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An Efficient Fe/H_2O Medium in situ Reduction and Cyclization Reaction for the Synthesis of Pyrazolo[3,4-*a*]acridin-10-one and Pyrazolo[4,3-*a*]acridin-10-one Derivatives

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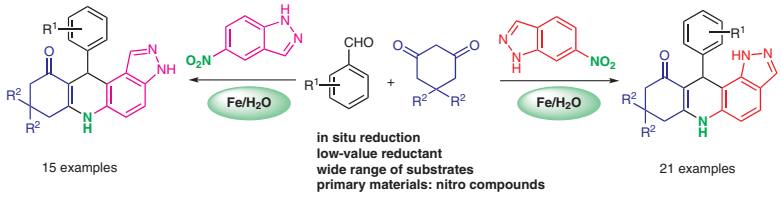

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Abstract An efficient and simple method for the synthesis of pyrazolo[3,4-*a*]acridine and pyrazolo[4,3-*a*]acridine derivatives directly form nitro compounds by in situ reduction and cyclization reaction under Fe/H_2O medium is reported. Compared to amino compounds, nitro compounds are more stable and easier to obtain. In addition, because iron is a nontoxic, inexpensive, and environmentally friendly reductant, this method is especially suitable for organic synthesis. The other advantages of this process are cheap raw materials, less pollution, and wide substrate range.

Key words in situ reductions, nitro compounds, iron, pyrazolo-[3,4-*a*]-acridin-10-one, pyrazolo[4,3-*a*]acridin-10-one

Nitrogen containing heterocycles are important organic compounds and exhibit numerous applications in many disciplines.[1–3] Pyrazole is an important part of nitrogen heterocyclic compounds and displays various chemical, biological, agrochemical, and pharmacological properties.[4] Therefore, several drugs have been developed from the pyrazole mother nucleus. Moreover, studies showed many fused pyrazoles also have biological and pharmacological activities.[5]

Acridine and its derivative are the other class of nitrogen containing heterocyclic compounds, which have demonstrated important biological activities, such as antiviral,[6] anti-inflammatory,[7] analgesic,[8] anticancer,[9] and so on. In fact, they widely exist in many natural products, and are also some drug candidate structures (Alzheimer's disease,[10] and HIV/AIDS[11]). Interestingly, they also have several significant optical properties, such as pigments and dyes,[12] and photochemical/physical properties.[13]

Nitro compounds are important reagents and famous precursors of many valuable organic products.[14] The reduction of nitro compounds always leads to various products, such as amines, hydroxylamines, hydrazines, nitroso, azo, or azoxy compounds. In addition, nitro compounds reacted with other reagents under the reduction condition could generate nitrogen heterocyclic compounds.[15]

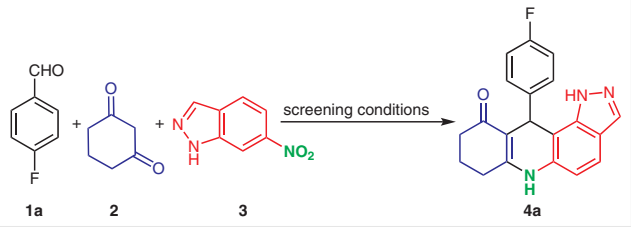
Iron [Fe(0)] can be naturally used as a reducing agent because it is one of the most inexpensive, abundant, nontoxic, and environmentally friendly transition metals. Recently, iron and iron salts are widely used in organic synthesis.[16] Considering that the nitro compounds are more stable and easier to obtain, we decided to use nitro compounds to synthesize some practically useful organic compounds. In this research, we planned to synthesize nitrogen containing heterocycles (pyrazolo[3,4-*a*]acridines and pyrazolo[4,3-*a*]acridines) with both pyrazole and acridine moieties from nitro compounds (5-nitro-1*H*-indazole and 6-nitro-1*H*-indazole) using Fe(0) as a reductant.

It could be found that amino compounds were the main reagents for the synthesis of nitrogen containing compounds. For instance, 4-aryl-1,4-dihydroquinolines, 4-aryl-dihydropyridines, and 5-arylpyrimido[4,5-*b*]quinoline-diones, and 2-amino-4-arylquinoline-3-carbonitriles were successfully synthesized by three research groups.[17] A one-pot synthesis of quinolines was reported from three-component reactions of amines, aldehydes, and alkynes.[18] Isoindolo[2,1-*a*]quinoline system was easily prepared using ethyl 4-aminobenzoate, 2-carboxybenzaldehyde, and cyclopentadiene as reagents.[19] The three-component reactions involving 1,3-diones, aldehydes, and aromatic amines to synthesize the corresponding acridine derivatives were successfully reported.[20] Similarly, amino compounds are also the main substance in the synthesis of acridine derivatives. For example, Nadaraj and co-workers reported the synthesis of acridine derivatives from the reactions of dimedone or cyclohexan-1,3-dione, α-naphthylamine, and various substituted benzaldehydes.[21] Similar reaction was carried out via the three-component coupling from aromatic aldehydes, 1-naphthylamine, and 5,5-dimethylcyclo-

hexane-1,3-dione.[22] Wang also reported a three-component reaction of an *o*-halogenated benzaldehyde, 1*H*-indazol-6-amine, and cyclohexane-1,3-dione for preparation of fused acridine derivatives.[23] Additionally, many other methods were reported for the synthesis of acridine derivatives from various amino compounds by different research groups.[24]

After reviewing the literature, we found that only few methods were reported for the synthesis of title compounds from aminoindazole.[25] It should be noted that these methods also used amino compounds as key substrates. In view of the importance of the previous detailed structures, we hope to find a simpler reagent to synthesize the target product.

Initially, 4-fluorobenzaldehyde (**1a**), cyclohexane-1,3-dione (**2**), and 6-nitro-1*H*-indazole (**3**) were chosen as model substrates to standardize the reaction conditions (Scheme 1). When the reaction was performed in the presence of two equivalents of Fe(0) and the additives H_2O (1 mL) and AcOH (1 mL) in different solvents at 60 °C, we were pleased to find that a better yield could be obtained in EtOH medium (Table 1 entries 1–3). Then, the reaction was tested in EtOH under different ratio of **3** and Fe(0). The results revealed that the superior yield (entries 4–6) was obtained when three equivalents of Fe(0) were used. The amount of the additives was also evaluated, and the results showed that water (1 mL) and AcOH (1 mL) could guarantee the excellent results (entries 7–9). We further found that 6 mL of EtOH and 80 °C could give the best results (entries 10–14). EtOH, H_2O, and AcOH all play decisive roles in the reaction, without any of them, the reaction did not work well (entries 15–17). The results are summarized in Table 1.



