

## Supplementary Information

### Synthesis, Photophysical and Electrochemical Properties of Donor-Acceptor Type Hydrazinyl Thiazolyl Coumarins

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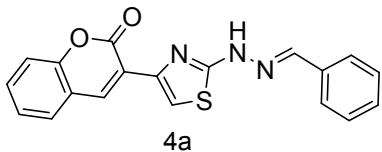
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## General information

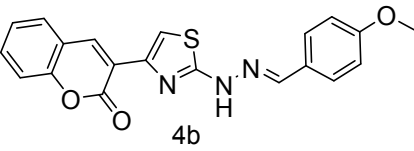
All chemicals and solvents were purchased from commercial sources (Sigma Aldrich, Acros Organics Ltd., and Merck) and were used as received. The  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR (500 MHz and 125 MHz) spectra were recorded on Bruker NMR spectrometer (500 MHz). The data are reported as follows: chemical shift (ppm) and multiplicity [s (singlet), d (doublet), t (triplet), q (quartet), m (multiplet or unresolved) and brs (broad singlet)] and coupling constant(s) are given in Hz. High-Resolution Mass Spectra were recorded on i) Bruker micrOTOF-QII mass spectrometer and ii) Micromass ESI-TOF MS. Mass Spectra were recorded on Shimadzu-LCMS-2010 A mass spectrometer. XRD pattern was recorded using a Rigaku Miniflex 600 diffractometer. Absorbance spectra were recorded on Shimadzu model UV-3100 or Carry 100 Bio UV-Visible spectrophotometer and fluorescence emission spectra were recorded on a Jobin Yvon Horiba model Fluoromax-3 spectrofluorimeter. The quantum yields of titled compounds (4) were calculated using quinine sulphate as standard reference compound ( $\Phi_{\text{em}} = 0.545$  in 1.0 N  $\text{H}_2\text{SO}_4$ ). For HOMO and LUMO calculations, Cyclic voltammetry experiments were performed on a CHI6002E (CH Instruments Inc., USA) electrochemical workstation equipped with a standard three-electrode assembly. Melting points were determined by MR-Vis+ instrument (Labindia) and are uncorrected. Thin layer chromatography (TLC) was performed on 0.25 mm Merck silica gel plates and the developed plates are visualized under UV light. Millipore double distilled water was used for the workup process.

Physical and spectral data (<sup>1</sup>H NMR, <sup>13</sup>C NMR, HRMS and Mass) of synthesized compounds (4a-4ak)

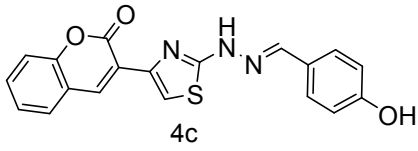
**(E)-3-(2-(2-benzylidenehydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4a):**

 <p>4a</p>	Yellow solid, yield: 98%, mp: 208-210°C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 12.19 (brs, 1H, -NH), 8.55 (s, 1H, Thiazole H), 8.07 (s, 1H, -N=C-H), 7.86 (dd, J=8.0 Hz, J=1.5 Hz, 1H, arom H), 7.78 (s, 1H, Coumarin C <sub>4</sub> -H), 7.68-7.66(m, 2H, arom H), 7.62 (td, J=7.0 Hz, J=1.5 Hz, 1H, arom H), 7.46-7.43 (m, 3H, arom H), 7.41-7.38 (m, 2H, arom H). HRMS (ESI): Anal. Calcd. For C <sub>19</sub> H <sub>14</sub> O <sub>2</sub> N <sub>3</sub> S [M +H] <sup>+</sup> 348.0801; Found: 348.0806.
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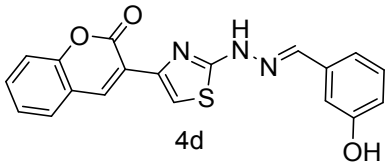
**(E)-3-(2-(2-(4-methoxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one(4b):**

 <p>4b</p>	Yellow solid, yield: 98%. mp: 241-242 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 12.06 (brs, 1H, -NH), 8.53 (s, 1H, Thiazole H), 8.01 (s, 1H, -N=C-H), 7.85 (dd, J=10.0 Hz, J =2.0 Hz, 1H, arom H), 7.75 (s, 1H, Coumarin C <sub>4</sub> -H), 7.63-7.59 (m, 3H, arom H), 7.45 (d, J =10.5 Hz, 1H, arom H), 7.39 (td, J =9.5 Hz, J=1.0 Hz, 1H, arom H), 7.0 (d, J =11.0 Hz, 2H, arom H), 3.80 (s, 3H, -OCH <sub>3</sub> ). HRMS(ESI): Anal. Calcd. For C <sub>20</sub> H <sub>16</sub> O <sub>3</sub> N <sub>3</sub> S [M+H] <sup>+</sup> 378.0907; Found: 378.0915
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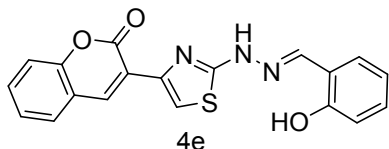
**(E)-3-(2-(2-(4-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4c):**

 <p>4c</p>	Dark brown solid, yield: 96%. mp: 248-250 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 11.94 (s, 1H NH), 8.54 (s, 1H, Thiazole H), 7.97 (s, 1H, -N=C-H), 7.85(dd, J=7.5 Hz, J=1.5 Hz, 1H, arom H), 7.74 (s, 1H, Coumarin C <sub>4</sub> -H), 7.63 (td, J =7.0 Hz, J =1.5 Hz, 1H, arom H), 7.50 (d, J = 8.5 Hz, 2H, arom H), 7.45 (d, J = 9.0 Hz, 1H, arom H), 7.39 (td, J = 8.0 Hz, J= 1.0 Hz, 1H, arom H), 6.82 (d, J=9.0 Hz, 2H, arom H).
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**(E)-3-(2-(2-(3-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4d):**

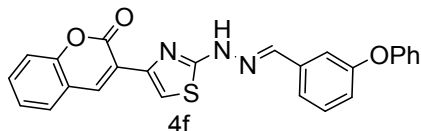
 <p>4d</p>	Yellow solid, yield: 96%. mp: 250-252 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 12.14 (s, 1H, -NH), 9.60 (brs, 1H, -OH), 8.53 (s, 1H, Thiazole H), 7.97 (s, 1H, -N=C-H), 7.85(d, J=8.0 Hz, 1H, arom H), 7.77 (s, 1H, Coumarin C <sub>4</sub> -H), 7.62 (td, J =8.5 Hz, J =1.5 Hz, 1H, arom H), 7.45 (d, J=8.0 Hz, 1H, arom H), 7.38 (t, J=7.5 Hz, 1H, arom H), 7.22 (t, J =8.0 Hz, 1H, arom H), 7.12 (s, 1H, arom H), 7.04 (d, J =8.0 Hz, 1H, arom H), 6.79 (dd, J=8.0 Hz, J=2.0 Hz 1H, arom H).
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**(E)-3-(2-(2-(2-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4e):**



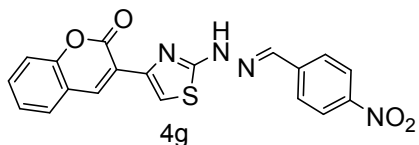
Yellow solid, yield: 94%. mp: 248-250 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.06 (brs, 1H, -NH), 10.09 (brs, 1H, OH), 8.50 (s, 1H, Thiazole **H**), 8.36 (s, 1H, -N=C-**H**), 7.81(d, *J*=8.0 Hz, 1H, arom H), 7.73 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.62 (d, *J*=7.5 Hz, 1H, arom H), 7.58 (t, *J*=7.5 Hz, 1H, arom H), 7.41 (d, *J*=7.5 Hz, 1H, arom H), 7.35 (t, *J*=7.5 Hz, 1H, arom H), 7.21 (t, *J*=8.0 Hz, 1H, arom H), 6.91(d, *J*=8.5 Hz, 1H, arom H), 6.86 (t, *J*=7.5 Hz, 1H, arom H).

**(E)-3-(2-(2-(3-phenoxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4f):**



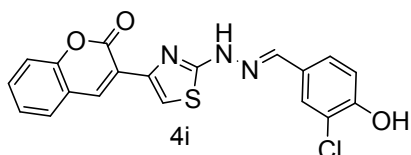
Yellow solid, yield: 95%. mp: 248-250 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.25 (brs, 1H, -NH), 8.51 (s, 1H, Thiazole **H**), 8.03 (s, 1H, -N=C-**H**), 7.83 (dd, *J*=9.5 Hz, *J*=1.5 Hz, 1H, arom H), 7.75 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.62 (td, *J*=9.5 Hz, *J*=2.0 Hz, 1H, arom H), 7.46-7.41 (m, 5H, arom H), 7.38 (td, *J*=10.0 Hz, *J*=1.5 Hz, 1H, arom H), 7.28 (s, 1H, arom H), 7.18 (tt, *J*=9.0 Hz, *J*=1.5 Hz, 1H, arom H), 7.06 (dd, *J*=11 Hz, *J*=1.0 Hz, 2H, arom H), 7.01 (dt, *J*=9.5 Hz, *J*=2.5 Hz, 1H, arom H). HRMS(ESI): Anal. Calcd. For C<sub>25</sub>H<sub>18</sub>O<sub>3</sub>N<sub>3</sub>S [M+H]<sup>+</sup> 440.1063; Found: 440.1065.

**(E)-3-(2-(2-(4-nitrobenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4g):**



Orange solid, yield: 96%, mp: 236-238 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.66 (brs, 1H, -NH), 8.55 (s, 1H, Thiazole **H**), 8.28 (d, *J*=9.0 Hz, 2H, arom H), 8.17 (s, 1H, -N=C-**H**), 7.91 (d, *J*=8.5 Hz, 2H, arom H), 7.87 (d, *J*=8.0 Hz, 1H, arom H), 7.84 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.64 (td, *J*=8.5 Hz, *J*=1.5 Hz, 1H, arom H), 7.46 (d, *J*=8.5 Hz, 1H, arom H), 7.40 (t, *J*=8.0 Hz, 1H, arom H).

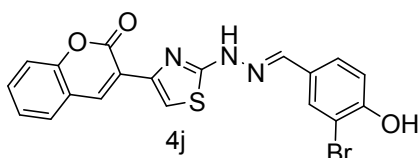
**(E)-3-(2-(2-(3-chloro-4-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4i):**



Yellow solid, yield: 93%, mp: 232-234 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.26 (brs, 1H, -NH), 10.36 (s, 1H, -OH), 8.54 (s, 1H, Thiazole **H**), 8.29 (s, 1H, -N=C-**H**), 7.86 (dd, *J*=9.5 Hz, *J*=2.0 Hz, 1H, arom H), 7.78 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.65-7.61 (m, 2H, arom H), 7.45 (d, *J*=10.0 Hz, 1H, arom H), 7.39 (td, *J*=9.5

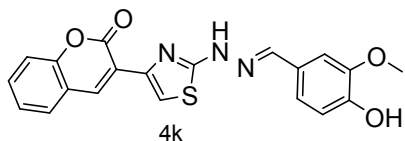
Hz,  $J=1.0$  Hz, 1H, arom H), 7.24 (dd,  $J=11.0$  Hz,  $J=3.5$  Hz, 1H, arom H), 6.92 (d,  $J=11.0$  Hz, 1H, arom H). HRMS (ESI): Anal. Calcd. For  $C_{19}H_{11}O_3N_3ClS[M-H]^+$  396.02; Found: 396.02.

**(E)-3-(2-(2-(3-bromo-4-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4j):**



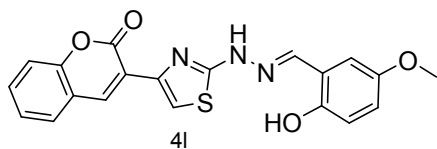
Yellow solid, yield: 92%, mp: 242-244 °C.  $^1H$  NMR (500MHz,  $d_6$ -DMSO):  $\delta$  12.28 (s, 1H, -NH), 10.40 (s, 1H, -OH), 8.54 (s, 1H, Thiazole **H**), 8.28 (s, 1H, -N=C-**H**), 7.86 (d,  $J=7.5$  Hz,  $J=1.5$  Hz, 1H, arom H), 7.78 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.76 (d,  $J=3.0$  Hz, 1H, arom H), 7.63 (td,  $J=7.5$  Hz,  $J=2.0$  Hz, 1H, arom H), 7.46 (d,  $J=8.0$  Hz, 1H, arom H), 7.39 (td,  $J=7.0$  Hz,  $J=1.0$  Hz, 1H, arom H), 7.36 (dd,  $J=9.0$  Hz,  $J=3.0$  Hz, 1H, arom H), 6.87 (d,  $J=8.5$  Hz, 1H, arom H). HRMS (ESI):  $[M-H]^+$ 441.2967.

**(E)-3-(2-(2-(4-hydroxy-3-methoxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4k):**

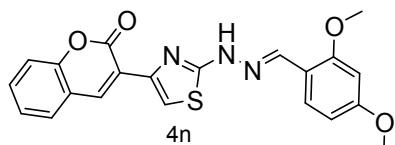


Dark yellow solid, yield: 96%. mp: 224-226 °C.  $^1H$  NMR (500MHz,  $d_6$ -DMSO):  $\delta$  12.05 (brs, 1H, -NH), 8.54 (s, 1H, Thiazole **H**), 7.97 (s, 1H, -N=C-**H**), 7.86 (d,  $J=9.5$  Hz, 1H, arom H), 7.75 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.63 (td,  $J=9.5$  Hz,  $J=2.0$  Hz, 1H, arom H), 7.46 (d,  $J=10.0$  Hz, 1H, arom H), 7.39 (t,  $J=9.0$  Hz, 1H, arom H), 7.25 (d,  $J=2.5$  Hz, 1H, arom H), 7.08 (dd,  $J=9.5$  Hz,  $J=2.0$  Hz, 1H, arom H), 6.83 (d,  $J=10.0$  Hz, 1H, arom H), 3.83 (s, 3H, -OCH<sub>3</sub>). HRMS (ESI): Anal. Calcd. For  $C_{20}H_{16}O_4N_3S[M+H]^+$  394.0856; Found: 394.0864.

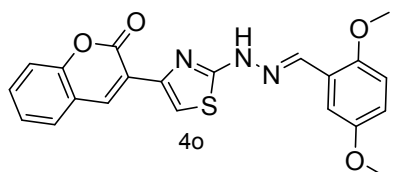
**(E)-3-(2-(2-(2-hydroxy-5-methoxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4l):**



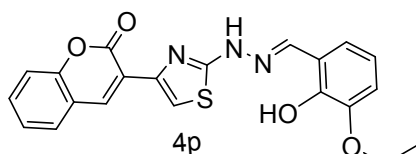
Light yellow solid, yield: 93%. mp: 245-247 °C.  $^1H$  NMR (500MHz,  $d_6$ -DMSO):  $\delta$  12.20 (s, 1H, -NH), 9.65 (s, 1H, -OH), 8.55 (s, 1H, Thiazole **H**), 8.32 (s, 1H, -N=C-**H**), 7.86 (dd,  $J=8.0$  Hz,  $J=1.5$  Hz, 1H, arom H), 7.77 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.63 (td,  $J=7.5$  Hz,  $J=1.5$  Hz, 1H, arom H), 7.46 (d,  $J=8.0$  Hz, 1H, arom H), 7.39 (td,  $J=8.0$  Hz,  $J=1.0$  Hz, 1H, arom H), 7.18 (d,  $J=2.5$  Hz, 1H, arom H), 6.87-6.82 (m, 2H, arom H), 3.72 (s, 3H, -OCH<sub>3</sub>). HRMS (ESI):  $[M+H]^+$ 394.0858.

**(E)-3-(2-(2-(2,4-dimethoxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4n):**

Brownish yellow solid, yield: 96%, mp: 242-244 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.02 (brs, 1H, -NH), 8.54(s, 1H, Thiazole **H**), 8.31 (s, 1H, -N=C-**H**), 7.87 (d, *J*=10.0 Hz, 1H, arom H), 7.74 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.72 (d, *J*=11.0 Hz, 1H, arom H), 7.63 (td, *J*=11.0 Hz, *J*=2.0 Hz, 1H, arom H), 7.46 (d, *J*=10.5Hz, 1H, arom H), 7.39 (td, *J*=9.5 Hz, *J*=1.0 Hz, 1H, arom H), 6.64-6.62 (m, 2H, arom H), 3.86 (s, 3H, -OCH<sub>3</sub>), 3.82 (s, 3H, -OCH<sub>3</sub>). HRMS (ESI): [M+H]<sup>+</sup>408.1021.

**(E)-3-(2-(2-(2,5-dimethoxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one(4o):**

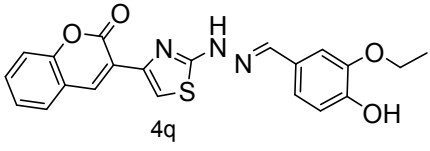
Yellow solid, yield: 95%. mp: 236-238 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.18 (brs, 1H, -NH), 8.53 (s, 1H, Thiazole **H**), 8.33 (s, 1H, -N=C-**H**), 7.85 (dd, *J*=7.5 Hz, *J*=1.5 Hz, 1H, arom H), 7.76 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.62 (td, *J*=7.0 Hz, *J*=1.5 Hz, 1H, arom H), 7.44 (d, *J*=8.5Hz, 1H, arom H), 7.38 (td, *J*=7.5 Hz, *J*=1.0 Hz, 1H, arom H), 7.29 (d, *J*=3.0 Hz, 1H, arom H), 7.02 (d, *J*=9.0 Hz, 1H, arom H), 6.95 (dd, *J*=9.0 Hz, *J*=3.0 Hz, 1H, arom H), 3.80 (s, 3H, -OCH<sub>3</sub>), 3.75 (s, 3H, -OCH<sub>3</sub>). HRMS (ESI): [M+H]<sup>+</sup>408.1017.

**(E)-3-(2-(2-(3-ethoxy-2-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4p):**

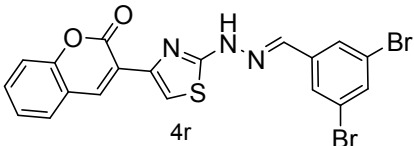
Yellow solid, yield: 93%, mp: 228-230 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.16 (brs, 1H, -NH), 9.32 (brs, 1H, -OH), 8.55 (s, 1H, Thiazole **H**), 8.38 (s, 1H, -N=C-**H**), 7.86 (dd, *J*=8.0 Hz, *J*=1.5 Hz, 1H, arom H), 7.77 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.63 (td, *J*=7.5 Hz, *J*=1.5 Hz, 1H, arom H), 7.46 (d, *J*=8.0 Hz, 1H, arom H), 7.39 (td, *J*=7.5 Hz, *J*=1.0 Hz, 1H, arom H), 7.24 (dd, *J*=8.0 Hz, *J*=1.0 Hz, 1H, arom H), 6.97 (dd, *J*=8.0 Hz, *J*=1.5 Hz, 1H, arom H), 6.82 (t, *J*=8.0 Hz, 1H, arom H), 4.07 (q, *J*=7.0 Hz, 2H, -OCH<sub>2</sub>), 1.36 (t, *J*=7.0 Hz, 3H, -CH<sub>3</sub>). HRMS (ESI): [M+H]<sup>+</sup> 408.1011.

**(E)-3-(2-(2-(3-ethoxy-4-hydroxybenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4q):**

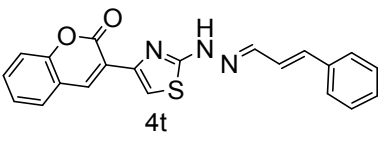
Off white solid, yield: 96%, mp: 265-267 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.03 (brs, 1H, -NH), 8.53 (s, 1H, Thiazole **H**), 7.96 (s, 1H, -N=C-**H**), 7.86 (d, *J*=9.5

 <p style="text-align: center;">4q</p>	<p>Hz, 1H, arom H), 7.75 (s, 1H, Coumarin C<sub>4</sub>-H), 7.63 (td, <i>J</i>=9.5 Hz, <i>J</i>=1.5 Hz, 1H, arom H), 7.46 (d, <i>J</i>=10.0 Hz, 1H, arom H), 7.39 (t, <i>J</i>=9.5 Hz, 1H, arom H), 7.23 (d, <i>J</i>=2.0 Hz, 1H, arom H), 7.07 (dd, <i>J</i>=10.0 Hz, <i>J</i>=2.0 Hz, 1H, arom H), 6.85 (d, <i>J</i>=10.5 Hz, 1H, arom H), 4.07 (q, <i>J</i>=9.0 Hz, 2H, -OCH<sub>2</sub>), 1.37 (t, <i>J</i>=9.0 Hz, 3H, -CH<sub>3</sub>). HRMS (ESI): Anal. Calcd. For C<sub>21</sub>H<sub>18</sub>O<sub>4</sub>N<sub>3</sub>S [M+H]<sup>+</sup>408.1013; Found: 408.1014.</p>
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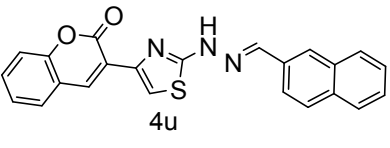
**(E)-3-(2-(2-(3,5-dibromobenzylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4r):**

 <p style="text-align: center;">4r</p>	<p>Yellow solid, yield: 90%, mp: 248-250 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 10.79 (brs, 1H, -NH), 8.55 (s, 1H, Thiazole H), 8.28 (s, 1H, -N=C-H), 7.86 (dd, <i>J</i>=9.5 Hz, <i>J</i>=1.5 Hz, 1H, arom H), 7.82 (s, 1H, Coumarin C<sub>4</sub>-H), 7.77 (s, 2H, arom H), 7.64 (td, <i>J</i>=10.5 Hz, <i>J</i>=2.0 Hz, 1H, arom H), 7.51-7.38 (m, 3H, arom H).</p>
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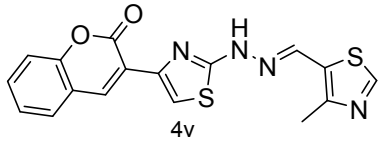
**(3-(2-((E)-2-((E)-3-phenylallylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one(4t):**

 <p style="text-align: center;">4t</p>	<p>Yellow solid, yield: 96%. mp: 245-247 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.14 (s, 1H, -NH), 8.50 (s, 1H, Thiazole H), 7.89 (d, <i>J</i>=10.0 Hz, 1H, -N=C-H), 7.82 (dd, <i>J</i>=9.5 Hz, <i>J</i>=1.5 Hz, 1H, arom H), 7.74 (s, 1H, Coumarin C<sub>4</sub>-H), 7.63-7.58 (m, 3H, arom H), 7.43 (d, <i>J</i>=10.0 Hz, 1H, arom H), 7.39-7.34 (m, 3H, arom H), 7.29 (tt, <i>J</i>=9.0 Hz, <i>J</i>=3.0 Hz, 1H, arom H), 6.98-6.95 (m, 2H). HRMS (ESI): Anal. Calcd. For C<sub>21</sub>H<sub>16</sub>O<sub>2</sub>N<sub>3</sub>S [M+H]<sup>+</sup> 374.0958; Found: 374.0965.</p>
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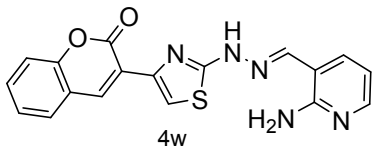
**(E)-3-(2-(2-(naphthalen-2-ylmethylene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one(4u):**

 <p style="text-align: center;">4u</p>	<p>Off-white solid, yield: 95%, mp: 254-256 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 12.33 (brs, 1H, -NH), 8.55 (s, 1H, Thiazole H), 8.23 (s, 1H, -N=C-H), 8.06 (s, 1H, arom H), 7.98-7.91 (m, 4H, arom H), 7.86 (dd, <i>J</i>=10.0 Hz, <i>J</i>=2.0 Hz, 1H, arom H), 7.81 (s, 1H, Coumarin C<sub>4</sub>-H), 7.63 (td, <i>J</i>=9.5 Hz, <i>J</i>=2.0 Hz, 1H, arom H), 7.58-7.52 (m, 2H, arom H), 7.46 (d, <i>J</i>=10.5 Hz, 1H, arom H), 7.40 (td, <i>J</i>=9.5 Hz, <i>J</i>=1.0 Hz, 1H, arom H). HRMS (ESI): [M+H]<sup>+</sup>398.0954.</p>
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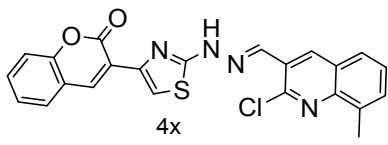
**(E)-3-(2-(2-((4-methylthiazol-5-yl)methylene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4v):**

	Yellow solid, yield: 95%. mp: 242-244 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 12.22 (s, 1H, -NH), 9.0 (s, 1H, Thiazole-H), 8.53 (s, 1H, Thiazole H), 8.29 (s, 1H, -N=C-H), 7.86 (dd, <i>J</i> =9.5 Hz, <i>J</i> =1.5 Hz, 1H, arom H), 7.77 (s, 1H, Coumarin C <sub>4</sub> -H), 7.63 (td, <i>J</i> =9.0 Hz, <i>J</i> =2.0 Hz, 1H, arom H), 7.46 (d, <i>J</i> =10.5 Hz, 1H, arom H), 7.39 (td, <i>J</i> =9.5 Hz, <i>J</i> =1.5 Hz, 1H, arom H), 2.08 (s, 3H, -CH <sub>3</sub> ). LCMS (ESI): [M+H] <sup>+</sup> 369.
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**(E)-3-(2-(2-((2-aminopyridin-3-yl)methylene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4w):**

	Light brownish yellow solid, yield: 93%. mp: 246-248 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 12.64 (s, 1H, -NH), 8.57 (s, 1H, Thiazole H), 8.34 (brs, 1H, NH <sub>2</sub> ), 8.26 (s, 1H, -N=C-H), 8.17 (d, <i>J</i> =9.0 Hz, 1H, arom H), 8.07 (dd, <i>J</i> =7.5 Hz, <i>J</i> =2.0 Hz, 1H, arom H), 7.88-7.86 (m, 2H, Coumarin C <sub>4</sub> -H & 1H, arom H), 7.65 (td, <i>J</i> =10.5 Hz, <i>J</i> =2.0 Hz, 1H, arom H), 7.46 (d, <i>J</i> =10.5 Hz, 1H, arom H), 7.40 (td, <i>J</i> =10.0 Hz, <i>J</i> =1.0 Hz, 1H, arom H), 7.02 (td, <i>J</i> =7.5Hz, <i>J</i> =1.5Hz, 1H, arom H). LCMS (ESI): [M+H] <sup>+</sup> 364.
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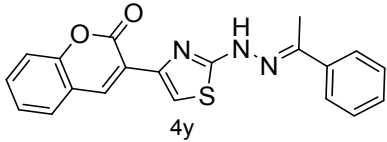
**(E)-3-(2-(2-((2-chloro-8-methylquinolin-3-yl)methylene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4x):**

	Yellow solid, yield: 92%, mp: 248-250°C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 12.64 (s, 1H, -NH), 8.79 (s, 1H, arom H), 8.56(s, 1H, Thiazole H), 8.50 (s, 1H, -N=C-H), 8.02 (d, <i>J</i> =10.5 Hz, 1H, arom H), 7.87(dd, <i>J</i> =10.0 <i>J</i> =2.0 Hz, 1H, arom H), 7.85(s, 1H, Coumarin C <sub>4</sub> -H), 7.69 (d, <i>J</i> =8.5 Hz, 1H, arom H), 7.64 (td, <i>J</i> =10.5 Hz, <i>J</i> =1.5 Hz, 1H, arom H), 7.57 (t, <i>J</i> =10.0 Hz, 1H, arom H), 7.46 (d, <i>J</i> =10.0 Hz, 1H, arom H), 7.40(td, <i>J</i> =9.5 Hz, <i>J</i> =1.0 Hz, 1H, arom H), 2.66 (s, 3H, -CH <sub>3</sub> ). LCMS (ESI): [M+H] <sup>+</sup> 447.
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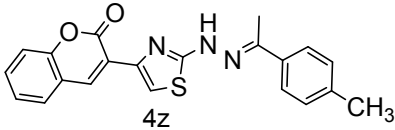
**(E)-3-(2-(2-(1-phenylethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4y):**

	Light yellow solid, yield: 97%, mp: 226-228 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 11.34 (brs, 1H, NH), 8.59 (s, 1H, Thiazole H), 7.83 (d, <i>J</i> =8.0 Hz, 1H, arom H), 7.80-7.78 (m, 3H, Coumarin C <sub>4</sub> -H & 2H, arom H), 7.64 (td, <i>J</i> =8.5 Hz, <i>J</i> =1.5 Hz, 1H, arom H), 7.47-7.38
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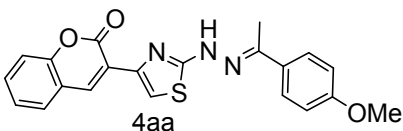


 <p style="text-align: center;">4y</p>	<p>(m, 5H, arom H), 2.34 (s, 3H, -CH<sub>3</sub>). HRMS (ESI): Anal. Calcd. For C<sub>20</sub>H<sub>16</sub>O<sub>2</sub>N<sub>3</sub>S[M+H]<sup>+</sup> 362.0958; Found: 362.0968.</p>
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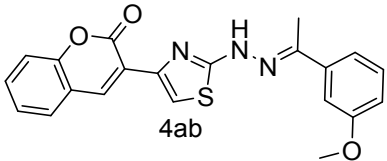
**(E)-3-(2-(2-(1-(p-tolyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4z):**

