Electronic Supplementary Information (ESI)

Highly Efficient One-pot Three-component Betti Reaction in water using Reverse Zinc Oxide Micelles as Recoverable and Reusable Catalyst

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1. Comparison of size distribution and TEM of reverse ZnO nanomicelles between prior to and after reaction.

The comparison of the size distributions of nanomicelles and TEM micrographs were shown in Figure 1 and Figure 2, respectively. The size distribution of nanomicelles was narrow and maintained the normal distribution between prior to and after reaction. After the reaction, the average size of the nanoparticles increased from about 116.8 to 176.0 nm. TEM images displayed clearly that most of ZnO nanomicelles was spherical. No remarkable changes occurred after reaction.

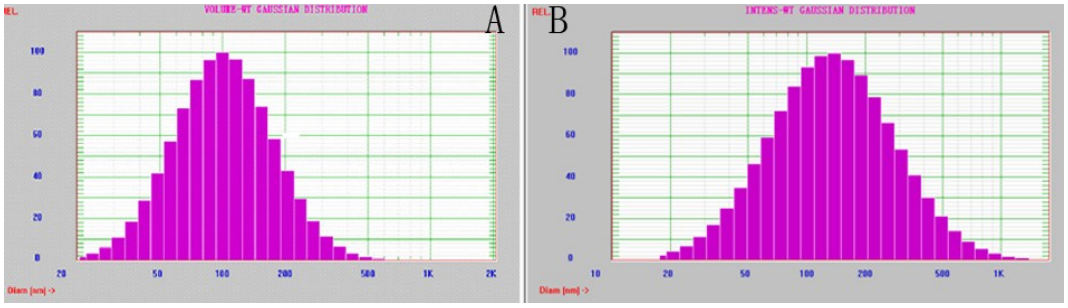


Fig.1 Size distribution for reversed ZnO nanomicelles. (A) Prior to the reaction, average diameter is 116.8 nm; (B) After the reaction, average diameter is 176.0 nm

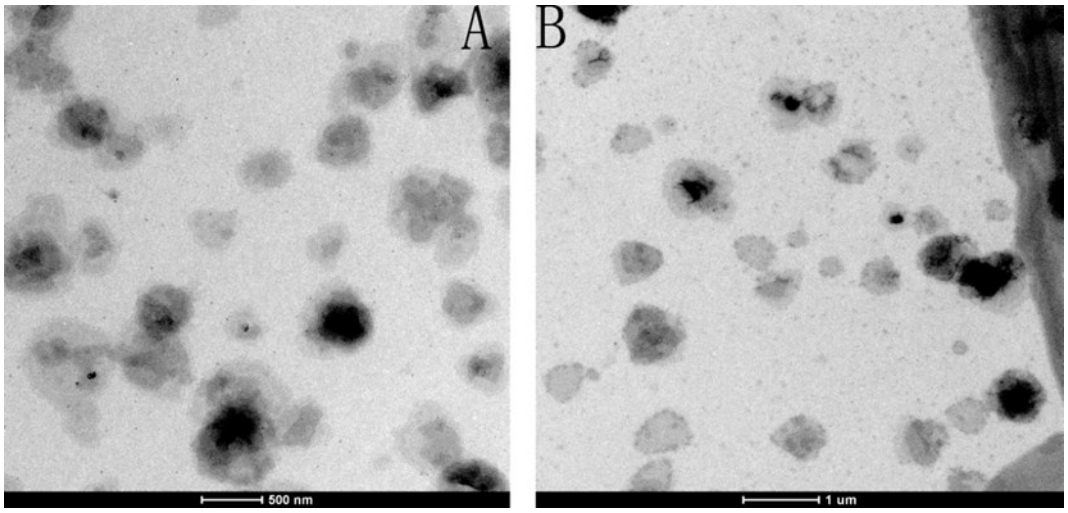


Fig.2 TEM image of reversed ZnO nanomicelles. (A) Prior to the reactions; (B) After the reactions.

2. Determination of the content of Znic oxide

According to the turbidity quantitative method, the relationship of the absorbance at 660 nm and the turbidity of the standard sample at the different concentration were investigated. The standard curve was plotted in Origin 8.0 to obtain the standard curve equation (Fig. 3). The reaction was cycled for six times. The content of ZnO of every cycle was achieved from the absorbance data. The variation tendency was shown on Fig 4. The curve illustrated that the content of ZnO maintained stable.

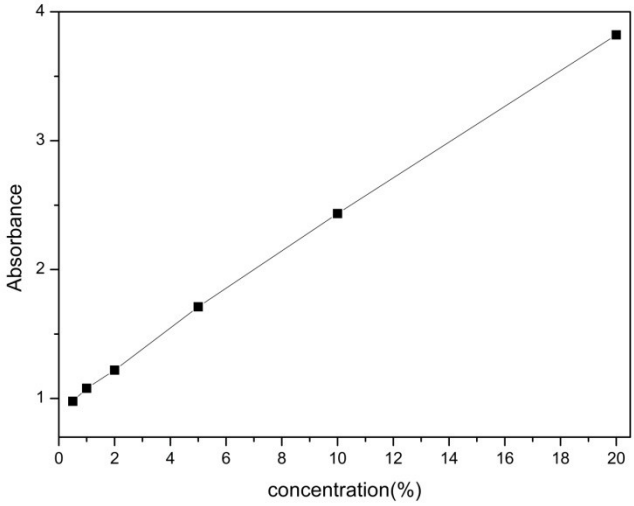


Fig.3 Standard curve

Y=0.1454x+0.941 R^2=0.99881

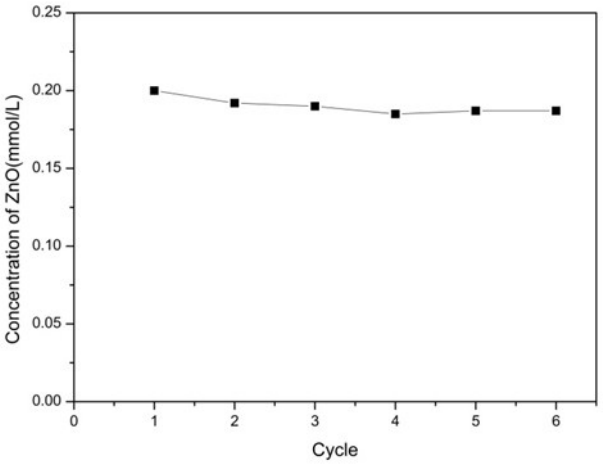
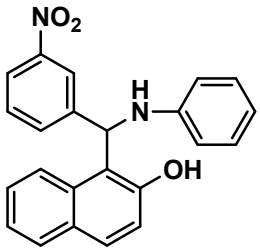


Fig.4 The contents of ZnO in six cycle

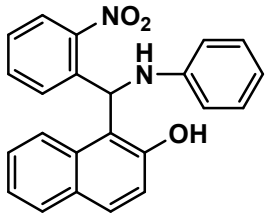
3. General procedure for the preparation of Betti base derivatives 4a~4j

To a mixture of aromatic aldehydes (1 mmol), β-naphthol (1 mmol, 0.144 g) and benzylamine (1 mmol, 0.094 g) in 30 mL water in a round bottom flask, catalytic amount of ZnO reversed nanomicelles was added and stirred at room temperature for appropriate time. After completion of the reaction as monitored by thin layer

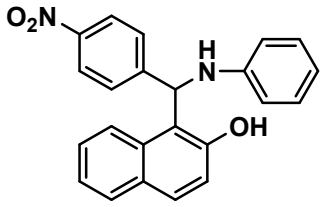
chromatography (TLC), the aqueous layer was decanted to recycle the catalyst for another reaction. The crude product was filtered and purified by recrystallization from ethanol (95%).



1-((3-nitrophenyl)(phenylamino)methyl)naphthalen)-2-ol (4a) yellow solid, yield: 0.355g, 96%; mp: 152 ºC(95%ethanol); IR (KBr, *v*, cm^{-1}): 3335.63 (s), 3092.03 (w), 2893.08 (w), 1601.28(m), 1528.37(s), 1498.62(m), 1344(m), 1231.93(m), 1216.73(m); ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 11.06 (s, 1H,Ar-OH), 8.39 (t, *J* = 2.0 Hz, 1H, Ar-H), 8.16 (m, 1H, Ar-H), 7.82-7.73 (m, 4H, Ar-H), 7.51 (dd, J_1 = 9.1 Hz, J_2 = 6.8 Hz, 1H, Ar-H), 7.42 (m, 1H, Ar-H), 7.33 (m, 1H, Ar-H), 7.20-7.14 (m, 3H, Ar-H), 6.98-6.92 (m, 1H, Ar-H), 6.80-6.76 (m, 2H,Ar-H), 6.31 (s, 1H, N-H), 4.16 (s, 1H,- CH -); ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.2, 146.2, 143.0, 134.2, 131.3, 130.8, 130.6, 129.7, 129.4, 127.3, 123.7, 123.3, 122.3, 120.9, 120.2, 116.4, 112.9, 61.5; HRMS (ESI) m/z [$M-H^-$]: 370.4007, found: 369.1229.

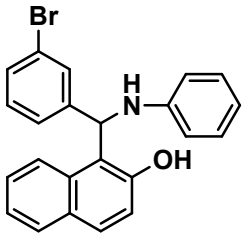


1-((2-nitrophenyl)(phenylamino)methyl)naphthalen)-2-ol (4b) yellow solid, yield: 0.341g, 92 %; mp: 131.7-133.9 ºC (95%ethanol); IR (KBr, *v*, cm^{-1}): 3335.02 (s), 1625.79 (w), 1597.2 (m), 1531.68 (s), 1497.49 (m), 1232.54 (m), 1218.5 (m), 813.76 (m) ; ^{1}H NMR(400 MHz, $CDCl_3$) δ (ppm):11.01 (s, 1H, Ar-OH), 8.39 (t, *J* = 2.0 Hz, 1H, Ar-H), 8.16 (m, 1H, Ar-H), 7.79 (m, 5H,Ar-H), 7.51 (dd, J_1= 9.4Hz, J_2= 6.5 Hz, 1H, Ar-H), 7.42 (m, 1H, Ar-H), 7.36 - 7.29 (m, 1H, Ar-H), 7.21 - 7.11 (m, 3H, Ar-H), 6.98 - 6.91 (m, 1H, Ar -H), 6.82 - 6.74 (m, 2H, Ar-H), 6.31 (s, 1H, N-H), 4.11 (s, 1H, -CH-); ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.2, 146.2, 143.0, 134.2, 131.3, 130.8, 130.6, 129.7, 129.4, 127.3, 123.7, 123.3, 122.3, 120.9, 120.2, 116.4, 112.9, 61.5; HRMS (ESI) m/z [$M-H^-$]: 370.4007, found:369.1248.

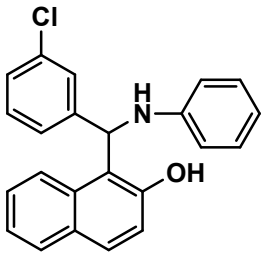


1-((4-nitrophenyl)(phenylamino)methyl)naphthalen)-2-ol (4c) yellow solid, yield:0.352g, 95%;mp: 136.1-138.4

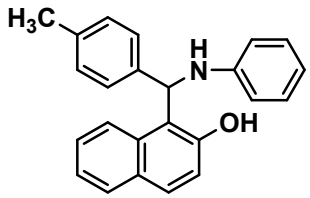
ºC(95%ethanol); IR (KBr, *v*, cm^{-1}): 3333.88 (m), 1625.89 (w), 1603.52 (m), 1525.25 (s), 1500.09 (m), 1234.52 (m), 818.01 (w); ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 11.92 (s, 1H, Ar-OH), 8.22-8.17 (m, 2H, Ar-H), 7.41 (m, 1H, Ar-H), 7.32 (m, 1H, Ar-H), 7.20-7.12 (m, 3H, Ar-H), 6.97-6.92 (m, 1H, Ar-H), 6.82-6.76 (m, 2H, Ar-H), 6.30(s, 1H, Ar-H), 5.29 (m, 1H, N-H),4.14 (s, 1H, -CH-); ^{13}CNMR (100 MHz, $CDCl_3$) δ 155.9, 147.9, 146.3, 131.3, 130.8, 129.7, 129.3, 129.1, 127.3, 124.7, 123.3, 122.3, 120.9, 120.1, 116.3, 113.1, 61.5; HRMS (ESI) m/z [$M-H^-$]: 370.4007, found: 369.1241.



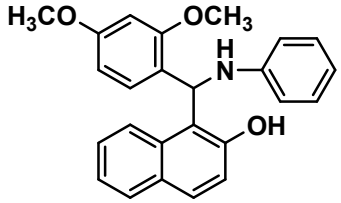
1-((3-bromophenyl)(phenylamino)methyl)naphthalen-2-ol (4d) brown solid, yield: 0.375g, 93 %; mp: 139.2-141.9 ºC (95%ethanol); IR (KBr, v, cm^{-1}): 3334.97 (s), 2887.41 (w), 1601.03 (s), 1496.65 (m), 1229.43 (s), 744.37 (m); ^{1}H NMR (400MHz,$CDCl_3$) δ (ppm): 11.34 (s, 1H, Ar-OH), 7.82-7.72 (m, 3H, Ar-H), 7.65 (d, *J* = 1.4 Hz, 1H, Ar-H), 7.46-7.37 (m, 3H, Ar-H), 7.35-7.28 (m,1H,Ar-H), 7.24-7.12 (m, 4H,Ar-H), 6.93 (t, *J* = 7.4 Hz, 1H, Ar-H), 6.76 (d, *J* = 8.2 Hz, 2H, Ar-H), 6.14 (s, 1H, N-H), 4.13 (s, 1H,-CH-); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 156.3, 146.5, 143.2, 131.9, 131.4, 131.1, 131.1, 130.4, 129.6, 129.2, 127.1, 126.8, 123.5, 123.1, 122.2, 121.3, 120.2, 116.4, 113.1, 62.2; HRMS (ESI) m/z [$M-H^-$]: 404.2992, found: 404.0485.



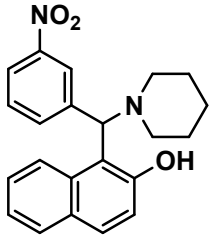
1-((3-chlorophenyl)(phenylamino)methyl)naphthalen-2-ol (4e) white solid, yield: 0.313g, 87 %; mp: 141.6 - 143.3 ºC (95%ethanol); IR (KBr, v, cm^{-1}): 1624.74 (m), 1601.08 (s), 1498.54 (m), 1269.79 (m), 1235.61 (m), 743.92 (m); ^{1}H NMR (400MHz, $CDCl_3$) δ (ppm): 11.63 (s, 1H, Ar-OH), 7.79 (dd, J_1 = 8.3Hz, J_2 =3.4 Hz, 2H, Ar-H), 7.58 (d, *J* = 8.5 Hz, 1H, Ar-H), 7.51 (d, *J* = 8.0 Hz, 1H, Ar-H), 7.41-7.34 (m, 1H, Ar-H), 7.31 (d, *J* = 7.7 Hz, 1H, Ar-H), 7.27 (dd, J_1 = 7.5Hz, J_2 = 1.8 Hz, 1H, Ar-H), 7.20-7.15 (m, 3H, Ar-H), 7.15-7.13 (m, 1H, Ar-H), 7.11 (dd, J_1 = 7.7Hz, J_2 = 0.9 Hz, 1H, Ar-H), 6.94 (t, *J* = 7.4 Hz, 1H, Ar-H), 6.82 (d, *J* = 7.8 Hz, 2H, Ar-H), 6.55 (s, 1H, -CH-), 3.99 d, *J* = 77.8 Hz, 1H, N-H); ^{13}C NMR (100 MHz, CDCl3) δ: 157.1, 147.0, 137.5, 133.9, 131.5, 130.3, 129.5, 129.1, 127.9, 127.2, 116.6, 112.8, 59.6; HRMS (ESI) m/z [$M-H^-$]: 359.8479, found: 358.0988.



