

## New Journal of Chemistry

### Supplementary Information

# **A novel organic-inorganic hybrid material: production, characterization and catalytic performance for the reaction of arylaldehydes, dimedone and 6-amino-1,3-dimethyluracil**

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This supplementary information includes:

- (i) Selected spectral data of the synthesized pyrimido[4,5-*b*]quinolines (pages S2-S4)
- (ii) The FT-IR, XRD, TGA and adsorption/desorption porosimetry diagrams of the catalyst (pages S5-S12)
- (iii) The original spectrums of the synthesized pyrimido[4,5-*b*]quinolines (pages S13-S30)

### Selected spectral data of the products

**Product 1a.**  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.89 (s, 3H, C-CH<sub>3</sub>), 1.05 (s, 3H, C-CH<sub>3</sub>), 2.05 (d,  $J = 16.0$  Hz, 1H, C=C-CH<sub>2</sub>), 2.23 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.52-2.65 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.10 (s, 3H, NCH<sub>3</sub>), 3.47 (s, 3H, NCH<sub>3</sub>), 4.89 (s, 1H, methine CH), 7.09 (t,  $J = 7.1$  Hz, 1H, H<sub>Ar</sub>), 7.17-7.25 (m, 4Hm H<sub>Ar</sub>), 9.02 (s, 1H, NH).

**Product 1b.**  $^1\text{H}$  NMR (500 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.90 (s, 3H, C-CH<sub>3</sub>), 1.03 (s, 3H, C-CH<sub>3</sub>), 2.03-2.10 (distorted AB system, 2H, C=C-CH<sub>2</sub>), 2.20-2.28 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.09 (s, 3H, NCH<sub>3</sub>), 3.45 (s, 3H, NCH<sub>3</sub>), 4.85 (s, 1H, methine CH), 7.18 (d,  $J = 8.4$  Hz, 2H, H<sub>Ar</sub>), 7.37 (d,  $J = 8.4$  Hz, 2H, H<sub>Ar</sub>), 9.04 (s, 1H, NH).

**Product 1c.**  $^1\text{H}$  NMR (250 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.85 (s, 3H, C-CH<sub>3</sub>), 1.00 (s, 3H, C-CH<sub>3</sub>), 2.01 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.19 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.47-2.55 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.06 (s, 3H, NCH<sub>3</sub>), 3.42 (s, 3H, NCH<sub>3</sub>), 4.83 (s, 1H, methine CH), 7.14-7.28 (m, 5H, H<sub>Ar</sub> and NH).

**Product 1d.**  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.90 (s, 3H, C-CH<sub>3</sub>), 1.05 (s, 3H, C-CH<sub>3</sub>), 1.98 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.21 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.49-2.62 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.06 (s, 3H, NCH<sub>3</sub>), 3.46 (s, 3H, NCH<sub>3</sub>), 5.14 (s, 1H, methine CH), 7.26 (dd,  $J = 8.4, 2.1$  Hz, 1H, H<sub>Ar</sub>), 7.33-7.36 (m, 2H, H<sub>Ar</sub>), 9.05 (s, 1H, NH).

**Product 1e.**  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.90 (s, 3H, C-CH<sub>3</sub>), 1.05 (s, 3H, C-CH<sub>3</sub>), 1.98 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.20 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.50-2.62 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.06 (s, 3H, NCH<sub>3</sub>), 3.47 (s, 3H, NCH<sub>3</sub>), 5.18 (s, 1H, methine CH), 7.09 (t,  $J = 7.7, 2.1$  Hz, 1H, H<sub>Ar</sub>), 7.16-7.24 (m, 2H, H<sub>Ar</sub>), 7.34 (d,  $J = 7.6$  Hz, 1H, H<sub>Ar</sub>), 9.03 (s, 1H, NH).

**Product 1f.**  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.91 (s, 3H, C-CH<sub>3</sub>), 1.06 (s, 3H, C-CH<sub>3</sub>), 2.06 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.26 (d,  $J = 16.4$  Hz, 1H, C=C-CH<sub>2</sub>), 2.57-2.69 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.09 (s, 3H, NCH<sub>3</sub>), 3.47 (s, 3H, NCH<sub>3</sub>), 4.98 (s, 1H, methine CH), 7.52 (m, 1H, H<sub>Ar</sub>), 7.72 (d,  $J = 7.7$  Hz, 1H, H<sub>Ar</sub>), 7.98 (dd,  $J = 8.2, 1.4$  Hz, 1H, H<sub>Ar</sub>), 8.05 (s, 1H, H<sub>Ar</sub>), 9.18 (s, 1H, NH);  $^{13}\text{C}$  NMR (100 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 26.8, 28.1, 29.5, 30.8, 31.2, 32.6, 35.0, 50.3, 89.7, 111.1, 121.6, 122.8, 129.8, 135.1, 144.6, 147.9, 148.9, 150.8, 150.9, 161.2, 195.1.

**Product 1g.**  $^1\text{H}$  NMR (500 MHz, DMSO- $d_6$ ):  $\delta$  (ppm) 0.84 (s, 3H, C-CH<sub>3</sub>), 1.02 (s, 3H, C-CH<sub>3</sub>), 1.95 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.18 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.57 (d,  $J$

= 5.4 Hz, 2H, O=C-CH<sub>2</sub>), 3.02 (s, 3H, NCH<sub>3</sub>), 3.44 (s, 3H, NCH<sub>3</sub>), 5.74 (s, 1H, methine CH), 7.29 (t,  $J = 7.3$  Hz, 1H, H<sub>Ar</sub>), 7.43 (d,  $J = 7.5$  Hz, 1H, H<sub>Ar</sub>), 7.52 (t,  $J = 7.5$  Hz, 1H, H<sub>Ar</sub>), 7.74 (d,  $J = 8.0$  Hz, 1H), 8.98 (s, 1H, NH); <sup>13</sup>C NMR (125 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm): 26.9, 28.0, 29.3, 30.5, 30.8, 32.2, 32.5, 50.4, 90.2, 111.6, 124.1, 127.3, 131.1, 133.2, 141.5, 144.7, 148.8, 150.5, 150.9, 160.9, 194.9. Mass:  $m/z$  410 (M<sup>+</sup>), 411 (M<sup>+</sup> + 1).

**Product 1h.** <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm) 0.90 (s, 3H, C-CH<sub>3</sub>), 1.05 (s, 3H, C-CH<sub>3</sub>), 2.04 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.20-2.24 (m, 4H, one hydrogen of C=C-CH<sub>2</sub> and CH<sub>3</sub>-Ar), 2.52-2.63 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.09 (s, 3H, NCH<sub>3</sub>), 3.46 (s, 3H, NCH<sub>3</sub>), 4.84 (s, 1H, methine CH), 6.98 (d,  $J = 7.9$  Hz, 2H, H<sub>Ar</sub>), 7.11 (d,  $J = 8.0$  Hz, 2H, H<sub>Ar</sub>), 8.99 (s, 1H, NH); <sup>13</sup>C NMR (100 MHz, DMSO- d<sub>6</sub>):  $\delta$  (ppm): 21.1, 26.9, 28.1, 29.6, 30.6, 31.2, 32.6, 33.8, 50.5, 90.8, 112.3, 128.0, 128.7, 135.2, 43.9, 144.2, 149.8, 151.0, 161.1, 195.0.

**Product 1i.** FT-IR (KBr):  $\nu_{max}$  (cm<sup>-1</sup>) 3289, 3231, 3095, 2953, 1699, 1665, 1614, 1506, 1381, 1239; <sup>1</sup>H NMR (500 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm) 0.92 (s, 3H, C-CH<sub>3</sub>), 1.04 (s, 3H, C-CH<sub>3</sub>), 2.04 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.21 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.52 (d,  $J = 18.0$  Hz, 1H, O=C-CH<sub>2</sub>), 2.60 (d,  $J = 17.5$  Hz, 1H, O=C-CH<sub>2</sub>), 3.09 (s, 3H, NCH<sub>3</sub>), 3.44 (s, 3H, NCH<sub>3</sub>), 3.65 (s, 3H, OCH<sub>3</sub>), 3.67 (s, 3H, OCH<sub>3</sub>), 4.85 (s, 1H, methine CH), 6.69 (dd,  $J = 1.9, 8.3$  Hz, 1H, H<sub>Ar</sub>), 6.74, d,  $J = 8.3$  Hz, 1H, H<sub>Ar</sub>), 6.83 (s, 1H, H<sub>Ar</sub>), 8.99 (s, 1H, NH); <sup>13</sup>C NMR (125 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm): 26.8, 28.1, 29.7, 30.6, 32.2, 32.5, 33.6, 50.5, 55.8, 55.9, 90.9, 111.8, 112.2, 112.5, 119.9, 139.6, 144, 147.6, 148.5, 149.9, 150.9, 161.2, 195.1. Mass:  $m/z$  425 (M<sup>+</sup>), 426 (M<sup>+</sup> + 1).

**Product 1j.** FT-IR (KBr):  $\nu_{max}$  (cm<sup>-1</sup>) 3476, 3208, 3083, 2956, 1697, 1666, 1636, 1503, 1377, 1217; <sup>1</sup>H-NMR (400 MHz, DMSO- d<sub>6</sub>):  $\delta$  (ppm): 0.85 (s, 3H, C-CH<sub>3</sub>), 1.04 (s, 3H, C-CH<sub>3</sub>), 1.96 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.18 (d,  $J = 16.2$  Hz, 1H, C=C-CH<sub>2</sub>), 2.47-2.57 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.06 (s, 3H, NCH<sub>3</sub>), 3.46 (s, 3H, NCH<sub>3</sub>), 3.63 (s, 3H, OCH<sub>3</sub>), 3.67 (s, 3H, OCH<sub>3</sub>), 4.88 (s, 1H, methine CH), 6.64 (dd,  $J = 3.1, 8.8$  Hz, 1H, H<sub>Ar</sub>), 6.75-6.79 (m, 2H), 8.97 (s, 1H, NH); <sup>13</sup>C NMR (125 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm): 25.4, 26.9, 28.4, 28.7, 29.4, 31.4, 32.3, 49.7, 54.6, 55.4, 88.3, 109.4, 110.4, 111.7, 117.6, 133.5, 143.7, 149.3, 150.0, 151.9, 159.9, 162.7, 193.7; Mass:  $m/z$  425 (M<sup>+</sup>), 426 (M<sup>+</sup> + 1).

**Product 1k.** <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm): 0.91 (s, 3H, C-CH<sub>3</sub>), 1.05 (s, 3H, C-CH<sub>3</sub>), 2.05 (d,  $J = 16.0$  Hz, 1H, C=C-CH<sub>2</sub>), 2.22 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.52-2.63 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.10 (s, 3H, NCH<sub>3</sub>), 3.46 (s, 3H, NCH<sub>3</sub>), 3.68 (s, 3H,

OCH<sub>3</sub>), 4.82 (s, 1H, methine CH), 6.74 (d,  $J = 8.6$  Hz, 2H, H<sub>Ar</sub>), 7.13 (d,  $J = 8.6$  Hz, 2H, H<sub>Ar</sub>), 8.99 (s, 1H, NH).

**Product 11.** <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>):  $\delta$  (ppm): 0.90 (s, 3H, C-CH<sub>3</sub>), 1.05 (s, 3H, C-CH<sub>3</sub>), 2.05 (d,  $J = 16.0$  Hz, 1H, C=C-CH<sub>2</sub>), 2.22 (d,  $J = 16.1$  Hz, 1H, C=C-CH<sub>2</sub>), 2.52-2.62 (distorted AB system, 2H, O=C-CH<sub>2</sub>), 3.11 (s, 3H, NCH<sub>3</sub>), 3.45 (s, 3H, NCH<sub>3</sub>), 4.78 (s, 1H, methine CH), 6.56 (d,  $J = 8.4$  Hz, 2H, H<sub>Ar</sub>), 7.00 (d,  $J = 8.4$  Hz, 2H, H<sub>Ar</sub>), 8.96 (s, 1H, NH), 9.09 (s, 1H, OH).

