Supplementary information

Fe₃O₄@SiO₂ core/shell functionalized by gallic acid: a novel, robust and water-compatible heterogeneous magnetic nanocatalyst for environmentally friendly synthesis of acridine-1,8-diones

Zahra Firoozi, Dariush Khalili,* and Ali Reza Sardarian*

^aDepartment of Chemistry, College of Sciences, Shiraz University, Shiraz 71467-13565, Iran

Table of content

| Experimental section | S2 |
|---|-----|
| Calculation of green chemistry metrics | S2 |
| Characterization of the products | S4 |
| Copy of FT-IR, ¹ H-NMR and ¹³ C-NMR of products | S12 |
| References | S56 |

Experimental section

Chemical and instrument

All chemicals were purchased from Merck, Acros, and Sigma-Aldrich and used without further purification. The power and oscillation of the used ultrasonic device in catalyst synthesis was 120 (V) and 40 (kHz) and the final catalyst was characterized as follows: The fourier transform infrared spectrometer (FT-IR) model Shimadzu FT-IR 8300 was applied to FT-IR measurement using KBr pellet in the range of 400 to 4000 wavenumbers/cm⁻¹. Also, the X-ray diffraction (XRD) patterns were recorded by a GNR (Italy) XRD explorer X-ray diffractometer using CuK α radiation (λ = 1.54178 Å) with a 20 scan range of 10° to 80°. Moreover, the presence of elements in the magnetic samples was proved by energy dispersive X-ray spectrometry (EDX) attached to a Philips scanning electron microscope (SEM), Transmission electron microscope (TEM) images were taken on a ZEISS EM10C-100KV microscope, and field emission scanning electron microscope (FE-SEM) images were taken on a ZEISS SIGMA VP microscope, Dynamic light scattering (DLS) measurements were performed on a HORIBA LB-550. To obtain thermal gravimetric analysis (TGA) data, a device from TA company model Q600 made in USA was used and vibrating sample magnetometer (VSM) measurements were analyzed using BHV-55 model vibrating sample magnetometer. Finally, zeta potential analysis was performed on a HORIBA Z-100. The reaction progress has been checked by thin layer chromatography (TLC). The final products characterized by melting points in open capillary tubes were determined with a Büchi B-545 melting point apparatus, and nuclear magnetic resonance (NMR) spectroscopy using Devices Brucker DPX-400 spectrometer that work for ¹³C at 101 MHz and for ¹H at 400 MHz and a spectrometer Brucker DPX-300 that work for ¹³C at 75 MHz and ¹H at 300 Hz in pure deuterated dimethyl sulfoxide $(DMSO-d_6)$ and deuterated chloroform $(CDCl_3)$.

Calculation of green chemistry metrics

In order to assess the environmental sustainability of our catalytic system, important green chemistry metrics such as the environmental factor (E-factor), atom economy, reaction mass efficiency (RME), process mass intensity (PMI) and eco-score (scale) were scrutinized. Taking the Fe₃O4@SiO₂-NH-GA-[(CH₂)₄-SO₃H]₃-catalyzed reaction involving dimedone, benzaldehyde, and ammonium acetate as a model reaction, these metrics were obtained as follows:



| Compound code | 1 | 2a | 3 | 4a |
|------------------|-------|-----|------|-----|
| M.W. (g/mol) | 140 | 106 | 77 | 349 |
| M.W. (mg) | 2×140 | 106 | 84.7 | 349 |

The total mass of reactants = 470.7

Obtained product = $349 \times 0.9 = 314.1$

Environmental factor (E-factor):

E-factor = Amount of waste /Amount of product

The Amount of waste = (total mass of raw materials - the total mass of product)

The Amount of waste = (470.7 - 314.1) = 156.6

E-factor = 156.6 /314.1 = 0.50 KgKg⁻¹

Molecular weight of the desired product

% Atom economy = 100 × *Molecular weight all of reactants*

 $\frac{349}{8 \text{ Atom economy} = 100 \times 280 + 106 + 77} = 75.4\%$

Reactionmassefficiency(RME)=mass of product $\times 100$ $\Sigma mass of stoichiometric reactants$

Reaction mass efficiency (RME) = $\frac{314.1}{470.7} \times 100$ = 67%

mass of product

 $\mathsf{PMI} = \frac{470.7 + 2}{314.1} = 1.51$

Ideal value of PMI = E-factor + 1 = 0.50 + 1 = 1.50

E-score has been calculated for the reaction based on the following 6 parameters below (See Beilstein Journal of Organic Chemistry 2006, 2, 3.)

=

| Entry | Parameter | Values | Penalty Points |
|----------------------|-------------------------|--------------------------|----------------|
| 1 | Yield | 100-90/2 | 5 |
| 2 | Cost of reactants | Inexpensive | 0 |
| 3 | Safety of reactants | 5+5+5+5 | 20 |
| 4 | Technical setup | Common setup | 0 |
| 5 | Temperature /time | 60 °C, <1 h | 2 |
| 6 | Workup and purification | Classical chromatography | 10 |
| Total penalty points | | 37 | |

Eco-Score = 100 - the sum of individual penalties = 100-37 = 63

Eco-scale from 0 to 100 using the following scores: > 75, excellent; > 50, acceptable; and < 50, inadequate.