1-((phenylamino)(p-tolyl)methyl)naphthalen-2-ol (4f) white solid, yield: 0.282g, 83 %, mp: 133.1-135.5 ºC (95%ethanol); IR (KBr, *v*, cm^{-1}): 3352.17(s), 2973.76 (w), 1603.48 (s), 1496.03 (m),1230.1(s), 1466.49 (m), 1378.51(m); ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 11.5 (s, 1H, Ar-OH), 7.76 (dd, J_1 = 16.4 Hz, J_2 = 8.6 Hz, 3H, Ar-H), 7.37 (t, *J* = 9.3 Hz, 3H, Ar-H), 7.28 (t, *J* = 7.5 Hz, 1H, Ar-H), 7.17-7.12 (m, 5H, Ar-H), 6.91 (t, *J* = 7.4 Hz, 1H, Ar-H), 6.76 (d, *J* = 7.9 Hz, 2H, Ar-H), 6.14 (s, 1H, N–H), 4.13 (s, 1H,-CH-), 2.31 (s, 3H, -CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 156.2, 146.8, 138.5, 131.6, 130.1, 130.0, 129.6, 129.2, 129.1, 128.0, 126.9, 122.9,121.9, 121.5, 120.1, 116.4, 114.0, 62.6, 21.2; HRMS (ESI) m/z [M-H^-]: 339.4297, found: 338.1531.

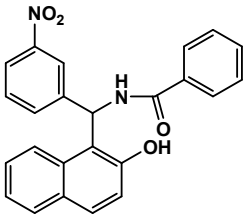


1-((2,4-dimethoxyphenyl)(phenylamino)methyl)naphthalen-2-ol (4g) yellow solid, yield: 0.112g, 29 % ; mp: 135.7-137.1 ºC (95%ethanol); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.88 (s, 1H, Ar–OH), 7.77 (t, *J* = 9.6 Hz, 2H, Ar-H), 7.67 (d, *J* = 8.1 Hz, 1H, Ar-H), 7.44 (dd, J_1 = 14.2 Hz, J_2=7.0 Hz, 2H, Ar-H), 7.37- 7.31 (m, 2H, Ar-H), 7.29 (s, 4H, Ar-H), 7.00 (t, *J* = 7.2 Hz, 1H, Ar-H), 6.92 (d, *J* = 7.5 Hz, 1H, Ar-H), 6.65 (dd, J_1 = 8.9 Hz, J_2 =2.3 Hz, 1H, Ar-H), 6.53 (d, *J* = 6.2 Hz, 1H, Ar-H), 6.49 (d, *J* = 2.3 Hz, 1H, Ar-H), 3.91 (s, 3H, -OCH_3), 3.52 (s, 3H, -OCH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ 153.6, 134.6, 129.8, 128.9, 128.7, 127.8, 126.4, 123.5, 121.1, 118.9, 117.9, 109.5, 98.1, 58.5, 55.7, 50.9; HRMS(ESI) m/z[M-H^-]: 385.4551, found: 384.1573.

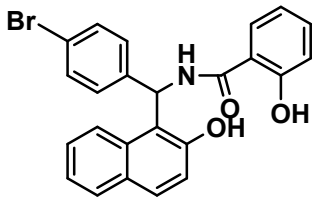


1-((3-nitrophenyl)(piperidin-1-yl)methyl)naphthalen-2-ol (4h) yellow solid, yield:0.337g, 93 %, mp: 185.8-186.5 ºC[(1)] (95%ethanol); IR (KBr, *v*, cm^{-1}): 3626.29 (w), 2953.47 (w), 2811.51(w), 1620.41(m), 1449.05 (m), 1343.15 (s), 1237.06 (m); ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 8.40 (s, 1H, Ar-OH), 8.06 (m, 1H, Ar-H), 7.93 (s, 1H,Ar-H), 7.78 (d, *J* = 8.6Hz, 1H,Ar-H), 7.70 (dd, J_1 = 15.9 Hz, J_2= 7.8 Hz, 2H, Ar-H), 7.41(m, 2H, Ar-H), 7.27-7.21 (m, 2H, Ar-H), 7.16 (d, *J* = 8.9Hz, 1H,Ar-H), 5.19 (s, 1H, -CH-), 3.34 (s, 1H, -CH_2-), 2.61 (s, 1H, -CH_2-), 2.08 (d, *J* = 49.1 Hz, 2H, -CH_2-), 1.65

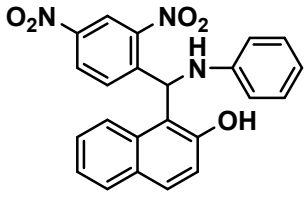
(d, *J* = 39.6 Hz, 6H,-CH_2-), 1.40-1.14 (m, 1H, -CH_2-); ^{13}C NMR (100 MHz, $CDCl_3$) δ 155.6, 142.1, 135.1, 132.1, 130.2, 129.2, 128.8, 126.9, 124.1, 123.2, 122.8, 120.5, 120.2, 115.2, 71.3, 54.9, 26.1, 24.1; HRMS (ESI) m/z [M-H^-]: 362.4218, found: 361.1544.



N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)benzamide(4i) white solid, yield: 0.291 g, 73 %; mp: 217.7-219.5ºC[(2)] (95 % ethanol), ^{1}H NMR (400MHz, DMSO-d_6) δ (ppm): 9.73 (s, 1H, Ar-OH), 8.63 (s, 1H, Ar-H), 7.89 (d, *J* = 8.4 Hz, 2H, Ar-H), 7.80 - 7.71 (m, 4H, Ar-H), 7.67 (d, *J* = 8.2 Hz, 1H,Ar-H), 7.41 (d, J_1 = 15.1 Hz, J_2= 8.0 Hz, 3H,Ar-H), 7.26 (dd, J_1= 15.4 Hz, J_2 = 7.6 Hz, 4H, Ar-H), 7.14 - 7.05 (m, 2H, -NH- and -CH-).

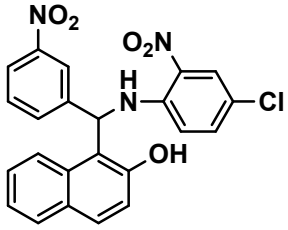


N-((4-bromophenyl)(2-hydroxynaphthalen-1-yl)methyl)-2-hydroxybenzamide (4j) white solid, yield: 0.318g, 71 %; mp: 181.2-183.1 ºC (95 % ethanol), IR (KBr, ν, cm^{-1}): 3312.32 (w), 3127.51 (w), 1600.81 (w), 1504,17 (w), 1401.65 (s), 1228.1 (m), 1068.96 (m), 1014.96 (m), 813.09 (m); ^{1}H NMR (400 MHz, $CDCl_3$) δ: 11.25 (s, 1H, Ar-OH), 8.60 (s, 1H, Ar-H), 7.80 (t, *J* = 11.6 Hz, 2H, Ar-H), 7.65 (dd, J_1 = 18.8 Hz, J_2 =8.4 Hz, 1H, Ar–H), 7.57-7.52 (m, 2H， Ar-H), 7.46 (dd, J_1 = 15.0 Hz,J_2 =7.4 Hz, 3H, Ar-H), 7.40 - 7.32 (m, 2H, Ar-H), 7.31-7.21 (m, 4H, Ar-H), 6.42 (s, 1H, N-H), 5.62 (s, 1H, -CH-); ^{13}C NMR (100 MHz, $CDCl_3$) δ 161.3, 152.3, 141.6, 137.9, 132.4, 132.1, 131.5, 131.1, 129.6, 129.2, 128.9, 128.7, 128.0, 126.9, 123.6, 122.7, 121.6, 119.3, 113.8, 81.7; HRMS (ESI) m/z [M-H^-]: 448.3086, found: 447.1367.

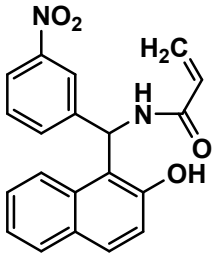


1-((2,4-dinitrophenyl)(phenylamino)methyl)naphthalen-2-ol (4k) yellow solid, yield: 0.219 g, 53 %; mp: 143.3 - 144.1 ºC (95 % ethanol);^{1}H NMR (400 MHz, $CDCl_3$) δ 10.30 (s, 1H, Ar-OH), 8.46 (s, 1H, Ar-H), 7.96-7.78 (m, 3H, Ar - H), 7.51 (d, *J* = 9.1 Hz, 1H, Ar-H), 7.47 - 7.32 (m, 4H, Ar-H), 7.24 (dd, J_1 = 17.3 Hz, J_2 = 8.2 Hz, 3H, Ar-H), 7.15-6.95

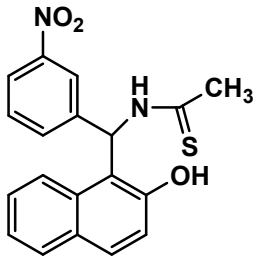
(m, 3H, Ar-H), 5.37 (s, 1H, Ar-H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 161.6, 155.7, 133.6, 133.2, 131.0, 128.8, 128.2, 127.9, 127.2, 123.6, 122.4, 118.5, 117.9, 110.7, 60.5; HRMS (ESI) m/z [M-H$^-$]: 415.3983, found: 414.0639.



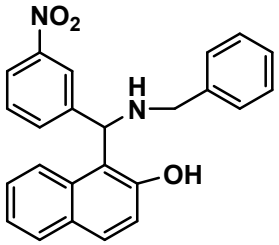
1-(((4-chloro-2-nitrophenyl)amino)(3-nitrophenyl)methyl)naphthalen-2-ol (4l) yellow solid, yield: 0.202g, 45%, mp: 150.9 - 151.8 ºC (95 % ethanol); IR (KBr, ν, cm^{-1}): 3322.6 (m), 1623.21 (m), 1598.18 (m), 1528.65 (s), 1470.24 (m), 1347.86 (s), 1211.58 (m), 710.94 (m); ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 10.08 (s, 1H, Ar-OH), 8.21 (s, 1H, Ar-H), 8.04-7.96 (m, 3H, Ar-H), 7.70 (dd, J_1 = 10.3Hz, J_2 = 4.9 Hz, 4H, Ar-H), 7.52 (t, J = 8.0 Hz, 1H, Ar-H), 7.24 - 7.10 (m, 4H, Ar-H), 6.77 (s, 1H, N-H), 6.44 (s, 1H, -CH-); ^{13}C NMR (100 MHz, DMSO-d_6) δ157.7, 153.4, 153.0, 137.3, 134.9, 134.1, 133.6, 130.9, 130.1, 127.7, 126.4, 125.7, 125.2, 123.4, 71.3; MS (ESI) m/z [M-H$^-$]: 449.8431, found: 448.0682.



N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)acrylamide (4m) brown solid, yield: 0.056 g, 16 %; mp: 119.2-121.2 ºC (253 – 254 ºC[3]) (95 % ethanol); IR (KBr, *v*, cm^{-1}): 3229.05 (m), 3107.44 (m), 1624.27 (m), 1596.4 (m), 1529.51 (s), 1462.62 (m), 1404.1 (m), 1283.64 (m), 1206.22 (m), 1013.92 (m), 913.59 (w); ^{1}H NMR (400 MHz,DMSO-d_6) δ (ppm) : 10.06 (s, 1H, Ar-OH), 8.22 (s, 1H, Ar-H), 8.00 (dd, J_1 = 8.2 Hz, J_2 = 2.6 Hz, 3H, Ar-H), 7.70 (m, 4H, Ar-H), 7.52 (t, J = 8.0 Hz, 1H, Ar-H), 7.23 - 7.12 (m, 4H, Ar-H), 6.77 (s, 1H, N–H), 6.44 (s, 1H, -CH-); ^{13}C NMR (100 MHz, DMSO-d_6) δ 152.9, 148.6, 148.2, 132.6, 130.1, 129.3, 128.8, 126.2, 125.4, 122.9, 121.8, 120.9, 120.4, 118.6, 100.0, 66.5.



N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)ethanethioamide (4n) brown solid, yield: 0.126 g, 36%; mp: 135.0 - 136.8 ºC (234 - 236 ºC[4]) (95 % ethanol); IR (KBr, ν, cm^{-1}): 2974.83 (w), 2896.7 (w), 1622.41 (m), 1606.24 (s), 1498.48 (m), 1409.48 (m), 1379.94 (m), 1312.59 (w), 1231.77 (s), 946.2 (m); ^{1}H NMR (400 MHz, $CDCl_3$) δ (ppm): 11.55 (s, 1H, Ar-OH), 7.75 (dd, J_1 = 16.3Hz,J_2 = 8.7 Hz, 3H, Ar-H), 7.37 (m, 3H, Ar-H), 7.14 (m, 5H, Ar-H), 6.94 - 6.88 (m, 1H, Ar-H), 6.76 (dd,J_1 = 8.6 Hz,J_2 = 1.0 Hz, 2H, Ar-H), 6.13 (s, 1H, N-H), 4.13 (s, 1H, -CH-), 2.33 - 2.27 (m, 3H, $-CH_3$); ^{13}C NMR (100 MHz, $CDCl_3$) δ 156.2, 146.8, 138.5, 138.2, 131.6, 130.1, 129.5, 129.1, 128.0, 126.9, 122.9, 121.9, 121.5, 120.1, 116.4, 114.0, 62.6, 21.2.



1-((benzylamino)(3-nitrophenyl)methyl)naphthalen-2-ol (4o) yellow solid, yield: 0.342 g, 87 %; mp: 136.1-138.4 ºC (95 % ethanol); IR (KBr, ν, cm^{-1}): 3312.73 (w), 2922.98 (w), 2851.77 (w), 1703.14 (s), 1619.83 (m), 1599.01 (m), 1530.58 (s), 1239.01 (m), 813.55 (m); ^{1}H NMR (400 MHz, $CDCl_3$) δ 10.12 (s, 1H, Ar-OH), 8.72 (s, 1H, Ar-H), 8.49 (d, *J* = 8.2 Hz, 1H, Ar-H), 8.32 (s, 1H, Ar-H), 8.24 (d, *J* = 7.6 Hz, 1H, Ar-H), 8.08 (d, *J* = 8.2 Hz, 1H, Ar-H), 7.70 (d, *J* = 8.6 Hz, 1H, Ar-H), 7.43 (dd, J_1 = 13.5 Hz, J_2 = 5.7 Hz, 2H, Ar-H), 7.41 - 7.36 (m, 3H, Ar-H), 7.35 - 7.30 (m, 3H, Ar-H), 7.29 - 7.23 (m, 2H, Ar-H), 5.91 (s, 1H, N-H), 4.12 (m, 2H, $-CH_2-$), 3.88 (d, *J* = 13.0 Hz, 1H, -CH-).^{13}C NMR (100 MHz, $CDCl_3$) δ 189.8, 171.2, 156.8, 148.5, 143.2, 137.5, 134.7, 134.1, 132.3, 130.7, 129.1, 128.7, 128.1, 126.9, 124.5, 123.2, 122.9, 120.6, 120.3, 112.0, 61.7, 52.7; MS (ESI) m/z [M $+H^-$]: 384.4273, found: 383.1394.