Scheme 1 The model reaction for the synthesis of compound **4a**

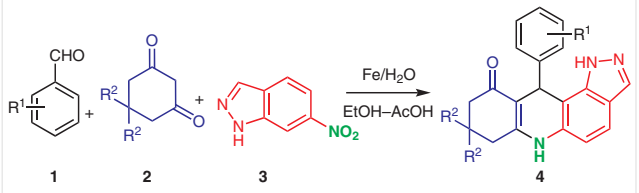
With the optimized conditions in hand, the scope of the reaction was then investigated using a wide range of aromatic aldehydes **1**, 6-nitro-1*H*-indazole (**3**) , and cyclohexane-1,3-diones **2** (Scheme 2). Aromatic aldehydes bearing electron-donating (OH, Me, MeO), or electron-withdrawing (F, Cl, Br,) substituents were successfully applied in this synthesis, and the desired pyrazolo[3,4-*a*]acridine derivatives **4a–g** were obtained in good yields. Dimedone was also used to react with aromatic aldehydes and 6-nitro-1*H*-indazole under preferred conditions, and to our delight, the corresponding products **4h–u** were obtained in excellent

Table 1 Screening of the Reaction Conditions[a]

Entry	Solvent	Additive	Ratio (**3**/Fe)	Temp (°C)	Yield (%)[b]
1	EtOH (4 mL)	H_2O (1 mL)/AcOH (1 mL)	1:2	60	35
2	MeOH (4 mL)	H_2O (1 mL)/AcOH (1 mL)	1:2	60	23
3	THF (4 mL)	H_2O (1 mL)/AcOH (1 mL)	1:2	60	12
4	EtOH (4 mL)	H_2O (1 mL)/AcOH (1 mL)	1:3	60	58
5	EtOH (4 mL)	H_2O (1 mL)/AcOH (1 mL)	1:4	60	55
6	EtOH (4 mL)	H_2O (1 mL)/AcOH (1 mL)	1:5	60	50
7	EtOH (4 mL)	H_2O (2 mL)/AcOH (1 mL)	1:3	60	45
8	EtOH (4 mL)	H_2O (1 mL)/AcOH (2 mL)	1:3	60	41
9	EtOH (4 mL)	H_2O (2 mL)/AcOH (2 mL)	1:3	60	30
10	EtOH (6 mL)	H_2O (1 mL)/AcOH (1 mL)	1:3	60	65
11	EtOH (8 mL)	H_2O (1 mL)/AcOH (1 mL)	1:3	60	63
12	EtOH (6 mL)	H_2O (1 mL)/AcOH (1 mL)	1:3	70	73
13	EtOH (6 mL)	H_2O (1 mL)/AcOH (1 mL)	1:3	80	86
14	EtOH (6 mL)	H_2O (1 mL)/AcOH (1 mL)	1:3	90	74
15	EtOH (6 mL)	H_2O (1 mL)	1:3	80	0
16	EtOH (6 mL)	AcOH (1 mL)	1:3	80	32
17	–	H_2O (1 mL)/AcOH (1 mL)	1:3	80	0

[a] Conditions: 4-fluorobenzaldehyde (**1a**; 1 mmol), cyclohexane-1,3-dione (**2**; 1 mmol), 6-nitro-1*H*-indazole (**3**; 1 mmol), reaction time: ca. 6 h (monitored by TLC).
[b] Isolated yield.

yields. The results show that different electronic properties of substituents have no effect on the reaction. The results are listed in Table 2.



Scheme 2 Synthesis of compounds **4**

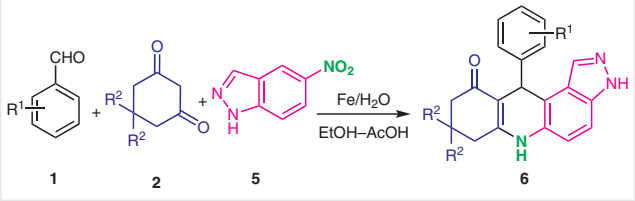
In order to test this synthetic method, further researches were performed with 5-nitro-1*H*-indazole (**5**), aromatic aldehydes **1** (1 mmol), and cyclohexane-1,3-diones **2** under optimized conditions (Scheme 3). Interestingly, the desired pyrazolo[4,3-*a*]acridine products **6a–o** were formed in good to excellent yields in all cases. Similarly, the electron-donating (Me, OCH_2O) and electron-withdrawing (F, Cl, Br) substituents of aromatic aldehydes have little effect on the reactions. Furthermore, the application of the current methodology to dimedone with aromatic aldehydes and 5-nitro-1*H*-indazole was investigated. As expected, the reactions could be carried out very well and the desired products were produced in high yields. The results are

Table 2 Compounds **4** Prepared[a]

Entry	R^1	R^2	Time (h)	Product	Yield (%)[b]
1	4-F	H	6	**4a**	86
2	4-Cl	H	6	**4b**	88
3	3-Br	H	6	**4c**	84
4	4-Me	H	6	**4d**	89
5	3-MeO	H	6	**4e**	85
6	3,4-(OCH_2O)-	H	6	**4f**	84
7	4-HO	H	6	**4g**	88
8	4-H	Me	7	**4h**	83
9	4-F	Me	6	**4i**	89
10	3-Cl	Me	6	**4j**	86
11	4-Cl	Me	6	**4k**	87
12	2,4-Cl_2	Me	7	**4l**	81
13	3-Br	Me	6	**4m**	83
14	4-Br	Me	6	**4n**	88
15	4-Me	Me	6	**4o**	86
16	3,4-Me_2	Me	7	**4p**	83
17	2-MeO	Me	6	**4q**	84
18	3-MeO	Me	6	**4r**	81
19	3-MeO-4-HO	Me	7	**4s**	85
20	4-MeO	Me	6	**4t**	87
21	3,4-(OCH_2O)-	Me	6	**4u**	86

[a] Conditions: aromatic aldehyde **1** (1 mmol), cyclohexane-1,3-dione (dimedone) **2** (1 mmol), 6-nitro-1*H*-indazole (**3**; 1 mmol), reaction time: ca. 6 h (monitored by TLC).
[b] Isolated yield.

summarized in Table 3. In our synthetic strategy, nitro compounds can be successfully reduced by iron to yield the expected products.



Scheme 3 Synthesis of compounds **6**

The structures of the products were characterized by ^1H NMR, ^{13}C NMR, IR, and HRMS. We take **4a** as an example to analyze their structures. In its ^1H NMR, the singlet signals appear at δ = 12.92 (s, 1 H), and 9.72 (s, 1 H) due to the NH protons. The tertiary hydrogen chemical shift occurs near the 5.58 ppm. Two methylene groups show as the multiple peaks at δ = 2.65–2.60 (m, 2H) and 2.29–2.21 (m, 2H), respectively, while, another methylene group was split into two parts, appearing at δ = 1.98–1.92 (m, 1 H) and 1.87–1.80 (m, 1 H). Seven aromatic protons appear from 7.93–6.88 ppm. The structure analysis process for compound **6** is similar to that of **4** (see Supporting Information).

