Supporting Information 2

A new MCM-41 supported HPF₆ catalyst for the library synthesis of highly substituted 1,4-dihydropyridines and oxidation to pyridines: report of one-dimensional packing towards LMSOMs and studies on their photophysical properties

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General synthetic procedure for the preparation of 6a-6s

All the reactions were carried out in a round bottom flask equipped with a magnetic stirrer. In a typical reaction a solution of ketone (1 mmol), 1,3-diketoneone (1 mmol), aldehyde (1 mmol), and ammonium carbonate (1.2 mmol) in water (2 ml) were stirred at room temperature till completion using 2 mmol 30% aqueous H₂O₂ in presence of 40 mg of silica-HPF₆ catalyst. The completion of the reaction was indicated by the disappearance of the starting material in thin layer chromatography. After completion of the reaction the crude product was taken in dichloromethane and filtered to separate the products as filtrate from the catalyst (residue). The solvent was evaporated in rotary evaporator and the crude product was further purified by silica gel column chromatography (15% ethyl acetate/85% petroleum ether). The products were characterized by IR, ¹H NMR, ¹³C NMR, CHN and X-ray single crystal analysis. The spectral and analytical data of all the novel fully oxidized pyridine compounds are given below.
Spectroscopic characterization for 6a-6s

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\text{7,8-dihydro-2-(4-methoxyphenyl)-7,7-dimethyl-4-(4-nitrophenyl)quinolin-5(6H)-one (6a):}
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White solid, mp 182-184 °C (CH2Cl2 + EtOAc, equal volumes); IR ν \text{max} (KBr) 3430, 3050, 2957, 2872, 1686, 1573, and 1530 cm\(^{-1}\); \(^1\)H NMR (300 MHz, CDCl\(_3\)) δ: 8.22 (2H, d, J = 8.7 Hz), 8.01 (2H, d, J = 8.7 Hz), 7.36-7.34 (3H, m), 6.95 (2H, d, J = 9.0 Hz), 3.81 (3H, s), 3.18 (2H, s), 2.47 (2H, s), 1.10 (6H, s); \(^{13}\)C NMR (75 MHz, CDCl\(_3\)) δ: 196.6, 163.2, 162.3, 158.7, 150.9, 147.5, 146.9, 129.7, 128.7, 123.3, 123.0, 120.9, 114.6, 55.5, 53.4, 46.5, 32.6, 28.2; Anal. Caled for C\(_{24}\)H\(_{22}\)N\(_2\)O\(_4\): C, 71.63; H, 5.51; N, 6.96. Found C, 71.92; H, 5.60; N, 7.01.

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\text{7,8-dihydro-2-(4-methoxyphenyl)-7,7-dimethyl-4-(3-nitrophenyl)quinolin-5(6H)-one (6b):}
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White solid, mp 156-158 °C (CH2Cl2 + EtOAc, equal volumes); IR ν \text{max} (KBr) 3432, 3052, 2957, 2872, 1686, 1576, and 1535 cm\(^{-1}\); \(^1\)H NMR (300 MHz, CDCl\(_3\)) δ: 8.14-8.03 (4H, m), 7.51 (2H, br s), 7.37 (1H, m), 6.92 (2H, br s), 3.83-3.75 (3H, m), 3.20 (2H, s), 2.46 (2H, s), 1.00 (6H, s); \(^{13}\)C NMR (75 MHz, CDCl\(_3\)) δ: 197.3, 163.7, 162.0, 159.3, 149.8, 148.0, 142.1, 140.8, 134.3, 131.0, 129.4, 128.8, 124.6, 124.4, 122.9, 122.7, 122.0, 120.8, 114.5, 114.1, 55.6, 53.4, 47.4, 32.6, 28.3; Anal. Caled for C\(_{24}\)H\(_{22}\)N\(_2\)O\(_4\): C, 71.63; H, 5.51; N, 6.96. Found C, 71.82; H, 5.45; N, 6.78.
2-(9H-fluoren-2-yl)-7,8-dihydro-4-(4-methoxyphenyl)-7,7-dimethylquinolin-5(6H)-one (6c):