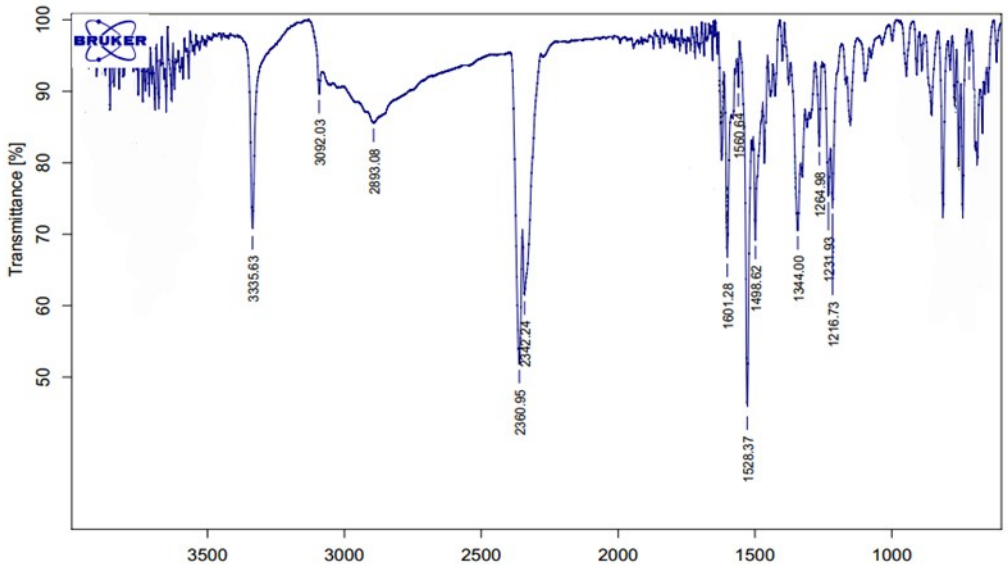


Fig. 5 IR spectrum of 4a

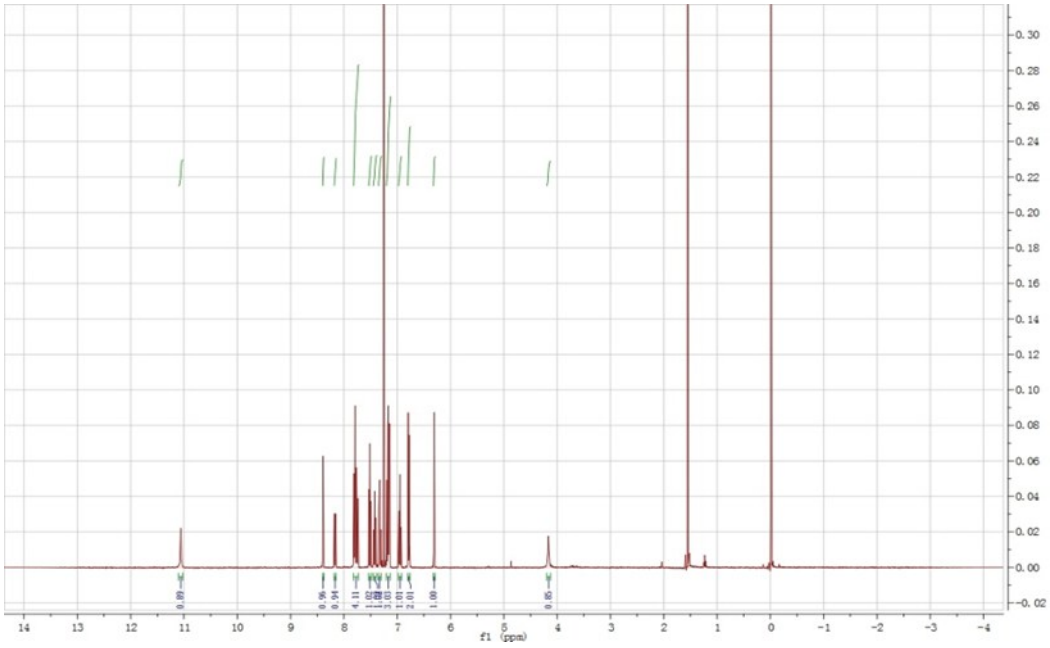


Fig. 6 ^{1}H NMR spectrum of 4a

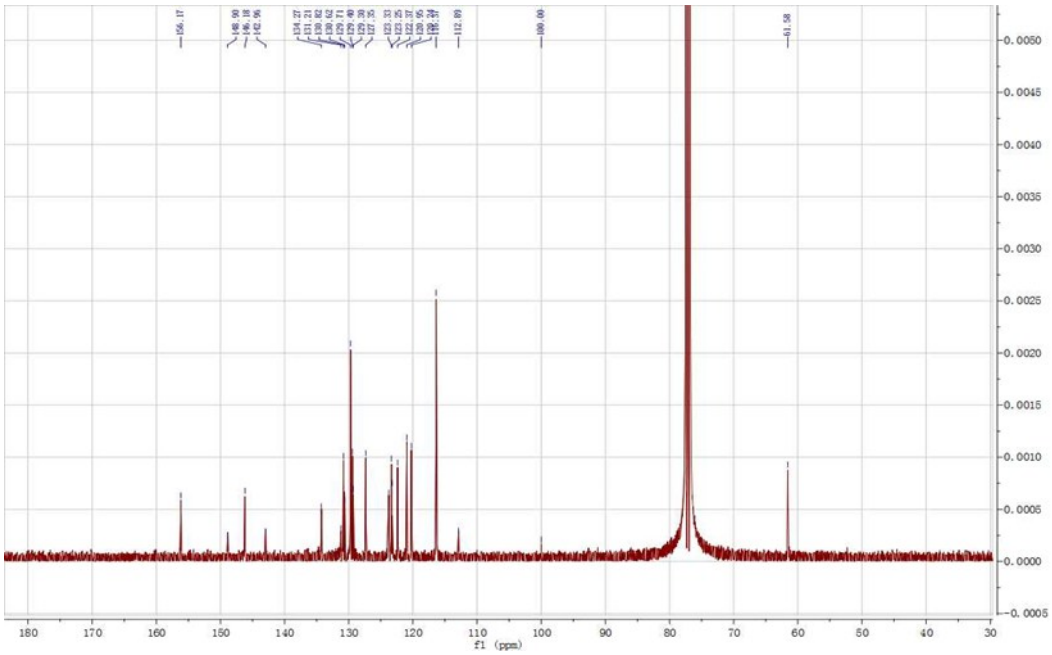


Fig. 7 ^{13}C NMR spectrum of 4a

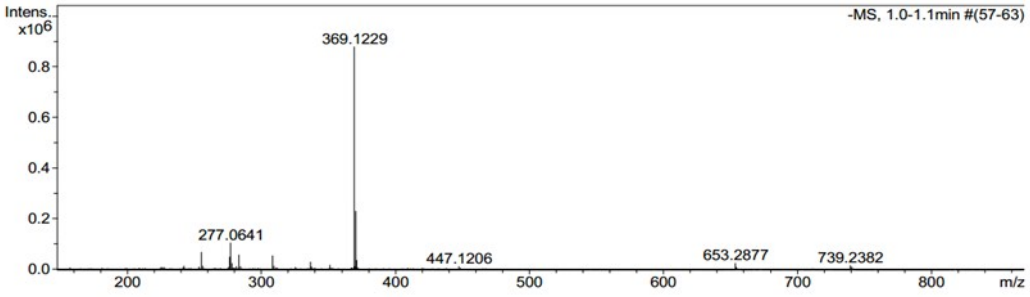


Fig. 8 HRMS spectrum of 4a

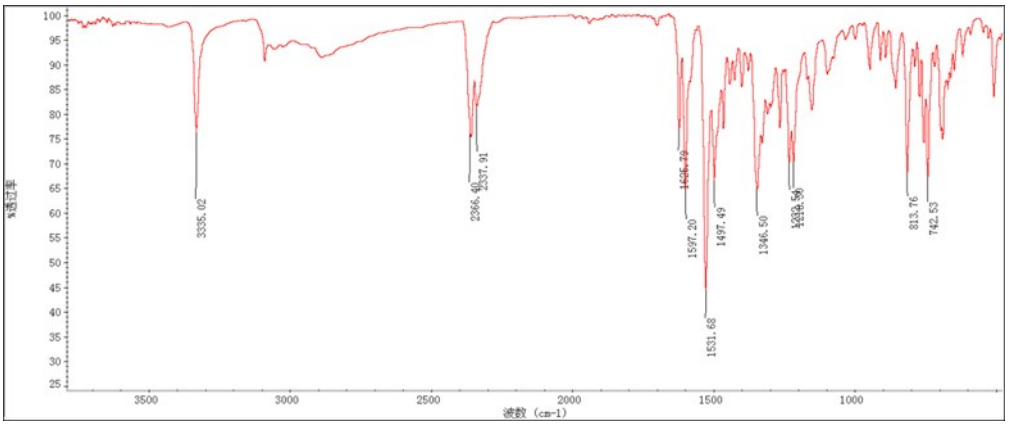


Fig. 9 IR spectrum of 4b

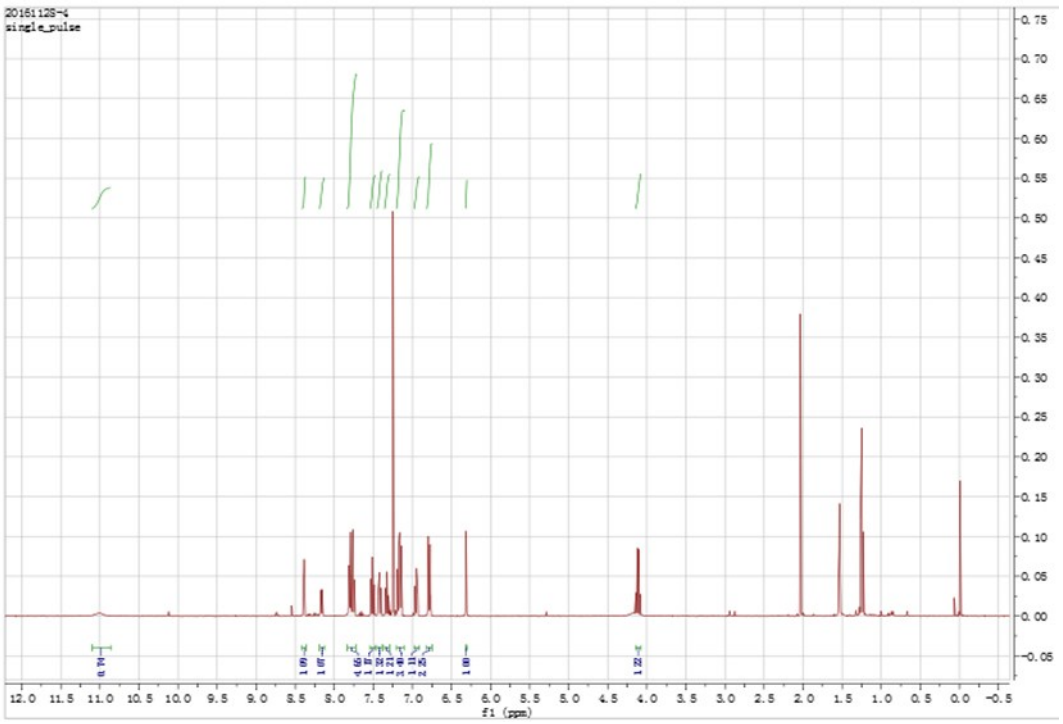


Fig. 10 ^{1}H NMR spectrum of 4b

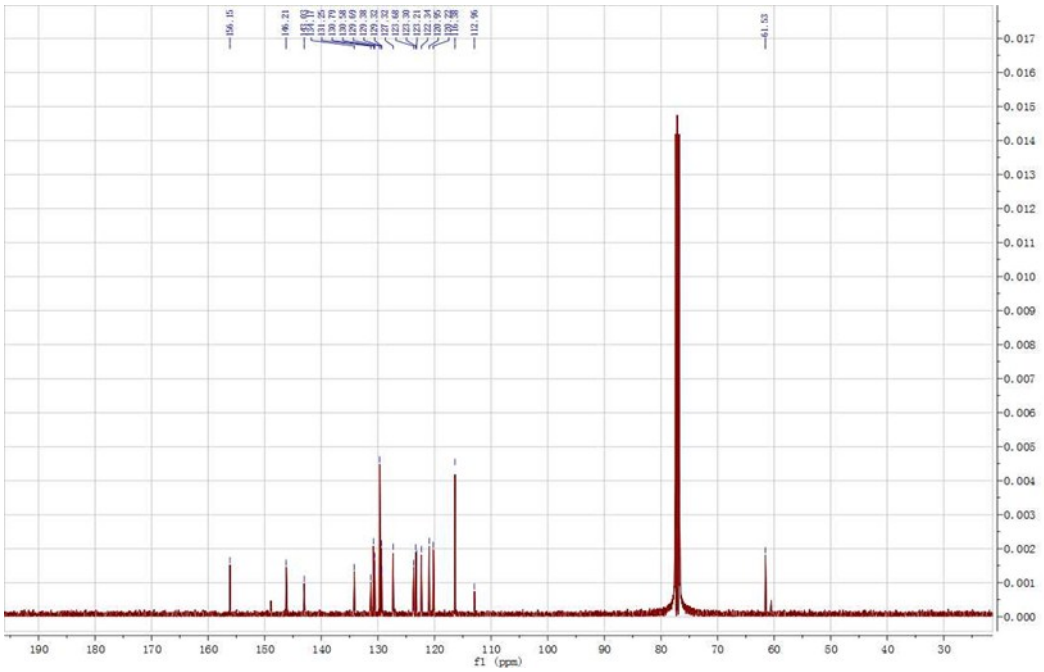


Fig. 11 ^{13}C NMR spectrum of 4b

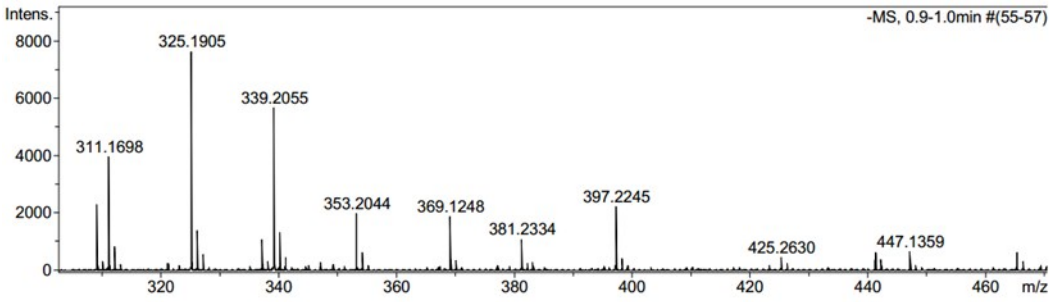


Fig. 12 HRMS spectrum of 4b

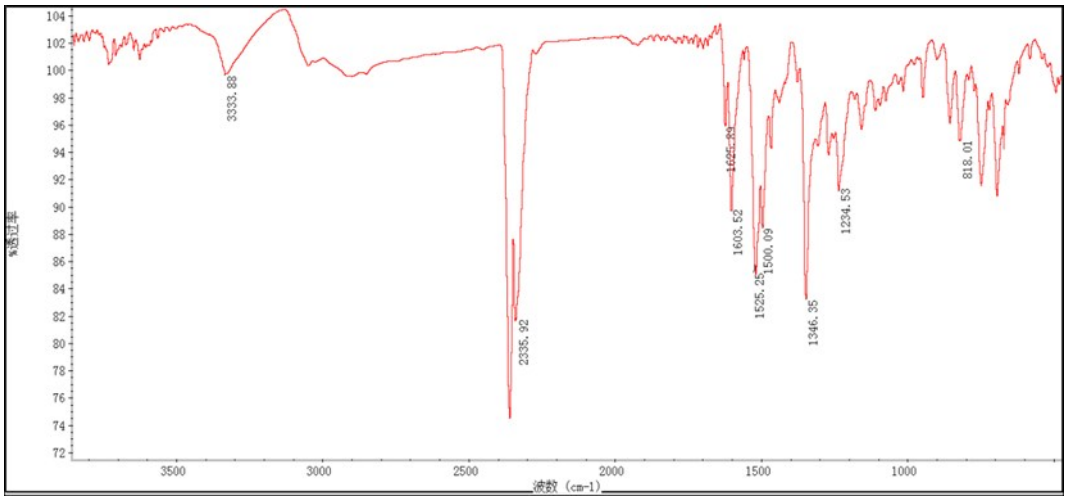


Fig. 13 IR spectrum of 4c

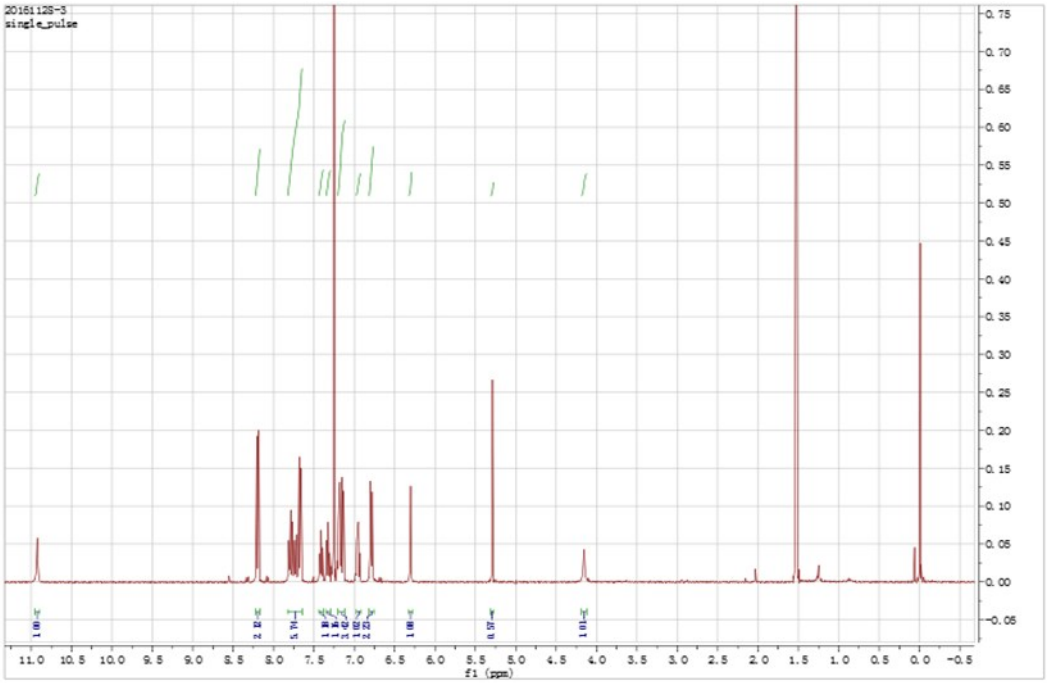