Table 3 Compounds **6** Prepared[a]

Entry	R^1	R^2	Time (h)	Product	Yield (%)[b]
1	4-F	H	6	**6a**	85
2	4-Cl	H	6	**6b**	83
3	3,4-Cl_2	H	6	**6c**	86
4	4-Br	H	6	**6d**	88
5	4-Me	H	7	**6e**	81
6	3,4-Me_2	H	7	**6f**	80
7	3,4-(OCH_2O)-	H	7	**6g**	89
8	4-F	Me	6	**6h**	87
9	4-Cl	Me	6	**6i**	85
10	3,4-Cl_2	Me	6	**6j**	82
11	4-Br	Me	6	**6k**	90
12	4-Me	Me	7	**6l**	84
13	3,4-Me_2	Me	6	**6m**	80
14	3,4,5-$(MeO)_3$	Me	6	**6n**	83
15	3,4-(OCH_2O)-	Me	7	**6o**	85

[a] Conditions: aromatic aldehyde **1** (1 mmol), cyclohexane-1,3-dione (dimedone) **2** (1 mmol), 5-nitro-1*H*-indazole (**5**; 1 mmol), reaction time: ca. 6 h (monitored by TLC).
[b] Isolated yield.

In conclusion, we have reported a simple and efficient method for the synthesis of pyrazolo[3,4-*a*]acridine and pyrazolo[4,3-*a*]acridine derivatives directly form nitro compounds by in situ reduction and cyclization reaction in Fe/H_2O medium. Compared with using amino compounds, nitro compounds are more stable and more readily available. Other advantages of this method are cheap raw materials, less pollution, simple operation, and use of wide range of substrates.

All reagents were purchased from Merck and Sigma-Aldrich and used without further purification. Melting points were determined on XT-5 microscopic melting point apparatus and are uncorrected. IR spectra were recorded on a FT Bruker Tensor 27 spectrometer. ^1H NMR and ^{13}C NMR spectra of **4** and **6** were obtained from solution in DMSO-d_6 with Me_4Si as internal standard using a Bruker-400 spectrometer under r.t. conditions. HRMS spectra were obtained with a Bruker microTOF-Q 134 instrument.

Pyrazolo[3,4-*a*]acridines 4 and Pyrazolo[4,3-*a*]acridines 6; General Procedure

Aromatic aldehyde **1** (1 mmol), cyclohexane-1,3-dione (dimedone) **2**, 6-nitro-1*H*-indazole (**3**) or 5-nitro-1*H*-indazole **5** (1 mmol), Fe (3 mmol), EtOH (6 mL), H_2O (1 mL), and AcOH (1 mL) were placed into a 25 mL round-bottom flask. Then, the mixture was stirred at 80 °C for about 6 h (monitored by TLC). After completion of the reaction, brine

(8 mL) was added to the reaction system. The mixture was transferred to a separatory funnel, and was extracted with EtOAc (3 × 15 mL). The organic layers were combined and washed thoroughly with brine, dried (anhyd Na_2SO_4), and filtered through Celite. Following removal of the solvent in vacuo, the residue was purified by crystallization from DMF to give the pure product **4** or **6**.

11-(4-Fluorophenyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]-acridin-10-one (4a)

Yellow solid; yield: 286.7 mg (86%); mp >280 °C.

IR (KBr): 3432, 1668, 1631, 1507, 1424, 1395, 1257, 1161, 1028, 928, 845, 763, 665, 535, 480 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.92 (s, 1 H, NH), 9.72 (s, 1 H, NH), 7.93 (s, 1 H, ArH), 7.52 (d, J = 8.4 Hz, 1 H, ArH), 7.37 (dd, J_1 = 8.4 Hz, J_2 = 6.0 Hz, 2 H, ArH), 6.97 (t, J = 8.8 Hz, 2 H, ArH), 6.88 (d, J = 8.4 Hz, 1 H, ArH), 5.58 (s, 1 H, CH), 2.65–2.60 (m, 2 H, CH_2), 2.29–2.21 (m, 2 H, CH_2), 1.98–1.92 (m, 1 H, CH), 1.87–1.80 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 162.1, 159.7, 153.8, 143.7 (d, $J_{C,F}$ = 2.9 Hz), 139.1, 134.6, 129.6 (d, $J_{C,F}$ = 7.9 Hz), 120.2, 119.8, 114.9 (d, $J_{C,F}$ = 20.9 Hz), 111.8, 108.4, 106.2, 37.2, 35.0, 27.5, 21.5.

HRMS (ESI-TOF): m/z calcd for $C_{20}H_{16}FN_3ONa$ [M + Na]$^+$: 356.1175; found: 356.1179.

11-(4-Chlorophenyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]-acridin-10-one (4b)

Yellow solid; yield: 307.8 mg (88%); mp 217–219 °C.

IR (KBr): 3421, 3269, 1703, 1610, 1539, 1488, 1393, 1320, 1224, 1128, 1086, 1013, 930, 738, 615 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.90 (s, 1 H, NH), 9.75 (s, 1 H, NH), 7.94 (s, 1 H, ArH), 7.53 (d, J = 8.8 Hz, 1 H, ArH), 7.36 (d, J = 8.4 Hz, 2 H, ArH), 7.21 (d, J = 8.4 Hz, 2 H, ArH), 6.89 (d, J = 8.4 Hz, 1 H, ArH), 5.57 (s, 1 H, CH), 2.63 (d, J = 4.0 Hz, 2 H, CH_2), 2.29–2.18 (m, 2 H, CH_2), 1.98–1.92 (m, 1 H, CH), 1.88–1.79 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 153.9, 146.4, 134.6, 130.8, 129.7, 128.2, 120.2, 119.9, 111.8, 108.1, 105.9, 37.2, 35.2, 27.5, 21.5.

HRMS (ESI-TOF): m/z calcd for $C_{20}H_{16}ClN_3ONa$ [M + Na]$^+$: 372.0880; found: 372.0885.

11-(3-Bromophenyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]-acridin-10-one (4c)

Yellow solid; yield: 331.2 mg (84%); mp >280 °C.

IR (KBr): 3327, 3180, 1660, 1595, 1493, 1385, 1285, 1199, 1102, 928, 842, 788, 729, 690, 594 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.94 (s, 1 H, NH), 9.77 (s, 1 H, NH), 7.95 (s, 1 H, NH), 7.64 (s, 1 H, ArH), 7.55 (d, J = 8.4 Hz, 1 H, ArH), 7.66–7.22 (m, 2 H, ArH), 7.11 (t, J = 8.0 Hz, 1 H, ArH), 6.89 (d, J = 8.8 Hz, 1 H, ArH), 5.55 (s, 1 H, CH), 2.67–2.60 (m, 2 H, CH_2), 2.31–2.20 (m, 2 H, CH_2), 1.98–1.92 (m, 1 H, CH), 1.87–1.79 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 154.0, 150.0, 134.7, 130.7, 130.7, 129.1, 126.9, 121.7, 120.2, 120.1, 111.8, 107.9, 37.1, 35.6, 27.5, 21.5.

HRMS (ESI-TOF): m/z calcd for $C_{20}H_{16}BrN_3ONa$ [M + Na]$^+$: 416.0374; found: 416.0399.

