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Supporting Information

Eco-friendly grinding synthesis of double layered nano material and correlation between basicity, calcinations and catalytic activity in green synthesis of novel fused pyrimidines

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Characterization Data

7-phenylchromeno[4,3-d]benzothiazolo[3,2-a]pyrimidin-6(7H)-one (**4a**)

White powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.93 (s, 1H, -CH), 7.05-7.17 (m, 3H, Ar-H), 7.17 (t, 2H, J = 8.0 Hz, Ar-H), 7.22-7.30 (m, 4H, Ar-H), 7.38-7.51 (m, 4H); ^{13}C NMR (125 MHz, DMSO d_6): 68, 103, 114, 115, 115, 119, 122, 122, 123, 123, 124, 126, 126, 127, 127, 131, 141, 152, 164, 167, 168; ESI-MS: m/z Calculated for $C_{23}H_{14}N_2O_2S$ 382.43 Found $[M+H]^+$ 383.

7-styrylchromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4b**)

Light brown crystal, mp 180-182; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 5.63 (s, 1H, -CH), 6.26 (d, 1H, =CH), 6.70 (d, 1H, =CH), 7.19-7.38 (m, 8H, Ar-H), 7.58 (t, 2H, J = 7.5 Hz, Ar-H), 7.84 (t, 3H, J = 8.0 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 65, 101, 102, 103, 112, 114, 116, 117, 120, 122, 124, 128, 129, 131, 133, 137, 144, 151, 152, 157, 161, 163, 164, 167; ESI-MS: m/z Calculated for $C_{25}H_{16}N_2O_2S$ 408.47 Found $[M+H]^+$ 409.

7-(4-chlorophenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4c**)

Light yellow powder, mp 188-189; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.39 (s, 1H, -CH), 6.94-7.09 (m, 2H, Ar-H), 7.16 (d, 1H, J = 8.0 Hz, Ar-H), 7.21 (t, 1H, J = 7.5 Hz, Ar-H), 7.39-7.62 (m, 4H, J = 8.0 Hz), 7.82 (d, 2H, J = 8.5 Hz, Ar-H), 7.90 (d, 2H, J = 7.0 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 68, 106, 113, 118, 118, 121, 125, 125, 125, 126, 129, 130, 134, 146, 155, 163, 167, 170; ESI-MS: m/z Calculated for $C_{23}H_{13}ClN_2O_2S$ 416.88 Found $[M+H]^+$ 418.

7-(4-dimethylaminophenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4d**)

Reddish brown powder, mp 160-162; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.08 (s, 6H, -N(CH_3)$_2$), 6.26 (s, 1H, -CH), 7.15-7.41 (m, 8H, Ar-H), 7.59 (t, 2H, J = 7.5 Hz, Ar-H), 7.86 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 45, 65, 103, 111, 114, 115, 118, 118, 119, 121, 123, 124, 125, 126, 128, 131, 131, 141, 152, 164, 167; ESI-MS: m/z Calculated for $C_{25}H_{19}N_3O_2S$ 425.50 Found $[M+H]^+$ 426.

7-(3-hydroxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4e**)

Off white powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.20 (s, 1H, -CH), 6.45-6.56 (m, 3H, Ar-H), 6.94 (t, 1H, J = 7.5 Hz, Ar-H), 7.22-7.28 (m, 4H, Ar-H), 7.41-7.51 (m, 4H, Ar-H), 9.26 (br, 1H, OH); ^{13}C NMR (125 MHz, DMSO d_6): 65, 103, 112, 113, 114, 115, 117, 119, 122, 122, 123, 123, 124, 127, 128, 131, 141, 143, 152, 157, 164, 167, 168; ESI-MS: m/z Calculated for $C_{23}H_{14}N_2O_3S$ 398.43 Found $[M+H]^+$ 399.

7-(4-methoxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4f**)

Off white powder, mp 234-236 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.73 (s, 3H, OCH_3), 6.24 (s, 1H, -CH), 6.80 (d, 2H, J = 7.0 Hz, Ar-H), 7.16-7.43 (m, 4H, Ar-H), 7.51 (t, 2H, J = 7.5 Hz, Ar-H), 7.68 (d, 2H, J = 7.0 Hz, Ar-H), 7.79 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 56, 66, 103, 118, 121, 125, 128, 133, 133, 133, 134, 138, 141, 142, 145, 150, 152, 164, 166; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_3S$ 412.46 Found $[M+H]^+$ 413.

7-(4-methylphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4g**)

Yellow powder, mp >250 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.80 (s, 3H, CH_3), 6.24 (s, 1H, -CH), 7.19-7.40 (m, 8H, Ar-H), 7.51 (t, 2H, J = 7.5 Hz, Ar-H), 7.80 (d, 2H, J = 7.0 Hz, Ar-H);

^{13}C NMR (125 MHz, DMSO d_6): 27, 65, 102, 111,116, 119, 119, 123, 123, 124, 124, 128, 131, 140, 145, 152, 154, 164, 167; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_2S$ 396.46 Found $[M]^+$ 397.

7-(4-nitrophenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4h**)

Off white powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.17 (s, 1H, -CH), 7.20-7.25 (m, 5H, Ar-H), 7.29 (d, 2H, J = 7.0 Hz, Ar-H), 7.50 (t, 2H, J = 7.5 Hz, Ar-H), 7.81 (d, 2H, J = 7.5 Hz, Ar-H), 8.13 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 67, 103, 111, 115, 118, 122, 123, 123, 124, 124, 126, 128, 128, 131, 144, 152, 161, 164, 167; ESI-MS: m/z Calculated for $C_{23}H_{13}N_3O_4S$ 427.43 Found $[M+H]^+$ 429.

7-(2-methylphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4i**)

Off white powder, mp >250 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.61 (s, 3H, CH_3), 6.37 (s, 1H, -CH), 7.21-7.25 (m, 4H, Ar-H), 7.29 (t, 2H, J = 7.0 Hz, Ar-H), 7.46 (t, 2H, J = 7.5 Hz, Ar-H), 7.86 (d, 2H, J = 7.5 Hz, Ar-H), 8.13 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 38, 67, 103, 115, 118, 123, 123, 124, 124, 128, 128, 130, 131, 140, 144, 150, 152, 164, 166; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_2S$ 396.46 Found $[M+H]^+$ 397.

7-(2-methoxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4j**)

Off white powder, mp 234-236 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.73 (s, 3H, OCH_3), 6.24 (s, 1H, -CH), 6.78 (d, 2H, J = 7.0 Hz, Ar-H), 7.19-7.31 (m, 4H, Ar-H), 7.53 (t, 2H, J = 7.5 Hz, Ar-H), 7.71 (d, 2H, J = 7.0 Hz, Ar-H), 7.79 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 55, 66, 106, 115, 119, 122, 122, 123, 123, 126, 131, 134, 135, 143, 149, 152, 156, 167, 169; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_3S$ 412.46 Found $[M+H]^+$ 413.

7-(4-hydroxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4k**)

Off white powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.16 (s, 1H, -CH), 6.54 (d, 1H, J = 7.5 Hz, Ar-H), 6.86 (d, 1H, J = 7.5 Hz, Ar-H), 7.19-7.26 (m, 4H, Ar-H), 7.35- 7.51 (m, 4H, Ar-H), 7.80 (d, 2H, J = 7.5 Hz, Ar-H), 8.91 (br, 1H, OH); ^{13}C NMR (125 MHz, DMSO d_6): 65, 101, 109, 111, 112, 113, 115, 117, 120, 120, 120, 121, 124, 125, 128, 138, 141, 149, 159, 154, 162, 164, 166, 167; ESI-MS: m/z Calculated for $C_{23}H_{14}N_2O_3S$ 398.43 Found $[M+H]^+$ 399.

7-(4-chloroyphenyl)-11-methylbenzo[4,5]thiazolo[3,2-*a*]chromeno[4,3-d]pyrimidin-6(7*H*)-one (**4l**)

Off white powder, mp 184-186 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.29 (s, 3H, CH_3), 6.29 (s, 1H, -CH), 7.14 (d, 2H, J = 7.5 Hz, Ar-H), 7.24-7.35 (m, 4H, Ar-H), 7.55-7.59 (m, 2H, Ar-H), 7.88 (d, 2H, J = 7.5 Hz, Ar-H), 7.93 (d, 1H, J = 8.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 23, 67, 104, 116, 116, 117, 117, 123, 128, 129, 129, 129, 130, 131, 132,138, 139, 152, 164, 166; ESI-MS: m/z Calculated for $C_{24}H_{15}ClN_2O_2S$ 430.91 Found $[M+H]^+$ 431.

11-nitro-7-phenyl-11-methylbenzo[4,5]thiazolo[3,2-*a*]chromeno[4,3-d]pyrimidin-6(7*H*)-one (**4m**)

Off white powder, mp 215-217 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.37 (s, 1H,-CH), 7.27 (t, 2H, J = 8.0 Hz, Ar-H), 7.33 (d, 2H, J = 8.5 Hz, Ar-H), 7.40 (d, 2H, J = 8.5 Hz, Ar-H), 7.56 (t, 2H, J = 8.0 Hz, Ar-H), 7.84 (d, 2H, J = 8.0 Hz, Ar-H), 8.09 (d, 2H, J = 9.0 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 69, 103, 111, 115, 118, 122, 122, 123, 127, 130, 131, 139, 145, 149, 151, 157, 164, 166; ESI-MS: m/z Calculated for $C_{23}H_{13}N_3O_4S$ 427.43 Found $[M]^+$ 427.5.

2-chloro-7-(p-tolyl)benzo[4,5]thiazolo[3,2-a]chromeno[4,3-d]pyrimidin-6(7H)-one **(4n)**

Off white powder, mp 225-227 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.39 (s, 3H, CH_3), 6.30 (s, 1H, -CH), 6.89-6.94 (dd, 2H, J = 8.0, 8.5 Hz, Ar-H), 7.06 (t, 2H, J = 8.0 Hz), 7.25-7.36 (m, 4H, Ar-H), 7.70 (d, 1H, J = 7.5 Hz, Ar-H), 8.10 (d, 2H, J = 8.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 21, 69, 102, 117, 118, 121, 121, 121, 126, 127, 128, 129, 129, 129, 130, 1132, 134, 143, 150, 151, 161, 163, 167; ESI-MS: m/z Calculated for $C_{24}H_{15}ClN_2O_2S$ 430.90 Found $[M]^+$ 430.9.