 <p style="text-align: center;">4z</p>	<p>Yellow solid, yield: 95%. mp: 218-220 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 11.27 (s, 1H, -NH), 8.58 (s, 1H, Thiazole H), 7.83 (d, J=8.0 Hz, 1H, arom H), 7.78 (s, 1H, Coumarin C<sub>4</sub>-H), 7.68 (d, J=8.5 Hz, 2H, arom H), 7.63 (td, J=7.0 Hz, J=1.5 Hz, 1H, arom H), 7.46 (d, J=8.5 Hz, 1H, arom H), 7.39 (td, J=7.5 Hz, J=1.0 Hz, 1H, arom H), 7.23 (d, J=8.0 Hz, 2H, arom H), 2.33 (s, 3H, -CH<sub>3</sub>), 2.31 (s, 3H, -CH<sub>3</sub>). HRMS (ESI): Anal. Calcd. For C<sub>21</sub>H<sub>18</sub>O<sub>2</sub>N<sub>3</sub>S [M+H]<sup>+</sup> 376.1114; Found: 376.1129</p>
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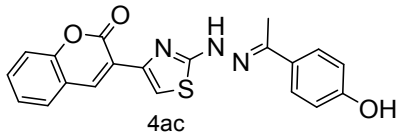
**(E)-3-(2-(2-(1-(4-methoxyphenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4aa):**

 <p style="text-align: center;">4aa</p>	<p>Pale yellow solid, yield: 96%, mp: 240-242 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 11.22 (brs, 1H, -NH), 8.56 (s, 1H, Thiazole H), 7.81 (dd, J=9.5 Hz, J=1.5 Hz, 1H, arom H), 7.75 (s, 1H, Coumarin C<sub>4</sub>-H), 7.73 (d, J=11.0 Hz, 2H, arom H), 7.62 (td, J=9.5 Hz, J=2.0 Hz, 1H, arom H), 7.44 (d, J=10.5 Hz, 1H, arom H), 7.38 (td, J=9.5 Hz, J=1.0 Hz, 1H, arom H), 6.97 (d, J=11.5 Hz, 1H, arom H), 3.78 (s, 3H, -OCH<sub>3</sub>), 2.3 (s, 3H, -CH<sub>3</sub>). HRMS (ESI): Anal. Calcd. For C<sub>21</sub>H<sub>18</sub>O<sub>3</sub>N<sub>3</sub>S [M+H]<sup>+</sup> 392.1063; Found: 392.1077</p>
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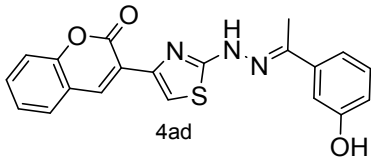
**(E)-3-(2-(2-(1-(3-methoxyphenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4ab):**

 <p style="text-align: center;">4ab</p>	<p>Yellow solid, yield: 95%, mp: 240-242 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 11.35 (brs, 1H, -NH), 8.57 (s, 1H, Thiazole H), 7.82 (dd, J=7.5 Hz, J=1.0 Hz, 1H, arom H), 7.78 (s, 1H, Coumarin C<sub>4</sub>-H), 7.63 (td, J=8.5 Hz, J=1.5 Hz, 1H, arom H), 7.45 (d, J=8.0 Hz, 1H, arom H), 7.39 (t, J=7.5 Hz, 1H, arom H), 7.35-7.32 (m, 3H, arom H), 6.96 (dt, J=7.0 Hz, J=2.0 Hz, 1H, arom H), 3.80 (s, 3H, -OCH<sub>3</sub>), 2.32 (s, 3H, -CH<sub>3</sub>). LCMS (ESI): [M+H]<sup>+</sup> 392.</p>
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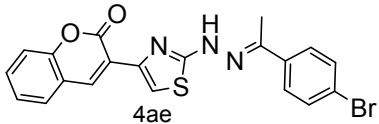
**(E)-3-(2-(2-(1-(4-hydroxyphenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4ac):**

	Brownish yellow solid, yield: 96%, mp: 248-250 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 11.14 (brs, 1H, -NH), 8.58 (s, 1H, Thiazole H), 7.82 (dd, <i>J</i> =7.5 Hz, <i>J</i> =1.0 Hz 1H, arom H), 7.76 (s, 1H, Coumarin C <sub>4</sub> -H), 7.65-7.61 (m, 3H, arom H), 7.46 (d, <i>J</i> =8.5 Hz, 1H, arom H), 7.39 (td, <i>J</i> =8.0 Hz, <i>J</i> =1.0 Hz, 1H, arom H), 6.80 (d, <i>J</i> =9.0 Hz, <i>J</i> =3.0 Hz, 2H, arom H), 2.27 (s, 3H, -CH <sub>3</sub> ). HRMS (ESI): Anal. Calcd. For C <sub>20</sub> H <sub>16</sub> O <sub>3</sub> N <sub>3</sub> S [M+H] <sup>+</sup> 378.0907; Found: 378.0919.
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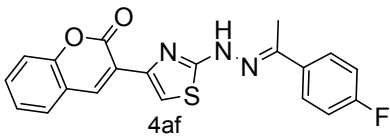
**(E)-3-(2-(2-(1-(3-hydroxyphenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4ad):**

	Yellow solid, yield: 94%, mp: 248-250 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 11.29 (brs, 1H, -NH), 8.58 (s, 1H, Thiazole H), 7.83 (dd, <i>J</i> =7.5 Hz, <i>J</i> =1.5 Hz, 1H, arom H), 7.79 (s, 1H, Coumarin C <sub>4</sub> -H), 7.63 (td, <i>J</i> =7.0 Hz, <i>J</i> =1.5 Hz, 1H, arom H), 7.46 (d, <i>J</i> =8.5 Hz, 1H, arom H), 7.40 (td, <i>J</i> =7.5 Hz, <i>J</i> =1.0 Hz, 1H, arom H), 7.23-7.18(m, 3H, arom H), 6.78 (dt, <i>J</i> =6.5 Hz, <i>J</i> =2.5 Hz, 1H, arom H), 2.29 (s, 3H, -CH <sub>3</sub> ). HRMS (ESI): Anal. Calcd. For C <sub>20</sub> H <sub>16</sub> O <sub>3</sub> N <sub>3</sub> S [M+H] <sup>+</sup> 378.0907; Found: 378.0920.
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**(E)-3-(2-(2-(1-(4-bromophenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4ae):**

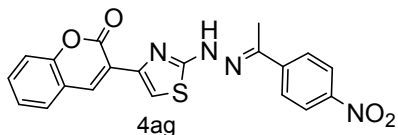
	Yellow solid, yield: 93%, mp: 240-242 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 11.45 (brs, 1H, -NH), 8.33 (s, 1H, Thiazole H), 7.84-7.81 (m, 1H, arom H), 7.74-7.65 (m, 4H, Coumarin C <sub>4</sub> -H & 3H, arom H), 7.62 (d, <i>J</i> =10.5 Hz, 2H, arom H), 7.47 (d, <i>J</i> =10.0 Hz, 1H, arom H), 7.41 (t, <i>J</i> =9.5 Hz, 1H, arom H), 2.29 (s, 3H, -CH <sub>3</sub> ). LCMS (ESI): [M+H] <sup>+</sup> 441.
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**(E)-3-(2-(2-(1-(4-fluorophenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4af):**

	Light yellow solid, yield: 95%, mp: 234-236 °C. <sup>1</sup> H NMR (500MHz, d <sub>6</sub> -DMSO): δ 11.32 (brs, 1H, -NH), 8.58 (s, 1H, Thiazole H), 7.85-7.81 (m, 3H, arom H), 7.78 (s, 1H, Coumarin C <sub>4</sub> -H), 7.63 (td, <i>J</i> =9.5 Hz, <i>J</i> =2.0 Hz, 1H, arom H), 7.46 (d, <i>J</i> =10.0 Hz, 1H, arom H), 7.39 (td, <i>J</i> =9.5 Hz, <i>J</i> =1.0 Hz, 1H, arom H), 7.26 (t, <i>J</i> =11.0 Hz, 2H, arom H), 2.33 (s, 3H, -CH <sub>3</sub> ). LCMS
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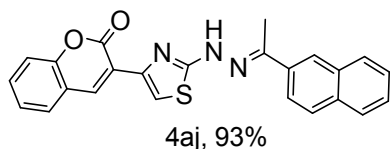
(ESI): [M+H]<sup>+</sup> 380.

**(E)-3-(2-(2-(1-(4-nitrophenyl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4ag):**



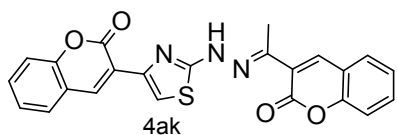
Orange yellow solid, yield: 94%, mp: 260-262 °C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 11.68 (s, 1H, -NH), 8.59 (s, 1H, Thiazole **H**), 8.28 (d, *J*=9.0 Hz, 2H, arom H), 8.03(d, *J*=9.0 Hz, 2H, arom H), 7.85-7.82(m, 2H, Coumarin C<sub>4</sub>-**H** & 1H, arom H), 7.64 (td, *J*=9.0 Hz, *J*=1.5 Hz, 1H, arom H), 7.47 (d, *J*=8.5 Hz, 1H, arom H), 7.40 (td, *J*=8.5 Hz, *J*=1.0 Hz, 1H, arom H), 2.40 (s, 3H, -CH<sub>3</sub>). LCMS (ESI): [M+H]<sup>+</sup> 407.

**(E)-3-(2-(2-(1-(naphthalen-2-yl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4aj):**



Brownish Yellow solid, yield: 93%, mp: 254-256°C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 11.64 (brs, 1H, -NH), 8.61 (s, 1H, Thiazole **H**), 8.23 (s, 1H, arom H), 8.10 (dd, *J*=10.5 Hz, *J*=2.0 Hz, 1H, arom H), 7.99 (d, *J*=11.5 Hz, 1H, arom H), 7.93 (d, *J*=10.5 Hz, 1H, arom H), 7.82 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.63 (d, *J*=10.5 Hz, 1H, arom H), 7.54 (t, *J*=5.0 Hz, 1H, arom H), 7.47 (d, *J*=11.5 Hz, 2H, arom H), 7.43-7.36 (m, 3H, arom H), 2.47 (s, 3H, -CH<sub>3</sub>).

**(E)-3-(2-(2-(1-(2-oxo-4a,8a-dihydro-2H-chromen-3-yl)ethylidene)hydrazinyl)thiazol-4-yl)-2H-chromen-2-one (4ak):**



Yellow solid, yield: 91%, mp: 252-254°C. <sup>1</sup>H NMR (500MHz, d<sub>6</sub>-DMSO): δ 11.47 (s, 1H, -NH), 8.60 (s, 1H, Thiazole **H**), 8.19 (s, 1H, Coumarin **H**), 7.88 (dd, *J*=8.0 Hz, *J*=1.5 Hz, 1H, arom H), 7.83 (dd, *J*=7.5Hz, *J*=1.0 Hz, 1H, arom H), 7.80 (s, 1H, Coumarin C<sub>4</sub>-**H**), 7.67-7.62 (m, 2H, arom H), 7.46 (t, *J*=9.0 Hz, 2H, arom H), 7.42-7.37 (m, 2H, arom H), 2.29 (s, 3H, -CH<sub>3</sub>). HRMS (ESI): Anal. Calcd. For C<sub>23</sub>H<sub>16</sub>O<sub>4</sub>N<sub>3</sub>S [M-H]<sup>+</sup> 430.0856; Found: 430.0872.

Copies of <sup>1</sup>H NMR spectra of synthesized compounds (4a-4ak)