Characterization of the products

3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4a) (Table 4, entry 1)

White solid; Yield = 92%, M.P. 272-275 °C (Lit. 274-276 °C ^[1]); IR (KBr): \bar{u} (cm⁻¹) = 3280, 3196, 3066, 2960, 1644, 1610, 1486, 1224. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 9.40 (s, 1H, N-*H*), 7.23-7.08 (m, 5H, Ar*H*), 4.88 (s, 1H, C*H*Ar), 2.53 (d, *J* = 16.96 Hz, 2H, CH₂), 2.40 (d, *J* = 17.12 Hz, 2H, CH₂), 2.25 (d, *J* = 16.12 Hz, 2H, CH₂), 2.06 (d, *J* = 16.32 Hz, 2H, CH₂), 1.08 (s, 6H, 2CH₃), 0.93 (s, 6H, 2CH₃). ¹³C NMR (101MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 26.4, 29.0, 32.1, 32.7, 50.1, 111.4, 125.4, 127.5, 127.5, 147.1, 149.3, 194.3.

3,3,6,6-tetramethyl-9-(2-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4b) (Table 4, entry 2)

Light yellow solid; Yield = 88%, M.P. 294-296 °C(Lit. 295-297 °C^[2]); IR (KBr): \bar{u} (cm⁻¹) = 3424, 3286, 3204, 2958, 1612, 1370, 1224. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 9.58 (s, 1H, N-*H*), 7.60-7.32 (m, 4H, Ar*H*), 5.64 (s, 1H, C*H*Ar), 2.29 (d, *J* = 8.68 Hz, 2H, C*H*₂), 2.20 (d, *J* = 16.60 Hz, 2H, C*H*₂), 2.13 (d, *J* = 16.16 Hz, 2H, C*H*₂), 2.06 (d, *J* = 16.24 Hz, 2H, C*H*₂), 1.06 (s, 6H, 2C*H*₃), 0.91 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 20.5, 22.6, 28.5, 39.8, 49.9, 67.1, 113.2, 127.7, 129.2, 129.9, 131.3, 139.9, 148.0, 148.0, 195.1.

3,3,6,6-tetramethyl-9-(3-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4c) (Table 4, entry 3)

Light yellow solid; Yield = 94%, M.P. 293-295 °C(Lit. 291-293 °C^[1]); IR (KBr): \bar{u} (cm⁻¹) = 3272, 3186, 3066, 2960, 1646, 1610, 1488, 1346, 1224. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 9.50 (s, 1H, N-*H*), 7.49-7.98 (m, 4H, Ar*H*), 4.92 (s, 1H, C*H*Ar), 2.49 (d, *J* = 16.64 Hz, 2H, C*H*₂), 2.37 (d, *J* = 17.08 Hz, 2H, C*H*₂), 2.21 (d, *J* = 16.16 Hz, 2H, C*H*₂), 2.00 (d, *J* = 16.12 Hz, 2H, C*H*₂), 1.02 (s, 6H, 2C*H*₃), 0.86 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 26.3, 29.0, 32.1, 33.4, 49.9, 110.5, 120.7, 121.9, 134.4, 147.3, 149.1, 150.0, 194.4.

3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4d) (Table 4, entry 4)

Light yellow solid; Yield = 96%, M.P. 299-301 °C(Lit. 297-299 °C^[1]); IR (KBr): \bar{u} (cm⁻¹) = 3386, 3074, 2960, 1644, 1480, 1342, 1222. ¹H NMR (400 MHz, DMSO-*d*₆) δ (ppm): 9.50 (s, 1H, N-*H*), 8.08 (d, *J* = 8.60 Hz, 2H, Ar*H*), 7.42 (d, *J* = 8.68 Hz, 2H , Ar*H*), 4.90 (s, 1H, C*H*Ar), 2.49 (d, *J* = 17.00 Hz, 2H, C*H*₂), 2.34 (d, *J* = 17.08 Hz, 2H, C*H*₂), 2.20 (d, *J* = 16.16 Hz, 2H, C*H*₂), 1.99 (d, *J* = 16.36 Hz, 2H, C*H*₂), 1.01 (s, 6H, 2C*H*₃), 0.86 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) δ (ppm): 27.1, 29.4, 32.7, 34.4, 50.5, 112.8, 123.4, 129.0, 130.9, 147.8, 153.7, 195.1.

9-(2-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4e) (Table 4, entry 5)

Light yellow solid; Yield = 85%, M.P. 292-294 °C(Lit. 290-299 °C^[4]); IR (KBr): \bar{u} (cm⁻¹) = 3280, 3202, 3072, 2954, 1636, 1608, 1486, 1224, 750. ¹H NMR (400 MHz, DMSO-*d*₆) δ (ppm): 9.35 (s, 1H, N-*H*), 7.29 (d, *J* = 7.08 Hz, 2H, Ar*H*), 7.20-7.13 (m, 2H, Ar*H*), 7.05 (d, *J* = 7.44 Hz, 2H, Ar*H*), 5.08 (s, 1H, C*H*Ar), 2.45 (d, *J* = 17.00 Hz, 2H, C*H*₂), 2.28 (d, *J* = 17.00 Hz, 2H, C*H*₂), 2.15 (d, *J* = 16.16 Hz, 2H, C*H*₂), 1.93 (d, *J* = 16.12 Hz, 2H, C*H*₂), 1.01 (s, 6H, 2C*H*₃), 0.87 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) δ (ppm): 26.3, 29.1, 31.9, 32.9, 50.2, 110.7, 126.1, 126.9, 129.0, 131.9, 132.2, 144.0, 149.6, 194.1.