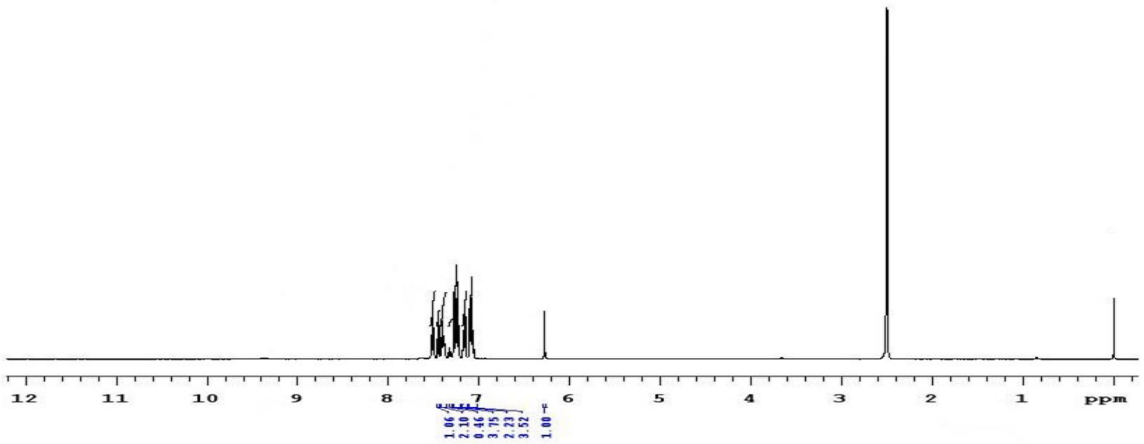
7-(4-bromophenyl)-2-chloro-11-methylbenzo[4,5]thiazolo[3,2-a]chromeno[4,3-d]pyrimidin-6(7H)-one **(4o)**

Off white powder, mp >250 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.23 (s, 3H, CH_3), 6.29 (s, 1H, -CH), 7.09-7.23 (m, 8H, Ar-H), 7.32 (t, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 22, 66, 104, 107, 113, 117, 125, 126, 128, 129, 130, 130, 134, 143, 151, 155, 162, 164; ESI-MS: m/z Calculated for $C_{24}H_{14}BrClN_2O_2S$ 509.80 Found $[M]^+$ 509.8.