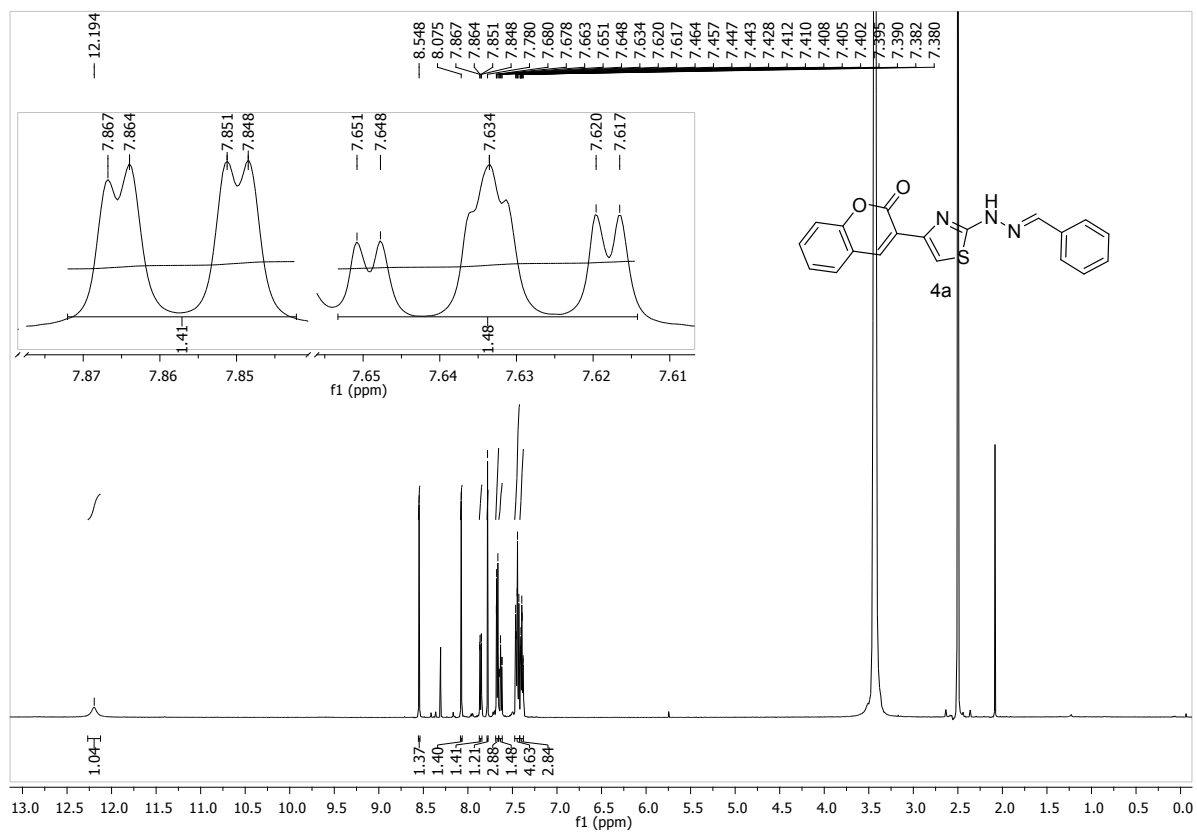


Figure S1-1. <sup>1</sup>H NMR Spectrum of 4a

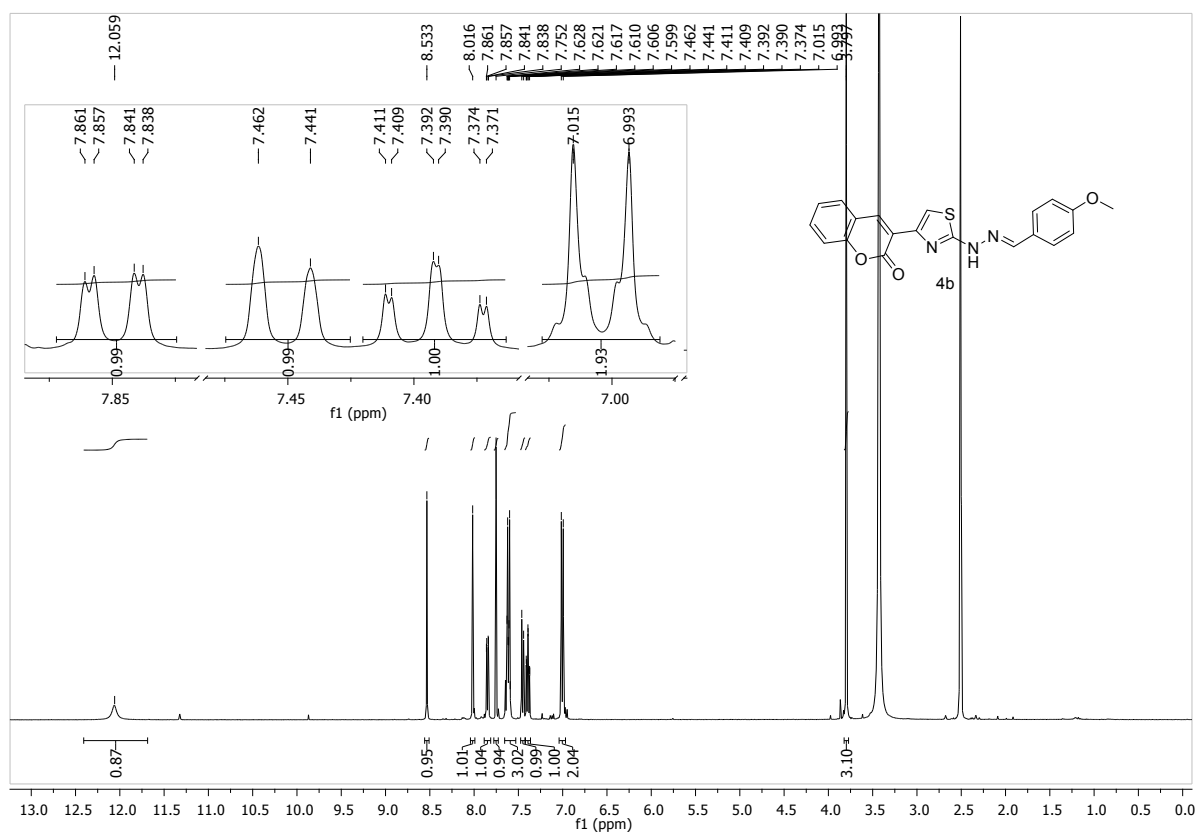


Figure S1-2. <sup>1</sup>H NMR Spectrum of 4b

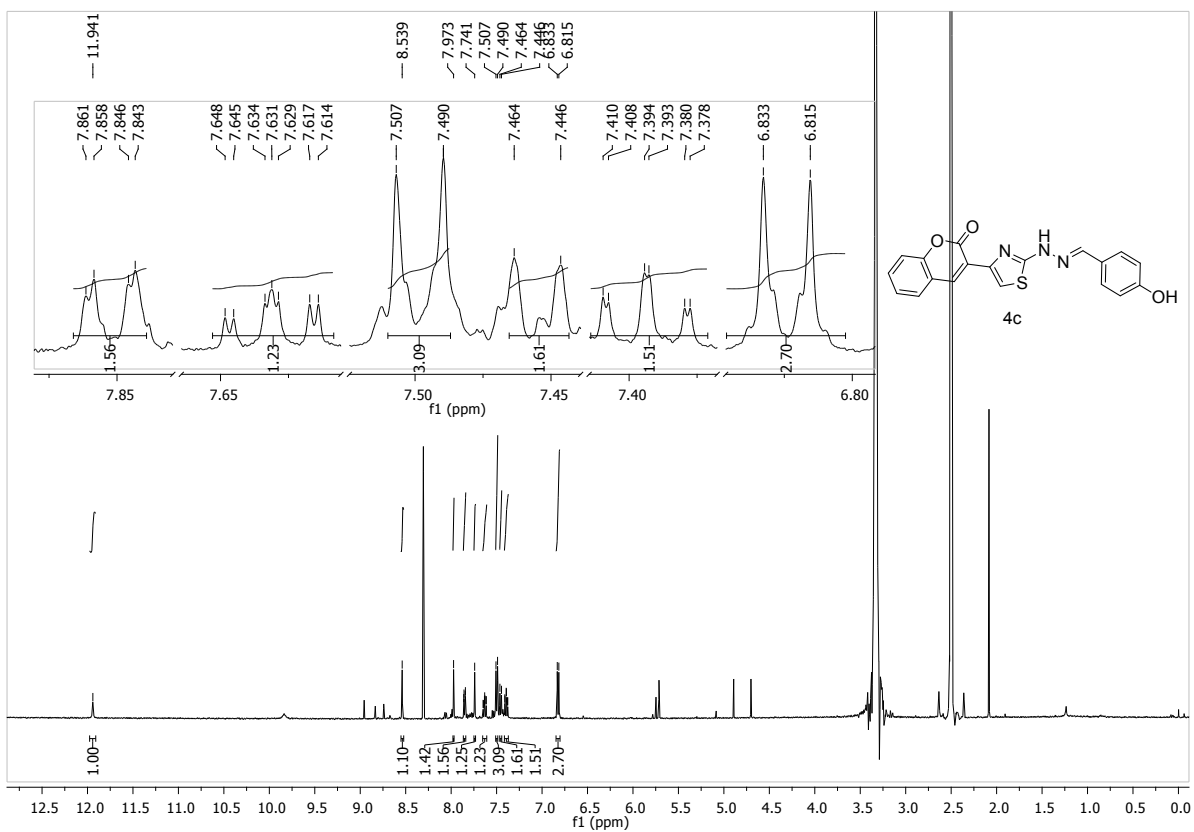


Figure S1-3. <sup>1</sup>H NMR Spectrum of 4c

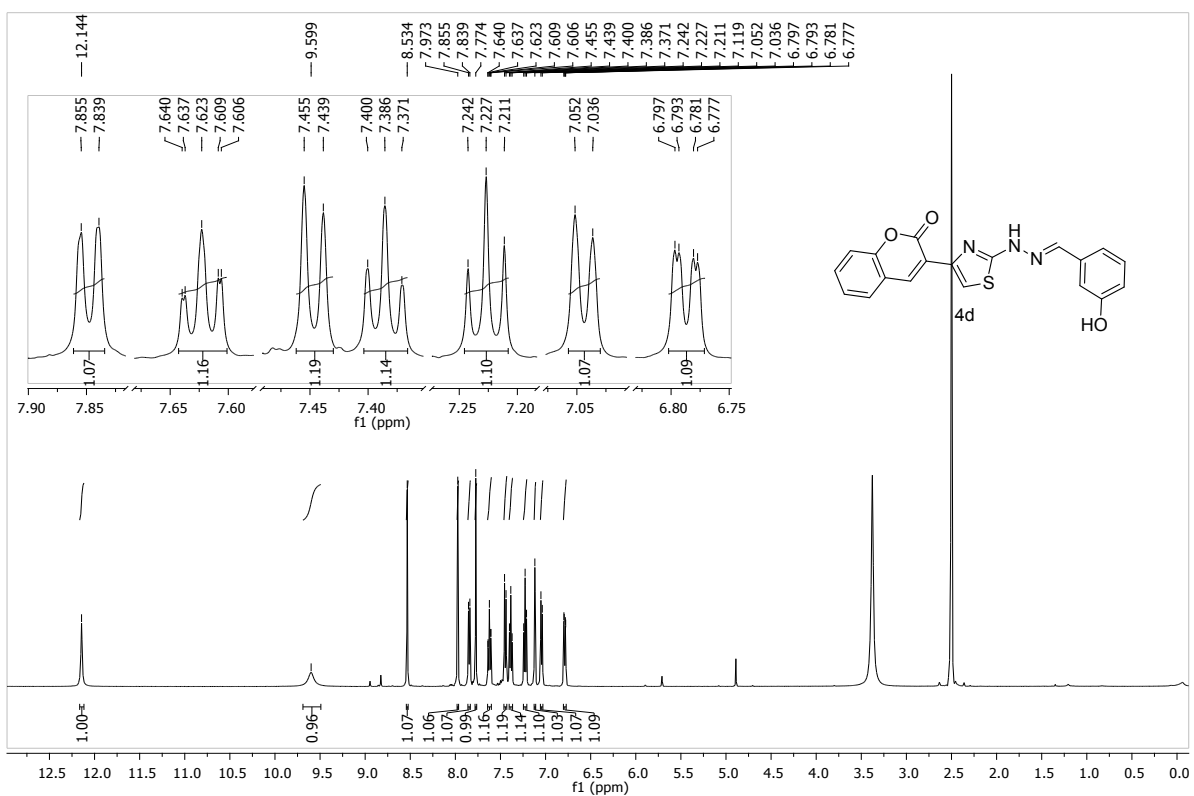


Figure S1-4. <sup>1</sup>H NMR Spectrum of 4d

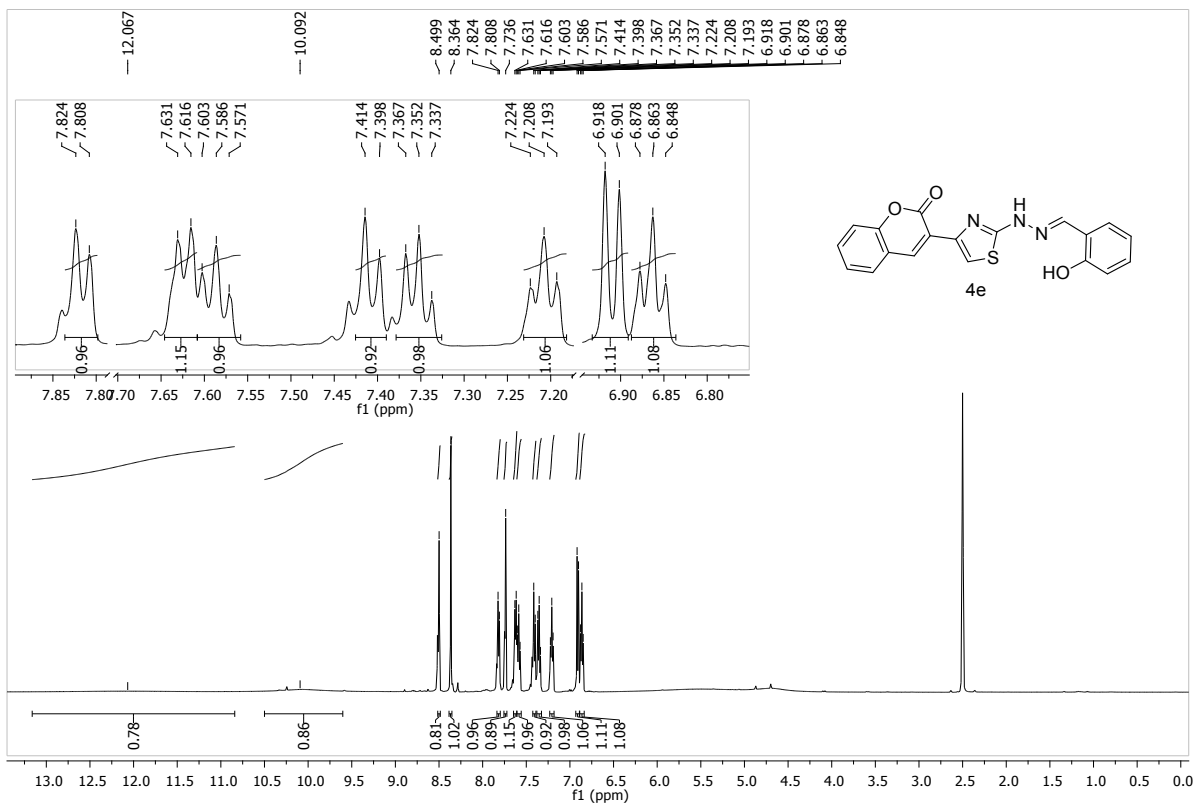


Figure S1-5. <sup>1</sup>H NMR Spectrum of 4e

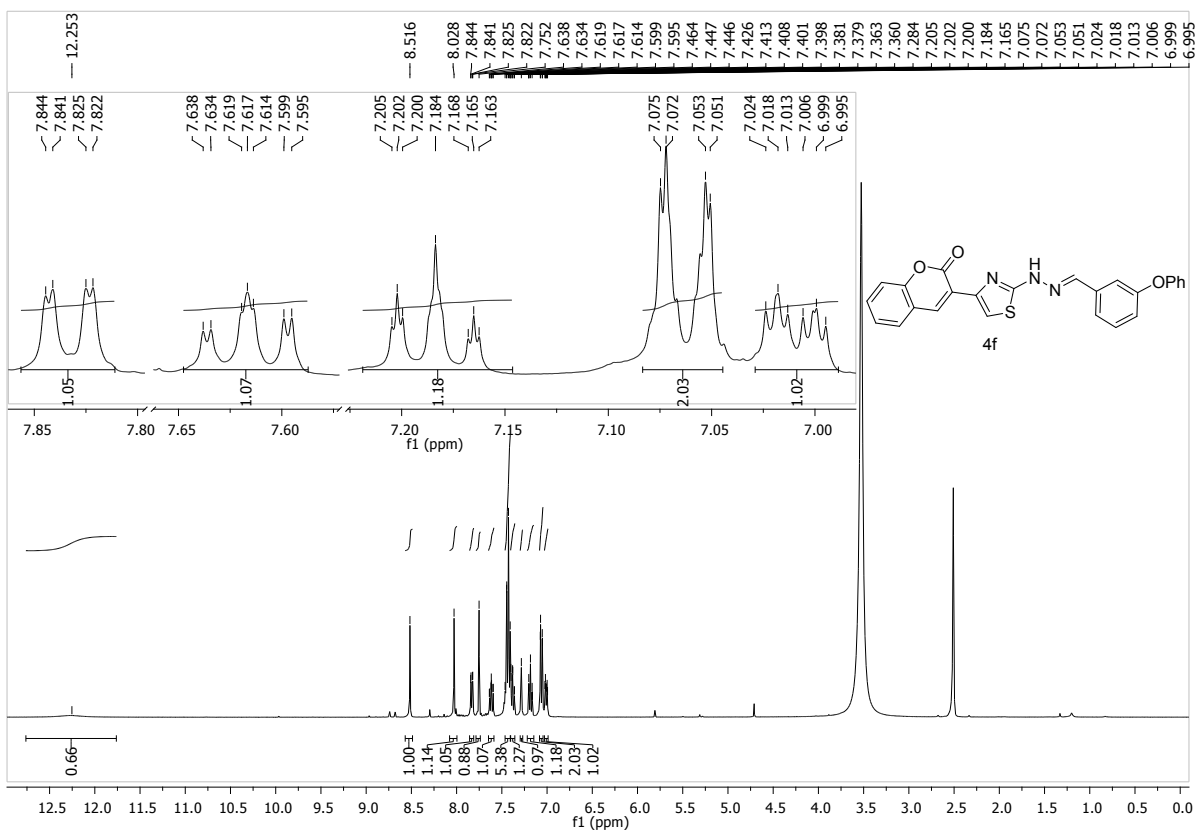


Figure S1-6. <sup>1</sup>H NMR Spectrum of 4f

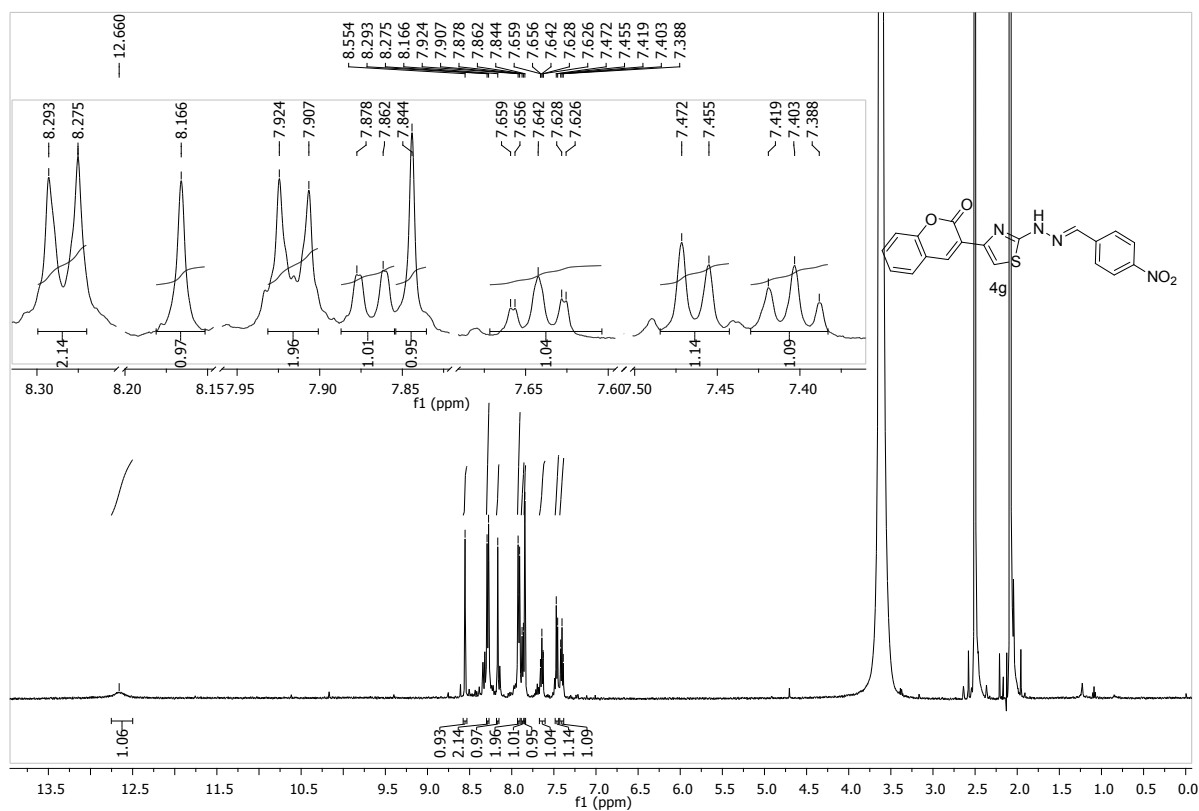


Figure S1-7. <sup>1</sup>H NMR Spectrum of 4g

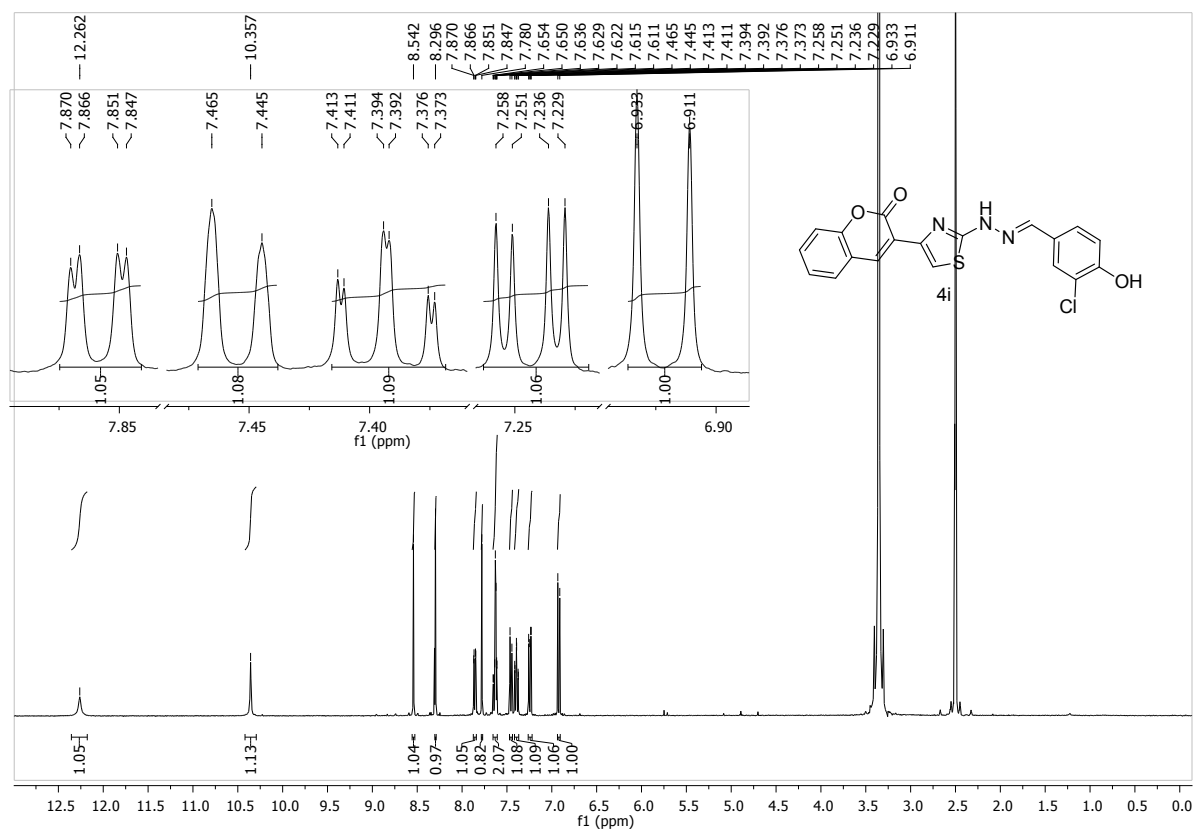


Figure S1-8. <sup>1</sup>H NMR Spectrum of 4i

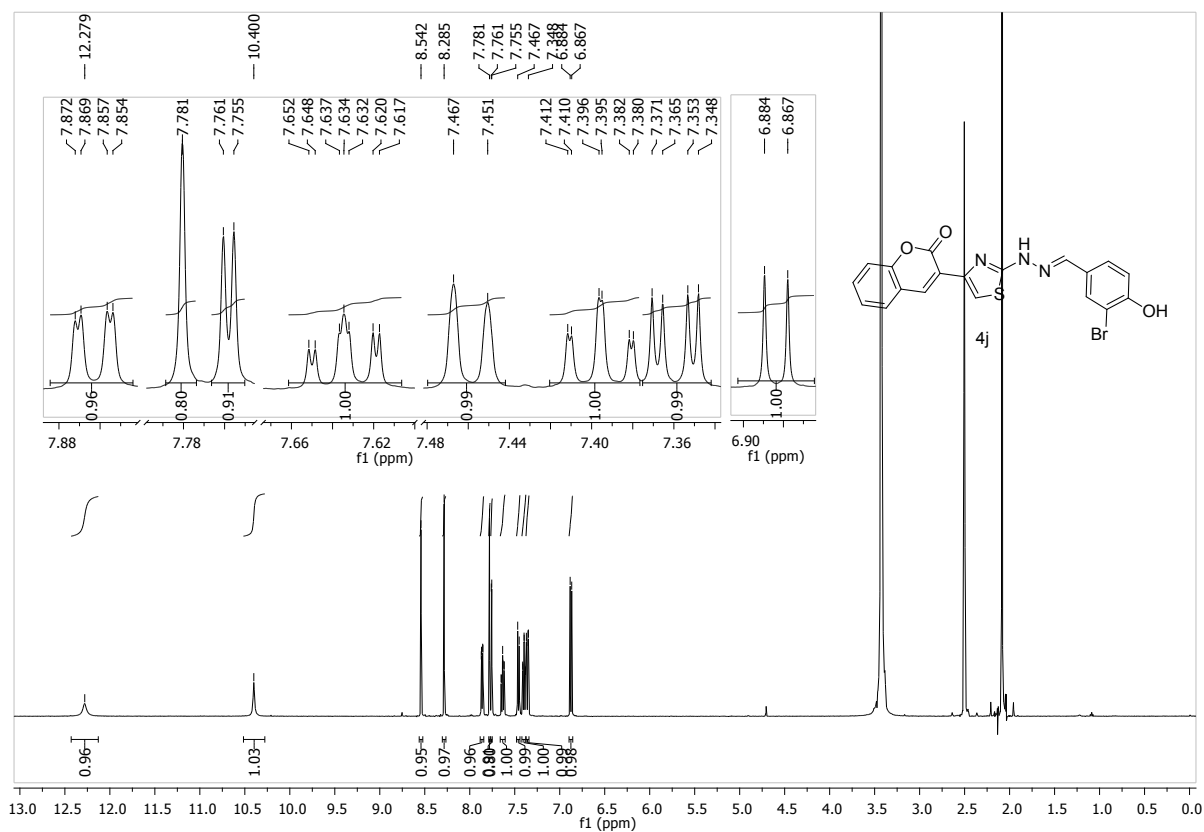


Figure S1-9. <sup>1</sup>H NMR Spectrum of 4j

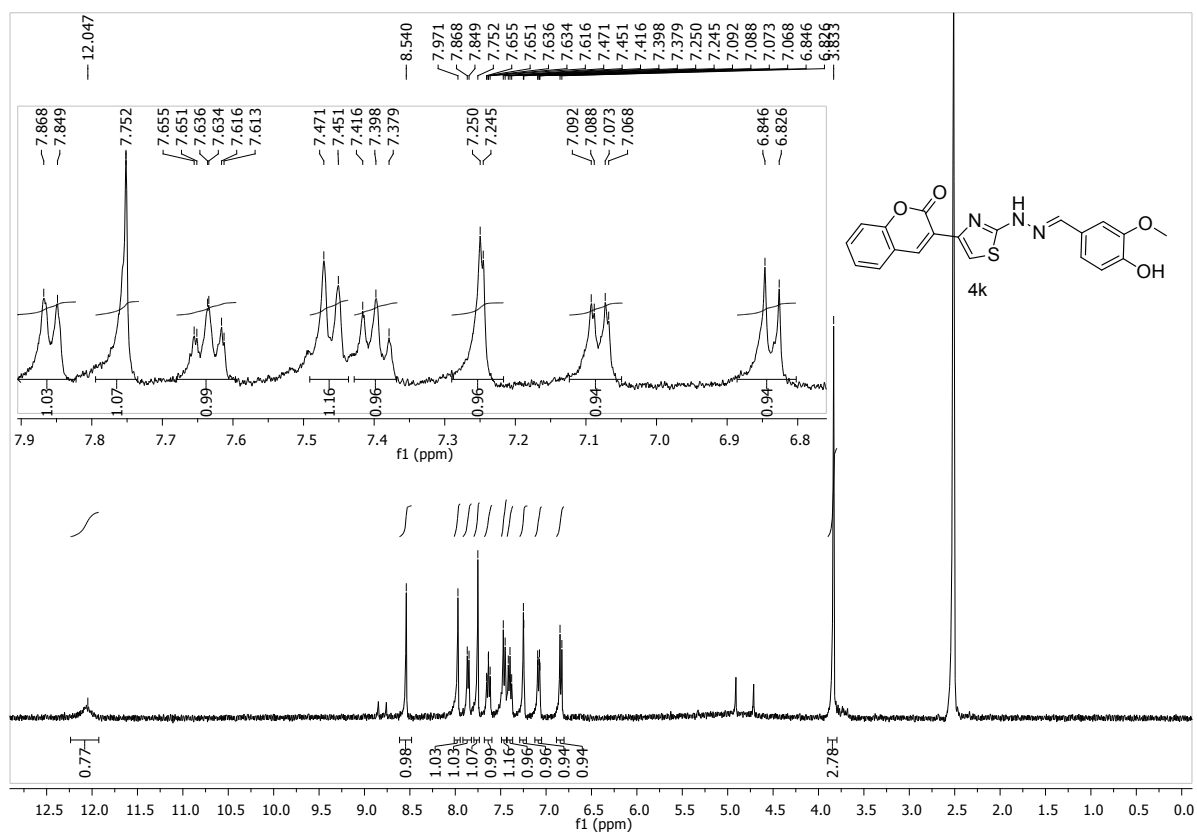


Figure S1-10. <sup>1</sup>H NMR Spectrum of 4k



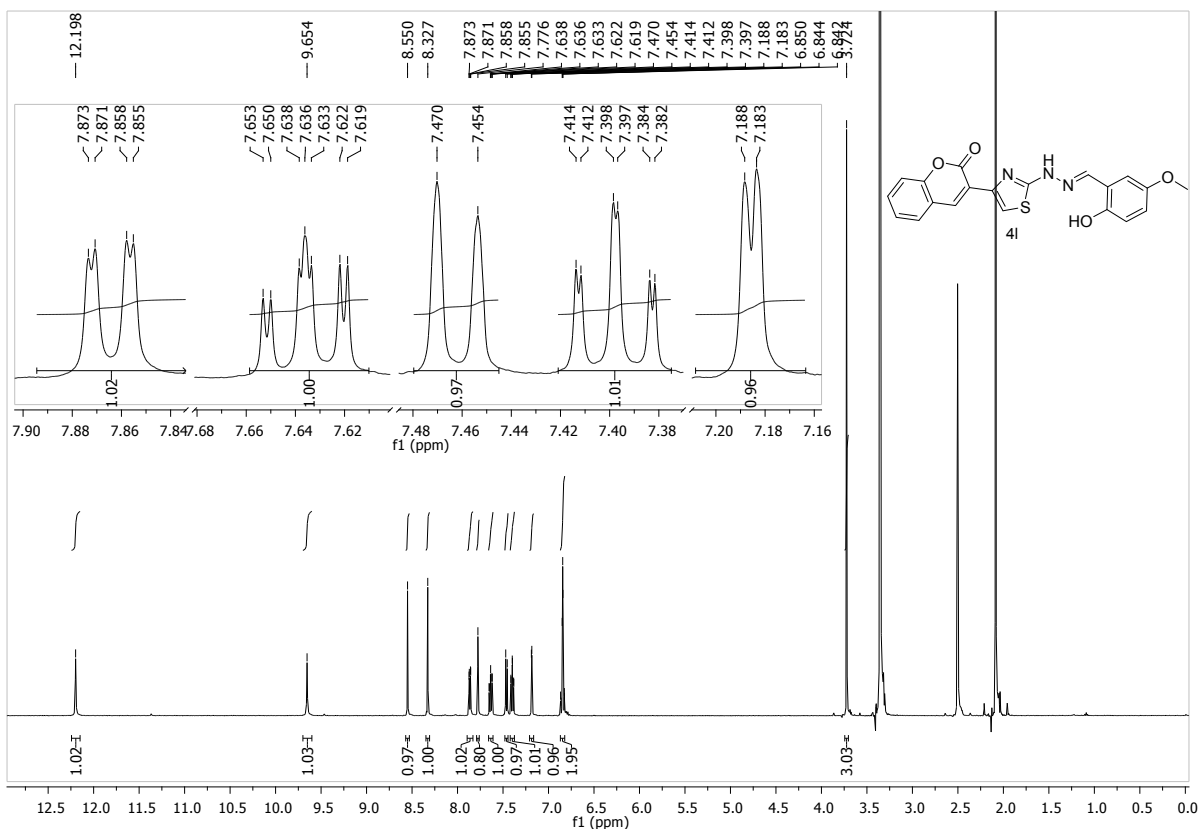


Figure S1-11. <sup>1</sup>H NMR Spectrum of 4l

