Supporting Information

For

One-Pot Magnetic Iron Oxide-Carbon Nanodots Composite Catalyzed Cyclooxidative Aqueous Tandem Synthesis of Quinazolinones in Presence of Tert-butyl Hydroperoxide

Biju Majumdar,[‡] Daisy Sarma,[‡] Siddarth Jain[‡] and Tridib K. Sarma[‡]*

[‡]Discipline of Chemistry, Indian Institute of Technology Indore, Simrol, Khandwa Road, Madhya Pradesh-453552

Corresponding Author E-mail: tridib@iiti.ac.in

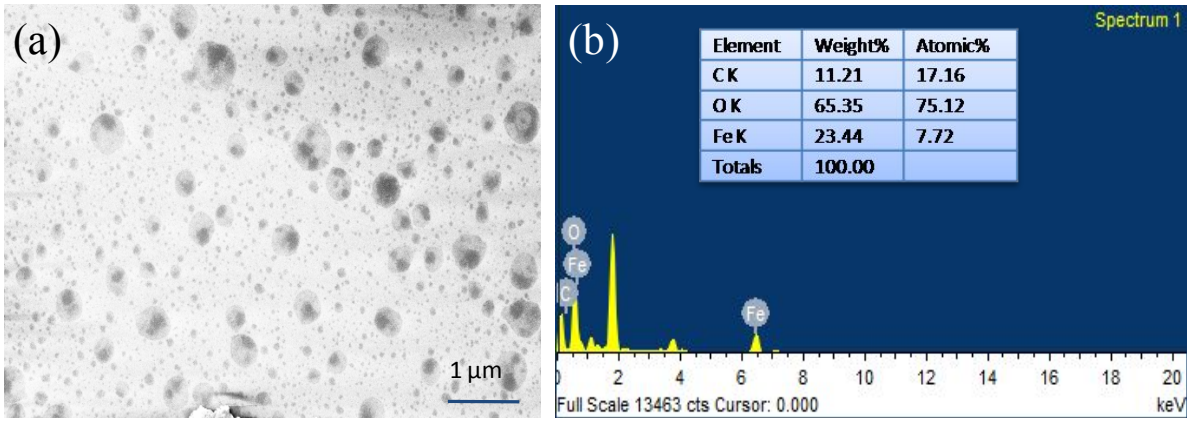
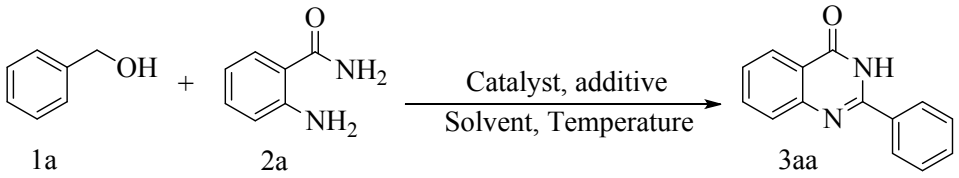


Figure S1: a) SEM image and b) EDS spectrum of Fe_3O_4-CND composite

Table S1. Oxidative coupling of (1a) and (2a) under various conditions[a]



1a 2a 3aa

entry	catalyst (wt%)	solvent	oxidant (eq.)	temp. (°C)	yield[b] (%)
1	Fe_3O_4-CND (10)	H_2O	-	50	trace
2	Fe_3O_4-CND (5)	H_2O	TBHP (2)	90	65
3	Fe_3O_4-CND (15)	H_2O	TBHP (2)	90	91
4	Fe_3O_4-CND (10)	CH_3CN	TBHP (2)	90	46
5	Fe_3O_4-CND (10)	H_2O	TBHP (4)	90	64
6	Fe_3O_4-CND (10)	H_2O	K_2CO_3 (2)	90	trace

[a]Reaction conditions: 1a (1.5 mmol), 2a (0.5 mmol), catalyst 5-15 wt%, oxidants1-4 equiv. and solvent 2mL, 16 hr, [b] yields of isolated product

Characterization data of quinazolinones

2-phenylquinazolin-4(3*H*)-one (3aa)[1]: Colourless solid, m.p. 231-233 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.28 (br, s, 1H), 8.33-8.31 (m, 1H), 8.23-8.20 (m, 2H), 7.84-7.78 (m, 2H), 7.60-7.56 (m, 3H), 7.52-7.48 (m, 1H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 163.6, 151.6, 149.4, 134.8, 132.8, 131.6, 129.0, 128.0, 127.2, 126.8, 126.3, 120.8; HRMS (ESI): calcd for [$C_{14}H_{10}N_2O^+$+ Na^+] 245.0685, found 245.0691.

2-(4-chlorophenyl)quinazolin-4(3*H*)-one (3ba)[2]: Colourless solid, m.p. >300 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.59 (br, s, 1H), 8.18 (d, *J*=8.56 Hz, 2H), 8.14 (d, *J*=7.56 Hz, 1H), 7.84 (t, *J*=7.0 Hz, 1H), 7.72 (d, *J*=8.0 Hz, 1H), 7.60 (d, *J*=8.8 Hz, 2H), 7.52 (t, *J*=7.0 Hz, 1H);^{13}C

NMR (DMSO-d^6, 100 MHz): δ = 163.5, 147.7, 140.6, 133.4, 128.8, 128.3, 127.4, 117.3, 114.9, 114.5; HRMS (ESI): calcd for [$C_{14}H_9ClN_2O^+$+ Na^+] 279.0296, found 279.0303.

2-(pyridin-2-yl)quinazolin-4(3*H*)-one (3ca)[3]: Yellow solid, m.p. 165-170 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 11.79 (br, s, 1H),8.75-8.72 (m, 1H), 8.46-8.41 (m, 1H), 8.19-8.15 (m, 1H), 8.09-8.02 (m, 1H), 7.89-7.76 (m, 2H), 7.66-7.61 (m, 1H), 7.58-7.52 (m, 1H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 160.3, 149.1, 148.8, 148.1, 147.6, 137.6, 135.0, 133.1, 129.6, 126.4, 126.2, 123.5, 122.0; HRMS (ESI): calcd for [$C_{13}H_9N_3O^+$+ Na^+] 223.0746, found 223.0752.

(*E*)-2-styrylquinazolin-4(3*H*)-one (3da)[2]: Colourless solid, m.p. 224-227 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.31 (br, s, 1H), 8.09 (d, *J*= 7.8 Hz, 1H), 7.92 (d, *J* = 16.3 Hz, 1H), 7.77 (t, *J* = 7.04 Hz, 1H), 7.70-7.64 (m, 3H), 7.51-7.36 (m, 4H), 6.98 (d, *J* = 15.2 Hz, 1H);^{13}C NMR (DMSO-d^6, 100 MHz): δ = 161.8, 159.9, 151.4, 149.0, 138.3, 135.0, 129.8, 129.1, 127.7, 126.3, 125.9, 121.1; HRMS (ESI): calcd for [$C_{16}H_{12}N_2O^+$+ Na^+] 271.0842, found 271.0853.