11-(*p*-Tolyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4d)

Yellow solid; yield: 293.2 mg (89%); mp 243–245 °C.

IR (KBr): 3269, 1706, 1590, 1491, 1450, 1393, 1283, 1197, 1049, 931, 852, 805, 759, 638 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.86 (s, 1 H, NH), 9.65 (s, 1 H, NH), 7.91 (s, 1 H, ArH), 7.50 (d, J = 8.4 Hz, 1 H, ArH), 7.24 (d, J = 8.0 Hz, 2 H, ArH), 6.93 (d, J = 8.0 Hz, 2 H, ArH), 6.86 (d, J = 8.4 Hz, 1 H, ArH), 5.52 (s, 1 H, CH), 2.64–2.59 (m, 2 H, CH_2), 2.28–2.21 (m, 2 H, CH_2), 2.14 (s, 3 H, CH_3), 1.98–1.92 (m, 1 H, CH), 1.87–1.79 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.8, 153.6, 144.7, 139.2, 135.1, 134.6, 134.4, 128.8, 127.8, 120.2, 119.5, 111.8, 108.6, 106.6, 37.2, 35.2, 27.5, 21.5, 21.0.

HRMS (ESI-TOF): m/z calcd for $C_{21}H_{19}N_3ONa$ [M + Na]$^+$: 352.1426; found: 352.1438.

11-(3-Methoxyphenyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]-acridin-10-one (4e)

Yellow solid; yield: 293.6 mg (85%); mp 270–272 °C.

IR (KBr): 3421, 3256, 1657, 1598, 1540, 1492, 1384, 1259, 1197, 1080, 1061, 1038, 928, 844, 754, 665 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.90 (s, 1 H, NH), 9.67 (s, 1 H, NH), 7.92 (s, 1 H, ArH), 7.51 (d, J = 8.4 Hz, 1 H, ArH), 7.03 (q, J = 8.0 Hz, 2 H, ArH), 6.86 (d, J = 8.4 Hz, 2 H, ArH), 6.61 (dd, J_1 = 8.0 Hz, J_2 = 2.0 Hz, 1 H, ArH), 5.52 (s, 1 H, CH), 3.66 (s, 3 H, CH_3), 2.68–2.61 (m, 2 H, CH_2), 2.28–2.23 (m, 2 H, CH_2), 1.99–1.94 (m, 1 H, CH), 1.85–1.84 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 159.3, 153.8, 149.0, 134.4, 129.3, 120.1, 119.6, 114.4, 111.8, 111.0, 108.3, 100.0, 55.3, 37.2, 35.6, 27.5, 21.3.

HRMS (ESI-TOF): m/z calcd for $C_{21}H_{19}N_3O_2Na$ [M + Na]$^+$: 368.1375; found: 368.1380.

11-(Benzo[*d*][1,3]dioxol-5-yl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4f)

Yellow solid; yield: 301.9 mg (84%); mp 269–271 °C.

IR (KBr): 3427, 3135, 1593, 1540, 1486, 1390, 1250, 1228, 1193, 1038, 929 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.87 (s, 1 H, NH), 9.76 (s, 1 H, NH), 7.93 (s, 1 H, ArH), 7.51 (d, J = 8.4 Hz, 1 H, ArH), 6.94 (d, J = 1.6 Hz, 1 H, ArH), 6.89 (d, J = 8.8 Hz, 1 H, ArH), 6.78 (dd, J_1 = 8.0 Hz, J_2 = 1.6 Hz, 1 H, ArH), 6.67 (d, J = 8.0 Hz, 1 H, ArH), 5.87 (d, J = 5.6 Hz, 2 H, CH_2), 5.48 (s, 1 H, CH), 2.68–2.58 (m, 2 H, CH_2), 2.29–2.21 (m, 2 H, CH_2), 1.97–1.91 (m, 1 H, CH), 1.88–1.78 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 153.7, 147.2, 145.6, 141.8, 139.2, 134.5, 134.4, 120.7, 120.1, 119.6, 111.8, 108.6, 108.5, 108.1, 106.6, 101.0, 37.2, 35.3, 27.5, 21.5.

HRMS (ESI-TOF): m/z calcd for $C_{21}H_{17}N_3O_3Na$ [M + Na]$^+$: 382.1168; found: 382.1181.

11-(4-Hydroxyphenyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]-acridin-10-one (4g)

Yellow solid; yield: 291.6 mg (88%), mp 240–242 °C.

IR (KBr): 3268, 3150, 1667, 1593, 1479, 1390, 1253, 1197, 1127, 976, 933, 839, 764, 640, 592 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.84 (s, 1 H, NH), 9.68 (s, 1 H, NH), 9.06 (s, 1 H, OH), 7.91 (s, 1 H, ArH), 7.48 (d, J = 8.4 Hz, 1 H, ArH), 7.13 (d, J = 8.4 Hz, 2 H, ArH), 6.86 (d, J = 8.8 Hz, 1 H, ArH), 6.52 (d, J = 8.4 Hz, 2 H, ArH), 5.44 (s, 1 H, CH), 2.68–2.55 (m, 2 H, CH_2), 2.28–2.21 (m, 2 H, CH_2), 1.98–1.92 (m, 1 H, CH), 1.87–1.83 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.9, 155.8, 153.5, 139.2, 138.3, 134.5, 134.4, 128.7, 120.1, 119.3, 114.9, 111.8, 108.8, 107.1, 37.3, 34.7, 27.5, 21.5.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{20}H_{17}N_3O_2Na$ [M + Na]$^+$: 354.1218; found: 354.1229.

8,8-Dimethyl-11-phenyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo-[3,4-*a*]acridin-10-one (4h)

Yellow solid; yield: 285.0 mg (83%); mp >280 °C.

IR (KBr): 3421, 3178, 1610, 1594, 1490, 1395, 1255, 1152, 1030, 930, 853, 797, 696, 612, 538 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.85 (s, 1 H, NH), 9.61 (s, 1 H, NH), 7.92 (d, *J* = 0.8 Hz, 1 H, ArH), 7.51 (d, *J* = 8.4 Hz, 1 H, ArH), 6.96 (d, *J* = 1.2 Hz, 1 H, ArH), 6.86 (d, *J* = 8.8 Hz, 1 H, ArH), 6.80 (dd, J_1 = 8.0 Hz, J_2 = 1.2 Hz, 1 H, ArH), 6.68 (d, *J* = 8.4 Hz, 1 H, ArH), 5.87 (d, *J* = 8.4 Hz, 2 H, ArH), 5.42 (s, 1 H, CH), 2.55 (d, *J* = 16.8 Hz, 1 H, CH), 2.43 (d, *J* = 16.4 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.04 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.95 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 152.0, 147.4, 139.1, 134.6, 134.5, 128.2, 127.8, 126.2, 120.2, 119.6, 111.8, 107.2, 106.5, 50.7, 35.8, 32.6, 29.7, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{21}N_3ONa$ [M + Na]$^+$: 366.1582; found: 366.1588.