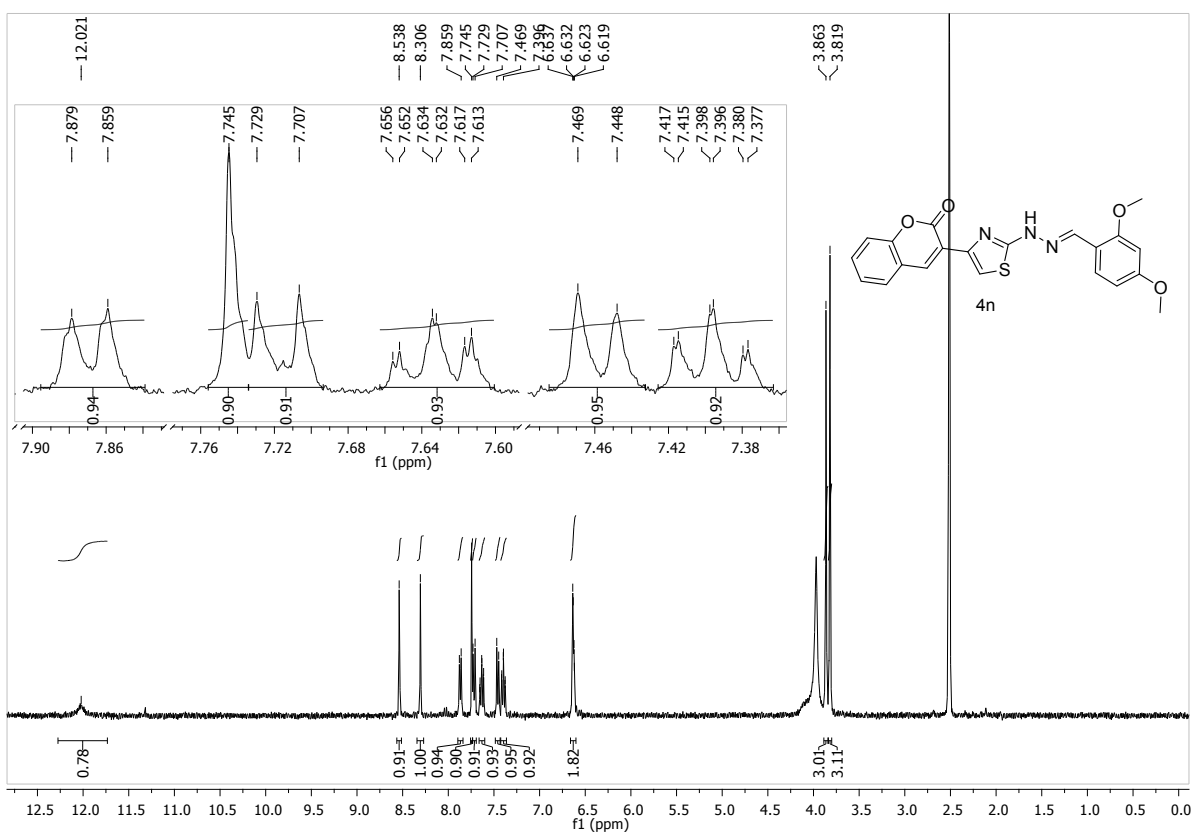


Figure S1-12. <sup>1</sup>H NMR Spectrum of Compound 4n

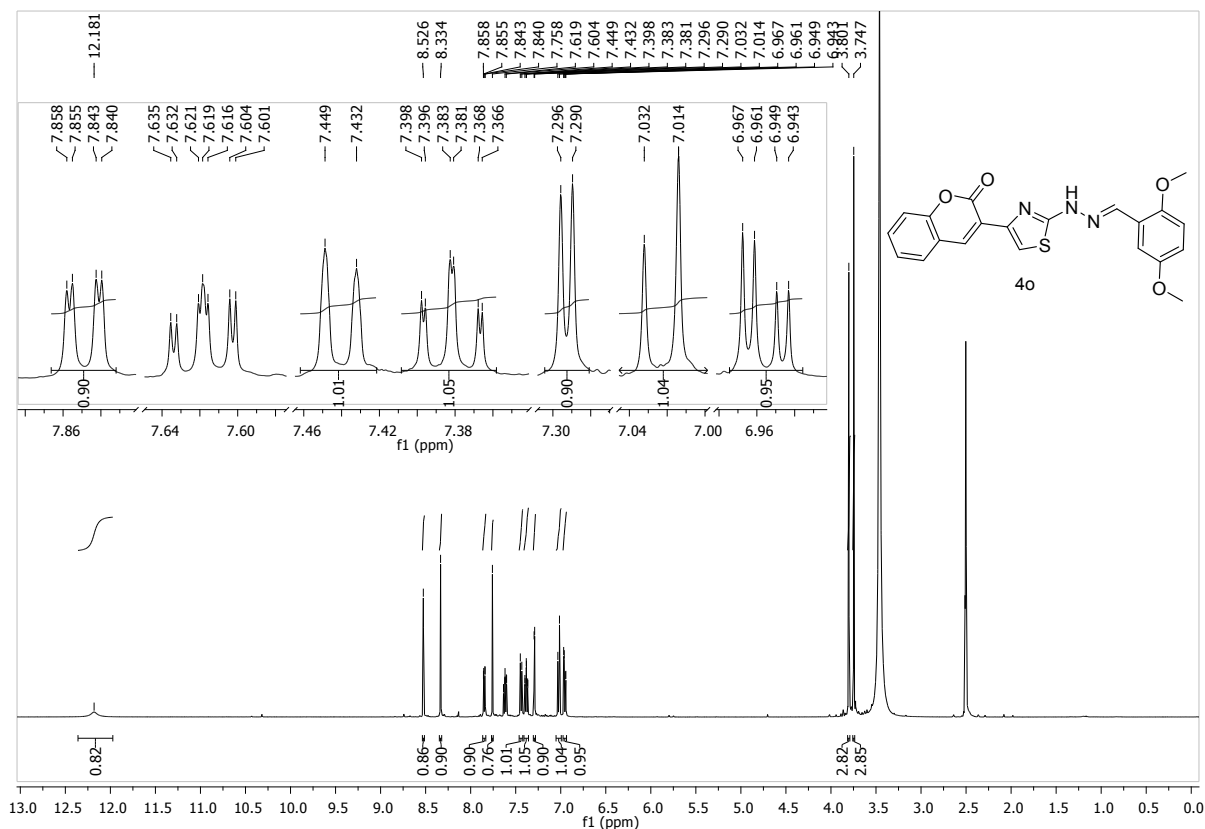


Figure S1-13. <sup>1</sup>H NMR Spectrum of 4o

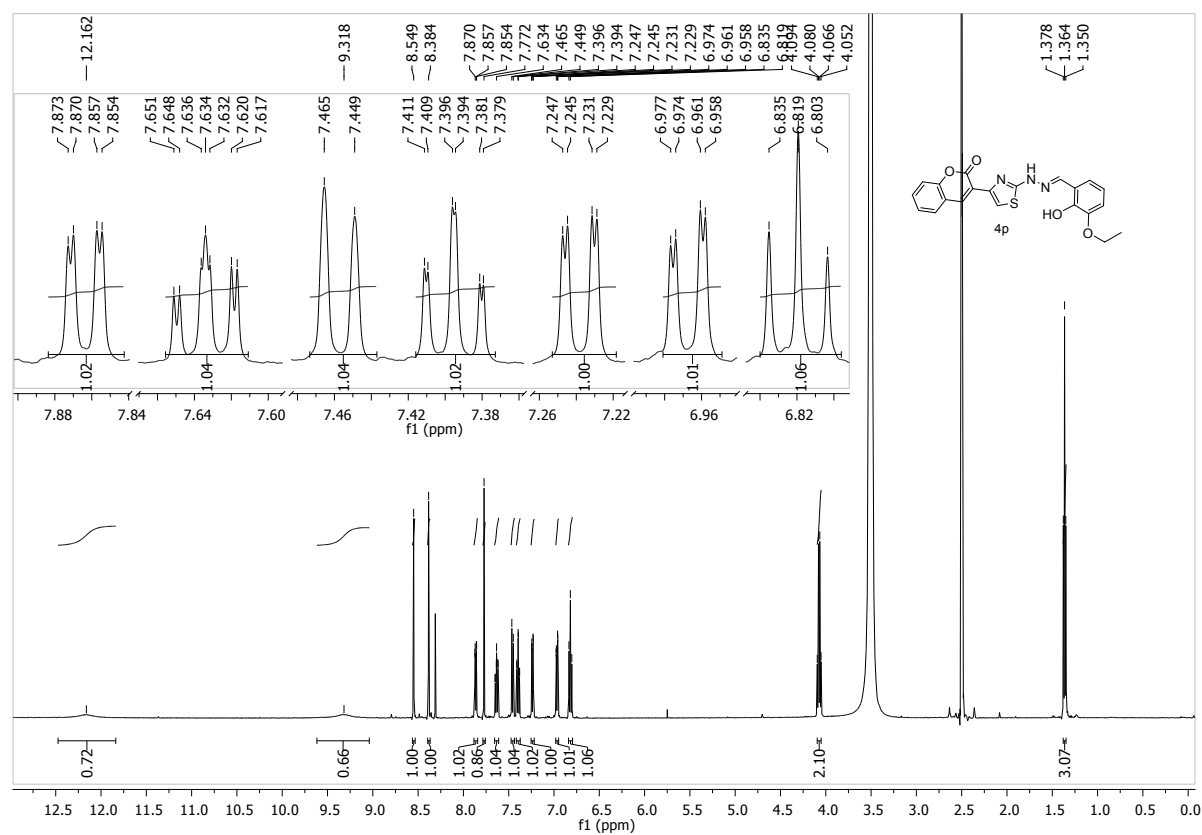


Figure S1-14. <sup>1</sup>H NMR Spectrum of 4p

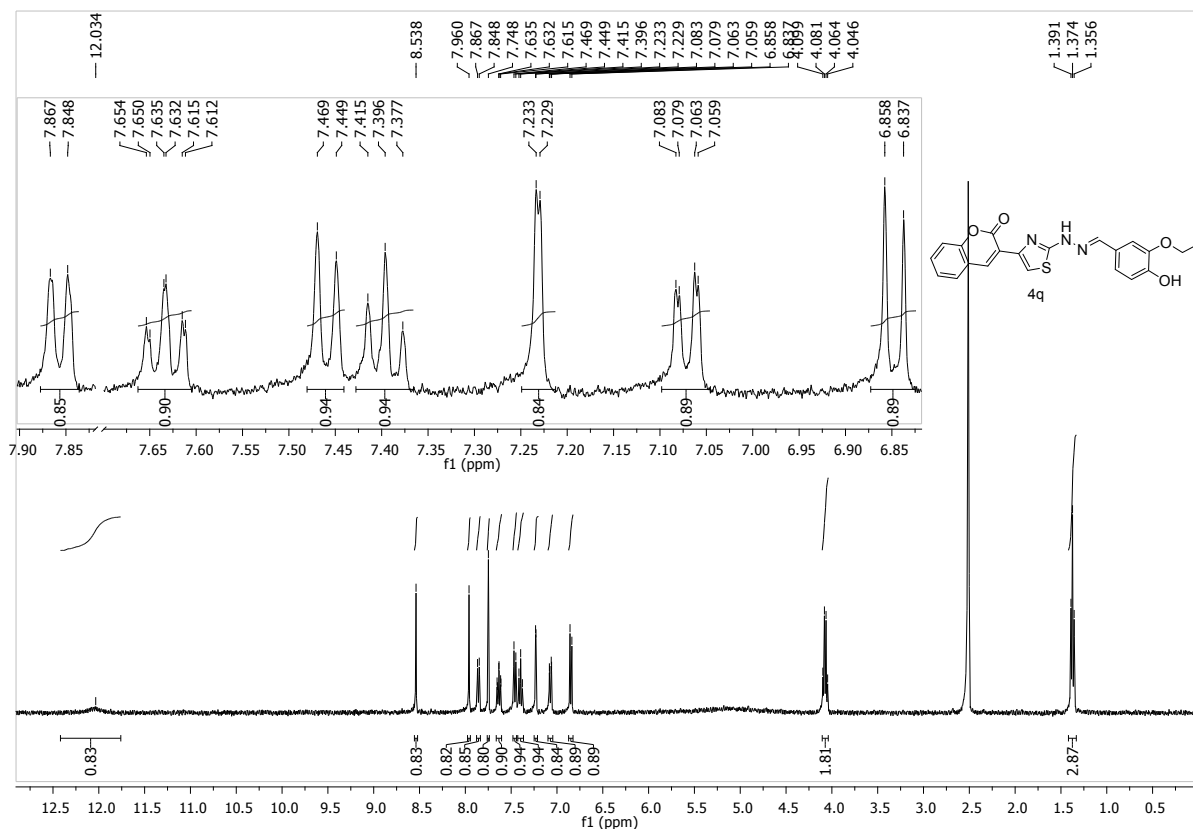


Figure S1-15. <sup>1</sup>H NMR Spectrum of Compound 4q

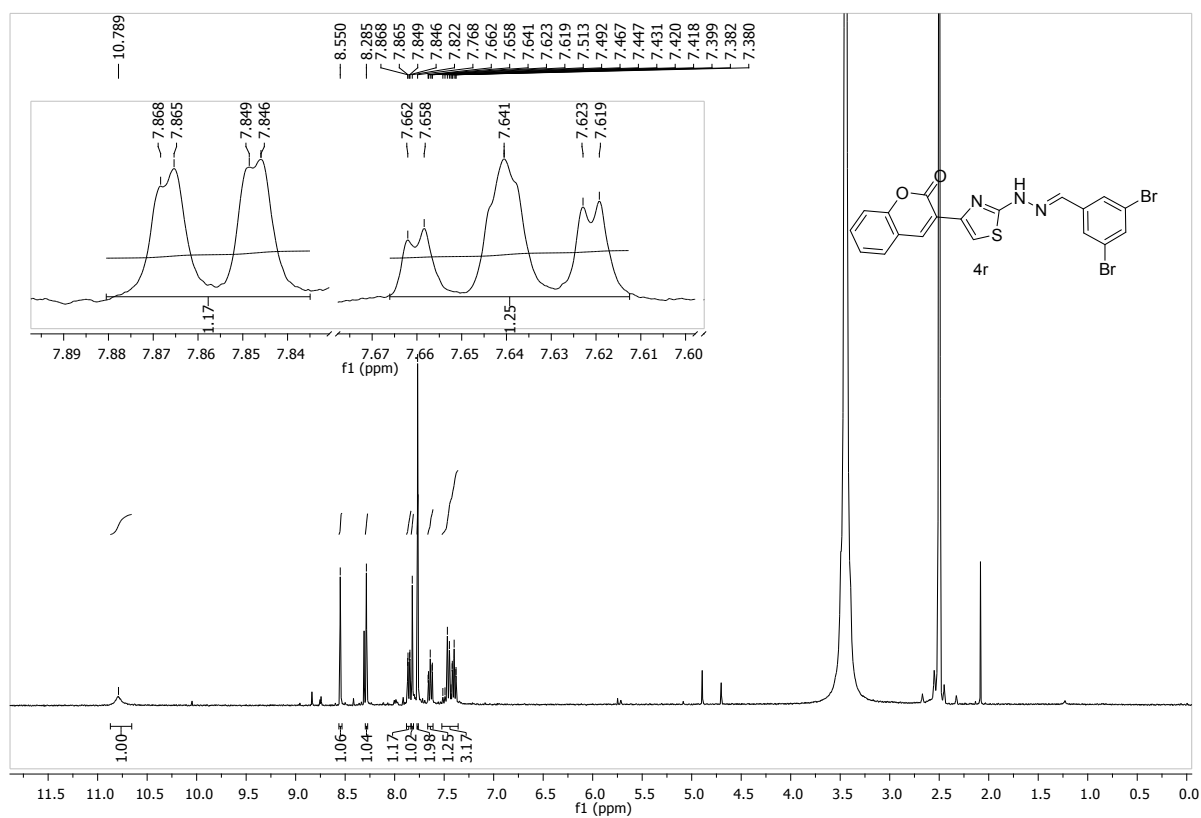


Figure S1-16. <sup>1</sup>H NMR Spectrum of 4r

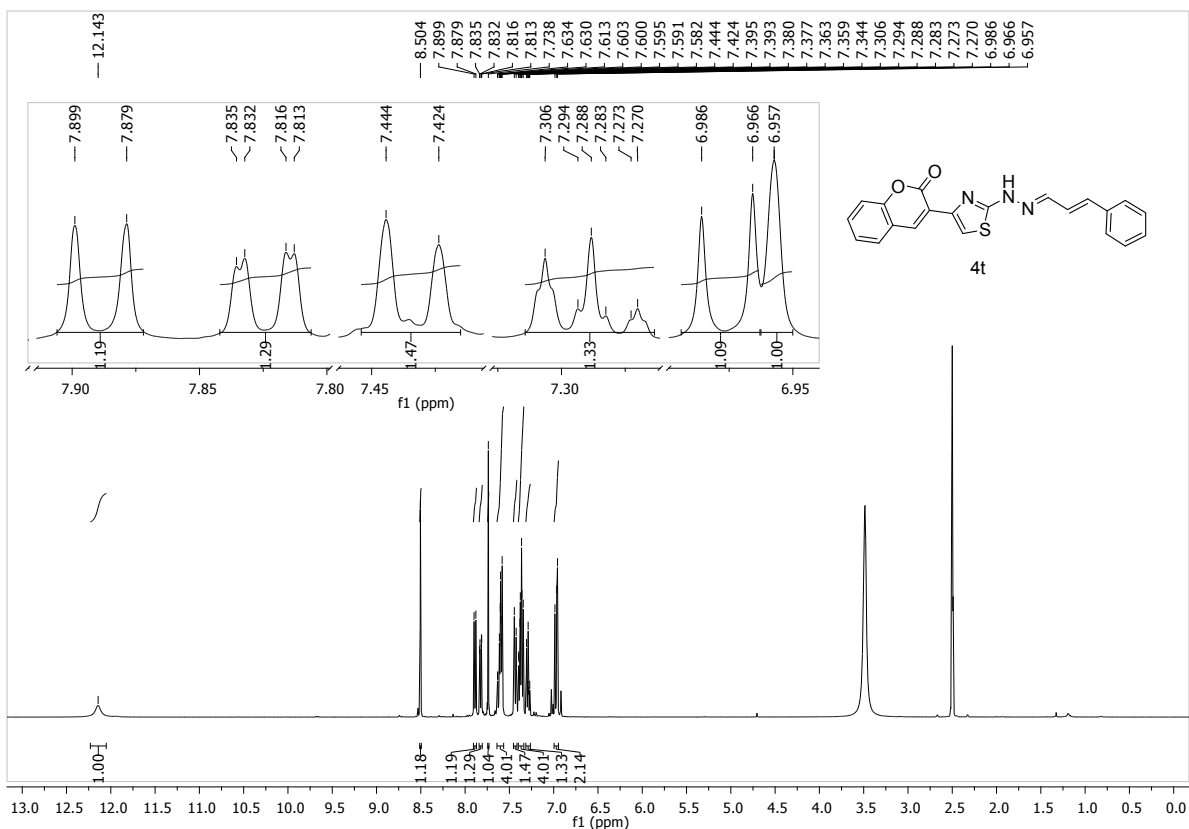


Figure S1-17. <sup>1</sup>H NMR Spectrum of 4t

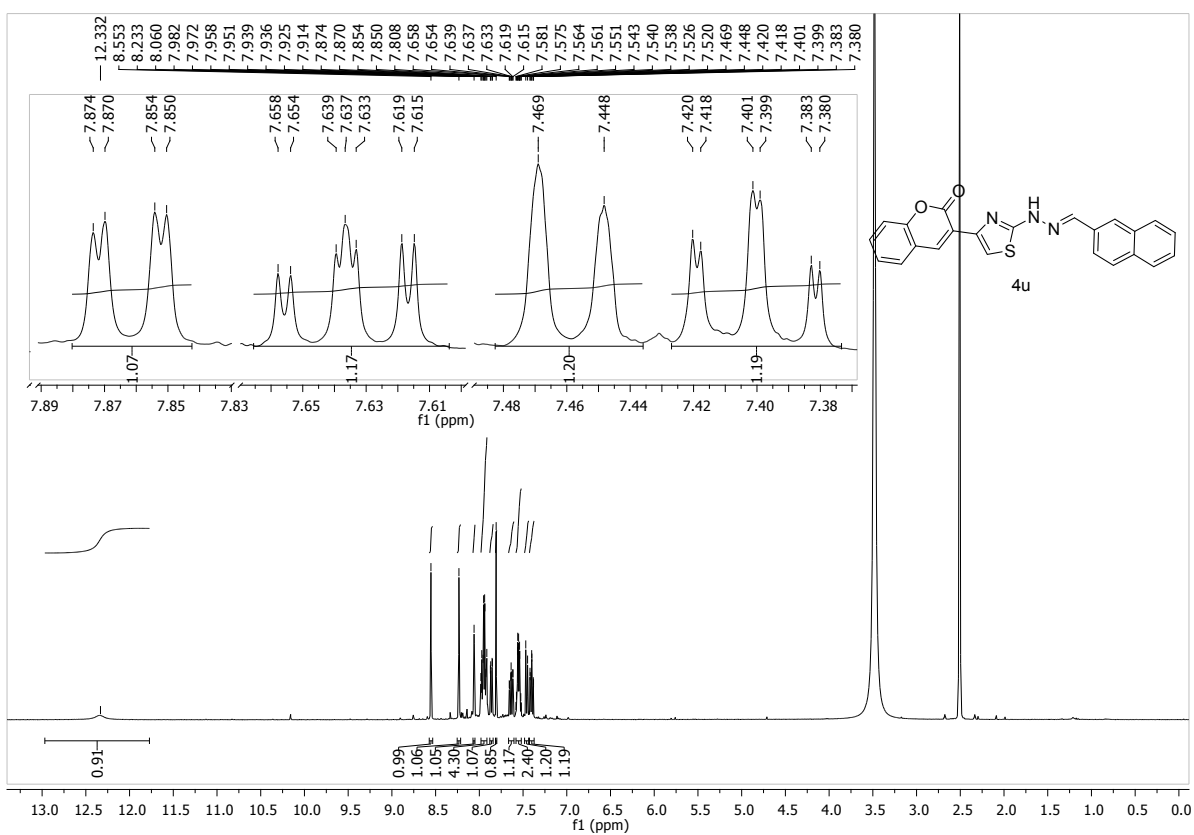


Figure S1-18. <sup>1</sup>H NMR Spectrum of 4u

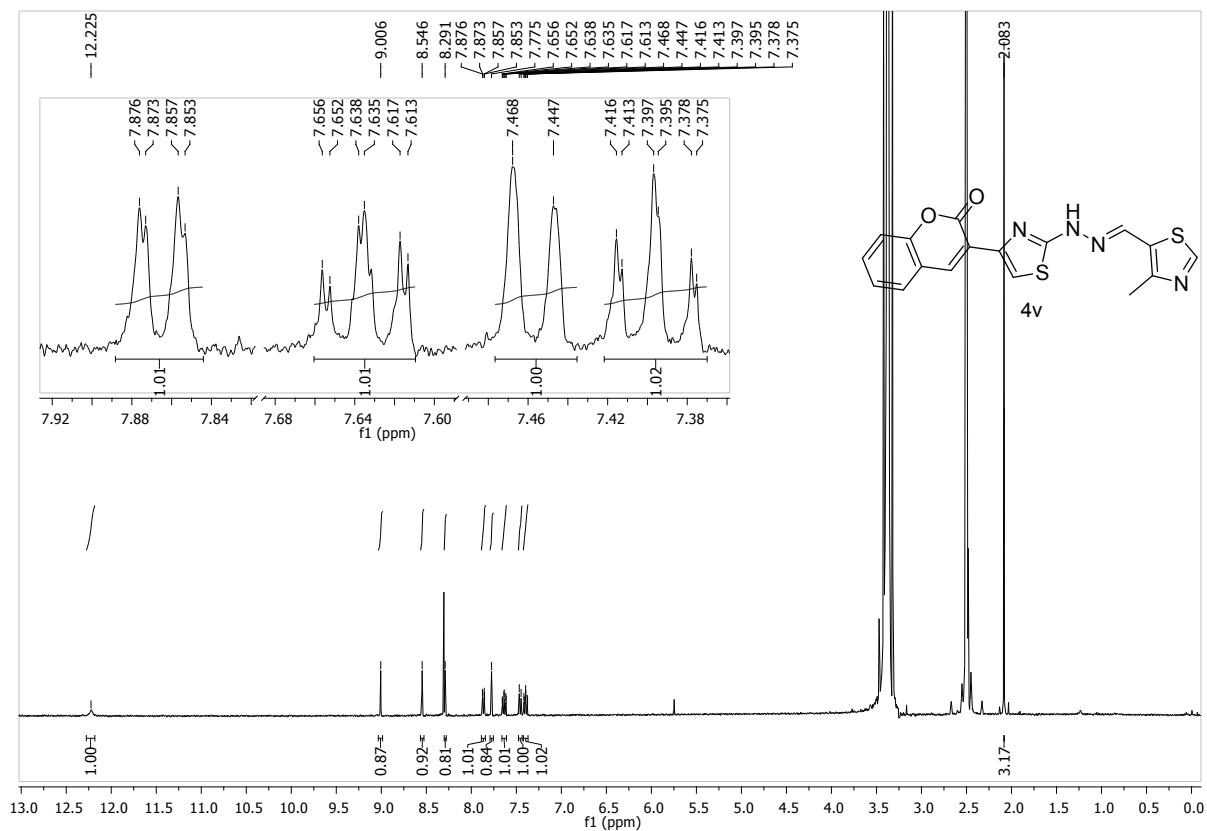


Figure S1-19. <sup>1</sup>H NMR Spectrum of 4v

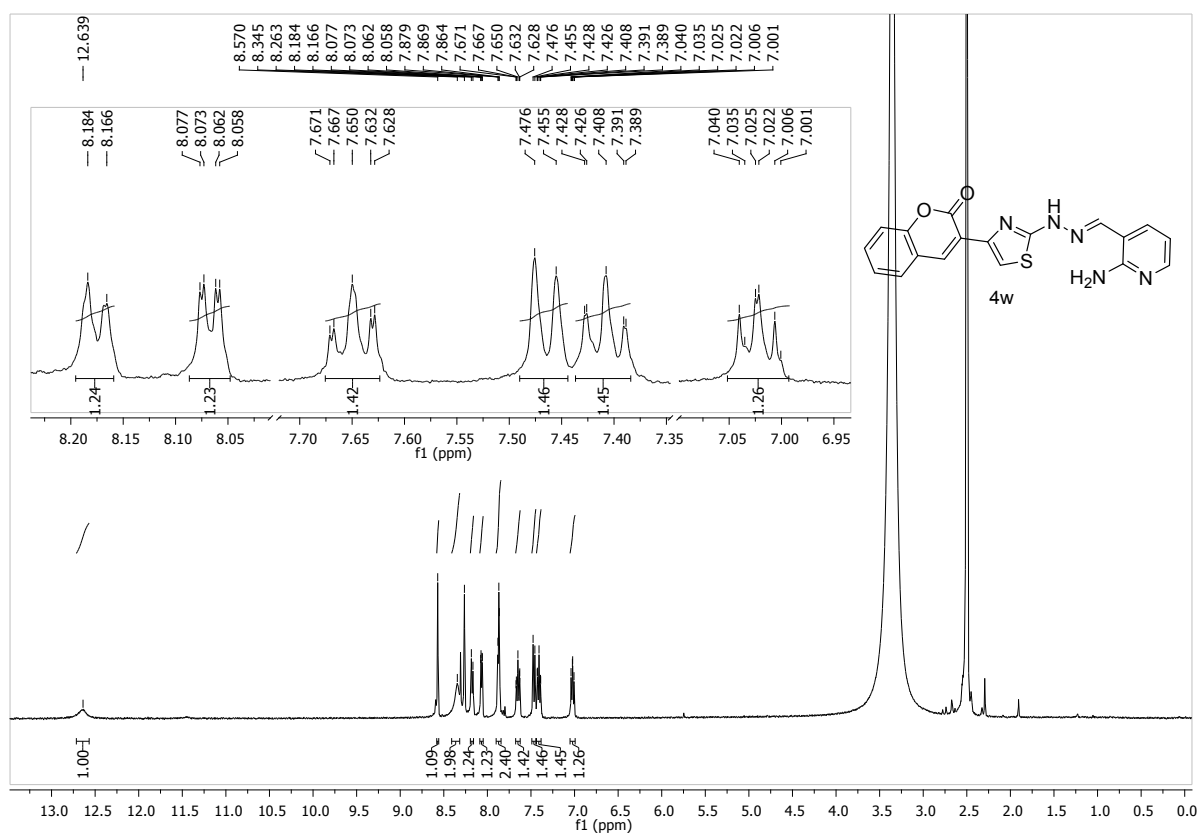


Figure S1-20. <sup>1</sup>H NMR Spectrum of 4w

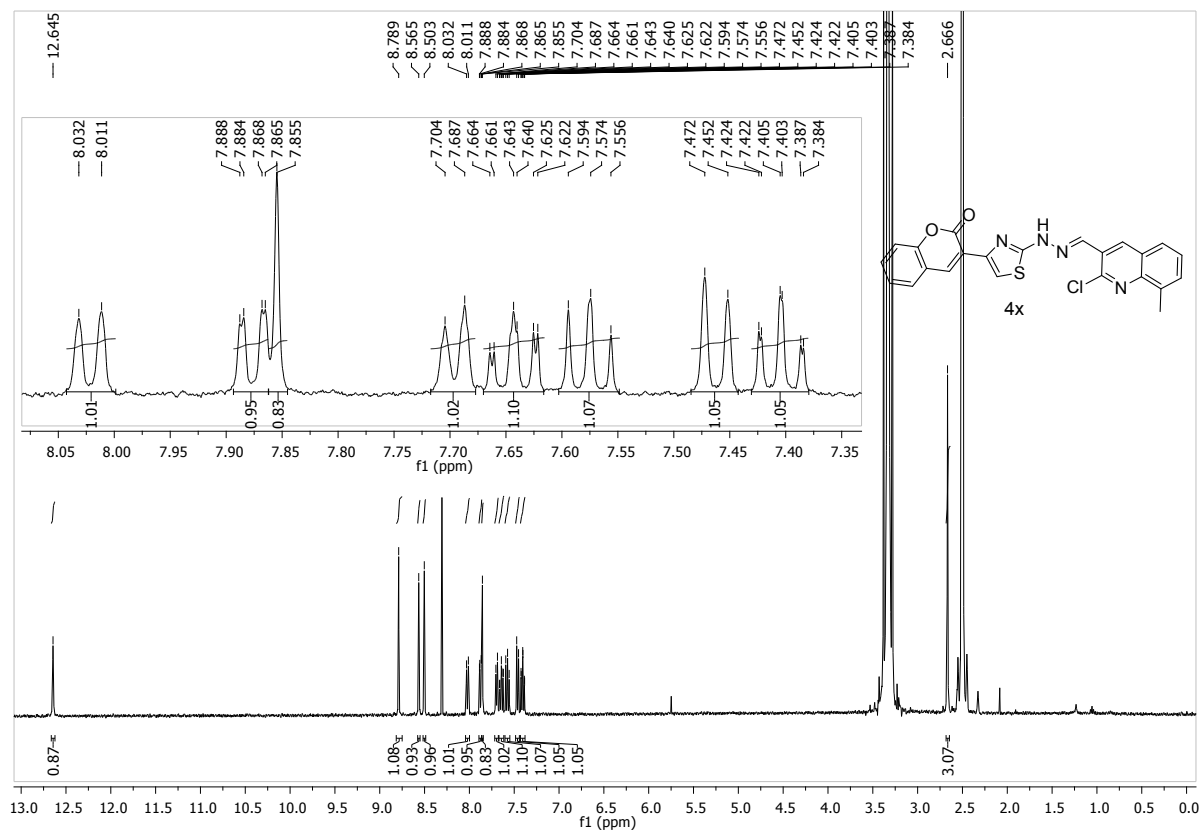


Figure S1-21. <sup>1</sup>H NMR Spectrum of 4x

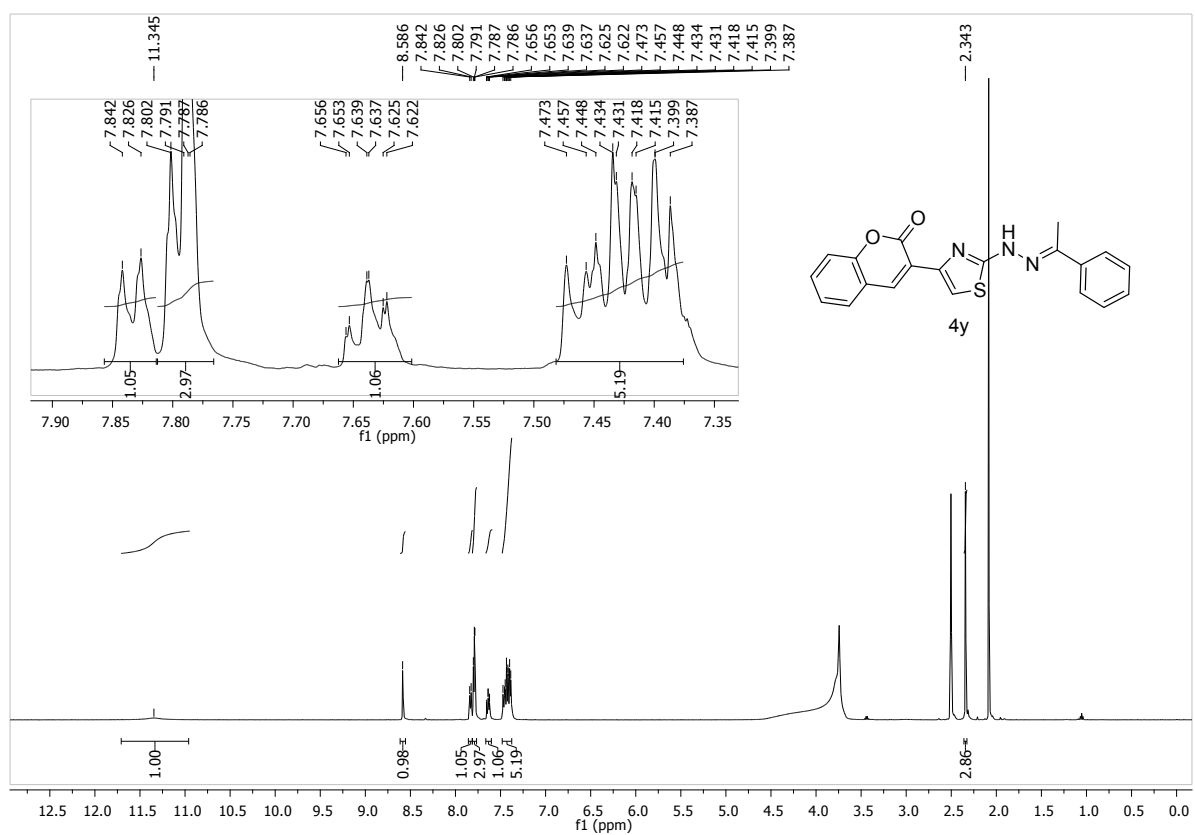


Figure S1-22. <sup>1</sup>H NMR Spectrum of 4y