Yellowish white solid, mp 140-142 °C (CH₂Cl₂ + EtOAc, equal volumes); IR \( \nu_{\text{max}} \) (KBr) 3430, 3050, 2957, 2872, 1686, 1573, and 1530 cm⁻¹; \(^1\)H NMR (300 MHz, CDCl₃) \( \delta \): 8.29 (1H, s), 8.08 (1H, d, \( J = 7.8 \) Hz), 7.85 (2H, q, \( J = 8.1 \) Hz), 7.57-7.55 (2H, m), 7.53 (1H, s), 7.41-7.36 (2H, m), 7.26 (2H, d, \( J = 8.4 \) Hz), 6.96 (2H, d, \( J = 8.4 \) Hz), 3.96 (2H, s), 3.85 (3H, s), 3.19 (2H, s), 2.55 (2H, s), 1.16 (6H, br s); \(^{13}\)C NMR (75 MHz, CDCl₃) \( \delta \): 197.8, 163.5, 159.6, 159.3, 152.2, 144.1, 143.9, 143.7, 141.1, 136.4, 132.5, 129.4, 127.4, 127.0, 126.5, 125.2, 124.2, 123.8, 122.0, 120.5, 120.2, 113.6, 55.3, 54.0, 47.7, 37.0, 32.6, 28.3; Anal. Calcd for C₃₁H₂₇NO₂: C, 83.57; H, 6.11; N, 3.14. Found C, 83.26; H, 6.01; N, 3.34.

4-(4-chlorophenyl)-2-(9H-fluoren-2-yl)-7,8-dihydro-7,7-dimethylquinolin-5(6H)-one (6d):

Yellowish white solid, mp 180-182 °C (CH₂Cl₂ + EtOAc, equal volumes); IR \( \nu_{\text{max}} \) (KBr) 3424, 3050, 2957, 2872, 1686, 1573, and 1530 cm⁻¹; \(^1\)H NMR (300 MHz, CDCl₃) \( \delta \): 8.32 (1H, s), 8.10 (1H, d, \( J = 7.8 \) Hz), 7.85 (2H, t, \( J = 8.1 \) Hz), 7.58 (1H, d, \( J = 6.9 \) Hz), 7.53 (1H, s), 7.44-7.33 (4H, m), 7.25 (2H, dd, \( J = 6.6 \) Hz and \( J = 1.8 \) Hz), 3.98 (2H, s), 3.23 (2H, s), 2.56 (2H, s), 1.19 (6H, br s); \(^{13}\)C NMR (75 MHz, CDCl₃) \( \delta \): 197.6, 163.6, 159.6, 150.9, 144.0, 143.9, 143.7, 140.9, 138.9, 136.3, 133.8, 129.2, 128.2, 127.4, 126.9,
126.4, 125.1, 124.0, 123.4, 121.4, 120.4, 120.1, 53.7, 47.7, 36.9, 32.5, 28.2; Anal. Calcd for C₃₀H₂₄ClNO: 
C, 80.08; H, 5.38; N, 3.11. Found C, 80.30; H, 5.38; N, 3.19.

**4-(4-bromophenyl)-2-(9H-fluoren-2-yl)-7,8-dihydro-7,7-dimethylquinolin-5(6H)-one (6e):**

Yellowish white solid, mp 192-194 °C (CH₂Cl₂ + EtOAc, equal volumes); IR $\nu_{\text{max}}$ (KBr) 3424, 3051, 2954, 2872, 1689, 1573, and 1526 cm⁻¹; $^1$H NMR (300 MHz, CDCl₃) $\delta$: 8.30 (1H, s), 8.09 (1H, d, J = 7.8 Hz), 7.86 (2H, t, J = 8.7 Hz), 7.58 (1H, d, J = 7.2 Hz), 7.53 (1H, s), 7.42-7.35 (4H, m), 7.23 (2H, d, J = 8.7 Hz), 3.98 (2H, s), 3.22 (2H, s), 2.55 (2H, s), 1.17 (6H, br s); $^{13}$C NMR (75 MHz, CDCl₃) $\delta$: 197.4, 163.4, 159.4, 144.2, 144.0, 140.1, 138.7, 134.1, 129.3, 128.4, 127.6, 127.0, 126.8, 125.2, 124.5, 123.7, 122.1, 120.6, 120.3, 53.8, 47.3, 37.1, 32.7, 28.3; Anal. Calcd for C₃₀H₂₄BrNO: C, 72.88; H, 4.89; N, 2.83. Found C, 72.90; H, 4.98; N, 2.99.