9-(3-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4f) (Table 4, entry 6)

Light yellow solid; Yield = 87%, M.P. 292-294 °C(Lit. 290 °C^[4]); IR (KBr): \bar{v} (cm⁻¹) = 3416, 3182, 3064, 2962, 1620, 1616, 1490, 1220, 694. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 9.50 (s, 1H, N-*H*), 7.22 (s, 1H, Ar*H*), 7.22-7.08 (m, 1H, Ar-*H*), 7.05 (t, *J* = 7.80 Hz, 1H, Ar*H*), 7.00-6.97 (m, 1H, Ar*H*), 5.00 (s, 1H, C*H*Ar), 2.25 (d, *J* = 16.88 Hz, 2H, C*H*₂), 2.19 (d, *J* = 4.92 Hz, 2H, C*H*₂), 2.15 (d, *J* = 5.52 Hz, 2H, C*H*₂), 2.10 (d, *J* = 16.36 Hz, 2H, C*H*₂), 1.01 (s, 6H, 2C*H*₃), 0.90 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 27.1, 29.5, 32.6, 40.8, 50.7, 112.8, 126.2, 126.6, 128.0, 129.1, 133.7, 148.5, 148.8, 195.67.

9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4g) (Table 4, entry 7)

White solid; Yield = 87%, M.P. 294-296 °C(Lit. 298-300 °C^[5]); IR (KBr): \bar{u} (cm⁻¹) = 3552, 3478, 3408, 2960, 1644, 1618, 1488, 1222, 844. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 9.38 (s, 1H, N-*H*), 7.23 (d, *J* = 8.40 Hz, 2H, Ar*H*), 7.15 (d, *J* = 8.48 Hz, 2H, Ar*H*), 4.78 (s, 1H, C*H*Ar), 2.46 (d, *J* = 17.04 Hz, 2H, C*H*₂), 2.32 (d, *J* = 17.04 Hz, 2H, C*H*₂), 2.18 (d, *J* = 16.12 Hz, 2H, C*H*₂), 1.98 (d, *J* = 16.08 Hz, 2H, - C*H*₂), 1.01 (s, 6H, 2C*H*₃), 0.86 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 27.1, 29.5, 39.6, 40.9, 50.7, 113.2, 128.1, 129.4, 131.5, 145.0, 148.2, 195.6.

9-(2-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-

dione (4h) (Table 4, entry 8)

White solid; Yield = 81%, M.P. 255-257 °C(Lit. 252-254 °C^[6]); IR (KBr): \bar{u} (cm⁻¹) = 3276, 3182, 3064, 2962, 1640, 1612, 1486, 1222, 560. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 9.39, (s, 1H, N-*H*), 7.23-7.09 (m, 4H, Ar*H*), 4.80 (s, 1H, C*H*Ar), 2.46 (d, *J* = 21.12 Hz, 2H, C*H*₂), 2.35 (d, *J* = 17.04

Hz, 2H, CH₂), 2.19 (d, J = 16.08 Hz, 2H, CH₂), 2.01 (d, J = 16.00 Hz, 2H, CH₂), 1.01 (s, 6H, 2CH₃), 0.87 (s, 6H, 2CH₃). ¹³C NMR (101MHz, DMSO- d_6) δ (ppm): 26.3, 29.0, 32.1, 33.0, 50.0, 110.8, 125.4, 126.1, 127.5, 129.5, 132.1, 149.3, 149.6, 194.4.

9-(5-bromo-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4i) (Table 4, entry 9)

Light yellow solid; Yield = 80%, M.P. 238-240 °C(Lit. 235-238 °C^[3]); IR (KBr): \bar{u} (cm⁻¹) = 3512, 3480, 3416, 2958, 1616, 1604, 1480, 1232, 626. ¹H NMR (300 MHz, DMSO-*d*₆) δ (ppm): 9.69 (s, 1H, O-H), 9.64 (s, 1H, N-*H*), 7.07 (d, *J* = 6.84 Hz, 1H, , Ar*H*), 6.98 (s, 1H, Ar*H*), 6.65 (d, *J* = 10.32 Hz, 1H, Ar*H*), 4.79 (s, 1H, CHAr), 2.48 (d, *J* = 12.57 Hz, 2H, C*H*₂), 2.37 (d, *J* = 17.10 Hz, 2H, C*H*₂), 2.24 (d, *J* = 16.20 Hz, 2H, C*H*₂), 2.05 (d, *J* = 16.08 Hz, 2H, C*H*₂), 1.00 (s, 6H, 2C*H*₃), 0.91 (s, 6H, 2C*H*₃). ¹³C NMR (75MHz, DMSO-*d*₆) δ (ppm): 26.7, 29.5, 32.6, 37.7, 50.2, 110.8, 113.4, 113.7, 119.6, 130.1, 131.9, 140.5, 143.9, 151.7, 153.7, 196.2.

3,3,6,6-tetramethyl-9-(m-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4j) (Table 4, entry 10)

Yellow solid; Yield = 80%, M.P. 214-217 °C(Lit. 211-213 °C^[7]); IR (KBr): \bar{u} (cm⁻¹) = 3278, 3182, 3066, 2956, 1642, 1604, 1488, 1220. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 9.33 (s, 1H, N-*H*), 7.12-6.90 (m, 4H, Ar*H*), 4.83 (s, 1H, C*H*Ar), 2.51 (d, *J* = 17.08 Hz, 2H, C*H*₂), 2.39 (d, *J* = 17.04 Hz, 2H, C*H*₂), 2.26 (s, 3H, C*H*₃), 2.23 (d, *J* = 16.28 Hz, 2H, C*H*₂), 2.05 (d, *J* = 16.28 Hz, 2H, -C*H*₂), 1.07 (s, 6H, 2C*H*₃), 0.93 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 19.3, 24.5, 27.2, 30.3, 30.8, 48.4, 109.6, 122.8, 124.2, 125.7, 126.6, 134.2, 145.2, 147.3, 192.5.

