

## SUPPLEMENTARY SECTION-1

### The Synthesis of New 8-Imino-1-one Acridine Derivatives Catalyzed by Calix[4]arene Monoacid Core

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## Experimental Section

**1. General methods:**  $^1\text{H}$  and  $^{13}\text{C}$  spectra were obtained on Bruker 300 MHz instrument at 300 MHz and 75 MHz respectively. DEPTQ-135 experiments were performed on Bruker 300 MHz instrument at 75 MHz. Chemical shifts are reported in parts per million (ppm) downfield from an internal TMS (tetramethylsilane) reference. Coupling constants ( $J$ ) are reported in hertz (Hz), and spin multiplicities are represented by the symbols s (singlet), br s (broad singlet), d (doublet), t (triplet), q (quartet) and m (multiplet). IR spectra were recorded on a Perkin Elmer Spectrophotometer RX / FT- IR system. Band positions are reported in reciprocal centimeters ( $\text{cm}^{-1}$ ). The CHN analyses were carried out on a 2400 Series II CHNS Analyzer, Perkin Elmer USA. Melting points were determined on an electrical melting point apparatus with an open capillary. The progress of the reaction was checked by TLC using 300-400 mesh silica gel. All the available reagents were purchased from commercial sources and used without purification. All the solvents used during reactions were distilled for purity.

### 2. Synthesis of *p*-*tert*-butylcalix[4]arene:

*p*-*tert*-Butylcalix[4]arene was synthesized from the mixture of *p*-*tert*-butylphenol (2 g, 13.32 mmol), 37% formaldehyde solution (1.24 mL, 16.6 mmol of HCHO), and NaOH (240 mg, 0.6 mmol) according to our reported procedure.<sup>6c</sup>

### 3. Synthesis of 25, 26, 27-trihydroxy-28-(4-carboxy-1-butoxy)-*p*-*tert*-butylcalix[4]arene (C4V1):

*p*-*tert*-Butylcalix[4]arene (200mg, 0.3 mmol) was mixed with NaH (120 mg, 5 mmol) in a 25 ml round bottom flask, 10 ml of dry DMF was added to it. The mixture was allowed to stir for 30 mins at room temperature (25-30 °C). 5-Bromoaleric acid (217.2 mg, 1.2 mmol) was diluted with dry DMF (2 ml). Then it was added drop wise (1 drop per 5 sec) maintaining the temperature at 20-25 °C. After the complete addition the reaction mixture was stirred for 4 h. As the reaction progressed, the suspended particles changed into a form of viscous mass. The disappearance of starting *p*-*tert*-Butylcalix[4]arene was checked by TLC. After complete conversion the reaction mixture was placed in an ice bath and was quenched via acidification using 5% HCl solution. Then 20 ml of chilled water was added to it and we got the crude product by filtration. 202.2 mg (90%) pure product, color less solid was gained via column chromatography at 15% ethyl acetate in petroleum ether as eluent. mp. 298 °C; IR (KBr,  $\nu \text{ cm}^{-1}$ ): 3350, 2961, 2872, 1710, 1486, 1362, 1201, 1123.  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  10.28-10.11 (m, 1H, -OH), 9.50 (s, 2H, -OH), 7.01-6.91 (m, 8H, Ar-H), 4.27-4.07 (m, 6H, -CH<sub>2</sub>), 3.38-3.34 (m, 4H, -CH<sub>2</sub>), 2.53(brs, 2H, -CH<sub>2</sub>), 2.14 (brs, 2H, -CH<sub>2</sub>), 2.14 (brs, 2H, -CH<sub>2</sub>), 1.18-1.14 (m, 36H, -CH<sub>3</sub>);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  177.9, 149.2, 148.4, 148.2, 147.7, 143.6, 143.2, 133.4, 128.3, 128.0, 127.7, 127.6, 126.5, 125.9, 125.8, 125.7, 114.7, 53.4, 34.2, 34.0, 33.9, 33.5, 33.0, 32.2, 31.9, 31.5, 31.2, 29.7, 29.2, 22.7, 21.2; HRMS (ESI-TOF) m/z: [M + Na]<sup>+</sup> Calcd for [C<sub>49</sub>H<sub>64</sub>O<sub>6</sub>Na]: 771.4595. Found 771.4593. Anal. calcd. for C<sub>49</sub>H<sub>64</sub>O<sub>6</sub>; C: 78.57; H: 8.61. Found: C: 78.59; H: 8.60.<sup>6b</sup>

### 4. Synthesis of 25, 27-dihydroxy-26, 28-bis(4-carboxy-1-butoxy)-*p*-*tert*-butylcalix[4]arene (C4V2):

C4V2 was synthesized from *p*-*tert*-Butylcalix[4]arene with the modified (slight vigorous) procedure than the C4V1. Addition of higher amount (double equivalent) of 5-bromoaleric acid (434.4 mg, 2.4 mmol) at

a higher temperature i.e., 50 °C produced the di-acid incorporated C4V2. 237 mg (93%) pure product, white solid was gained via column chromatography at 45% ethyl acetate in petroleum ether as eluent. IR (KBr,  $\nu$  cm<sup>-1</sup>): 3355, 2959, 2864, 1711, 1485, 1358, 1220, 1110. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.95 (s, 2H, -OH), 6.95-6.85 (m, 8H, Ar-H), 4.40 (d,  $J$  = 12.3 Hz, 1H, -CH<sub>2</sub>), 4.21 (d,  $J$  = 12.9 Hz, 3H, -CH<sub>2</sub>), 4.03-4.01 (m, 2H, -CH<sub>2</sub>), 3.88-3.83 (m, 2H, -CH<sub>2</sub>), 3.31-3.27 (m, 4H, -CH<sub>2</sub>), 2.48 (brs, 4H, -CH<sub>2</sub>), 2.07 (brs, 4H, -CH<sub>2</sub>), 1.86-1.83 (m, 4H, -CH<sub>2</sub>), 1.15-1.05 (m, 36H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  179.4, 151.3, 148.9, 146.5, 142.6, 133.8, 132.9, 129.0, 128.2, 126.0, 125.4, 125.2, 76.1, 34.0, 33.9, 32.7, 32.6, 31.5, 31.2, 31.0, 30.5, 29.7, 29.4, 21.3; HRMS (ESI-TOF) m/z: [M + Na]<sup>+</sup> Calcd for [C<sub>54</sub>H<sub>72</sub>O<sub>8</sub>Na]: 871.5119. Found 871.5121. Anal. calcd. for C<sub>54</sub>H<sub>72</sub>O<sub>8</sub>; C: 76.38; H: 8.55. Found: C: 76.35; H: 8.53.

## 5. General procedure for the synthesis of 8-imino-1-one acridine derivatives (all entries of Scheme 3):

37.5 mg (0.05 mmol) **C4V1** was added in 2 ml water and stirred for 5 min. A mixture of enaminoketone (2 mmol) and aldehyde (1 mmol) was added to that solution and then it was again stirred at 100 °C for 4-5 h. After that, the reaction mixture was filtered and the pure product was separated out by preparative TLC from the crude.

## 6. Spectral and Analytical Data of the products:

**9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-p-tolyl-8-p-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4a):** Yellow solid (74%), mp. 165 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2955, 2927, 2868, 1638, 1599, 1578, 1511, 1367, 1220, 1014, 843; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.49 (d,  $J$  = 7.4 Hz, 2H, Ar-H), 7.34 (d,  $J$  = 6.6 Hz, 2H, Ar-H), 7.27 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 7.12 (d,  $J$  = 7.2 Hz, 2H, Ar-H), 7.06 (d,  $J$  = 6.6 Hz, 2H, Ar-H), 6.46 (d,  $J$  = 7.2 Hz, 2H, Ar-H), 5.62 (s, 1H, Benzylic-CH), 2.48(s, 3H, -CH<sub>3</sub>), 2.31 (s, 3H, -CH<sub>3</sub>), 2.23-1.73 (m, 8H, -CH<sub>2</sub>), 0.98 (s, 3H, -CH<sub>3</sub>), 0.86 (s, 3H, -CH<sub>3</sub>), 0.83 (s, 3H, -CH<sub>3</sub>), 0.72 (s, 3H, -CH<sub>3</sub>). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.7, 162.8, 150.8, 149.5, 145.7, 141.9, 139.0, 136.6, 131.3, 130.7, 130.4, 129.7, 129.1, 127.5, 119.3, 114.4, 112.5, 50.2, 41.7, 41.6, 40.6, 33.2, 32.2, 31.2, 29.8, 29.4, 26.5, 21.2, 20.7; HRMS (ESI-TOF) m/z: [M + H]<sup>+</sup> Calcd for [C<sub>37</sub>H<sub>40</sub>ClN<sub>2</sub>O]: 563.2824. Found 563.2828. Anal. calcd. for C<sub>37</sub>H<sub>39</sub>ClN<sub>2</sub>O; C: 78.91; H: 6.98; N: 4.97. Found: C: 78.95; H: 5.01; N: 4.95.

**9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-m-tolyl-8-m-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4b):** Yellow solid (73%), mp. 186 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2953, 2927, 1638, 1596, 1575, 1370, 1227, 1010; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.34-7.21 (m, 6H, Ar-H), 7.02 (t,  $J$  = 8.1 Hz, 1H, Ar-H), 6.93 (d,  $J$  = 6.3 Hz, 2H, Ar-H), 6.70 (d,  $J$  = 7.2 Hz, 2H, Ar-H), 6.27-6.21 (m, 2H, Ar-H), 5.49 (s, 1H, Benzylic-CH), 2.39 (s, 3H, -CH<sub>3</sub>), 2.20 (s, 3H, -CH<sub>3</sub>), 2.17-1.61 (m, 8H, -CH<sub>2</sub>), 0.87 (s, 3H, -CH<sub>3</sub>), 0.76 (s, 3H, -CH<sub>3</sub>), 0.72 (s, 3H, -CH<sub>3</sub>), 0.62 (s, 3H, -CH<sub>3</sub>). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.6, 162.6, 152.1, 150.6, 146.1, 142.0, 140.0, 139.2, 138.2, 130.5, 130.1, 129.8, 129.5, 128.3, 122.8, 120.0, 119.0, 116.3, 114.2, 112.4, 50.2, 41.7, 40.7, 33.3, 32.2, 31.3, 29.8, 29.4, 26.5, 21.4; HRMS (ESI-TOF) m/z: [M + H]<sup>+</sup> Calcd for [C<sub>37</sub>H<sub>40</sub>BrN<sub>2</sub>O]: 607.2319. Found 607.2316. Anal. calcd. for C<sub>37</sub>H<sub>39</sub>BrN<sub>2</sub>O; C: 73.14; H: 6.47; N: 4.61. Found: C: 73.11; H: 6.49; N: 4.59.