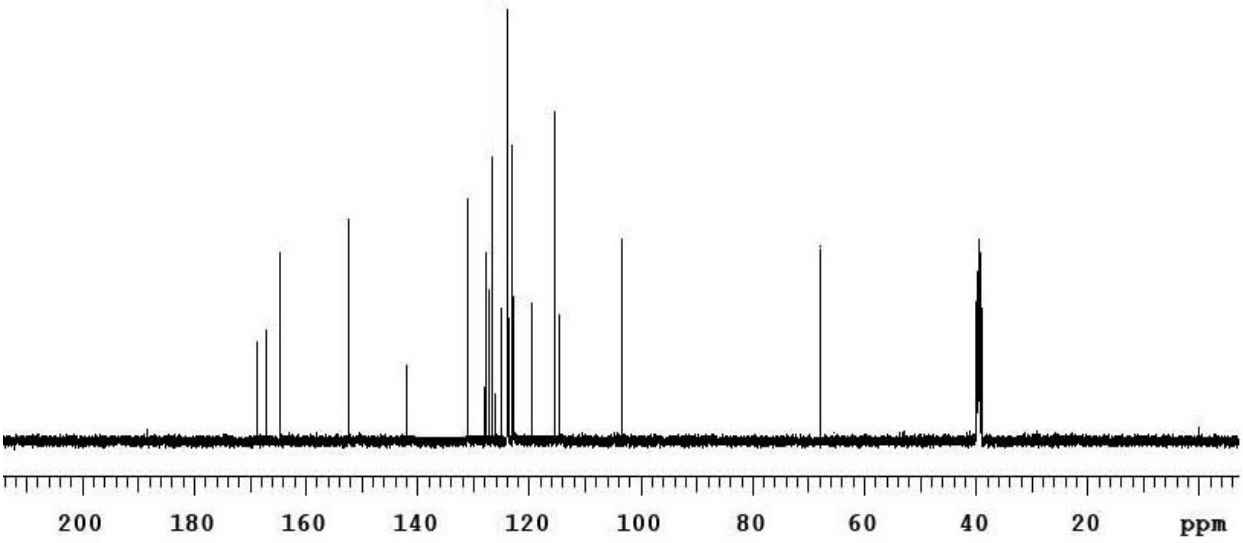
Scanned NMR Spectra

Compound 4a

H NMR

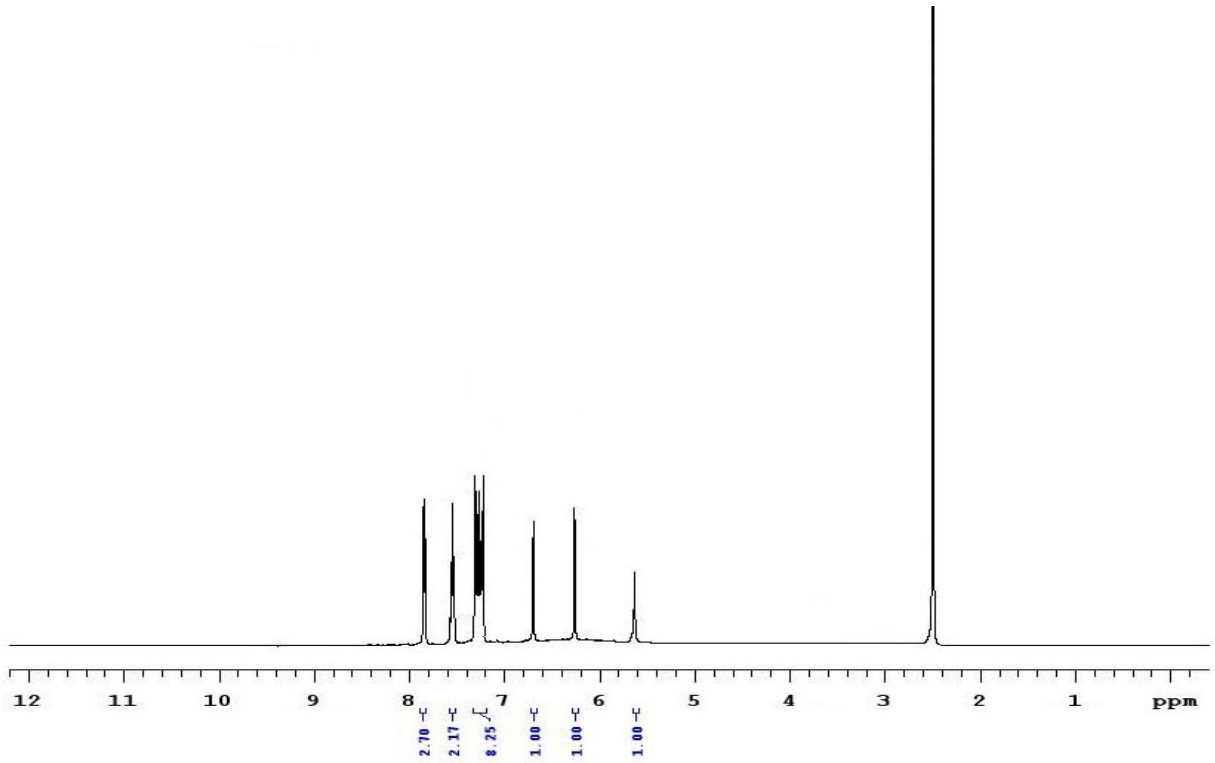


^{13}C NMR

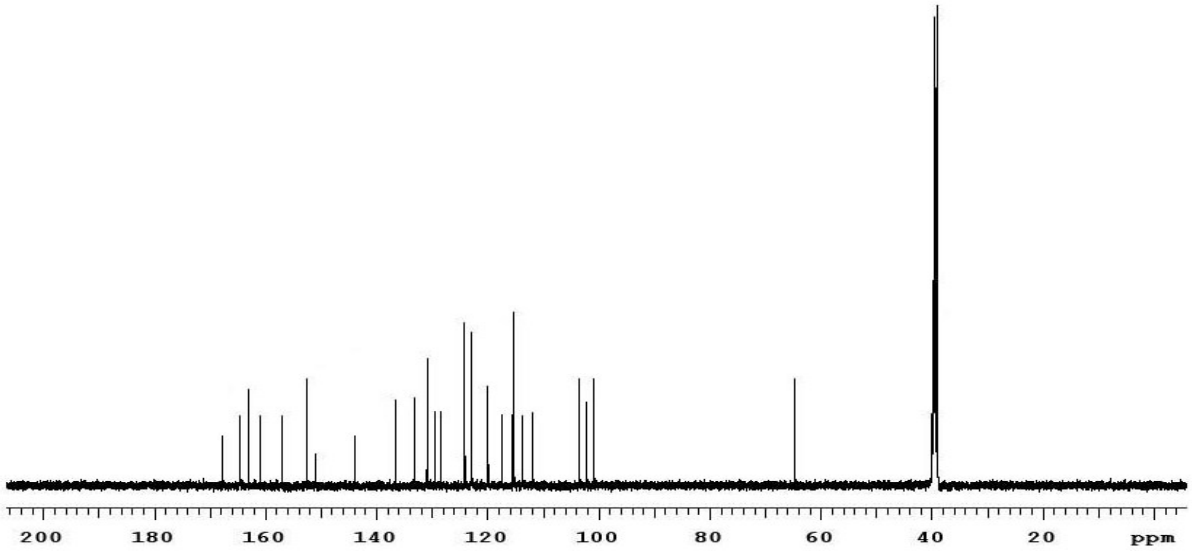


Compound 4b

H NMR

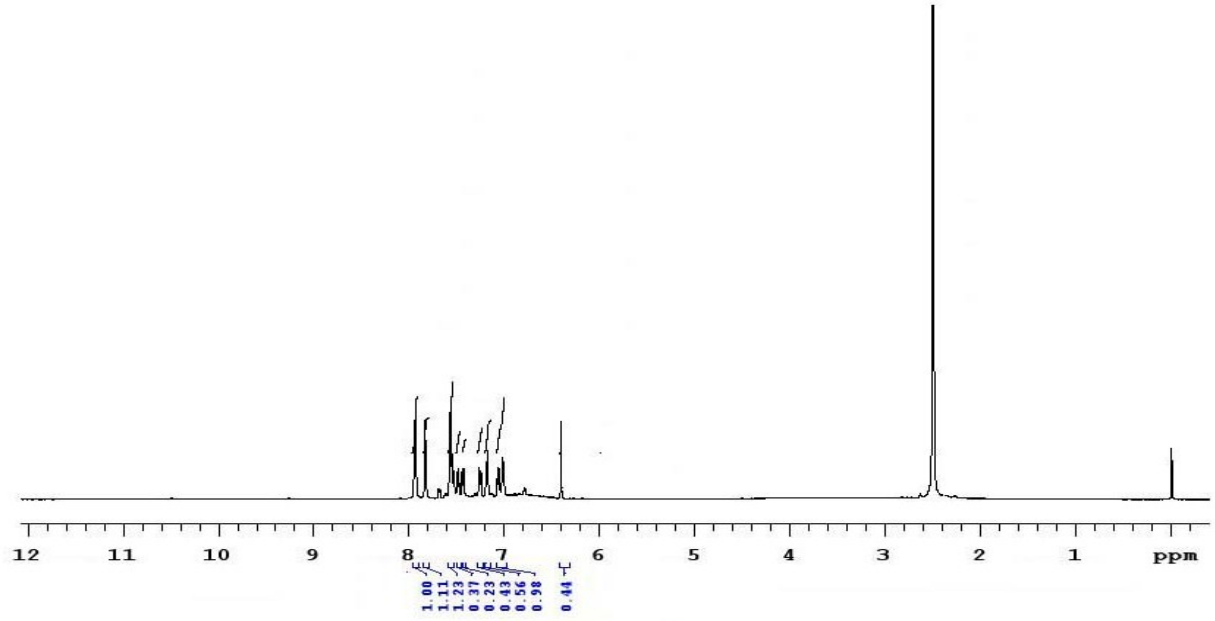


^{13}C NMR

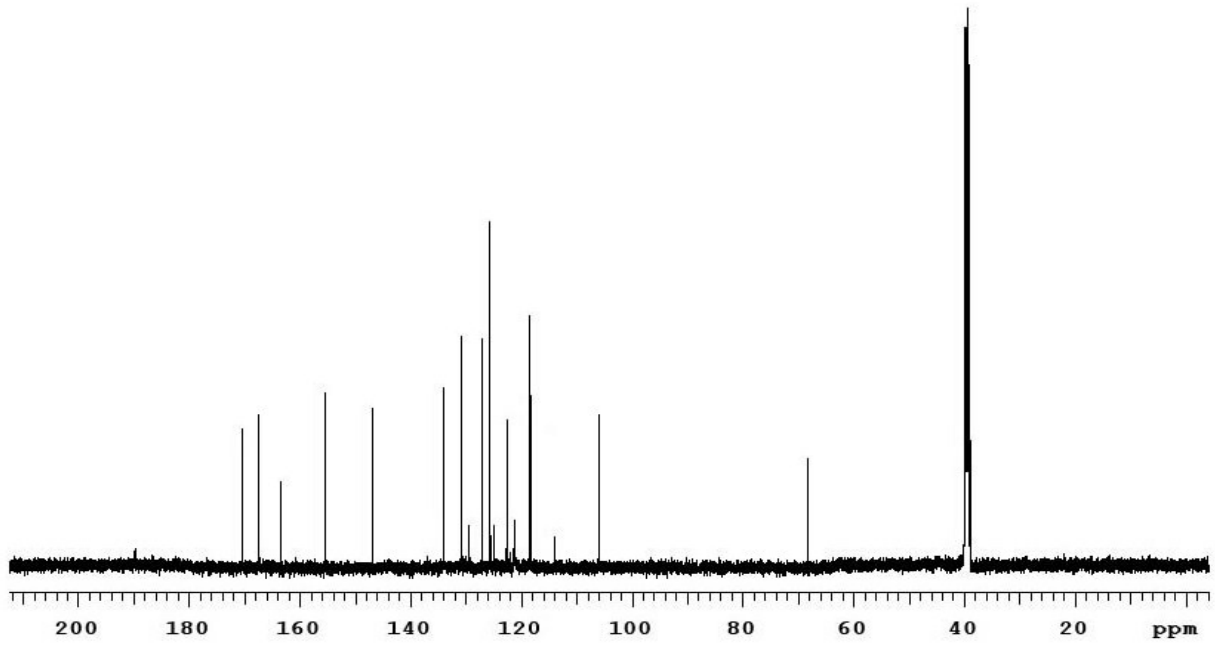


Compound 4c

H NMR



^{13}C NMR



Compound 4d

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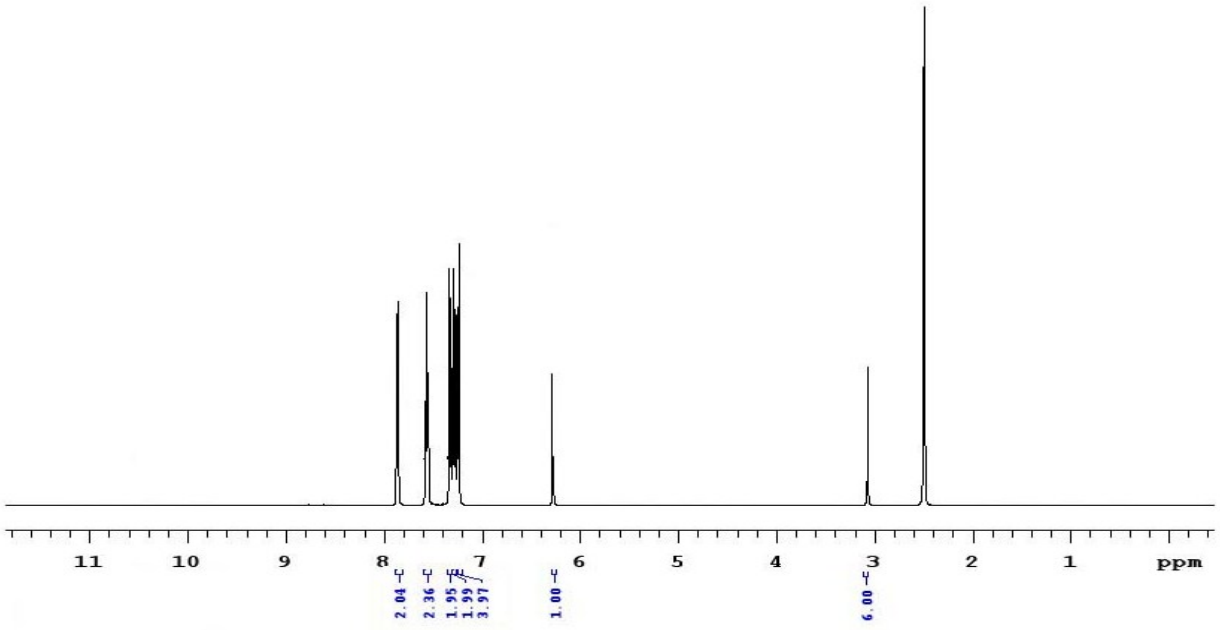
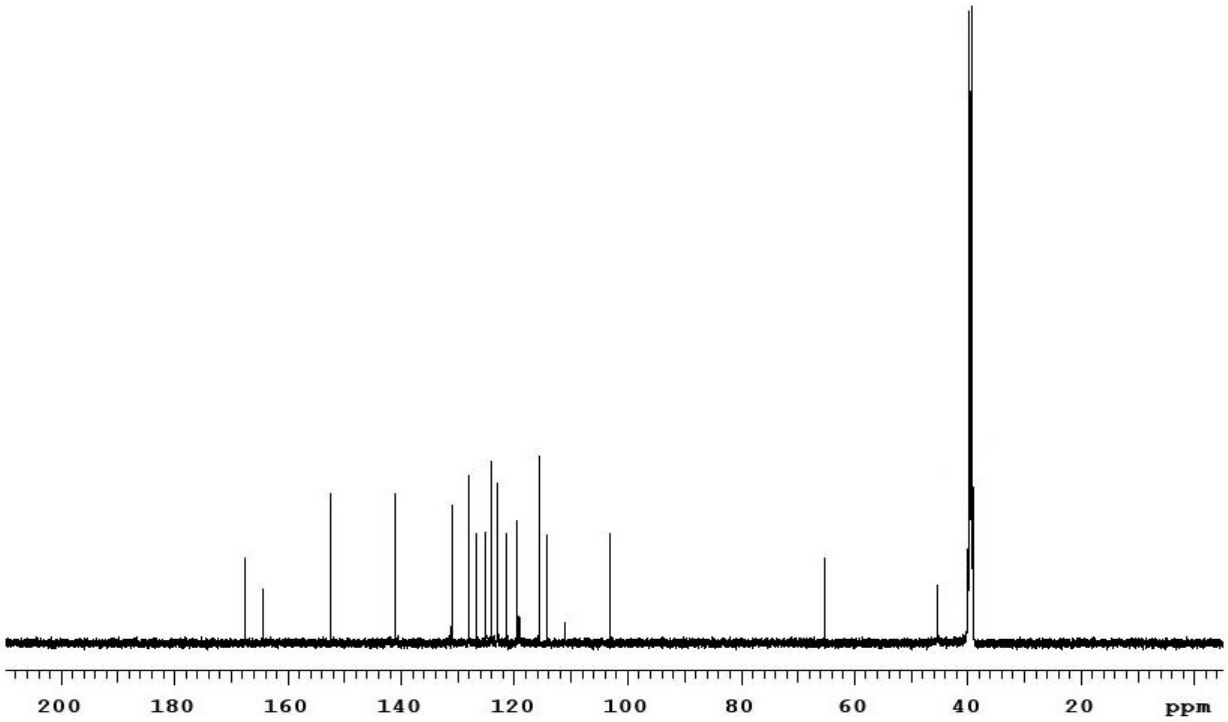