Fig. 14 ^{1}H NMR spectrum of 4c

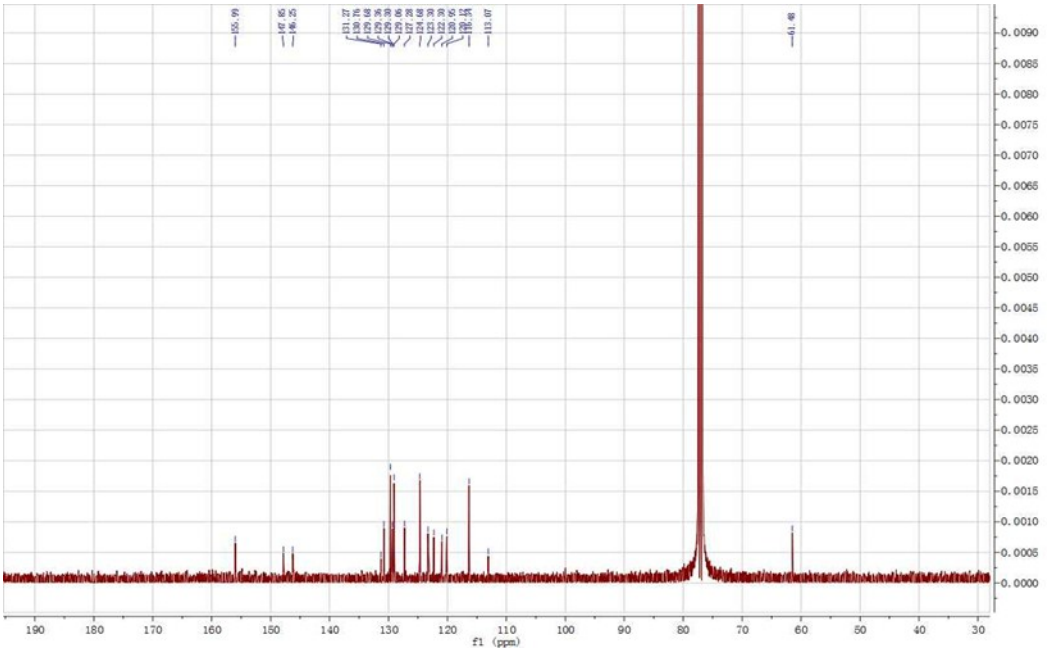


Fig. 15 ^{13}C NMR spectrum of 4c

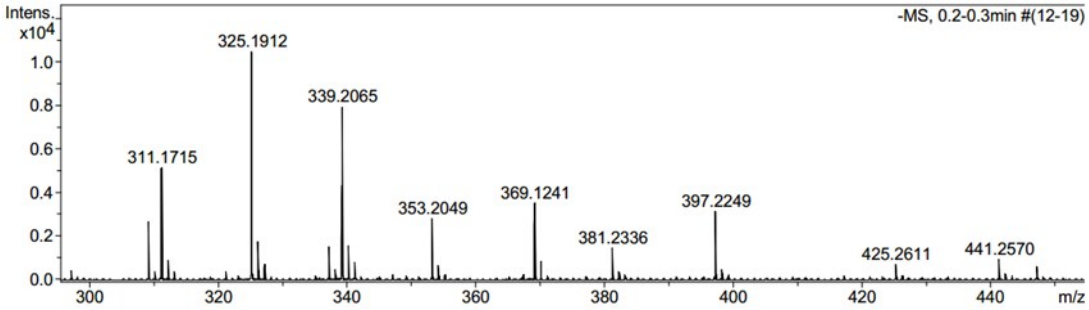


Fig. 16 HRMS spectrum of 4c

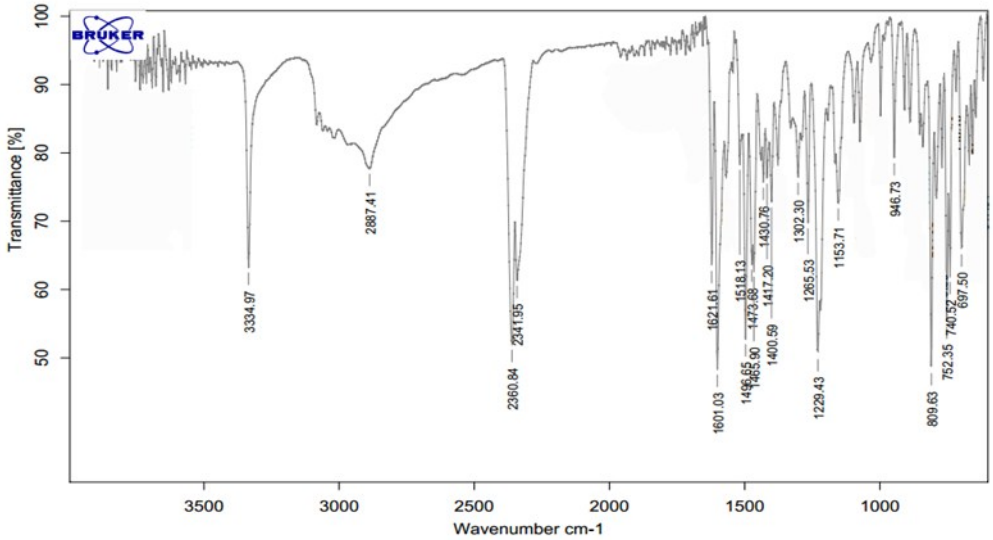


Fig. 17 IR spectrum of 4d

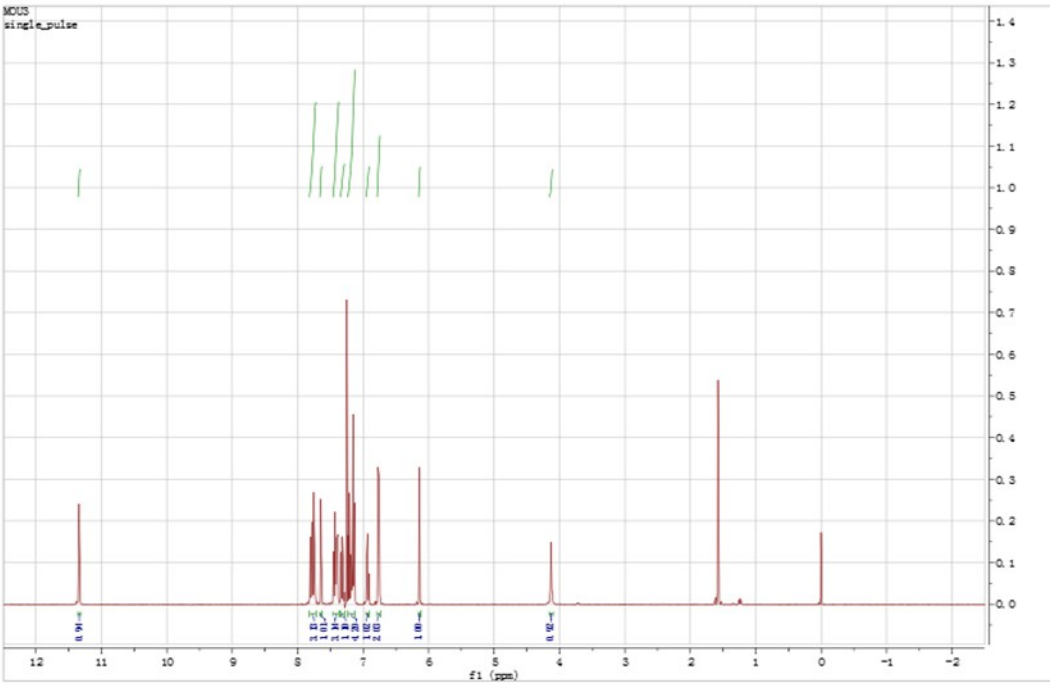


Fig. 18 ^{1}H NMR spectrum of 4d

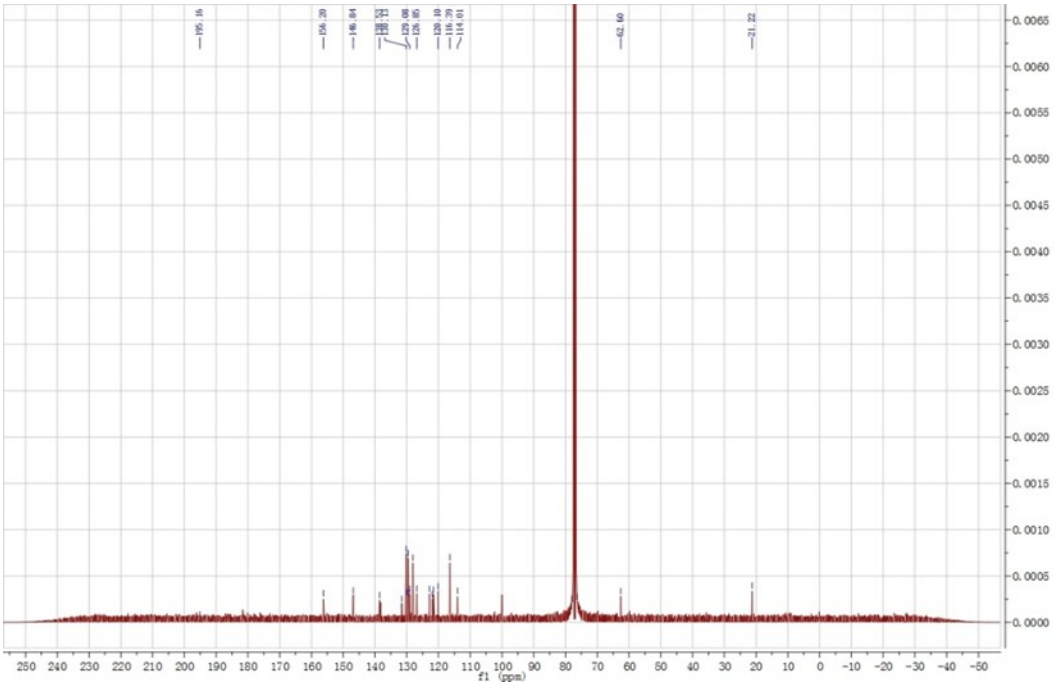


Fig. 19 ^{13}C NMR spectrum of 4d

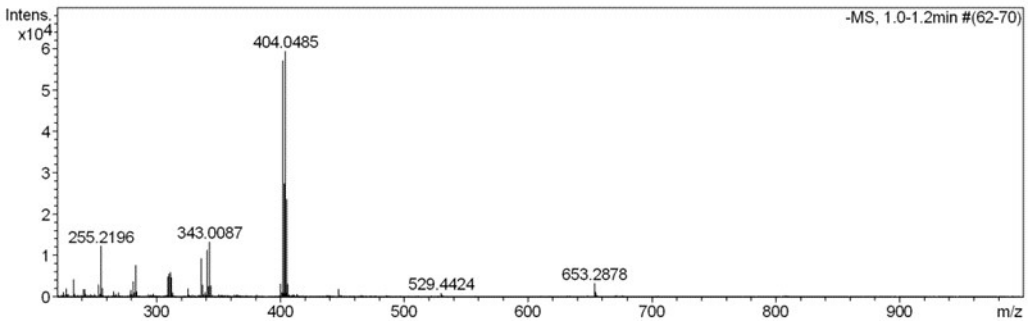


Fig.20 HRMS spectrum of 4d

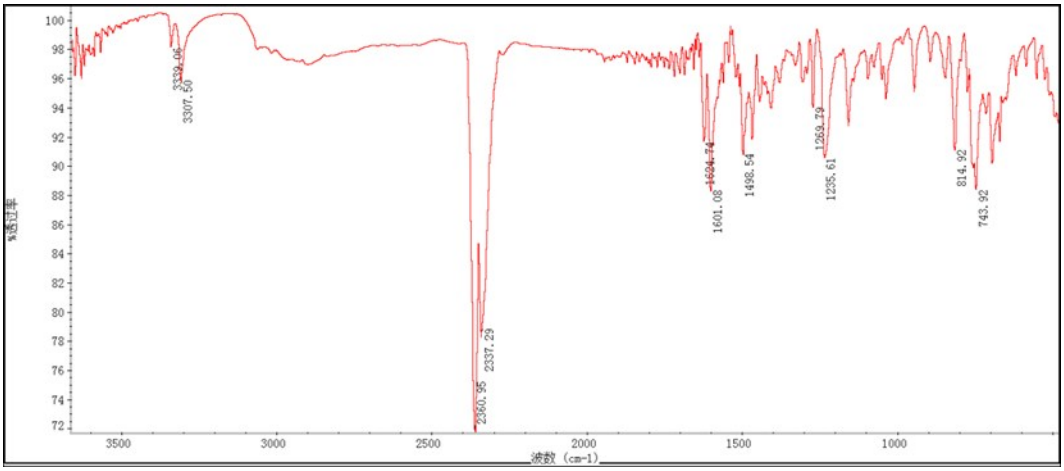


Fig. 21 IR spectrum of 4e

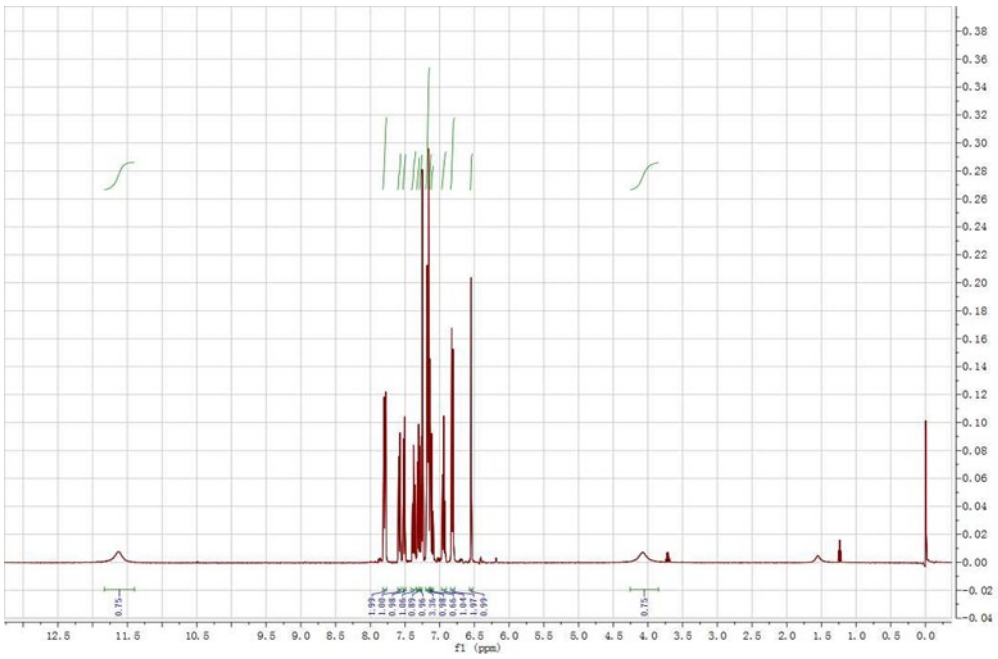


Fig. 22 ^{1}H NMR spectrum of 4e

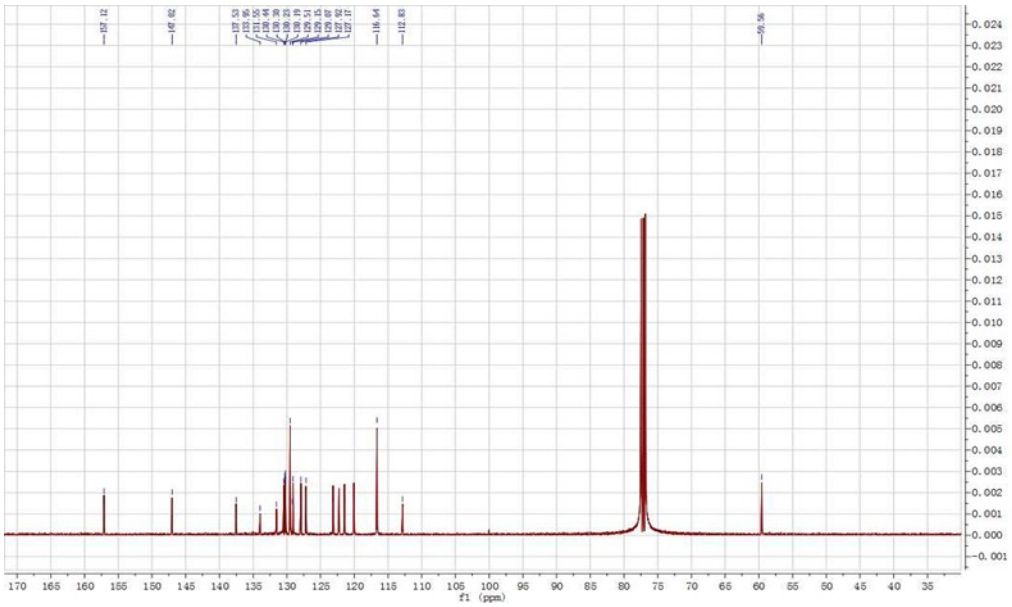


Fig. 23 ^{13}C NMR spectrum of 4e