3,3,6,6,9-pentamethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4k) (Table 4, entry 11)

Light yellow solid; Yield = 74%, M.P. 270-273 °C(Lit. 269-279 °C^[8]); IR (KBr): \bar{u} (cm⁻¹) = 3416, 3280, 3204, 2958, 1630, 1608, 1488, 1232. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 9.51 (s, 1H, N-*H*), 6.50 (s, 1H, CHAr), 2.27 (d, *J* = 14.64 Hz, 2H, C*H*₂), 2.18 (s, 3H, C*H*₃), 2.14 (d, *J* = 8.60 Hz, 2H, C*H*₂), 1.93 (d, *J* = 6.28 Hz, 2H, C*H*₂), 1.66 (d, *J* = 8.64 Hz, 2H, C*H*₂), 1.02 (s, 6H, 2C*H*₃), 0.90 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 21.5, 22.5, 27.0, 29.5, 41.1, 50.9, 114.5, 148.3, 195.9.

9-ethyl-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4I) (Table 4, entry 12)

Light yellow solid; Yield = 70%, M.P. 291-293 °C(Lit. 282-283 °C^[9]); IR (KBr): \bar{u} (cm⁻¹) =3468, 2951, 1608, 1395. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 9.61 (s, 1H, N-*H*), 3.14 (t, *J* = 11.38 Hz, 1H, C*H*CH₂), 2.28 (d, *J* = 9.12 Hz, 2H, C*H*₂), 2.20 (d, *J* = 12.64 Hz, 2H, C*H*₂), 2.11 (d, *J* = 8.40 Hz, 2H, C*H*₂), 2.03 (d, *J* = 14.56 Hz, 2H, C*H*₂), 1.40-1.31 (m, 2H, C*H*₂), 1.10 (s, 6H, 2C*H*₃), 0.99 (s, 6H, 2C*H*₃), 0.89 (t, *J* = 4.86 Hz, 3H, CH₂C*H*₃). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 11.1, 24.7, 29.4, 31.8, 32.6, 40.5, 46.4, 50.0, 115.4, 125.8, 127.6, 128.0, 145.7, 150.2, 195.6.

3,3,6,6-tetramethyl-9-(pyridin-3-yl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4m) (Table 4, entry 13)

Light yellow solid; Yield = 83%, M.P. 301-303 °C(Lit. 298-300 °C^[10]); IR (KBr): \bar{u} (cm⁻¹) = 3361, 2936, 2877, 3815, 1612, 1601, 1545, 1403. ¹H NMR (400 MHz, DMSO-*d*₆) δ (ppm): 9.48 (s, 1H, N-*H*), 7.37(d, *J* = 52.08 Hz, 2H, Ar*H*), 7.64-7.54 (m, 1H, Ar*H*), 7.33 (s, 1H, Ar*H*), 4.82 (s, 1H, CHAr), 2.36 (d, *J* = 17.00 Hz, 2H, C*H*₂), 2.23 (d, *J* = 13.80 Hz, 2H, C*H*₂), 2.10 (d, *J* = 7.80 Hz, 2H, C*H*₂), 1.99 (d, *J* = 15.88Hz, 2H, C*H*₂), 1.01 (s, 6H, 2C*H*₃), 0.85 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) δ (ppm): 26.9, 28.9, 29.4, 31.8, 32.3, 32.6, 50.4, 110.8, 127.5, 136.8, 145.9, 148.1, 150.6, 163.9, 195.0.

3,3,6,6-tetramethyl-9,10-diphenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4n) (Table 4, entry 14)

White solid; Yield = 84%, M.P. 250-252 °C(Lit. 253-255 °C^[11]); IR (KBr): \bar{u} (cm⁻¹) = 3272, 3024, 2954, 1598, 1484, 1262. ¹H NMR (400 MHz, DMSO-*d*₆) δ (ppm): 7.87-7.11 (m, 10H, Ar*H*), 5.11 (s, 1H, C*H*Ar), 2.28 (d, *J* = 7.64 Hz, 2H, C*H*₂), 2.07 (d, *J* = 9.04 Hz, 2H, C*H*₂), 1.81 (d, *J* = 17.02 Hz, 2H, C*H*₂), 1.32 (d, *J* = 17.84 Hz, 2H, C*H*₂), 0.93 (s, 6H, 2C*H*₃), 0.77 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) δ (ppm): 25.5, 26.0, 29.2, 29.6, 49.5, 112.8, 113.0, 125.7, 127.4, 127.8, 128.2, 129.3, 138.4, 149.3, 150.2, 195.0.