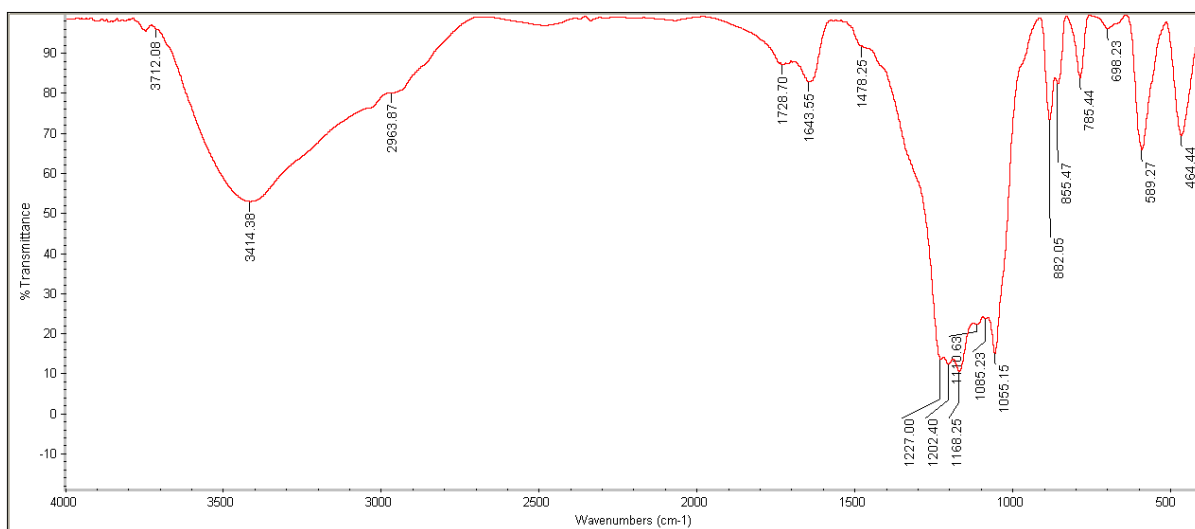


Fig. S1 The FT-IR spectrum of [TSSECM].

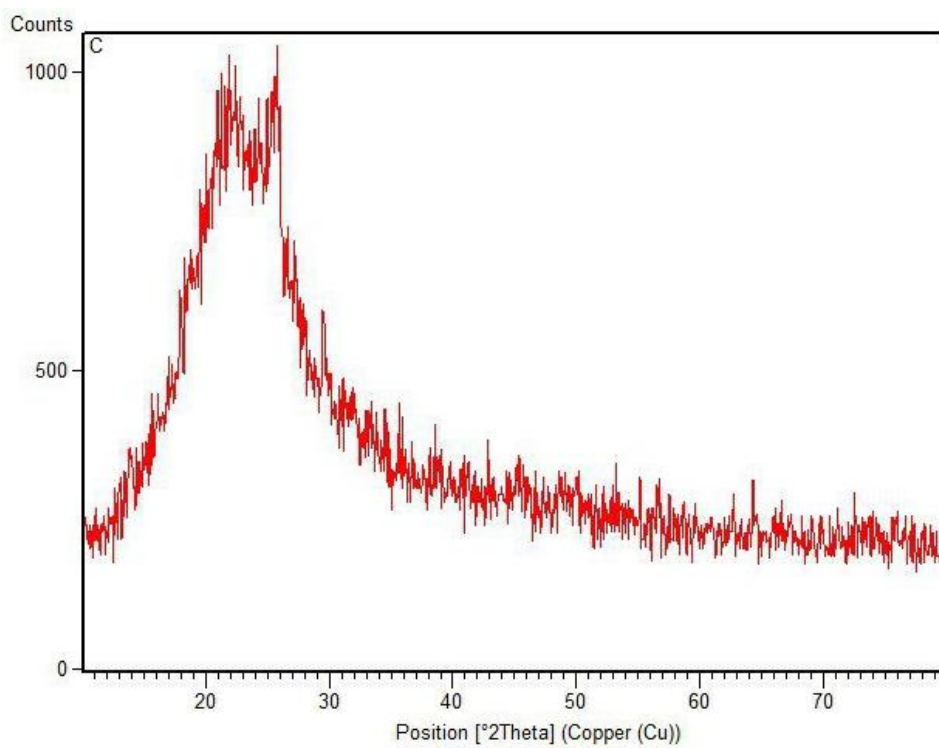


Fig. S2 The XRD pattern of the catalyst.

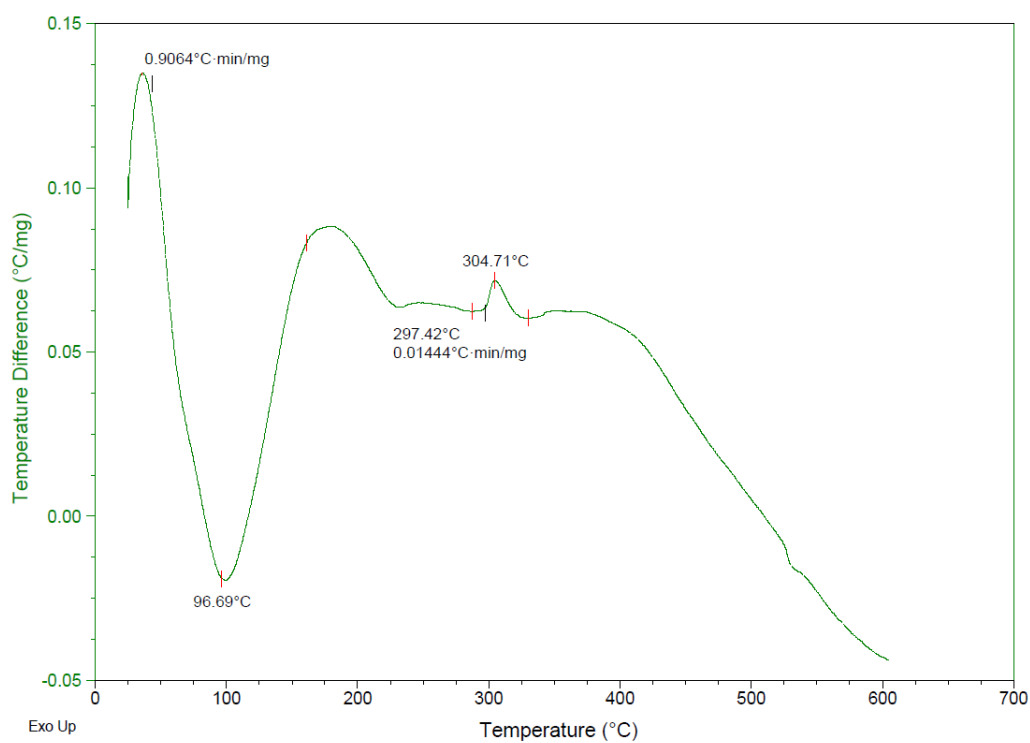
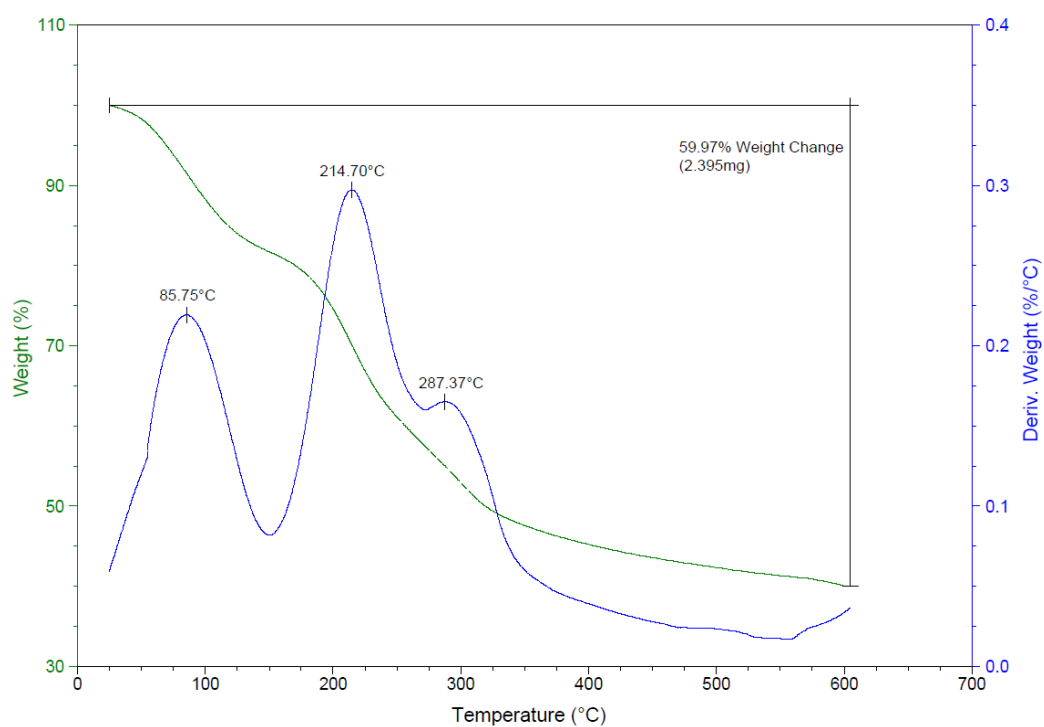


Fig. S3 The TG, DTG and DTA diagrams of the catalyst.

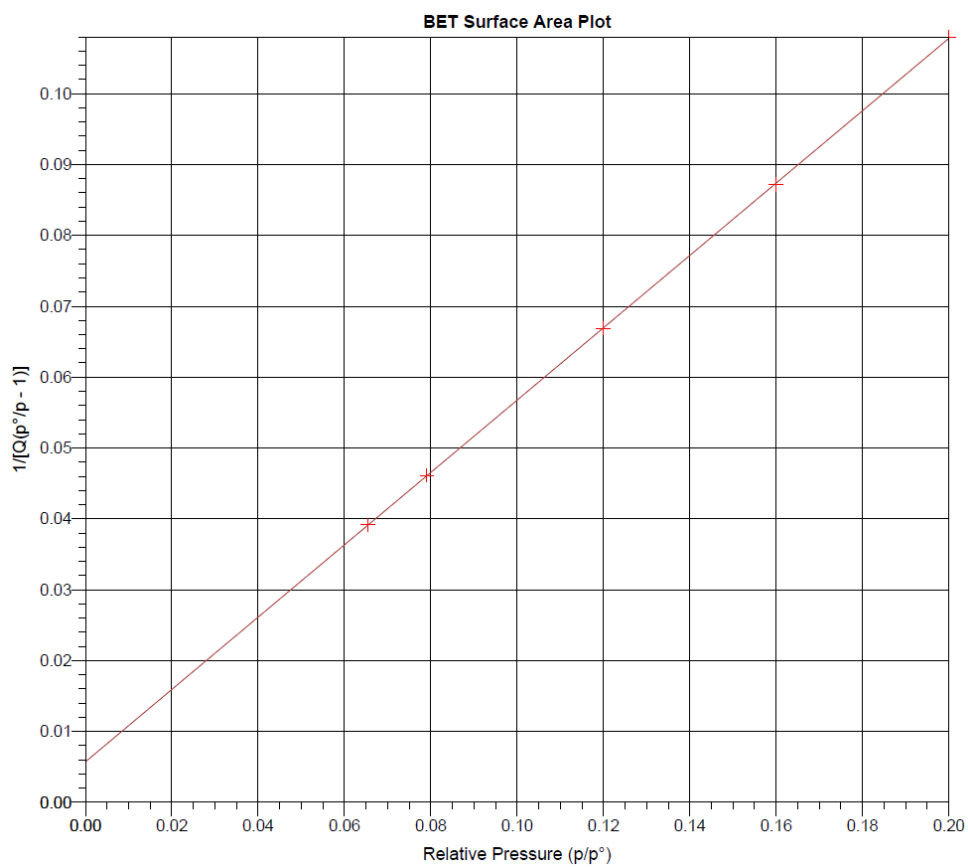


Fig. S4 The BET surface area plot of [TSSECM].