6-chloro-2-phenylquinazolin-4(3*H*)-one (3ab)[4]: m.p. 294-296 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.69 (br, s, 1H), 8.17-8.15 (m, 2H), 8.08 (s, 1H), 7.87-7.85 (m, 1H), 7.78-7.76 (m, 1H), 7.64-7.51 (m, 3H); HRMS (ESI): calcd for [$C_{14}H_9ClN_2O^+$+ Na^+] 279.0296, found 279.0302.

2-(2-nitrophenyl)quinazolin-4(3*H*)-one (3ea): Pale Yellow Solid, 230-240 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.81 (br, s, 1H), 8.19 (d, *J* = 8.28 Hz, 1H), 8.16 (d, *J* = 7.28 Hz, 1H), 7.92-7.80 (m, 4H), 7.63 (d, *J* = 8.04 Hz, 1H), 7.55 (t, *J* = 7.2 Hz, 1H);^{13}C NMR (DMSO-d^6, 100 MHz): δ = 161.5, 151.6, 148.5, 147.4, 134.7, 133.9, 131.5, 129.1, 127.4, 127.1, 125.9, 124.5, 121.2; HRMS (ESI): calcd for [$C_{14}H_9N_3O_3^+$+ Na^+] 290.0536, found 290.0540.

2-pentylquinazolin-4(3*H*)-one (3fa)[2]: Colourless solid, m.p. 152-154 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 12.28 (br, s, 1H), 8.27 (d, *J* = 6.86 Hz, 1H), 7.77-7.75 (m, 1H), 7.68 (d, *J* = 7.45 Hz, 1H), 7.45 (t,*J* = 7.62 Hz, 1H), 2.80 (t, *J* = 7.76 Hz, 2H), 1.92-1.87 (m, 2H), 1.46-1.39 (m, 4H), 0.92 (t, *J* = 7.76 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 165.5, 158.2, 150.6, 135.2, 128.3, 127.8, 127.2, 121.1, 36.6, 31.9, 28.1, 22.8, 14.2; HRMS (ESI): calcd for [$C_{13}H_{16}N_2O^+$+ Na^+] 239.1155, found 239.1167.

2-hexylquinazolin-4(3*H*)-one (3ga)[5]: Colourless solid, m.p. 140-145 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ 12.47 (br, s, 1H), 8.32 (d, *J* = 8.27 Hz, 1H), 7.78-7.75 (m, 1H), 7.75 (d, *J* = 8.32 Hz,

1H), 7.46-7.47 (m, 1H), 2.80 (t, *J* = 7.62 Hz, 2H), 1.90-1.87 (m, 2H), 1.47-1.45 (m, 2H), 1.37-1.30 (m, 4H), 0.88 (t, *J* = 6.90 Hz, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ165.3, 156.1, 149.3, 134.5, 127.4, 126.2, 126.7, 120.1, 35.8, 31.4, 28.6, 27.4, 22.4,14.0; HRMS (ESI): calcd for [$C_{14}H_{18}N_2O^+$+ Na^+] 253.1311, found 253.1319.

2-(4-bromophenyl)quinazolin-4(3*H*)-one (3ha)[2]: Colourless solid, m.p. 290-292 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.59 (br, s, 1H), 8.15-8.10 (m, 3H), 7.82 (t, *J* = 7.04 Hz, 1H), 7.76-7.72 (m, 3H), 7.51 (t, *J* = 7.04 Hz, 1H); ^{13}C NMR (DMSO-d^6, 100 MHz): δ = 159.7, 151.4, 148.2, 134.6, 131.6, 129.7, 127.5, 126.7, 125.8, 125.2, 121.0; HRMS (ESI): calcd for [$C_{14}H_9BrN_2O^+$+ Na^+] 322.9790, found 322.9798.

2-(*p*-tolyl)quinazolin-4(3*H*)-one (3ia)[1]: Colourless solid, m.p. 230-232 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 11.40 (br, s, 1H), 8.30 (d, *J* = 7.52 Hz, 1H), 8.10 (d, *J* = 8.28 Hz, 2H), 7.82-7.76 (m, 2H), 7.46 (t, *J* = 7.76 Hz, 1H), 7.35 (d, *J* = 8.0 Hz, 2H);^{13}C NMR ($CDCl_3$, 100 MHz): δ = 164.1, 151.7, 149.6, 142.1, 134.8, 130.1, 129.9, 129.7, 129.0, 127.8, 127.3, 126.5, 126.3, 120.6, 21.5; HRMS (ESI): calcd for [$C_{15}H_{12}N_2O^+$+ Na^+] 259.0842, found 259.0848.

2-(4-methoxyphenyl)quinazolin-4(3*H*)-one (3ja)[1]: Colourless solid, m.p. 230-233 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 10.80 (br, s, 1H), 8.28 (d, *J* = 7.52 Hz, 1H), 8.12 (d, *J* = 8.0 Hz, 2H), 7.78(m, 2H), 7.46 (m, 1H), 7.05 (d, *J* = 8.04 Hz, 2H), 3.90 (s, 3H); HRMS (ESI): calcd for [$C_{15}H_{12}N_2O_2^+$+ Na^+] 275.0791, found 275.0798.

2-(furan-2-yl)quinazolin-4(3*H*)-one (3la)[2]: Colourless solid, m.p. 272-275 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.48 (br, s, 1H), 8.10 (d, *J* = 8.04 Hz, 1H), 7.99 (m, 1H), 7.78 (t, *J* = 8.52 Hz, 1H), 7.66 (d, *J* = 8.0 Hz, 1H), 7.61 (d, *J* = 3.52 Hz, 1H), 7.46 (t, *J* = 7.04 Hz, 1H), 6.74-6.73 (m, 1H); ^{13}C NMR (DMSO-d^6, 100 MHz): δ = 161.5, 148.6, 146.5, 146.0, 144.0, 134.6, 127.2, 126.4, 125.9, 121.1, 114.5, 112.5; HRMS (ESI): calcd for [$C_{12}H_8N_2O_2^+$+ Na^+] 235.0478, found 235.0479.

2-(thiophen-2-yl)quinazolin-4(3*H*)-one (3ma)[2]: Colourless solid, m.p. 220-222 °C; ^{1}H NMR (DMSO-d^6, 400 MHz): δ = 12.63 (br, s, 1H), 8.21 (d, *J* = 4.76 Hz, 1H), 8.10 (d, *J* = 7.8 Hz, 1H), 7.85 (d, *J* = 5.76 Hz, 1H), 7.77 (t, *J* = 8.52 Hz, 1H), 7.63 (d, *J* = 8.0 Hz, 1H), 7.45 (t, *J* = 8.04 Hz, 1H), 7.23-7.21 (m, 1H); ^{13}C NMR (DMSO-d^6, 100 MHz): δ = 161.8, 148.6, 147.8, 137.3, 134.7,

132.1, 129.4, 128.5, 126.9, 126.3, 125.9, 120.8; HRMS (ESI): calcd for [$C_{12}H_8N_2OS^+$+ Na^+] 251.0250, found 251.0252.