**2-(9H-fluoren-2-yl)-7,8-dihydro-7,7-dimethyl-4-(3-nitrophenyl)quinolin-5(6H)-one (6f):**

Yellowish white solid, mp 190-192 °C (CH₂Cl₂ + EtOAc, equal volumes); IR $\nu_{\text{max}}$ (KBr) 3432, 3059, 2946, 2872, 1680, 1574, 1524, and 1346 cm⁻¹; $^1$H NMR (300 MHz, CDCl₃) $\delta$: 8.32 (1H, s), 8.27 (1H, d, J = 8.1 Hz), 8.10 (1H, d, J = 8.1 Hz), 7.99 (1H, s), 7.85 (2H, t, J = 8.1 Hz), 7.67-7.57 (3H, m), 7.53 (1H, s), 7.41-7.35 (2H, m), 4.03 (2H, s), 3.25 (2H, s), 2.56 (2H, s), 1.19 (6H, br s); $^{13}$C NMR (75 MHz, CDCl₃) $\delta$: 197.6, 164.0, 158.2, 149.5, 148.7, 144.0, 143.4, 142.7, 140.8, 135.4, 134.2, 133.7, 129.0, 127.3, 127.0, 125.1,
124.0, 122.8, 122.0, 121.2, 120.2, 53.5, 47.7, 37.1, 32.6, 28.2; Anal. Calcd for C_{30}H_{24}N_{2}O_{3}: C, 78.24; H, 5.25; N, 6.08. Found C, 78.24; H, 5.25; N, 6.08.

4-(4-Bromo-phenyl)-3,7,7-trimethyl-2-phenyl-7,8-dihydro-6H-quinolin-5-one (6g):
White solid, mp 142-144 °C (CH_{2}Cl_{2} + EtOAc, equal volumes); IR \nu_{\text{max}} (KBr) 3421, 3061, 2954, 2872, 1689, 1569, and 1525 cm\(^{-1}\); \(^{1}\)H NMR (300 MHz, CDCl\(_3\)) \delta: 8.01-7.84 (7H, m), 7.40 (2H, d, J = 8.1 Hz), 3.56 (2H, s), 2.89 (2H, s), 2.36 (3H, s), 1.55 (6H, s); \(^{13}\)C NMR (75 MHz, CDCl\(_3\)) \delta: 197.8, 162.4, 159.7, 150.7, 139.7, 138.5, 131.7, 129.1, 128.9, 128.8, 128.4, 124.0, 121.3, 53.9, 47.0, 32.6, 28.3, 17.7; Anal. Calcd for C\(_{24}\)H\(_{22}\)BrNO: C, 68.58; H, 5.28; N, 3.33. Found C, 68.30; H, 5.28; N, 3.53.

4-(5,6,7,8-tetrahydro-3,7,7-trimethyl-5-oxo-2-phenylquinolin-4-yl)benzonitrile (6h):
Yellow solid, mp 152-154 °C (CH_{2}Cl_{2} + EtOAc, equal volumes); IR \nu_{\text{max}} (KBr) 3428, 3051, 2945, 2872, 1684, 1574, 1530, and 1345 cm\(^{-1}\); \(^{1}\)H NMR (300 MHz, CDCl\(_3\)) \delta: 7.72 (2H, d, J = 7.8 Hz), 7.52-7.42 (5H, m), 7.19 (2H, d, J = 7.8 Hz), 3.16 (2H, s), 2.44 (2H, s), 1.89 (3H, s), 1.11 (6H, s); \(^{13}\)C NMR (75 MHz, CDCl\(_3\)) \delta: 197.9, 162.9, 160.1, 149.6, 145.1, 139.7, 132.3, 129.0, 128.9, 128.5, 128.3, 128.1, 123.4, 118.9, 111.1, 53.7, 47.1, 28.2, 17.6; Anal. Calcd for C\(_{25}\)H\(_{24}\)N\(_{2}\)O: C, 81.94; H, 6.05; N, 7.64. Found C, 82.20; H, 6.15; N, 7.44.
3,7,7-Trimethyl-4-(4-nitro-phenyl)-2-phenyl-7,8-dihydro-6H-quinolin-5-one (6i):