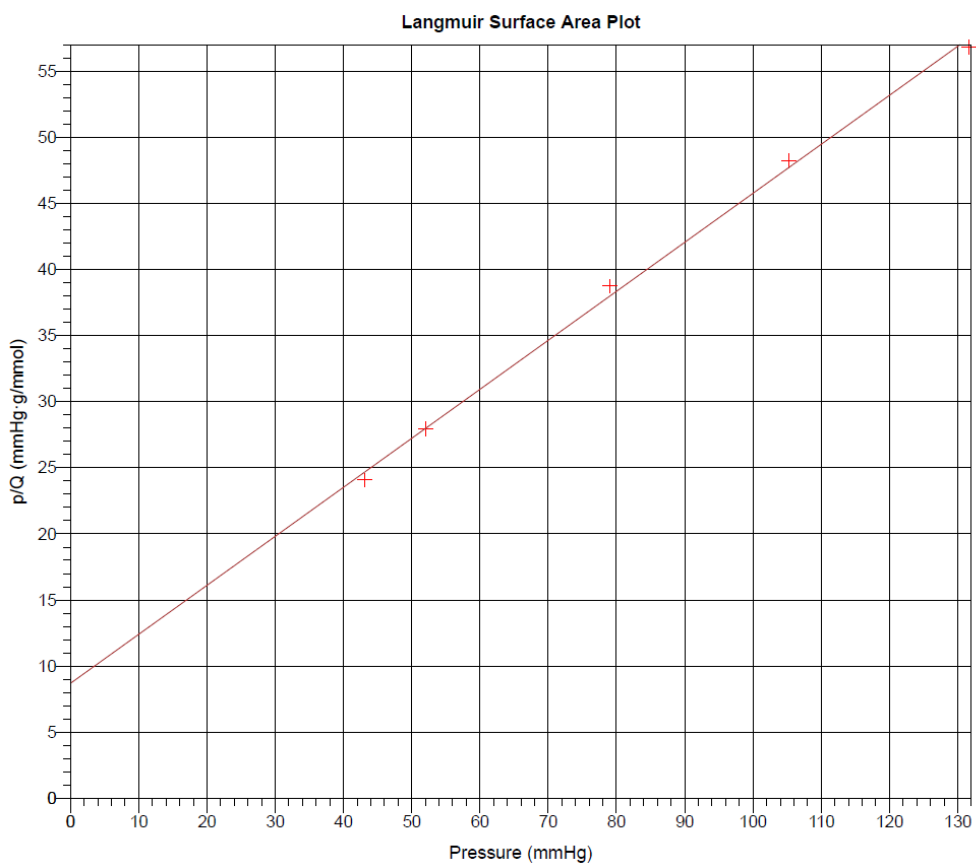


Fig. S5 The Langmuir surface area plot of [TSSECM].

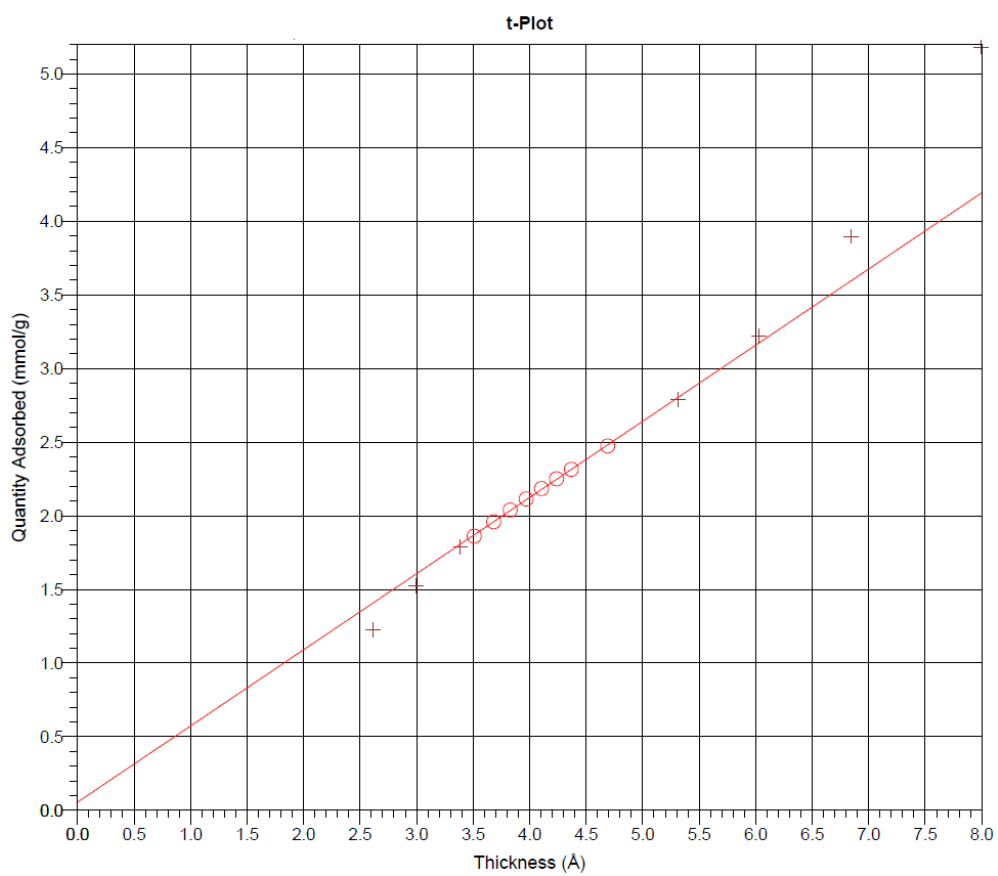


Fig. S6 The t-plot of [TSSECM].

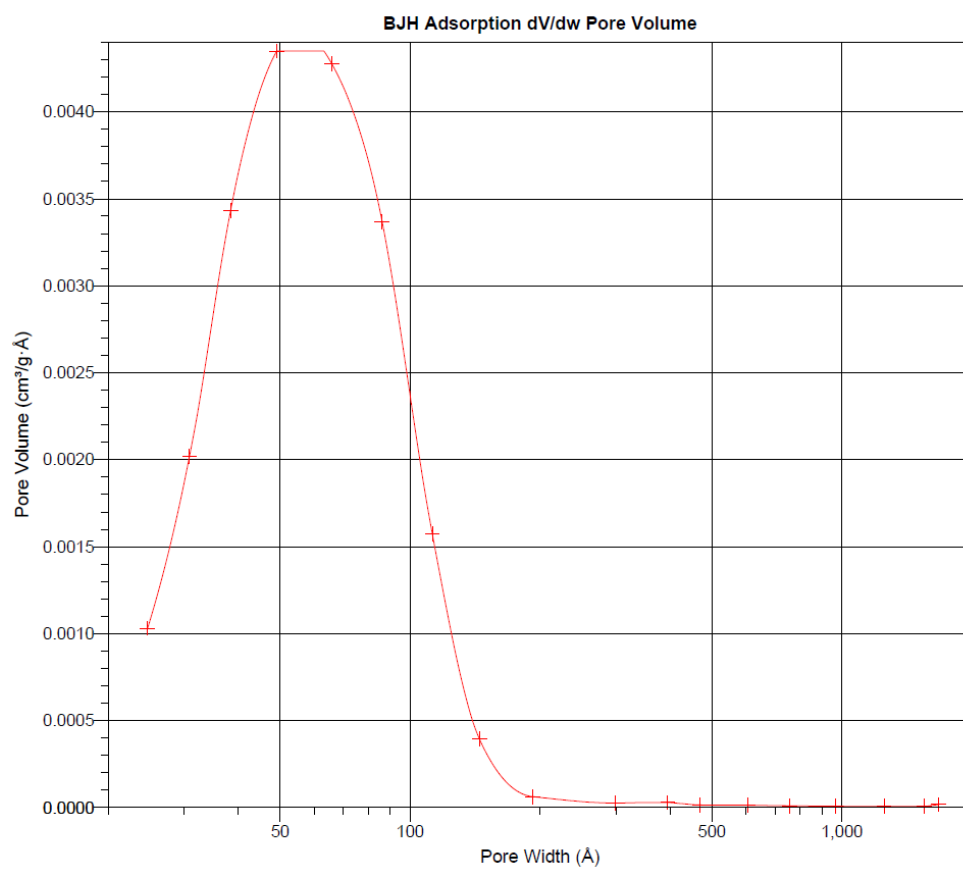


Fig. S7 The BJH plot of [TSSECM].

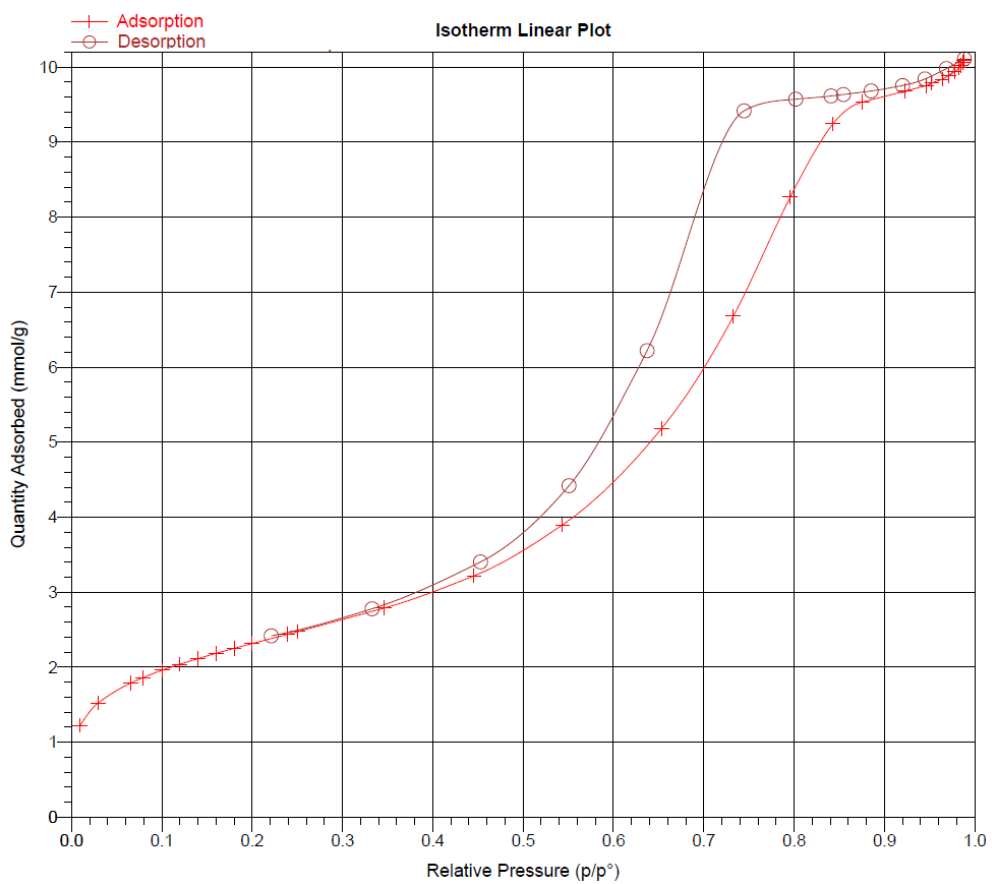


Fig. S8 The  $N_2$  adsorption-desorption isotherm plot of [TSSECM].