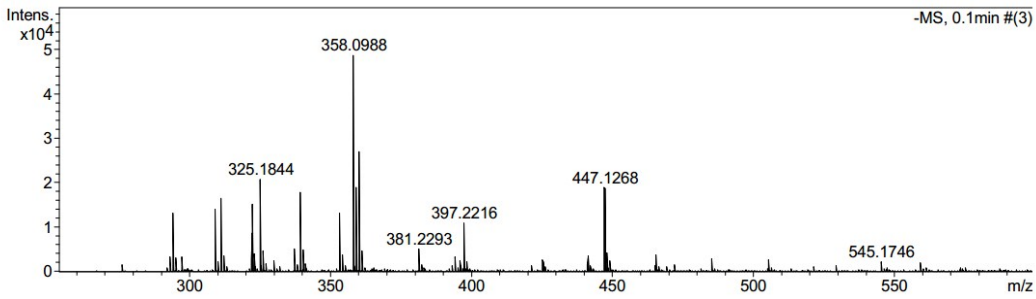


Fig. 24 HRMS spectrum of 4e

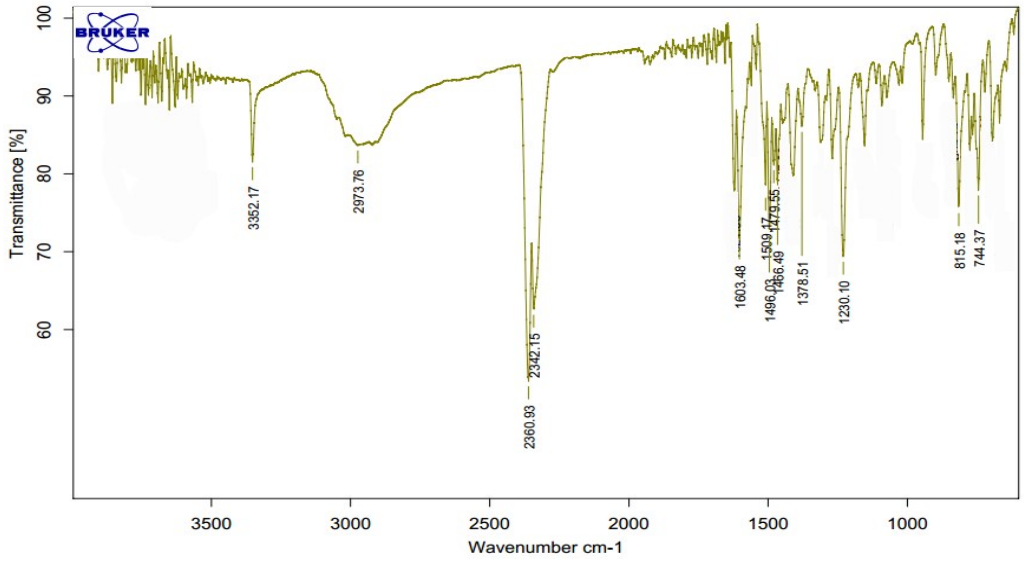


Fig. 25 IR spectrum of 4f

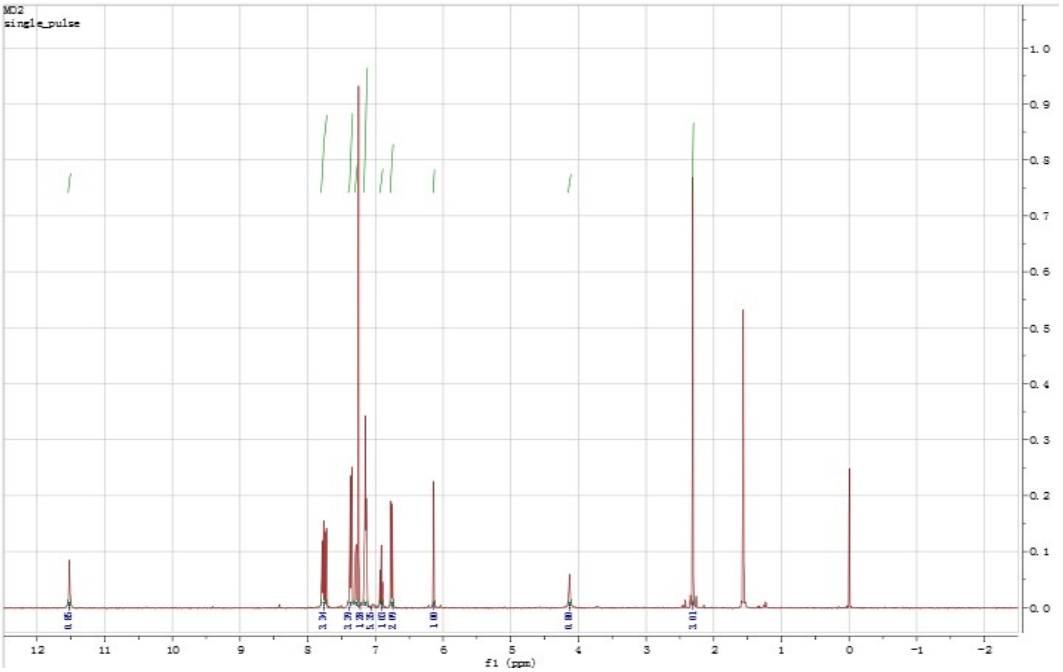


Fig. 26 ^{1}H NMR spectrum of 4f

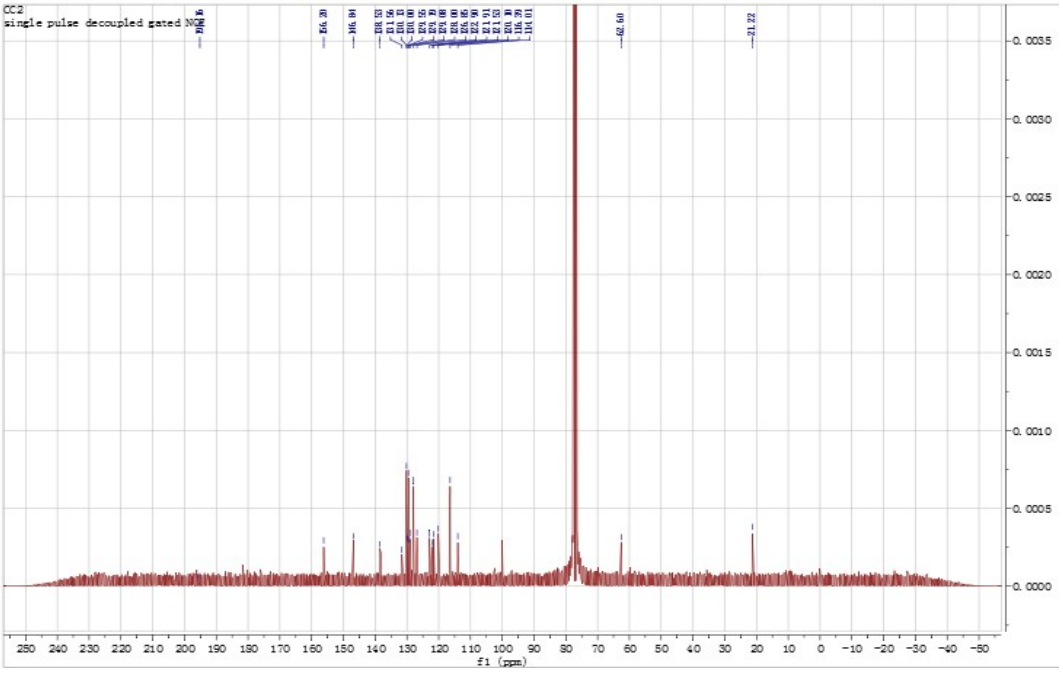


Fig. 27 ^{13}C NMR spectrum of 4f

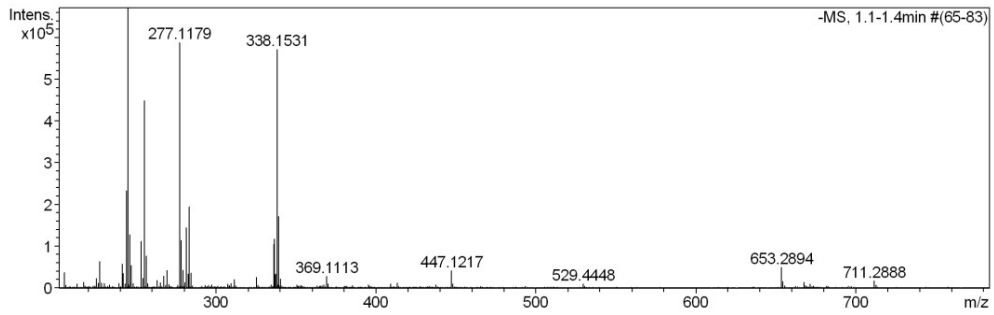


Fig. 28 HRMS spectrum of 4f

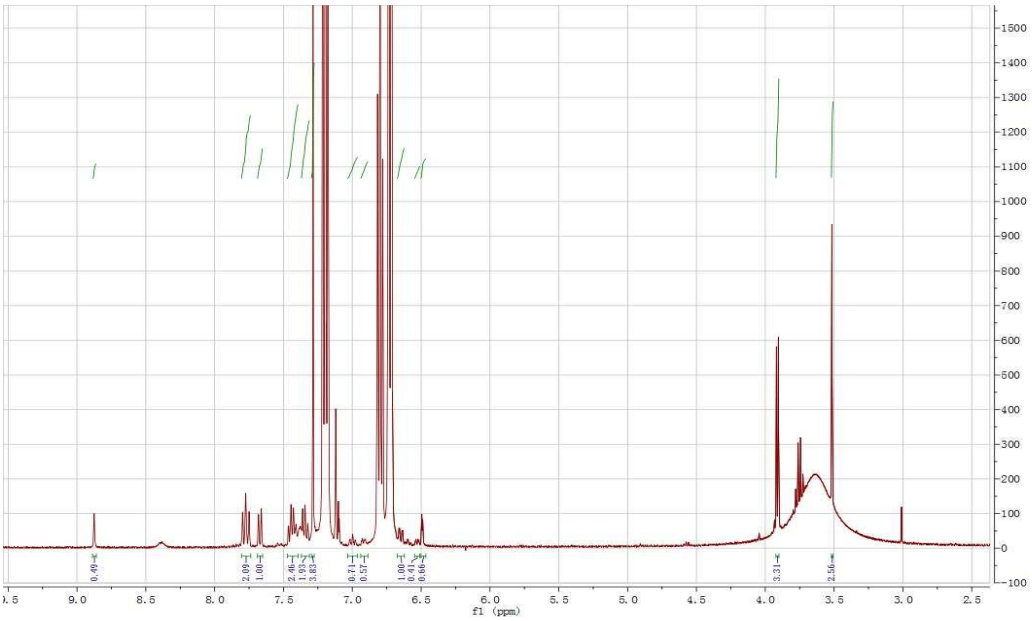


Fig. 29 ^{1}H NMR spectrum of 4g

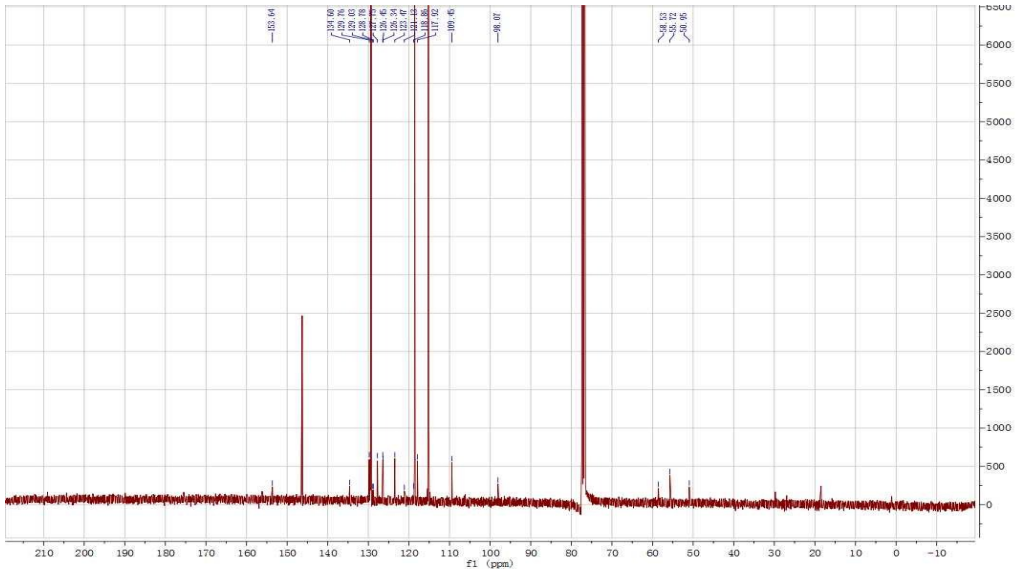


Fig. 30 ^{13}C NMR spectrum of 4g

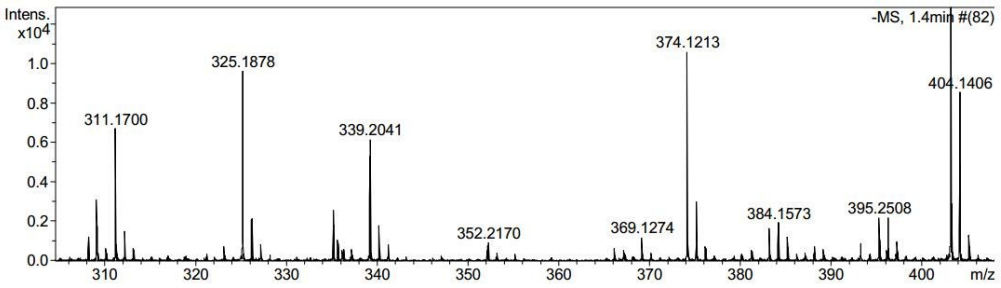


Fig. 31 HRMS spectrum of 4g

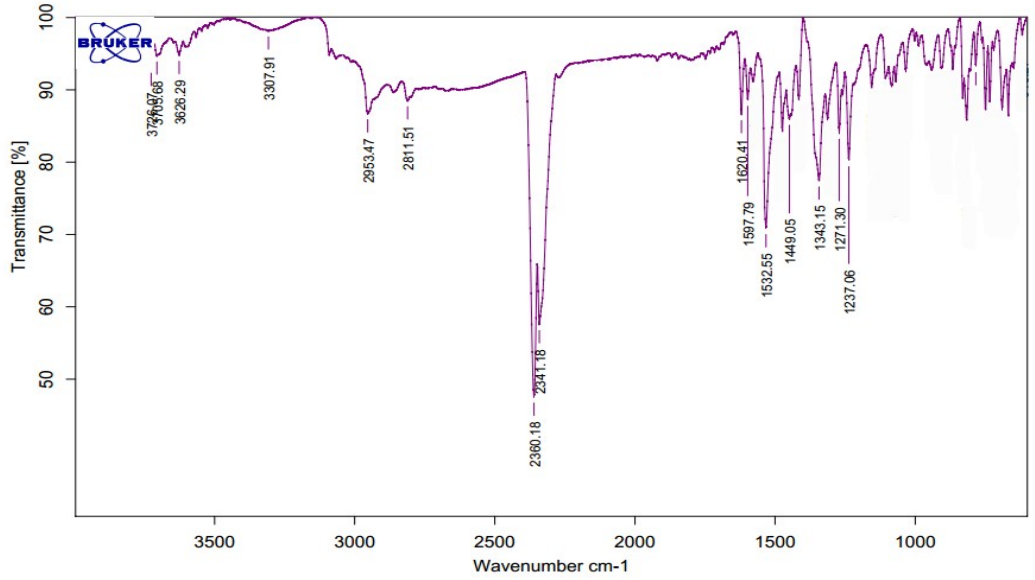


Fig. 32 IR spectrum of 4h

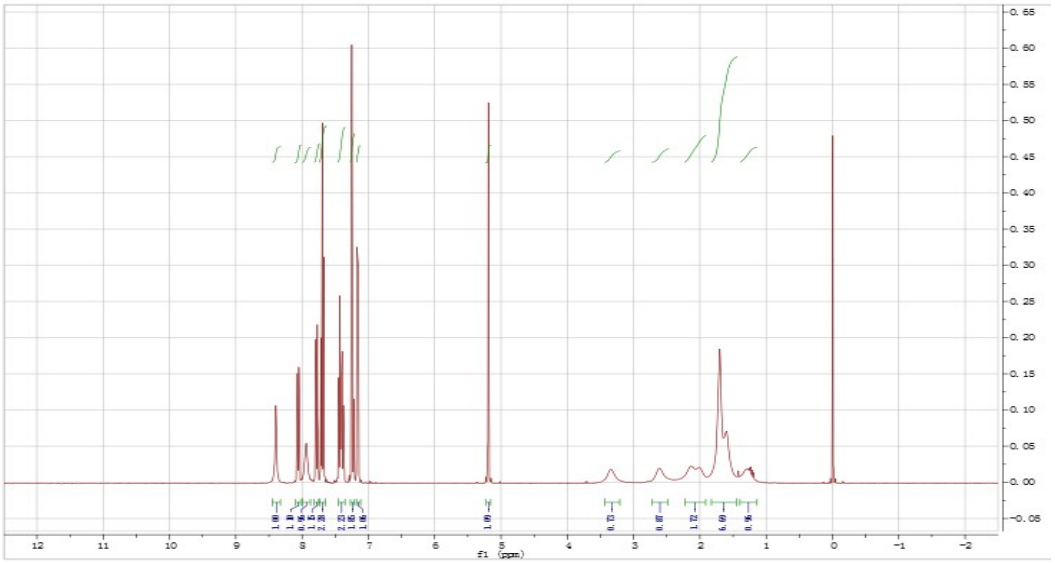