**3,3,6,6-Tetramethyl-10-m-tolyl-9-p-tolyl-8-m-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4c):** Yellow solid (66%), mp. 156 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2957, 2930, 2865, 1622, 1599, 1554, 1349, 1195, 1011; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.34-7.10 (m, 4H, Ar-H), 7.00-6.94 (m, 5H, Ar-H), 6.66 (d, J = 6.3 Hz, 1H, Ar-H), 6.29-6.23 (m, 2H, Ar-H), 5.53 (s, 1H, Benzylic-CH), 2.37 (s, 3H, -CH<sub>3</sub>), 2.17 (s, 6H, -CH<sub>3</sub>), 2.14-1.61 (m, 8H, -CH<sub>2</sub>), 0.86 (s, 3H, -CH<sub>3</sub>), 0.73 (s, 6H, -CH<sub>3</sub>), 0.62 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 162.7, 152.2, 150.6, 143.8, 141.5, 139.4, 138.0, 134.4, 129.6, 128.2, 128.0, 122.7, 120.1, 116.4, 114.8, 112.9, 50.3, 41.6, 41.6, 40.8, 32.8, 32.2, 31.3, 29.8, 29.3, 26.7, 26.6, 21.3, 21.0; Anal. calcd. for C<sub>38</sub>H<sub>42</sub>N<sub>2</sub>O; C: 84.09; H: 7.80; N: 5.16. Found: C: 84.13; H: 7.81; N: 5.18.

**10-(4-Bromophenyl)-8-(4-bromophenylimino)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4d):** Yellow solid (77%), mp. 220 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2944, 2915, 1601, 1534, 1513, 1349, 1198, 1004; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.59 (d, J = 6.9 Hz, 2H, Ar-H), 7.32 (d, J = 6.6 Hz, 2H, Ar-H), 7.23 (d, J = 7.2 Hz, 2H, Ar-H), 7.12 (d, J = 7.2 Hz, 2H, Ar-H), 7.04 (d, J = 7.2 Hz, 2H, Ar-H), 6.30 (d, J = 7.2 Hz, 2H, Ar-H), 5.44 (s, 1H, Benzylic-CH), 2.17-1.85 (m, 6H, -CH<sub>2</sub>), 1.78-1.60 (m, 2H, -CH<sub>2</sub>), 0.88 (s, 3H, -CH<sub>3</sub>), 0.77 (s, 3H, -CH<sub>3</sub>), 0.72 (s, 3H, -CH<sub>3</sub>), 0.62 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 163.4, 150.9, 149.9, 145.1, 141.9, 138.3, 133.2, 131.5, 131.4, 131.0, 129.5, 127.7, 123.2, 121.2, 115.0, 114.5, 113.1, 50.1, 41.8, 40.8, 33.2, 32.3, 31.4, 29.8, 29.4, 26.6; Anal. calcd. for C<sub>35</sub>H<sub>33</sub>Br<sub>2</sub>ClN<sub>2</sub>O; C: 60.67; H: 4.80; N: 4.04. Found: C: 60.69; H: 4.79; N: 4.02.

**10-(4-Bromophenyl)-8-(4-bromophenylimino)-3,3,6,6-tetramethyl-9-p-tolyl-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4e):** Yellow solid (69%), mp. 208 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2956, 2925, 2867, 1642, 1618, 1479, 1369, 1220, 1010; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.69 (d, J = 8.1 Hz, 2H, Ar-H), 7.39-7.32 (m, 4H, Ar-H), 7.16 (d, J = 8.4 Hz, 2H, Ar-H), 7.07 (d, J = 7.2 Hz, 2H, Ar-H), 6.44 (d, J = 8.1 Hz, 2H, Ar-H), 5.58 (s, 1H, Benzylic-CH), 2.30 (s, 3H, -CH<sub>3</sub>), 2.23-1.72 (m, 8H, -CH<sub>2</sub>), 0.99 (s, 3H, -CH<sub>3</sub>), 0.88 (s, 3H, -CH<sub>3</sub>), 0.84 (s, 3H, -CH<sub>3</sub>), 0.74 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 163.4, 151.1, 149.7, 143.4, 141.6, 138.5, 134.7, 133.0, 131.4, 128.3, 127.8, 122.9, 121.3, 114.9, 114.7, 113.5, 50.1, 41.7, 40.8, 32.8, 32.2, 31.3, 29.8, 29.3, 26.7, 26.6, 21.0; Anal. calcd. for C<sub>36</sub>H<sub>36</sub>Br<sub>2</sub>N<sub>2</sub>O; C: 64.30; H: 5.40; N: 4.17. Found: C: 64.33; H: 5.38; N: 4.15.

**9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-m-tolyl-8-m-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4f):** Yellow solid (74%), mp. 162-164 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2952, 2924, 2851, 1632, 1601, 1588, 1247, 1160, 1014; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.40-7.32 (m, 3H, Ar-H), 7.25-7.13 (m, 3H, Ar-H), 7.04 (t, J = 7.6 Hz, 1H, Ar-H), 6.95 (d, J = 6.6 Hz, 2H, Ar-H), 6.73 (d, J = 7.5 Hz, 1H, Ar-H), 6.28-6.23 (m, 2H, Ar-H), 5.50 (s, 1H, Benzylic-CH), 2.41 (s, 3H, -CH<sub>3</sub>), 2.22 (s, 3H, -CH<sub>3</sub>), 2.19-1.62 (m, 8H, -CH<sub>2</sub>), 0.80 (s, 3H, -CH<sub>3</sub>), 0.78 (s, 3H, -CH<sub>3</sub>), 0.74 (s, 3H, -CH<sub>3</sub>), 0.64 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 162.7, 152.2, 150.6, 145.6, 141.9, 139.3, 138.3, 130.8, 129.8, 129.7, 128.4, 127.6, 122.9, 120.1, 116.4, 114.4, 112.6, 50.3, 41.8, 40.8, 33.2, 32.3, 31.3, 29.8, 29.5, 26.9, 26.6, 21.4; Anal. calcd. for C<sub>37</sub>H<sub>39</sub>ClN<sub>2</sub>O; C: 78.91; H: 6.98; N: 4.97. Found: C: 78.95; H: 6.95; N: 4.99.

**9-(4-Bromophenyl)-10-(4-methoxyphenyl)-8-(4-methoxyphenylimino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4g):** Yellow solid (65%), mp. 151-152 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2957, 2928, 2859, 1638, 1609, 1425, 1369, 1202, 1013; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.33-7.25

(m, 4H, Ar-H), 7.06-6.92 (m, 4H, Ar-H), 6.71 (d,  $J$  = 8.4 Hz, 2H, Ar-H), 6.39 (d,  $J$  = 8.4 Hz, 2H, Ar-H), 5.48 (s, 1H, Benzylic-CH), 3.81 (s, 3H, -OCH<sub>3</sub>), 3.68 (s, 3H, -OCH<sub>3</sub>), 2.20-1.68 (m, 8H, -CH<sub>2</sub>), 0.87 (s, 3H, -CH<sub>3</sub>), 0.77 (s, 3H, -CH<sub>3</sub>), 0.72 (s, 3H, -CH<sub>3</sub>), 0.61 (s, 3H, -CH<sub>3</sub>); Anal. calcd. for C<sub>37</sub>H<sub>39</sub>BrN<sub>2</sub>O<sub>3</sub>; C: 69.48; H: 6.15; N: 4.38. Found: C: 69.45; H: 6.16; N: 4.40.

**9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-p-tolyl-8-p-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4h):** Yellow solid (73%), mp. 184 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2954, 2925, 2855, 1638, 1601, 1576, 1367, 1219, 1009; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.34-7.22 (m, 6H, Ar-H), 7.02-6.94 (m, 4H, Ar-H), 6.35 (d,  $J$  = 7.1 Hz, 2H, Ar-H), 5.49 (s, 1H, Benzylic-CH), 2.39 (s, 3H, -CH<sub>3</sub>), 2.21 (s, 3H, -CH<sub>3</sub>), 2.17-1.60 (m, 8H, -CH<sub>2</sub>), 0.87 (s, 3H, -CH<sub>3</sub>), 0.76 (s, 3H, -CH<sub>3</sub>), 0.72 (s, 3H, -CH<sub>3</sub>), 0.61 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 162.8, 150.8, 149.5, 146.2, 142.0, 139.1, 136.8, 131.9, 131.4, 130.6, 130.2, 129.2, 119.4, 119.1, 114.5, 112.6, 50.3, 41.8, 41.7, 40.7, 33.4, 32.3, 31.4, 29.9, 29.5, 26.9, 26.6, 21.3, 20.8; Anal. calcd. for C<sub>37</sub>H<sub>39</sub>BrN<sub>2</sub>O; C: 73.14; H: 6.47; N: 4.61. Found: C: 73.17; H: 6.45; N: 4.60.

**9-(4-Chlorophenyl)-10-(4-methoxyphenyl)-8-(4-methoxyphenylimino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4i):** Yellow solid (74%), mp. 146 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2961, 2929, 1654, 1610, 1501, 1404, 1196, 1026; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.23-6.82 (m, 9H, Ar-H), 6.58 (brs, 2H, Ar-H), 6.26 (brs, 1H, Ar-H), 5.35-5.01 (m, 1H, Benzylic-CH), 3.69 (s, 3H, -OCH<sub>3</sub>), 3.55 (s, 3H, -OCH<sub>3</sub>), 1.92-1.48 (m, 8H, -CH<sub>2</sub>), 0.73-0.47 (m, 12H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 163.2, 159.6, 155.2, 151.1, 150.4, 145.7, 145.4, 144.8, 142.1, 132.0, 131.4, 130.8, 129.7, 129.2, 128.1, 127.6, 120.6, 114.9, 114.6, 114.2, 113.9, 112.6, 55.5, 55.4, 50.3, 50.1, 41.8, 40.7, 33.2, 32.3, 31.3, 29.9, 29.7, 26.7, 26.6; Anal. calcd. for C<sub>37</sub>H<sub>39</sub>ClN<sub>2</sub>O<sub>3</sub>; C: 74.67; H: 6.60; N: 4.71. Found: C: 74.64; H: 6.61; N: 4.68.

**9,10-Bis-(4-chlorophenyl)-8-(4-chlorophenylimino)-3,3,6,6-tetramethyl-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4j):** Yellow solid (77%), mp. 224 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2955, 2927, 2870, 1644, 1618, 1471, 1379, 1240, 1013; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.51 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 7.39 (d,  $J$  = 7.2 Hz, 2H, Ar-H), 7.20-7.16 (m, 6H, Ar-H), 6.42 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 5.51 (s, 1H, Benzylic-CH), 2.24-1.67 (m, 8H, -CH<sub>2</sub>), 0.96 (s, 3H, -CH<sub>3</sub>), 0.84 (s, 3H, -CH<sub>3</sub>), 0.79 (s, 3H, -CH<sub>3</sub>), 0.69 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.7, 163.4, 150.5, 149.9, 145.2, 142.0, 137.8, 135.1, 131.0, 130.2, 129.5, 128.6, 127.7, 127.4, 120.8, 114.5, 113.1, 50.2, 41.8, 40.8, 33.2, 32.3, 3.4, 29.8, 29.4, 26.6; Anal. calcd. for C<sub>35</sub>H<sub>33</sub>Cl<sub>3</sub>N<sub>2</sub>O; C: 69.60; H: 5.51; N: 4.64. Found: C: 69.65; H: 5.53; N: 4.66.