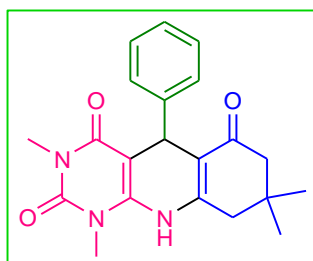
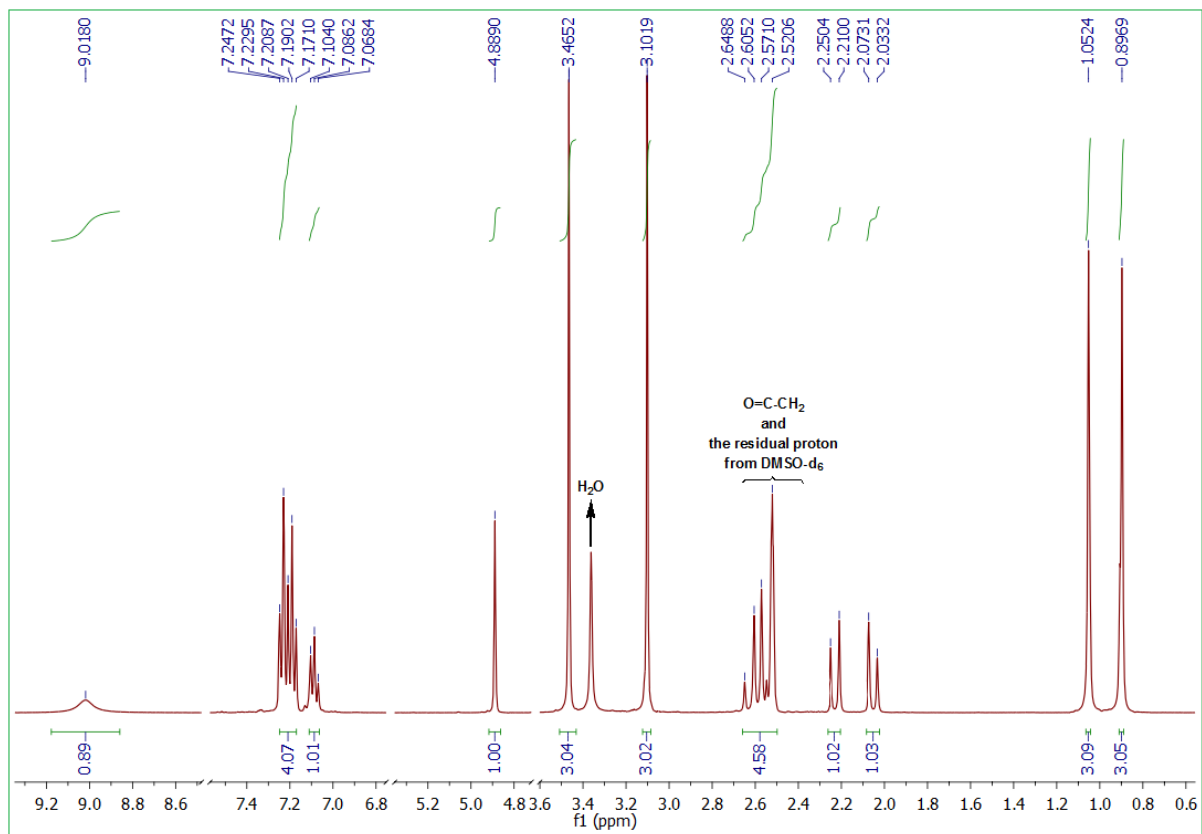


Fig. S9 The <sup>1</sup>H NMR spectrum of compound **1a**.

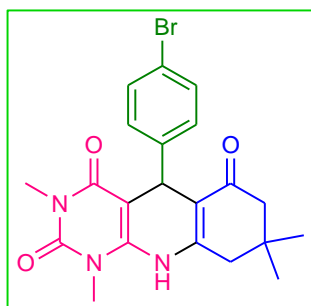
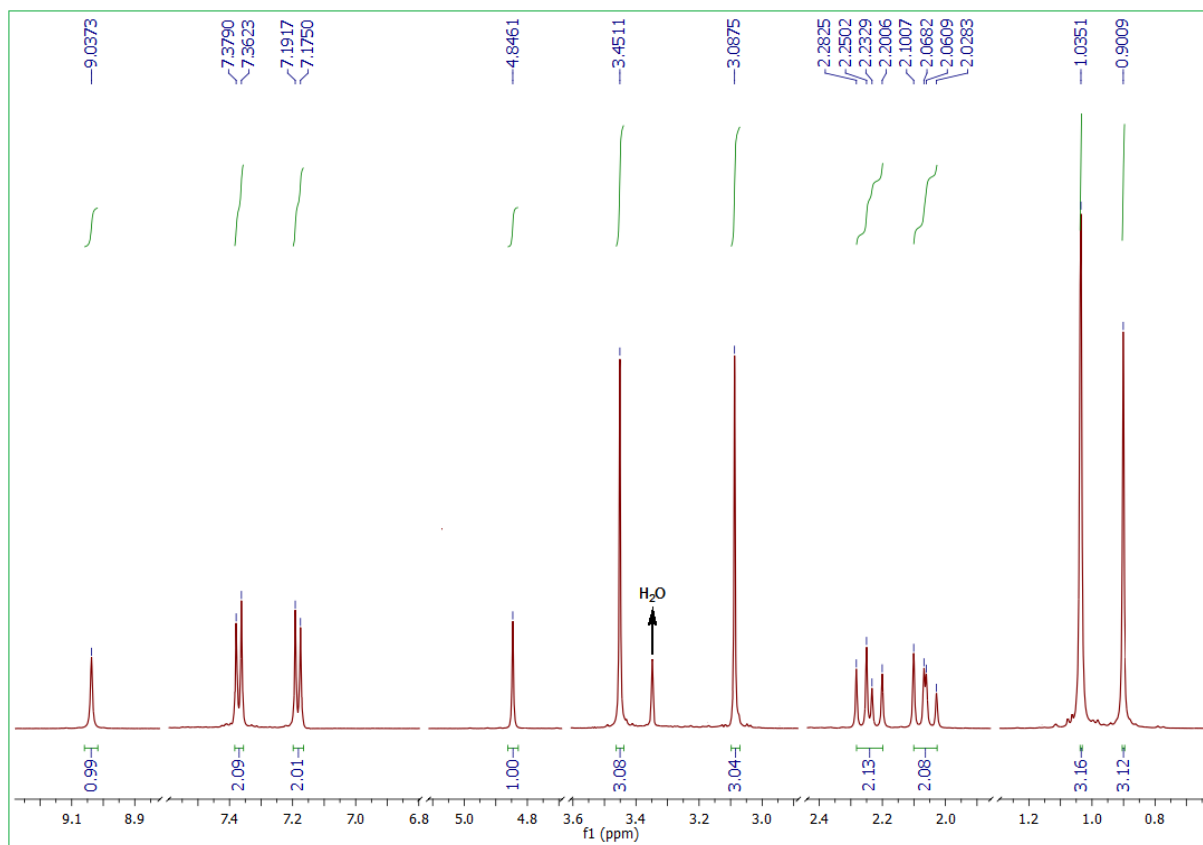


Fig. S10 The <sup>1</sup>H NMR spectrum of compound **1b**.

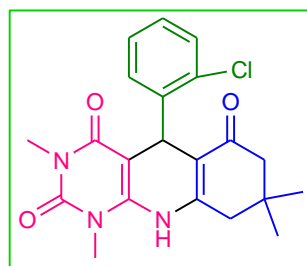
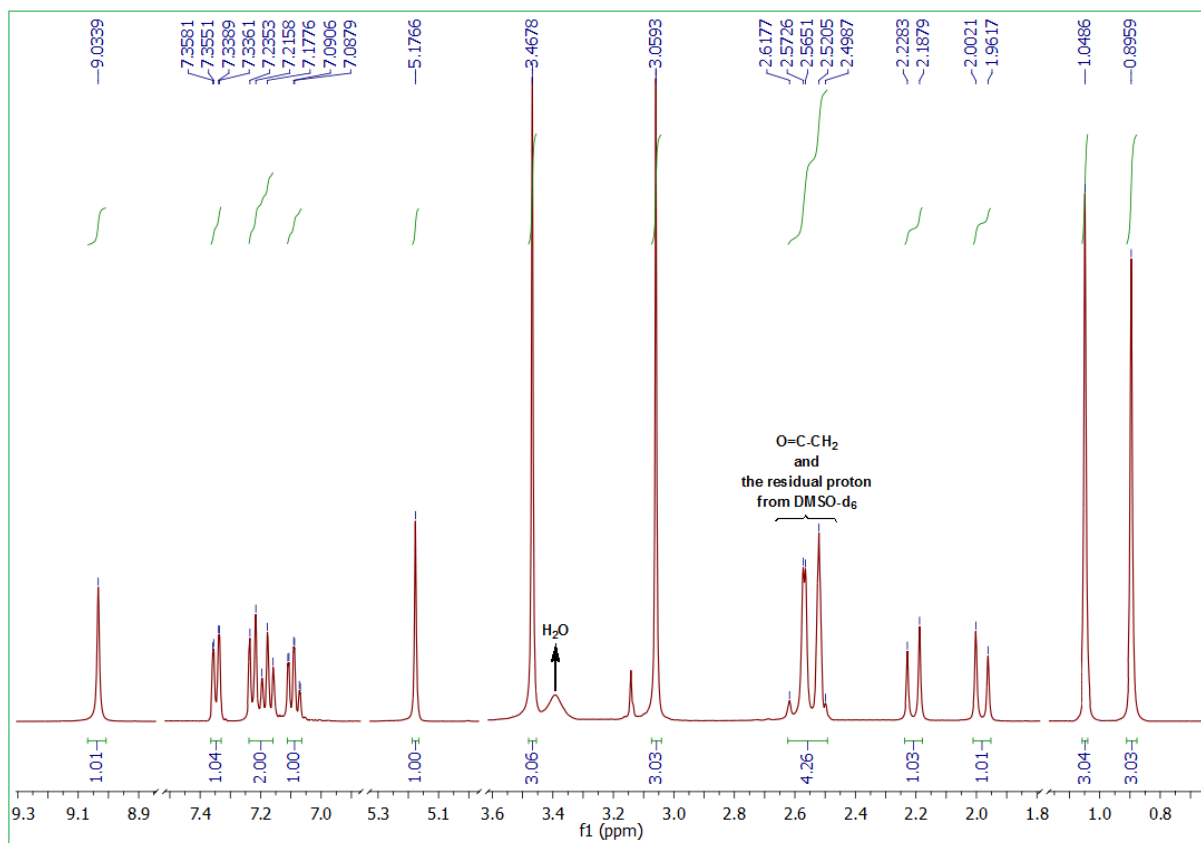


Fig. S11 The <sup>1</sup>H NMR spectrum of compound **1e**.