Yellowish white solid, mp 172-174 °C (CH₂Cl₂ + EtOAc, equal volumes); IR ν max (KBr) 3434, 3044, 2946, 2871, 1680, 1574, 1530, and 1353 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.30 (2H, d, J = 8.7 Hz), 7.54-7.41 (5H, m), 7.25 (2H, d, J = 9.3 Hz), 3.15 (2H, s), 2.45 (2H, s), 1.90 (3H, s), 1.11 (6H, s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.5, 162.5, 159.9, 149.8, 147.1, 146.8, 138.8, 129.1, 129.0, 128.6, 128.4, 128.1, 123.8, 123.6, 53.6, 46.6, 32.6, 28.1, 17.5; Anal. Calcd for C₂₄H₂₂N₂O₃: C, 74.59; H, 5.74; N, 7.25. Found C, 74.59; H, 5.80; N, 7.45.

7,8-dihydro-3,7,7-trimethyl-4-(3-nitrophenyl)-2-phenylquinolin-5(6H)-one (6j):

Yellowish white solid, mp 162-164 °C (CH₂Cl₂ + EtOAc, equal volumes); IR ν max (KBr) 3427, 3051, 2945, 2872, 1680, 1574, 1530, and 1349 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.27 (1H, d, J = 8.4 Hz), 7.99 (1H, s), 7.64 (1H, t, J = 7.8 Hz), 7.56-7.44 (6H, m), 3.16 (2H, s), 2.48 (2H, s), 1.95 (3H, s), 1.13 (6H, br s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.6, 162.7, 160.0, 149.3, 148.5, 141.2, 139.1, 133.5, 129.4, 129.0, 128.8, 128.4, 123.8, 122.3, 53.7, 46.8, 32.6, 28.4, 28.1, 17.7; Anal. Calcd for C₂₄H₂₂N₂O₃: C, 74.59; H, 5.74; N, 7.25. Found C, 74.89; H, 5.84; N, 7.25.
2-(4-chlorophenyl)-7,8-dihydro-7,7-dimethyl-4-(4-nitrophenyl)quinolin-5(6H)-one (6k):

White solid, mp 186-188 °C (CH2Cl2 + EtOAc, equal volumes); IR νmax (KBr) 3430, 3048, 2946, 2871, 1680, 1574, 1530, and 1335 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.28 (2H, d, J = 8.4 Hz), 8.04 (2H, d, J = 8.4 Hz), 7.51-7.40 (5H, m), 3.22 (2H, s), 2.57 (2H, s), 1.17 (6H, s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.5, 164.0, 158.6, 147.4, 136.8, 136.0, 129.2, 129.1, 128.8, 128.7, 123.4, 120.7, 53.5, 47.6, 32.7, 28.3; Anal. Calcd for C₂₃H₁₉ClN₂O₃: C, 67.90; H, 4.71; N, 6.89. Found C, 68.04; H, 4.71; N, 6.71.