Fig. 33 ^{1}H NMR spectrum of 4h

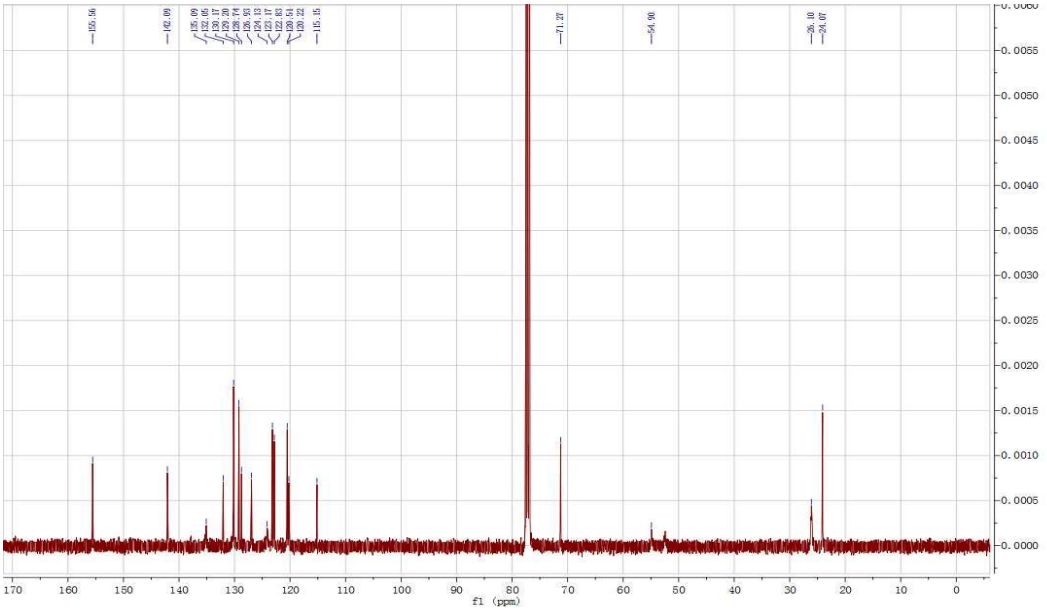


Fig. 34 ^{13}C NMR spectrum of 4h

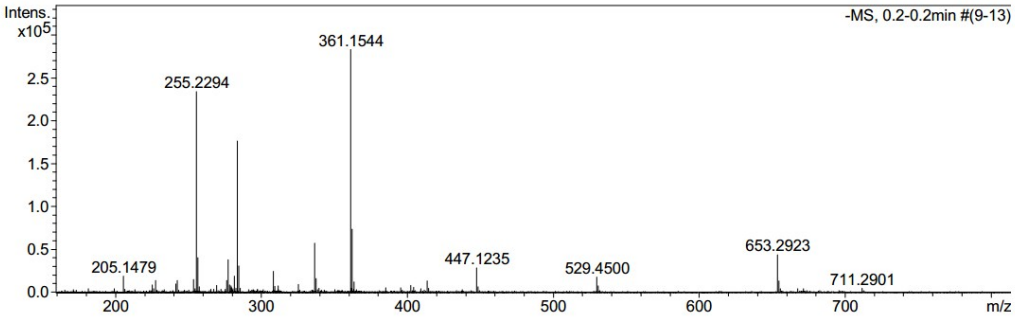


Fig. 35 HRMS spectrum of 4h

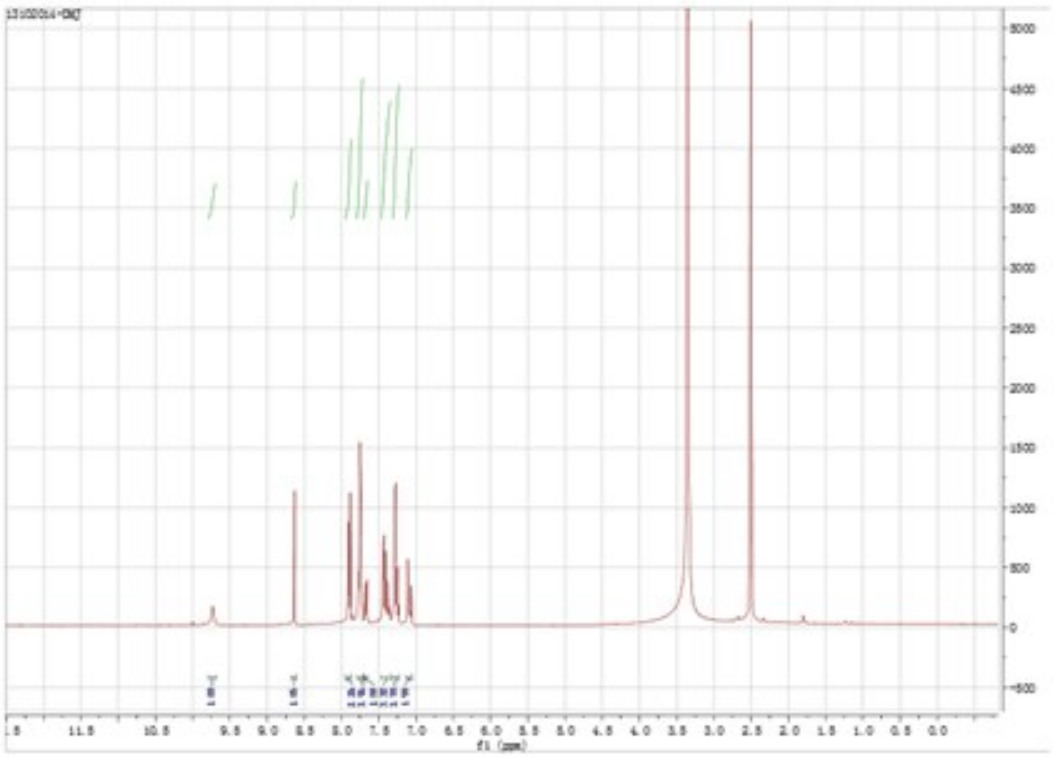


Fig. 36 ^{1}H NMR spectrum of 4i

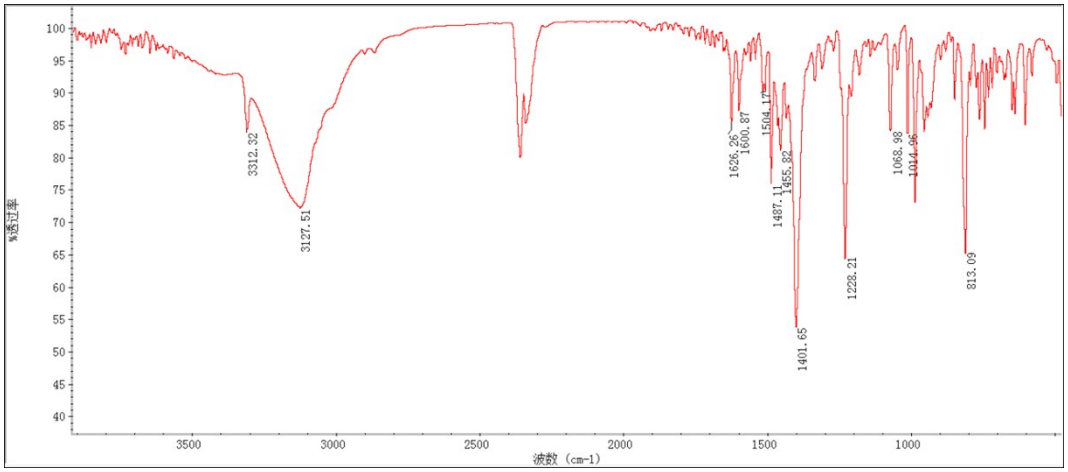


Fig. 37 IR spectrum of 4j

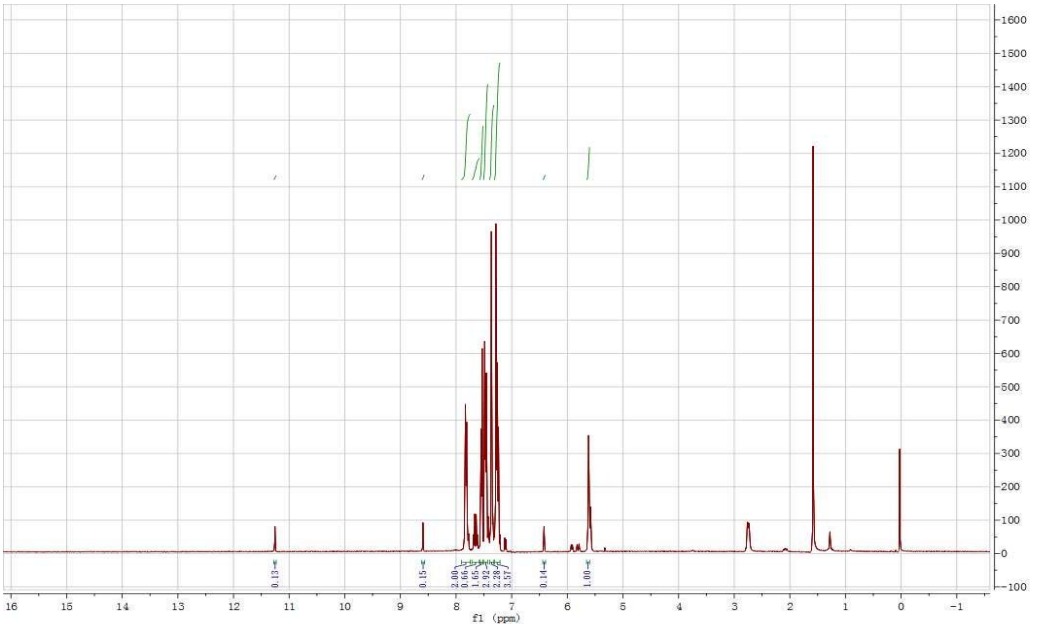


Fig.38 ^{1}H NMR spectrum of 4j

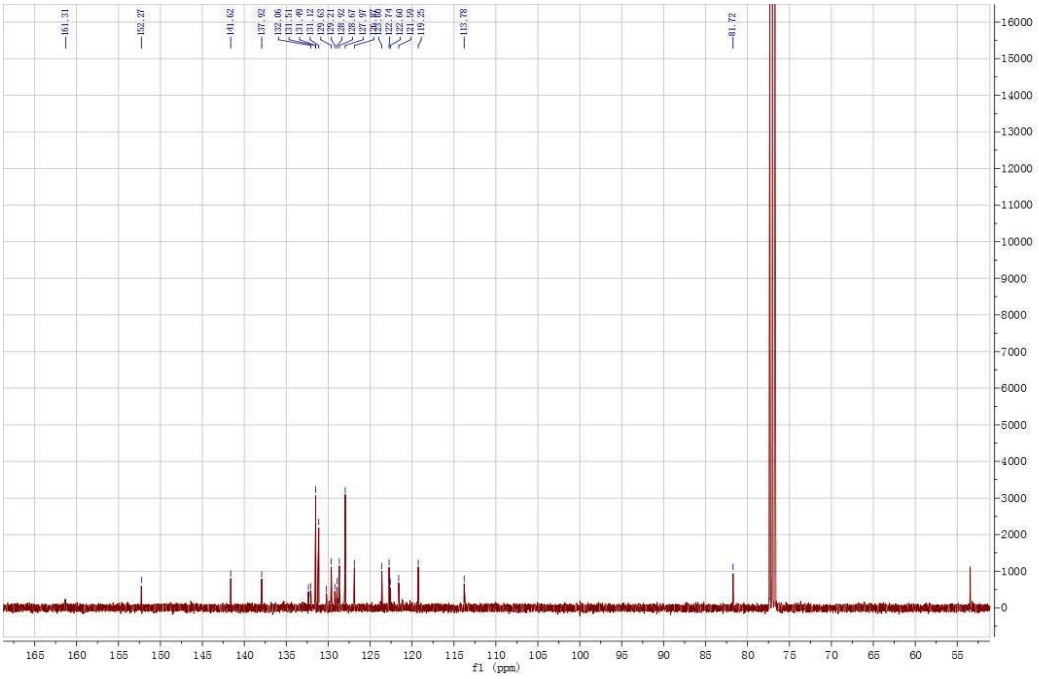


Fig. 39 ^{13}C NMR spectrum of 4j

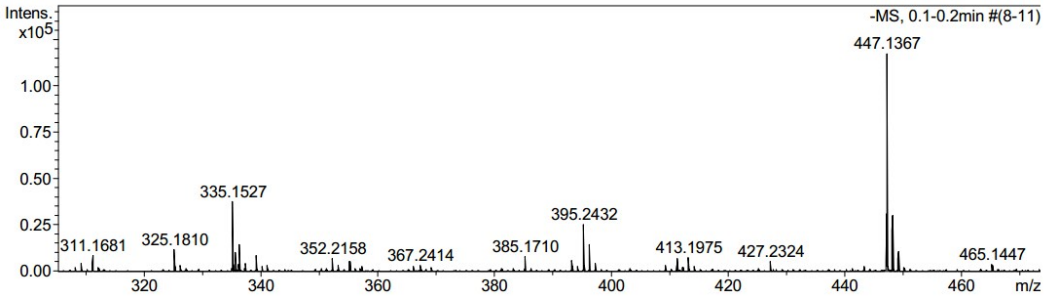


Fig. 40 HRMS spectrum of 4j

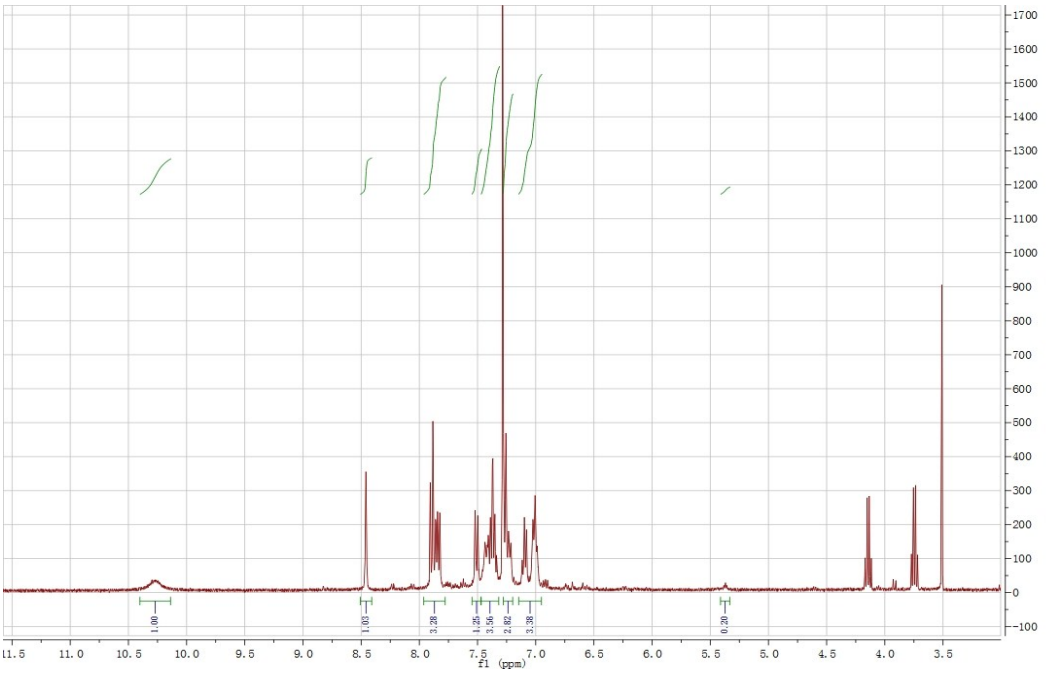


Fig. 41 ^{1}H NMR spectrum of 4k

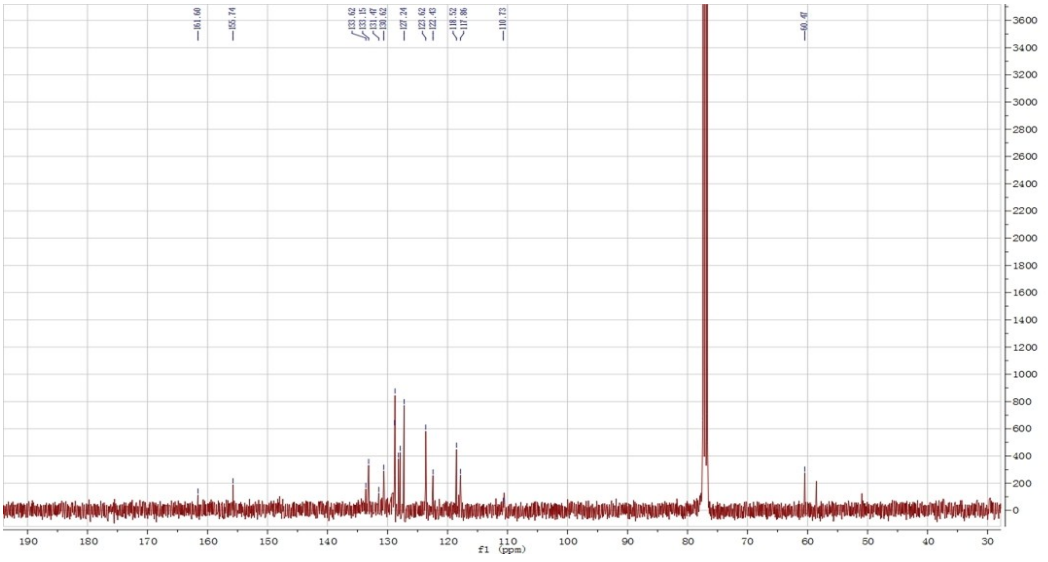
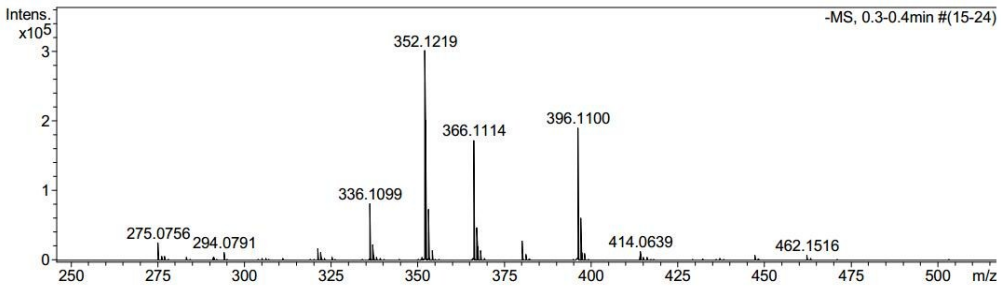