11-(4-Fluorophenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4i)

Yellow solid; yield: 321.7 mg (89%); mp >280 °C.

IR (KBr): 3428, 3174, 1676, 1496, 1392, 1265, 1120, 1030, 930, 848, 600, 558, 550, 489 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.89 (s, 1 H, NH), 9.65 (s, 1 H, NH), 7.93 (s, 1 H, ArH), 7.52 (d, *J* = 8.4 Hz, 1 H, ArH), 7.38 (dd, J_1 = 8.8 Hz, J_2 = 5.6 Hz, 2 H, ArH), 6.97 (t, *J* = 8.8 Hz, 2 H, ArH), 6.88 (d, *J* = 8.4 Hz, 1 H, ArH), 5.53 (s, 1 H, CH), 2.56 (d, *J* = 16.8 Hz, 1 H, CH), 2.42 (d, *J* = 16.4 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.02 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.92 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 162.1, 159.7, 152.0, 143.5, 134.6, 129.5 (d, $J_{C,F}$ = 7.9 Hz), 120.0 (d, $J_{C,F}$ = 37.9 Hz), 119.8, 115.0, 114.8 (d, $J_{C,F}$ = 20.8 Hz), 111.8, 107.1, 106.2, 50.7, 35.1, 32.6, 29.7, 28.3, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}FN_3ONa$ [M + Na]$^+$: 384.1488; found: 384.1470.

11-(3-Chlorophenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4j)

Yellow solid; yield: 325.0 mg (86%); mp >280 °C.

IR (KBr): 3447, 3247, 1675, 1594, 1490, 1387, 1151, 1095, 1030, 931, 885, 703, 658 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.93 (s, 1 H, NH), 9.70 (s, 1 H, NH), 7.94 (d, *J* = 1.2 Hz, 1 H, ArH), 7.55 (d, *J* = 8.8 Hz, 1 H, ArH), 7.50 (s, 1 H, ArH), 7.25 (d, *J* = 7.6 Hz, 1 H, ArH), 7.18 (t, *J* = 7.6 Hz, 1 H, ArH), 7.10 (d, *J* = 8.0 Hz, 1 H, ArH), 6.89 (d, *J* = 8.4 Hz, 1 H, ArH), 5.52 (s, 1 H, CH), 2.57 (d, *J* = 16.4 Hz, 1 H, CH), 2.44 (d, *J* = 16.4 Hz, 1 H, CH), 2.24 (d, *J* = 16.0 Hz, 1 H, CH), 2.03 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.92 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.5, 152.2, 149.6, 139.1, 134.7, 134.6, 132.9, 130.3, 127.8, 126.5, 126.3, 120.3, 120.1, 111.9, 106.7, 105.6, 50.6, 35.8, 32.7, 29.7, 26.9.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}ClN_3ONa$ [M + Na]$^+$: 400.1193; found: 400.1207.

11-(4-Chlorophenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4k)

Yellow solid; yield: 328.7 mg (87%); mp >280 °C.

IR (KBr): 3443, 1631, 1489, 1397, 1156, 1080, 1028, 708, 577 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.89 (s, 1 H, NH), 9.68 (s, 1 H, NH), 7.93 (s, 1 H, ArH), 7.53 (d, *J* = 8.4 Hz, 1 H, ArH), 7.37 (d, *J* = 8.4 Hz, 2 H, ArH), 7.21 (d, *J* = 8.4 Hz, 2 H, ArH), 6.88 (d, *J* = 8.8 Hz, 1 H, ArH), 5.53 (s, 1 H, CH), 2.56 (s, *J* = 16.4 Hz, 1 H, CH), 2.42 (d, *J* = 16.4 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.02 (d, *J* = 16.0 Hz, 1 H, CH), 1.04 (s, 3 H, CH_3), 0.91 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 152.1, 146.2, 139.1, 134.7, 134.6, 130.8, 129.7, 128.2, 120.2, 112.0, 111.8, 106.9, 105.8, 50.7, 35.4, 32.6, 29.7, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}ClN_3ONa$ [M + Na]$^+$: 400.1193; found: 400.1175.

11-(2,4-Dichlorophenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4l)

Yellow solid; yield: 334.0 mg (81%); mp >280 °C.

IR (KBr): 3421, 3246, 1676, 1584, 1507, 1473, 1388, 1264, 1150, 1044, 1030, 669, 572, 448 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.06 (s, 1 H, NH), 9.83 (s, 1 H, NH), 7.94 (s, 1 H, ArH), 7.91 (d, *J* = 9.2 Hz, 1 H, ArH), 7.54 (d, *J* = 8.4 Hz, 1 H, ArH), 7.32 (dd, J_1= 6.4 Hz, J_2 = 2.4 Hz, 2 H, ArH), 6.82 (d, *J* = 8.4 Hz, 1 H, ArH), 5.77 (s, 1 H, CH), 2.54 (d, *J* = 17.6 Hz, 1 H, CH), 2.38 (d, *J* = 16.4 Hz, 1 H, CH), 2.20 (d, *J* = 16.4 Hz, 1 H, CH), 1.98 (d, *J* = 16.0 Hz, 1 H, CH), 1.04 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.3, 152.7, 141.3, 139.5, 135.6, 135.0, 133.5, 131.9, 129.7, 126.8, 120.5, 120.4, 111.8, 104.4, 103.4, 50.6, 37.1, 32.4, 31.2, 29.7, 27.1.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{19}Cl_2N_3ONa$ [M + Na]$^+$: 434.0803; found: 434.0825.

11-(3-Bromophenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4m)

Yellow solid; yield: 350.5 mg (83%); mp >280 °C.

IR (KBr): 3422, 3170, 1633, 1592, 1495, 1394, 1255, 1071, 1030, 931, 850, 734, 658 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.93 (s, 1 H, NH), 9.70 (s, 1 H, NH), 7.95 (s, 1 H, ArH), 7.66 (s, 1 H, ArH), 7.55 (d, *J* = 8.4 Hz, 1 H, ArH), 7.29 (d, *J* = 7.8 Hz, 1 H, ArH), 7.23 (d, *J* = 7.8 Hz, 1 H, ArH), 7.12 (t, *J* = 7.8 Hz, 1 H, ArH), 6.89 (d, *J* = 8.4 Hz, 1 H, ArH), 5.51 (s, 1 H, CH), 2.57 (d, *J* = 16.8 Hz, 1 H, CH), 2.44 (d, *J* = 16.4 Hz, 1 H, CH), 2.24 (d, *J* = 16.0 Hz, 1 H, CH), 2.03 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.5, 152.3, 149.8, 139.1, 134.6, 130.7, 130.6, 129.2, 126.8, 121.7, 120.3, 120.1, 111.9, 106.7, 105.7, 50.6, 35.8, 32.7, 29.7, 26.9.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}BrN_3ONa$ [M + Na]$^+$: 444.0687; found: 444.0695.

11-(4-Bromophenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4n)

Yellow solid; yield: 371.6 mg (88%); mp >280 °C.

