Supplementary Information

A comparative study between Co- and $CoFe_2O_4$-NPs catalytic activity in C–N and C–O bond formation: synthesis of benzimidazoles and benzoxazoles from o-haloanilides

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1. Characterization of Catalysts:

Co-NPs

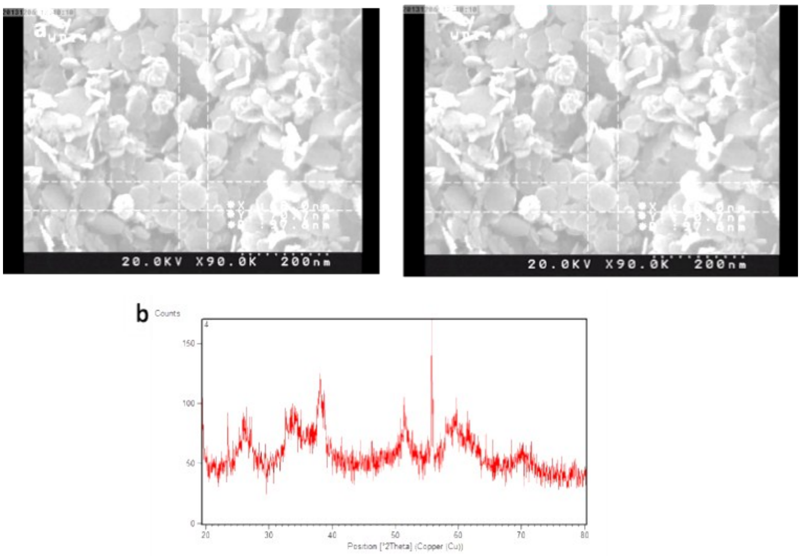


Figure 1s. The FE-SEM images and b: XRD patterns of Co-NPs

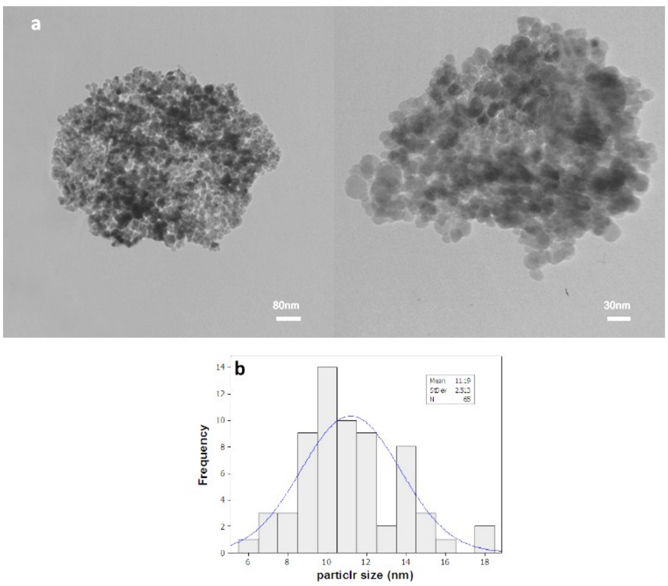
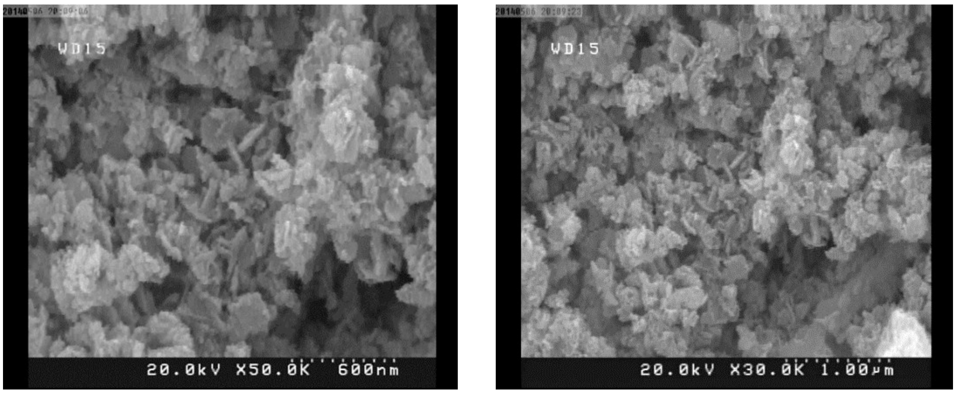


Figure 2s a: TEM images and b: particle size distribution results for Co-NPs

$CoFe_2O_4$-NPs:



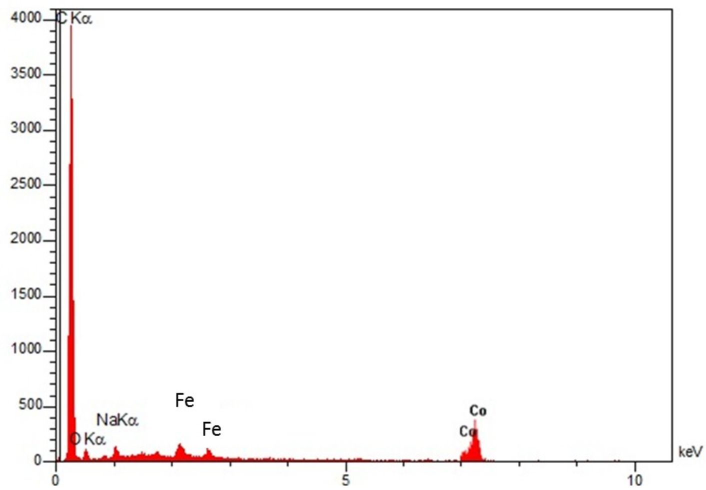


Figure 3s The FE-SEM images and SEM-EDX spectra of $CoFe_2O_4$-NPs.