Fig. 42 ^{13}C NMR spectrum of 4k



14

Fig. 43 HRMS spectrum of 4k

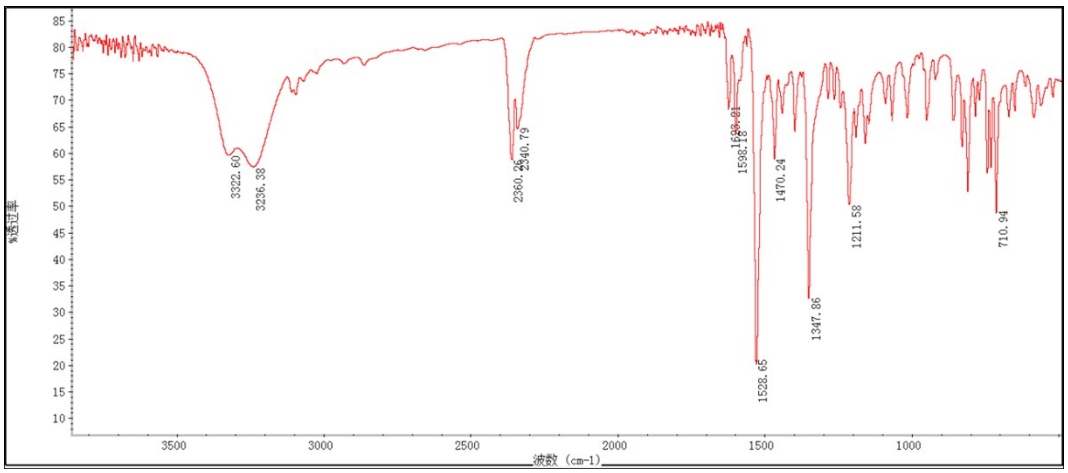


Fig.44 IR spectrum of 4l

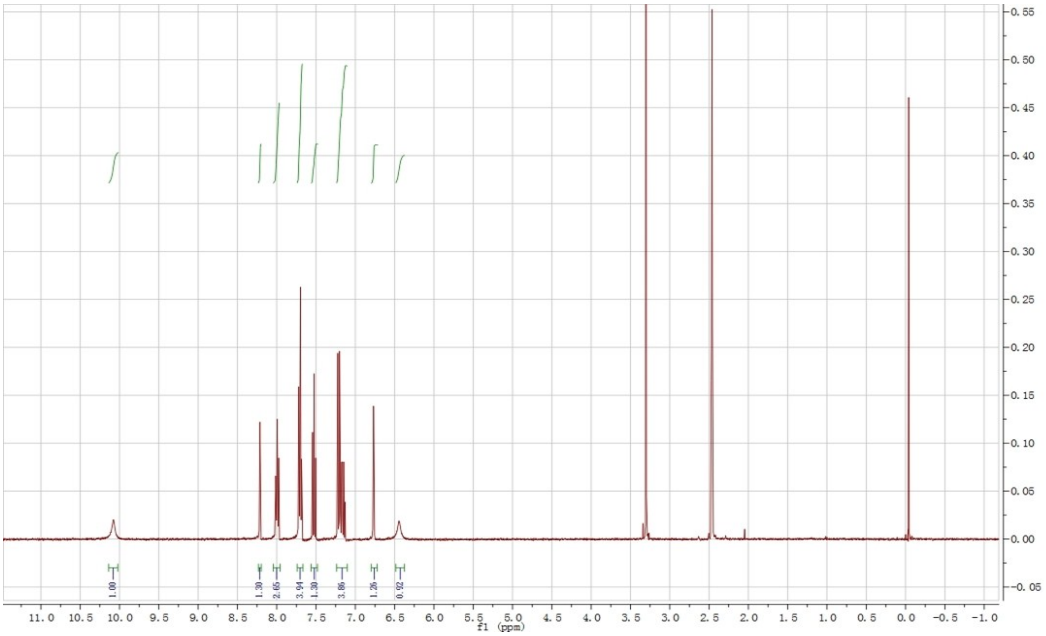


Fig. 45 ^{1}H NMR spectrum of 4l

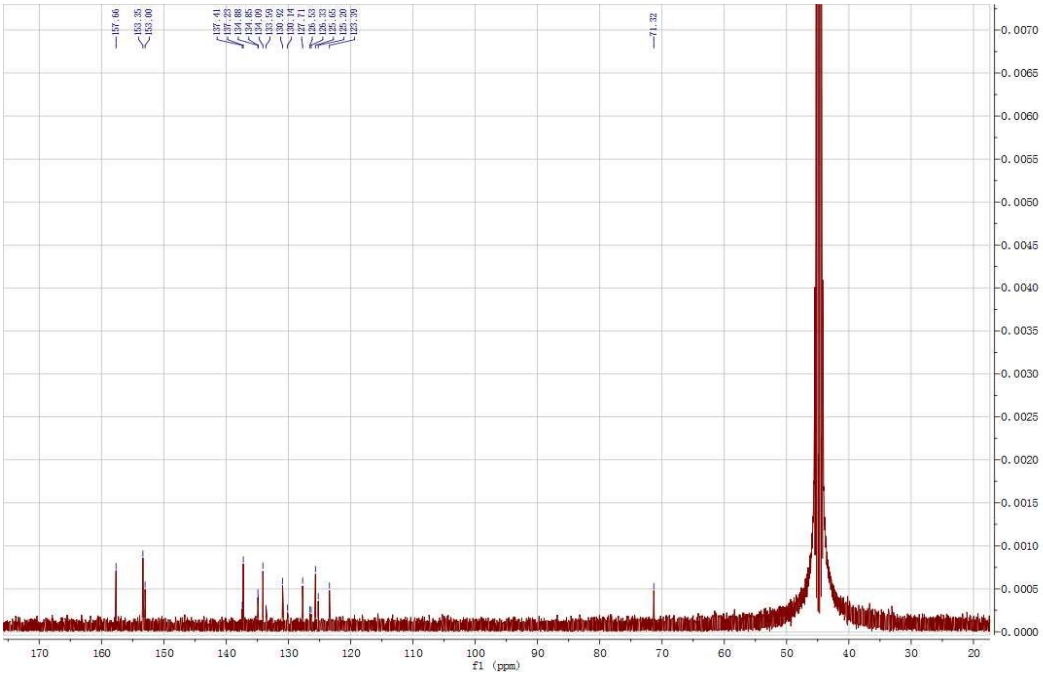


Fig. 46 ^{13}C NMR spectrum of 4l

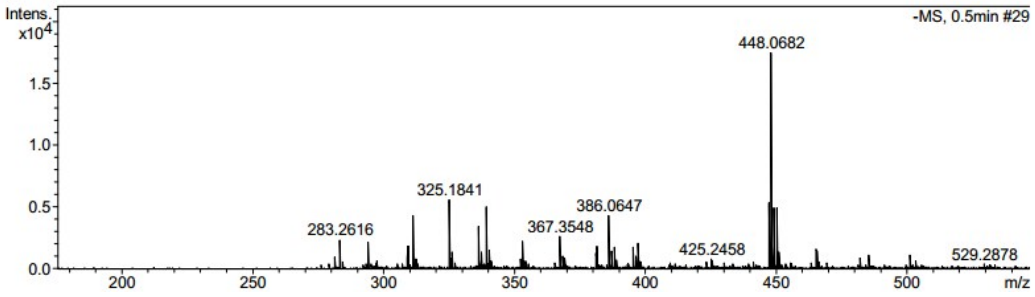


Fig.47 HRMS spectrum of 4l

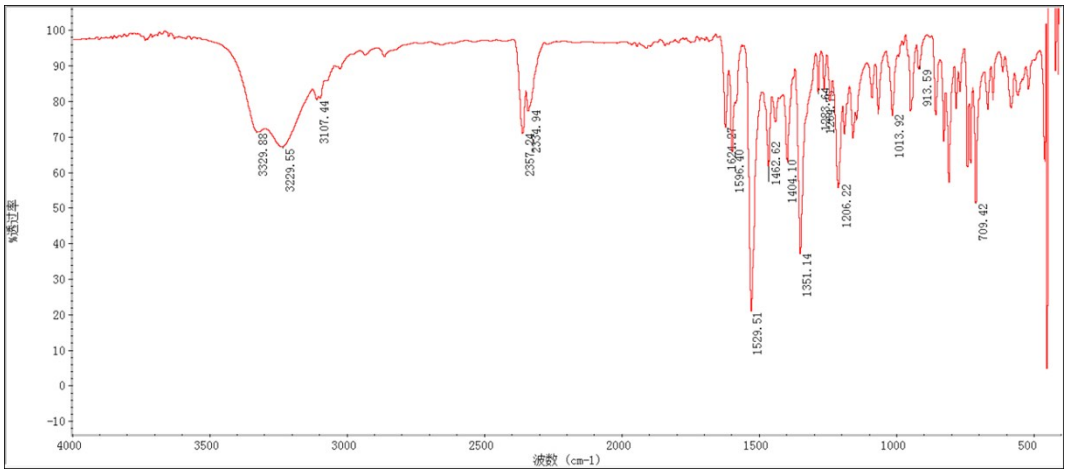


Fig. 48 IR spectrum of 4m

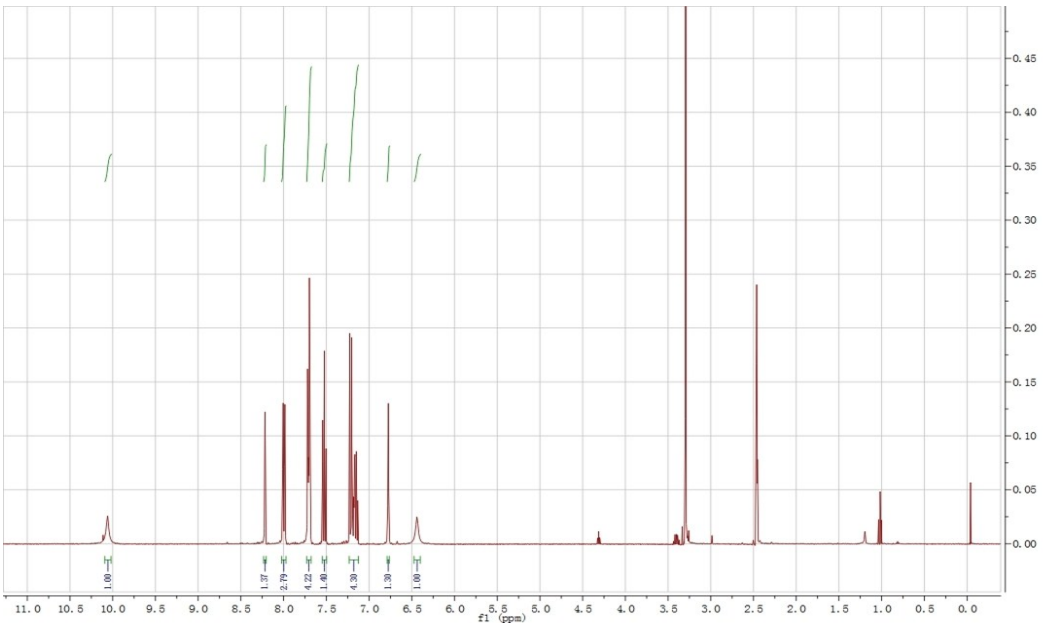


Fig. 49 ^{1}H NMR spectrum of 4m

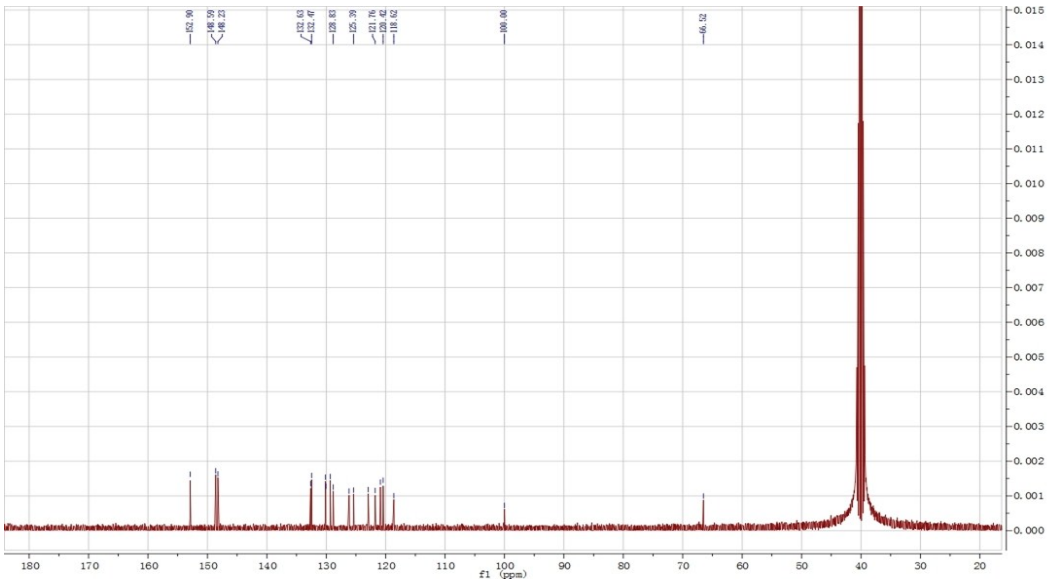


Fig. 50 ^{13}C NMR spectrum of 4m