2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one (A)[6]: Colourless crystal, m.p. 215-220 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.94 (d, *J* = 7.76 Hz, 1H), 7.60 (m, 2H), 7.44 (m, 3H), 7.33 (t, *J* = 7.52 Hz, 1H), 6.90 (t, *J* = 7.76 Hz, 1H), 6.67 (d, *J* = 8.04 Hz, 1H), 5.90 (s, 1H), 5.88 (br, 1H, NH), 4.35 (br, 1H, NH), ^{13}C NMR (100 MHz, DMSO-d_6): δ 164.0, 148.3, 142.1, 133.8, 128.9, 128.8, 127.8, 127.3, 117.6, 115.4, 114.8, 67.0; HRMS (ESI): calcd for [$C_{14}H_{12}N_2O$ + Na^+] 247.0842, found 247.0864.

References:

1. Hikawa, H.; Ino, Y.; Suzuki, H.; Yokoyama, Y. *J. Org. Chem.* **2012**, *77*, 7046.
2. Upadhyaya, K.; Thakur, R. K.; Shukla, S. K.; Tripathi, R. P. *J. Org. Chem*. **2016**, *81*, 5046.
3. Siddiki, S. M. A. H.; Kon, K. A.; Touchy, S.; Shimizu, K. *Catal. Sci. Technol.* **2014**, *4*, 1716.
4. Zhou, J.; Fang, J. *J. Org. Chem.* **2011**, *76*, 7730.
5. Sharif, M.; Opalach, J.; Langer, P.; Beller, M.; Wu, X.-F. *RSC Adv.* **2014**, *4*, 8.
6. Majumdar, B.; Mandani, S.; Bhattacharya, T.; Sarma, D.; Sarma, T. K. *J. Org. Chem*. **2017**, *82*, 2097.