10-(3-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine -1,8(2H,5H)-dione (4o) (Table 4, entry 15)

White solid; Yield = 84%, M.P. 260-262 °C(Lit. 184-187 °C^[12]); IR (KBr): \bar{u} (cm⁻¹) = 3260, 2952, 1598, 1480, 1282. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.16-7.01 (m, 5H, Ar*H*), 6.99-6.94 (m, 2H, Ar*H*), 6.44 (dd, *J* = 8.2 Hz, *j*= 2.52, 2H, ArH), 6.26 (s 1H, Ar*H*), 5.15 (s, 1H, C*H*Ar), 3.68 (s, 3H, OC*H*₃), 2.34 (d, *J* = 16.28 Hz, 2H, C*H*₂), 2.24 (d, *J* = 16.40 Hz, 2H, C*H*₂), 2.19 (d, *J* = 16.16 Hz, 2H, C*H*₂), 2.12 (d, *J* = 16.28 Hz, 2H, C*H*₂), 1.02 (s, 6H, 2C*H*₃), 0.92 (s, 6H, 2C*H*₃). ¹³C NMR (101

MHz, CDCl₃) δ (ppm): 27.2, 29.4, 39.4, 42.1, 50.6, 55.3, 100.5, 109.3, 109.6, 118.8, 125.8, 127.3, 128.2, 131.1, 136.3, 148.1, 150.1, 158.6, 192.9, 194.6.

10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10 hexahydroacridine **-1,8(2H,5H)-dione (4p)** (Table 4, entry 16)

Light yellow solid; Yield = 87%, M.P. 209-211 °C(Lit. 213-215 °C^[13]); IR (KBr): \bar{u} (cm⁻¹) = 3286, 2966, 1586, 1490, 1234. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 7.16 (d, *J* = 5.72 Hz, 4H, Ar*H*), 7.07-7.03 (m, 1H, Ar*H*), 6.89-6.86 (m, 1H, Ar*H*), 6.71-6.67 (m, 3H, Ar*H*), 5.02 (s, 1H, C*H*Ar), 3.74 (s, 3H, OC*H*₃), 2.49 (d, *J* = 14.08 Hz, 2H, C*H*₂), 2.38 (d, *J* = 21.88 Hz, 2H, C*H*₂), 2.16 (d, *J* = 21.16 Hz, 2H, C*H*₂), 1.96 (d, *J* = 21.52 Hz, 2H, C*H*₂), 1.01 (s, 6H, 2C*H*₃), 0.93 (s, 6H, 2C*H*₃). ¹³C NMR (101 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 27.1, 29.6, 31.1, 32.4, 50.5, 55.6, 113.0, 114.8, 116.7, 126.1, 127.2, 127.6, 128.5, 130.0, 149.0, 152.6, 155.7, 193.4.

10-(2-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4q) (Table 4, entry 17)

White solid; Yield = 73%, M.P. 208-211 °C(Lit. 188-190 °C^[12]); IR (KBr): \bar{u} (cm⁻¹) = 3416, 2956, 1640, 1364, 1222, 698. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.65-7.00 (m, 9H, Ar*H*), 5.16 (s, 1H, CHAr), 2.07 (d, *J* = 8.36 Hz, 2H, C*H*₂), 1.94 (d, *J* = 17.44 Hz, 2H, C*H*₂), 1.54 (d, *J* = 17.24 Hz, 2H, C*H*₂), 1.22 (d, *J* = 25.92 Hz, 2H, C*H*₂) 0.85 (s, 6H, 2C*H*₃), 0.76 (s, 6H, 2C*H*₃). ¹³C NMR (101 MHz, CDCl₃) δ (ppm): 30.1, 32.2, 38.7, 41.7, 50.1, 114.8, 125.8, 127.7, 128.2, 128.7, 128.8, 130.8, 130.9, 130.9, 131.6, 146.1, 148.4, 195.7.

10-(3-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4q) (Table 4, entry 18)

White solid; Yield = 77%, M.P. 262-264 °C(Lit. 177-180 °C^[12]); IR (KBr): \bar{u} (cm⁻¹) = 3278, 3192, 3070, 1600, 1480, 1284, 696. ¹H NMR (300 MHz, DMSO-*d*₆) δ (ppm): 7.25-7.07 (m, 5H, Ar*H*), 6.95-6.88 (m, 4H, Ar*H*), 5.05 (s, 1H, C*H*Ar), 2.53 (d, *J* = 10.32 Hz, 2H, C*H*₂), 2.40 (d, *J* = 16.83 Hz, 2H, C*H*₂), 2.19 (d, *J* = 15.99 Hz, 2H, C*H*₂), 2.00 (d, *J* = 16.11 Hz, 2H, C*H*₂), 1.03 (s, 6H, 2C*H*₃), 0.95 (s, 6H, 2C*H*₃). ¹³C NMR (75MHz, DMSO-*d*₆) δ (ppm): 27.1, 29.5, 30.5, 32.5, 50.5, 107.1, 115.0, 122.8, 125.2, 126.3, 127.3, 128.6, 131.4, 131.7, 138.0, 148.6, 152.0, 193.6.

10-(4-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4q) (Table 4, entry 19)

White solid; Yield = 78%, M.P. 295-297 °C(Lit. 301-303 °C^[13]); IR (KBr): \bar{u} (cm⁻¹) = 3416, 3058, 2956, 1636, 1364, 1222, 698. ¹H NMR (400 MHz, DMSO-*d*₆) δ (ppm): 7.48 (d, *J* = 8.24 Hz, 2H, Ar*H*), 7.34 (d, *J* = 7.52 Hz, 2H, Ar*H*), 7.17-7.12 (m, 4H, Ar*H*), 7.04 (t, *J* = 7.30 Hz, 1H, Ar*H*), 5.20 (s, 1H, CHAr), 2.13 (d, *J* = 16.28 Hz, 2H, C*H*₂), 2.06 (d, *J* = 16.60 Hz, 2H, C*H*₂), 1.98 (d, *J* = 17.36 Hz, 2H, C*H*₂), 1.74 (d, *J* = 18.16 Hz, 2H, C*H*₂), 0.89 (s, 6H, 2C*H*₃), 0.74 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) δ (ppm): 26.8, 29.7, 32.4, 32.6, 50.1, 114.9, 118.1, 122.1, 126.0, 127.8, 128.1, 135.4, 137.6, 145.9, 149.1, 195.7.