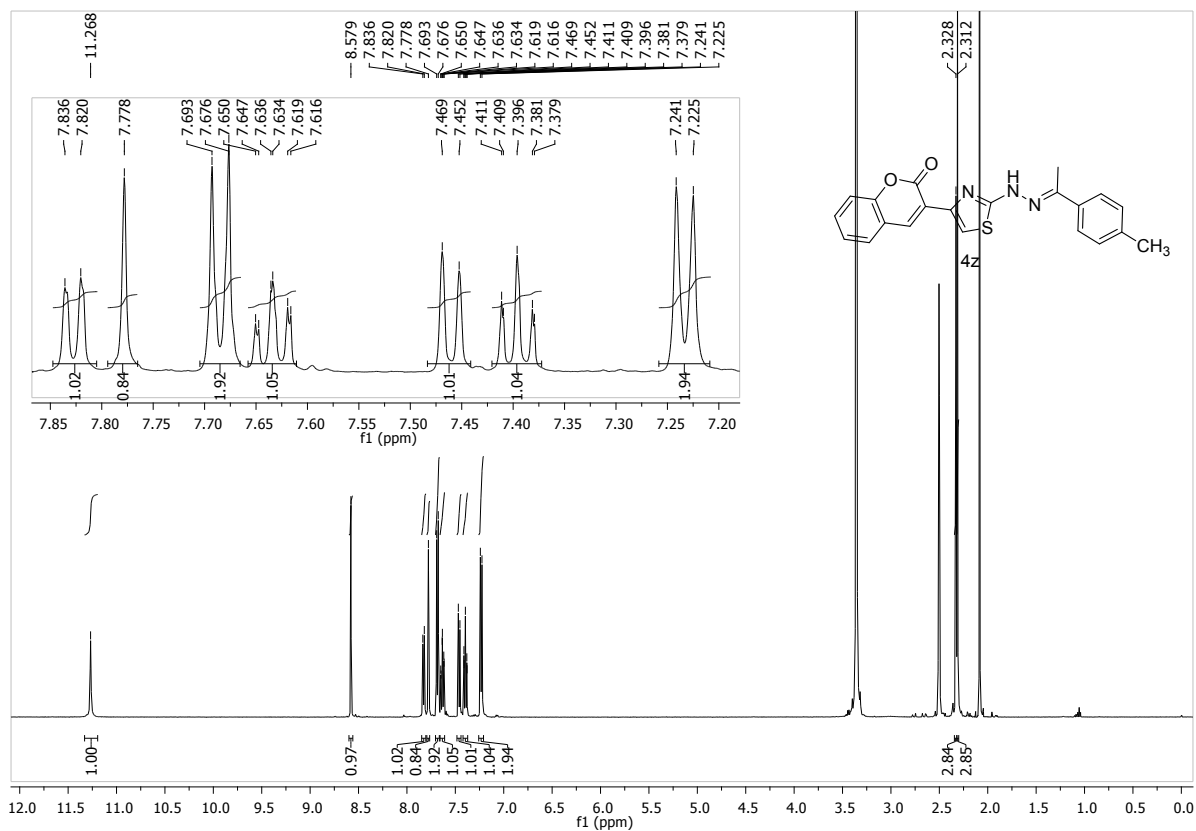


Figure S1-23. <sup>1</sup>H NMR Spectrum of 4z

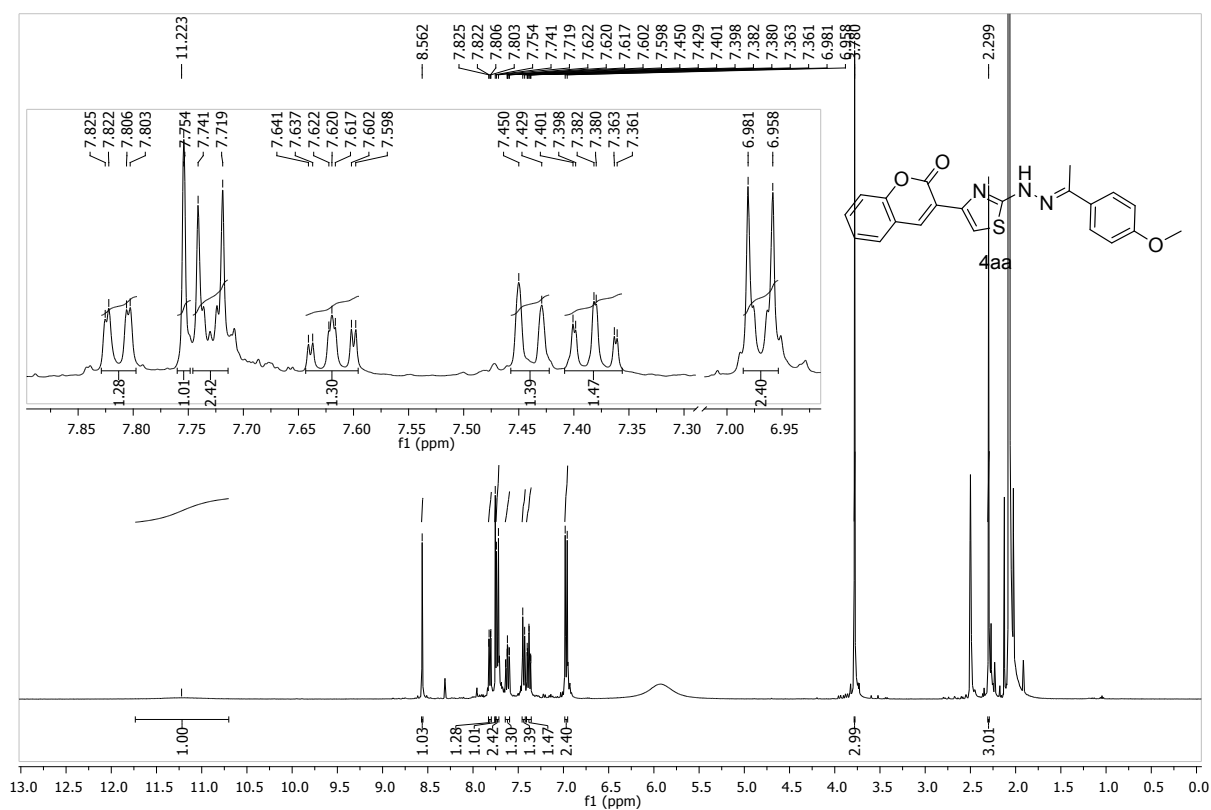


Figure S1-24. <sup>1</sup>H NMR Spectrum of 4aa

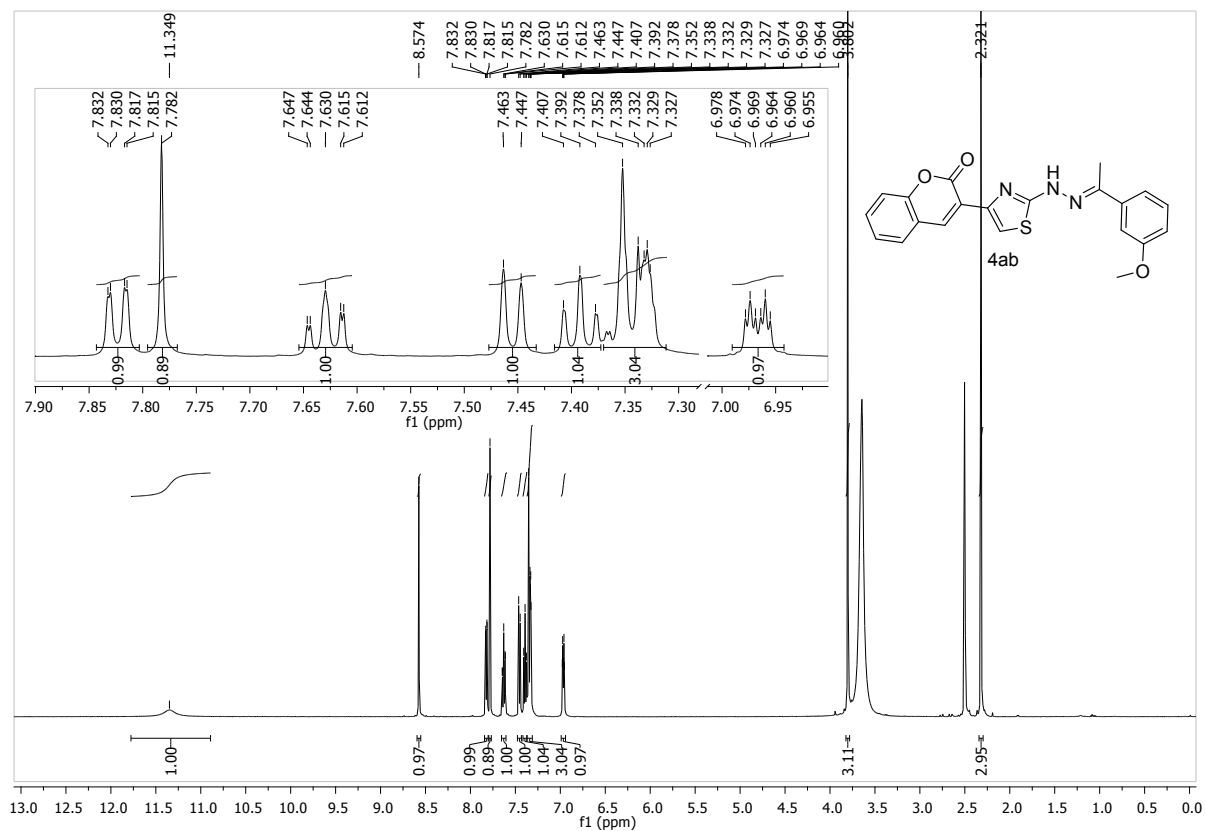


Figure S1-25. <sup>1</sup>H NMR Spectrum of 4ab

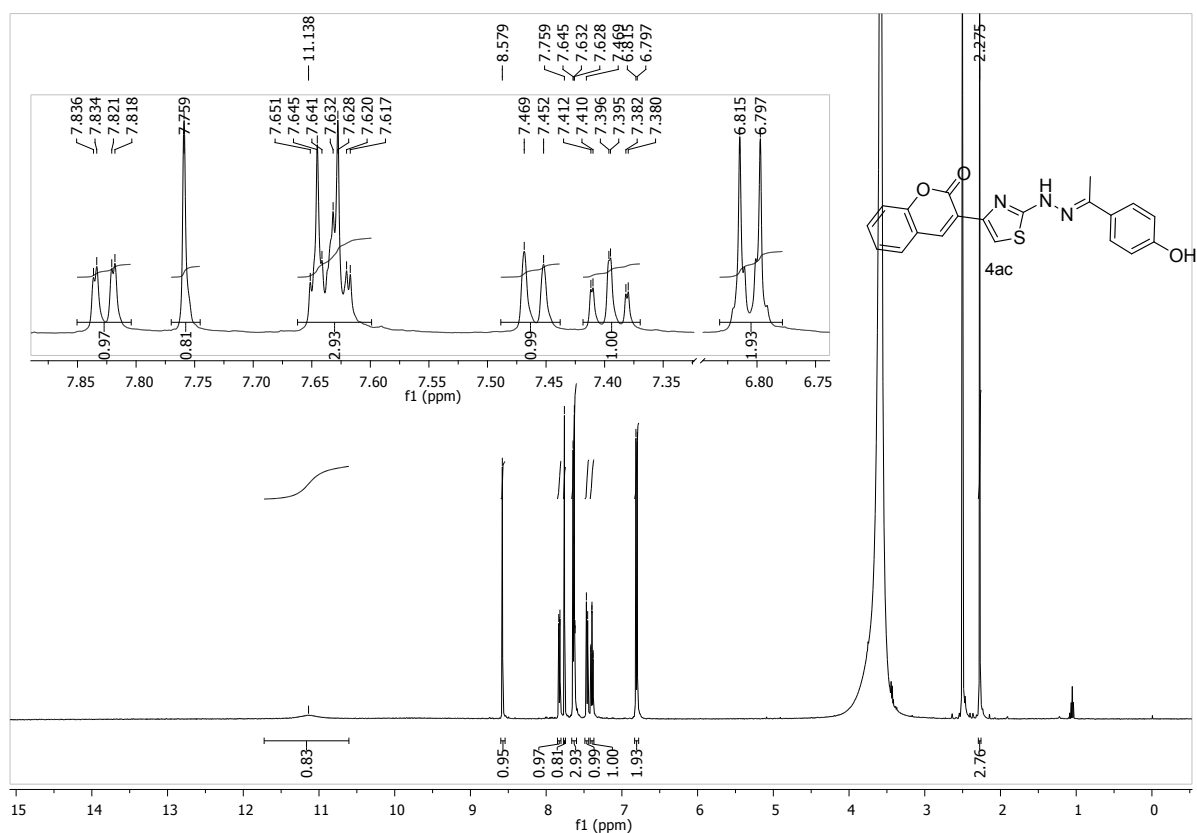


Figure S1-26. <sup>1</sup>H NMR Spectrum of 4ac



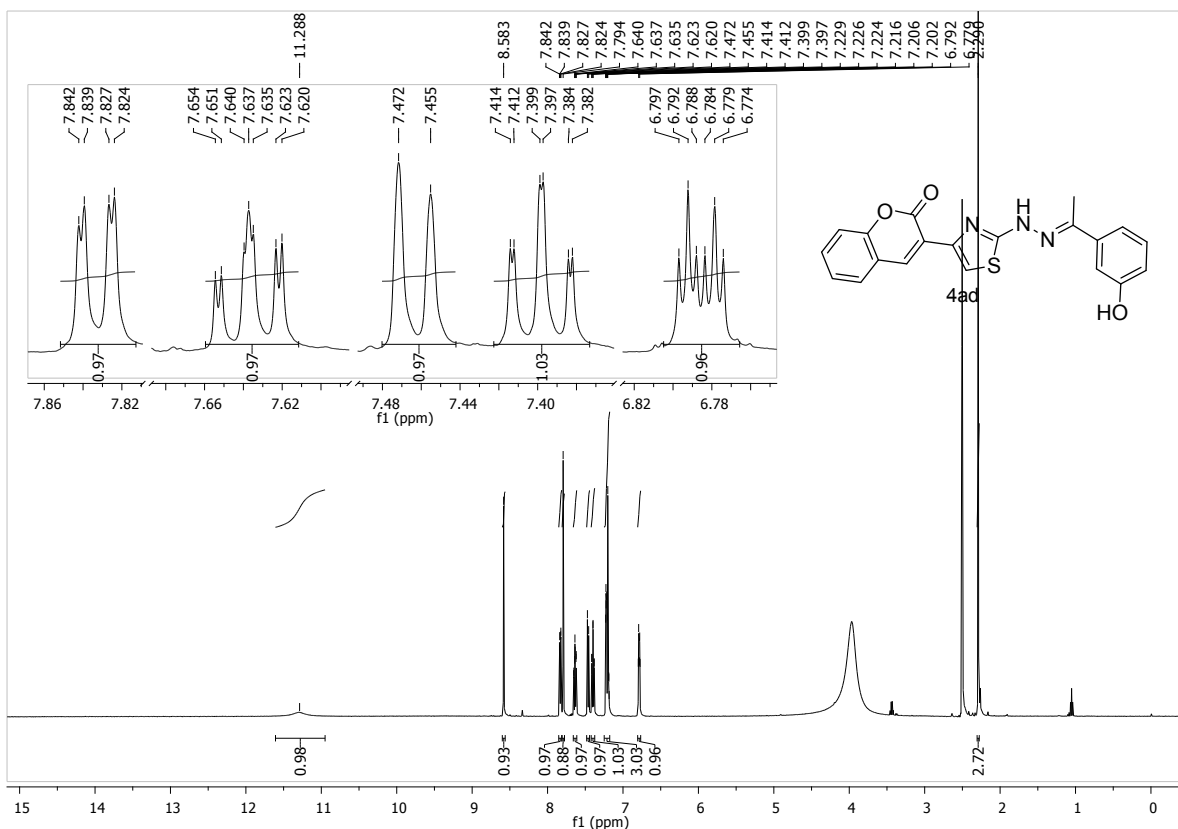


Figure S1-27. <sup>1</sup>H NMR Spectrum of 4ad

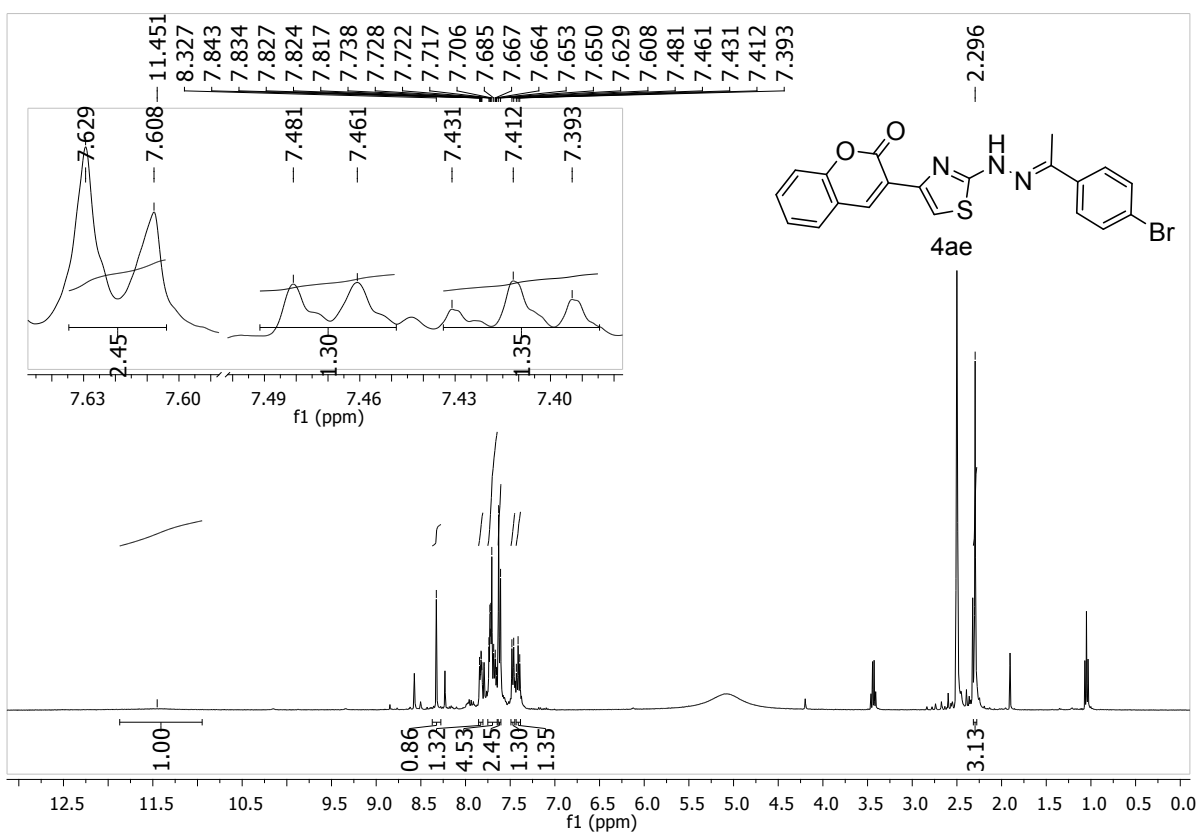


Figure S1-28. <sup>1</sup>H NMR Spectrum of Compound 4ae

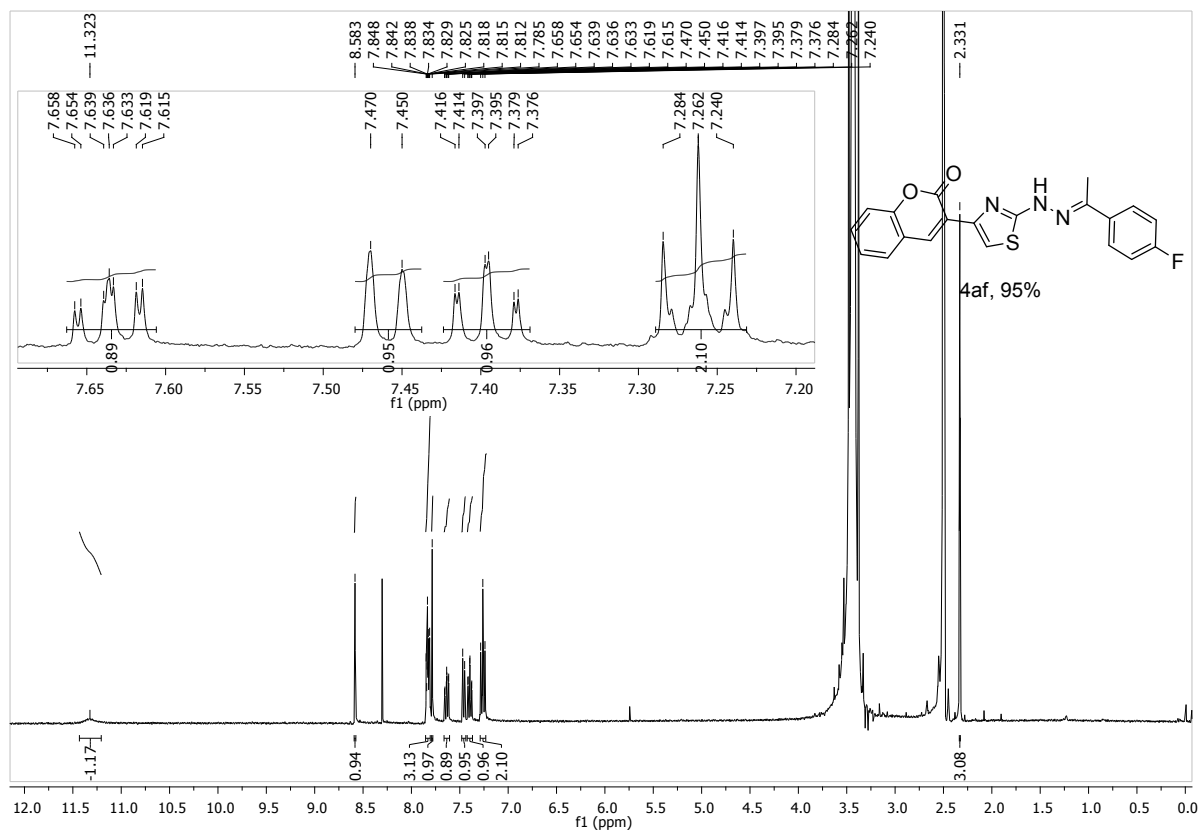


Figure S1-29. <sup>1</sup>H NMR Spectrum of 4af

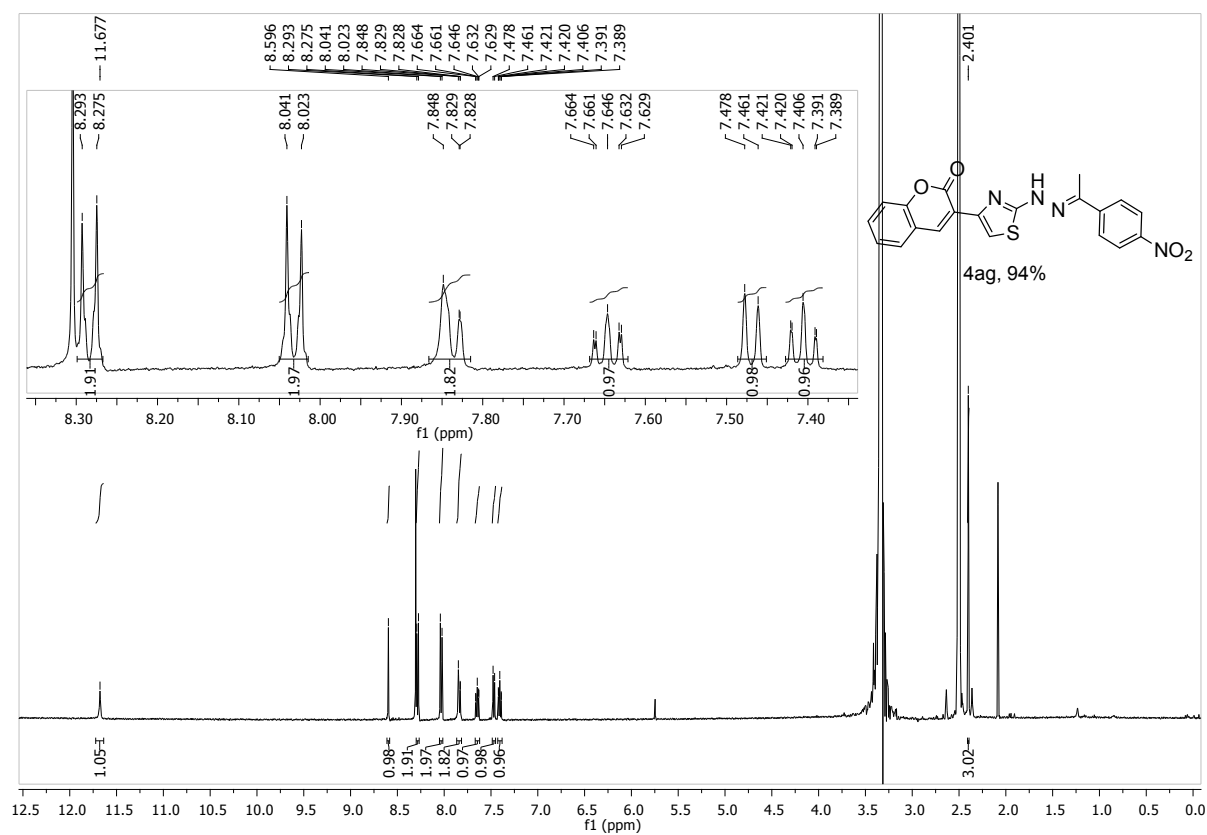


Figure S1-30. <sup>1</sup>H NMR Spectrum of 4ag

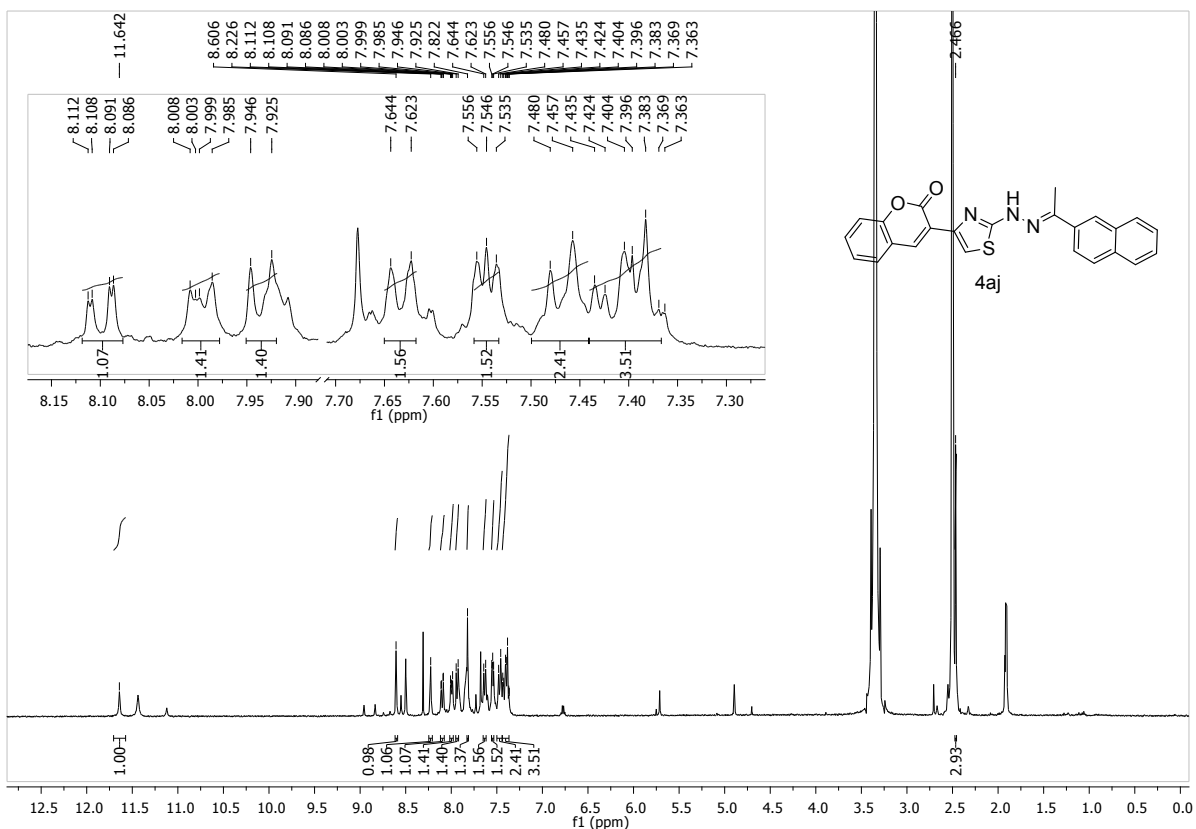


Figure S1-31. <sup>1</sup>H NMR Spectrum of Compound 4aj

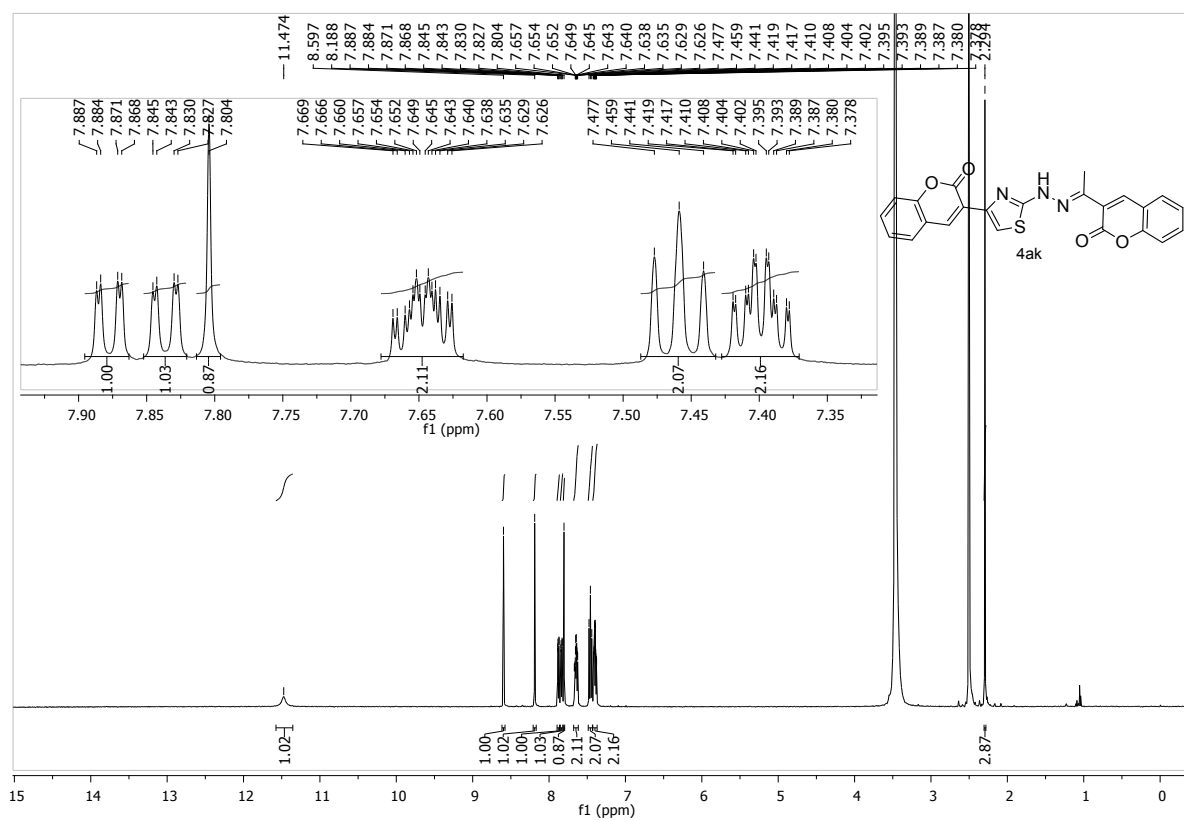


Figure S1-32. <sup>1</sup>H NMR Spectrum of Compound 4ak

Copies of HRMS and Mass spectra of synthesized compounds (4a-4ak)