^{1}H and ^{13}C NMR Spectra of Quinazolinones

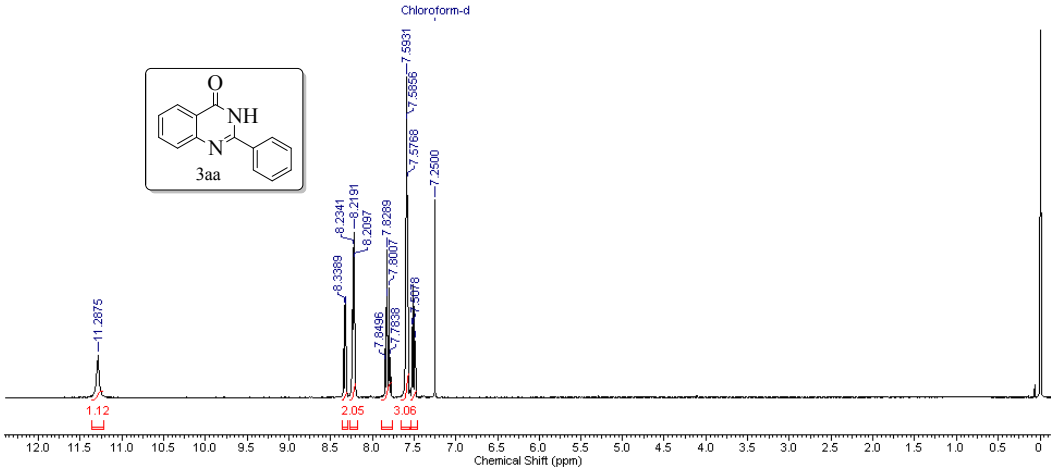


Figure S2: ^{1}H NMR of 3aa

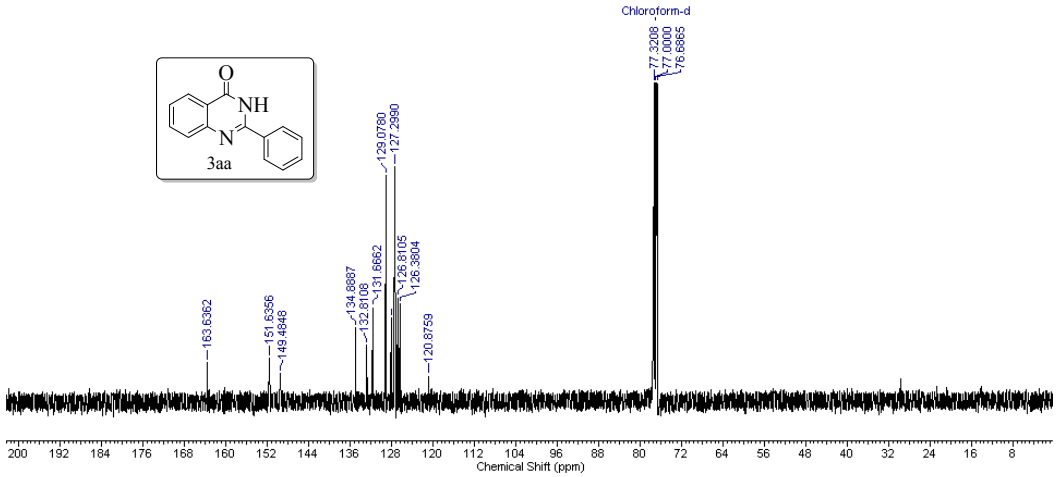


Figure S3: ^{13}C NMR of 3aa

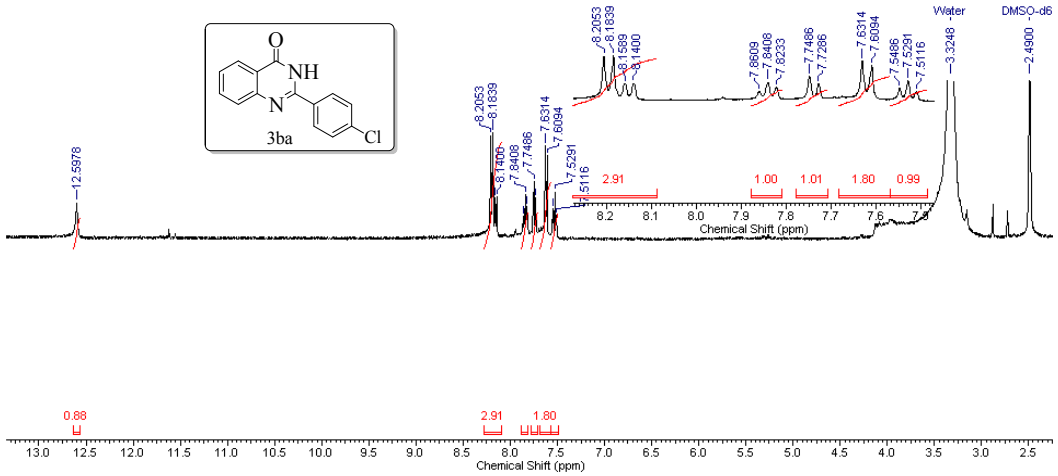


Figure S4: ^{1}H NMR of 3ba

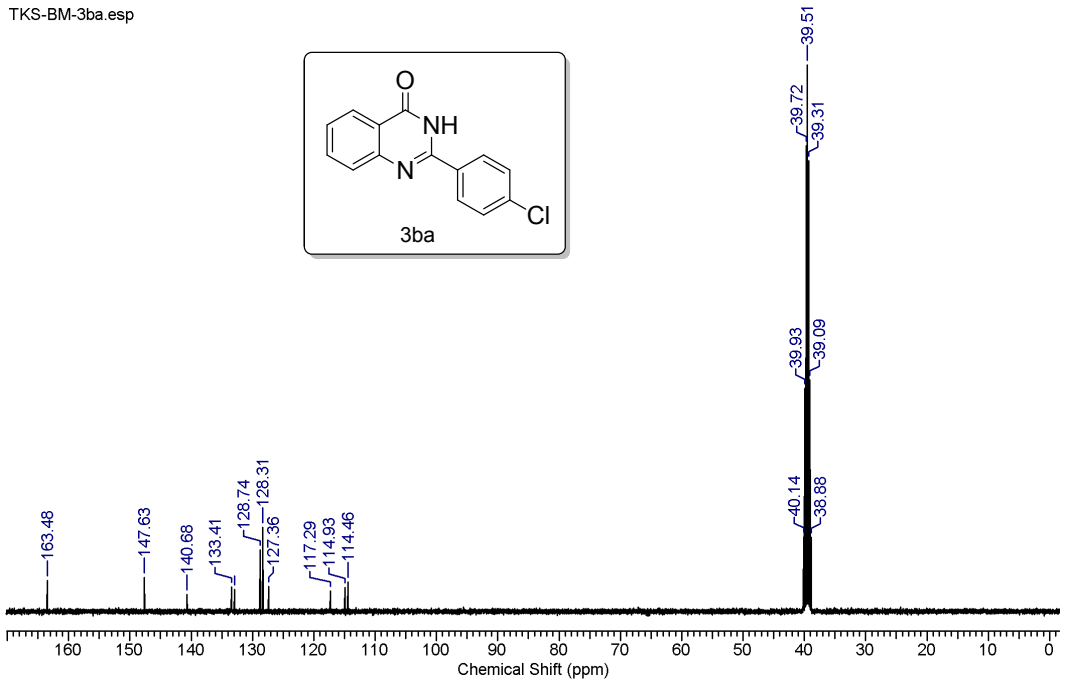


Figure S5: ^{13}C NMR of 3ba