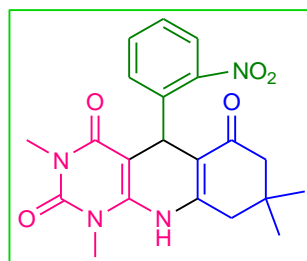
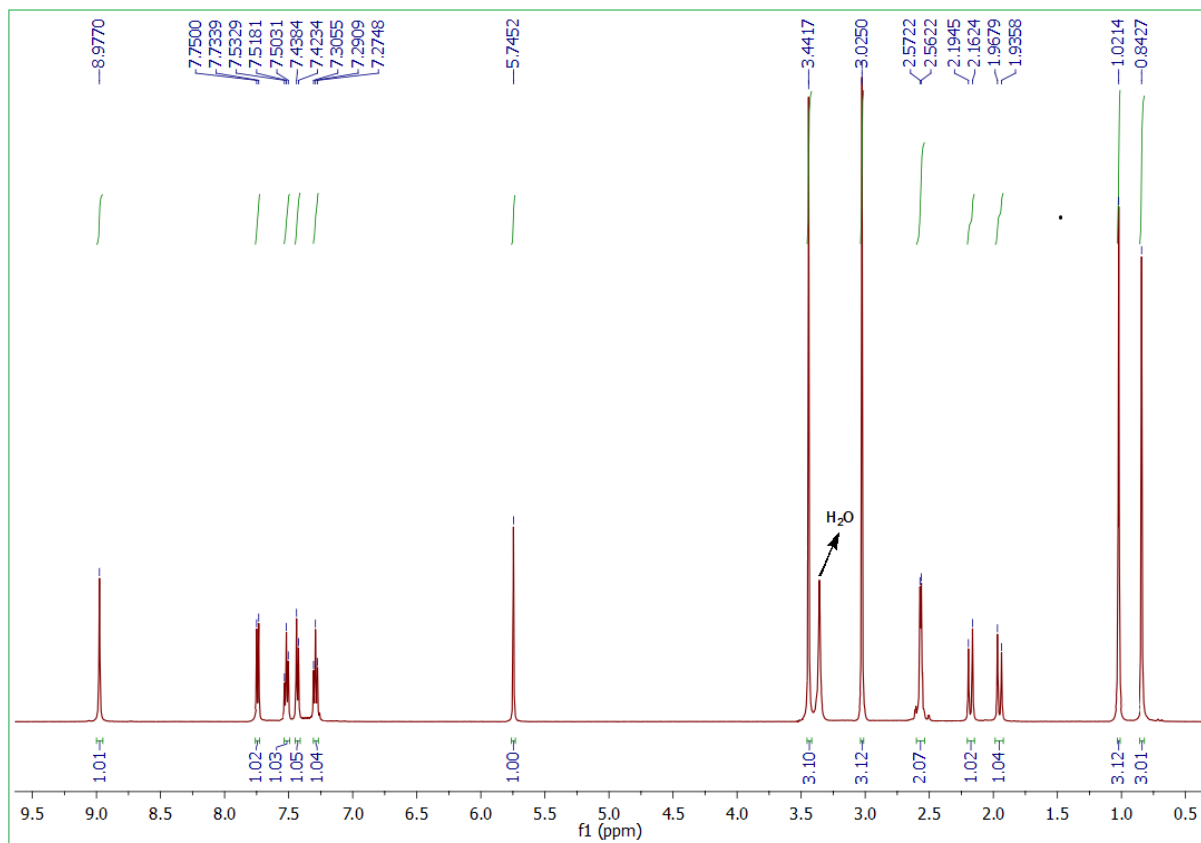


Fig. S12 The <sup>1</sup>H NMR spectrum of compound **1g**.



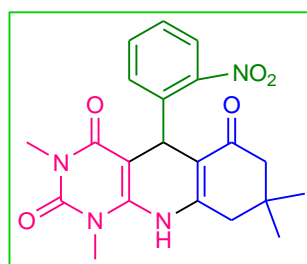
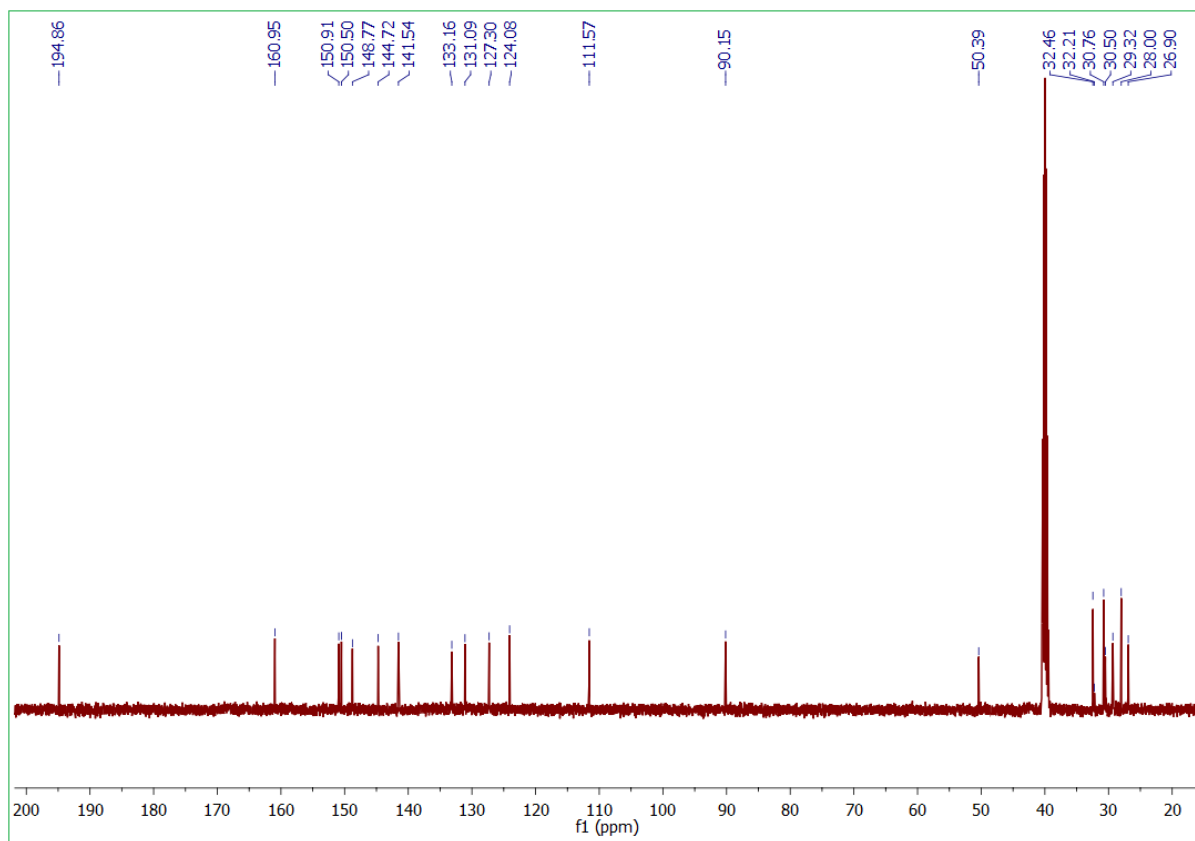


Fig. S13 The  $^{13}\text{C}$  NMR spectrum of compound **1g**.

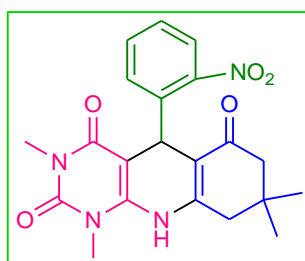
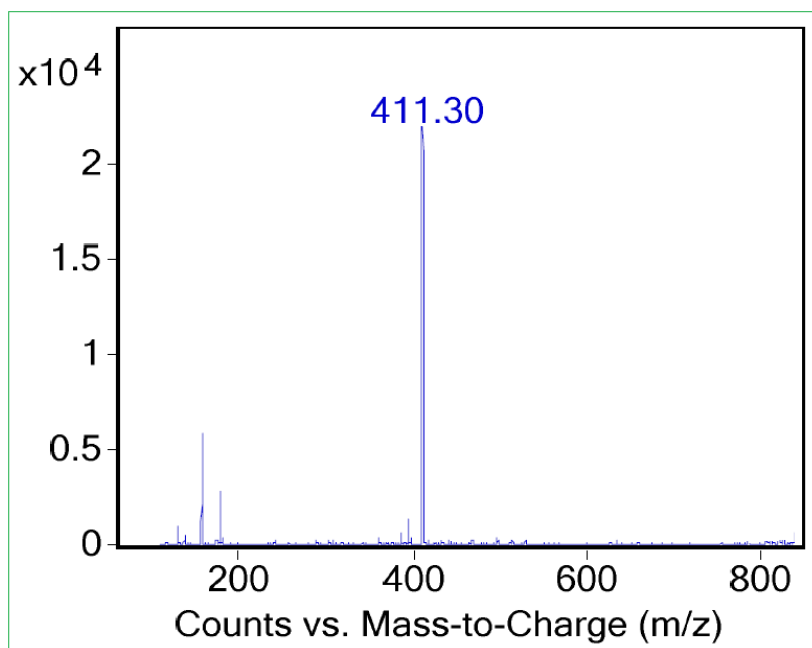


Fig. S14 The mass spectrum of compound **1g**.

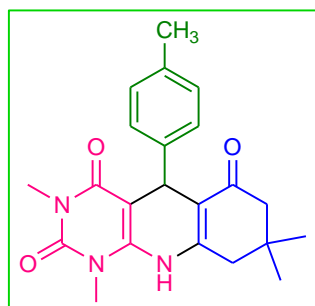
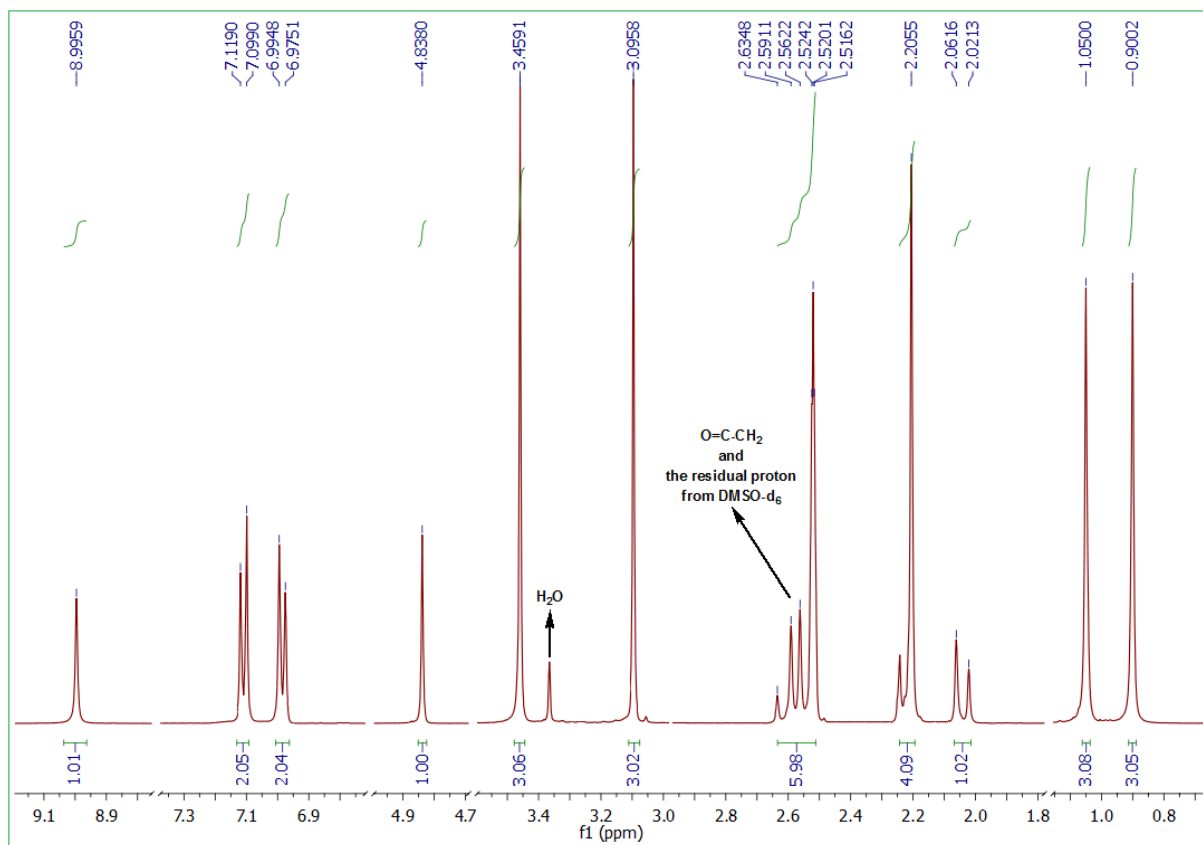


Fig. S15 The <sup>1</sup>H NMR spectrum of compound **1h**.