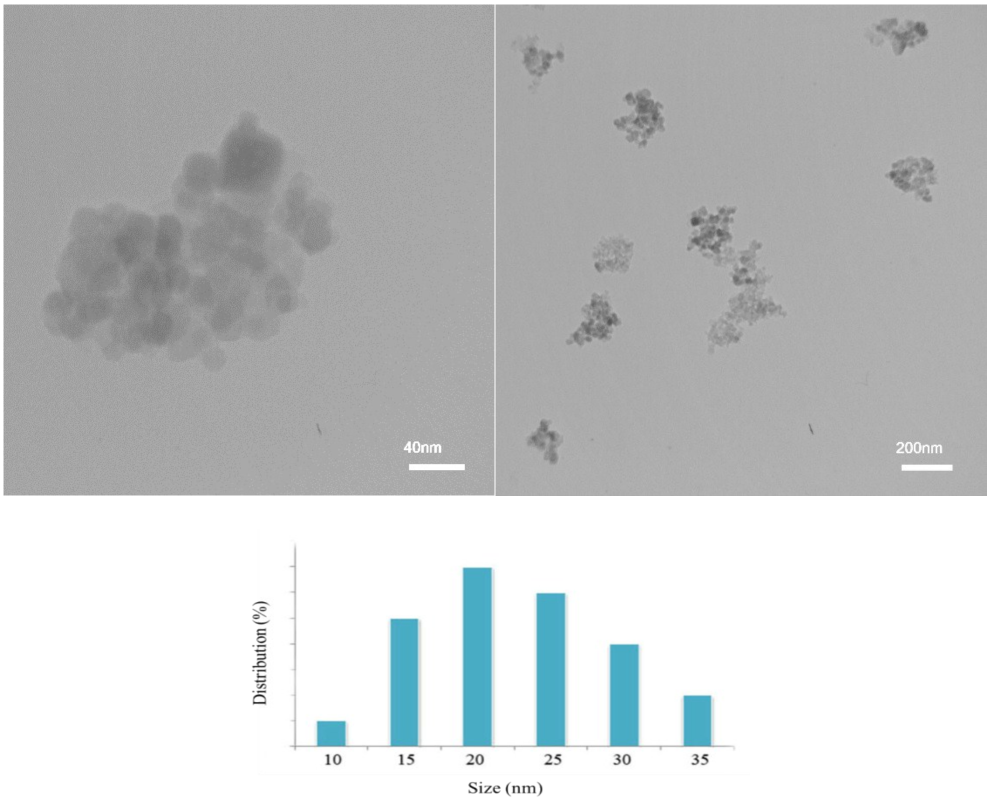


Figure 4s TEM images and particle size distribution results for $CoFe_2O_4$-NPs

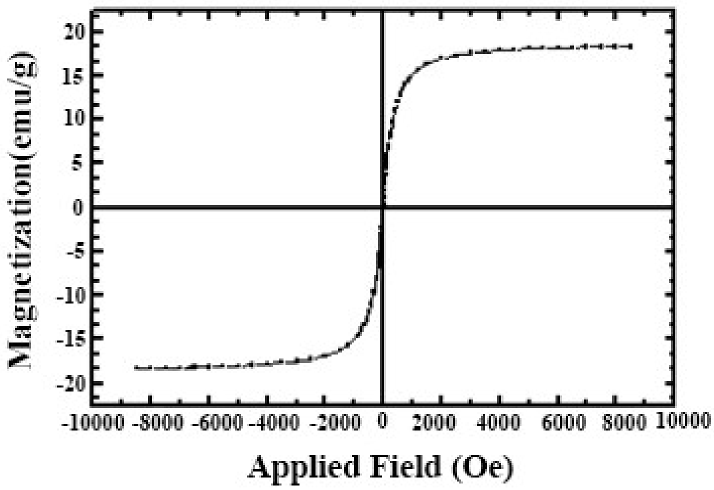


Figure 5s Room temperature magnetization curves of $CoFe_2O_4$-NPs

Table 1s. Recyclability of Co-NPs.

Run	Yield[a] (%)	Run	Yield[a] (%)
1	94	4	88
2	93	5	86
3	90	6	74

[a]Isolated yield.

Table 2s. Recyclability of $CoFe_2O_4$-NPs

Run	Yield[a] (%)	Run	Yield[a] (%)[a]
1	73	4	69
2	73	5	69
3	71	6	67

[a]Isolated yield.

2. Characterization of products

1 (**Table 2**) White solid; mp 101–106ºC; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.28-7.58 (m, 9H), 7; ^{13}C NMR (120 MHz, $CDCl_3$): δ 141.6, 139.9, 138.3, 134.4, 133.9, 129.7, 126.0, 121.4, 110.6, .FT-IR (neat) 3518, 3060, 2949, 2920, 2857, 1642, 1610, 1501, 1461, 1455, 1441, 1358, 1326, 1305, 1265, 1074, 1136, 1155, 1103 cm^{-1}.

2 (2) White solid; mp 93–94 ºC; ^{1}HNMR ($CDCl_3$, 400 MHz): δ 7.68–7.19 (m, 8H); ^{13}C NMR ($CDCl_3$, 120 MHz): δ 162.7, 150.1, 140.3, 134.7, 130.9, 128.4, 127.9, 126.8, 125.3, 123.9, 121.2; FT-IR (KBr) 3054, 2920, 2856, 1424, 1657, 1615, 1554, 1481, 1448, 1351, 1337, 1242, 1021, 1175, cm-1. Anal. Calcd (%) for $C_{13}H_8N_2O_3$: C, 60; H, 3; N, 17; found: C, 62.1; N, 16.8. [4]

3 (2) White solid; mp; 103–104 ºC ^{1}HNMR ($CDCl_3$, 400 MHz): δ 8.54–8.52 (m, 1H), 7.59-6.59 (m, 7H) ^{13}C NMR ($CDCl_3$, 120 MHz): δ 162.1, 150.7, 142.3, 136.7, 131.9, 128.9, 127.9, 126.8, 126.3,; FT-IR (KBr) 3058, 2921, 2856, 1428, 1647, 1605, 1524, 1431, 1458, 1361, 1327, 1252, 1021, 1165, cm^{-1}.

4 (2) White solid; mp 86–88 ºC; ^{1}HNMR ($CDCl_3$, 400 MHz): δ 7.94–7.07 (m, 8H), 2.33–2.23 (s, 3H); FT-IR (KBr) 3064, 2920, 2846, 1424, 1657, 1610, 1554, 1483, 1440, 1350, 1339, 1248, 1041, 1185, cm_{-1}.

5 (2) White solid; mp 156–157 ºC; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.89-7.10 (m, 8H); ^{13}C NMR (120 MHz, $CDCl_3$): δ 156.5, 151.2, 142.6, 133.4, 129.6, 126.8, 153.4, 143.9, 139.9, 138.3, 110.6, .FT-IR (neat) 3520, 3060, 2949, 2920, 2857, 1642, 1610, 1505, 1472, 1455, 1441, 1378, 1326, 1305, 1265, 1074, 1136, 1155, 1192 cm^{-1}.

6 (2) White solid; mp 126–128 ºC; ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.49-6.91 (m, 8H), 3.91 (s, 3 H), ^{13}C NMR (120 MHz, $CDCl_3$): δ 158.5, 152.2, 145.9, 142.6, 139.0, 138.9, 133.4, 129.6, 126.8, 93, .FT-IR (neat) 3420, 3050, 2949, 2920, 2857, 1662, 1610, 1505, 1472, 1450, 1449, 1368, 1328, 1315, 1265, 1172, 1136, 1165, 1074,cm^{-1}.

7 (2) White solid; yield 92%; mp 116–117 ºC; 1H NMR CDCl3, 400 MHz: δ 7.92–7.31 (m, 8H); ^{13}C NMR ($CDCl_3$, 120 MHz): δ 164.5, 156.8, 145.3, 134.4, 130.1, 127.6, 127.6, 124.2, 120,7, 110.3; FT-IR (KBr) 3430, 3281, 2967, 2924, 2855, 1651, 1459, 1437, 1411, 1346, 1242, 1175, 1021 cm^{-1}, Anal. Calcd (%) for $C_{13}H_{10}N_2O$: C, 74.3; H, 4.8; N, 13.3; found: C, 74.8; N, 13.9.3.

1 (Table 3) Yellow solid; mp 123–124 ºC, ^{1}H NMR (400 MHz, $CDCl_3$):δ 7.94-7.07 (m, 12H), 2.33-2.23 (s, 3H), ^{13}C NMR (100 MHz, $CDCl_3$): δ 153.6, 143.8, 139.4, 136.2, 130.2, 129.8, 128.0, 127.2, 124.6, 123.4, 121.3; FT-IR (KBr) 3521, 3053, 3012, 2985, 1658, 1590, 1490, 1472, 1459, 1377, 1332, 1311, 1284, 1245, 1178, 1081, 1032 cm^{-1}.

2 (3) Yellow solid; mp 117–119 ºC, [1]H NMR (400 MHz, $CDCl_3$):δ 7.63-7.33 (m, 12H), FT-IR (KBr) 3518, 3054, 3029, 2982, 1660, 1587, 1491, 1486, 1441, 1377, 1329, 1305, 1279, 1265, 1168, 1075, 1034 cm^{-1}.