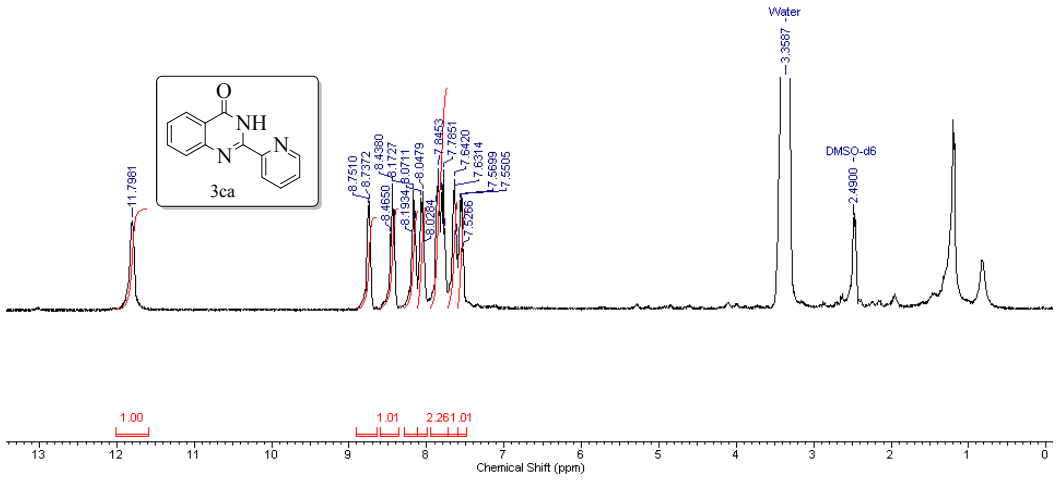


Figure S6: ^{1}H NMR of 3ca

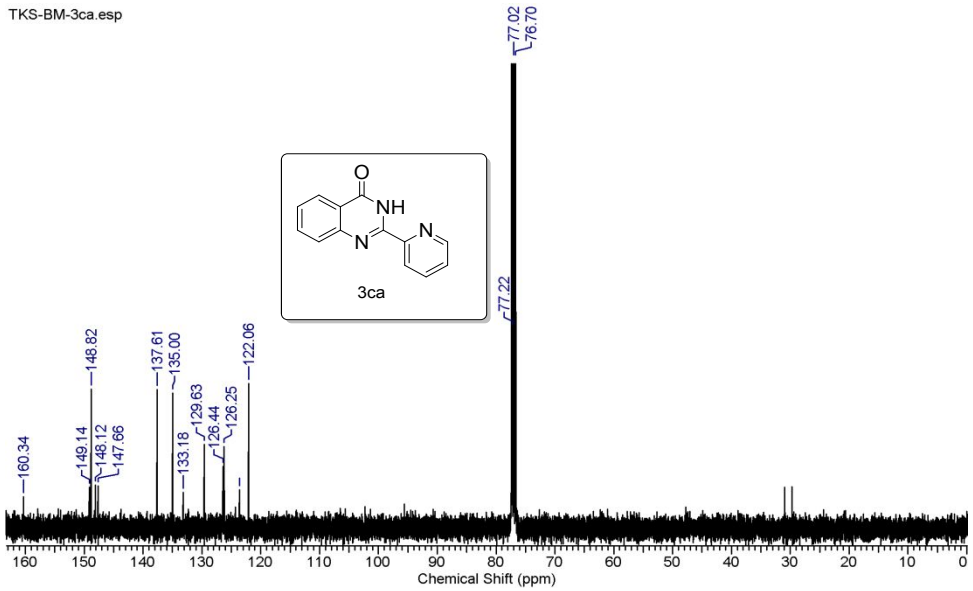


Figure S7: ^{13}C NMR of 3ca

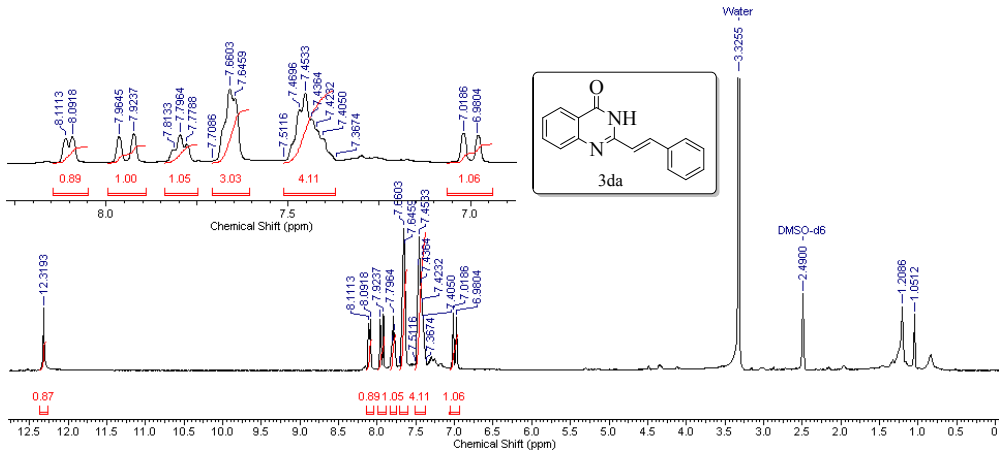


Figure S8: ^{1}H NMR of 3da

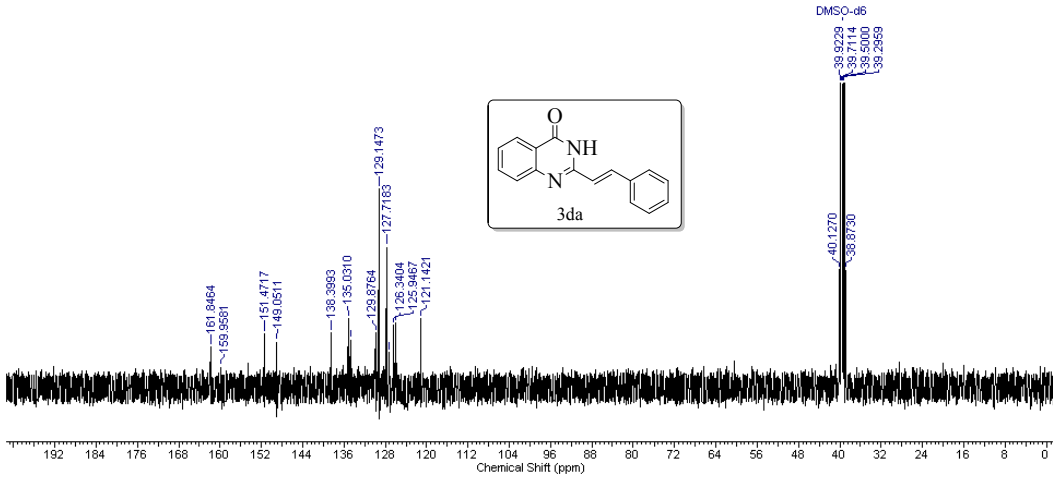


Figure S9: ^{13}C NMR of 3da

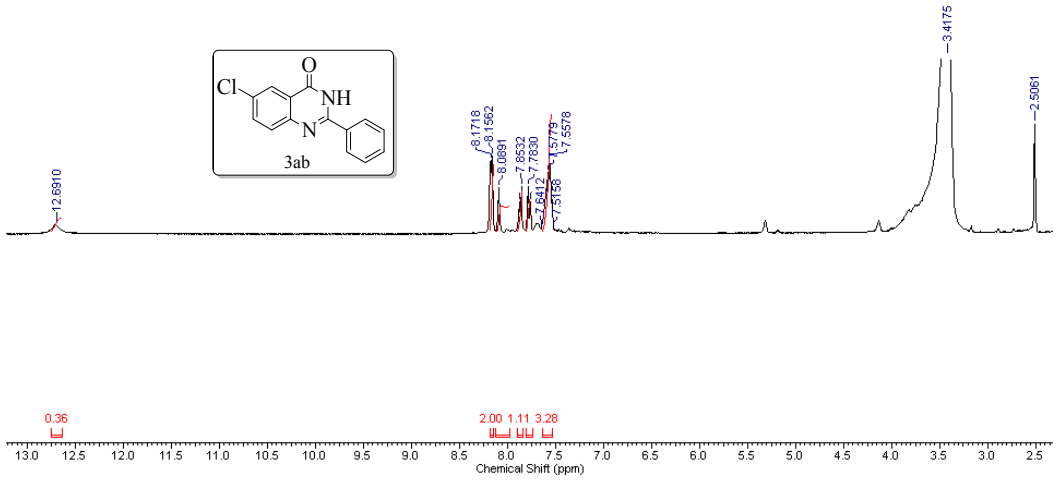