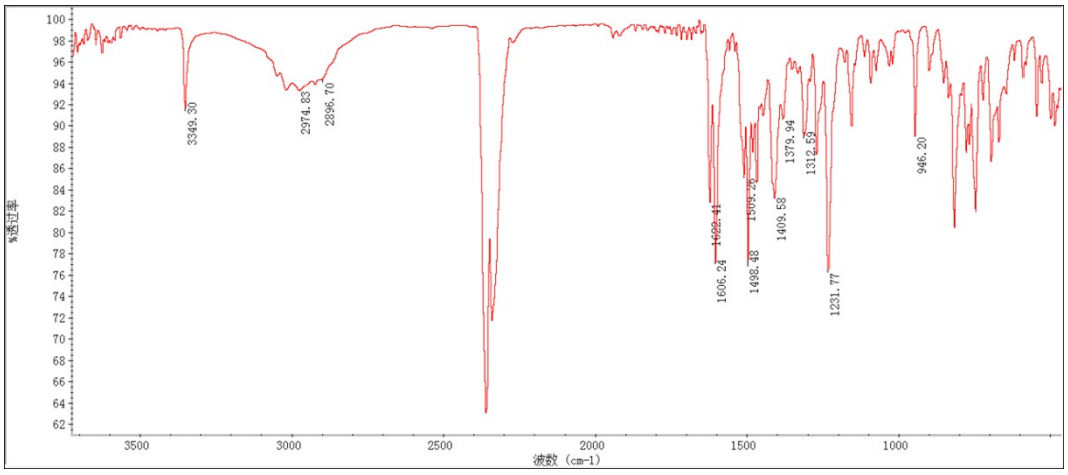


Fig. 51 IR spectrum of 4n

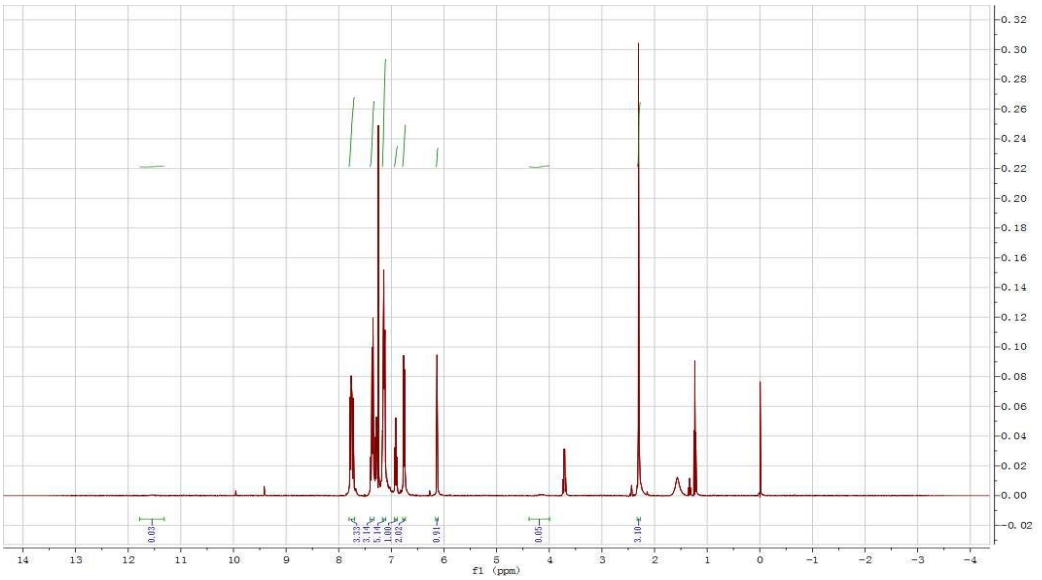


Fig. 52 ^{1}H NMR spectrum of 4n

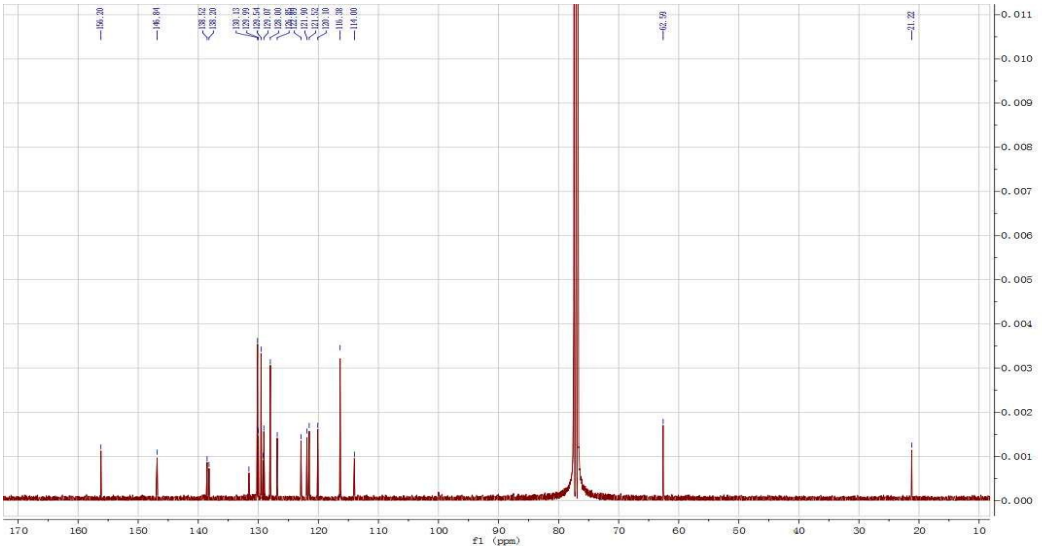


Fig. 53 ^{13}C NMR spectrum of 4n

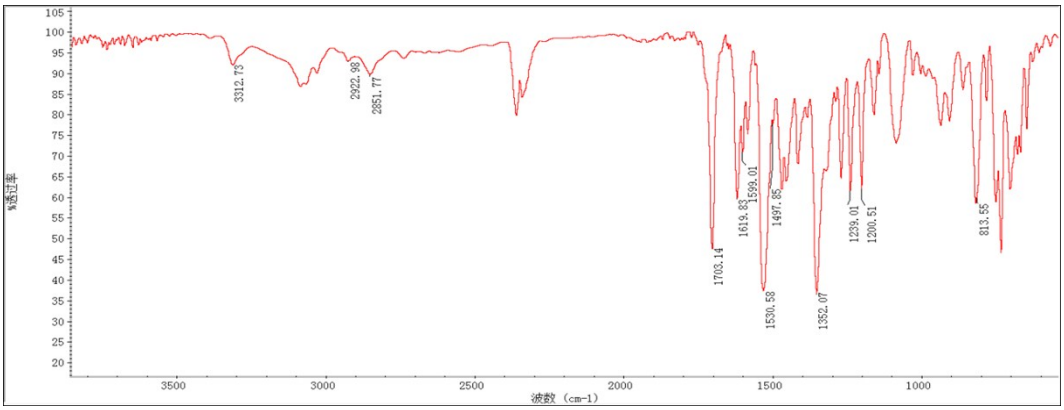


Fig. 54 IR spectrum of 4o

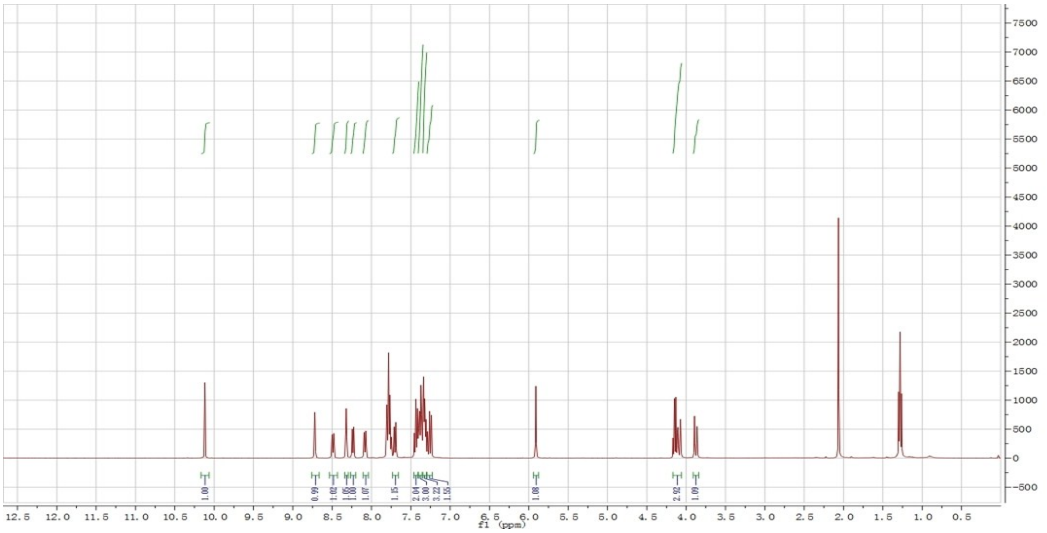


Fig.55 ^{1}H NMR spectrum of 4o

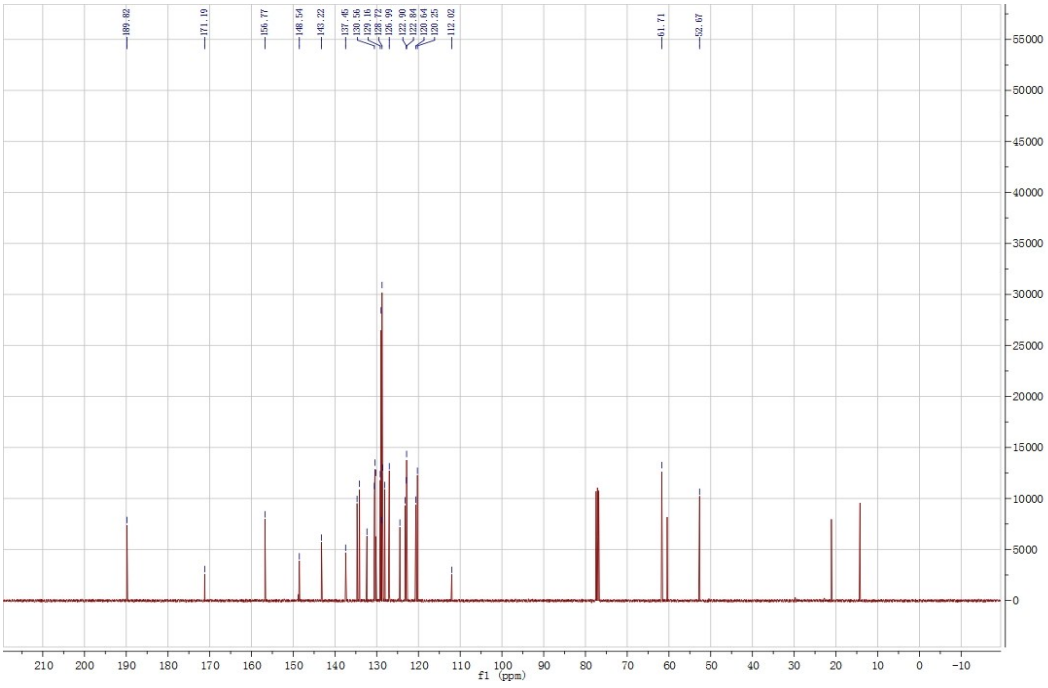


Fig. 56 ^{13}C NMR spectrum of 4o

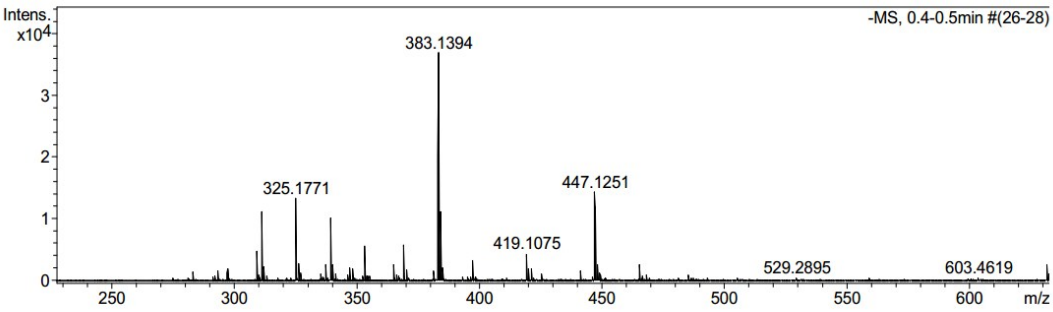


Fig. 57 HRMS spectrum of 4o

4. References

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