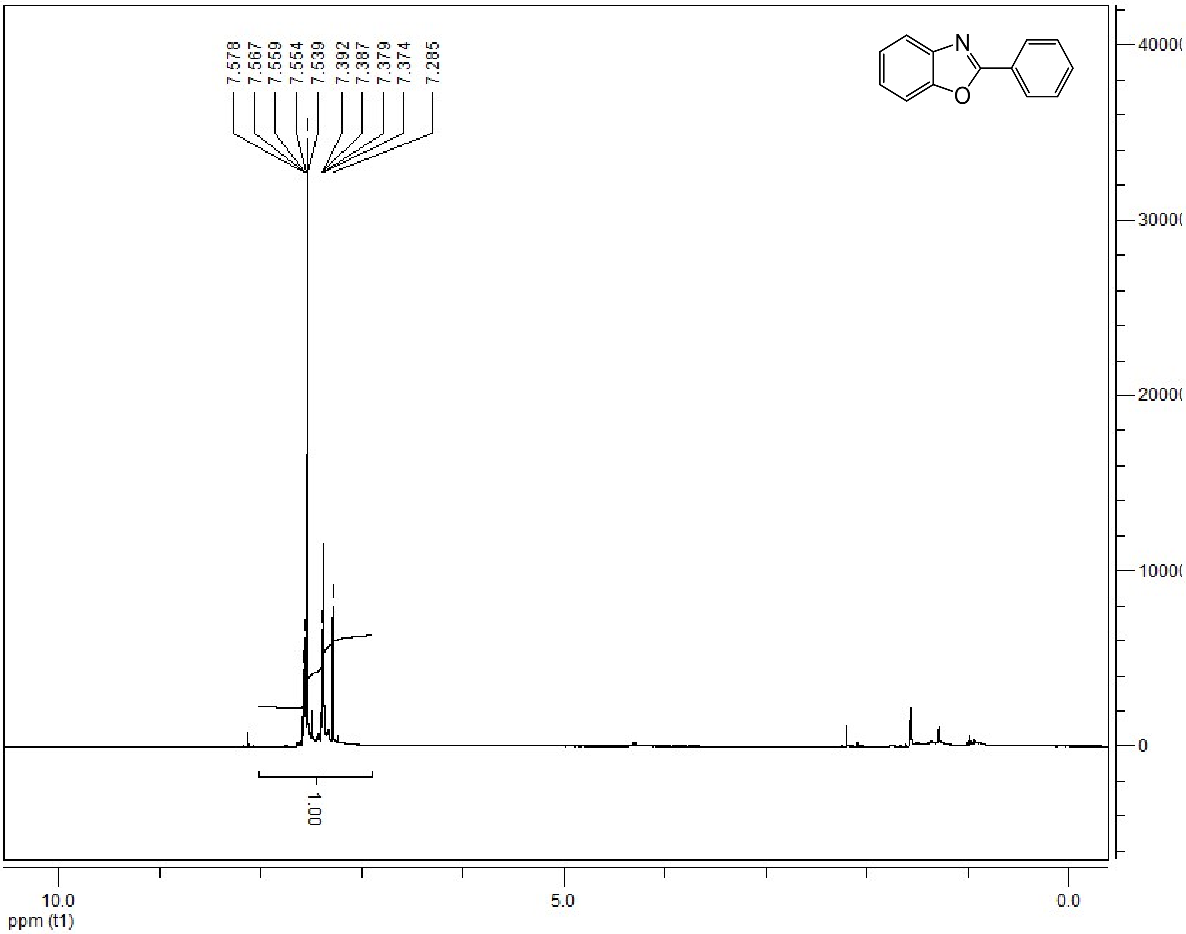
3 (3) Yellow solid; mp 115–116 ºC, [1]H NMR (400 MHz, CDCl3):δ 7.38-7.28 (m, 12 H), 3.88 (s, 3H); [13]C NMR (100 MHz, $CDCl_3$): δ 152.6, 143.2, 137.4, 137.2, 131.2, 129.3, 128.6, 127.5, 124.6, 123.8, 120.3; FT-IR (KBr) 3522, 3054, 3032, 2972, 1662, 1577, 1495, 1486, 1454, 1389, 1322, 1321, 1274, 1265, 1178, 1081, 1034 cm^{-1}.

4 (3) White solid; mp 119–120 ◦C; [1]H NMR (400 MHz, $CDCl_3$): δ 7.89–7.87 (m, 2H), 7.61(m, 2H), 7.35–7.22 (m, 2H), 7.02–6.99 (m, 4H), 3.88 (s, 3H) [13]C NMR (120 MHz, $CDCl_3$): δ 159.5, 152.6, 143.0, 137.7, 130.1, 129.9, 127.9, 127.4, 124.0, 123.8, 123.1, 118.9, 114.8, 110.3; FT-IR (KBr) 3543, 3062, 2957, 2839, 1606, 1546, 1514, 1472, 1458, 1442, 1353, 1325, 12285, 1210, 1186, 1074, 1028 cm^{-1}. Anal. Calcd (%) for $C_{19}H_{14}N_3O_3$: C, 68.68; H, 4.22; N, 12.65; found: C, 70.01; H, 4.35; N, 12.24.

5 (3) Yellow solid; mp 109–110 ºC, [1]H NMR (400 MHz, $CDCl_3$):δ 7.79-7.83(m, 2H), 7.28-7.22 (m, 4H), 7.01-7.09 (m, 2H), 6.84-6.91 (m, 2H), 6.79-6.76 (m, 2H); [13]C NMR (100 MHz, $CDCl_3$): δ 152.6, 143.2, 137.4, 137.2, 131.2, 129.3, 128.6, 127.5, 124.6, 123.8, 120.3; FT-IR (KBr) 3520, 3050, 3022, 2982, 1658, 1597, 1490, 1476, 1451, 1387, 1322, 1301, 1274, 1255, 1178, 1071, 1024 cm^{-1}. Anal. Calcd (%) for $C_{19}H_{14}N_2O_2$: C, 75.50; H, 4.63; N, 9.27; found: C, 74.89; H, 4.23; N, 9.28.

6 (3) Yellow liquid; [1]H NMR (400 MHz, $CDCl_3$): δ 7.82-7.86 (m, 2H), 7.20-7.16 (m, 4H), 7.05-7.09 (m, 2H), 6.74-6.82 (m, 2H), 6.75-6.72 (m, 2H). [13]C NMR (100 MHz, $CDCl_3$): δ 153.5, 145.3, 139.7, 135.4, 132.1, 130.4, 129.8, 128.7, 127.3, 123.4, 129.9, 120.0, 110.9. FT-IR (neat) 3451, 3058, 2932, 1649, 1606, 1554, 1447, 1401.1, 1372.1, 1261, 1019 cm^{-1}. Anal. Calcd (%) for $C_{19}H_{15}N_3O$: C, 75.75; H, 5.00; N, 13.95; found: C, 74.60; H, 5.04; N, 14.06.

1 (2)



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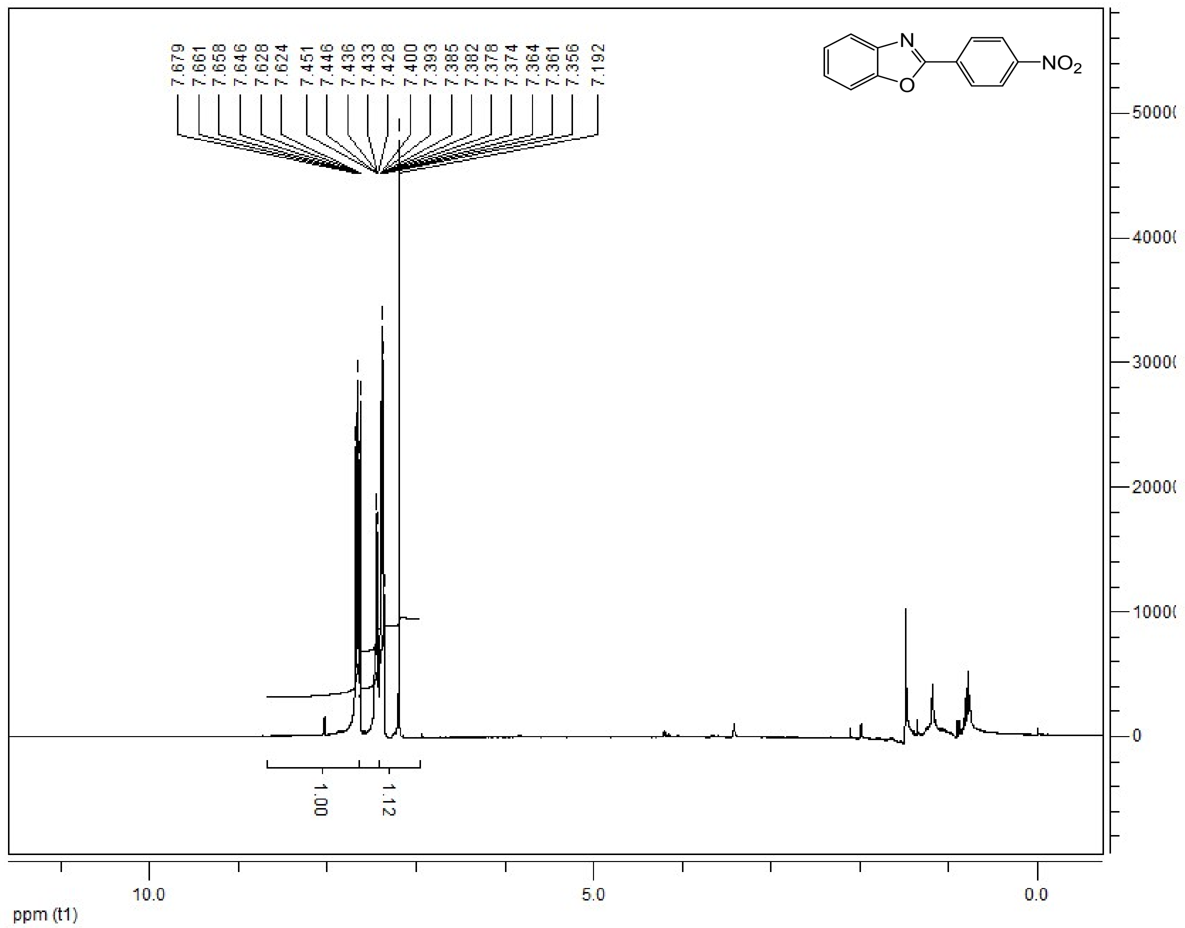