**3,3,6,6-Tetramethyl-10-(3-nitrophenyl)-8-(3-nitrophenylimino)-9-p-tolyl-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4k):** Yellow solid (84%), mp. 236 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2959, 2925, 2845, 1642, 1601, 1573, 1369, 1213, 1008; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.43 (q,  $J$  = 7.5 Hz,  $J$  = 7.9 Hz, 1H, Ar-H), 8.17 (s, 1H, Ar-H), 7.85-7.79 (m, 2H, Ar-H), 7.68 (d,  $J$  = 6.9 Hz, 1H, Ar-H), 7.42-7.35 (m, 4H, Ar-H), 7.09 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 6.89-6.87 (m, 1H, Ar-H), 5.55 (s, 1H, Benzylic-CH), 2.31 (s, 3H, -CH<sub>3</sub>), 2.24-1.97 (m, 6H, -CH<sub>2</sub>), 1.81-1.69 (m, 2H, -CH<sub>2</sub>), 1.00 (s, 3H, -CH<sub>3</sub>), 0.89 (s, 3H, -CH<sub>3</sub>), 0.84 (s, 3H, -CH<sub>3</sub>), 0.77 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.8, 164.5, 153.1, 148.9, 148.8, 148.6, 142.9, 141.9, 140.7, 136.1, 135.2, 130.9, 129.4, 128.6, 127.8, 126.0, 125.0, 124.2, 117.1, 115.3, 114.4,

114.2, 50.2, 42.0, 41.0, 32.9, 32.5, 31.7, 29.7, 29.3, 26.9, 26.8, 21.0; Anal. calcd. for C<sub>36</sub>H<sub>36</sub>N<sub>4</sub>O<sub>5</sub>; C: 71.50; H: 6.00; N: 9.27. Found: C: 71.55; H: 6.01; N: 9.30.

**9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-(3-nitrophenyl)-8-(3-nitrophenylimino)-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4l):** Yellow solid (86%), mp. 254 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2955, 2924, 1637, 1597, 1573, 1358, 1226, 1013; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.35 (d, J = 8.1 Hz, 1H, Ar-H), 8.07 (s, 1H, Ar-H), 7.77-7.72 (m, 2H, Ar-H), 7.58 (d, J = 7.5 Hz, 1H, Ar-H), 7.34-7.26 (m, 6H, Ar-H), 6.76 (d, J = 7.8 Hz, 1H, Ar-H), 5.44 (s, 1H, Benzylic-CH), 2.21-1.90 (m, 6H, -CH<sub>2</sub>), 1.73-1.60 (m, 2H, -CH<sub>2</sub>), 0.91 (s, 3H, -CH<sub>3</sub>), 0.81 (s, 3H, -CH<sub>3</sub>), 0.74 (s, 3H, -CH<sub>3</sub>), 0.68 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.7, 164.4, 152.8, 149.0, 148.7, 145.1, 142.3, 140.5, 136.0, 131.0, 130.9, 129.9, 129.5, 126.0, 124.9, 124.3, 119.7, 117.3, 114.6, 114.1, 113.9, 50.1, 42.0, 41.0, 33.4, 32.5, 31.6, 29.7, 29.4, 26.7; Anal. calcd. for C<sub>35</sub>H<sub>33</sub>BrN<sub>4</sub>O<sub>5</sub>; C: 62.78; H: 4.97; N: 8.37. Found: C: 62.80; H: 4.99; N: 8.35.

**9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-(3-nitrophenyl)-8-(3-nitrophenylimino)-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4m):** Yellow solid (86%), mp. 248 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2959, 2925, 2856, 1645, 1604, 1582, 1332, 1215, 1010; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.45 (d, J = 7.5 Hz, 1H, Ar-H), 8.16 (d, J = 1.5 Hz, 1H, Ar-H), 7.87-7.81 (m, 2H, Ar-H), 7.65 (d, J = 6.9 Hz, 1H, Ar-H), 7.43-7.37 (m, 4H, Ar-H), 7.28-7.25 (m, 2H, Ar-H), 6.84 (d, J = 7.2 Hz, 1H, Ar-H), 5.55 (s, 1H, Benzylic-CH), 2.30-1.97 (m, 5H, -CH<sub>2</sub>), 1.82-1.68 (m, 3H, -CH<sub>2</sub>), 1.00 (s, 3H, -CH<sub>3</sub>), 0.90 (s, 3H, -CH<sub>3</sub>), 0.84 (s, 3H, -CH<sub>3</sub>), 0.77 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.5, 164.1, 152.5, 148.7, 148.4, 144.3, 141.9, 140.3, 135.7, 131.2, 130.8, 129.2, 127.8, 125.8, 124.6, 124.1, 117.1, 114.5, 113.8, 49.8, 41.8, 40.7, 33.0, 32.3, 31.4, 29.4, 29.1, 26.5; Anal. calcd. for C<sub>35</sub>H<sub>33</sub>ClN<sub>4</sub>O<sub>5</sub>; C: 67.25; H: 5.32; N: 8.96. Found: C: 67.29; H: 5.30; N: 9.00.

**9-(4-Chlorophenyl)-2,2,5,5-tetramethyl-10-p-tolyl-8-p-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4n):** Yellow solid (65%), mp. 162 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2961, 2931, 2863, 1622, 1601, 1565, 1377, 1220, 1011; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): 7.19-6.90 (m, 12H, Ar-H), 5.84 (s, 1H, Benzylic-CH), 2.55 (brs, 2H, -CH<sub>2</sub>), 2.25 (s, 6H, -CH<sub>3</sub>), 1.73 (brs, 4H, -CH<sub>2</sub>), 1.56 (brs, 2H, -CH<sub>2</sub>), 1.21-1.02 (m, 12H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  200.9, 139.8, 136.5, 135.2, 130.1, 129.8, 129.5, 127.9, 127.6, 125.1, 39.4, 35.4, 34.8, 25.7, 25.5, 24.5, 20.9; Anal. calcd. for C<sub>37</sub>H<sub>39</sub>ClN<sub>2</sub>O; C: 78.91; H: 6.98; N: 4.97. Found: C: 78.94; H: 5.01; N: 4.93.

**9-(4-Chlorophenyl)-3,6-diphenyl-10-p-tolyl-8-p-tolylimino-3,4,5,6,7,8,9,10-octahydro-2H-acridin-1-one (4o):** Yellow solid (62%), mp. 246 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2955, 2925, 2860, 1642, 1597, 1567, 1365, 1223, 1023; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.06 (brs, 1H, Ar-H), 7.45-6.93 (m, 20H, Ar-H), 6.54-6.38 (m, 1H, Ar-H), 5.80-5.74 (m, 1H, Benzylic-CH), 3.37-3.11 (m, 2H, Benzylic-CH), 2.78-2.28 (m, 14H, -CH<sub>2</sub> and -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.2, 143.2, 142.6, 131.5, 130.6, 129.8, 129.4, 129.0, 128.8, 128.3, 127.8, 127.0, 126.8, 119.7, 115.3, 114.9, 55.4, 39.6, 39.2, 35.9, 35.2, 21.2; Anal. calcd. for C<sub>45</sub>H<sub>39</sub>ClN<sub>2</sub>O; C: 81.98; H: 5.96; N: 4.25. Found: C: 81.95; H: 5.95; N: 4.26.

**(E)-9-(4-bromophenyl)-3,6-dimethyl-10-p-tolyl-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2H)-one (4p):** Yellow solid (64%), mp. 175-176 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2961, 2930, 2852, 1625, 1599, 1545, 1344, 1263, 1190, 972; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.02 (d, J = 8.4 Hz, 1H, Ar-H), 7.45-7.03 (m, 10H, Ar-H), 6.50 (d, J = 8.1 Hz, 1H, Ar-H), 5.63 (d, J = 17.1 Hz, 1H, Benzylic-CH), 2.46 (s, 3H, -CH<sub>3</sub>), 2.30 (s, 3H, -CH<sub>3</sub>), 2.20-1.73 (m, 10H, -CH<sub>2</sub> and -CH), 0.90 (s, 3H, -CH<sub>3</sub>), 0.89 (s, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.0, 151.8, 149.8, 146.0, 143.9, 139.1, 137.0, 131.7, 131.5, 131.0, 130.4, 129.7, 129.5, 129.2, 128.8, 127.8, 127.7, 127.7, 122.8, 119.8, 119.5, 115.0, 114.4, 113.5, 45.6, 44.7, 36.7, 36.4, 36.1, 35.9, 33.2, 29.7, 29.2, 28.6, 21.2, 21.0, 20.8; Anal. calcd. for C<sub>35</sub>H<sub>35</sub>BrN<sub>2</sub>O; C: 72.53; H: 6.09; N: 4.83. Found: C: 72.59; H: 6.08; N: 4.85.

**(E)-3,6-dimethyl-9-propyl-10-p-tolyl-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2H)-one (4q):** Yellow solid (72%), mp. 136 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2951, 2926, 1648, 1599, 1501, 1377, 1240, 1053; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.17 (d, J = 7.5 Hz, 2H, Ar-H), 7.06-6.95 (m, 4H, Ar-H), 6.51-6.48 (m, 2H, Ar-H), 4.48 (s, 1H, -CH), 2.34 (s, 3H, -CH<sub>3</sub>), 2.28-2.26 (m, 1H, -CH<sub>2</sub> or -CH), 2.20 (s, 3H, -CH<sub>3</sub>), 2.20-1.90 (m, 5H, -CH<sub>2</sub> or -CH), 1.83-1.69 (m, 2H, -CH<sub>2</sub> or -CH), 1.64-1.16 (m, 6H, -CH<sub>2</sub> or -CH), 0.90-0.77 (m, 9H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.8, 196.6, 196.3, 196.2, 164.4, 164.4, 163.7, 152.6, 152.4, 152.4, 152.2, 151.6, 149.8, 149.7, 144.2, 144.0, 142.9, 138.7, 137.0, 136.9, 136.8, 131.2, 130.1, 129.1, 119.8, 119.6, 116.1, 115.4, 115.3, 115.1, 114.7, 114.5, 45.4, 45.2, 44.6, 44.4, 38.4, 37.9, 37.3, 36.4, 36.2, 35.4, 35.2, 28.9, 28.7, 28.6, 28.5, 27.3, 27.0, 26.9, 26.8, 26.6, 26.4, 21.1, 21.0, 21.0, 20.7, 18.9, 18.6, 14.6; Anal. calcd. for C<sub>32</sub>H<sub>38</sub>N<sub>2</sub>O; C: 82.36; H: 8.21; N: 6.00. Found: C: 82.41; H: 8.19; N: 6.02.

**(E)-3,3,6,6-tetramethyl-9-propyl-10-p-tolyl-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2H)-one (4r):** Yellow solid (83%), mp. 142 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2957, 2928, 2870, 1633, 1599, 1575, 1366, 1150, 971; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.28-7.26 (m, 2H, Ar-H), 7.08-7.00 (m, 4H, Ar-H), 6.58-6.56 (m, 2H, Ar-H), 4.53 (d, J = 4.5 Hz, 1H, -CH), 2.44 (s, 3H, -CH<sub>3</sub>), 2.30 (s, 3H, -CH<sub>3</sub>), 2.23 (s, 3H, -CH<sub>2</sub>), 2.18 (s, 1H, -CH<sub>2</sub>), 2.09-1.81 (m, 4H, -CH<sub>2</sub>), 1.66-1.54 (m, 2H, -CH<sub>2</sub>), 1.44-1.39 (m, 2H, -CH<sub>2</sub>), 0.96-0.84 (m, 15H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.3, 163.7, 151.8, 149.9, 142.6, 138.7, 137.0, 131.2, 130.2, 129.4, 129.1, 119.6, 114.5, 112.2, 50.6, 41.7, 41.6, 40.9, 38.0, 32.0, 31.1, 30.2, 29.8, 26.9, 26.5, 26.4, 21.1, 20.7, 19.0, 14.6; Anal. calcd. for C<sub>34</sub>H<sub>42</sub>N<sub>2</sub>O; C: 82.55; H: 8.56; N: 5.66. Found: C: 82.52; H: 8.58; N: 5.67.