3,3,6,6-tetramethyl-9-phenyl-10-propyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)dione (4t) (Table 4, entry 20)

Light yellow solid; Yield = 94%, M.P. 180-182 °C (Lit. new); IR (KBr): \bar{u} (cm⁻¹) = 2958, 1640, 1384, 1212. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.27 (d, *J* = 6.08 Hz, 2H, Ar*H*), 7.18 (t, *J* = 15.00 Hz, 2H, Ar*H*), 7.06 (t, *J* = 7.24 Hz, 1H, Ar*H*), 5.28 (s, 1H, C*H*Ar), 3.62 (t, *J* = 7.68 Hz, 2H, C*H*₂N), 2.54 (d, *J* = 16.60 Hz, 2H, C*H*₂), 2.41 (d, *J* = 16.72 Hz, 2H, C*H*₂), 2.31 (d, *J* = 7.24 Hz, 2H, C*H*₂), 2.23 (d, *J* = 8.24 Hz, 2H, C*H*₂), 1.68 (sext, *J* = 8.00 Hz, 2H, C*H*₂CH₃), 1.11 (s, 6H), 1.01 (s, 6H, 2C*H*₃), 0.90 (t, *J* = 3.06 Hz, 3H, 2C*H*₃), 2.41 (d, *J* = 16.72 Hz, 2H, C*H*₃). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 11.05, 24.70, 29.36, 31.84, 32.55, 40.47, 46.44, 49.96, 115.41, 125.81, 127.62, 127.95, 145.74, 150.16, 195.63.

3,3,6,6,10-pentamethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4u) (Table 4, entry 21)

Light yellow solid; Yield = 96%, M.P. 232-233 °C (Lit. 200-202 °C^[14]); IR (KBr): \bar{u} (cm⁻¹) = 3470, 29.31, 16.17, 1325. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 7.44 (t, *J* = 8.34 Hz, 4H, Ar*H*), 7.33-7.36 (m, 1H, Ar*H*), 4.89 (s, 1H, C*H*Ar), 3.72 (s, 1H, C*H*₃N), 2.19 (d, *J* = 8.20 Hz, 2H, C*H*₂), 2.14 (d, *J* = 5.28 Hz, 2H, C*H*₂), 2.09 (d, *J* = 5.76 Hz, 2H, C*H*₂), 2.05 (d, *J* = 5.28 Hz, 2H, C*H*₂), 1.19 (s, 6H, 2C*H*₃), 1.07 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 26.81, 28.99, 34.99, 38.00, 50.62, 113.26, 128.14, 128.47, 128.61, 146.84, 153.70, 188.98.

9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4v) (Table 4, entry 22)

Light yellow solid; Yield = 92%, M.P. 307-308 °C (Lit. 310 °C ^[3]); IR (KBr): \bar{u} (cm⁻¹) = 3449, 2929, 1620, 1372. ¹H NMR (400 MHz, CDCl₃) δ (ppm): 9.33 (s, 1H, N-*H*), 7.33 (d, *J* = 7.80 Hz, 4H, Ar*H*), 7.13-7.05 (m, 1H, Ar*H*), 4.96 (s, 1H, C*H*Ar), 3.26 (t, *J* = 7.32 Hz, 4H C*H*₂CO), 2.98 (t, *J* = 6.12 Hz, 4H C*H*₂), 1.83 (quin, 4H, C*H*₂CH₂CO). ¹³C NMR (101MHz, CDCl₃) δ (ppm): 21.05, 28.39, 30.05, 35.43, 112.19, 131.17, 132.78, 134.19, 142.67, 150.61, 195.72.

1,1'-(2,6-dimethyl-4-phenyl-1,4-dihydropyridine-3,5-diyl)bis(ethan-1-one)(4w)(Table 4, entry 23)

Yellow solid; Yield = 94%, M.P. 186-187 °C (Lit. 184-186 °C ^[15]); IR (KBr): \bar{u} (cm⁻¹) = 3407, 2915, 1621, 1397. ¹H NMR (400 MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 8.98 (s, 1H, N-*H*), 7.33 (d, *J* = 19.88 Hz, 4H, Ar*H*), 7.17-7.10 (m, 1H, Ar*H*), 4.88 (s, 1H, C*H*Ar), 2.38 (s, 6H, 2C*H*₃CO), 2.16 (s, 6H, 2C*H*₃). ¹³C NMR (101MHz, DMSO-*d*₆) $\bar{\delta}$ (ppm): 20.06, 27.21, 33.96, 112.85, 128.09, 128.78, 129.61, 143.48, 147.28, 198.59.