Figure S10: ^{1}H NMR of 3ab

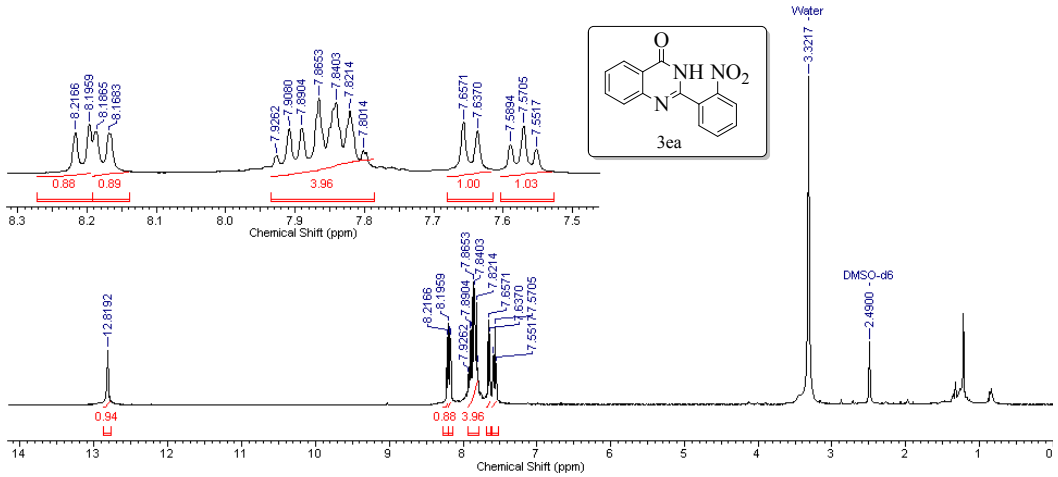


Figure S11: ^{1}H NMR of 3ea

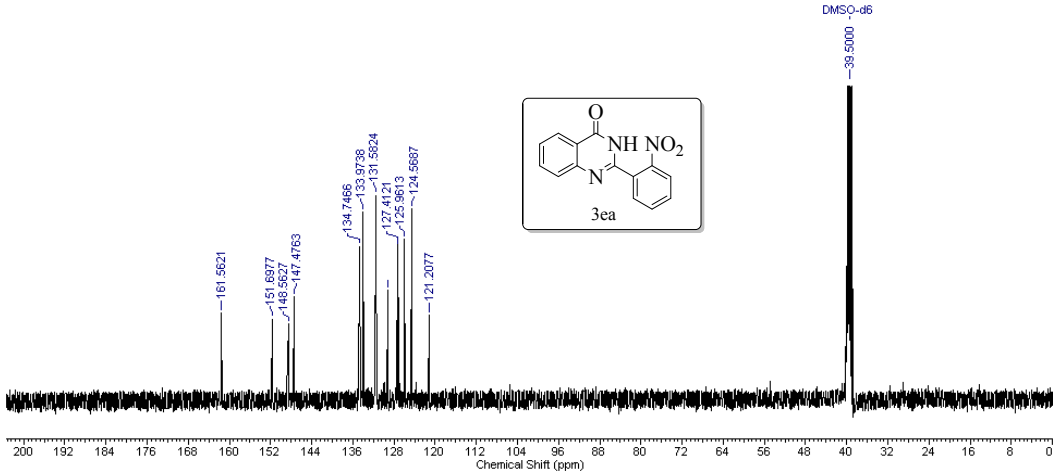


Figure S12: ^{13}C NMR of 3ea

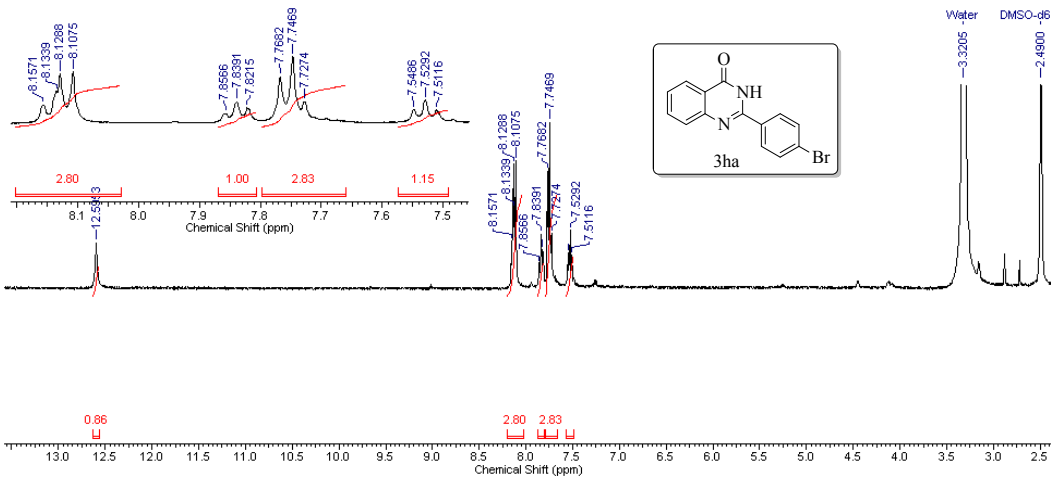


Figure S13: ^{1}H NMR of 3ha

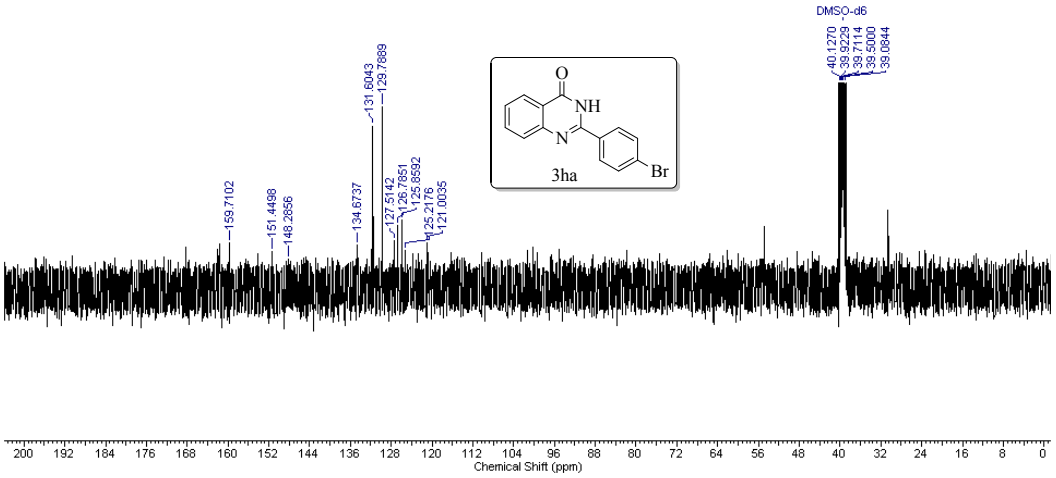


Figure S14: ^{13}C NMR of 3ha