2-(4-chlorophenyl)-7,8-dihydro-7,7-dimethyl-4-(3-nitrophenyl)quinolin-5(6H)-one (6l):

Yellowish white solid, mp 152-154 °C (CH₂Cl₂ + EtOAc, equal volumes); IR νmax (KBr) 3424, 3051, 2954, 2872, 1681, 1572, and 1525 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.30-8.27 (1H, m), 8.15 (1H, s), 8.05 (2H, d, J = 8.4 Hz), 7.61-7.59 (2H, m), 7.50-7.47 (3H, m), 3.28 (2H, s), 2.56 (2H, s), 1.17 (6H, s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.5, 164.0, 158.6, 149.8, 148.0, 141.9, 136.9, 135.9, 134.1, 129.2, 128.9, 123.5, 122.9, 121.2, 53.6, 47.6, 32.7, 28.3; Anal. Calcd for C₂₃H₁₉ClN₂O₃: C, 67.90; H, 4.71; N, 6.89. Found C, 67.99; H, 4.81; N, 6.80.
7,8-dihydro-7,7-dimethyl-4-(3-nitrophenyl)-2-(4-nitrophenyl)quinolin-5(6H)-one (6m):
Yellowi solid, mp 182-184 °C (CH₂Cl₂ + EtOAc, equal volumes); IR νmax (KBr) 3427, 3051, 2945, 2872, 1680, 1574, 1530, and 1349 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.36-8.26 (5H, m), 8.15 (1H, s), 7.65-7.58 (3H, m), 3.26 (2H, s), 2.58 (2H, s), 1.19 (6H, s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.9, 164.7, 157.5, 150.4, 149.3, 148.5, 143.9, 143.8, 141.9, 134.9, 134.4, 130.7, 130.0, 129.5, 128.8, 125.7, 124.7, 124.5, 124.2, 123.4, 123.3, 123.0, 122.4, 54.0, 48.1, 33.1, 28.7; Anal. Calcd for C₂₃H₁₉N₃O₅: C, 66.18; H, 4.59; N, 10.07. Found C, 66.48; H, 4.59; N, 10.17.

4-(4-chlorophenyl)-7,8-dihydro-2-(4-methoxyphenyl)-7,7-dimethylquinolin-5(6H)-one (6n):
White solid, mp 122-124 °C (CH₂Cl₂ + EtOAc, equal volumes); IR νmax (KBr) 3424, 3058, 2945, 1680, 1574, 1530, 1346 and 1259 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.05 (2H, d, J = 9.0 Hz), 8.15 (1H, s), 7.40-7.37 (3H, m), 7.21 (2H, d, J = 8.4 Hz), 7.00 (2H, d, J = 8.7 Hz), 3.87 (3H, s), 3.18 (2H, s), 2.53 (2H, s), 1.56 (6H, s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.6, 163.6, 161.6, 160.0, 151.1, 138.9, 133.9, 130.9, 129.5, 129.3, 129.1, 128.3, 123.1, 120.9, 114.4, 113.9, 55.5, 53.8, 47.6, 32.6, 28.3; Anal. Calcd for C₂₄H₂₂ClNO₂: C, 73.56; H, 5.66; N, 3.57. Found C, 73.85; H, 5.66; N, 3.80.

7,8-dihydro-7,7-dimethyl-4-(3-nitrophenyl)-2-(thiophen-2-yl)quinolin-5(6H)-one (6o):
Yellowish white solid, mp 202-204 °C (CH₂Cl₂ + EtOAc, equal volumes); IR νmax (KBr) 3427, 3051, 2945, 2872, 1680, 1574, 1530, and 1346 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.28-8.24 (1H, m), 8.13 (1H, s), 7.69 (1H, d, J = 3.6 Hz), 7.61-7.51 (3H, m), 7.37 (1H, s), 7.14 (1H, t, J = 4.8 Hz), 3.16 (2H, s), 2.51 (2H, s),
1.15 (6H, br s); Anal. Calcd for C₂₁H₁₈N₂O₃S: C, 66.65; H, 4.79; N, 7.40. Found C, 67.01; H, 4.78; N, 7.22.