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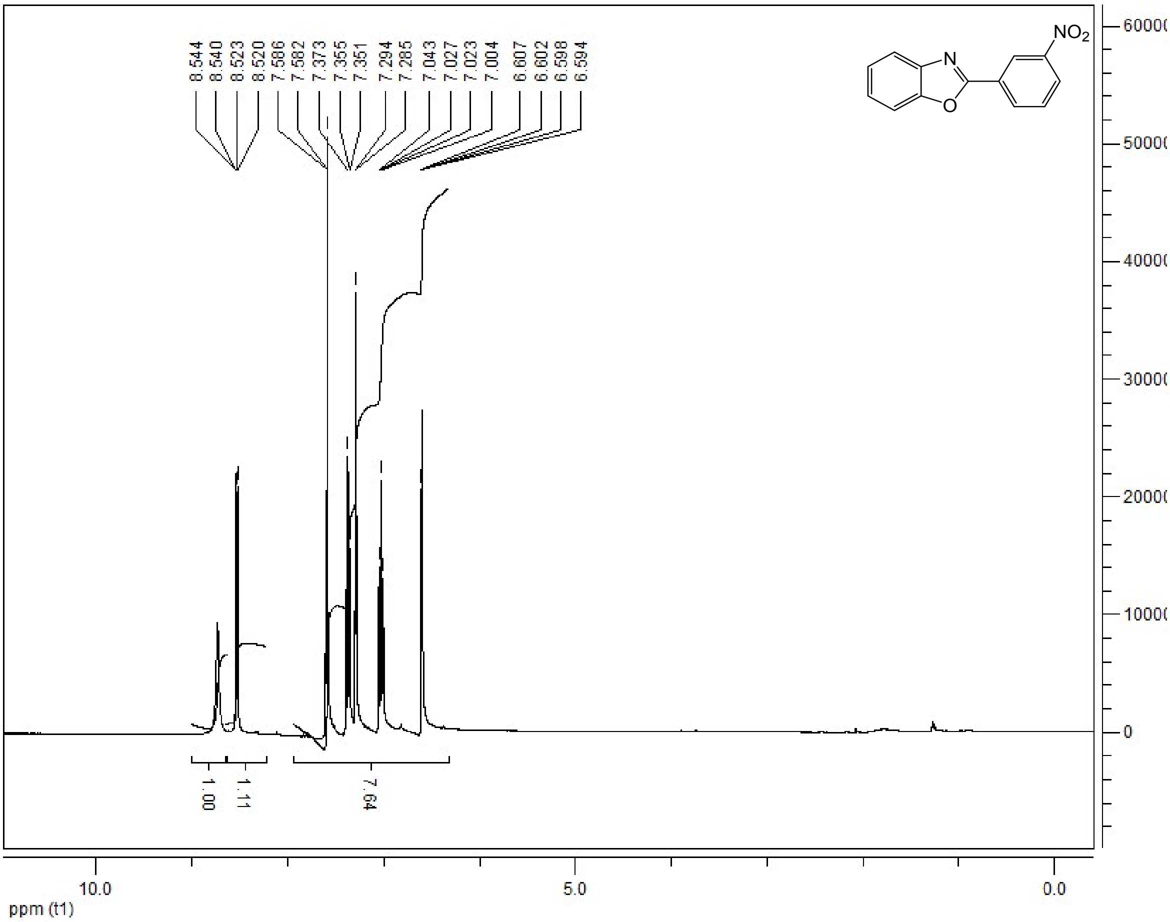

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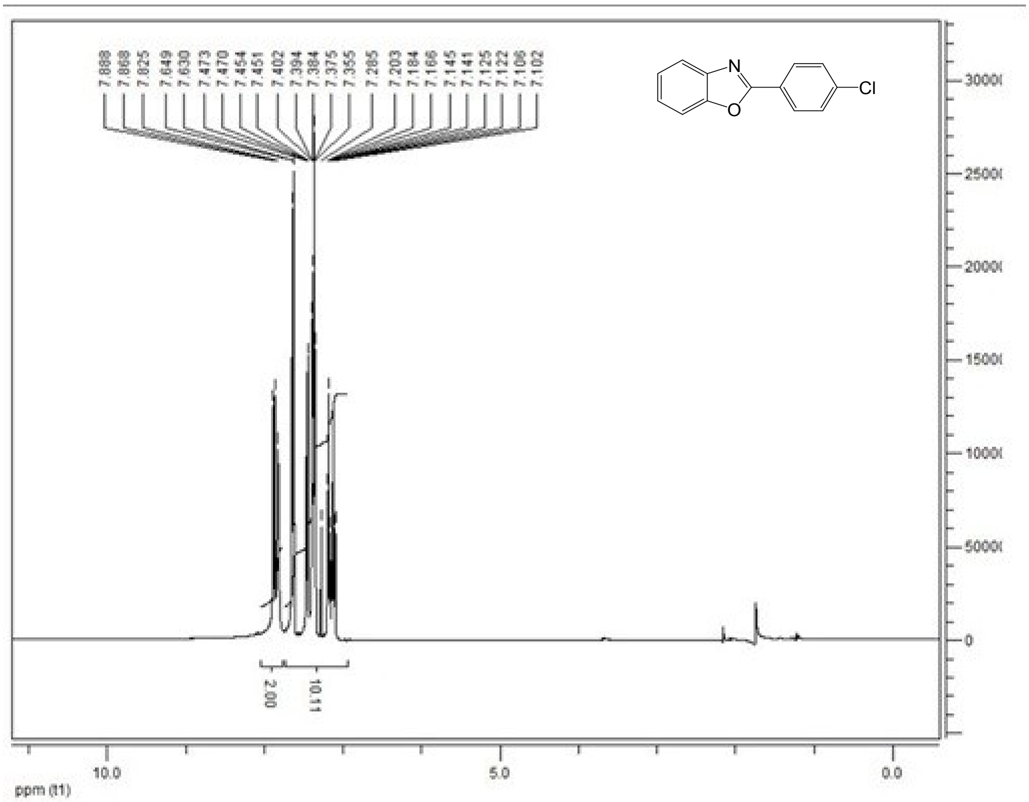
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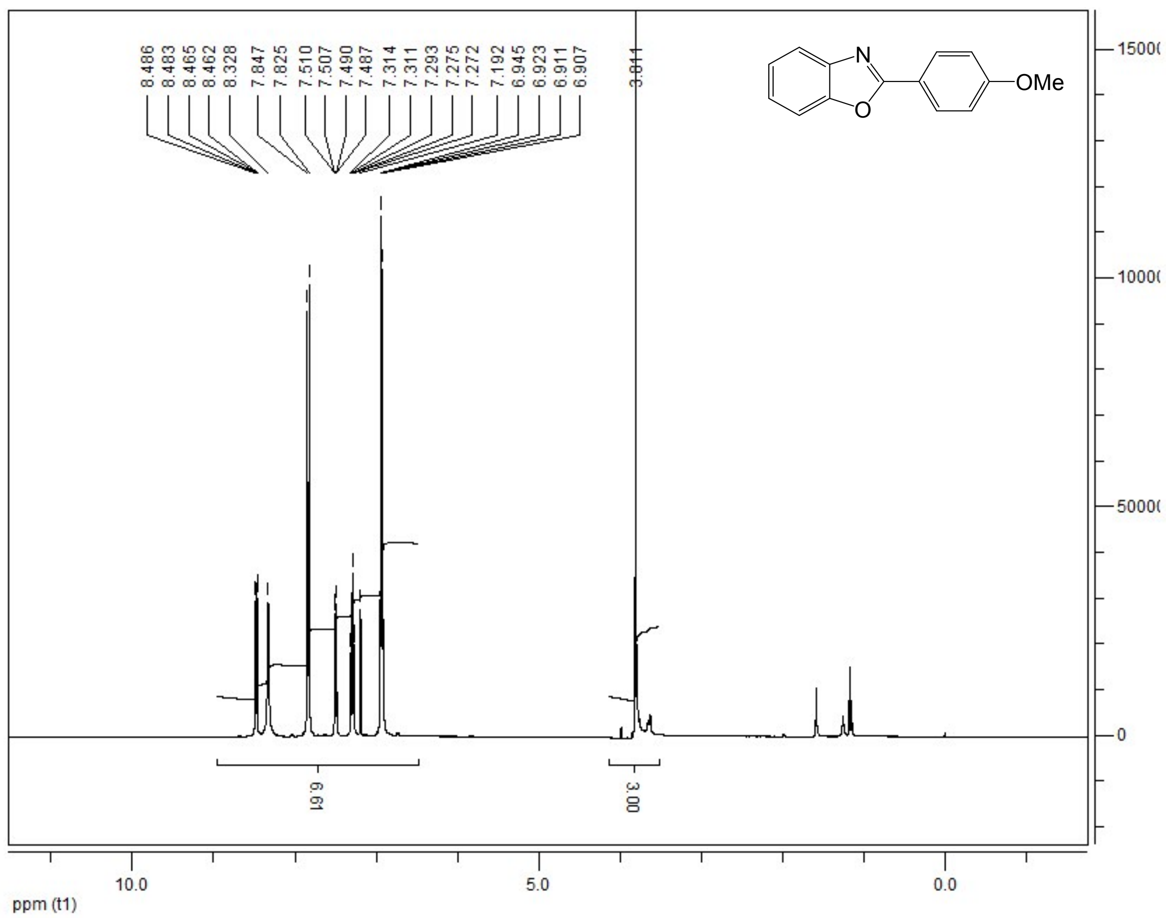