IR (KBr): 3421, 3177, 1680, 1593, 1482, 1450, 1391, 1152, 1030, 929, 848, 664, 596 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.89 (s, 1 H, NH), 9.68 (s, 1 H, NH), 7.93 (d, *J* = 0.8 Hz, 1 H, ArH), 7.53 (d, *J* = 8.4 Hz, 1 H, ArH), 7.37–7.34 (m, 2 H, ArH), 7.33–7.30 (m, 2 H, ArH), 6.88 (d, *J* = 8.4 Hz, 1 H, ArH), 5.51 (s, 1 H, CH), 2.56 (d, *J* = 16.8 Hz, 1 H, CH), 2.42 (d, *J* = 16.8 Hz, 1 H, CH), 2.23 (d, *J* = 16.4 Hz, 1 H, CH), 2.02 (d, *J* = 15.6 Hz, 1 H, CH), 1.04 (s, 3 H, CH_3), 0.91 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 152.1, 146.6, 139.1, 134.6, 134.6, 131.1, 130.1, 120.2, 112.0, 119.4, 111.8, 106.8, 105.8, 50.7, 35.5, 32.6, 29.7, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}BrN_3ONa$ [M + Na]$^+$: 444.0687; found: 444.0689.

8,8-Dimethyl-11-(*p*-tolyl)-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo-[3,4-*a*]acridin-10-one (4o)

Yellow solid; yield: 307.4 mg (86%); mp >280 °C.

IR (KBr): 3417, 3174, 1675, 1597, 1494, 1450, 1393, 1149, 1074, 1031, 930, 888, 843, 739, 663, 602 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.84 (s, 1 H, NH), 9.59 (s, 1 H, NH), 7.91 (s, 1 H, ArH), 7.50 (d, *J* = 8.4 Hz, 1 H, ArH), 7.27 (d, *J* = 7.6 Hz, 2 H, ArH), 6.94 (d, *J* = 7.6 Hz, 2 H, ArH), 6.86 (d, *J* = 8.4 Hz, 1 H, ArH), 5.47 (s, 1 H, CH), 2.56 (d, *J* = 16.8 Hz, 1 H, CH), 2.42 (d, *J* = 16.4 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.14 (s, 3 H, CH_3), 2.01 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 151.8, 144.6, 139.1, 135.1, 134.5, 134.4, 128.8, 127.7, 120.2, 119.5, 111.8, 107.3, 106.7, 50.8, 35.4, 32.6, 29.8, 27.0, 21.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{23}N_3ONa$ [M + Na]$^+$: 380.1739; found: 380.1750.

11-(3,4-Dimethoxyphenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4p)

Yellow solid; yield: 308.3 mg (83%); mp >280 °C.

IR (KBr): 3172, 1647, 1593, 1493, 1449, 1389, 1264, 1118, 1072, 1031, 919, 839, 813, 745, 642, 607 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.81 (s, 1 H, NH), 9.56 (s, 1 H, NH), 7.91 (s, 1 H, ArH), 7.49 (d, *J* = 8.4 Hz, 1 H, ArH), 7.23 (s, 1 H, ArH), 7.06 (d, *J* = 7.6 Hz, 1 H, ArH), 6.86 (dd, J_1= 10.8 Hz, J_2 = 8.0 Hz, 2 H, ArH), 5.41 (s, 1 H, CH), 2.56 (d, *J* = 16.8 Hz, 1 H, CH), 2.43 (d, *J* = 16.8 Hz, 1 H, CH), 2.22 (d, *J* = 16.0 Hz, 1 H, CH), 2.09 (s, 3 H, CH_3), 2.05 (s, 3 H, CH_3), 2.01 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 151.8, 145.0, 139.1, 135.5, 134.4, 133.9, 129.4, 129.1, 125.3, 120.2, 119.4, 111.8, 107.3, 106.9, 50.8, 35.5, 32.6, 29.8, 27.0, 20.0, 19.3.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{24}H_{25}N_3ONa$ [M + Na]$^+$: 394.1895; found: 394.1873.

11-(2-Methoxyphenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4q)

Yellow solid; yield: 313.7 mg (84%); mp >280 °C.

IR (KBr): 3389, 3296, 1621, 1586, 1484, 1391, 1234, 1151, 1019, 887, 842, 750, 630, 567 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 11.79 (s, 1 H, NH), 9.61 (s, 1 H, NH), 7.91 (d, *J* = 0.8 Hz, 1 H, ArH), 7.46 (d, *J* = 8.8 Hz, 1 H, ArH), 7.40 (dd, J_1 = 7.8 Hz, J_2 =1.2 Hz, 1 H, ArH), 7.05 (t, *J* = 7.2 Hz, 1 H, ArH), 6.91 (d, *J* = 8.0 Hz, 1 H, ArH), 6.82 (dd, J_1 = 8.4 Hz, J_2 = 5.2 Hz, 2 H, ArH), 5.71 (s, 1 H, CH), 3.87 (s, 3 H, CH_3), 2.58 (d, *J* = 16.4 Hz, 1 H, CH), 2.45 (d, *J* = 16.4 Hz, 1 H, CH), 2.21 (d, *J* = 16.0 Hz, 1 H, CH), 1.98 (d, *J* = 16.0 Hz, 1 H, CH), 1.06 (s, 3 H, CH_3), 1.00 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.2, 156.1, 152.6, 139.4, 135.4, 134.8, 134.6, 130.2, 127.8, 121.2, 119.8, 119.4, 112.6, 111.9, 105.9, 105.9, 56.7, 50.8, 32.5, 31.5, 29.8, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{23}N_3O_2Na$ [M + Na]$^+$: 396.1688; found: 396.1681.

11-(3-Methoxyphenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4r)

Yellow solid; yield: 302.5 mg (81%); mp >280 °C.

IR (KBr): 3443, 3170, 1595, 1483, 1451, 1394, 1264, 1150, 1032, 931, 853, 805, 748, 614 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.89 (s, 1 H, NH), 9.62 (s, 1 H, NH), 7.92 (d, *J* = 1.2 Hz, 1 H, ArH), 7.51 (d, *J* = 8.4 Hz, 1 H, ArH), 7.05 (t, *J* = 8.0 Hz, 2 H, ArH), 6.90 (d, *J* = 7.8 Hz, 1 H, ArH), 6.86 (d, *J* = 8.4 Hz, 1 H, ArH), 6.60 (dd, J_1 = 7.8 Hz, J_2 = 2.4 Hz, 1 H, ArH), 5.48 (s, 1 H, CH), 3.66 (s, 3 H, CH_3), 2.57 (d, *J* = 16.8 Hz, 1 H, CH), 2.44 (d, *J* = 16.4 Hz, 1 H, CH), 2.24 (d, *J* = 16.0 Hz, 1 H, CH), 2.03 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.95 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.5, 159.3, 152.1, 148.9, 139.1, 134.6, 134.5, 129.3, 120.2, 120.1, 119.6, 114.2, 111.8, 111.1, 107.0, 106.4, 55.3, 50.7, 35.9, 32.6, 29.8, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{23}N_3O_2Na$ [M + Na]$^+$: 396.1688; found: 396.1697.