^{13}C NMR



Compound 4e

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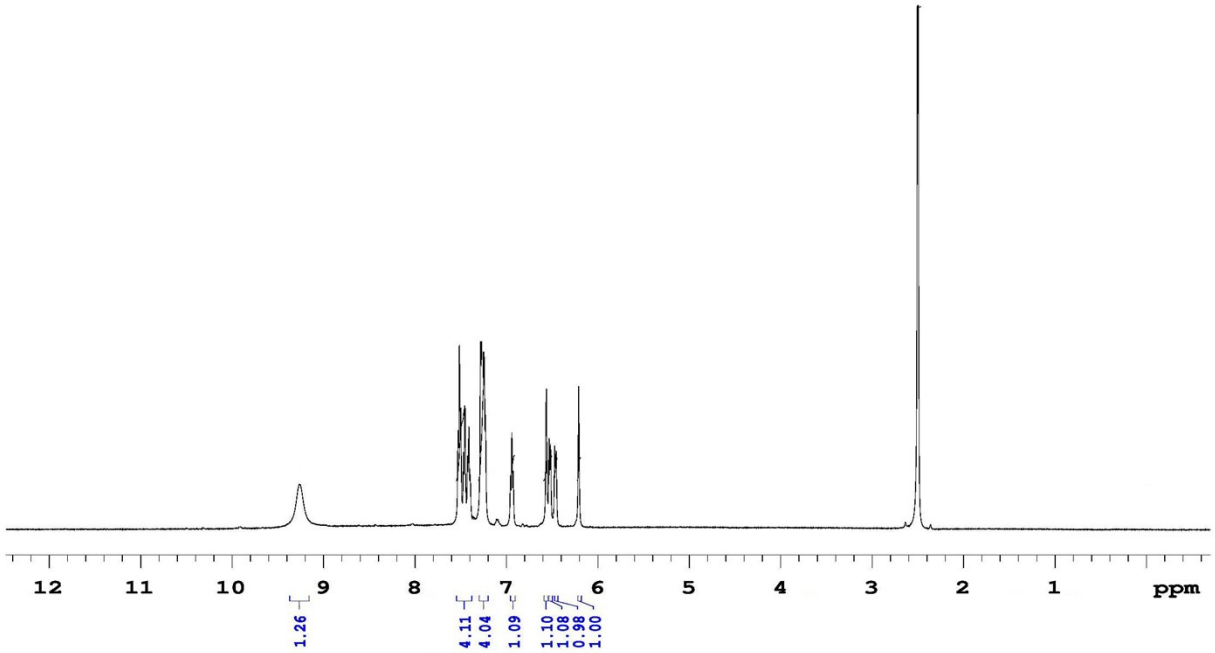

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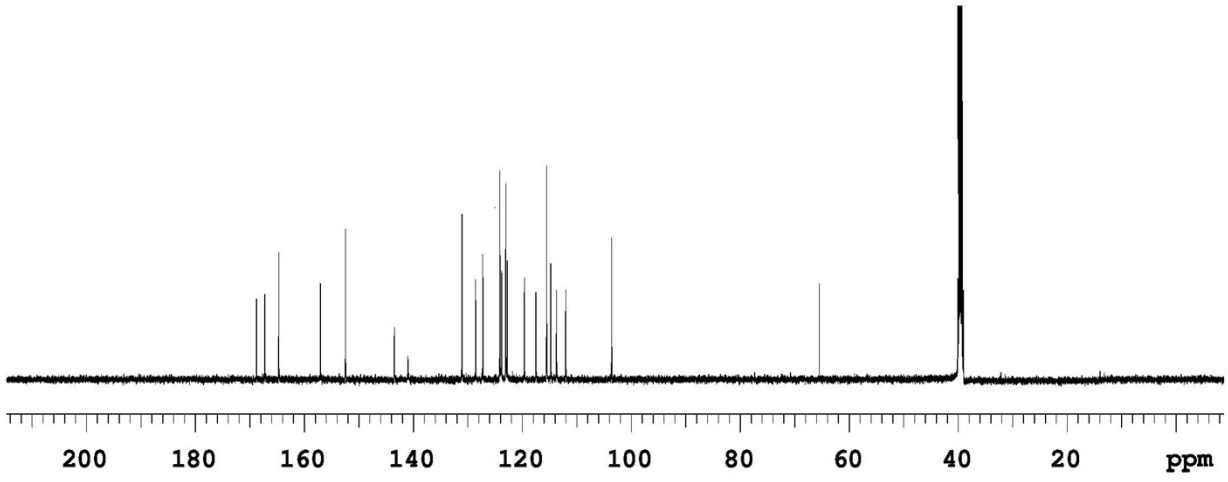

Compound 4f

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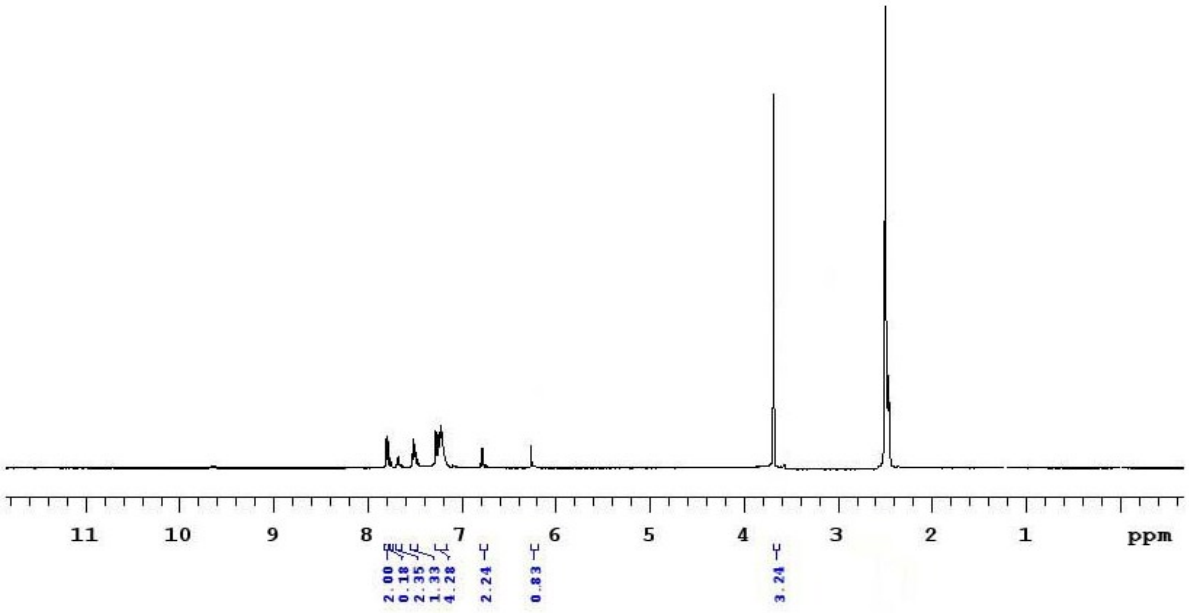

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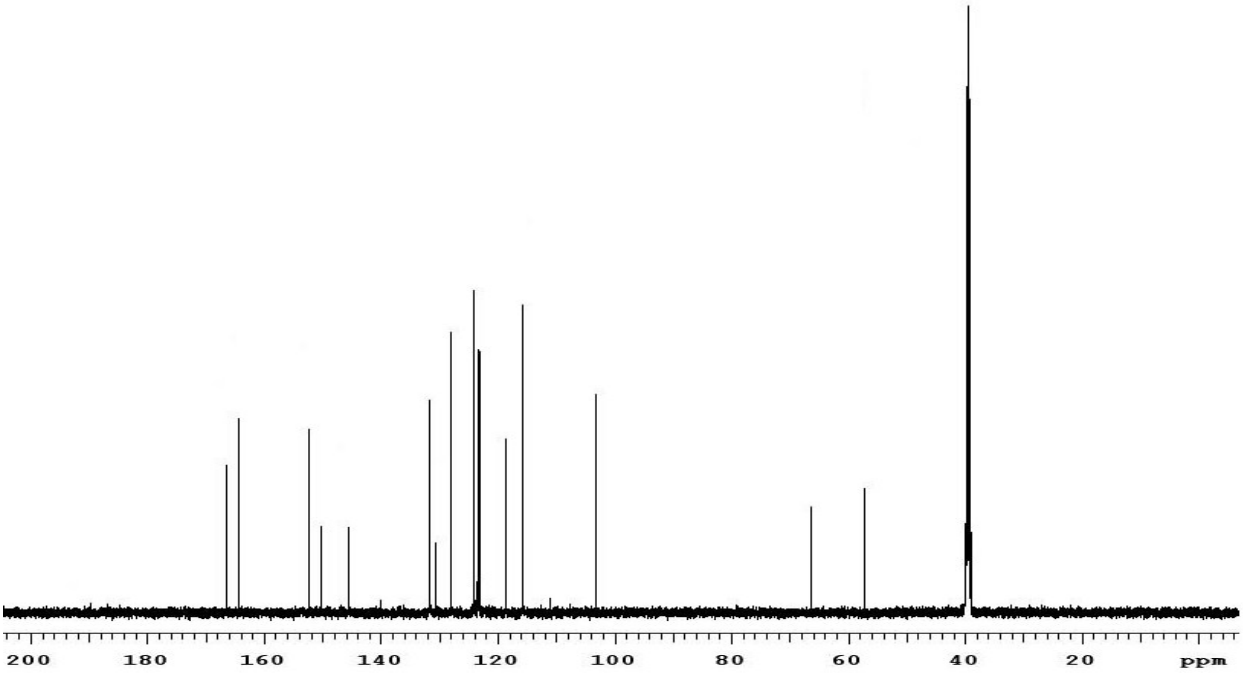
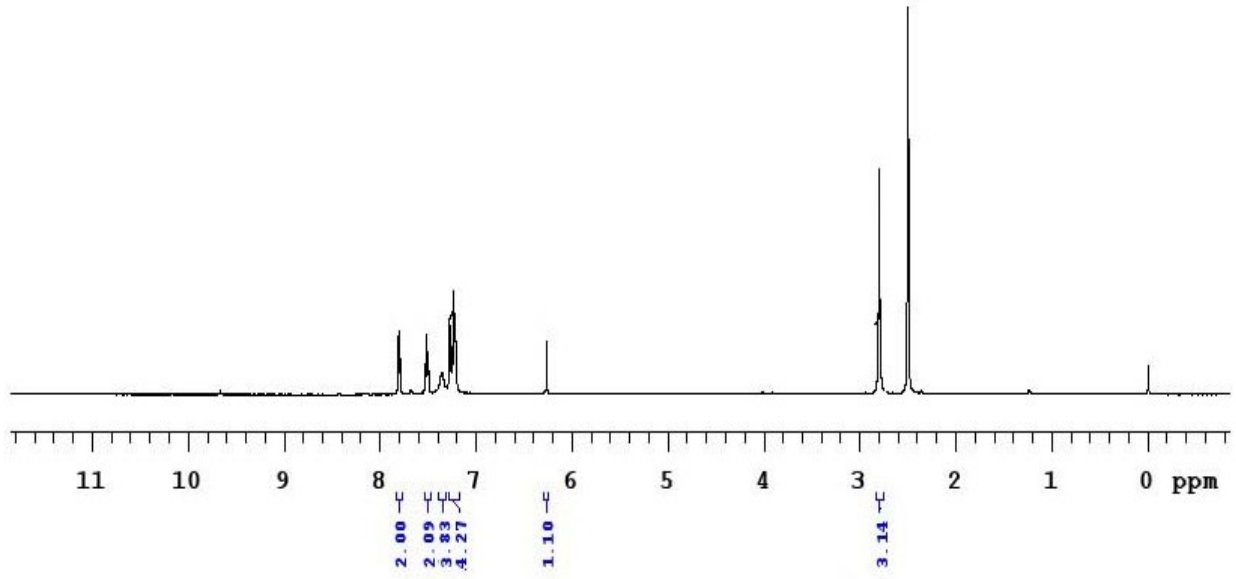