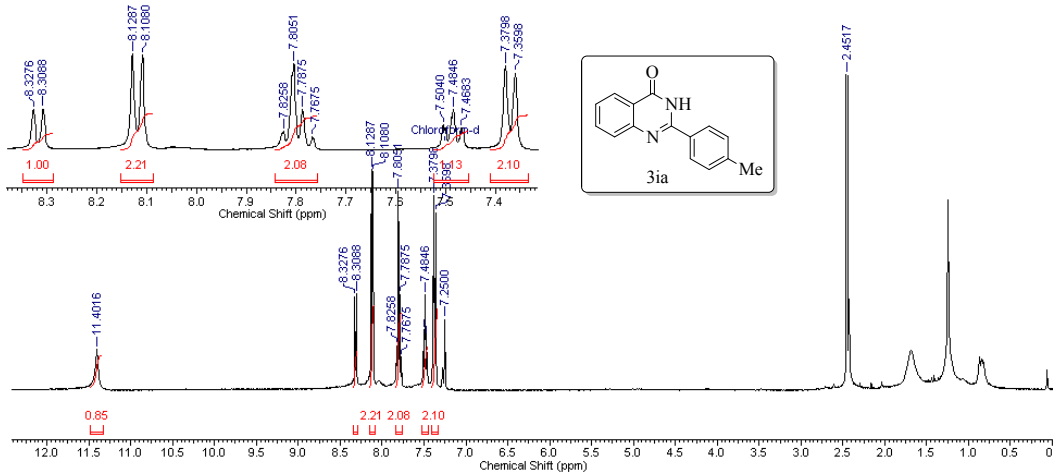


Figure S15: ^{1}H NMR of 3ia

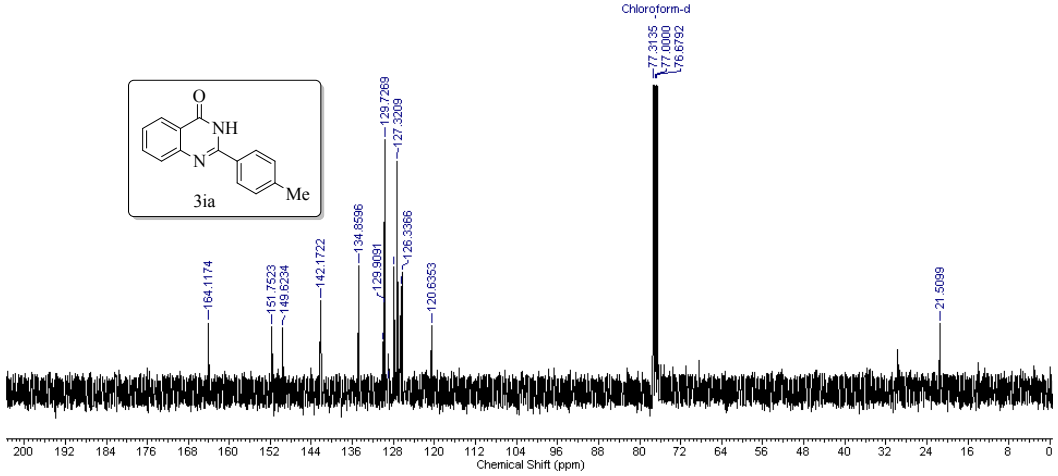


Figure S16: ^{13}C NMR of 3ia

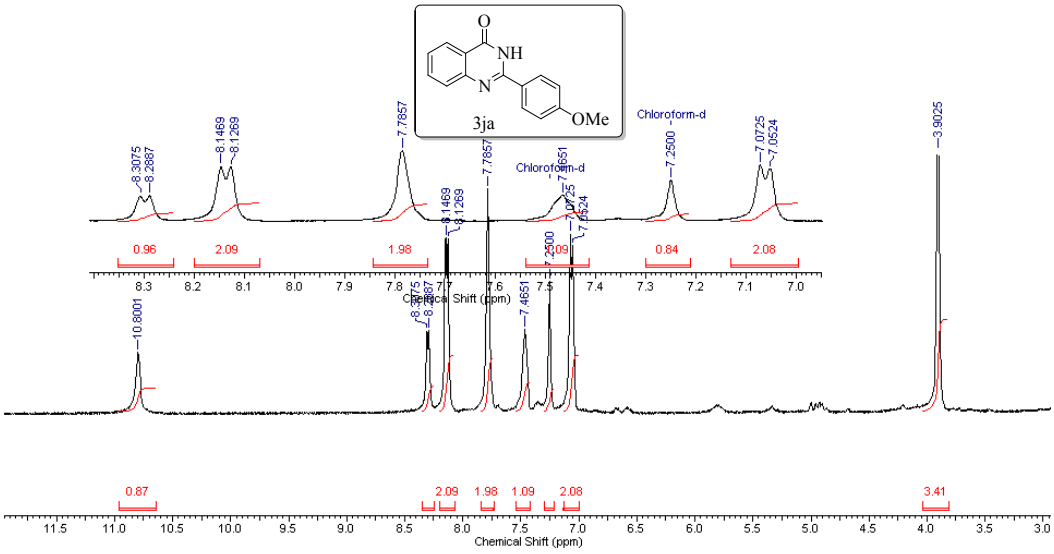


Figure S17: ^{1}H NMR of 3ja

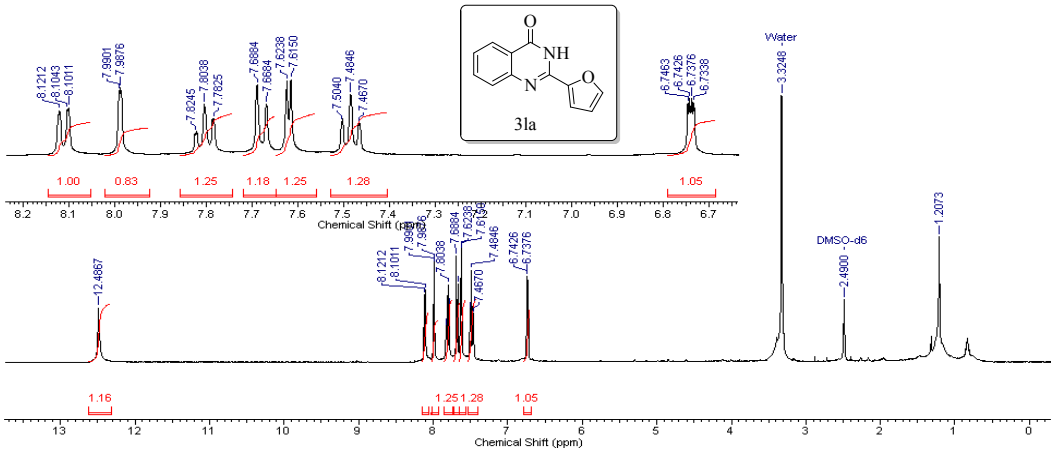


Figure S18: ^{1}H NMR of 3la

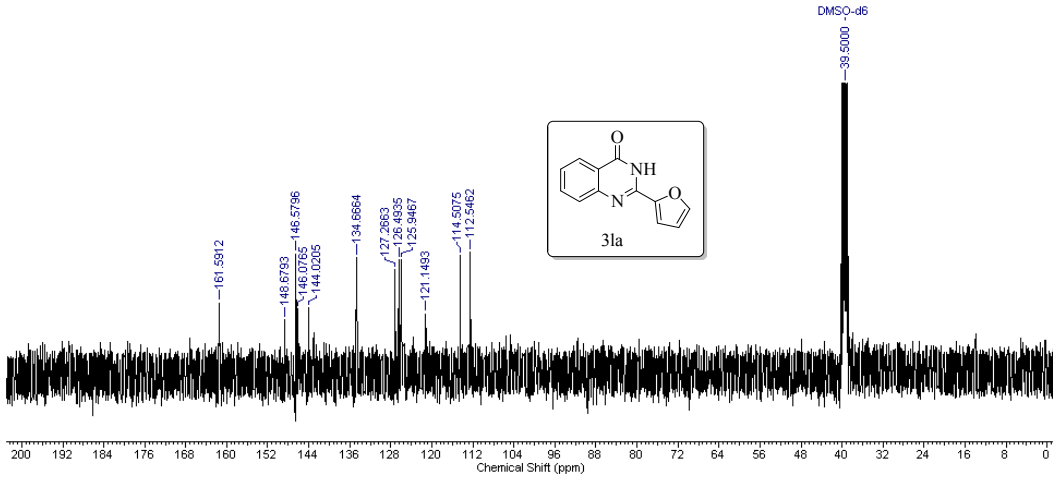


