (d, d, 4H, **3d**), 9.88 (s, H, **5d**), 10.15 (s, 2H, **3d**); ^{13}C NMR δ (ppm): 46.3 ($\text{CH}_2 - \mathbf{1d}$), 54.3, 55, 55.3, 55.4 ($\text{CH}_3 - \mathbf{3d, 4d, 1d, 5d}$), 118.49 ($\text{Cq} - \mathbf{4d}$), 113.9, 114.1, 114.3, 115.1, 125.6, 128.2, 131.6, 134.1 (CH, aromatics), 105.76, 129.65, 130.3, 135.85, 158.79, 161.36, 164.25, 164.7 ($\text{Cq aromatics, } \mathbf{3d, 4d, 1d, 5d}$), 150.53 ($\text{CH-}\mathbf{3d}$), 190.4 ($\text{CH-}\mathbf{5d}$)

Mixture: 1e, 2e, 4e, 5e

^1H -NMR δ (ppm):
vw 3.85 (s, 4H, **2e**), vw 5.1 (s, H, **2e**), vw 6.51 (s, H, **1e**), 7.15, 7.25 (d,d, 8H, **2e**), 7.25, 7.43 (d, d, 4H, **1e**), 7.38, 7.48 (d, d, 2H, **4e**), 7.45, 7.75 (d, d, 2H, **5e**), 9.8 (s, H, **5e**), 10.15 (s, H, **1e**); ^{13}C NMR δ (ppm): 46.3 ($\text{CH}_2 - \mathbf{1e}$), 54.7 ($\text{CH}_2 - \mathbf{2e}$), 114 ($\text{Cq-}\mathbf{4e}$) 108, 114, 131.03, 131.71, 134.9, 135.61, 139.23, 140.18, 140.8, ($\text{Cq aromatics, } \mathbf{1e, 2e, 4e, 5e}$), 127.1, 127.86, 128.4, 128.86, 129.02, 129.4, 130.8, 131.15 ($\text{CH- aromatics, } \mathbf{1e, 2e, 4e, 5e}$), 119.06 ($\text{CH} - \mathbf{5e}$)

^1H -NMR : chemical shift (δ ppm), J (Hz), multiplicity, number of protons

^{13}C NMR : chemical shift (δ ppm), numbering

vw – very weak peak,