Compound 4g

H NMR



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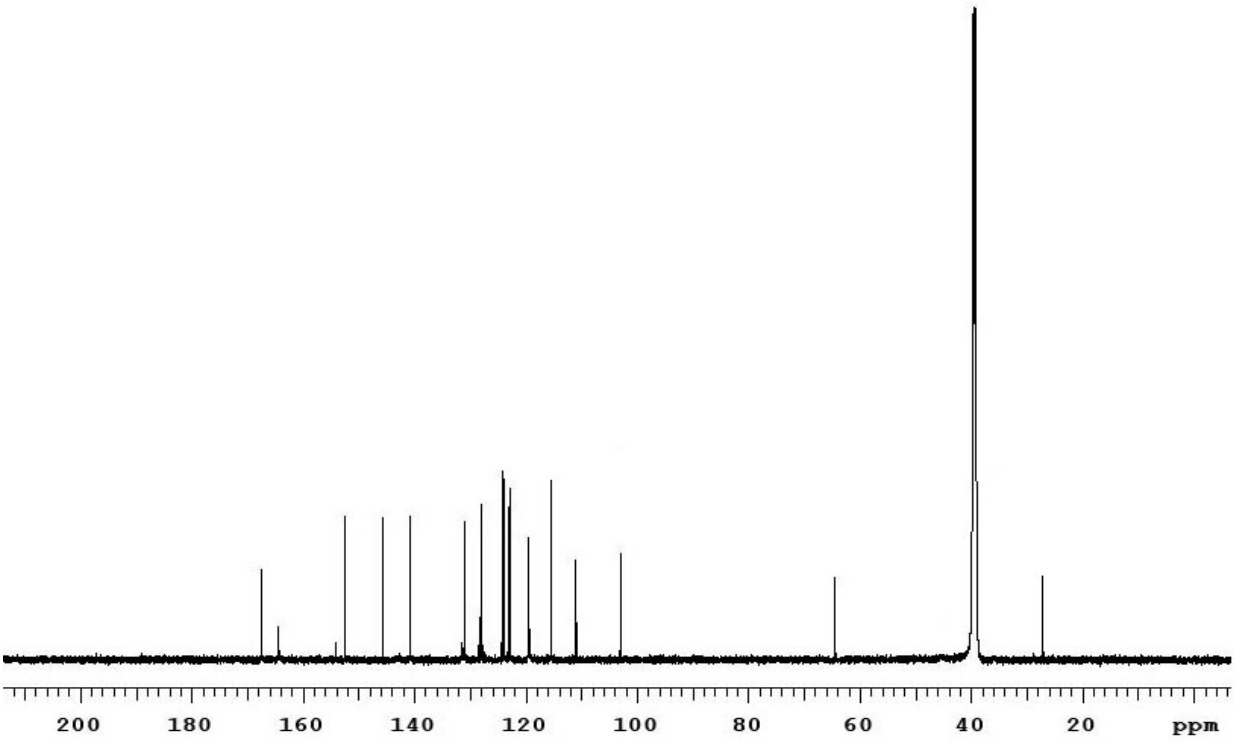

Compound 4h

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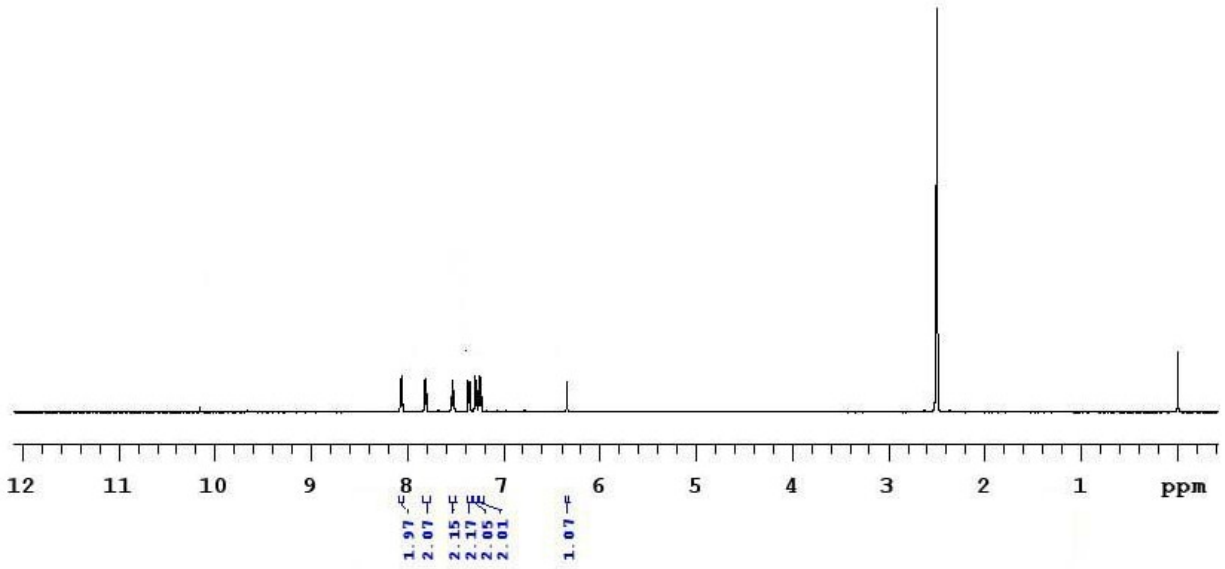