Figure S19: ^{13}C NMR of 3la

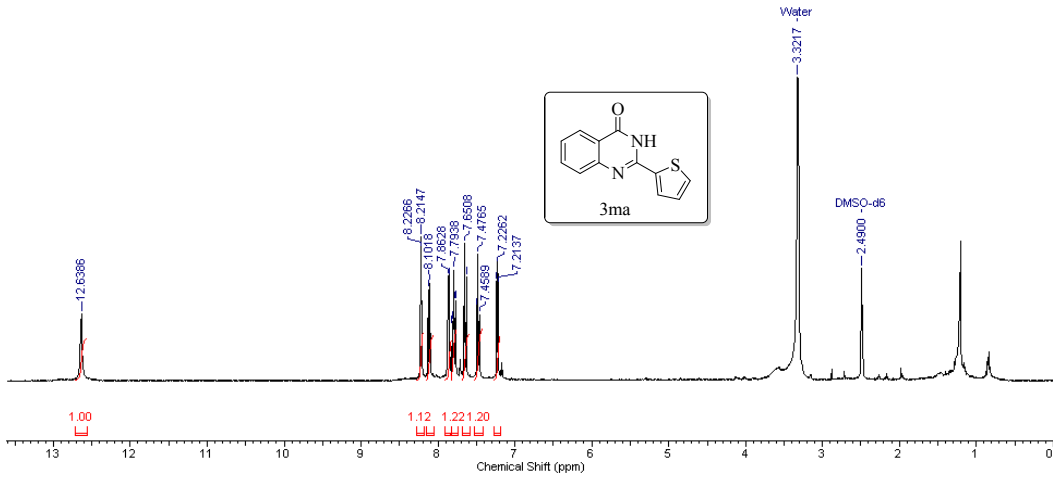


Figure S20: ^{1}H NMR of 3ma

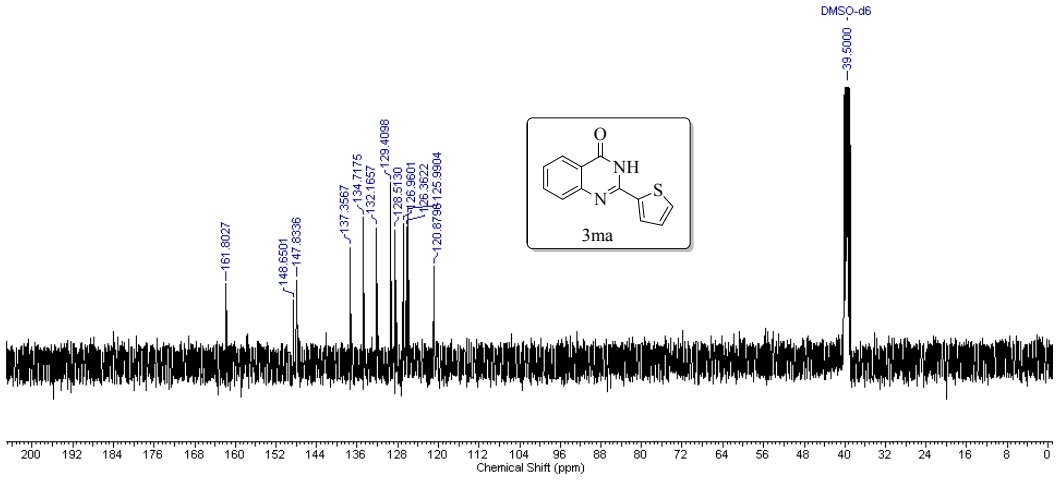


Figure S21: ^{13}C NMR of 3ma

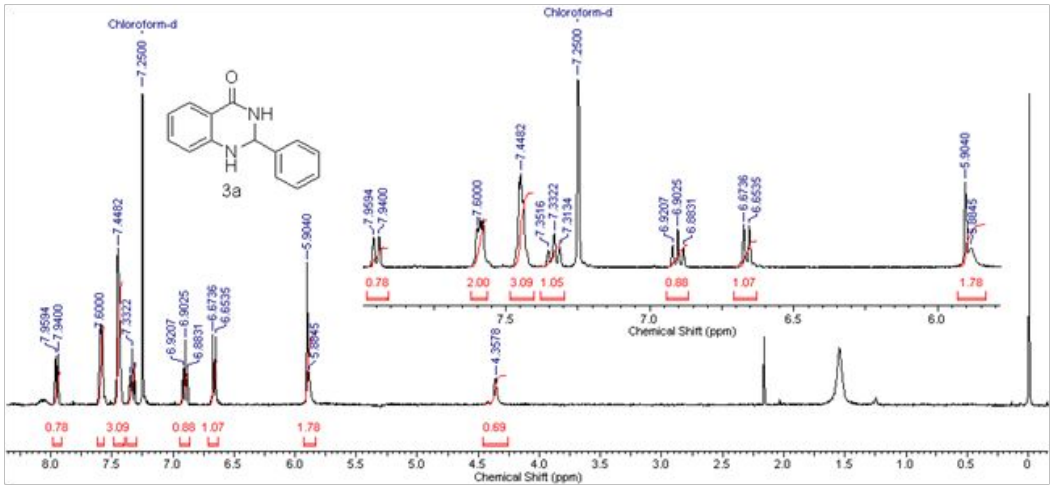


Figure S22: ^{1}H NMR of A

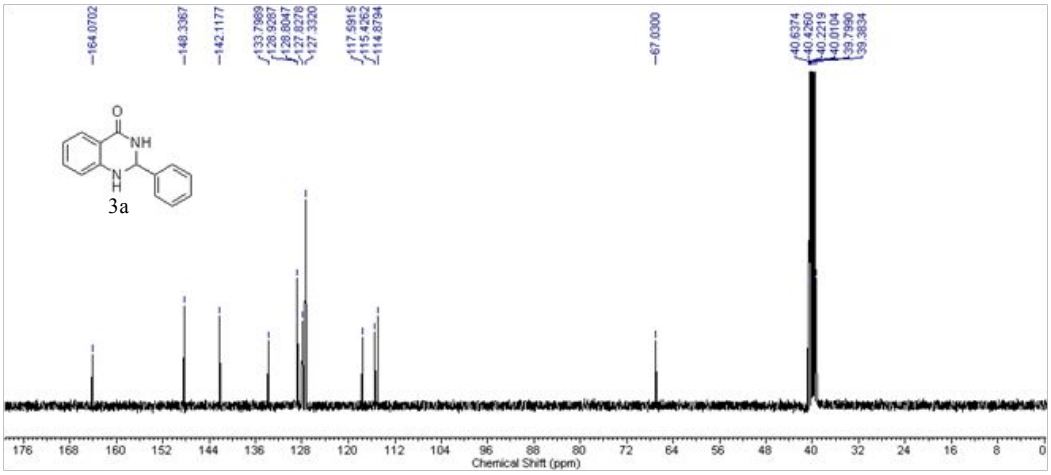


Figure S23: ^{13}C NMR of A