7,8-dihydro-7,7-dimethyl-4-(4-nitrophenyl)-2-(thiophen-2-yl)quinolin-5(6H)-one (6p):

Yellowish white solid, mp 198-200 °C (CH₂Cl₂ + EtOAc, equal volumes); IR νₘₐₓ (KBr) 3423, 3059, 2945, 2872, 1680, 1574, 1530, 1346 and 1259 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.27 (2H, d, J = 8.7 Hz), 7.69 (1H, d, J = 3.3 Hz), 7.52 (1H, d, J = 4.8 Hz), 7.40 (2H, d, J = 8.7 Hz), 7.34 (1H, s), 7.14 (1H, t, J = 4.4 Hz), 3.16 (2H, s), 2.51 (2H, s), 1.15 (6H, br s); Anal. Calcd for C₂₁H₁₈N₂O₃S: C, 66.65; H, 4.94; N, 7.41.

4-(4-chlorophenyl)-7,8-dihydro-7,7-dimethyl-2-(thiophen-2-yl)quinolin-5(6H)-one (6q):

Yellowish white solid, mp 172-174 °C (CH₂Cl₂ + EtOAc, equal volumes); IR νₘₐₓ (KBr) 3432, 3050, 2957, 2872, 1686, 1573, 1523 and 1347 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 7.66 (1H, d, J = 5.1 Hz), 7.48 (1H, d, J = 5.1 Hz), 7.37 (2H, d, J = 9.0, Hz), 7.33 (1H, s), 7.18 (2H, d, J = 9.0 Hz), 7.11 (1H, t, J = 5.0 Hz), 3.12 (2H, s), 2.49 (2H, s), 1.10 (6H, br s); ¹³C NMR (75 MHz, CDCl₃) δ: 197.3,163.9, 154.4, 150.9, 143.5, 138.6, 133.9,129.9, 129.1, 128.4, 128.2, 126.8, 123.4, 119.7, 53.6, 47.4, 32.5, 28.2; Anal. Calcd for C₂₁H₁₈ClNOS: C, 68.56; H, 4.93; N, 3.81. Found C, 68.85; H, 5.01; N, 4.01.
4-(4-bromophenyl)-7,8-dihydro-7,7-dimethyl-2-(thiophen-2-yl)quinolin-5(6H)-one (6r):

Yellowish white solid, mp 182-184 °C (CH₂Cl₂ + EtOAc, equal volumes); IR ν_max (KBr) 3427, 3050, 2957, 2872, 1686, 1574, 1530, 1346 and 1269 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 7.62 (1H, d, J = 3.2 Hz), 7.47 (2H, d, J = 9.6 Hz), 7.43 (1H, d, J = 4.7 Hz), 7.28 (1H, s), 7.08-7.04 (3H, m), 3.08 (2H, s), 2.44 (2H, s), 1.08 (6H, b rs); Anal. Calcd for C₂₁H₁₈BrNOS: C, 61.17; H, 4.40; N, 3.40. Found C, 61.17; H, 4.51; N, 3.59.

1,2,3,4,5,6,7,8-octahydro-9-(3-nitrophenyl)acridine (6s):

Yellow liquid; IR ν_max (Neat) 3432, 3061, 2872, 1684, 1573, 1523 and 1333 cm⁻¹; ¹H NMR (300 MHz, CDCl₃) δ: 8.34 (1H, s), 8.21 (1H, d, J = 8.1 Hz), 7.80 (1H, d, J = 7.2, Hz), 7.59 (1H, t, J = 8.1 Hz), 2.94 (2H, s), 2.66-2.58 (7H, m), 1.87-1.82 (7H, m), 1.72-1.64 (2H, m); ¹³C NMR (75 MHz, CDCl₃) δ: 153.5, 152.5, 148.1, 146.4, 142.1, 135.4, 130.7, 129.1, 128.0, 124.3, 122.6, 32.6, 27.9, 26.4, 25.5, 22.7, 22.5, 22.3; Anal. Calcd for C₁₉H₂₃N₂O₂: C, 74.00; H, 6.54; N, 9.08. Found C, 74.30; H, 6.60; N, 9.19.
Copy of spectra for 6a-6s

[Diagram of spectra with chemical structures and data points]

196.43
16.63 158.70 150.94 147.49 133.11 121.18 77.42 76.97 74.33 66.48 55.45 46.50 44.65 33.63 28.19
Electronic Supplementary Material (ESI) for Green Chemistry
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