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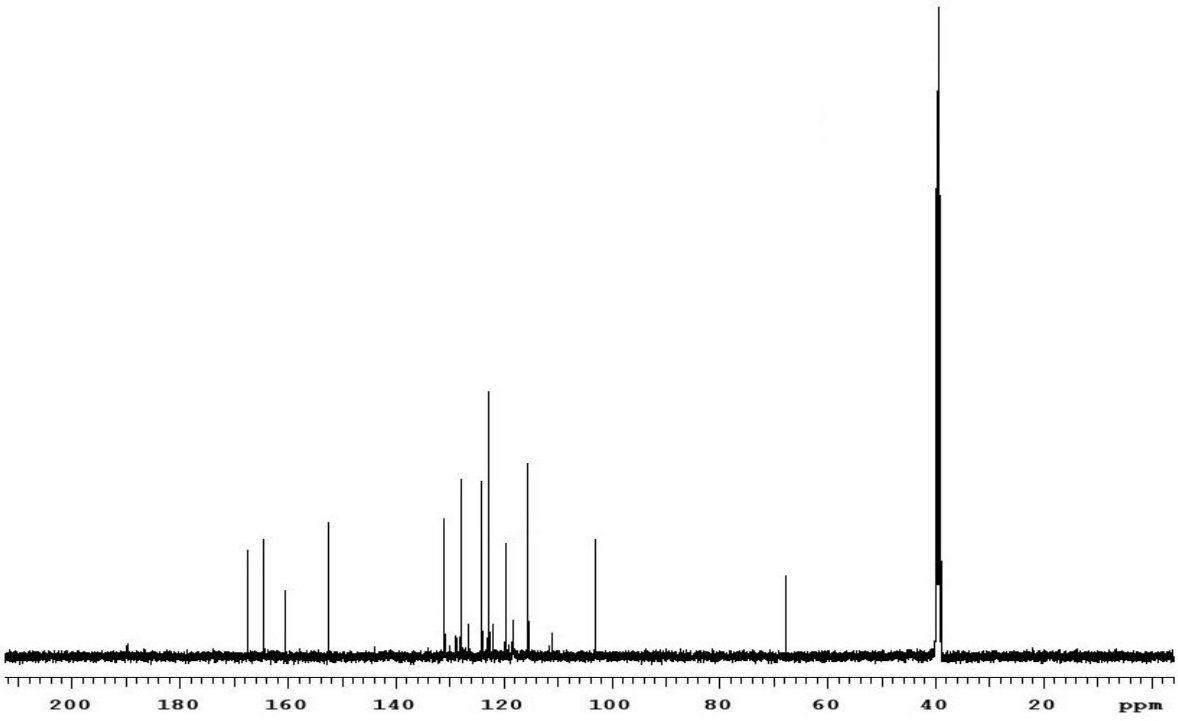
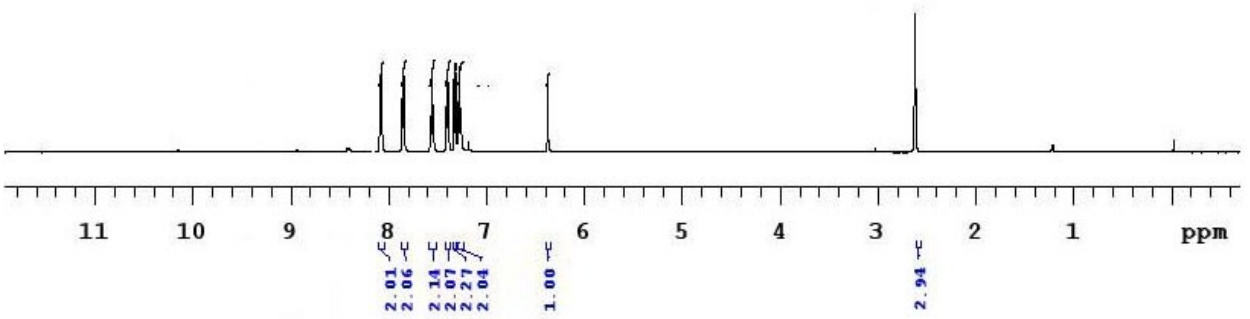
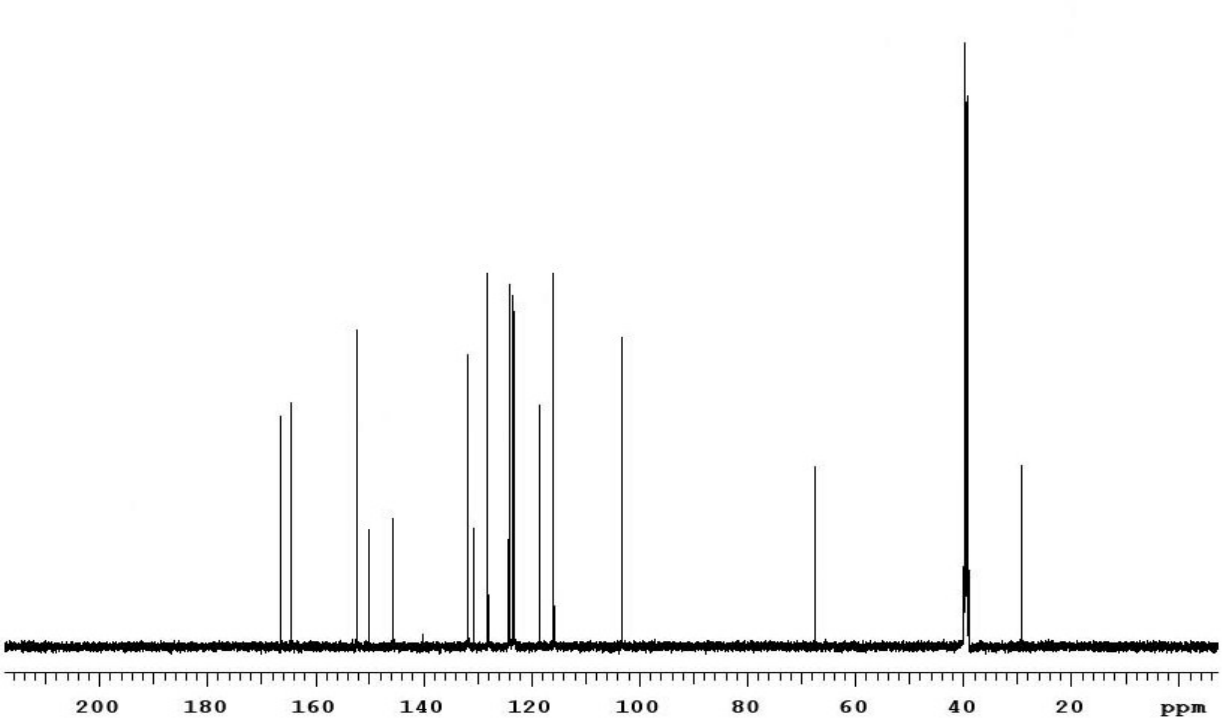