**(E)-8-(4-bromophenyl)-4-p-tolyl-7-(p-tolylimino)-2,3,5,6,7,8-hexahydrodicyclopenta[b,e]pyridin-1(4H)-one (4s):** Yellow solid (61%), mp. 162-164 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2951, 2910, 1601, 1544, 1395, 1170, 1020; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.38-7.29 (m, 4H, Ar-H), 7.16-7.06 (m, 8H, Ar-H), 5.21 (s, 1H, Benzylic-CH), 2.86 (brs, 2H, -CH<sub>2</sub>), 2.54 (brs, 4H, -CH<sub>2</sub>), 2.48-2.45 (m, 1H, -CH<sub>2</sub>), 2.39 (s, 6H, -CH<sub>3</sub>), 2.27 (s, 1H, -CH<sub>2</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.3, 166.2, 149.4, 136.8, 134.5, 131.0, 130.1, 129.8, 129.4, 128.7, 127.6, 121.8, 119.7, 116.3, 39.5, 34.8, 24.9, 23.9, 21.2; Anal. calcd. for C<sub>31</sub>H<sub>27</sub>BrN<sub>2</sub>O; C: 71.13; H: 5.20; N: 5.35. Found: C: 71.08; H: 5.19; N: 5.33.

**(E)-10-(4-bromophenyl)-8-(4-bromophenylimino)-9-propyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2H)-one (4t):** Yellow solid (73%), mp. 192-194 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2970, 2934, 2868, 1659, 1611,

1479, 1371, 1225, 1016;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.53 (d,  $J = 8.1$  Hz, 2H, Ar-H), 7.26 (d,  $J = 8.1$  Hz, 2H, Ar-H), 7.00 (d,  $J = 7.8$  Hz, 2H, Ar-H), 6.48 (d,  $J = 8.1$  Hz, 2H, Ar-H), 4.44 (s, 1H, -CH), 2.40-2.05 (m, 4H, -CH<sub>2</sub>), 1.93-1.65 (m, 7H, -CH<sub>2</sub>), 1.51-1.47 (m, 1H, -CH<sub>2</sub>), 1.40-1.36 (m, 2H, -CH<sub>2</sub>), 1.27-1.20 (m, 2H, -CH<sub>2</sub>), 0.82 (t,  $J = 6.8$  Hz, 3H, -CH<sub>3</sub>);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  196.6, 164.7, 152.3, 151.3, 144.1, 138.8, 132.8, 131.5, 131.4, 122.9, 121.7, 115.8, 115.0, 114.0, 38.3, 37.0, 28.3, 28.0, 26.7, 21.6, 21.3, 18.6, 14.6; Anal. calcd. for  $\text{C}_{28}\text{H}_{28}\text{Br}_2\text{N}_2\text{O}$ ; C: 59.17; H: 4.97; N: 4.93. Found: C: 59.20; H: 4.98; N: 4.92.

**(E)-3,3,6,6-tetramethyl-10-(4-nitrophenyl)-8-(4-nitrophenylimino)-9-p-tolyl-3,4,5,6,7,8,9,10-octahydroacridin-1(2H)-one (4u):** Yellow solid (82%), mp. 162-164 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2961, 2921, 2841, 1640, 1601, 1585, 1274, 1137, 1016;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.44 (d,  $J = 8.7$  Hz, 2H, Ar-H), 8.12 (d,  $J = 8.7$  Hz, 2H, Ar-H), 7.48 (d,  $J = 9$  Hz, 2H, Ar-H), 7.32 (d,  $J = 7.2$  Hz, 2H, Ar-H), 7.07 (d,  $J = 7.5$  Hz, 2H, Ar-H), 6.59 (d,  $J = 8.7$  Hz, 2H, Ar-H), 5.50 (s, 1H, Benzylic-CH), 2.30 (s, 3H, -CH<sub>3</sub>), 2.22-1.75 (m, 8H, -CH<sub>2</sub>), 0.98 (s, 3H, -CH<sub>3</sub>), 0.88 (s, 3H, -CH<sub>3</sub>), 0.83 (s, 3H, -CH<sub>3</sub>), 0.76 (s, 3H, -CH<sub>3</sub>); (due to very low solubility  $^{13}\text{C}$  could not be done); Anal. calcd. for  $\text{C}_{36}\text{H}_{36}\text{N}_4\text{O}_5$ ; C: 71.50; H: 6.00; N: 9.27. Found: C: 71.55; H: 6.01; N: 9.30.

**(E)-9-ethyl-3,3,6,6-tetramethyl-10-p-tolyl-8-(p-tolylimino)-3,4,5,6,7,8,9,10-octahydroacridin-1(2H)-one (4v):** Yellow solid (81%), mp. 148 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2955, 2930, 2859, 1635, 1609, 1435, 1360, 1197, 1011;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.17 (d,  $J = 8.1$  Hz, 2H, Ar-H), 6.97-6.91 (m, 4H, Ar-H), 6.47 (d,  $J = 8.1$  Hz, 2H, Ar-H), 4.46 (s, 1H, -CH), 2.24 (s, 3H, -CH<sub>3</sub>), 2.28-2.24 (m, 1H, -CH<sub>2</sub>), 2.20 (s, 3H, -CH<sub>3</sub>), 2.14-2.10 (m, 2H, -CH<sub>2</sub>), 2.01-1.81 (m, 3H, -CH<sub>2</sub>), 1.73-1.53 (m, 2H, -CH<sub>2</sub>), 1.04-0.74 (m, 17H, -CH<sub>2</sub> and -CH<sub>3</sub>);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  196.2, 163.7, 151.9, 149.7, 142.8, 138.6, 136.8, 131.0, 130.1, 129.4, 129.2, 129.0, 119.4, 113.4, 111.2, 50.4, 41.5, 41.4, 40.8, 31.8, 31.0, 30.0, 29.7, 27.8, 27.1, 26.4, 26.2, 21.0, 20.6, 9.8; Anal. calcd. for  $\text{C}_{33}\text{H}_{40}\text{N}_2\text{O}$ ; C: 82.46; H: 8.39; N: 5.83. Found: C: 82.50; H: 8.41; N: 5.82.

**9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3a):** Off white solid (72%), mp. 276 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2971, 2933, 2856, 1640, 1580, 1485, 1360, 1221, 1156, 1015;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.28 (brs, 4H, Ar-H), 7.14 (brs, 2H, Ar-H), 7.03 (brs, 2H, Ar-H), 5.16 (s, 1H, Benzylic-CH), 2.41 (s, 3H, -CH<sub>3</sub>), 2.09-1.98 (m, 6H, -CH<sub>2</sub>), 1.79-1.74 (m, 2H, -CH<sub>2</sub>), 0.86 (s, 6H, -CH<sub>3</sub>), 0.72 (s, 6H, -CH<sub>3</sub>);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  195.7, 150.2, 144.8, 139.6, 136.1, 131.3, 130.7, 129.2, 128.0, 114.0, 50.1, 41.7, 32.4, 32.3, 29.6, 26.6, 21.2; Anal. calcd. for  $\text{C}_{30}\text{H}_{32}\text{ClNO}_2$ ; C: 76.01; H: 6.80; N: 2.95. Found: C: 76.05; H: 6.82; N: 2.96.

**9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-m-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3b):** Off white solid (71%), mp. 238 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2960, 2929, 1639, 1571, 1488, 1357, 1222, 1155, 1010;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.37-7.24 (m, 6H, Ar-H), 6.94 (d,  $J = 6.0$  Hz, 2H, Ar-H), 5.15 (s, 1H, Benzylic-CH), 2.42 (s, 3H, -CH<sub>3</sub>), 2.15-1.98 (m, 6H, -CH<sub>2</sub>), 1.79-1.73 (m, 2H, -CH<sub>2</sub>), 0.87 (s, 6H, -CH<sub>3</sub>), 0.73 (s, 6H, -CH<sub>3</sub>);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  195.7, 150.0, 145.3, 138.6, 131.0, 130.2, 129.6, 119.6, 113.9, 50.1, 41.6, 32.4, 32.3, 29.6, 26.7, 21.4; Anal. calcd. for  $\text{C}_{30}\text{H}_{32}\text{BrNO}_2$ ; C: 69.50; H: 6.22; N: 2.70. Found: C: 69.46; H: 6.21; N: 2.69.

**3,3,6,6-Tetramethyl-10-m-tolyl-9-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3c):** Off white solid (77%), mp. 234 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2962, 2925, 2868, 1642, 1567, 1489, 1361, 1217, 1164, 1010;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.34-7.22 (m, 4H, Ar-H), 6.98-6.92 (m, 4H, Ar-H), 5.15 (s, 1H, Benzylic-CH), 2.39 (s, 3H, - $\text{CH}_3$ ), 2.17 (s, 3H, - $\text{CH}_3$ ), 2.14-1.94 (m, 6H, - $\text{CH}_2$ ), 1.76-1.70 (m, 2H, - $\text{CH}_2$ ), 0.86 (s, 6H, - $\text{CH}_3$ ), 0.72 (s, 6H, - $\text{CH}_3$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  195.8, 149.6, 143.4, 139.1, 135.2, 130.1, 129.6, 128.8, 127.8, 127.7, 114.7, 50.3, 41.8, 32.4, 32.3, 29.7, 26.9, 21.4, 21.0; Anal. calcd. for  $\text{C}_{31}\text{H}_{35}\text{NO}_2$ ; C: 82.08; H: 7.78; N: 3.09. Found: C: 82.11; H: 7.75; N: 3.11.

**10-(4-Bromophenyl)-9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3d):** Off white solid (65%), mp. 294 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2962, 2933, 1644, 1568, 1516, 1358, 1214, 1109;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.67 (d,  $J = 8.4$  Hz, 2H, Ar-H), 7.31 (d,  $J = 8.1$  Hz, 2H, Ar-H), 7.17 (d,  $J = 8.1$  Hz, 2H, Ar-H), 7.09 (d,  $J = 8.1$  Hz, 2H, Ar-H), 5.18 (s, 1H, Benzylic-CH), 2.19-2.00 (m, 6H, - $\text{CH}_2$ ), 1.81-1.75 (m, 2H, - $\text{CH}_2$ ), 0.92 (s, 6H, - $\text{CH}_3$ ), 0.77 (s, 6H, - $\text{CH}_3$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  195.7, 149.4, 144.5, 137.9, 133.5, 131.6, 131.2, 129.2, 128.2, 123.6, 114.4, 50.1, 41.8, 32.4, 29.7, 26.7; Anal. calcd. for  $\text{C}_{29}\text{H}_{29}\text{BrClNO}_2$ ; C: 64.63; H: 5.42; N: 2.60. Found: C: 64.66; H: 5.45; N: 2.62.