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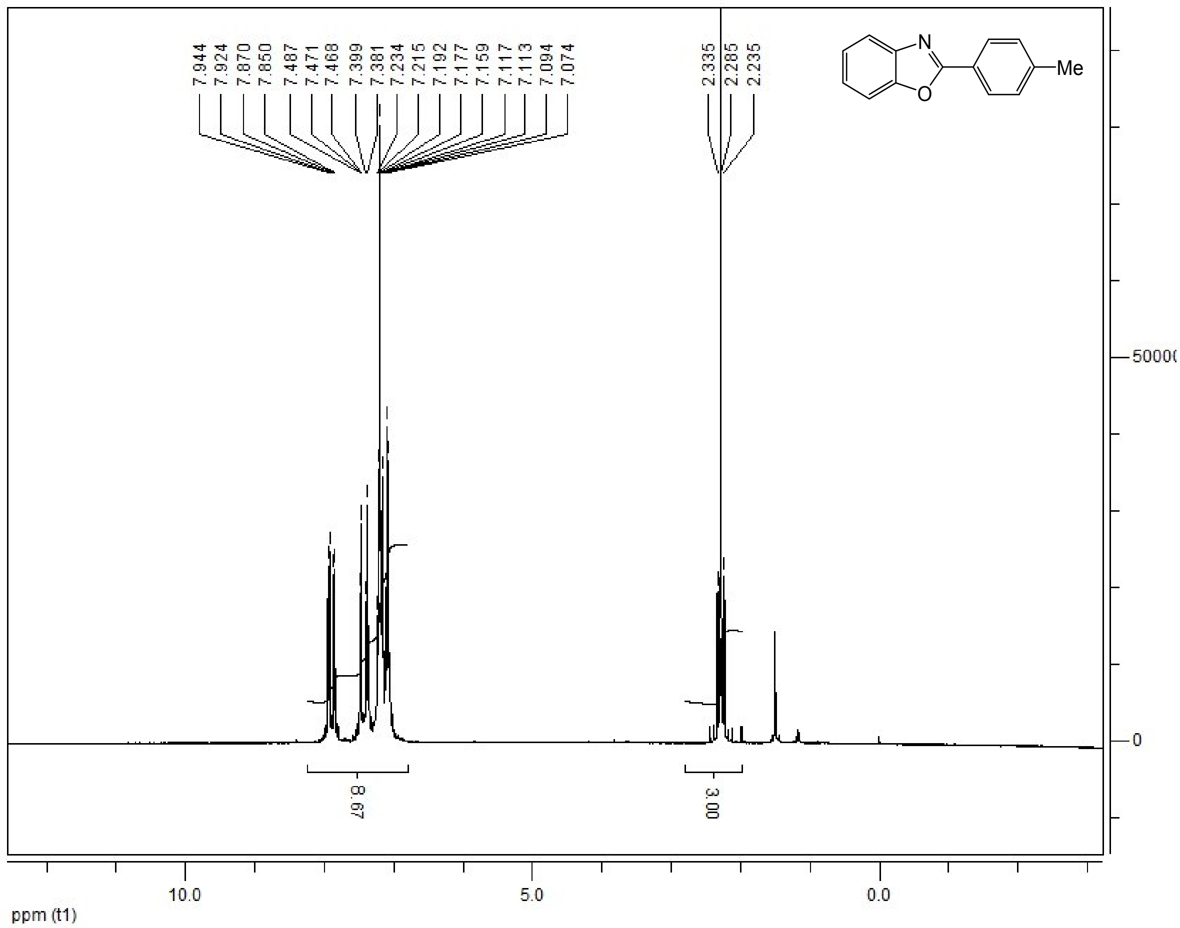