Compound 4i

H NMR

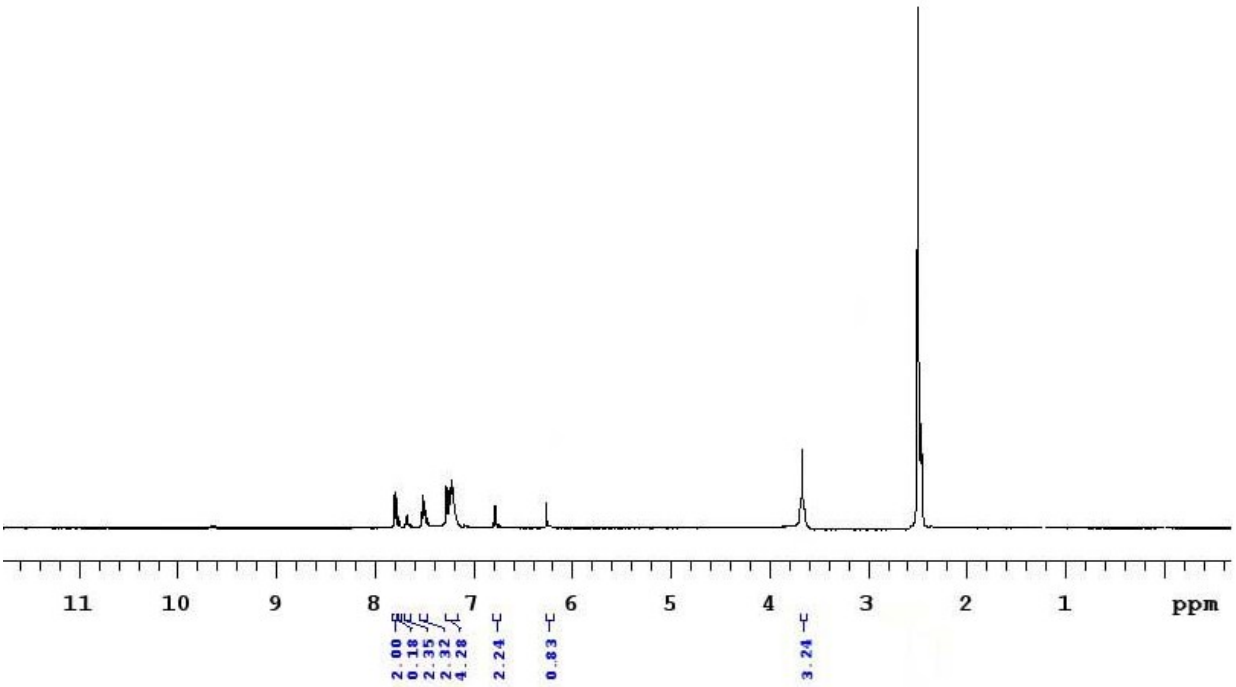


^{13}C NMR

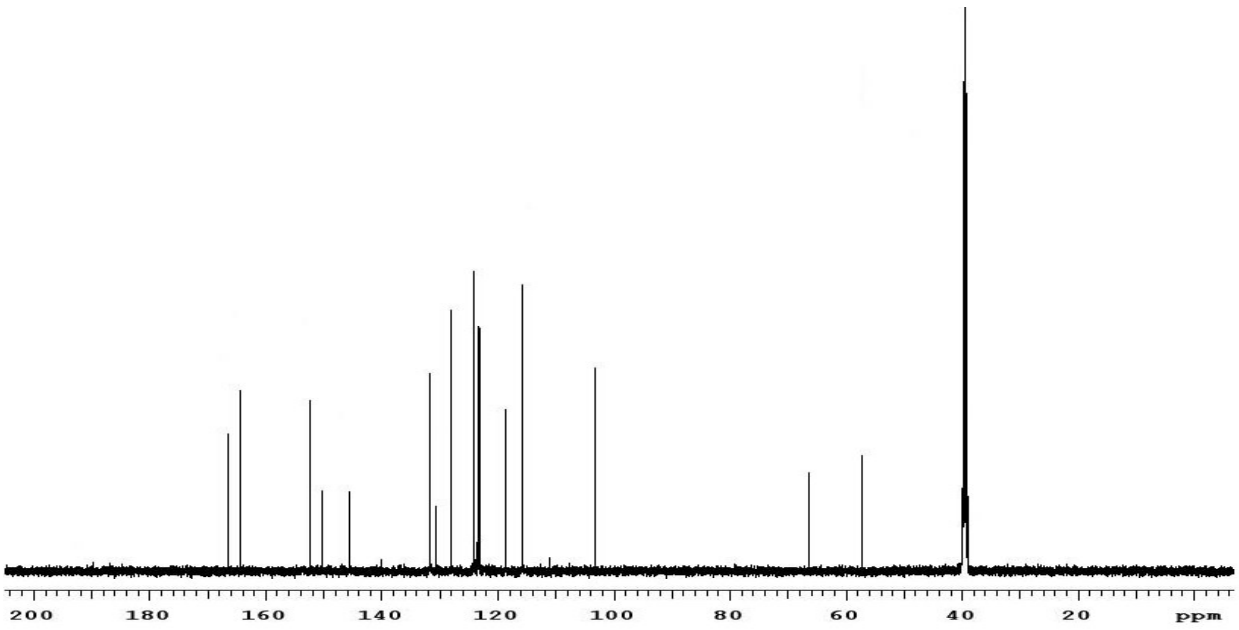


Compound 4j

H NMR

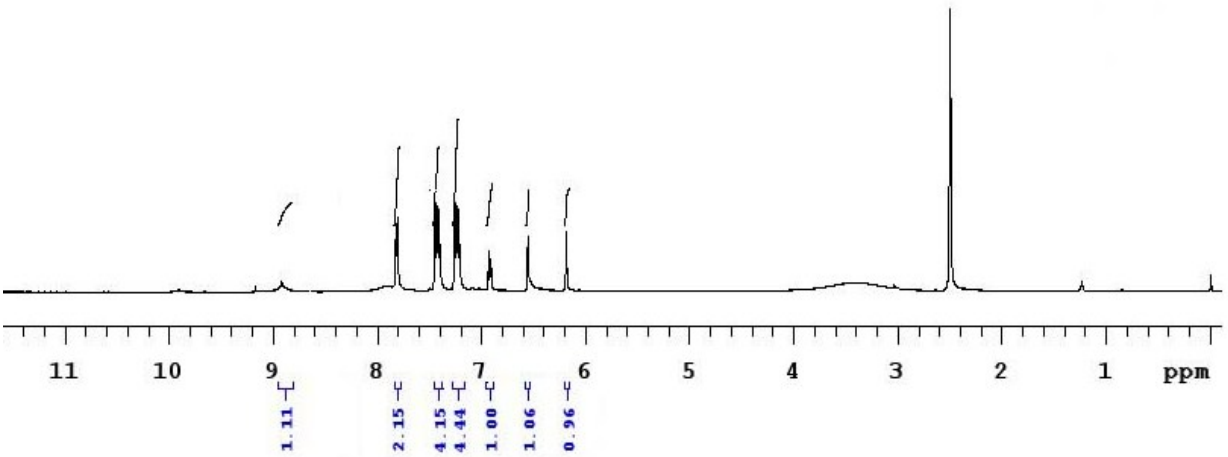


^{13}C NMR



Compound 4k

H NMR



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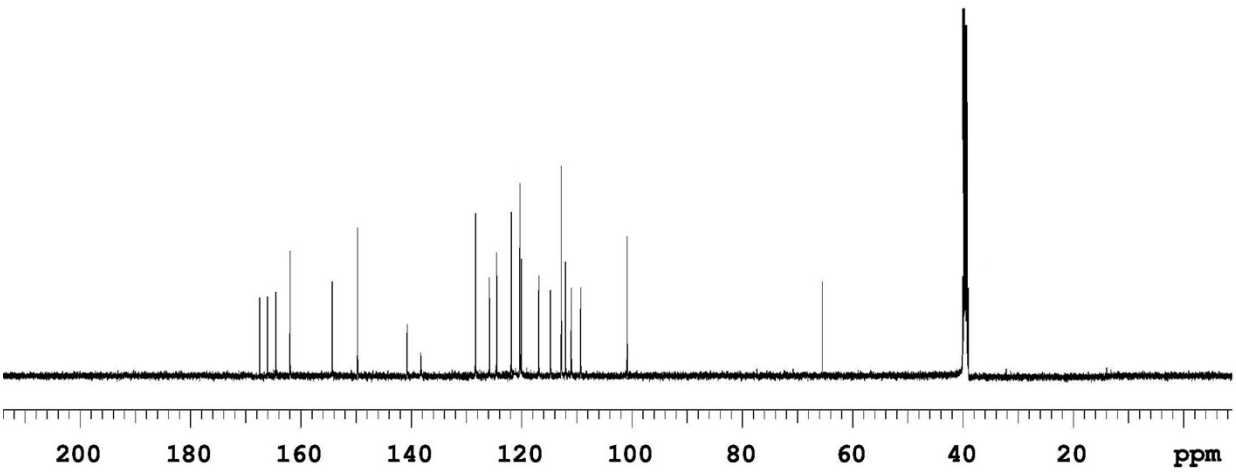

Compound 4l

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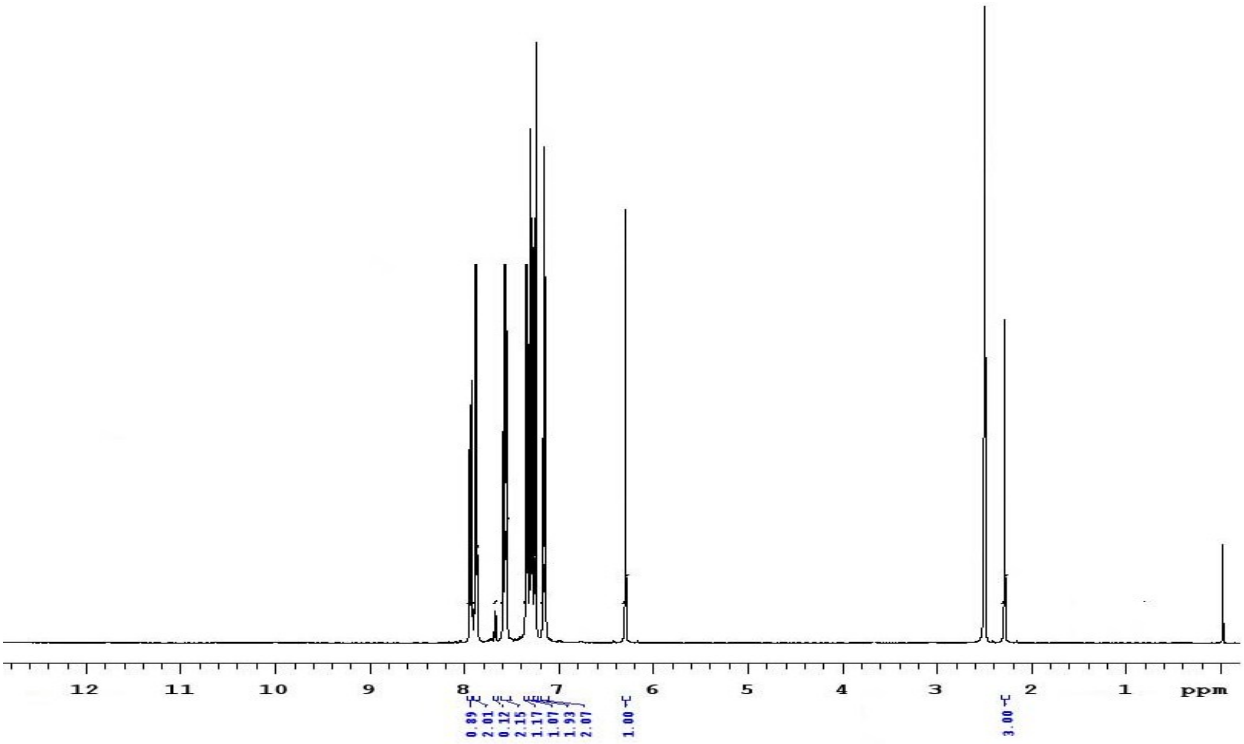

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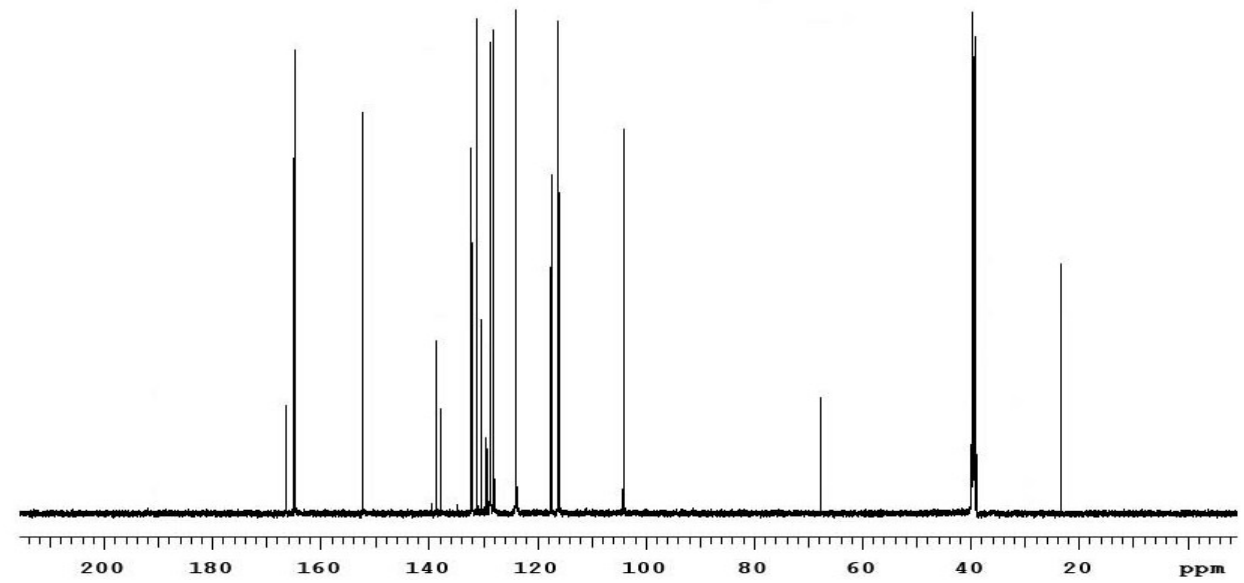
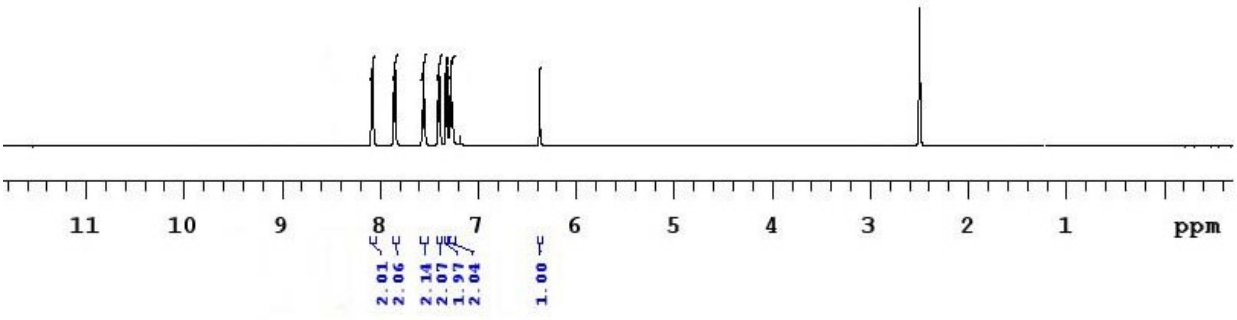
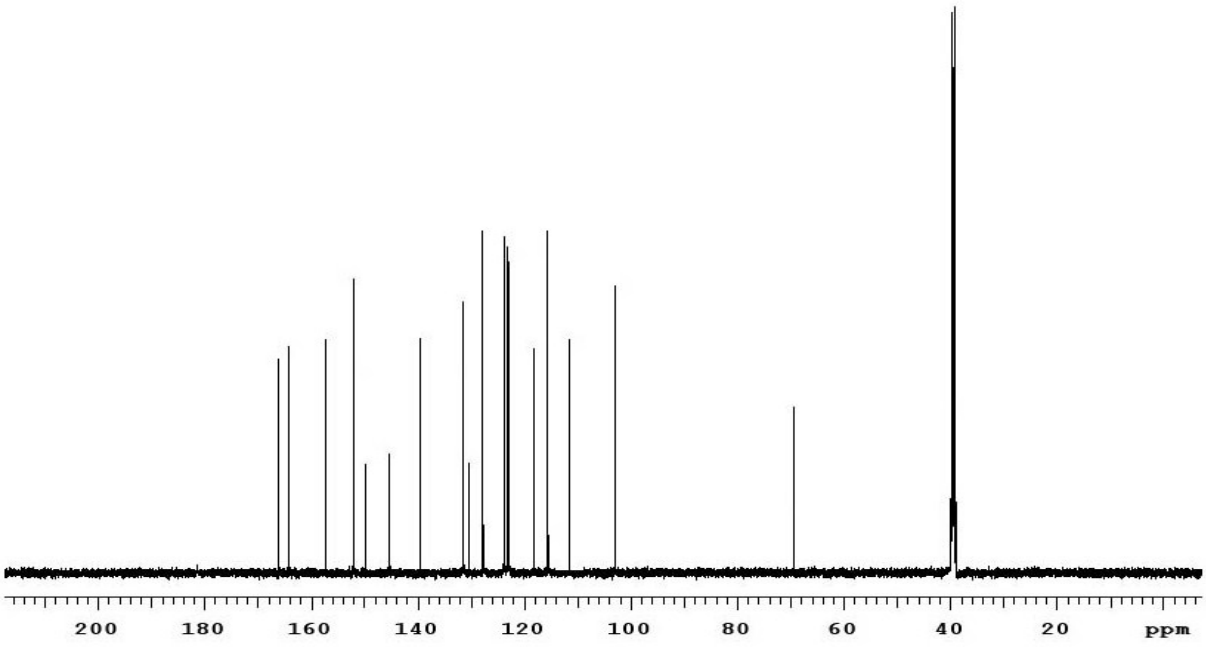