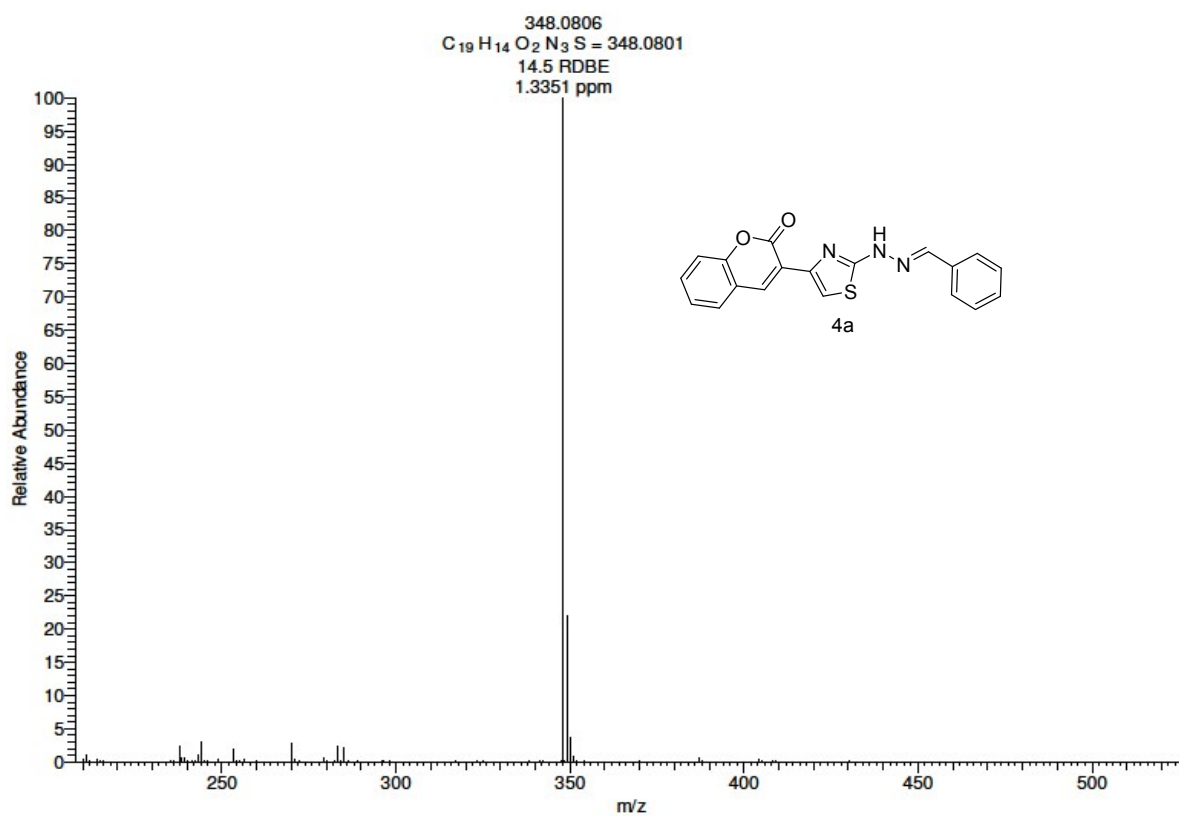


Figure S2-1. HRMS Spectrum of 4a

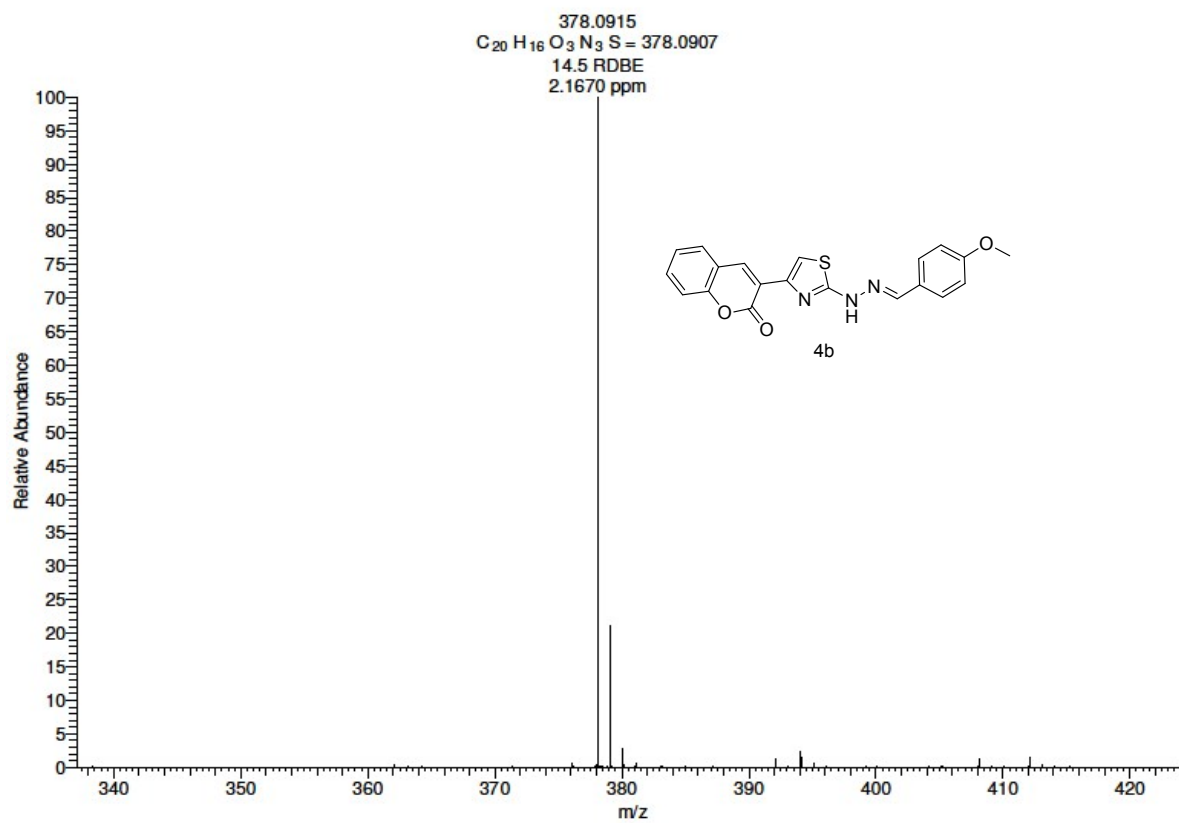


Figure S2-2. HRMS Spectrum of 4b

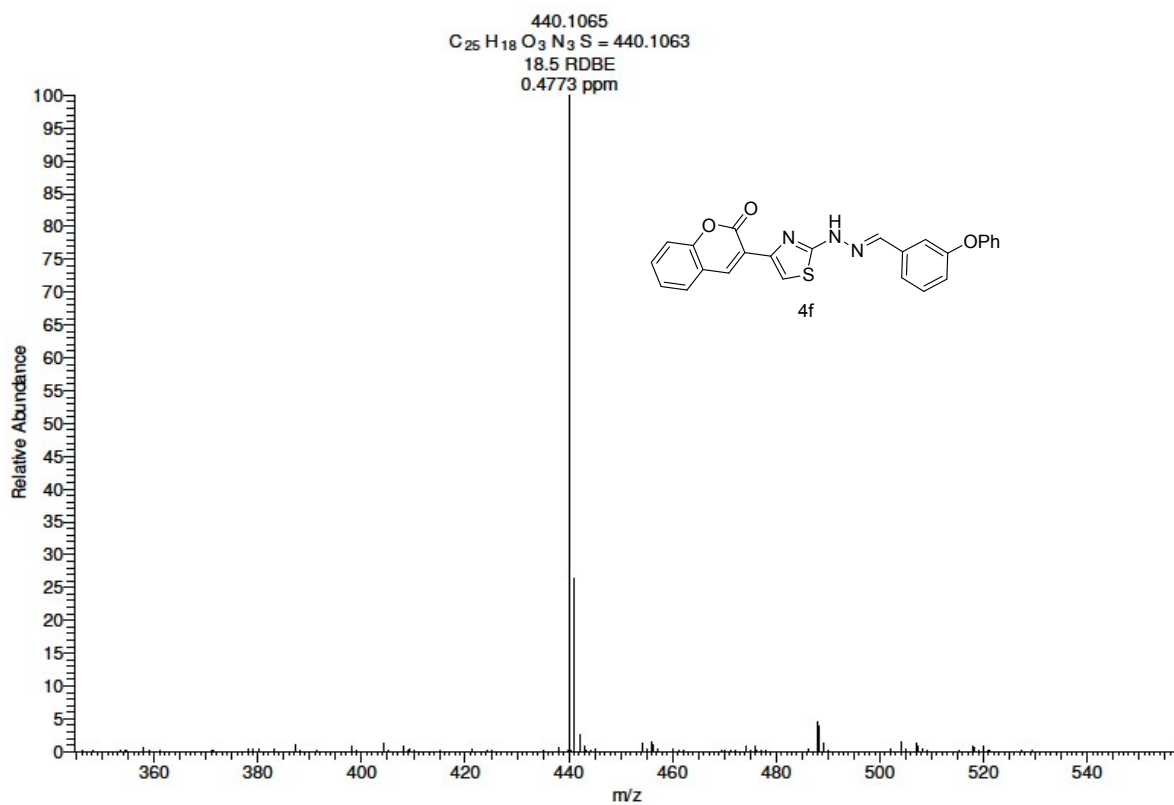


Figure S2-3. HRMS Spectrum of 4f

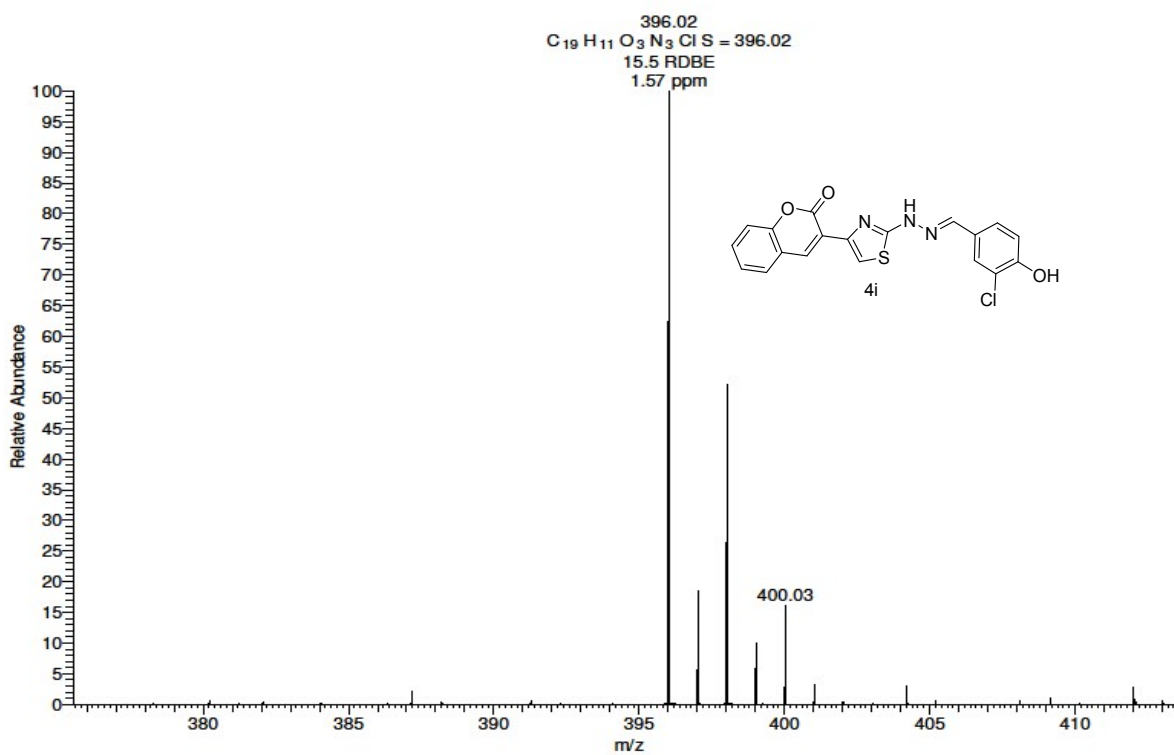


Figure S2-4. HRMS Spectrum of 4i

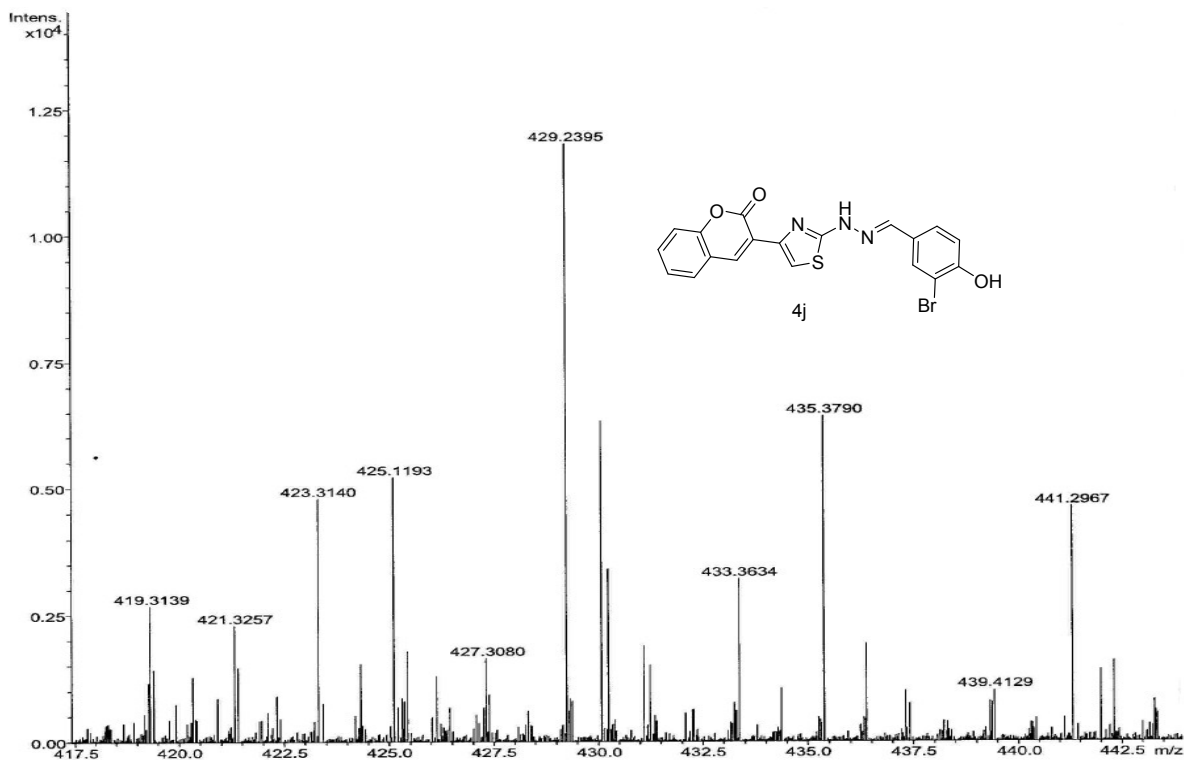


Figure S2-5. HRMS Spectrum of 4j

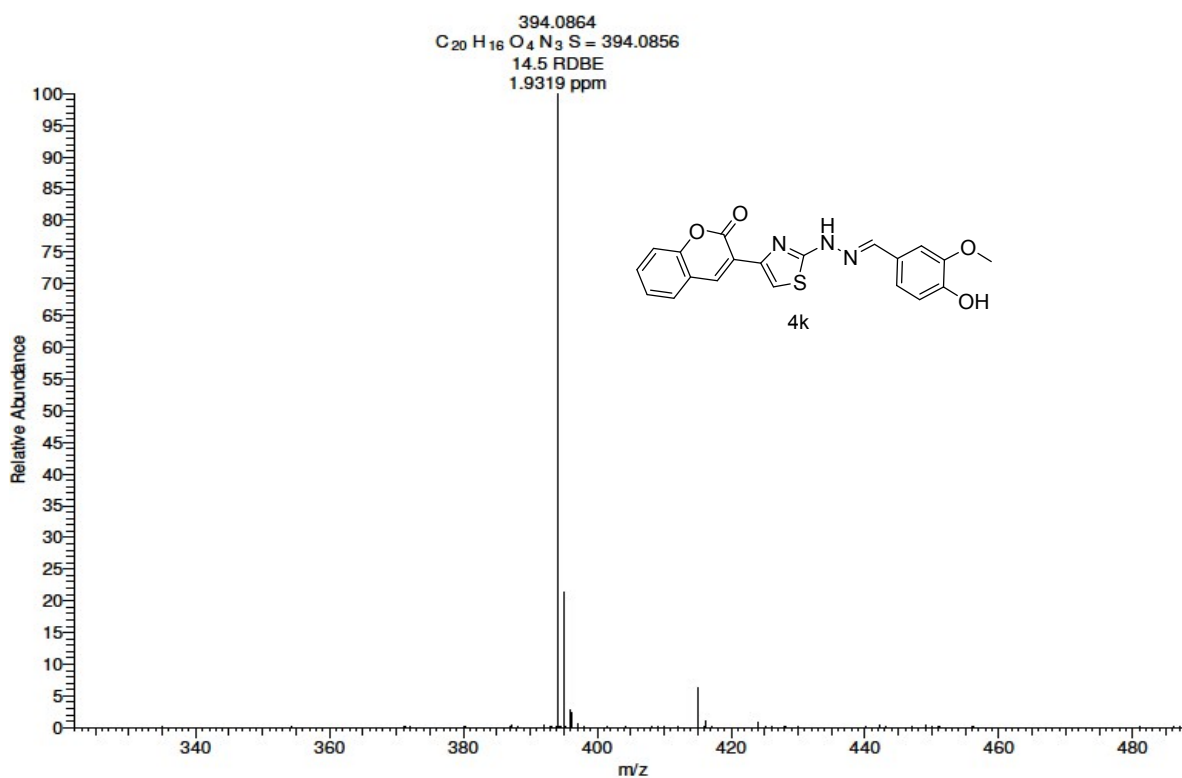


Figure S2-6. HRMS Spectrum of 4k

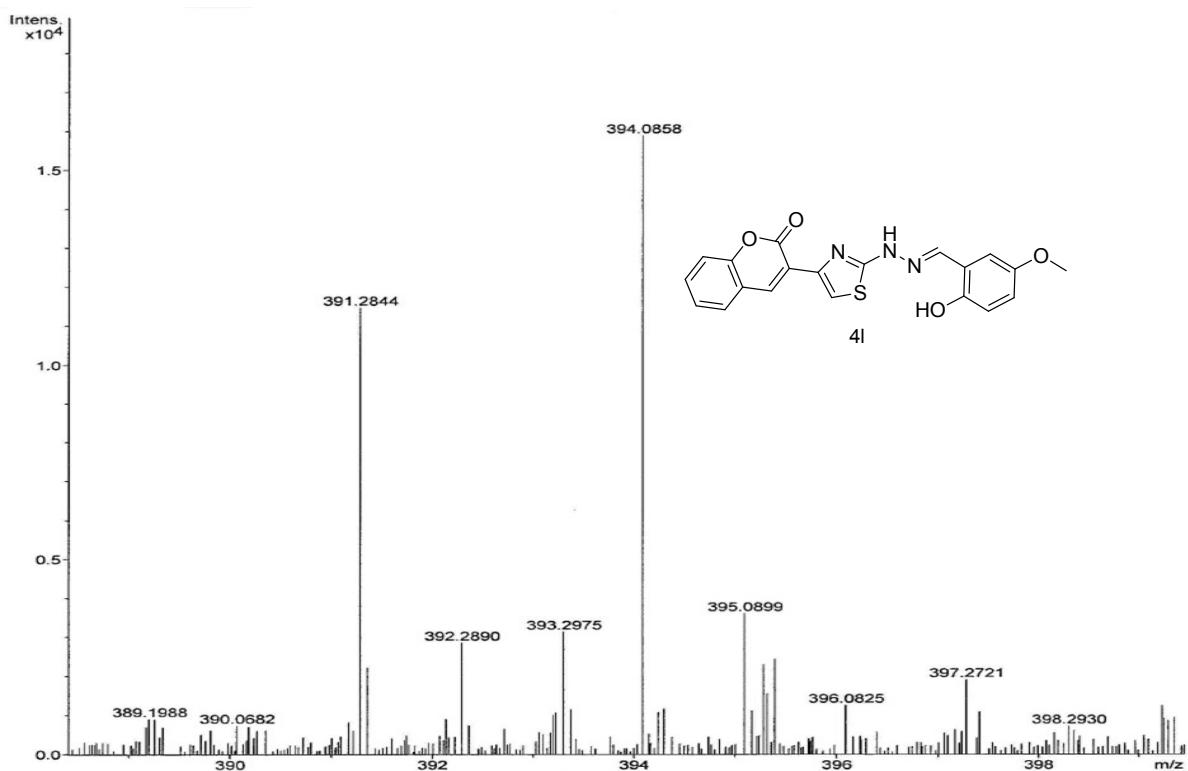


Figure S2-7. HRMS Spectrum of 4l

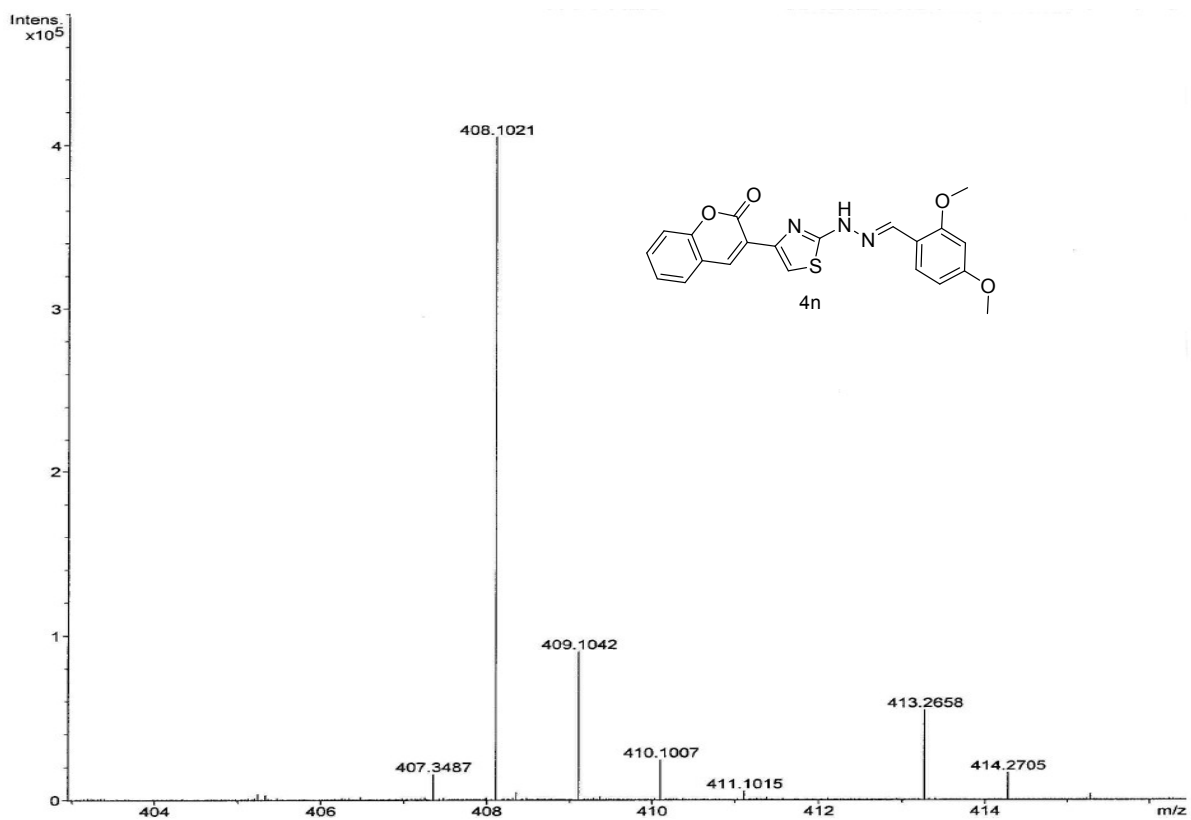


Figure S2-8. HRMS Spectrum of 4n

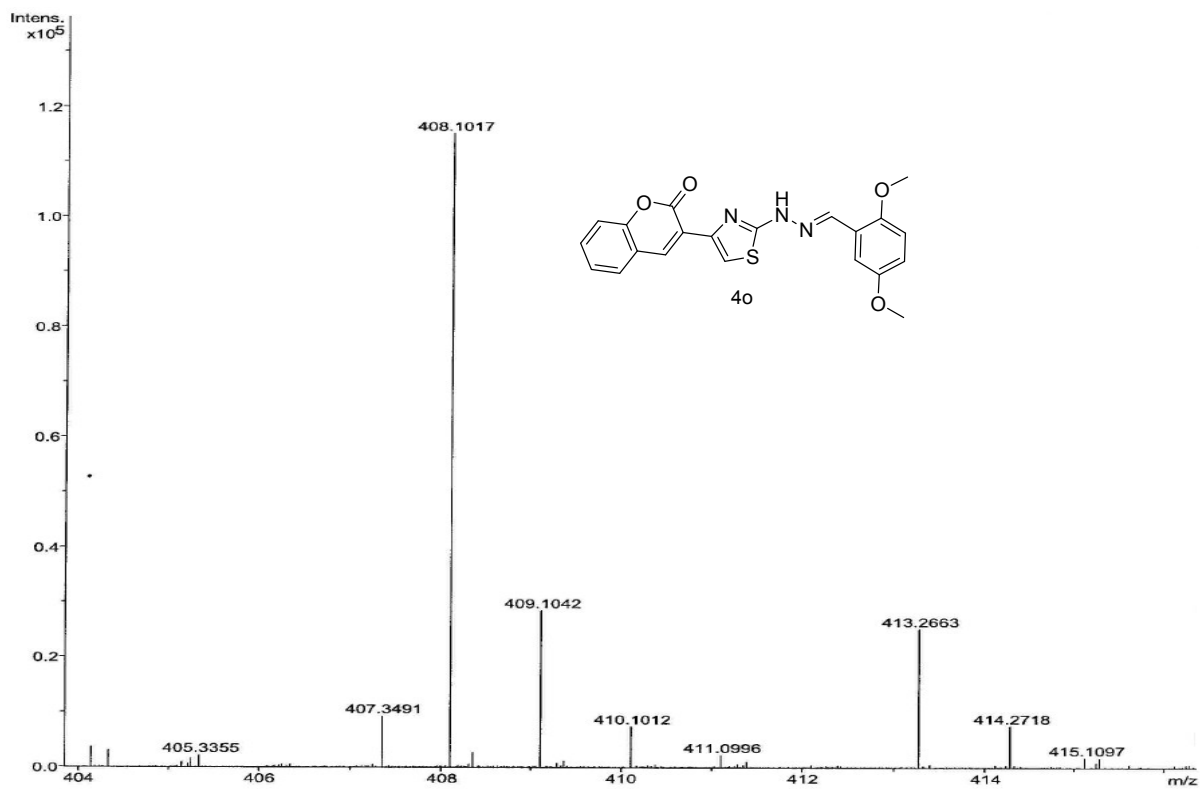


Figure S2-9. HRMS Spectrum of 4o

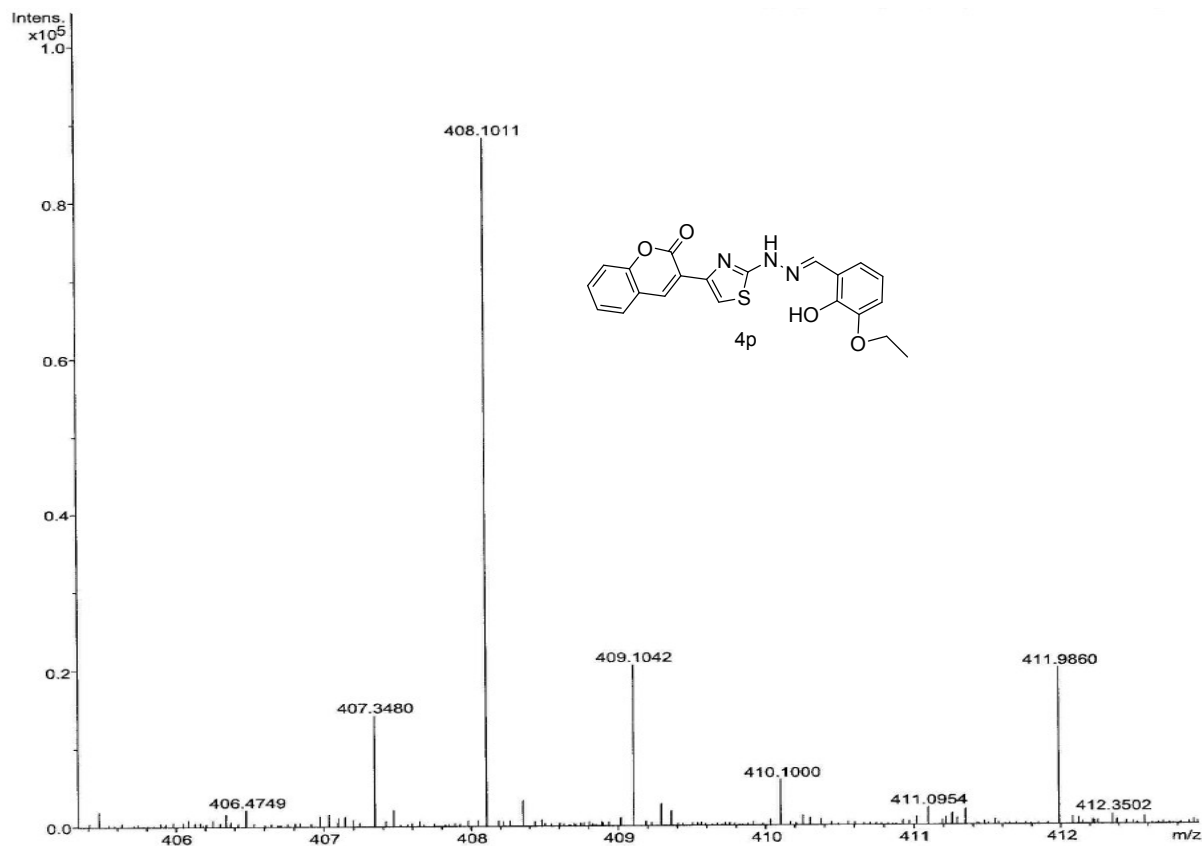


Figure S2-10. HRMS Spectrum of 4p



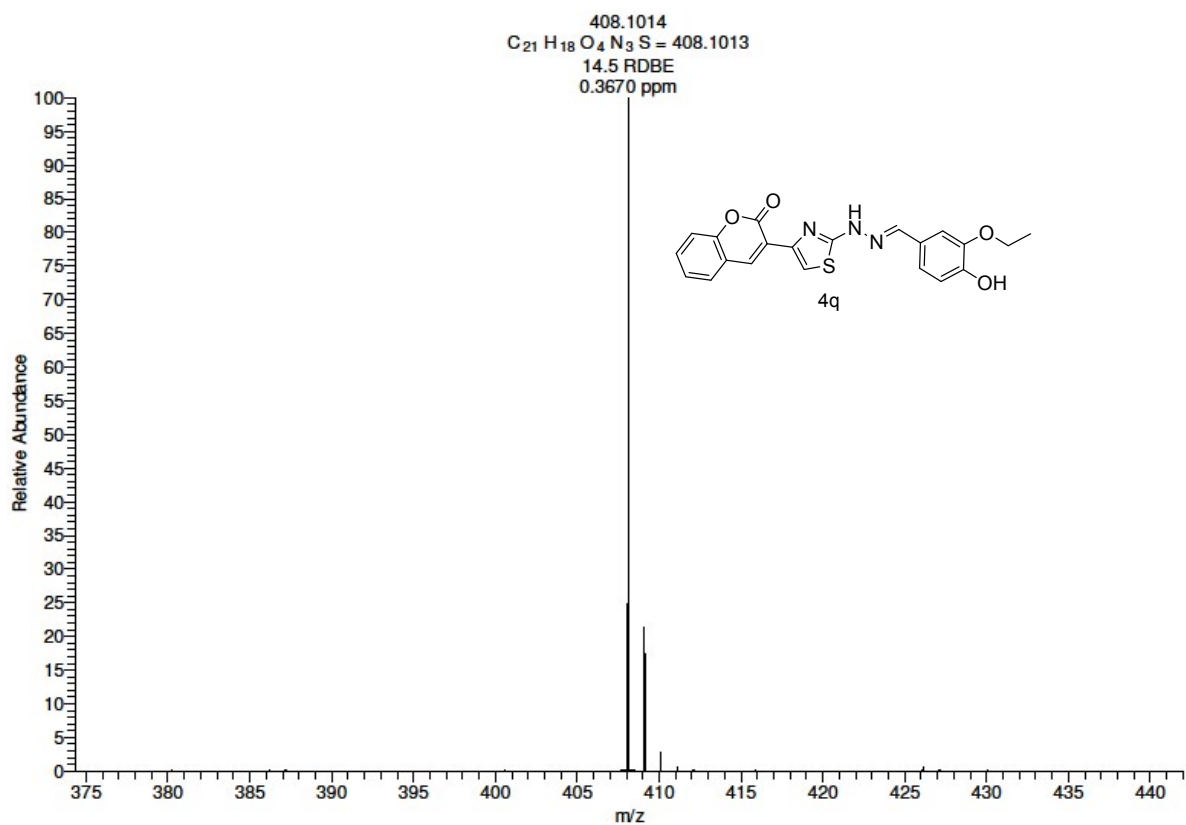


Figure S2-11. HRMS Spectrum of 4q

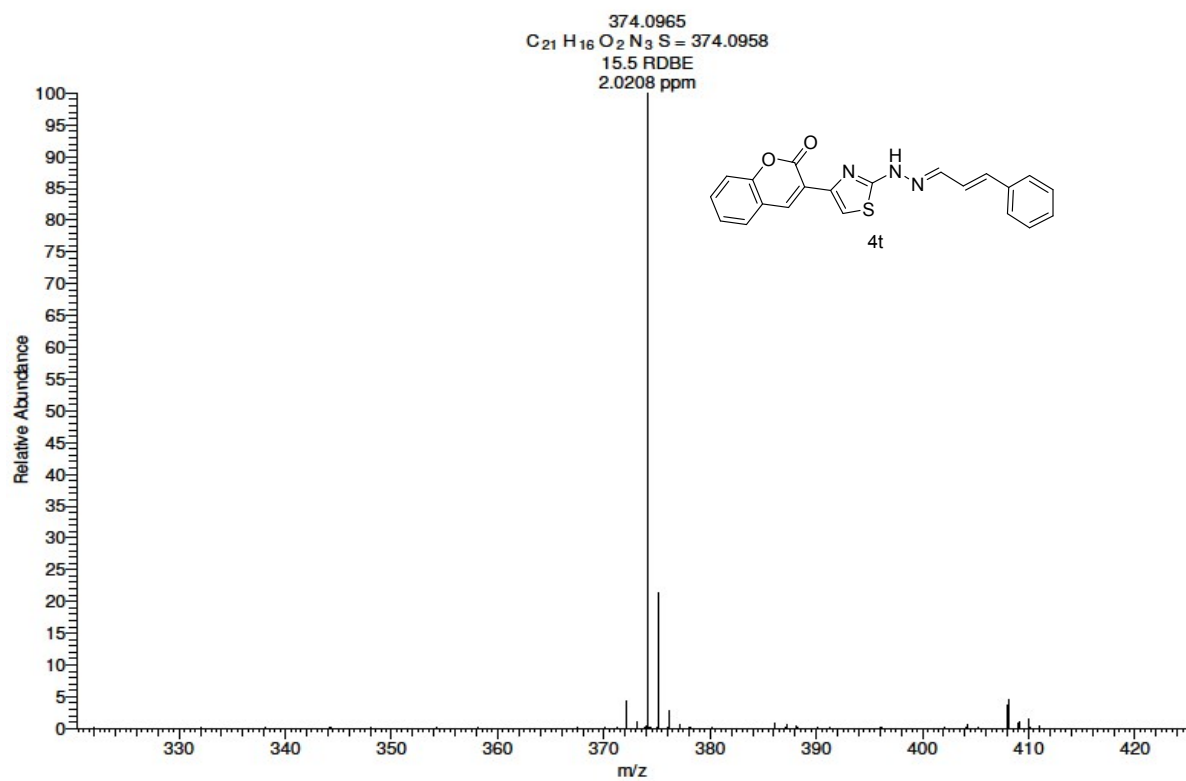


Figure S2-12. HRMS Spectrum of 4t

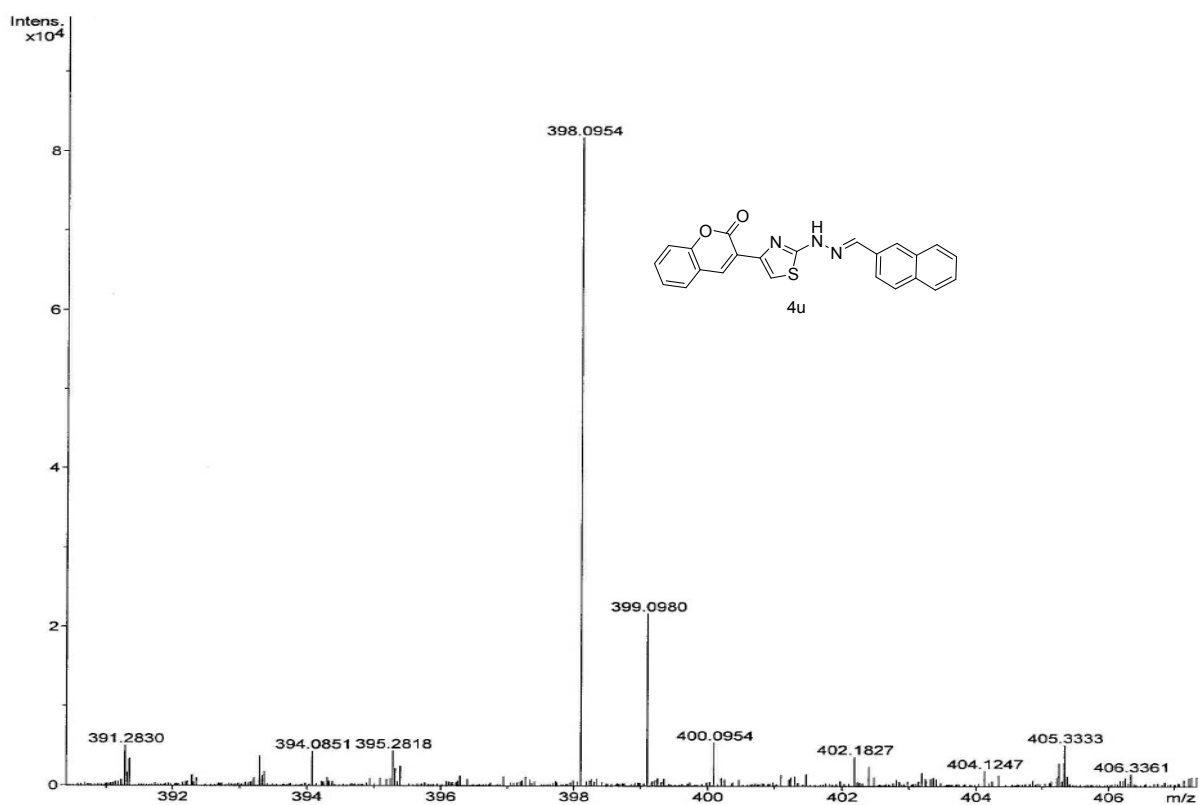
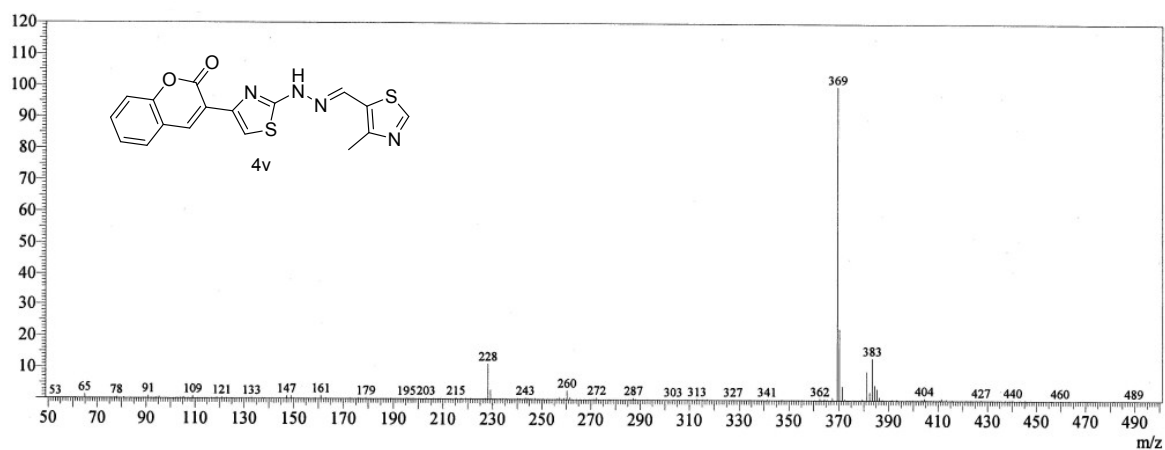
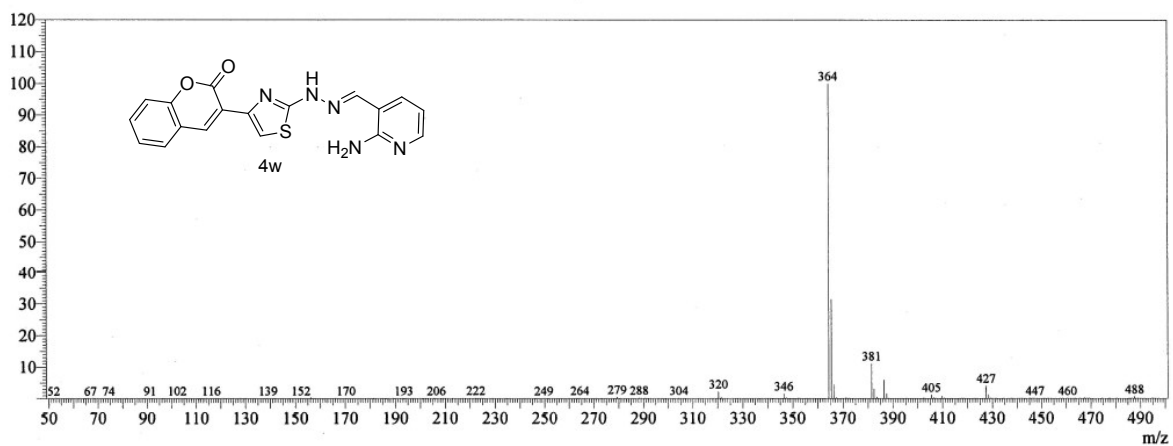


Figure S2-13. HRMS Spectrum of 4u



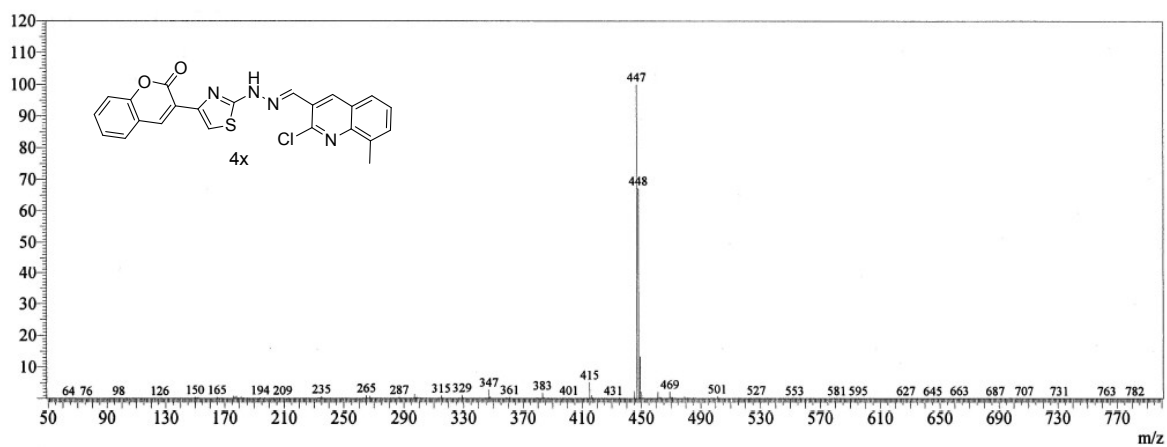
Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.757	0.543	1.043	144297678	11667039	12.36		100.00		369.45	5468346
				144297678	11667039			100.00			