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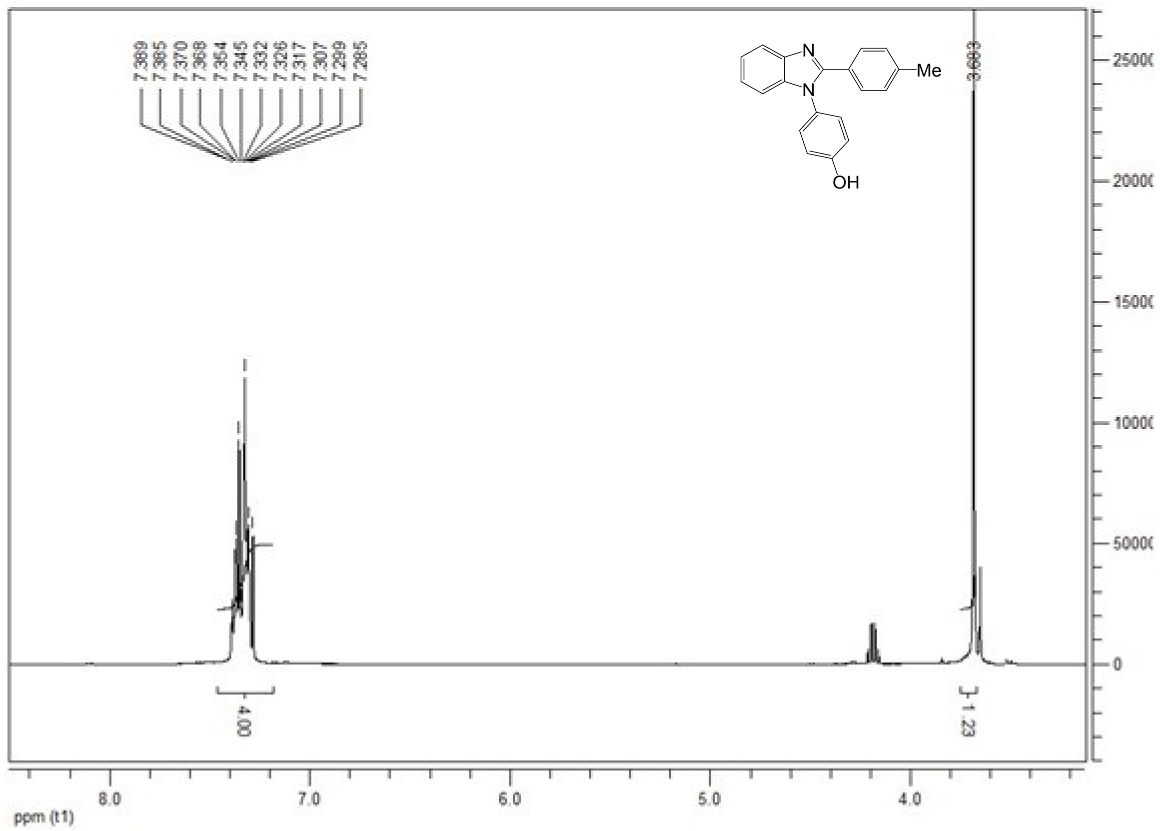
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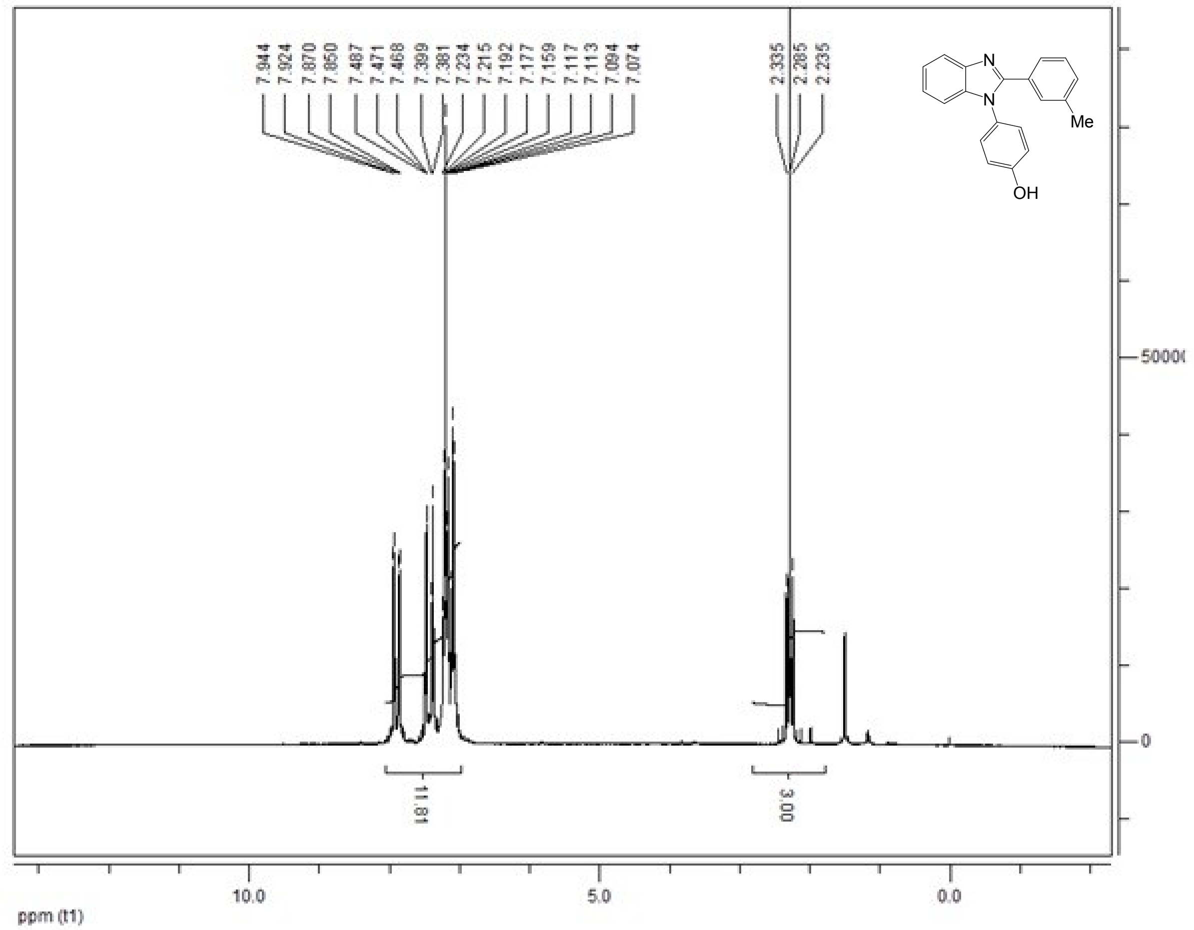

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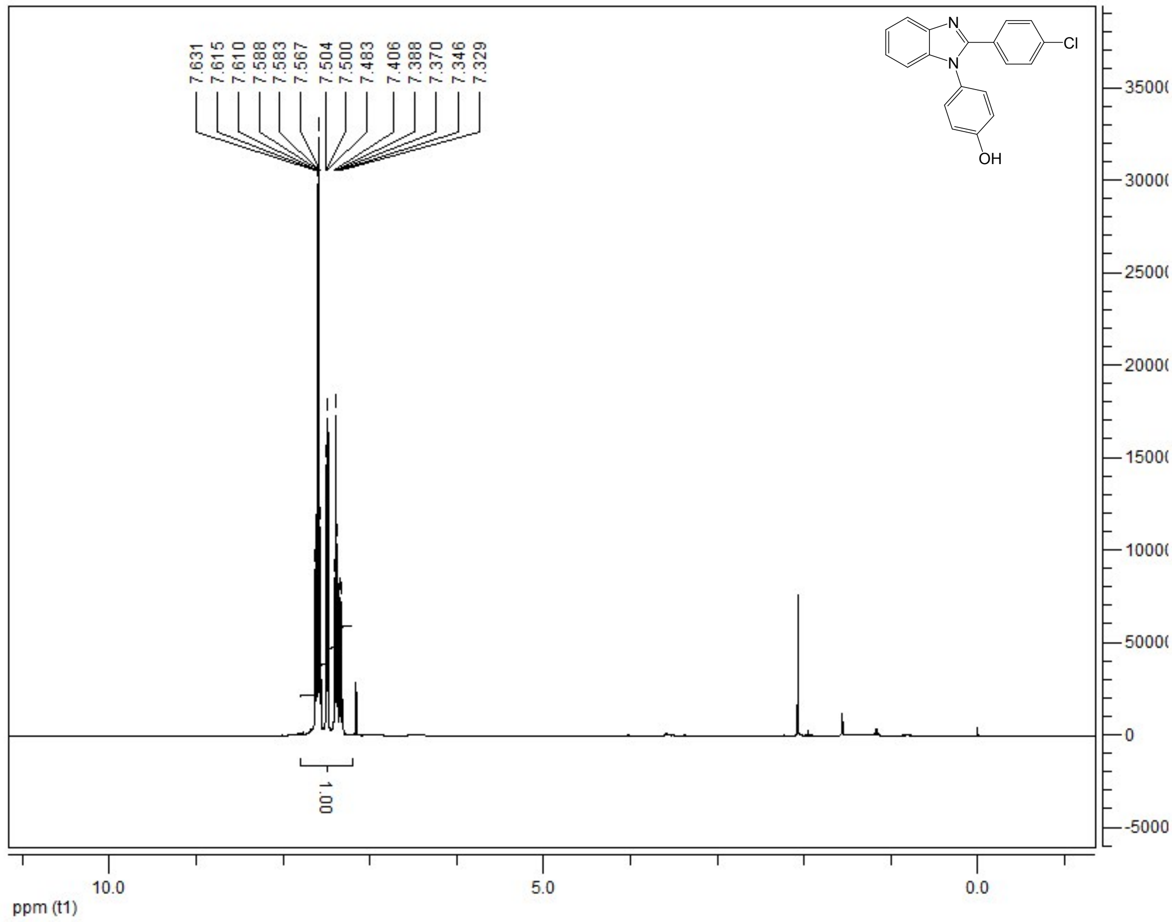