**10-(4-Bromophenyl)-3,3,6,6-tetramethyl-9-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3e):** Off white solid (74%), mp. 250 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2954, 2926, 2868, 1639, 1578, 1488, 1362, 1223, 1154, 1011;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.62 (d,  $J = 8.4$  Hz, 2H, Ar-H), 7.21 (d,  $J = 8.1$  Hz, 2H, Ar-H), 7.06 (d,  $J = 8.4$  Hz, 2H, Ar-H), 6.97 (d,  $J = 7.5$  Hz, 2H, Ar-H), 5.14 (s, 1H, Benzylic-CH), 2.18 (s, 3H, - $\text{CH}_3$ ), 2.15-1.95 (m, 6H, - $\text{CH}_2$ ), 1.76-1.70 (m, 2H, - $\text{CH}_2$ ), 0.88 (s, 6H, - $\text{CH}_3$ ), 0.74 (s, 6H, - $\text{CH}_3$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  195.5, 148.7, 142.7, 137.9, 135.1, 133.1, 131.0, 128.5, 127.4, 123.1, 114.7, 49.9, 41.5, 32.2, 31.9, 29.4, 26.6, 20.8; Anal. calcd. for  $\text{C}_{30}\text{H}_{32}\text{BrNO}_2$ ; C: 69.50; H: 6.22; N: 2.70. Found: C: 69.53; H: 6.21; N: 2.71.

**9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-m-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3f):** Off white solid (73%), mp. 266 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2962, 2931, 2869, 1658, 1571, 1510, 1355, 1220, 1133, 989;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.42-7.34 (m, 4H, Ar-H), 7.18 (d,  $J = 7.2$  Hz, 2H, Ar-H), 7.00 (d,  $J = 6.6$  Hz, 2H, Ar-H), 5.22 (s, 1H, Benzylic-CH), 2.47 (s, 3H, - $\text{CH}_3$ ), 2.20-2.04 (m, 6H, - $\text{CH}_2$ ), 1.84-1.77 (m, 2H, - $\text{CH}_2$ ), 0.92 (s, 6H, - $\text{CH}_3$ ), 0.78 (s, 6H, - $\text{CH}_3$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  195.8, 150.1, 114.9, 138.7, 131.4, 130.3, 129.7, 129.3, 128.1, 126.0, 114.1, 50.2, 41.7, 32.4, 32.3, 29.7, 26.7, 21.4; Anal. calcd. for  $\text{C}_{30}\text{H}_{32}\text{ClNO}_2$ ; C: 76.01; H: 6.80; N: 2.95. Found: C: 76.06; H: 6.81; N: 2.96.

**9-(4-Bromophenyl)-10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3g):** Off white solid (77%), mp. 198 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu \text{ cm}^{-1}$ ): 2961, 2875, 1641, 1565, 1510, 1359, 1233, 1122, 1014;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.38-7.27 (m, 4H, Ar-H), 7.10 (d,  $J = 8.7$  Hz, 2H, Ar-H), 7.02 (d,  $J = 6.9$  Hz, 2H, Ar-H), 5.19 (s, 1H, Benzylic-CH), 3.88 (s, 3H, - $\text{OCH}_3$ ), 2.20-2.03 (m, 6H, - $\text{CH}_2$ ), 1.86-1.81 (m, 2H, - $\text{CH}_2$ ), 0.92 (s, 6H, - $\text{CH}_3$ ), 0.78 (s, 6H, - $\text{CH}_3$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  200.4, 164.5, 155.1, 150.0, 135.9, 135.6, 134.3, 124.2, 119.9, 119.6, 118.6, 60.2, 54.7, 46.3,

37.1, 36.69, 34.3, 31.3; Anal. calcd. for  $C_{30}H_{32}BrNO_3$ ; C: 67.41; H: 6.03; N: 2.62. Found: C: 67.44; H: 6.01; N: 2.61.

**9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3h):** Off white solid (72%), mp. 244 °C ( $CDCl_3$ ); IR (KBr,  $\nu cm^{-1}$ ): 2960, 2930, 2872, 1640, 1574, 1511, 1361, 1223, 1141;  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  7.36-7.31 (m, 6H, Ar-H), 7.09 (d,  $J$  = 7.2 Hz, 2H, Ar-H), 5.23 (s, 1H, Benzylic-CH), 2.49 (s, 3H, -CH<sub>3</sub>), 2.23-2.05 (m, 6H, -CH<sub>2</sub>), 1.87-1.81 (m, 2H, -CH<sub>2</sub>), 0.95 (s, 6H, -CH<sub>3</sub>), 0.80 (s, 6H, -CH<sub>3</sub>);  $^{13}C$  NMR (75 MHz,  $CDCl_3$ ):  $\delta$  195.6, 150.1, 145.3, 139.5, 136.1, 131.0, 129.6, 119.6, 114.0, 50.1, 41.7, 32.4, 32.3, 29.6, 26.6, 21.2; Anal. calcd. for  $C_{30}H_{32}BrNO_2$ ; C: 69.50; H: 6.22; N: 2.70. Found: C: 69.47; H: 6.19; N: 2.71.

**9-(4-Chlorophenyl)-10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3i):** Off white solid (73%), mp. 222 °C (EtOAc); IR (KBr,  $\nu cm^{-1}$ ): 2960, 2869, 1638, 1572, 1512, 1360, 1250, 1221, 1143;  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  7.29 (d,  $J$  = 8.1 Hz, 2H, Ar-H), 7.13 (d,  $J$  = 8.4 Hz, 2H, Ar-H), 7.05 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 6.97 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 5.16 (s, 1H, Benzylic-CH), 3.85 (s, 3H, -OCH<sub>3</sub>), 2.10-1.98 (m, 6H, -CH<sub>2</sub>), 1.81-1.75 (m, 2H, -CH<sub>2</sub>), 0.88 (s, 6H, -CH<sub>3</sub>), 0.73 (s, 6H, -CH<sub>3</sub>);  $^{13}C$  NMR (75 MHz,  $CDCl_3$ ):  $\delta$  195.7, 159.8, 150.4, 144.8, 131.4, 131.4, 129.2, 128.1, 114.2, 55.6, 50.1, 41.8, 32.4, 32.3, 29.7, 26.7; Anal. calcd. for  $C_{30}H_{32}ClNO_3$ ; C: 73.53; H: 6.58; N: 2.86. Found: C: 73.57; H: 6.56; N: 2.84.

**9,10-Bis-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3j):** Off white solid (66%), mp. 274 °C ( $CDCl_3$ ); IR (KBr,  $\nu cm^{-1}$ ): 2961, 2870, 1642, 1578, 1523, 1365, 1250, 1173, 1013;  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  7.49-7.47 (m, 2H, Ar-H), 7.27 (d,  $J$  = 5.1 Hz, 2H, Ar-H), 7.19-7.13 (m, 4H, Ar-H), 5.15 (s, 1H, Benzylic-CH), 2.15-1.96 (m, 6H, -CH<sub>2</sub>), 1.77-1.71 (m, 2H, -CH<sub>2</sub>), 0.88 (s, 6H, -CH<sub>3</sub>), 0.74 (s, 6H, -CH<sub>3</sub>);  $^{13}C$  NMR (75 MHz,  $CDCl_3$ ):  $\delta$  195.6, 149.4, 144.5, 137.4, 135.6, 131.6, 130.9, 130.4, 129.2, 128.2, 114.5, 50.1, 41.8, 32.4, 29.7, 26.8; Anal. calcd. for  $C_{29}H_{29}Cl_2NO_2$ ; C: 70.44; H: 5.91; N: 2.83. Found: C: 70.39; H: 5.89; N: 2.85.

**3,3,6,6-Tetramethyl-10-(3-nitrophenyl)-9-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3k):** Yellowish solid (61%), mp. 276 °C ( $CDCl_3$ ); IR (KBr,  $\nu cm^{-1}$ ): 2964, 2936, 2876, 1643, 1363, 1234, 1137, 1010;  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  8.35 (d,  $J$  = 7.5 Hz, 1H, Ar-H), 8.07 (s, 1H, Ar-H), 7.76 (t,  $J$  = 8.0 Hz, 1H, Ar-H), 7.60 (d,  $J$  = 6.9 Hz, 1H, Ar-H), 7.22 (d,  $J$  = 7.8 Hz, 2H, Ar-H), 6.99 (d,  $J$  = 7.5 Hz, 2H, Ar-H), 5.16 (s, 1H, Benzylic-CH), 2.18 (s, 3H, -CH<sub>3</sub>), 2.11-1.97 (m, 6H, -CH<sub>2</sub>), 1.71-1.66 (m, 2H, -CH<sub>2</sub>), 0.88 (s, 6H, -CH<sub>3</sub>), 0.75 (s, 6H, -CH<sub>3</sub>);  $^{13}C$  NMR (75 MHz,  $CDCl_3$ ):  $\delta$  195.6, 149.0, 148.4, 142.7, 140.4, 136.1, 135.6, 131.2, 129.0, 127.7, 124.8, 124.5, 115.4, 50.1, 42.0, 32.6, 32.2, 29.7, 27.0, 21.1; Anal. calcd. for  $C_{30}H_{32}N_2O_4$ ; C: 74.36; H: 6.66; N: 5.78. Found: C: 74.34; H: 6.65; N: 5.76.

**9-(4-Bromophenyl)-3,3,6,6-tetramethyl-10-(3-nitro-phenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3l):** Yellowish solid (60%), mp. 258 °C (EtOAc); IR (KBr,  $\nu cm^{-1}$ ): 2968, 2930, 1643, 1565, 1522, 1372, 1241, 1146, 1019;  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  8.46 (d,  $J$  = 7.8 Hz, 1H, Ar-H), 8.14 (s, 1H, Ar-H), 7.86 (d,  $J$  = 7.8 Hz, 1H, Ar-H), 7.64 (d,  $J$  = 7.2 Hz, 1H, Ar-H), 7.40 (d,  $J$  = 8.4 Hz, 2H, Ar-H), 7.31

(d,  $J = 8.4$  Hz, 2H, Ar-H), 5.24 (s, 1H, Benzylic-CH), 2.21-2.04 (m, 5H, -CH<sub>2</sub>), 1.80-1.72 (m, 3H, -CH<sub>2</sub>), 0.98 (s, 6H, -CH<sub>3</sub>), 0.84 (s, 6H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ 195.5, 149.1, 148.5, 144.7, 140.2, 135.9, 131.3, 129.7, 124.8, 124.6, 120.0, 115.0, 50.0, 42.1, 32.6, 29.6, 26.9; Anal. calcd. for C<sub>29</sub>H<sub>29</sub>BrN<sub>2</sub>O<sub>4</sub>; C: 63.39; H: 5.32; N: 5.10. Found: C: 63.43; H: 5.31; N: 5.11.

**9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-10-(3-nitrophenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3m):** Yellowish solid (62%), mp. 280 °C (CDCl<sub>3</sub>); IR (KBr, ν cm<sup>-1</sup>): 2957, 2926, 1639, 1564, 1517, 1364, 1241, 1145, 1011; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ 8.45 (d,  $J = 8.1$  Hz, 1H, Ar-H), 8.14 (s, 1H, Ar-H), 7.84 (t,  $J = 8.0$  Hz, 1H, Ar-H), 7.66 (d,  $J = 7.2$  Hz, 1H, Ar-H), 7.36 (d,  $J = 8.4$  Hz, 2H, Ar-H), 7.24-7.21 (m, 2H, Ar-H), 5.24 (s, 1H, Benzylic-CH), 2.25-2.05 (m, 6H, -CH<sub>2</sub>), 1.83-1.74 (m, 2H, -CH<sub>2</sub>), 0.97 (s, 6H, -CH<sub>3</sub>), 0.83 (s, 6H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ 195.5, 149.0, 148.5, 144.2, 140.1, 135.8, 131.7, 131.2, 129.2, 128.2, 124.7, 124.5, 114.9, 50.0, 42.0, 32.5, 32.4, 29.6, 26.8; Anal. calcd. for C<sub>29</sub>H<sub>29</sub>ClN<sub>2</sub>O<sub>4</sub>; C: 68.97; H: 5.79; N: 5.55. Found: C: 68.94; H: 5.77; N: 5.57.