Figure S2-14. Mass Spectrum of 4v



Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.897	0.527	1.043	83035536	3466592	23.95		100.00		364.40	10893080
				83035536	3466592			100.00			

Figure S2-15. 4w



Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.723	0.527	1.143	532960548	34447698	15.47		100.00		447.05	14824899
				532960548	34447698			100.00			

Figure S2-16. Mass Spectrum of 4x

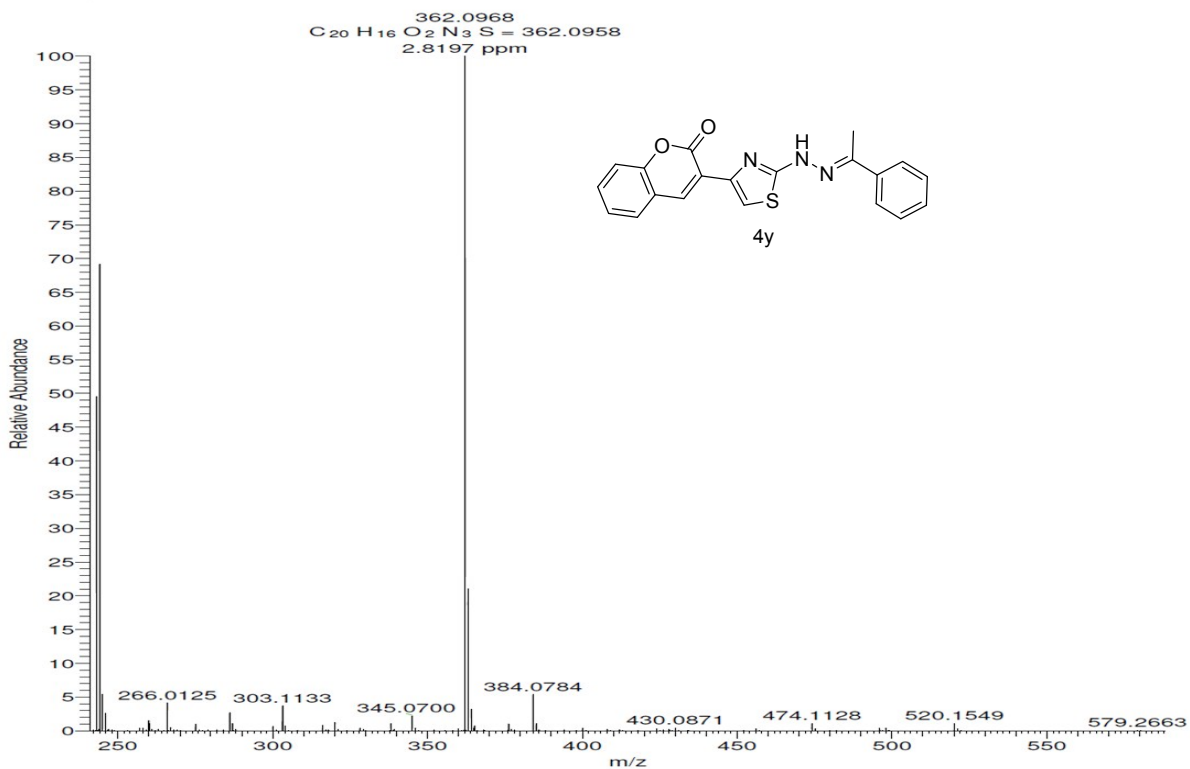


Figure S2-17. HRMS Spectrum of 4y

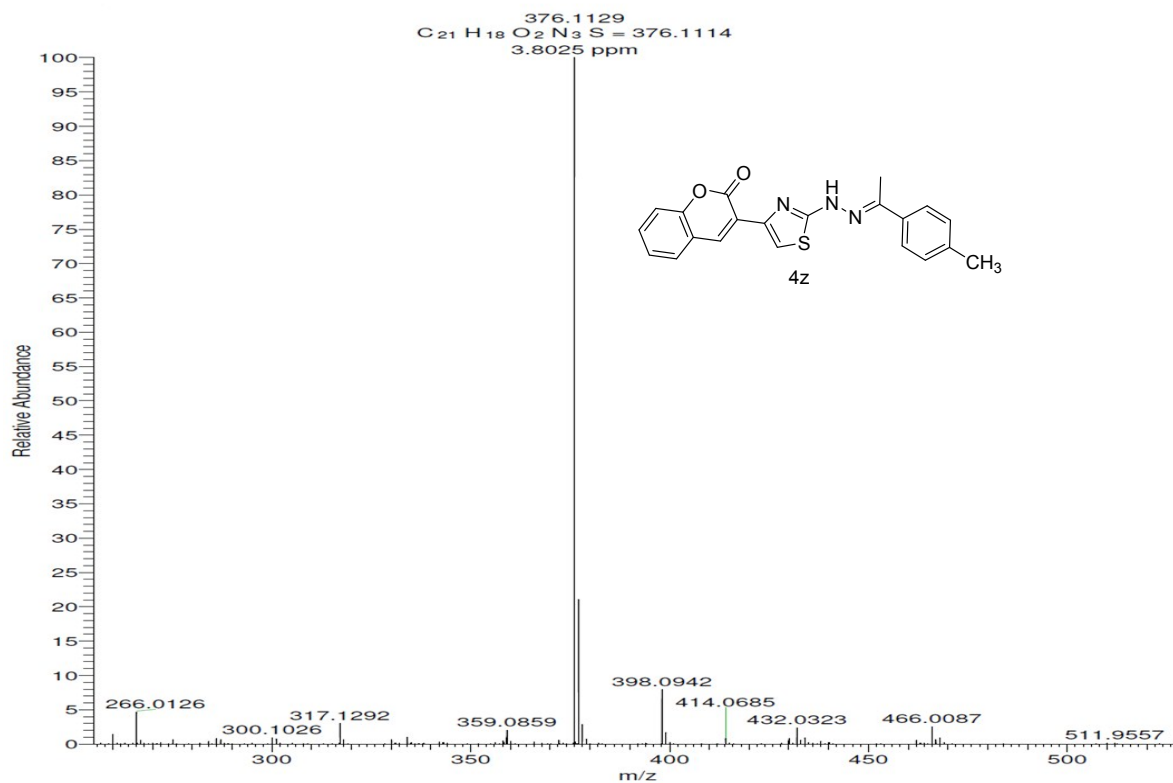


Figure S2-18. HRMS Spectrum of 4z

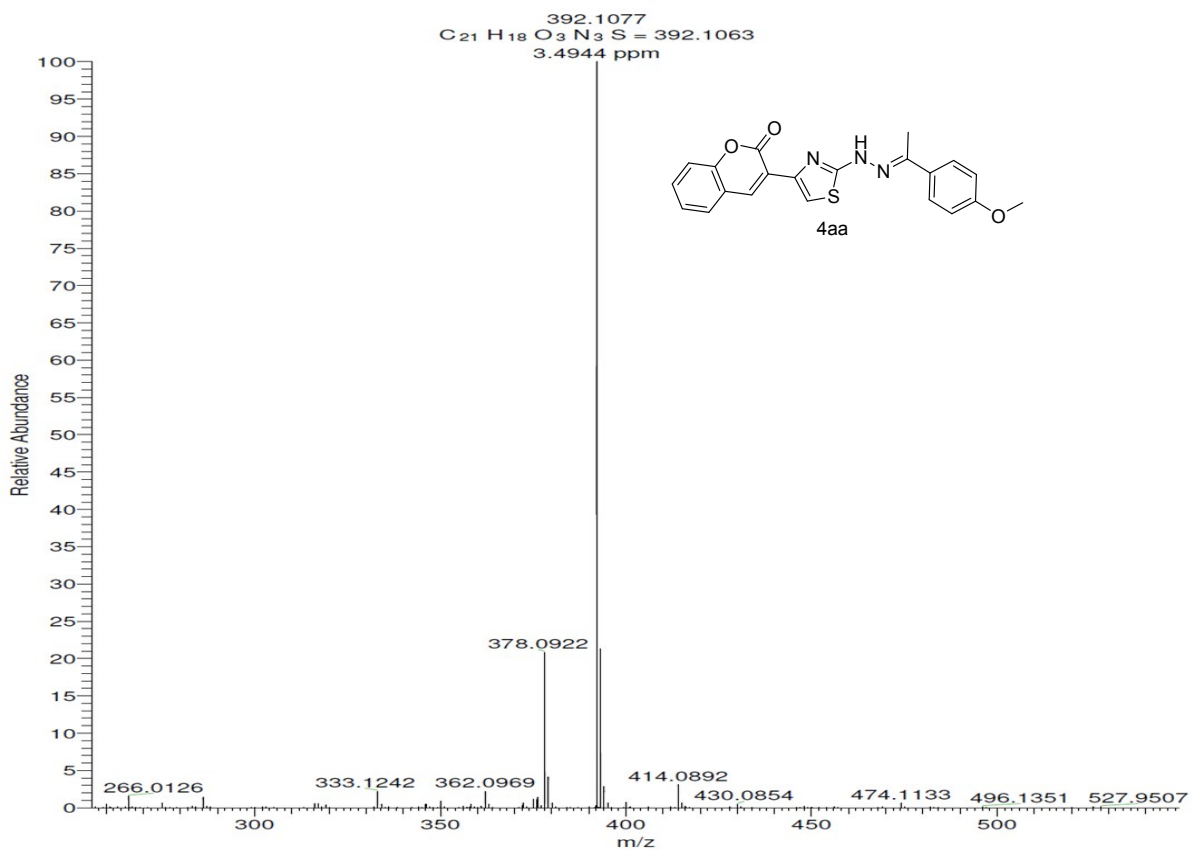
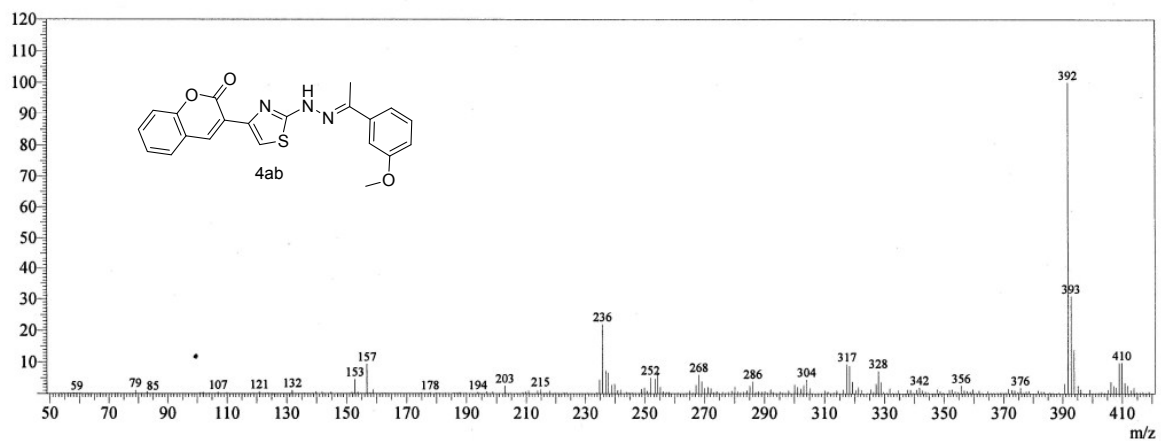


Figure S2-19. HRMS Spectrum of 4aa



Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.721	0.427	1.310	19790929	795722	24.87		100.00		391.80	150977
				19790929	795722			100.00			

Figure S2-20. Mass Spectrum of 4ab

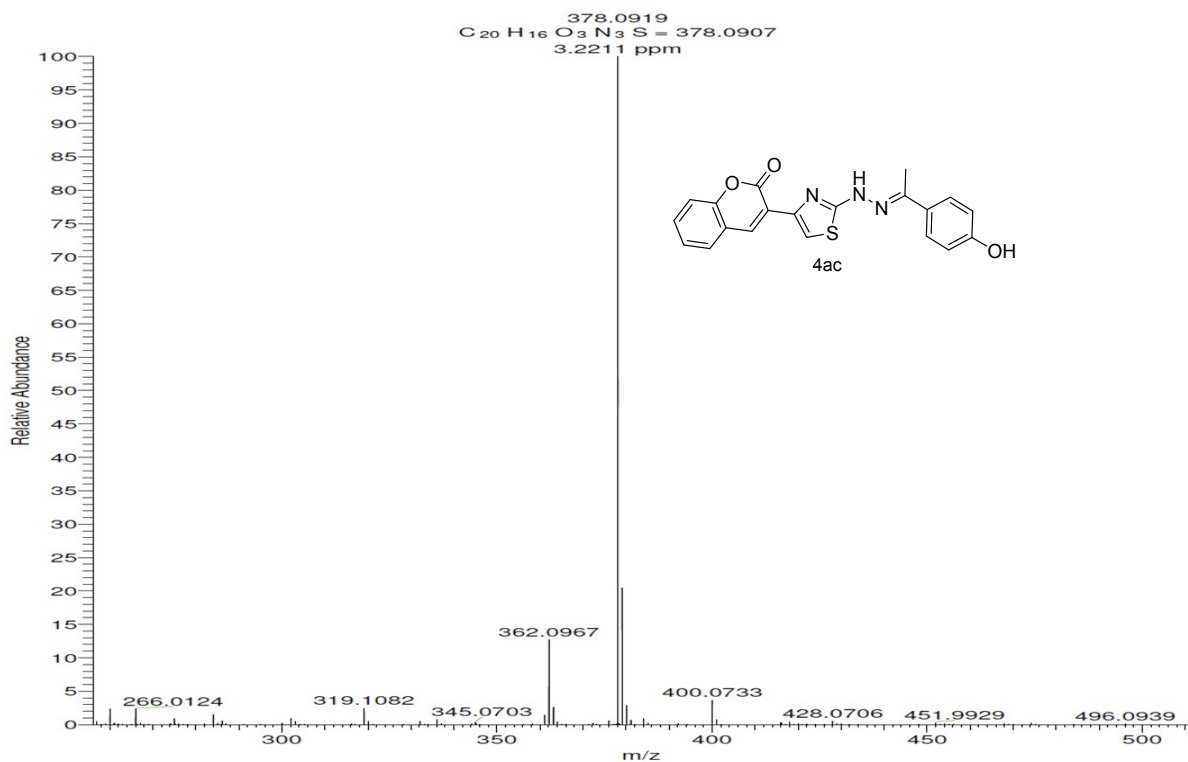


Figure S2-21. HRMS Spectrum of 4ac

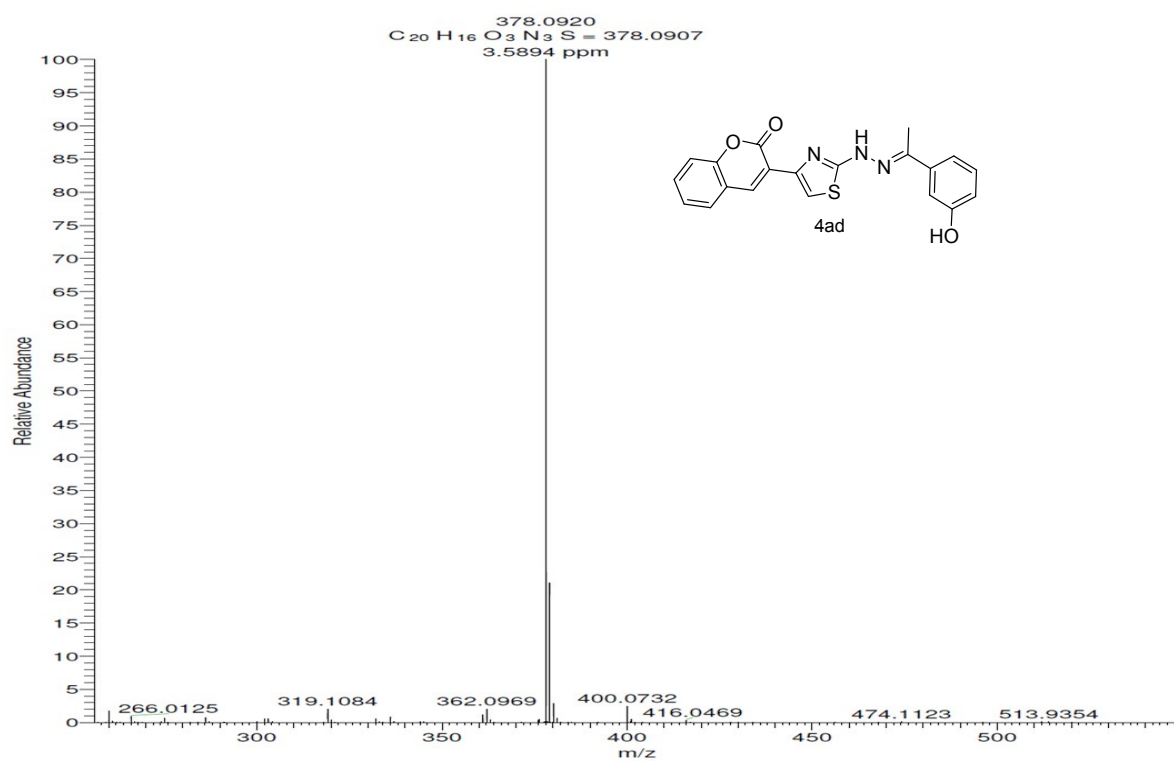
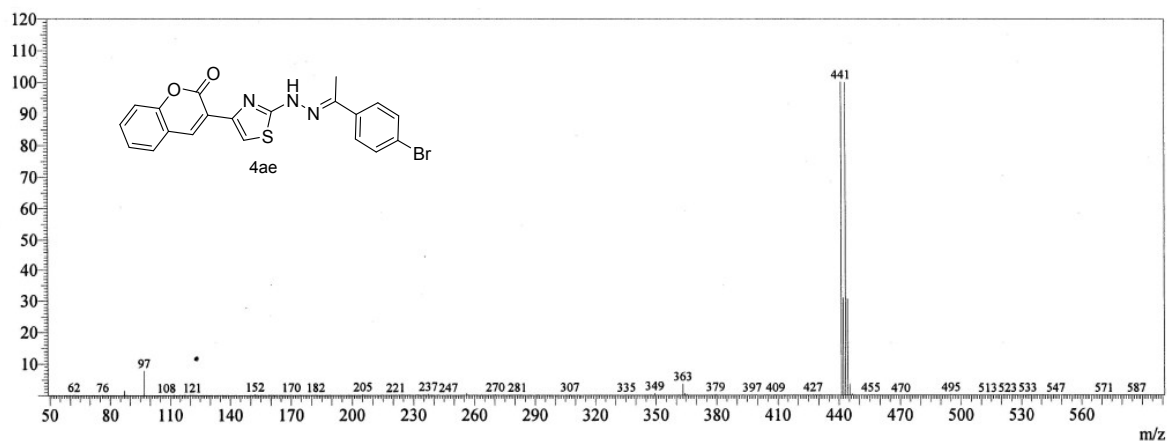
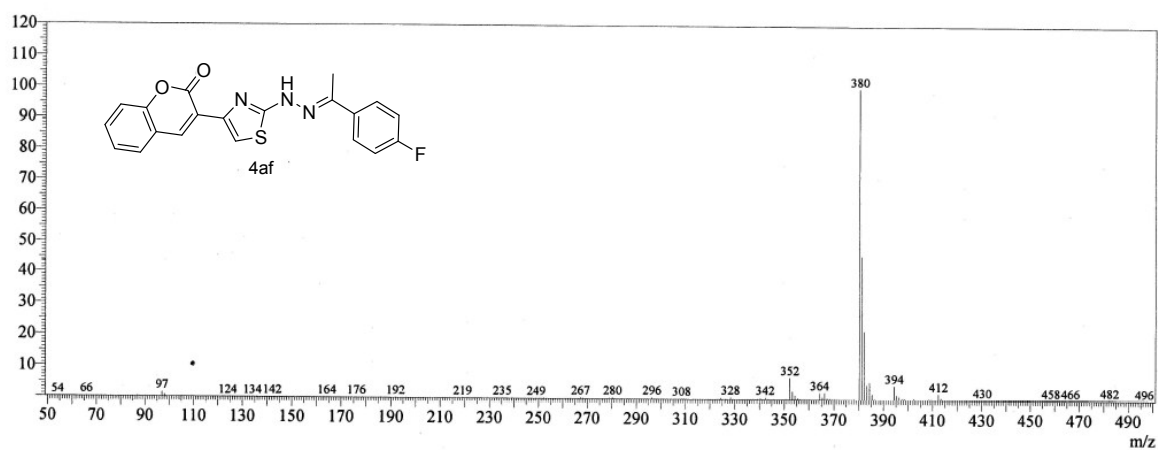


Figure S2-22. HRMS Spectrum of 4ad



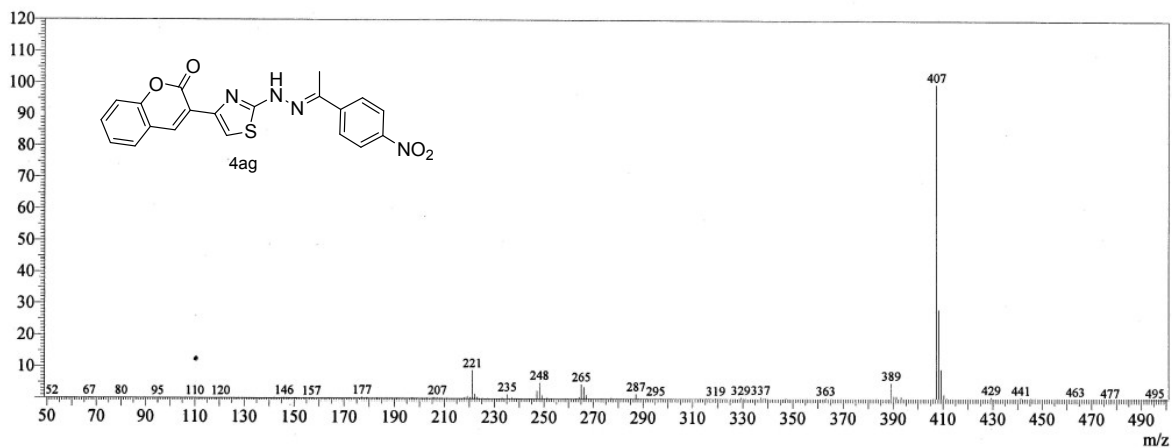
Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.774	0.543	1.044	103453966	9084372	11.38		100.00		441.25	5042040
				103453966	9084372			100.00			