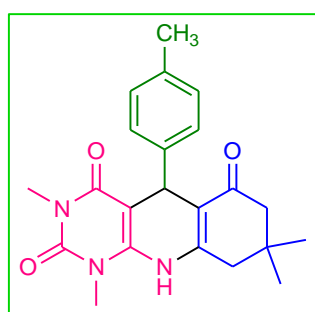
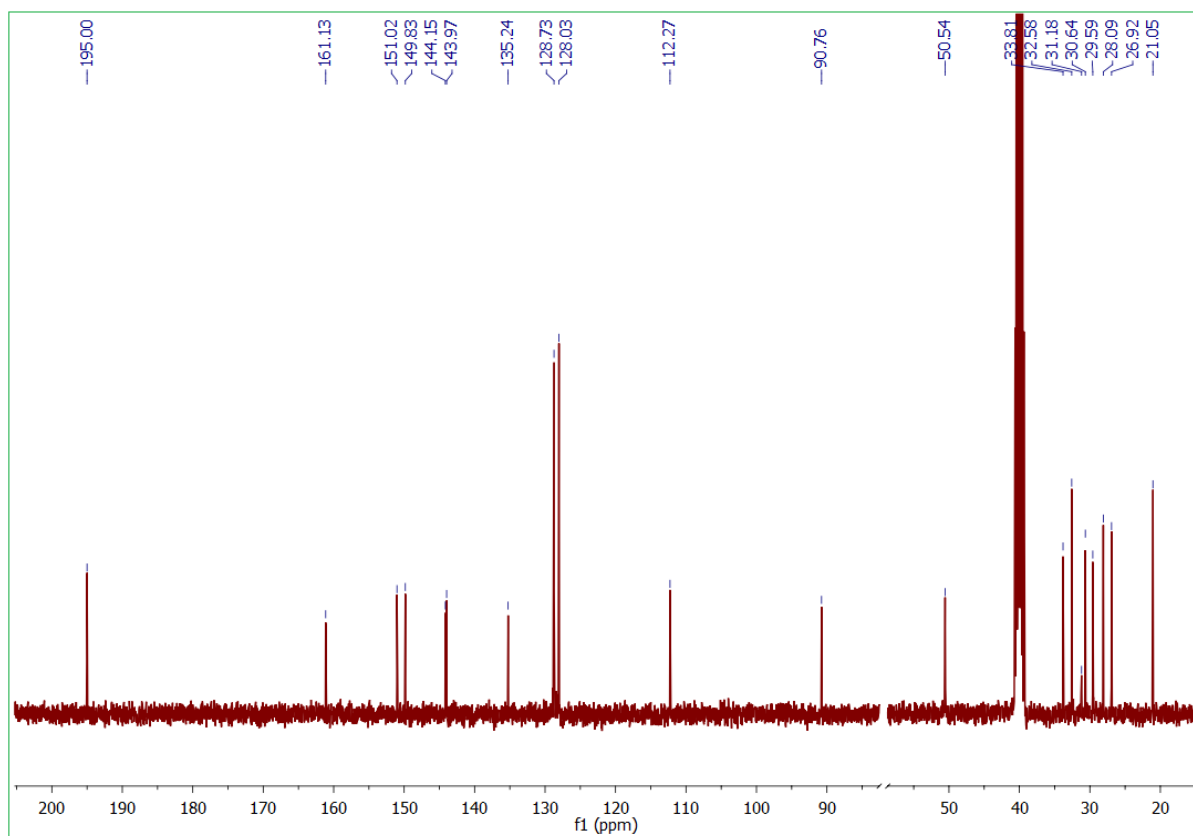


Fig. S16 The  $^{13}\text{C}$  NMR spectrum of compound **1h**.

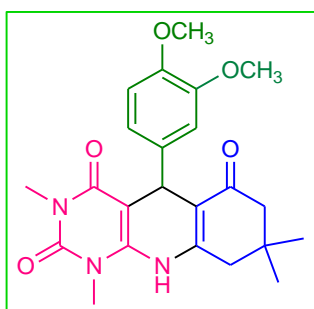
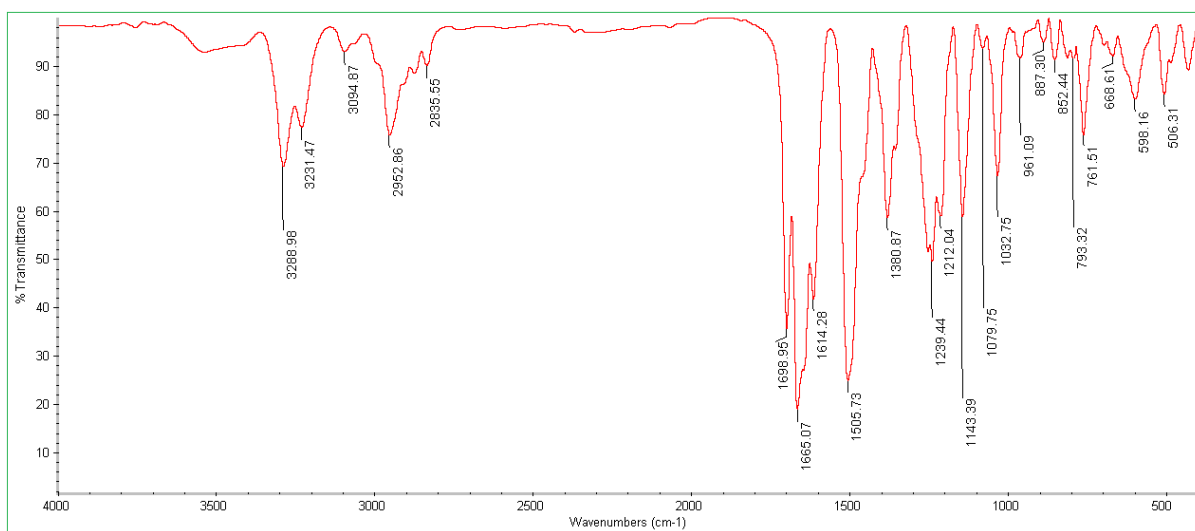


Fig. S17 The FT-IR spectrum of compound **1i**.

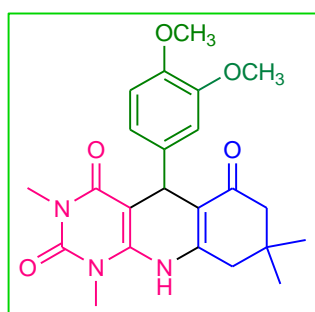
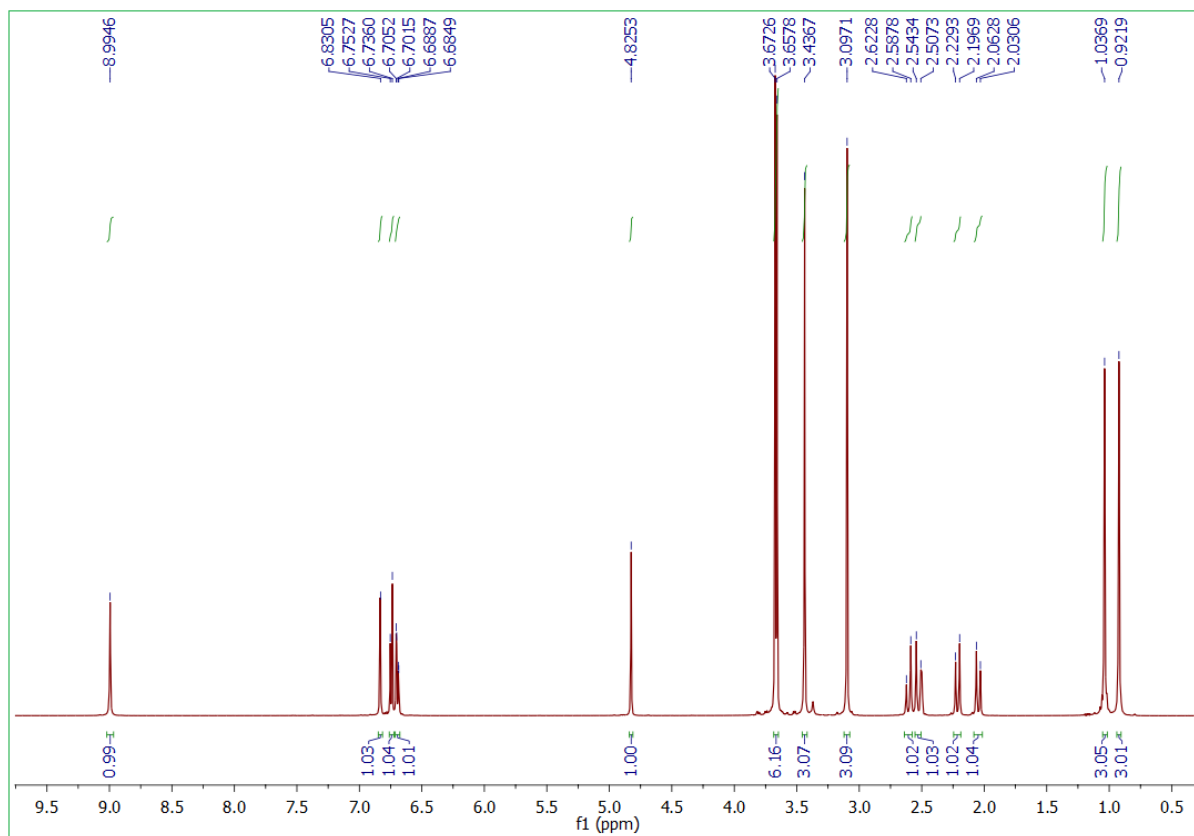


Fig. S18 The  $^1\text{H}$  NMR spectrum of compound **1i**.