Figure S1: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in DMSO- d_6



Figure S2: The ¹³C NMR spectrum (101 MHz) of spectrum of 3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in DMSO- d_6



Figure S3: The FT-IR spectrum of 3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S4: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9-(2-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in CDCl₃



Figure S5: The ¹³C NMR spectrum (101 MHz) of spectrum of 3,3,6,6-tetramethyl-9-(2-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S6: The FT-IR spectrum of 3,3,6,6-tetramethyl-9-(2-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S7: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9-(3-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S8: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6-tetramethyl-9-(3-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in DMSO- d_6



Figure S9: The FT-IR spectrum of 3,3,6,6-tetramethyl-9-(3-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S10: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S11: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S12: The FT-IR spectrum of 3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S13: The ¹H NMR spectrum of (400 MHz) 9-(2-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S14: The ¹³C NMR spectrum (101 MHz) of 9-(2-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S15: The FT-IR spectrum of 9-(2-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S16: The ¹H NMR spectrum (400 MHz) of 9-(3-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in $CDCl_3$



Figure S17: The ¹³C NMR spectrum (101 MHz) of 9-(3-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione (4f) in CDCl₃



Figure S18: The FT-IR spectrum of 9-(3-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S19: The ¹H NMR spectrum (400 MHz) of 9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S20: The ¹³C NMR spectrum (101 MHz) of 9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S21: The FT-IR spectrum of 9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S22: The ¹H NMR spectrum (400 MHz) of 9-(2-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S23: The ¹³C NMR spectrum (101 MHz) of 9-(2-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S24: The FT-IR spectrum of hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr

9-(2-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-



Figure S25: The ¹H NMR spectrum (300 MHz)of 9-(5-bromo-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO-d₆



Figure S26: The ¹³C NMR spectrum (76 MHz) of 9-(5-bromo-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in DMSO- d_6



Figure S27: The FT-IR spectrum of 9-(5-bromo-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S28: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9-(*m*-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S29: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6-tetramethyl-9-(*m*-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S30: The FT-IR spectrum of 3,3,6,6-tetramethyl-9-(*m*-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S31: The ¹H NMR spectrum (400 MHz) of 3,3,6,6,9-pentamethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S32: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6,9-pentamethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S33: The FT-IR spectrum of 3,3,6,6,9-pentamethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S34: The ¹H NMR spectrum (400 MHz) of 9-ethyl-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in $CDCI_3$



Figure S35: The ^{13}C NMR spectrum (101 MHz) of 9-ethyl-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione CDCl_3



Figure S36: The FT-IR spectrum of 9-ethyl-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in KBr



Figure S37: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9,10-diphenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S38: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6-tetramethyl-9,10-diphenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S39: The FT-IR spectrum of 3,3,6,6-tetramethyl-9,10-diphenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S40: The ¹H NMR spectrum (400 MHz) of 10-(3-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S41: The ¹³C NMR spectrum (101 MHz) of 10-(3-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S42: The FT-IR spectrum of 10-(3-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S43: The ¹H NMR spectrum of (400 MHz) 10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S44: The ¹³C NMR spectrum (101 MHz) of 10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in DMSO- d_6



Figure S45: The FT-IR spectrum of 10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S46: The ¹H NMR spectrum (400 MHz) of 10-(2-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in CDCl₃



Figure S47: The ¹³C NMR spectrum (101 MHz) of 10-(2-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in CDCl₃



Figure S48: The FT-IR spectrum of 10-(2-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S49: The ¹H NMR spectrum of (300 MHz) 10-(3-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in DMSO- d_6



Figure S50: The ¹³C NMR spectrum (76 MHz) of 10-(3-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in DMSO- d_6



Figure S51: The FT-IR spectrum of 10-(3-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S52: The ¹H NMR spectrum (400 MHz) of 10-(4-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in CDCl₃



Figure S53: The ¹³C NMR spectrum (101 MHz) of 10-(4-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in CDCl₃



Figure S54: The FT-IR spectrum of 10-(4-chlorophenyl)-3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2*H*,5*H*)-dione in KBr



Figure S55: The ¹H NMR spectrum (400 MHz) of 3,3,6,6-tetramethyl-9-phenyl-10-propyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in $CDCl_3$



Figure S56: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6-tetramethyl-9-phenyl-10-propyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in $CDCl_3$



Figure S57: The FT-IR spectrum of 3,3,6,6-tetramethyl-9-phenyl-10-propyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in KBr



Figure S58: The ¹H NMR spectrum (400 MHz) of 3,3,6,6,10-pentamethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S59: The ¹³C NMR spectrum (101 MHz) of 3,3,6,6,10-pentamethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S60: The FT-IR spectrum of 3,3,6,6,10-pentamethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in KBr



Figure S61: The ¹H NMR spectrum (400 MHz) of 9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in $CDCI_3$



Figure S62: The 13 C NMR spectrum (101 MHz) of 9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in CDCl₃



Figure S63: The FT-IR spectrum of 9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione in KBr



Figure S64: The ¹H NMR spectrum (400 MHz) of 1,1'-(2,6-dimethyl-4-phenyl-1,4-dihydropyridine-3,5-diyl)bis(ethan-1-one) in DMSO- d_6



Figure S65: The ¹³C NMR spectrum (101 MHz) of 1,1'-(2,6-dimethyl-4-phenyl-1,4-dihydropyridine-3,5-diyl)bis(ethan-1-one) in DMSO- d_6



Figure S66: The FT-IR spectrum of 1,1'-(2,6-dimethyl-4-phenyl-1,4-dihydropyridine-3,5-diyl)bis(ethan-1-one) in KBr

References

[1] Aher, D. S.; Khillare, K. R.; Shankarwar, S. G. Incorporation of Keggin-Based $H_3PW_7Mo_5O_{40}$ into Bentonite: Synthesis, Characterization and Catalytic Applications. *RSC Advances* **2021**, *11* (19), 11244-11254. https://doi.org/10.1039/d1ra01179k.

[2] Zolfigol, M. A.; Karimi, F.; Yarie, M.; Torabi, M. Catalytic Application of Sulfonic acid-functionalized titana-coated Magnetic Nanoparticles for the Preparation of 1,8-dioxodecahydroacridines and 2,4,6-triarylpyridines via anomeric-based Oxidation. *Applied Organometallic Chemistry* **2017**, *32* (2). https://doi.org/10.1002/aoc.4063.