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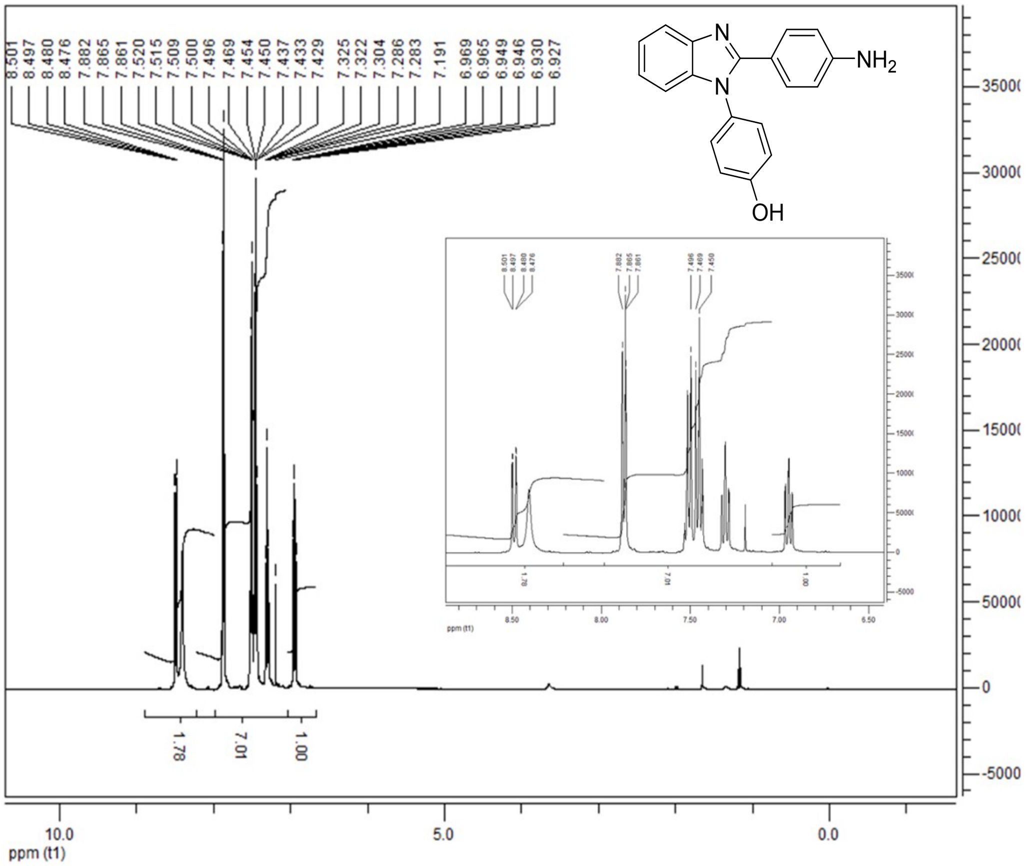
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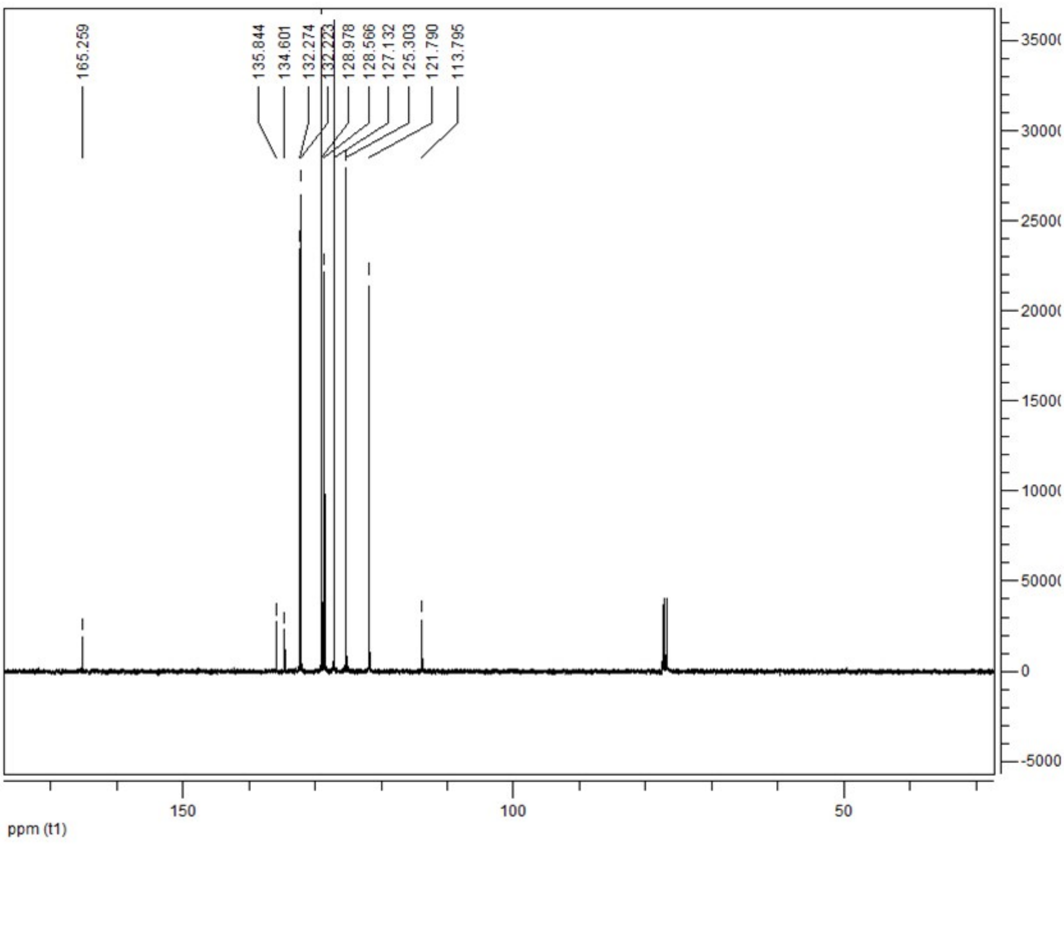
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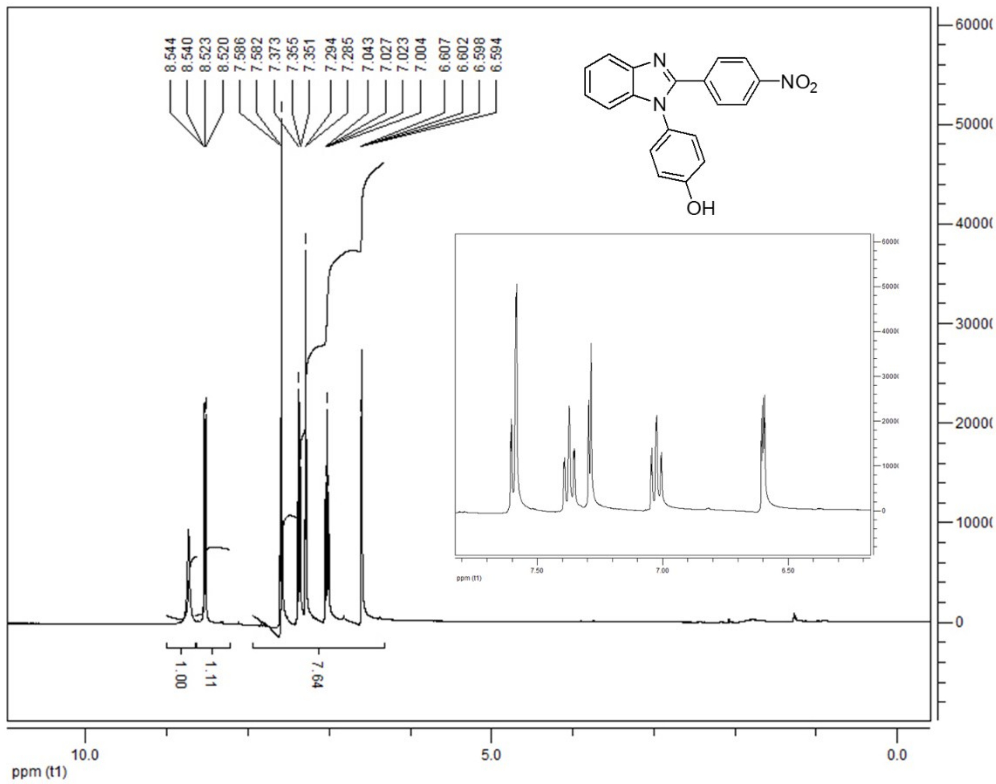
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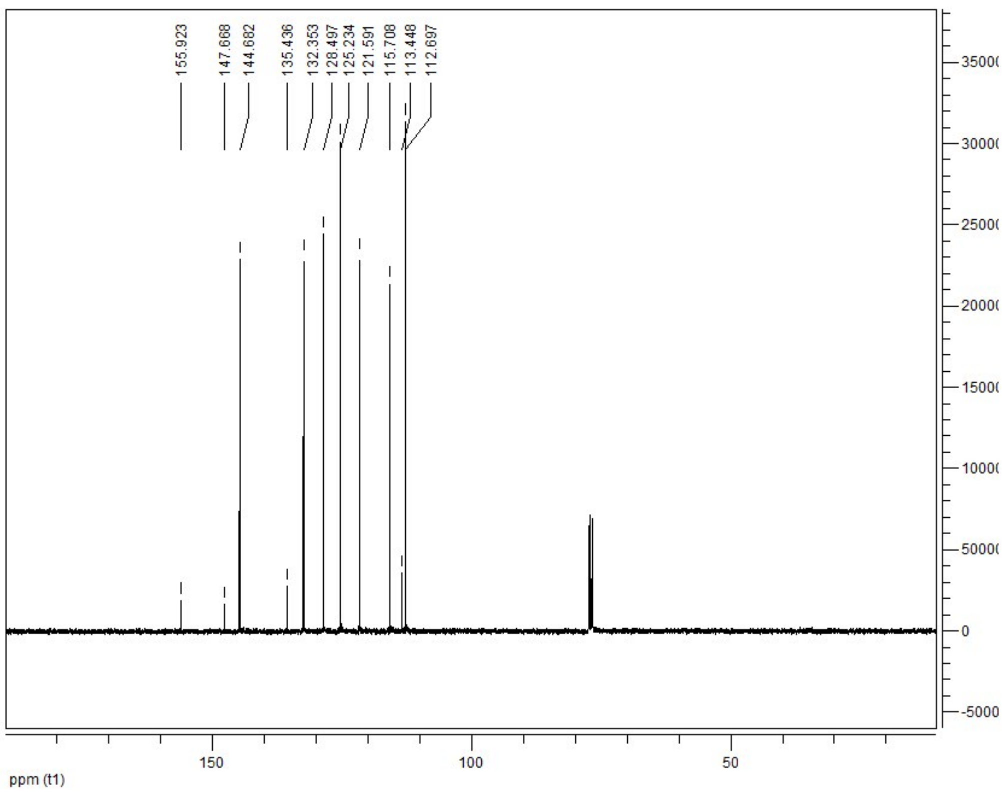
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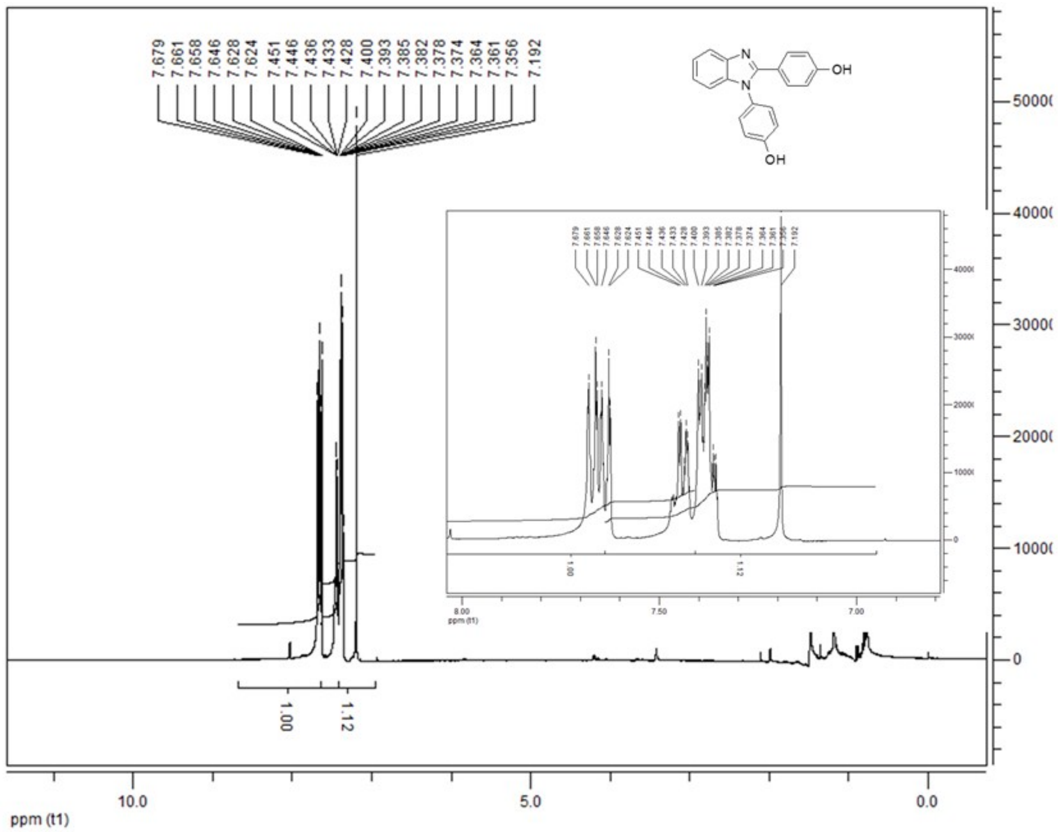