**9-(4-Chlorophenyl)-2,2,5,5-tetramethyl-10-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3n):** White solid (83%), mp. 256 °C (CDCl<sub>3</sub>); IR (KBr, ν cm<sup>-1</sup>): 2962, 2931, 1643, 1580, 1515, 1348, 1220, 1119, 997; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ 7.13 (d,  $J = 7.2$  Hz, 4H, Ar-H), 6.96 (d,  $J = 7.5$  Hz, 4H, Ar-H), 5.01 (s, 1H, Benzylic-CH), 2.24 (s, 3H, -CH<sub>3</sub>), 2.06-2.00 (m, 2H, -CH<sub>2</sub>), 1.83-1.77 (m, 2H, -CH<sub>2</sub>), 1.45 (brs, 4H, -CH<sub>2</sub>), 0.84 (s, 6H, -CH<sub>3</sub>), 0.74 (s, 6H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ 200.7, 149.8, 145.6, 139.4, 135.8, 131.0, 130.7, 130.1, 129.5, 129.0, 128.7, 127.9, 113.0, 39.3, 34.5, 32.8, 24.7, 23.7, 21.0; Anal. calcd. for C<sub>30</sub>H<sub>32</sub>ClNO<sub>2</sub>; C: 76.01; H: 6.80; N: 2.95. Found: C: 75.98; H: 6.79; N: 2.94.

**9-(4-Chlorophenyl)-3,6-diphenyl-10-p-tolyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (3o):** Off white solid (81%), mp. 262 °C (CDCl<sub>3</sub>); IR (KBr, ν cm<sup>-1</sup>): 2954, 2934, 1626, 1576, 1512, 1358, 1233, 1134, 1014; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ 7.49 (d,  $J = 8.4$  Hz, 1H, Ar-H), 7.39-7.27 (m, 12H, Ar-H), 7.15-7.08 (m, 5H, Ar-H), 5.48-5.43 (m, 1H, Benzylic-CH), 3.37-3.18 (m, 2H, Benzylic-CH), 2.66-2.35 (m, 11H, -CH<sub>2</sub> and -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ 195.1, 151.3, 151.1, 150.7, 145.0, 144.6, 142.5, 142.4, 139.8, 135.7, 135.6, 131.8, 131.6, 130.9, 130.7, 129.3, 129.2, 128.7, 128.6, 128.4, 128.2, 127.0, 126.7, 115.9, 114.8, 43.8, 43.1, 39.5, 39.2, 35.8, 35.1, 32.3, 31.8, 21.2; Anal. calcd. for C<sub>38</sub>H<sub>32</sub>ClNO<sub>2</sub>; C: 80.05; H: 5.66; N: 2.46. Found: C: 80.08; H: 5.67; N: 2.45.

**9-(4-bromophenyl)-3,6-dimethyl-10-p-tolyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (3p):** Off white solid (74%), mp. 248 °C (CDCl<sub>3</sub>); IR (KBr, ν cm<sup>-1</sup>): 2967, 2930, 2856, 1644, 1577, 1485, 1351, 1219, 1156, 993; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ 7.28-7.20 (m, 4H, Ar-H), 7.12-7.09 (m, 4H, Ar-H), 5.24-5.14 (m, 1H, Benzylic-CH), 2.39 (s, 3H, -CH<sub>3</sub>), 2.33-2.07 (m, 5H, -CH<sub>2</sub> or -CH), 1.92-1.69 (m, 5H, -CH<sub>2</sub> or -CH), 0.83-0.78 (m, 6H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ 196.0, 195.8, 195.7, 151.8, 151.6, 151.0, 150.7, 145.0, 144.9, 139.6, 135.9, 131.3, 130.5, 129.3, 129.1, 129.0, 128.0, 127.9, 115.3, 115.0, 114.5, 44.9, 44.5, 44.3, 36.3, 35.6, 35.3, 32.6, 32.1, 31.5, 29.0, 28.6, 28.4, 21.2, 20.8, 20.6, 20.3; Anal. calcd. for C<sub>28</sub>H<sub>28</sub>BrNO<sub>2</sub>; C: 68.57; H: 5.75; N: 2.86. Found: C: 68.61; H: 5.74; N: 2.87.

**3,6-dimethyl-9-propyl-10-p-tolyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (3q):** Off white solid (65%), mp. 185-186 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2961, 2929, 1633, 1574, 1358, 1232, 1159, 1010; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.29 (d, J = 7.2 Hz, 2H, Ar-H), 7.06 (brs, 2H, Ar-H), 4.23 (s, 1H, -CH), 2.45 (s, 3H, -CH<sub>3</sub>), 2.45-2.40 (m, 2H, -CH<sub>2</sub> or -CH), 2.29-1.95 (m, 7H, -CH<sub>2</sub> or -CH), 1.89-1.69 (m, 1H, -CH<sub>2</sub> or -CH), 1.41-1.24 (m, 4H, -CH<sub>2</sub>), 0.94 (s, 6H, -CH<sub>3</sub>), 0.87-0.84 (m, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.6, 196.3, 196.2, 152.6, 152.3, 151.4, 139.2, 136.3, 130.3, 129.1, 115.4, 114.8, 114.5, 114.1, 45.3, 44.6, 44.4, 38.6, 37.9, 37.4, 36.4, 35.5, 35.1, 28.9, 28.6, 28.5, 26.4, 26.1, 25.6, 21.2, 21.0, 20.5, 20.4, 18.6, 18.3, 14.4; Anal. calcd. for C<sub>25</sub>H<sub>31</sub>NO<sub>2</sub>; C: 79.54; H: 8.28; N: 3.71. Found: C: 79.50; H: 8.30; N: 3.70.

**3,3,6,6-tetramethyl-9-propyl-10-p-tolyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (3r):** Off white solid (68%), mp. 192 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2964, 2930, 2862, 1655, 1567, 1476, 1370, 1223, 1144, 1011; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.27 (d, J = 7.5 Hz, 2H, Ar-H), 7.05 (d, J = 5.7 Hz, 2H, Ar-H), 4.19 (s, 1H, -CH), 2.43-2.36 (m, 4H, -CH<sub>3</sub> or -CH<sub>2</sub>), 2.17 (s, 3H, -CH<sub>3</sub> or -CH<sub>2</sub>), 2.01 (d, J = 17.4 Hz, 2H, -CH<sub>2</sub>), 1.73 (d, J = 17.4 Hz, 2H, -CH<sub>2</sub>), 1.41-1.39 (m, 2H, -CH<sub>2</sub>), 1.27 (brs, 2H, -CH<sub>2</sub>), 0.92 (s, 12H, -CH<sub>3</sub>), 0.84 (t, J = 6.8 Hz, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.4, 151.3, 139.2, 136.3, 129.6, 123.9, 113.8, 50.4, 41.6, 32.1, 29.9, 28.2, 26.5, 18.7, 14.4; Anal. calcd. for C<sub>27</sub>H<sub>35</sub>NO<sub>2</sub>; C: 79.96; H: 8.70; N: 3.45. Found: C: 79.98; H: 8.72; N: 3.46.

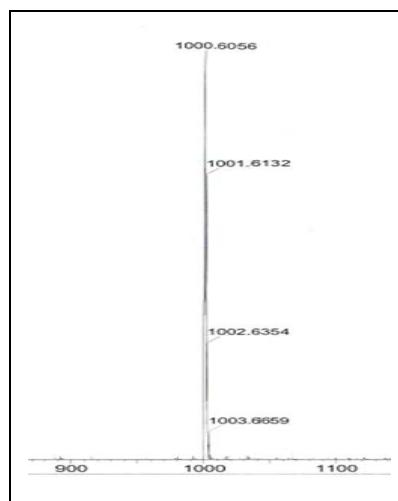
**8-(4-bromophenyl)-4-p-tolyl-2,3,5,6-tetrahydropyridocyclopenta[b,e]pyridine-1,7(4H,8H)-dione (3s):** Off white solid (61%), mp. 194 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2966, 1655, 1538, 1495, 1304, 1214, 1109; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.35 (brs, 4H, Ar-H), 7.24 (brs, 4H, Ar-H), 4.80 (s, 1H, Benzylic-CH), 2.45 (brs, 5H, -CH<sub>2</sub>), 2.36 (brs, 6H, -CH<sub>3</sub> or -CH<sub>2</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  201.8, 166.2, 142.2, 140.0, 133.9, 131.1, 130.6, 129.7, 127.9, 127.5, 120.2, 120.1, 34.1, 33.9, 24.7, 21.1; Anal. calcd. for C<sub>24</sub>H<sub>20</sub>BrNO<sub>2</sub>; C: 66.37; H: 4.64; N: 3.22. Found: C: 66.41; H: 4.65; N: 3.21.

**10-(4-bromophenyl)-9-propyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (3t):** Off white solid (67%), mp. 194 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2961, 2879, 1636, 1581, 1480, 1357, 1224, 1136, 1003; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.64 (d, J = 8.4 Hz, 2H, Ar-H), 7.09 (d, J = 8.4 Hz, 2H, Ar-H), 4.25 (d, J = 4.2 Hz, 1H, -CH), 2.56-2.41 (m, 3H, -CH<sub>2</sub>), 2.29-1.99 (m, 9H, -CH<sub>2</sub>), 1.42-1.18 (m, 4H, -CH<sub>2</sub>), 0.86 (t, J = 6.4 Hz, 3H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  196.5, 151.9, 133.0, 132.3, 131.2, 125.2, 115.4, 110.1, 38.5, 36.8, 28.2, 25.7, 21.2, 18.3, 14.3; Anal. calcd. for C<sub>22</sub>H<sub>24</sub>BrNO<sub>2</sub>; C: 63.77; H: 5.84; N: 3.38. Found: C: 63.74; H: 5.85; N: 3.39.

**3,3,6,6-tetramethyl-10-(4-nitrophenyl)-9-p-tolyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (3u):** Off white solid (63%), mp. 254-256 °C (CDCl<sub>3</sub>); IR (KBr,  $\nu$  cm<sup>-1</sup>): 2963, 2931, 2868, 1660, 1574, 1518, 1359, 1220, 1135, 996; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  8.43 (d, J = 8.7 Hz, 2H, Ar-H), 7.53 (d, J = 8.7 Hz, 2H, Ar-H), 7.28 (d, J = 7.8 Hz, 2H, Ar-H), 7.04 (d, J = 7.8 Hz, 2H, Ar-H), 5.21 (s, 1H, Benzylic-CH), 2.24 (s, 3H, -CH<sub>3</sub>), 2.17-2.03 (m, 6H, -CH<sub>2</sub>), 1.78 (d, J = 17.1 Hz, 2H, -CH<sub>2</sub>), 0.95 (s, 6H, -CH<sub>3</sub>), 0.82 (s, 6H, -CH<sub>3</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>):  $\delta$  195.6, 148.3, 147.9, 144.9, 142.6, 135.4, 131.2, 128.8, 127.6, 125.3, 115.2, 50.1, 41.8, 32.5, 32.5, 32.1, 29.6, 26.8, 21.0; Anal. calcd. for C<sub>30</sub>H<sub>32</sub>N<sub>2</sub>O<sub>4</sub>; C: 74.36; H: 6.66; N: 5.78. Found: C: 74.31; H: 6.68; N: 5.80.