[3] Zhu, A.; Liu, R.; Du, C.; Li, L. Betainium-Based Ionic Liquids Catalyzed Multicomponent Hantzsch Reactions for the Efficient Synthesis of Acridinediones. *RSC Advances* **2017**, 7 (11), 6679-6684. <u>https://doi.org/10.1039/c6ra25709g</u>.

[4] Navarro, C. A.; Sierra, C. A.; Ochoa-Puentes, C. Evaluation of Sodium Acetate trihydrate–urea DES As a Benign Reaction Media for the Biginelli Reaction. Unexpected Synthesis of methylenebis(3-Hydroxy-5,5-Dimethylcyclohex-2-enones), Hexahydroxanthene-1,8-Diones and Hexahydroacridine-1,8-Diones. *RSC Advances* **2016**, *6* (70), 65355-65365. <u>https://doi.org/10.1039/c6ra13848a</u>.

[5] Tiwari, K. N.; Uttam, M. R.; Kumari, P.; Vatsa, P.; Prabhakaran, S. M. Efficient Synthesis of Acridinediones in Aqueous Media. *Synthetic Communications* **2017**, *47* (10), 1013-1019. https://doi.org/10.1080/00397911.2017.1304556.

[6] Eidi, E.; Kassaee, M. Z.; Nasresfahani, Z. Nanocrystalline TiO₂, via Green Combustion Synthesis, As an Efficient and Reusable Catalyst for the Preparation of 1,8-Dioxooctahydroxanthenes and 1,8-Dioxodecahydroacridines. *Applied Organometallic Chemistry* **2015**, 29 (12), 793-797. <u>https://doi.org/10.1002/aoc.3370</u>.

[7] Taheri-Ledari, R.; Esmaeili, M. S.; Varzi, Z.; Eivazzadeh-Keihan, R.; Maleki, A.; Shalan, A. E. Facile Route to Synthesize Fe₃O₄@acacia–SO₃H Nanocomposite As a Heterogeneous Magnetic System for Catalytic Applications. *RSC Advances* **2020**, *10* (66), 40055-40067. <u>https://doi.org/10.1039/d0ra07986c</u>.

[8] Kumar, A.; Madderla, S.; Dharavath, R.; Nalaparaju, N.; Katta, R.; Gundu, S.; Thumma, V.; Prashanth, B.; Ashok, D. Microwave assisted synthesis of N-substituted acridine-1, 8-dione derivatives: Evaluation of antimicrobial activity. *Journal of Heterocyclic Chemistry* **2022**, *59* (7), 1180-1190.

[9] Tu, S., Miao, C., Gao, Y., Fang, F., Zhuang, Q., Feng, Y., & Shi, D. (2004). A Novel Cascade reaction of aryl aldoxime with dimedone under microwave irradiation: The synthesis of N-hydroxylacridine. *Synlett*, *2004*(02), 0255-0258.

[10] Patil, D., Chandam, D., Mulik, A., Patil, P., Jagadale, S., Kant, R., ... & Deshmukh, M. (2014). Novel Brønsted acidic ionic liquid ([CMIM][CF₃COO]) prompted multicomponent hantzsch reaction for the eco-friendly synthesis of acridinediones: An efficient and recyclable catalyst. *Catalysis letters*, *144*, 949-958.

[11] Gholami Dehbalaei, M.; Foroughifar, N.; Pasdar, H.; Khajeh-Amiri, A. *N*-Propyl Benzoguanamine Sulfonic Acid Supported on Magnetic Fe₃O₄ Nanoparticles: A Novel and Efficient Magnetically Heterogeneous Catalyst for the Synthesis of 1,8-Dioxo-Decahydroacridine Derivatives. *New Journal of Chemistry* **2018**, *42* (1), 327-335. https://doi.org/10.1039/c7nj03508j.

[12] Chate, A. V.; Sukale, S. B.; Ugale, R. S.; Gill, C. H. Baker's Yeast: An Efficient, Green,
and Reusable Biocatalyst for the One-Pot Synthesis of Biologically Important *N*-
Substituted Decahydroacridine-1,8-Dione Derivatives. Synthetic
Communications 2017, 47 (5), 409-420.
https://doi.org/10.1080/00397911.2016.1266501.

[13] Zarei, Z.; Akhlaghinia, B. Zn^{II} Doped and Immobilized on Functionalized Magnetic Hydrotalcite (Fe₃O₄/HT-SMTU-Zn^{II}): A Novel, Green and Magnetically Recyclable Bifunctional Nanocatalyst for the One-Pot Multi-Component Synthesis of Acridinediones under Solvent-Free Conditions. *New Journal of Chemistry* **2017**, *41* (24), 15485-15500. https://doi.org/10.1039/c7nj03281a.

[14] Srividya, N., Ramamurthy, P., Shanmugasundaram, P., & Ramakrishnan, V. T. (1996). Synthesis, characterization, and electrochemistry of some acridine-1, 8-dione dyes. *The Journal of Organic Chemistry*, *61*(15), 5083-5089.

[15] Wang, J. L., Liu, B. K., Yin, C., Wu, Q., & Lin, X. F. (2011). Candida antarctica lipase B-catalyzed the unprecedented three-component Hantzsch-type reaction of aldehyde with acetamide and 1, 3-dicarbonyl compounds in non-aqueous solvent. *Tetrahedron*, *67*(14), 2689-2692.