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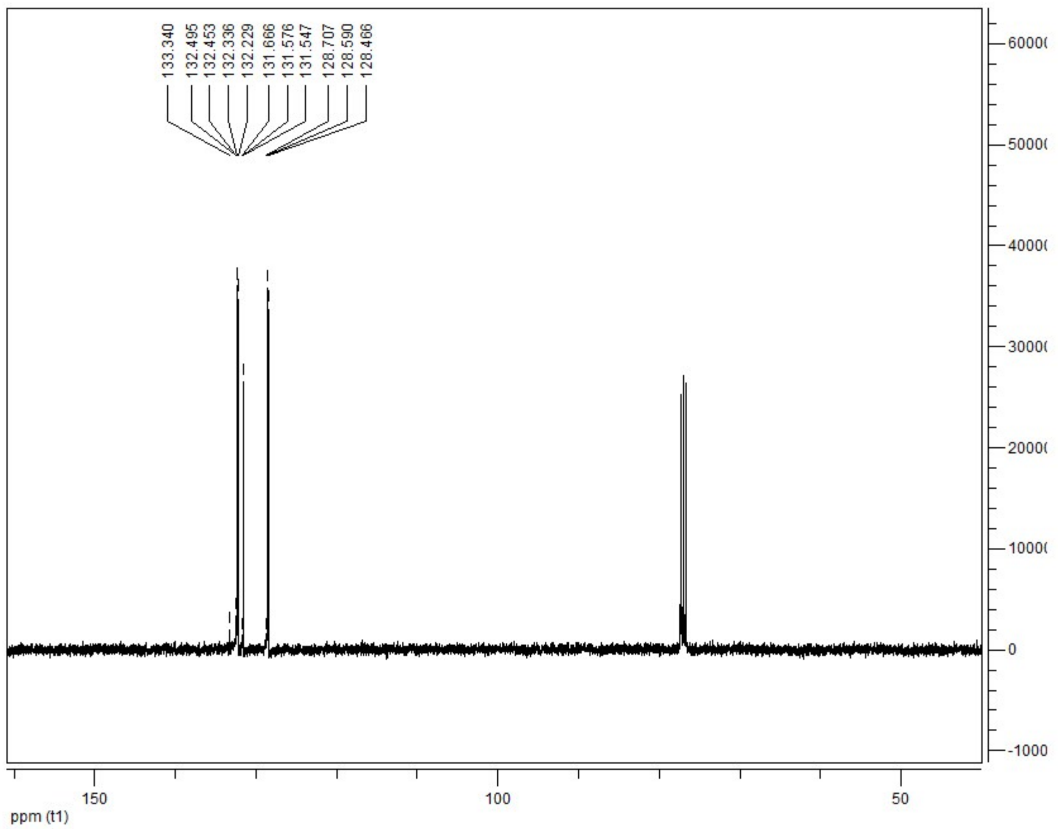
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3. References

1. C. G. Mortimer, G. Wells, J. P. Crochard, E. L. Stone, T.D. Bradshaw, M. F. G. Stevens and A. D. Westwel, *J. Med. Chem*. 2006, **49**, 179-185.
2. D. Alagille, R. M. Baldwina, G. D. Tamagnan,. *Tetrahedron Lett.* 2005, **46**, 1349–1351.
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