**9-ethyl-3,3,6,6-tetramethyl-10-p-tolyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (3v):** Off white solid (68%), mp. 178 °C ( $\text{CDCl}_3$ ); IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 2958, 2878, 1635, 1569, 1511, 1354, 1269, 1143, 1012;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.29 (d,  $J = 8.1$  Hz, 2H, Ar-H), 7.00 (d,  $J = 8.1$  Hz, 2H, Ar-H), 4.24 (brs, 1H, -CH), 2.46 (s, 3H, -CH<sub>3</sub>), 2.22 (s, 4H, -CH<sub>2</sub>), 2.06-2.00 (m, 2H, -CH<sub>2</sub>), 1.78-1.73 (m, 1H, -CH<sub>2</sub>), 1.54-1.50 (m, 1H, -CH<sub>2</sub>), 1.27 (brs, 2H, -CH<sub>2</sub>), 0.96 (s, 12H, -CH<sub>3</sub>), 0.86 (t,  $J = 7.8$  Hz, 3H, -CH<sub>3</sub>);  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta$  196.4, 151.4, 136.5, 129.8, 124.0, 113.3, 50.3, 41.7, 32.1, 30.0, 28.3, 27.6, 27.0, 26.6, 21.2, 14.0, 9.6; Anal. calcd. for  $\text{C}_{26}\text{H}_{33}\text{NO}_2$ ; C: 79.76; H: 8.50; N: 3.58. Found: C: 79.81; H: 8.49; N: 3.59.

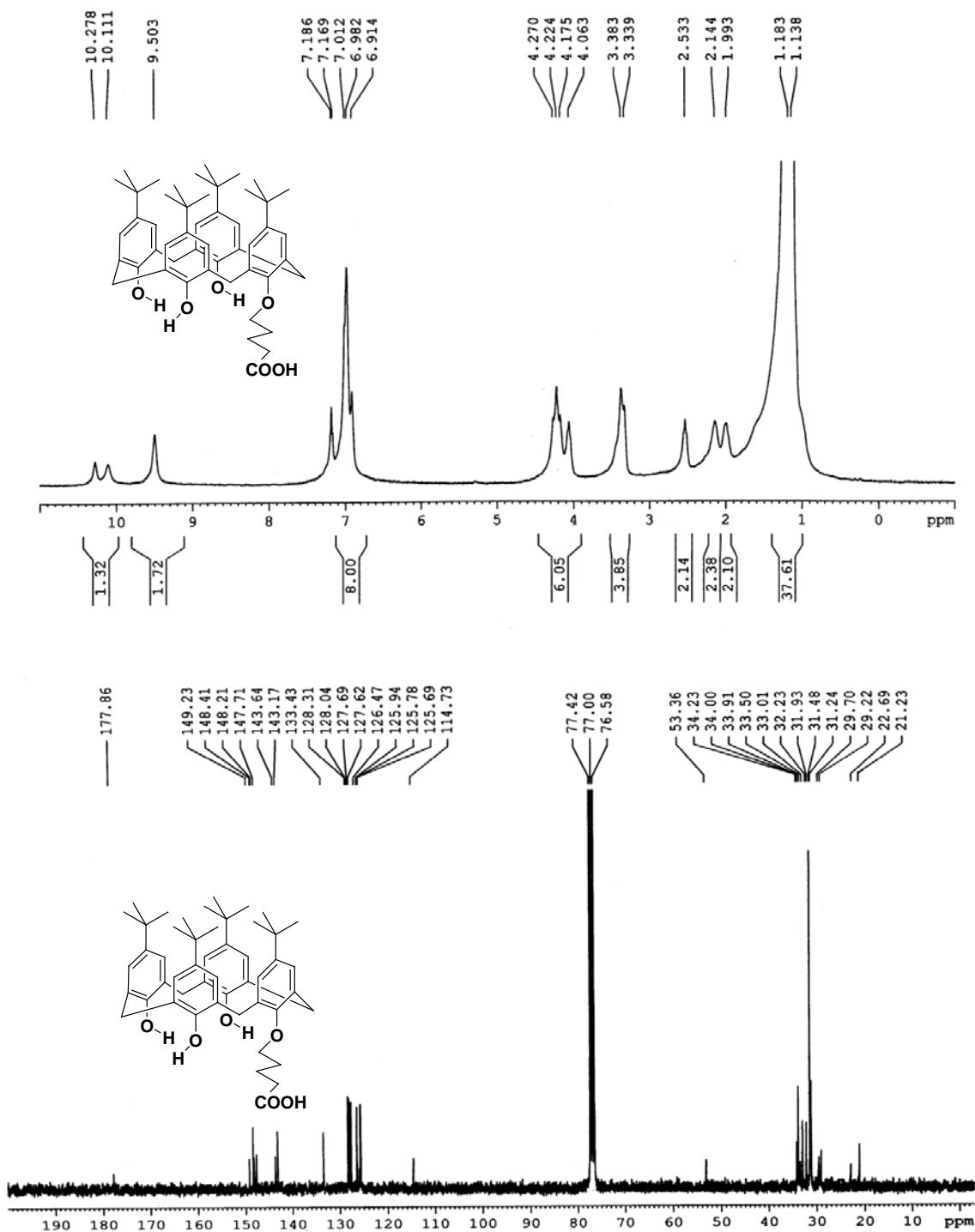
## 7. The mass peak for 1:1 enaminoketone 2a ⊂ calixarene C4V1:



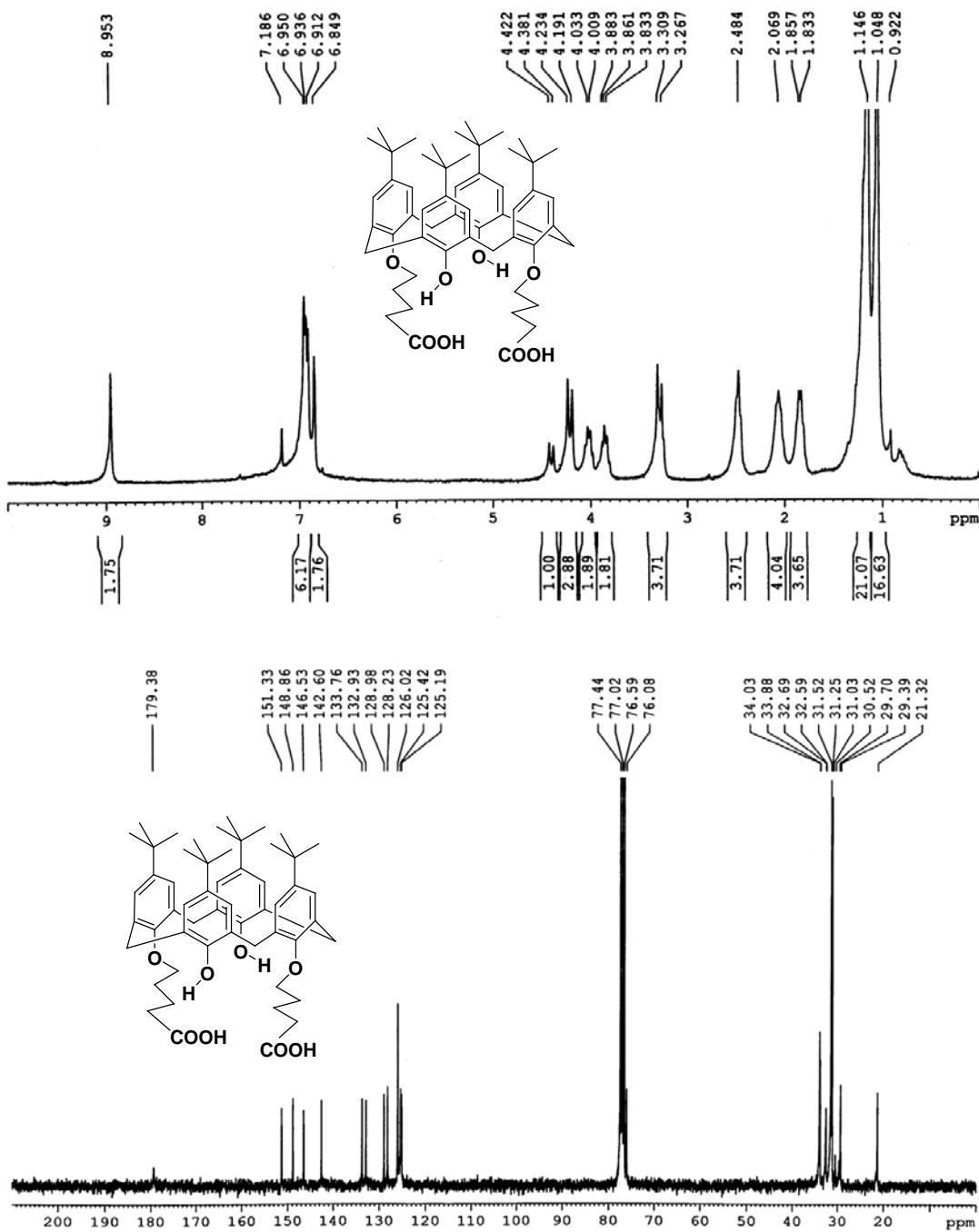
## 8. HPLC data of the crude product of reaction of 1b and 2b:



**9.  $^1\text{H}$  and  $^{13}\text{C}$  -NMR spectra of the mono acid grafted *p*-*tert*-butylcalix[4]arene (C4V1)**

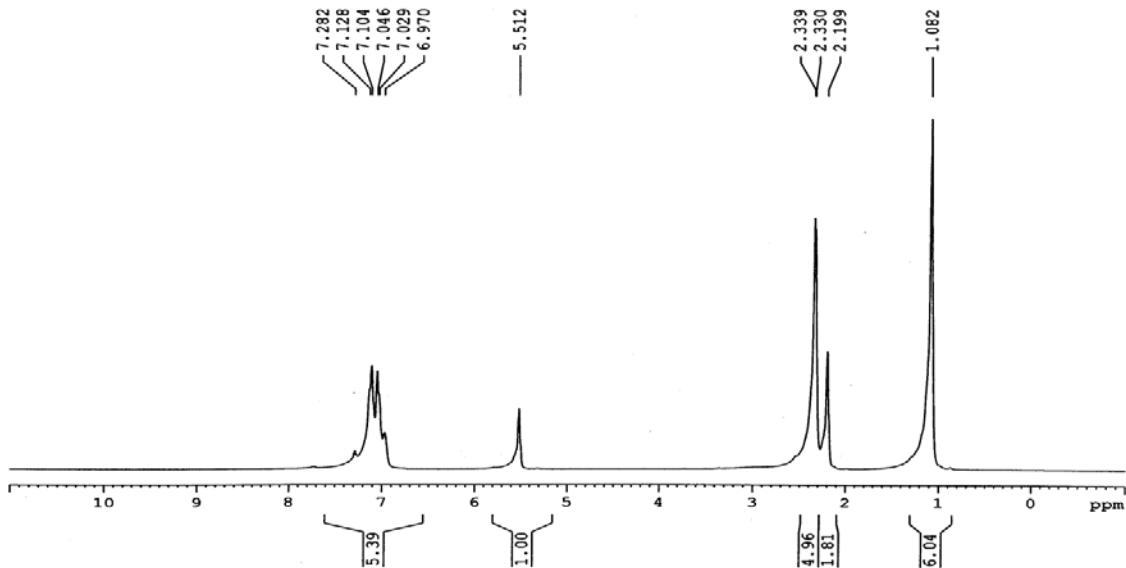


**10.  $^1\text{H}$  and  $^{13}\text{C}$  -NMR spectra of the di acid grafted *p*-*tert*-butylcalix[4]arene (C4V2):**

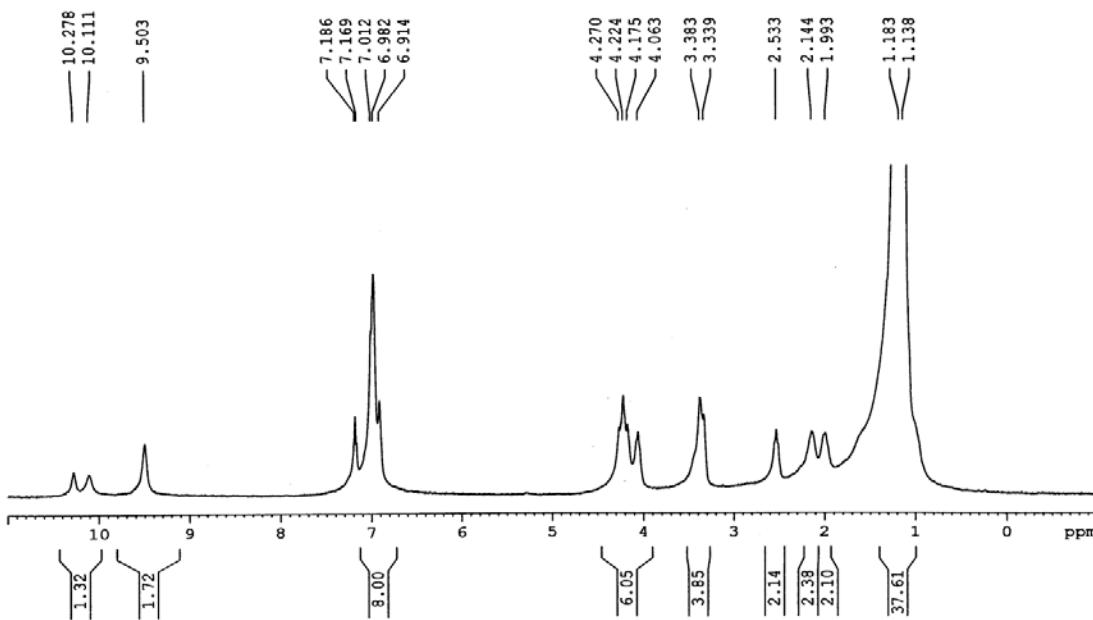


**11. Corresponding  $^1\text{H}$ -NMRs of Fig. 1 (Manuscript)** [Stacked  $^1\text{H}$  NMR spectra ( $\text{CDCl}_3$ , room temperature; 303 K) of the guest enaminoketone **2a**, the host cavitand **C4V1**, and enaminoketone **2a**  $\subset$  calixarene **C4V1**]

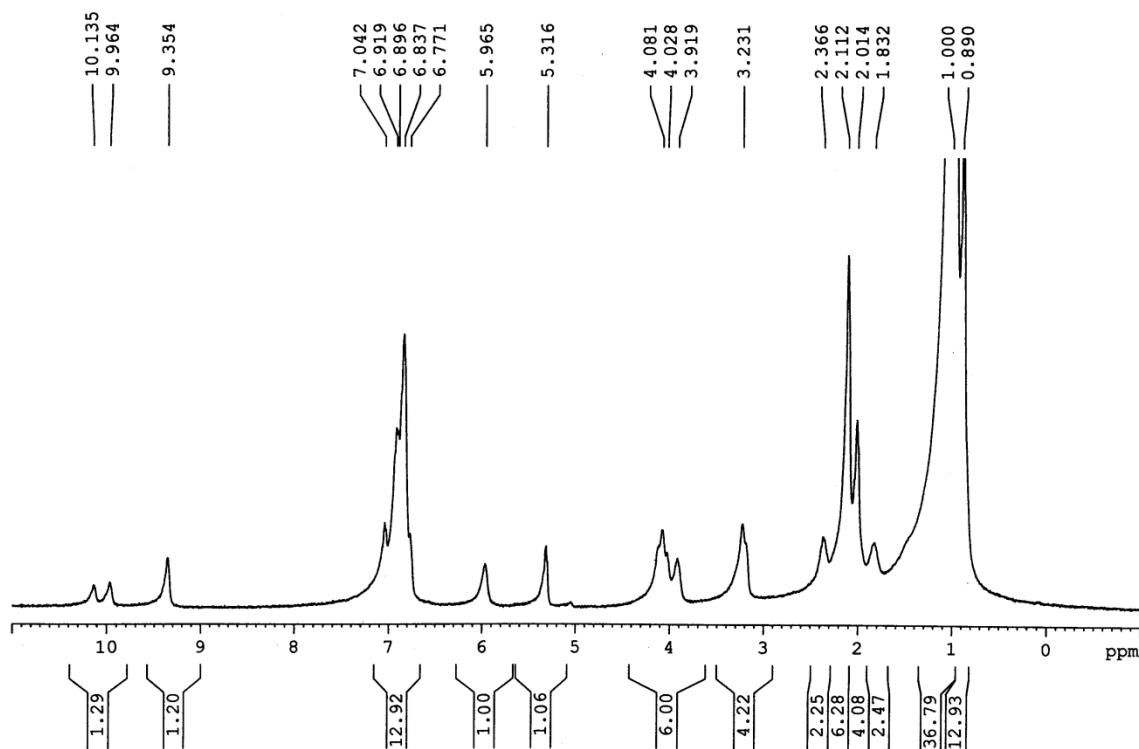
**Enaminoketone 1a**



**Cavitand C4V1**

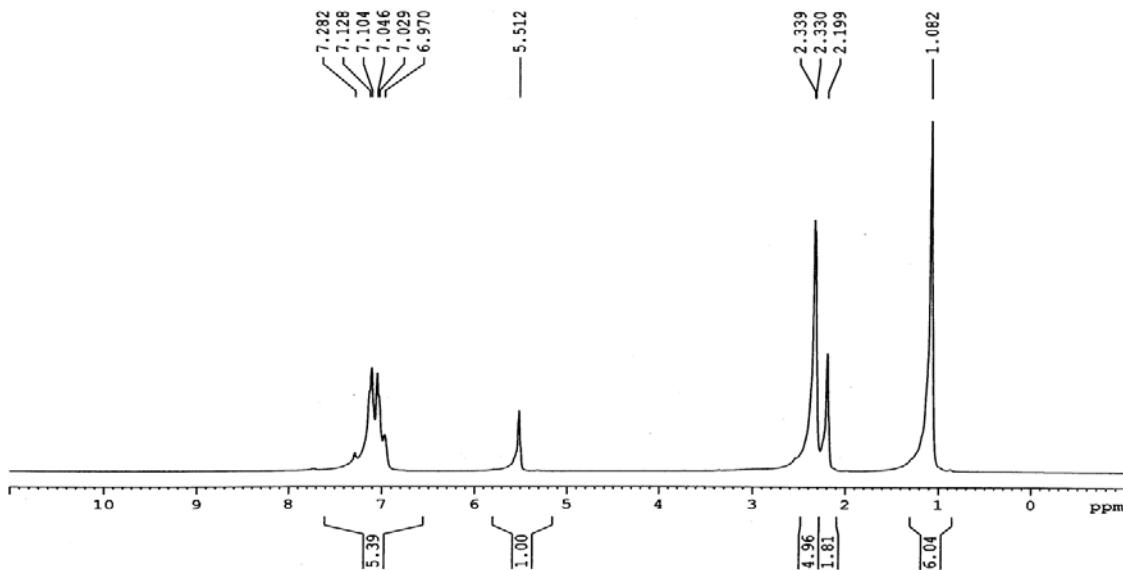


**Enaminoketone 1a ⊂ Calixarene C4V1**

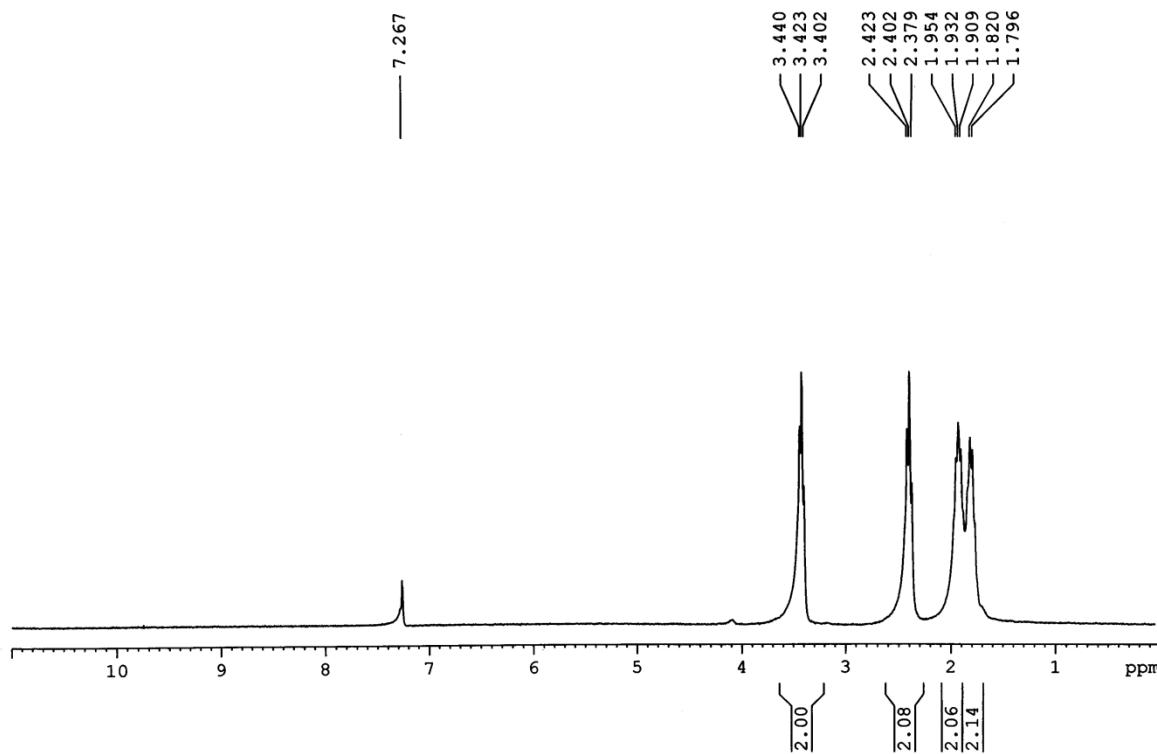


**12. Corresponding  $^1\text{H}$ -NMRs of Fig. 3 (Manuscript)** [Stacked  $^1\text{H}$  NMR spectra ( $\text{CDCl}_3$ , room temperature; 303 K) of the enaminoketone **1a**, 5-bromoaleric acid **5-BVA**, and **5-BVA** + enaminoketone **1a**]

**Enaminoketone **1a****



**5-bromoaleric acid **5-BVA****



**5-BVA + enaminoketone 1a**