Compound 4m

H NMR

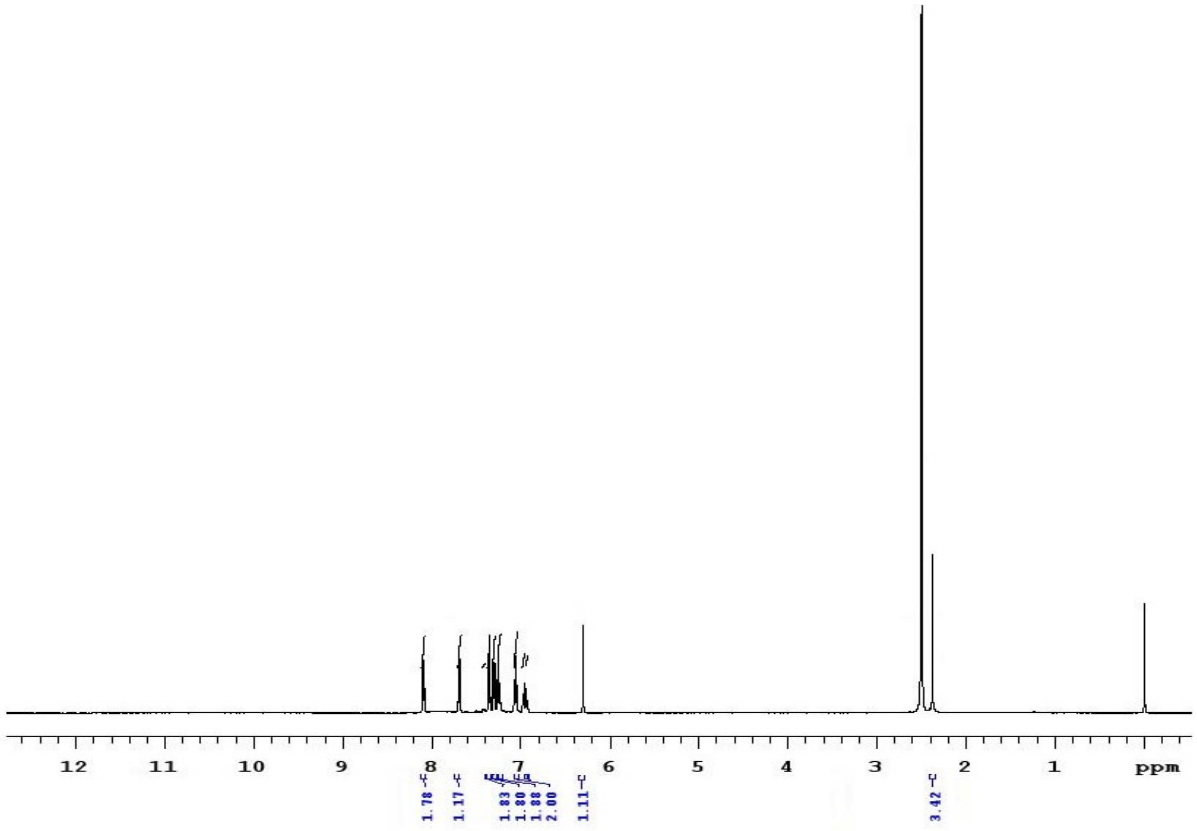


^{13}C NMR

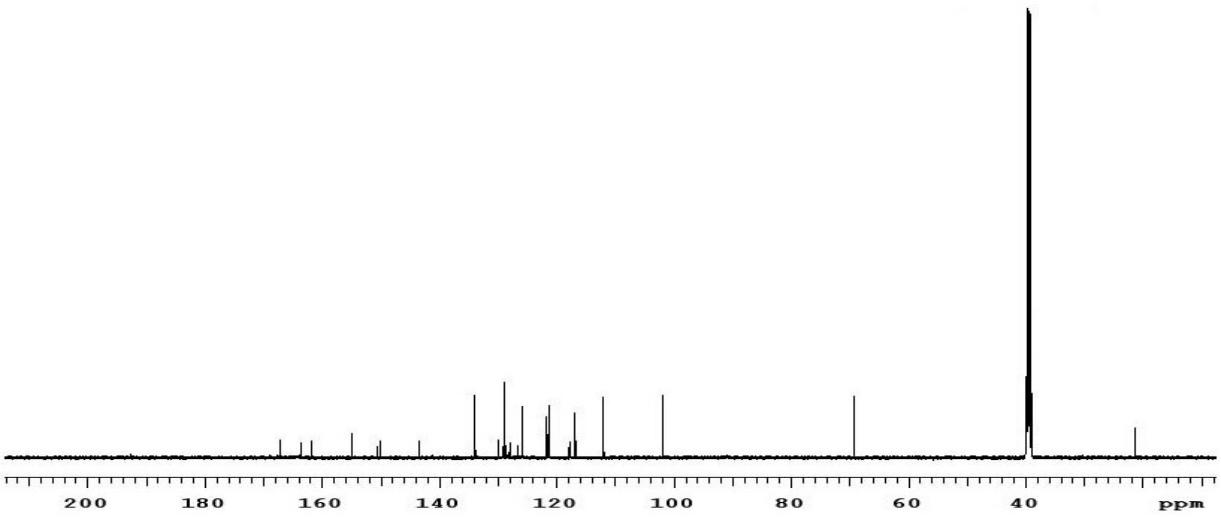


Compound 4n

H NMR

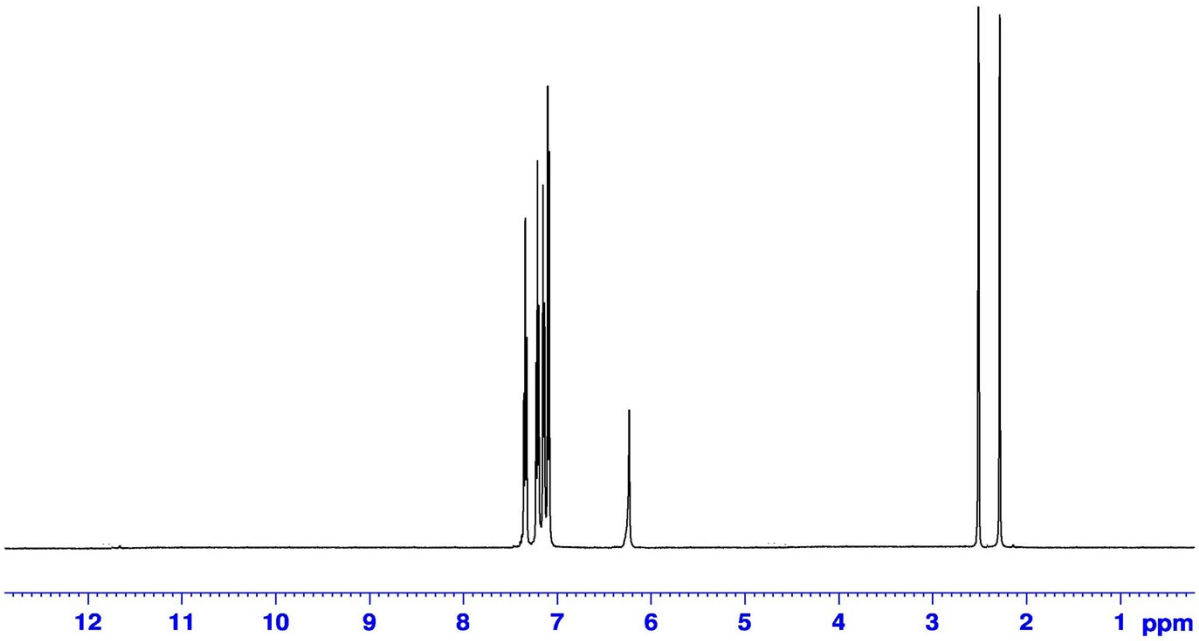


^{13}C NMR



Compound 4o

H NMR



^{13}C NMR