11-(4-Hydroxy-3-methoxyphenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4s)

Yellow solid; yield: 331.0 mg (85%); mp >280 °C.

IR (KBr): 3176, 1608, 1593, 1490, 1392, 1255, 1228, 1150, 1032, 939, 801, 749, 592 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.87 (s, 1 H, NH), 9.55 (s, 1 H, NH), 8.59 (s, 1 H, OH), 7.91 (d, *J* = 1.2 Hz, 1 H, ArH), 7.48 (d, *J* = 8.4 Hz, 1 H, ArH), 7.12 (d, *J* = 2.0 Hz, 1 H, ArH), 6.84 (d, *J* = 8.4 Hz, 1 H, ArH), 6.64 (dd, J_1 = 8.0 Hz, J_2 = 1.6 Hz, 1 H, ArH), 6.51 (d, *J* = 8.0 Hz, 1 H, ArH), 5.38 (s, 1 H, CH), 3.70 (s, 3 H, CH_3), 2.57 (d, *J* = 16.4 Hz, 1 H, CH), 2.42 (d, *J* = 16.8 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.02 (d, *J* = 16.0 Hz, 1 H, CH), 1.06 (s, 3 H, CH_3), 0.97 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.5, 151.7, 147.2, 145.0, 139.1, 138.9, 134.4, 120.1, 119.9, 119.3, 115.4, 113.7, 112.8, 111.8, 107.5, 107.1, 56.1, 50.8, 35.3, 32.6, 31.2, 29.9, 26.9.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{23}N_3O_3Na$ [M + Na]$^+$: 412.1637; found: 412.1649.

11-(4-Methoxyphenyl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4t)

Yellow solid; yield: 324.9 mg (87%); mp >280 °C.

IR (KBr): 3171, 1668, 1610, 1592, 1505, 1392, 1303, 1264, 1149, 1031, 931, 839, 742, 654, 594 cm^{-1}.

^1H NMR (400 MHz, DMSO-d_6): δ = 12.85 (s, 1 H, NH), 9.59 (s, 1 H, NH), 7.91 (d, *J* = 1.2 Hz, 1 H, ArH), 7.50 (d, *J* = 8.8 Hz, 1 H, ArH), 7.29 (d, *J* = 8.4 Hz, 2 H, ArH), 6.86 (d, *J* = 8.4 Hz, 1 H, ArH), 6.70 (d, *J* = 8.4 Hz, 2 H, ArH), 5.46 (s, 1 H, CH), 3.62 (s, 3 H, CH_3), 2.56 (d, *J* = 16.4 Hz, 1 H, CH), 2.42 (d, *J* = 16.8 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.02 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.94 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 157.8, 151.7, 139.8, 139.1, 134.5, 134.4, 128.8, 120.2, 119.5, 113.6, 111.8, 107.4, 106.9, 55.3, 50.8, 34.9, 32.6, 29.7, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{23}N_3O_2Na$ [M + Na]$^+$: 396.1688; found: 396.1690.

11-(Benzo[*d*][1,3]dioxol-5-yl)-8,8-dimethyl-1,6,7,8,9,11-hexahydro-10*H*-pyrazolo[3,4-*a*]acridin-10-one (4u)

Yellow solid; yield: 333.2 mg (86%); mp >280 °C.

IR (KBr): 3418, 1632, 1482, 1395, 1264, 1228, 1154, 1032, 934, 629 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.87 (s, 1 H, NH), 9.62 (s, 1 H, NH), 7.92 (s, 1 H, ArH), 7.51 (d, *J* = 8.8 Hz, 1 H, ArH), 7.38 (d, *J* = 7.8 Hz, 2 H, ArH), 7.14 (t, *J* = 7.8 Hz, 2 H, ArH), 7.03 (d, *J* = 7.8 Hz, 1 H, ArH), 6.87 (d, *J* = 8.4 Hz, 1 H, ArH), 5.51 (s, 1 H, CH), 2.57 (d, *J* = 16.4 Hz, 1 H, CH), 2.43 (d, *J* = 16.8 Hz, 1 H, CH), 2.23 (d, *J* = 16.0 Hz, 1 H, CH), 2.02 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.5, 151.8, 147.2, 145.6, 141.6, 139.1, 134.5, 120.7, 120.2, 119.6, 111.8, 108.6, 108.1, 107.3, 106.6, 101.0, 50.7, 35.5, 32.6, 31.2, 29.7, 27.1.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{21}N_3O_3Na$ [M + Na]$^+$: 410.1481; found: 410.1496.

11-(4-Fluorophenyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]-acridin-10-one (6a)

Yellow solid: yield: 283.4 mg (85%); mp 267–269 °C.

IR (KBr): 3259, 1584, 1492, 1228, 1192, 1088, 1041, 940, 838, 798, 705, 565, 487 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.96 (s, 1 H, NH), 9.58 (s, 1 H, NH), 8.04 (s, 1 H, ArH), 7.36–7.30 (m, 3 H, ArH), 7.11 (d, *J* = 8.8 Hz, 1 H, ArH), 6.96 (t, *J* = 8.8 Hz, 2 H, ArH), 5.53 (s, 1 H, CH), 2.67–2.60 (m, 2 H, CH_2), 2.27–2.18 (m, 2 H, CH_2), 1.97–1.92 (m, 1 H, CH), 1.88–1.80 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.7, 162.0, 159.6, 153.6, 144.65 (d, $J_{C,F}$ = 2.9 Hz), 129.7 (d, $J_{C,F}$ = 7.9 Hz), 129.4, 122.1, 117.2, 115.3, 115.0, 114.8, 109.8, 107.2, 37.7, 37.2, 27.6, 21.5.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{20}H_{16}FN_3ONa$ [M + Na]$^+$: 356.1175; found: 356.1184.

11-(4-Chlorophenyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]-acridin-10-one (6b)

Yellow solid: yield: 290.3 mg (83%); mp 218–220 °C.

IR (KBr): 3433, 3264, 1660, 1603, 1539, 1487, 1383, 1285, 1124, 1084, 1014, 932, 803, 736, 664 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.99 (s, 1 H, NH), 9.65 (s, 1 H, NH), 7.96 (s, 1 H, ArH), 7.36 (d, *J* = 8.8 Hz, 1 H, ArH), 7.31 (d, *J* = 8.4 Hz, 2 H, ArH), 7.20 (d, *J* = 8.4 Hz, 2 H, ArH), 7.12 (d, *J* = 8.8 Hz, 1 H, ArH), 5.52 (s, 1 H, CH), 2.65–2.59 (m, 2 H, CH_2), 2.27–2.18 (m, 2 H, CH_2), 1.97–1.92 (m, 1 H, CH), 1.87–1.79 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.7, 153.7, 147.4, 130.6, 129.8, 129.5, 129.5, 128.2, 122.1, 117.2, 114.9, 106.9, 37.9, 37.2, 27.6, 21.5.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{20}H_{16}ClN_3ONa$ [M + Na]$^+$: 372.0880; found: 372.0895.