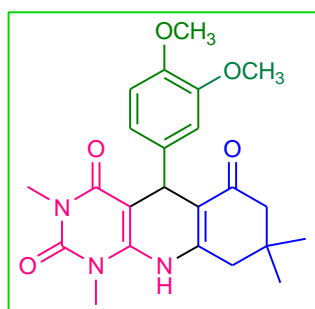
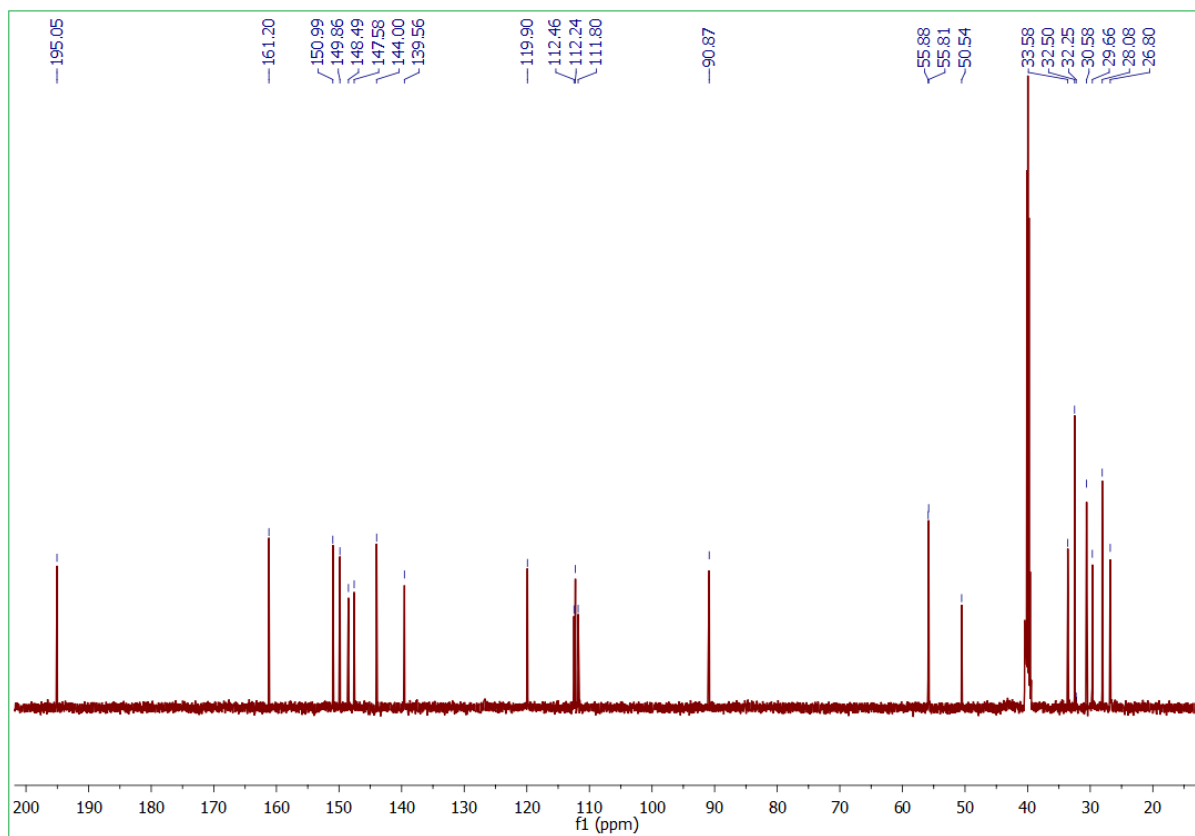


Fig. S19 The  $^{13}\text{C}$  NMR spectrum of compound **1i**.

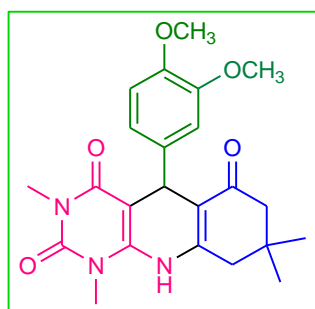
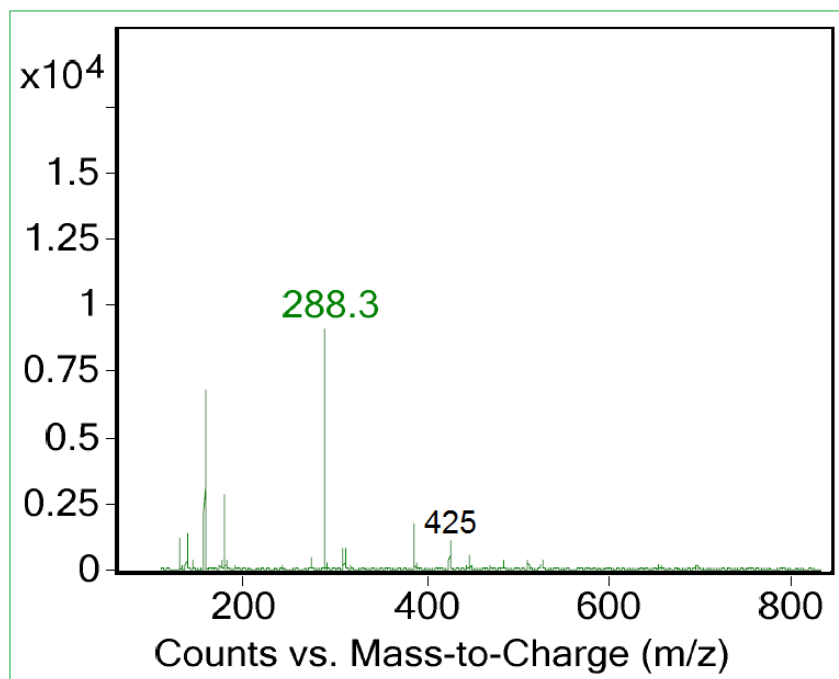


Fig. S20 The mass spectrum of compound **1i**.



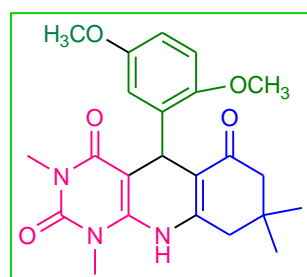
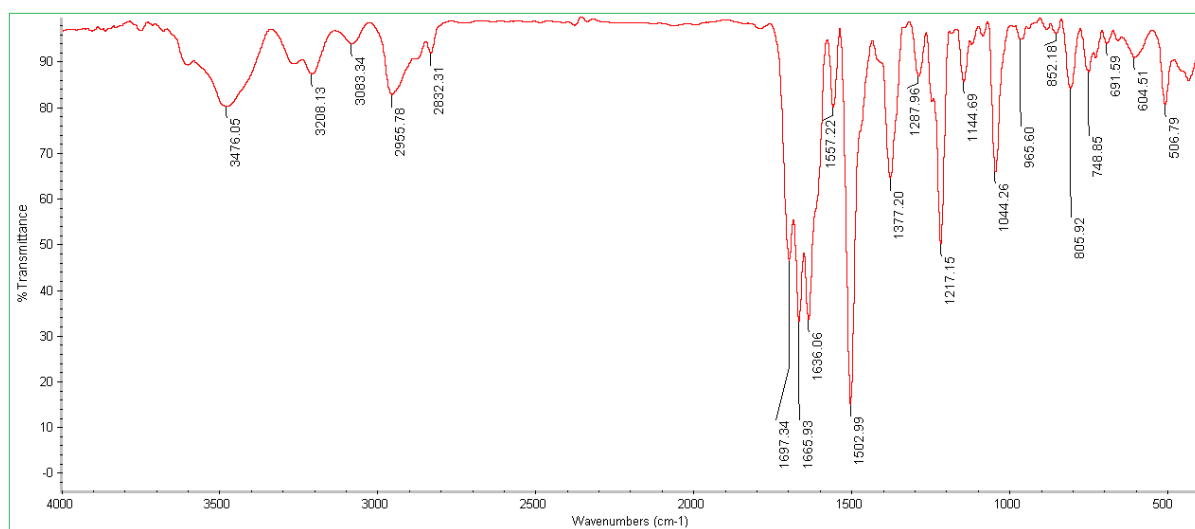


Fig. S21 The FT-IR spectrum of compound **1j**.

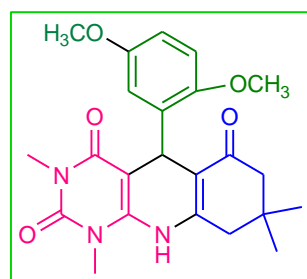
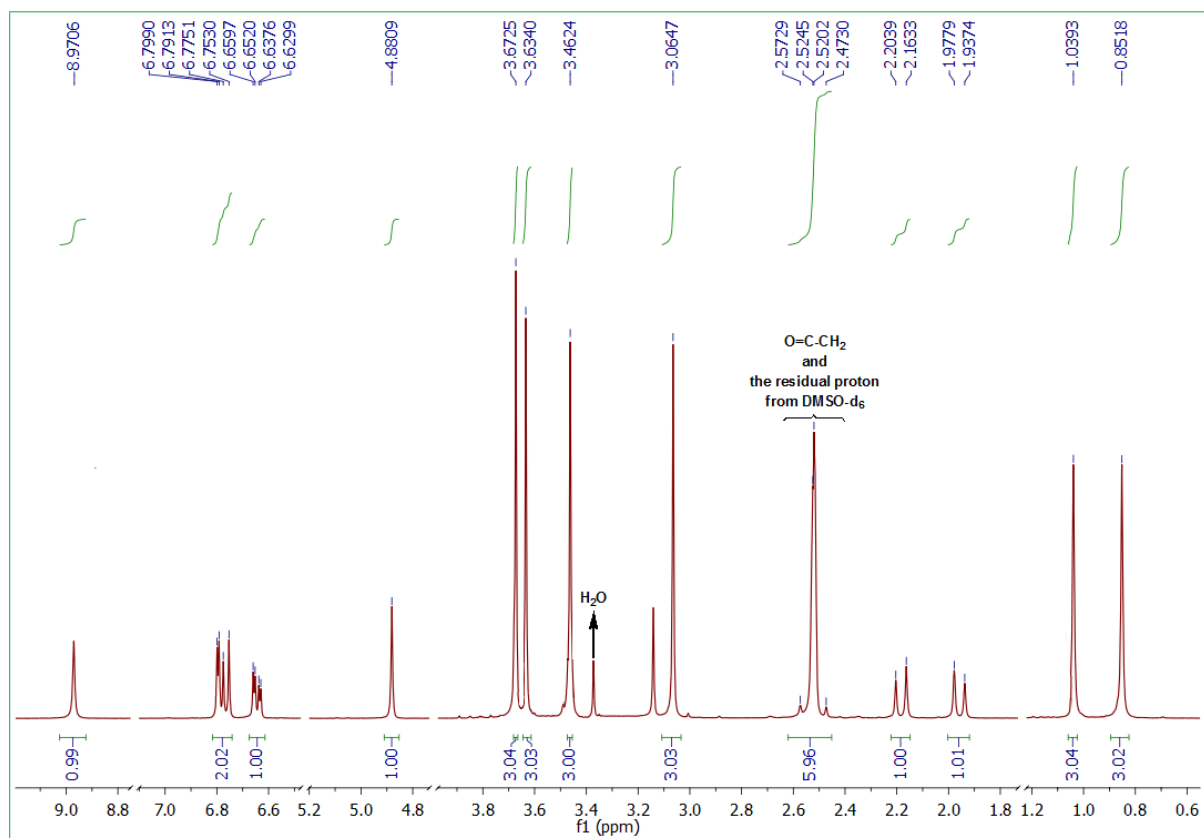


Fig. S22 The <sup>1</sup>H NMR spectrum of compound **1j**.