Figure S2-23. Mass Spectrum of 4ae



Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.795	0.577	1.010	304473890	24751492	12.30		100.00		380.25	12304297
				304473890	24751492			100.00			

Figure S2-24. Mass Spectrum of 4af



Peak#	R.Time	I.Time	F.Time	Area	Height	A/H	Mark	%Total	Name	Base m/z	Base Int.
1	0.788	0.543	1.143	319395880	22705501	14.06		100.00		407.25	9909644
				319395880	22705501			100.00			

Figure S2-25. Mass Spectrum of 4ag

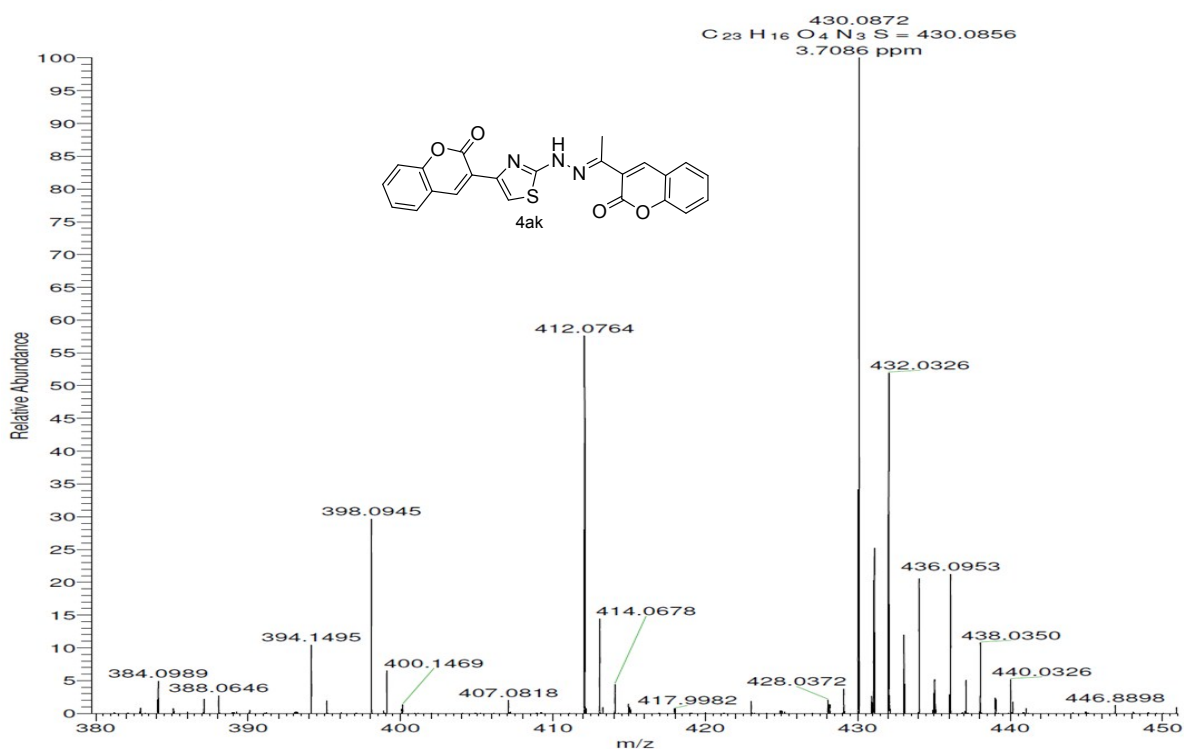
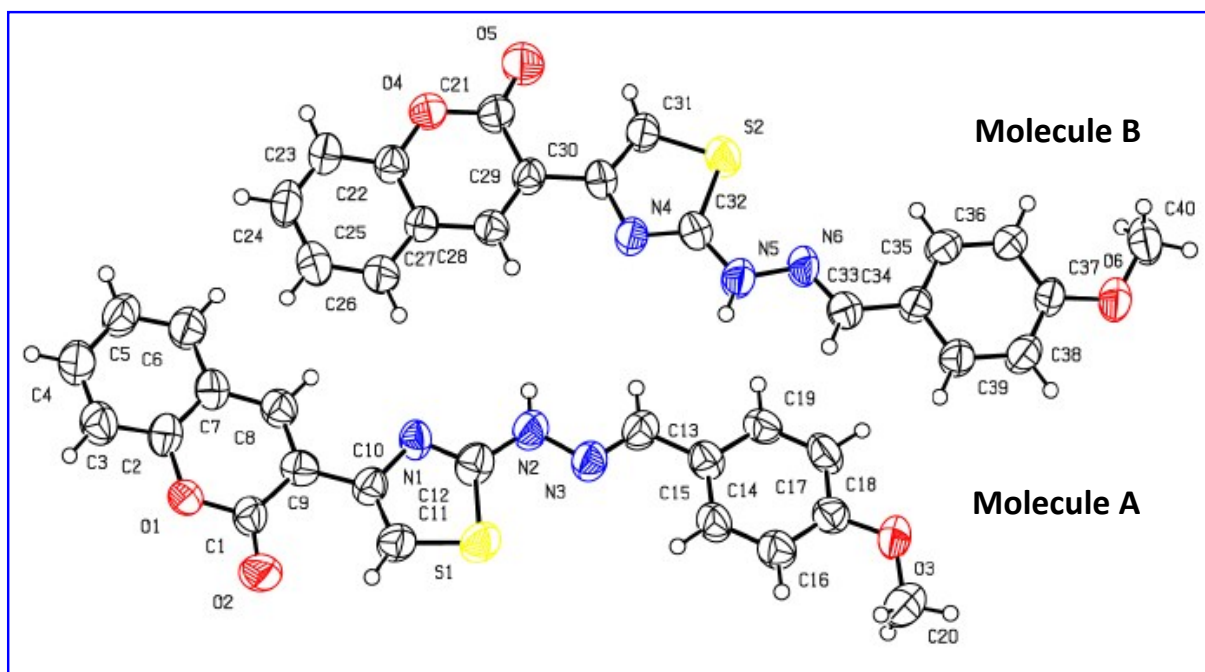


Figure S2-26. HRMS Spectrum of 4ak



## X-ray crystal structure and data of compound 4b



**Figure S3:** ORTEP diagram of (E)-3-(2-(2-(4-methoxybenzylidene) hydrazinyl)thiazol-4-yl)-2H- chromen-2-one (**4b**) (50 % probability)

**Table S1.** Crystal data and structure refinement for compound **4b**.

Identification code	shelx	
Empirical formula	C <sub>20</sub> H <sub>15</sub> N <sub>3</sub> O <sub>3</sub> S	
Formula weight	377.41	
Temperature	293(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	P 21/c	
Unit cell dimensions	a = 13.1185(7) Å	a = 90°.
	b = 20.3561(7) Å	b = 115.387(7)°.
	c = 14.7486(10) Å	g = 90°.
Volume	3558.2(4) Å <sup>3</sup>	
Z	8	
Density (calculated)	1.409 Mg/m <sup>3</sup>	
Absorption coefficient	0.209 mm <sup>-1</sup>	
F(000)	1568	
Crystal size	? x ? x ? mm <sup>3</sup>	
Theta range for data collection	1.988 to 24.998°.	
Index ranges	-15 ≤ h ≤ 15, -24 ≤ k ≤ 24, -17 ≤ l ≤ 17	

Reflections collected	33901
Independent reflections	6260 [R(int) = 0.0642]
Completeness to theta = 24.998°	100.0 %
Refinement method	Full-matrix least-squares on F <sup>2</sup>
Data / restraints / parameters	6260 / 0 / 489
Goodness-of-fit on F <sup>2</sup>	0.914
Final R indices [I>2sigma(I)]	R1 = 0.0878, wR2 = 0.2082
R indices (all data)	R1 = 0.1842, wR2 = 0.2701
Extinction coefficient	n/a
Largest diff. peak and hole	1.034 and -0.280 e.Å <sup>-3</sup>

**Table S2.** Atomic coordinates ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for skd19a.  $U(\text{eq})$  is defined as one third of the trace of the orthogonalized  $U_{ij}$  tensor.

	x	y	z	U(eq)
S(2)	6104(1)	3150(1)	6430(1)	66(1)
S(1)	-1072(1)	4916(1)	3631(1)	73(1)
O(1)	-1134(3)	7884(2)	3415(2)	64(1)
O(4)	6038(3)	6127(2)	6400(2)	65(1)
O(6)	4795(3)	-935(2)	6078(3)	78(1)
O(3)	197(3)	842(2)	3991(3)	80(1)
N(1)	518(3)	5709(2)	3720(3)	57(1)
N(3)	638(4)	3963(2)	3904(3)	62(1)
N(4)	4466(3)	3932(2)	6236(3)	57(1)
N(6)	4447(3)	2194(2)	6244(3)	60(1)
O(5)	7126(3)	5264(2)	6598(3)	92(1)
N(5)	4050(3)	2815(2)	6243(3)	71(1)
O(2)	-2209(3)	7031(2)	3238(3)	85(1)
N(2)	1009(4)	4590(2)	3883(3)	72(1)
C(32)	4746(4)	3312(3)	6292(3)	57(1)
C(30)	5366(4)	4319(2)	6302(3)	51(1)
C(37)	4644(4)	-287(2)	6166(3)	56(1)
C(17)	397(4)	1501(3)	3973(3)	58(1)
C(34)	4108(4)	1032(2)	6261(3)	53(1)
C(14)	989(4)	2812(2)	3988(3)	58(1)
C(10)	-416(4)	6084(2)	3599(3)	57(1)
C(28)	4220(4)	5322(2)	6088(3)	55(1)

C(27)	4081(4)	6019(2)	6078(3)	51(1)
C(22)	5020(4)	6408(2)	6255(3)	57(1)
C(7)	802(4)	7788(2)	3703(3)	59(1)
C(29)	5214(4)	5031(2)	6258(3)	52(1)
C(12)	265(4)	5090(3)	3752(4)	65(2)
C(15)	-64(4)	2643(2)	3903(4)	65(1)
C(2)	-113(5)	8179(2)	3554(4)	61(1)
C(35)	5181(4)	842(2)	6407(3)	62(1)
C(8)	672(4)	7099(2)	3701(3)	58(1)
C(38)	3579(4)	-114(2)	6045(4)	67(1)
C(33)	3798(4)	1718(2)	6274(3)	62(1)
C(39)	3310(4)	537(2)	6094(3)	61(1)
C(9)	-300(4)	6802(2)	3574(3)	55(1)
C(18)	1445(4)	1656(3)	4049(4)	68(1)
C(36)	5457(4)	193(2)	6361(3)	62(1)
C(23)	5003(5)	7078(3)	6291(4)	70(2)
C(21)	6193(4)	5454(3)	6438(4)	63(1)
C(13)	1320(5)	3509(2)	3986(4)	68(2)
C(1)	-1266(4)	7221(2)	3409(4)	61(1)
C(16)	-360(4)	1984(3)	3896(4)	67(1)
C(19)	1739(4)	2309(3)	4057(3)	66(1)
C(3)	-105(5)	8852(3)	3544(4)	73(2)
C(24)	4034(5)	7384(2)	6171(4)	73(2)
C(31)	6298(4)	3974(2)	6407(3)	60(1)
C(6)	1788(5)	8124(3)	3848(4)	71(2)
C(26)	3094(4)	6339(3)	5942(4)	71(2)
C(4)	873(5)	9163(3)	3680(4)	76(2)
C(25)	3083(5)	7017(3)	5985(4)	77(2)
C(5)	1823(5)	8797(3)	3837(4)	79(2)
C(11)	-1318(4)	5742(2)	3554(3)	68(2)
C(40)	5895(5)	-1142(3)	6216(4)	89(2)
C(20)	-933(5)	640(3)	3742(4)	103(2)

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**Table S3.** Bond lengths [Å] and angles [°] for skd19a.

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S(2)-C(31)	1.699(4)
S(2)-C(32)	1.737(5)
S(1)-C(11)	1.707(5)
S(1)-C(12)	1.722(5)
O(1)-C(1)	1.361(5)
O(1)-C(2)	1.401(6)
O(4)-C(21)	1.382(5)
O(4)-C(22)	1.383(5)
O(6)-C(37)	1.349(5)
O(6)-C(40)	1.432(5)
O(3)-C(17)	1.369(5)
O(3)-C(20)	1.427(6)
N(1)-C(12)	1.309(6)
N(1)-C(10)	1.390(5)
N(3)-C(13)	1.256(6)
N(3)-N(2)	1.371(5)
N(4)-C(32)	1.308(5)
N(4)-C(30)	1.387(5)
N(6)-C(33)	1.303(5)
N(6)-N(5)	1.368(5)
O(5)-C(21)	1.207(5)
N(5)-C(32)	1.343(6)
N(5)-H(5)	0.8600
O(2)-C(1)	1.215(5)
N(2)-C(12)	1.365(6)
N(2)-H(2)	0.8600
C(30)-C(31)	1.361(6)
C(30)-C(29)	1.460(6)
C(37)-C(38)	1.376(6)
C(37)-C(36)	1.383(6)
C(17)-C(16)	1.367(6)
C(17)-C(18)	1.368(6)
C(34)-C(35)	1.384(6)
C(34)-C(39)	1.396(6)
C(34)-C(33)	1.457(6)
C(14)-C(15)	1.376(6)
C(14)-C(19)	1.393(6)

C(14)-C(13)	1.484(6)
C(10)-C(11)	1.350(6)
C(10)-C(9)	1.471(6)
C(28)-C(29)	1.354(6)
C(28)-C(27)	1.429(6)
C(28)-H(28)	0.9300
C(27)-C(26)	1.385(6)
C(27)-C(22)	1.392(6)
C(22)-C(23)	1.365(6)
C(7)-C(2)	1.377(6)
C(7)-C(6)	1.396(6)
C(7)-C(8)	1.412(6)
C(29)-C(21)	1.473(6)
C(15)-C(16)	1.395(6)
C(15)-H(15)	0.9300
C(2)-C(3)	1.370(6)
C(35)-C(36)	1.379(6)
C(35)-H(35)	0.9300
C(8)-C(9)	1.351(6)
C(8)-H(8)	0.9300
C(38)-C(39)	1.380(6)
C(38)-H(38)	0.9300
C(33)-H(33)	0.9300
C(39)-H(39)	0.9300
C(9)-C(1)	1.459(6)
C(18)-C(19)	1.383(6)
C(18)-H(18)	0.9300
C(36)-H(36)	0.9300
C(23)-C(24)	1.357(6)
C(23)-H(23)	0.9300
C(13)-H(13)	0.9300
C(16)-H(16)	0.9300
C(19)-H(19)	0.9300
C(3)-C(4)	1.366(7)
C(3)-H(3)	0.9300
C(24)-C(25)	1.376(7)
C(24)-H(24)	0.9300
C(31)-H(31)	0.9300
C(6)-C(5)	1.371(6)

C(6)-H(6)	0.9300
C(26)-C(25)	1.382(6)
C(26)-H(26)	0.9300
C(4)-C(5)	1.383(7)
C(4)-H(4)	0.9300
C(25)-H(25)	0.9300
C(5)-H(5A)	0.9300
C(11)-H(11)	0.9300
C(40)-H(40A)	0.9600
C(40)-H(40B)	0.9600
C(40)-H(40C)	0.9600
C(20)-H(20A)	0.9600
C(20)-H(20B)	0.9600
C(20)-H(20C)	0.9600
C(31)-S(2)-C(32)	88.0(2)
C(11)-S(1)-C(12)	87.7(3)
C(1)-O(1)-C(2)	122.5(4)
C(21)-O(4)-C(22)	122.2(4)
C(37)-O(6)-C(40)	117.1(4)
C(17)-O(3)-C(20)	117.6(4)
C(12)-N(1)-C(10)	108.1(4)
C(13)-N(3)-N(2)	116.2(4)
C(32)-N(4)-C(30)	109.7(4)
C(33)-N(6)-N(5)	115.7(4)
C(32)-N(5)-N(6)	116.5(4)
C(32)-N(5)-H(5)	121.7
N(6)-N(5)-H(5)	121.7
C(12)-N(2)-N(3)	117.2(4)
C(12)-N(2)-H(2)	121.4
N(3)-N(2)-H(2)	121.4
N(4)-C(32)-N(5)	123.9(4)
N(4)-C(32)-S(2)	115.9(4)
N(5)-C(32)-S(2)	120.2(4)
C(31)-C(30)-N(4)	114.2(4)
C(31)-C(30)-C(29)	128.1(4)
N(4)-C(30)-C(29)	117.7(4)
O(6)-C(37)-C(38)	114.9(4)
O(6)-C(37)-C(36)	125.4(5)
C(38)-C(37)-C(36)	119.7(5)

C(16)-C(17)-C(18)	120.7(5)
C(16)-C(17)-O(3)	124.6(5)
C(18)-C(17)-O(3)	114.7(5)
C(35)-C(34)-C(39)	117.5(4)
C(35)-C(34)-C(33)	122.4(4)
C(39)-C(34)-C(33)	120.1(4)
C(15)-C(14)-C(19)	118.2(5)
C(15)-C(14)-C(13)	121.5(4)
C(19)-C(14)-C(13)	120.3(5)
C(11)-C(10)-N(1)	115.4(5)
C(11)-C(10)-C(9)	127.5(5)
N(1)-C(10)-C(9)	117.0(4)
C(29)-C(28)-C(27)	123.0(4)
C(29)-C(28)-H(28)	118.5
C(27)-C(28)-H(28)	118.5
C(26)-C(27)-C(22)	117.1(5)
C(26)-C(27)-C(28)	125.1(4)
C(22)-C(27)-C(28)	117.7(4)
C(23)-C(22)-O(4)	116.0(5)
C(23)-C(22)-C(27)	123.2(5)
O(4)-C(22)-C(27)	120.8(4)
C(2)-C(7)-C(6)	115.4(5)
C(2)-C(7)-C(8)	118.5(5)
C(6)-C(7)-C(8)	126.1(5)
C(28)-C(29)-C(30)	123.0(4)
C(28)-C(29)-C(21)	118.3(4)
C(30)-C(29)-C(21)	118.7(4)
N(1)-C(12)-N(2)	123.1(5)
N(1)-C(12)-S(1)	117.2(4)
N(2)-C(12)-S(1)	119.7(4)
C(14)-C(15)-C(16)	120.5(5)
C(14)-C(15)-H(15)	119.8
C(16)-C(15)-H(15)	119.8
C(3)-C(2)-C(7)	124.7(5)
C(3)-C(2)-O(1)	116.0(5)
C(7)-C(2)-O(1)	119.3(5)
C(36)-C(35)-C(34)	122.0(5)
C(36)-C(35)-H(35)	119.0
C(34)-C(35)-H(35)	119.0

C(9)-C(8)-C(7)	123.5(5)
C(9)-C(8)-H(8)	118.3
C(7)-C(8)-H(8)	118.3
C(37)-C(38)-C(39)	120.4(5)
C(37)-C(38)-H(38)	119.8
C(39)-C(38)-H(38)	119.8
N(6)-C(33)-C(34)	121.5(4)
N(6)-C(33)-H(33)	119.3
C(34)-C(33)-H(33)	119.3
C(38)-C(39)-C(34)	120.8(4)
C(38)-C(39)-H(39)	119.6
C(34)-C(39)-H(39)	119.6
C(8)-C(9)-C(1)	117.5(5)
C(8)-C(9)-C(10)	122.9(4)
C(1)-C(9)-C(10)	119.6(4)
C(17)-C(18)-C(19)	119.3(5)
C(17)-C(18)-H(18)	120.4
C(19)-C(18)-H(18)	120.4
C(35)-C(36)-C(37)	119.4(5)
C(35)-C(36)-H(36)	120.3
C(37)-C(36)-H(36)	120.3
C(24)-C(23)-C(22)	118.9(5)
C(24)-C(23)-H(23)	120.6
C(22)-C(23)-H(23)	120.6
O(5)-C(21)-O(4)	116.5(4)
O(5)-C(21)-C(29)	125.5(5)
O(4)-C(21)-C(29)	118.0(4)
N(3)-C(13)-C(14)	120.4(5)
N(3)-C(13)-H(13)	119.8
C(14)-C(13)-H(13)	119.8
O(2)-C(1)-O(1)	115.6(4)
O(2)-C(1)-C(9)	125.6(5)
O(1)-C(1)-C(9)	118.8(4)
C(17)-C(16)-C(15)	120.0(5)
C(17)-C(16)-H(16)	120.0
C(15)-C(16)-H(16)	120.0
C(18)-C(19)-C(14)	121.4(5)
C(18)-C(19)-H(19)	119.3
C(14)-C(19)-H(19)	119.3



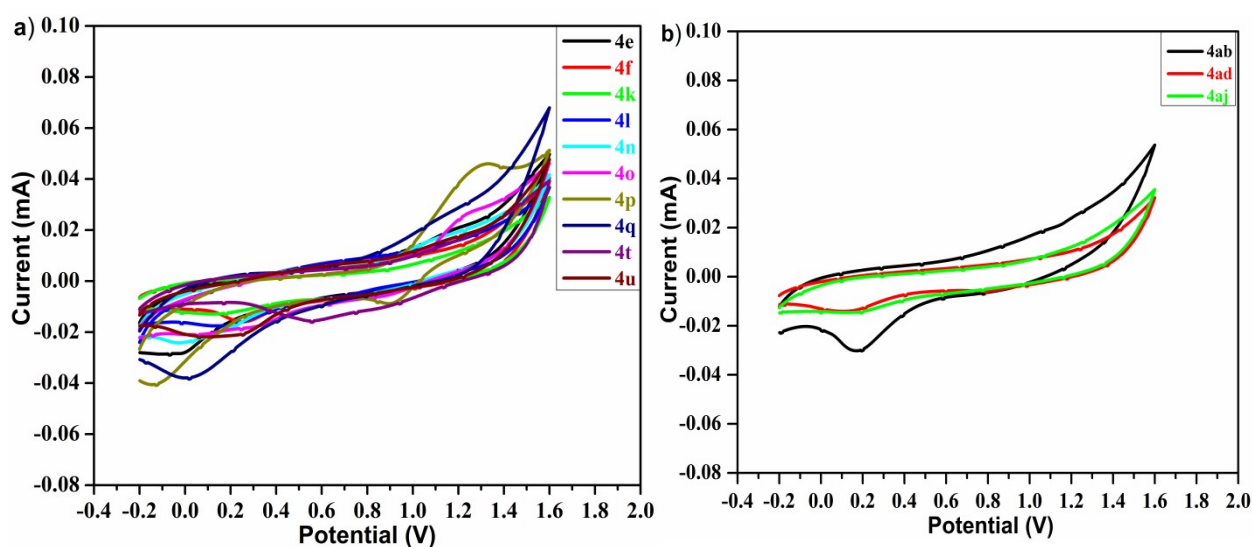
C(4)-C(3)-C(2)	118.2(5)
C(4)-C(3)-H(3)	120.9
C(2)-C(3)-H(3)	120.9
C(23)-C(24)-C(25)	119.7(5)
C(23)-C(24)-H(24)	120.1
C(25)-C(24)-H(24)	120.1
C(30)-C(31)-S(2)	112.2(4)
C(30)-C(31)-H(31)	123.9
S(2)-C(31)-H(31)	123.9
C(5)-C(6)-C(7)	121.3(5)
C(5)-C(6)-H(6)	119.3
C(7)-C(6)-H(6)	119.3
C(25)-C(26)-C(27)	119.4(5)
C(25)-C(26)-H(26)	120.3
C(27)-C(26)-H(26)	120.3
C(3)-C(4)-C(5)	119.8(5)
C(3)-C(4)-H(4)	120.1
C(5)-C(4)-H(4)	120.1
C(24)-C(25)-C(26)	121.6(5)
C(24)-C(25)-H(25)	119.2
C(26)-C(25)-H(25)	119.2
C(6)-C(5)-C(4)	120.6(5)
C(6)-C(5)-H(5A)	119.7
C(4)-C(5)-H(5A)	119.7
C(10)-C(11)-S(1)	111.6(4)
C(10)-C(11)-H(11)	124.2
S(1)-C(11)-H(11)	124.2
O(6)-C(40)-H(40A)	109.5
O(6)-C(40)-H(40B)	109.5
H(40A)-C(40)-H(40B)	109.5
O(6)-C(40)-H(40C)	109.5
H(40A)-C(40)-H(40C)	109.5
H(40B)-C(40)-H(40C)	109.5
O(3)-C(20)-H(20A)	109.5
O(3)-C(20)-H(20B)	109.5
H(20A)-C(20)-H(20B)	109.5
O(3)-C(20)-H(20C)	109.5
H(20A)-C(20)-H(20C)	109.5
H(20B)-C(20)-H(20C)	109.5

**Table S4.** Anisotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for skd19a. The anisotropic displacement factor exponent takes the form:  $-2p^2 [ h^2 a^* 2U^{11} + \dots + 2 h k a^* b^* U^{12} ]$

	U11	U22	U33	U23	U13	U12
S(2)	65(1)	55(1)	89(1)	1(1)	43(1)	9(1)
S(1)	68(1)	64(1)	100(1)	-2(1)	48(1)	-11(1)
O(1)	51(2)	55(2)	87(3)	-6(2)	32(2)	5(2)
O(4)	55(2)	60(2)	86(3)	4(2)	35(2)	-3(2)
O(6)	83(3)	46(2)	110(3)	-1(2)	45(2)	3(2)
O(3)	77(3)	43(2)	123(3)	9(2)	44(2)	-1(2)
N(1)	67(3)	42(3)	70(3)	-2(2)	39(2)	5(2)
N(3)	67(3)	51(3)	76(3)	3(2)	38(2)	-2(2)
N(4)	63(3)	45(3)	70(3)	0(2)	34(2)	-1(2)
N(6)	67(3)	43(3)	75(3)	-7(2)	34(2)	5(2)
O(5)	67(3)	74(3)	153(4)	11(2)	64(3)	4(2)
N(5)	63(3)	47(3)	111(4)	-7(2)	46(3)	-2(2)
O(2)	57(2)	79(3)	124(3)	-21(2)	44(2)	-1(2)
N(2)	66(3)	51(3)	110(4)	-1(3)	50(3)	-4(2)
C(32)	60(3)	52(3)	62(3)	1(3)	28(3)	9(3)
C(30)	63(3)	44(3)	55(3)	4(2)	32(3)	8(3)
C(37)	65(4)	38(3)	67(3)	-2(2)	33(3)	-2(2)
C(17)	54(3)	59(4)	60(3)	7(3)	25(3)	3(3)
C(34)	56(3)	45(3)	65(3)	-2(2)	33(3)	-2(2)
C(14)	59(3)	57(3)	64(3)	5(3)	31(3)	9(3)
C(10)	63(3)	51(3)	64(3)	-2(3)	34(3)	-4(3)
C(28)	53(3)	53(3)	62(3)	2(2)	28(3)	2(2)
C(27)	55(3)	46(3)	54(3)	3(2)	24(3)	-1(2)
C(22)	62(3)	51(3)	62(3)	2(2)	32(3)	-1(3)
C(7)	67(4)	50(3)	64(3)	-2(3)	31(3)	5(3)
C(29)	57(3)	44(3)	65(3)	4(2)	34(3)	-2(2)
C(12)	76(4)	61(4)	72(4)	0(3)	46(3)	-6(3)
C(15)	59(3)	54(3)	94(4)	6(3)	44(3)	10(3)
C(2)	81(4)	48(3)	58(3)	-3(3)	34(3)	0(3)
C(35)	56(3)	57(3)	73(4)	-9(3)	28(3)	-10(3)
C(8)	59(3)	56(3)	66(3)	-2(3)	32(3)	4(3)
C(38)	69(4)	51(3)	87(4)	-9(3)	38(3)	-13(3)
C(33)	59(3)	53(3)	74(4)	-6(3)	30(3)	4(3)
C(39)	59(3)	51(3)	82(4)	-1(3)	39(3)	-1(3)

C(9)	52(3)	64(4)	52(3)	-6(2)	25(3)	2(3)
C(18)	60(3)	62(4)	82(4)	9(3)	31(3)	21(3)
C(36)	54(3)	50(3)	82(4)	-6(3)	28(3)	-6(3)
C(23)	76(4)	47(4)	82(4)	4(3)	30(3)	-1(3)
C(21)	60(4)	58(4)	84(4)	5(3)	43(3)	-2(3)
C(13)	69(4)	57(4)	84(4)	2(3)	38(3)	-8(3)
C(1)	64(4)	49(4)	76(4)	-7(3)	36(3)	3(3)
C(16)	62(3)	64(4)	84(4)	6(3)	39(3)	6(3)
C(19)	48(3)	68(4)	85(4)	9(3)	30(3)	7(3)
C(3)	79(4)	60(4)	85(4)	-5(3)	40(3)	10(3)
C(24)	94(5)	39(3)	83(4)	0(3)	34(4)	0(3)
C(31)	61(3)	45(3)	81(4)	1(3)	38(3)	0(2)
C(6)	74(4)	59(4)	86(4)	1(3)	39(3)	-2(3)
C(26)	58(3)	58(4)	99(4)	7(3)	35(3)	2(3)
C(4)	86(5)	58(4)	90(4)	2(3)	43(4)	2(3)
C(25)	77(4)	58(4)	93(4)	9(3)	35(4)	14(3)
C(5)	80(4)	58(4)	111(5)	2(3)	52(4)	-11(3)
C(11)	59(3)	64(4)	93(4)	-5(3)	43(3)	-6(3)
C(40)	90(5)	52(4)	131(5)	13(3)	52(4)	18(3)
C(20)	84(5)	69(4)	156(6)	-18(4)	52(4)	-18(3)

### Cyclic voltammograms of HTC of aromatic aldehydes/ketones



**Figure. S4** a) Cyclic voltammograms of HTCs of aromatic aldehydes (4e, 4f, 4k, 4l, 4n, 4o, 4p, 4q, 4t & 4u) in  $\text{CHCl}_3$ ; b) Cyclic voltammograms of HTCs of aromatic ketones (4ab, 4ad, & 4aj) in  $\text{CHCl}_3$  (0.1 M  $\text{Bu}_4\text{NPF}_6$  in  $\text{CHCl}_3$ ) at a scan rate of 100 mV/s.