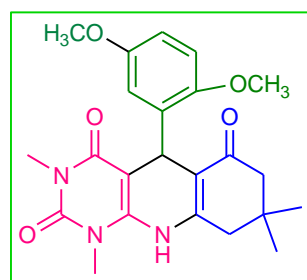
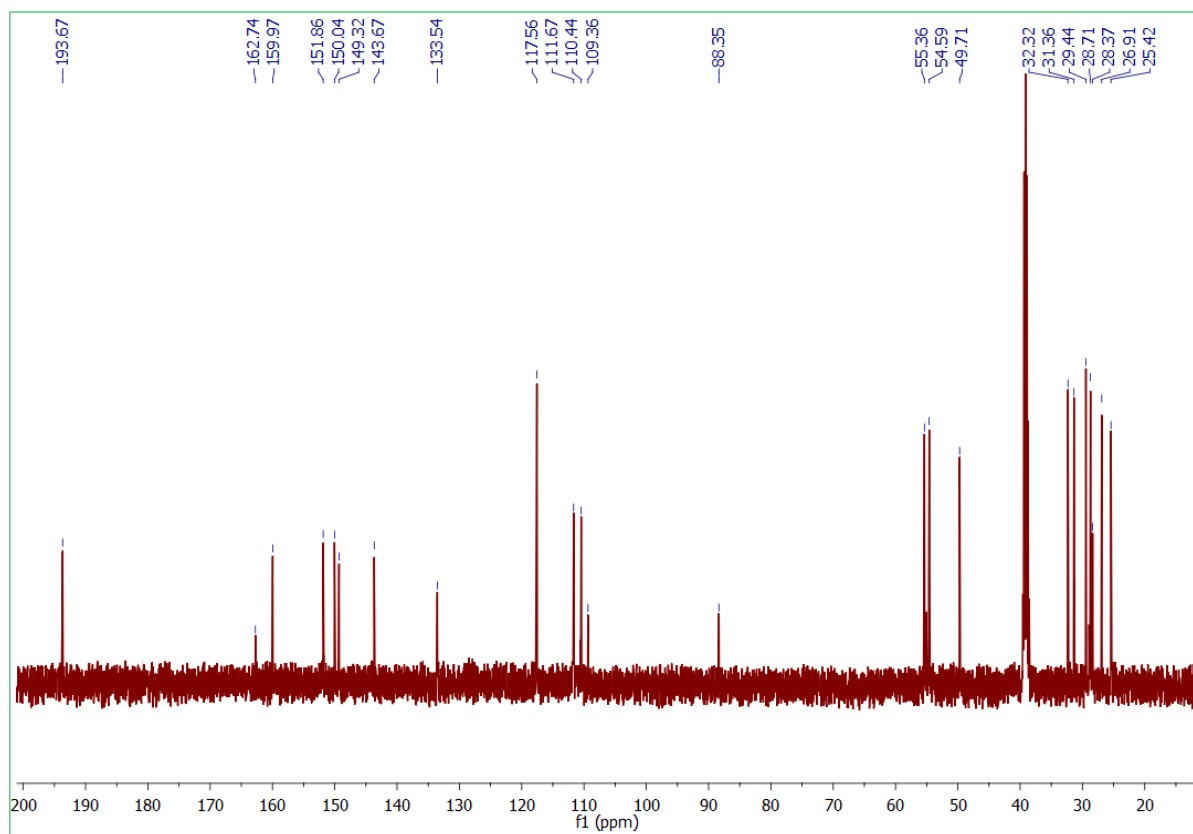


Fig. S23 The  $^{13}\text{C}$  NMR spectrum of compound **1j**.

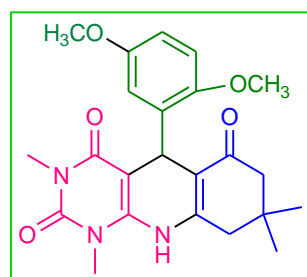
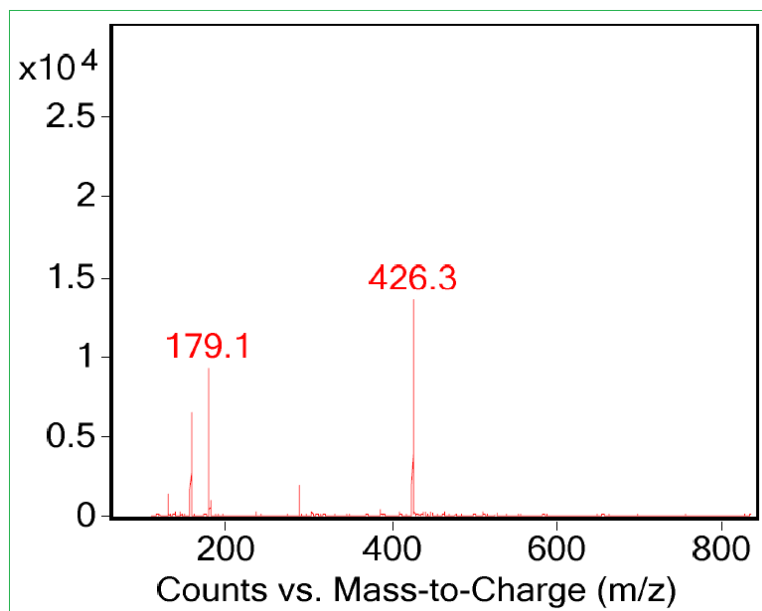


Fig. S24 The mass spectrum of compound **1j**.

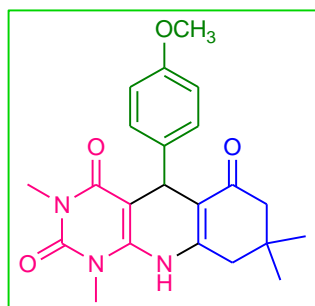
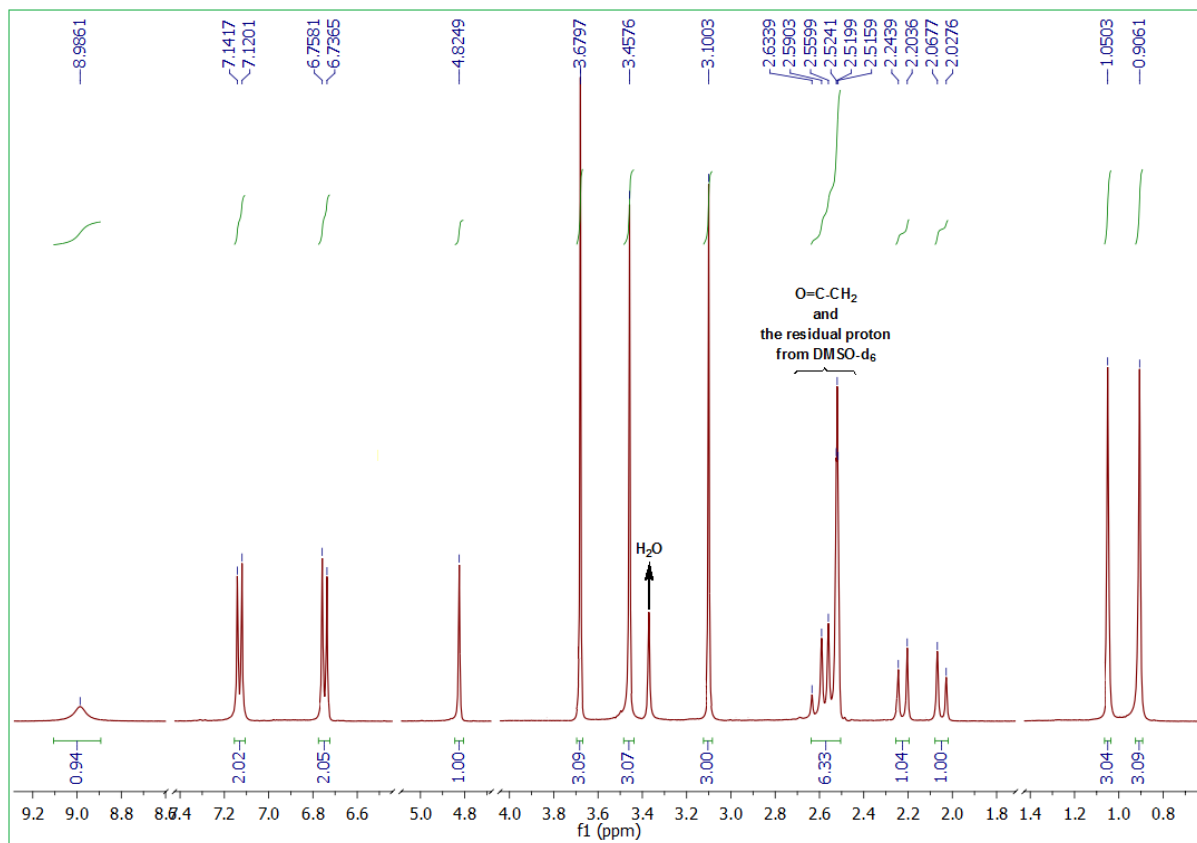


Fig. S25 The <sup>1</sup>H NMR spectrum of compound **1k**.

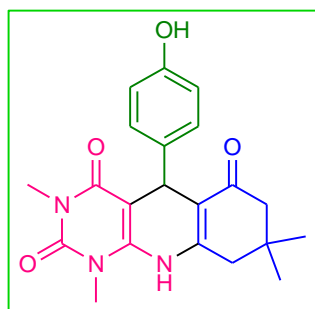
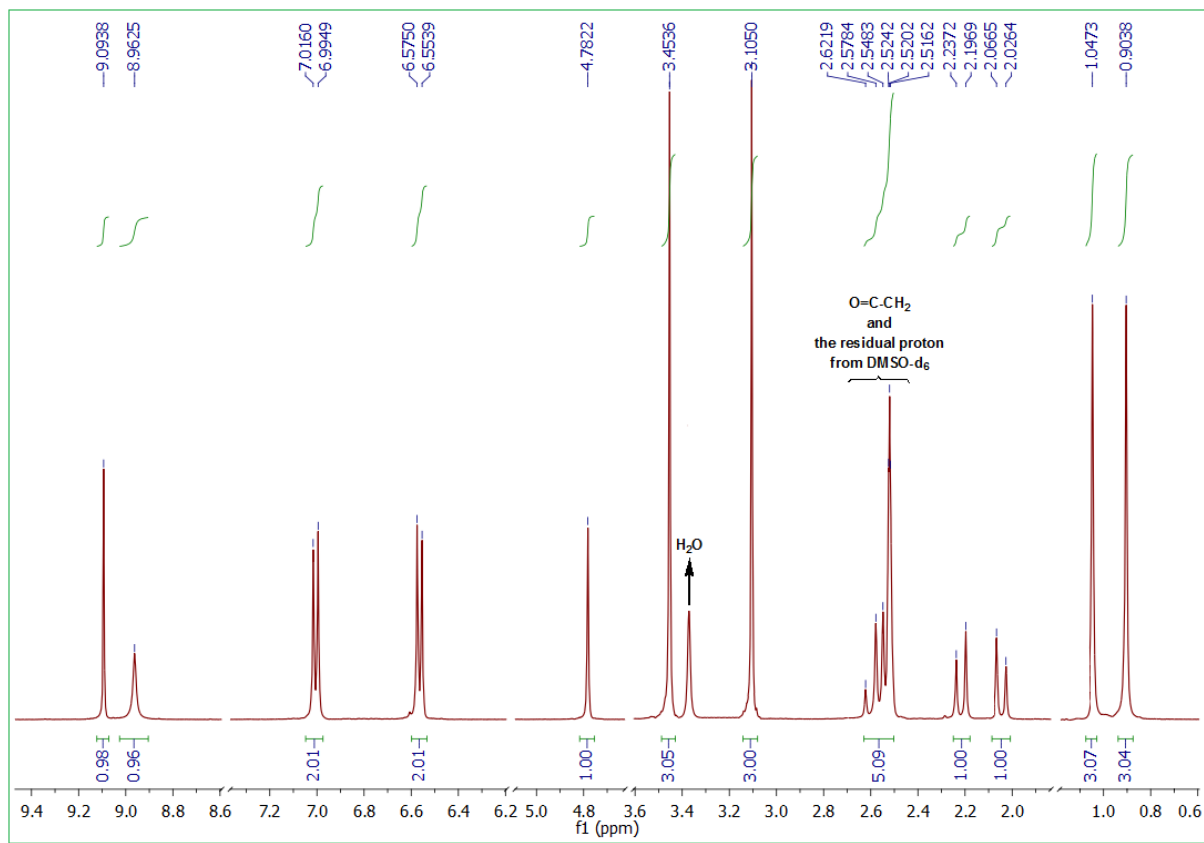


Fig. S26 The <sup>1</sup>H NMR spectrum of compound **11**.