11-(3,4-Dichlorophenyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6c)

Yellow solid: yield: 330.5 mg (86%); mp 234–236 °C.

IR (KBr): 3133, 1636, 1541, 1507, 1458, 1229, 1191, 1076, 936, 757, 567 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.02 (s, 1 H, NH), 9.66 (s, 1 H, NH), 8.08 (s, 1 H, ArH), 7.56 (s, 1 H, ArH), 7.40 (s, 2 H, ArH), 7.23 (s, 1 H, ArH), 7.13 (s, 1 H, ArH), 5.55 (s, 1 H, CH), 2.63 (s, 2 H, CH_2), 2.23 (s, 2 H, CH_2), 1.94 (s, 1 H, CH), 1.84 (s, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.7, 153.9, 149.3, 132.2, 131.7, 130.7, 130.6, 129.9, 129.8, 129.5, 128.6, 128.5, 128.4, 122.1, 117.3, 114.2, 106.5, 37.9, 37.1, 27.6, 21.5.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{20}H_{15}Cl_2N_3ONa$ [M + Na]$^+$: 406.0490; found: 406.0483.

11-(4-Bromophenyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]-acridin-10-one (6d)

Yellow solid: yield: 347.0 mg (88%); mp 212–214 °C.

IR (KBr): 3447, 3261, 1659, 1540, 1507, 1476, 1396, 1285, 1193, 1094, 931, 839, 803, 663, 531 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.98 (s, 1 H, NH), 9.63 (s, 1 H, NH), 7.96 (s, 1 H, ArH), 7.37–7.32 (m, 3 H, ArH), 7.25 (d, *J* = 8.4 Hz, 2 H, ArH), 7.11 (d, *J* = 8.8 Hz, 1 H, ArH), 5.50 (s, 1 H, CH), 2.63–2.60 (m, 2 H, CH_2), 2.27–2.18 (m, 2 H, CH_2), 1.97–1.91 (m, 1 H, CH), 1.86–1.79 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.7, 153.7, 147.8, 132.4, 131.2, 130.3, 130.1, 129.4, 122.1, 119.1, 117.2, 114.8, 106.8, 38.0, 37.2, 27.6, 21.5.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{20}H_{16}BrN_3ONa$ [M + Na]$^+$: 416.0374; found: 416.0382.

11-(*p*-Tolyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6e)

Yellow solid: yield: 266.8 mg (81%); mp 230–232 °C.

IR (KBr): 3446, 1697, 1646, 1482, 1429, 1386, 1159, 1029, 758, 531, 488 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.93 (s, 1 H, NH), 9.52 (s, 1 H, NH), 8.00 (s, 1 H, ArH), 7.32 (d, *J* = 8.4 Hz, 1 H, ArH), 7.17 (d, *J* = 7.6 Hz, 2 H, ArH), 7.09 (d, *J* = 8.8 Hz, 1 H, ArH), 6.94 (d, *J* = 7.2 Hz, 2 H, ArH), 5.46 (s, 1 H, CH), 2.62 (s, 2 H, CH_2), 2.24–2.20 (m, 2 H, CH_2), 2.14 (s, 3 H, CH_3), 1.92 (s, 1 H, CH), 1.82 (s, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.6, 153.4, 145.6, 137.9, 134.9, 132.0, 130.0, 129.4, 128.9, 127.9, 127.5, 122.2, 117.2, 115.8, 109.5, 107.4, 38.0, 37.2, 27.6, 21.6, 21.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{21}H_{19}N_3ONa$ [M + Na]$^+$: 352.1426; found: 352.1447.

11-(3,4-Dimethylphenyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo-[4,3-*a*]acridin-10-one (6f)

Yellow solid; yield: 274.7 mg (80%); mp >280 °C.

IR (KBr): 3255, 1587, 1492, 1395, 1229, 1191, 1128, 1013, 936, 801, 696, 547 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.92 (s, 1 H, NH), 9.48 (s, 1 H, NH), 8.00 (s, 1 H, ArH), 7.31 (d, *J* = 8.4 Hz, 1 H, ArH), 7.08 (s, 2 H, ArH), 6.92 (dd, J_1 = 36.0 Hz, J_2 = 6.8 Hz, 2 H, ArH), 5.41 (s, 1 H, CH), 2.62 (s, 2 H, CH_2), 2.19 (s, 2 H, CH_2), 2.09 (s, 3 H, CH_3), 2.05 (s, 3 H, CH_3), 1.93 (s, 1 H, CH), 1.82 (s, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.6, 153.4, 146.0, 135.6, 133.7, 129.5, 129.4, 129.4, 125.5, 122.3, 117.2, 115.9, 107.4, 38.0, 37.2, 27.6, 21.6, 20.0, 19.3.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{21}N_3ONa$ [M + Na]$^+$: 366.1582; found: 366.1593.

11-(Benzo[*d*][1,3]dioxol-5-yl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6g)

Yellow solid; yield: 319.8 mg (89%); mp 264–266 °C.

IR (KBr): 3269, 1629, 1565, 1421, 1376, 1186, 1033, 919, 810, 764, 673, 579, 522, 487 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.95 (s, 1 H, NH), 9.55 (s, 1 H, NH), 8.05 (s, 1 H, ArH), 7.34 (d, *J* = 8.8 Hz, 1 H, ArH), 7.10 (d, *J* = 8.8 Hz, 1 H, ArH), 6.83 (d, *J* = 1.6 Hz, 1 H, ArH), 6.75 (dd, J_1 = 8.0 Hz, J_2 = 1.6 Hz, 1 H, ArH), 6.67 (d, *J* = 8.0 Hz, 1 H, ArH), 5.86 (dd, J_1 = 8.0 Hz, J_2 = 0.8 Hz, 2 H, CH_2), 5.44 (s, 1 H, CH), 2.64–2.59 (m, 2 H, CH_2), 2.26–2.20 (m, 2 H, CH_2), 1.97–1.92 (m, 1 H, CH), 1.86–1.81 (m, 1 H, CH).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.8, 153.4, 147.3, 145.5, 142.7, 138.0, 132.1, 129.4, 122.2, 120.8, 117.2, 115.7, 109.7, 108.6, 108.0, 107.3, 101.0, 38.0, 37.2, 27.6, 21.6.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{21}H_{17}N_3O_3Na$ [M + Na]$^+$: 382.1168; found: 382.1181.

11-(4-Fluorophenyl)-8,8-dimethyl-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6h)

Yellow solid; yield: 334.4 mg (87%); mp 217–219 °C.

IR (KBr): 3339, 3157, 1701, 1602, 1504, 1486, 1413, 1386, 1232, 1091, 933, 846, 796, 591 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.98 (s, 1 H, NH), 9.58 (s, 1 H, NH), 8.03 (s, 1 H, ArH), 7.36–7.31 (m, 3 H, ArH), 7.12 (d, *J* = 8.8 Hz, 1 H, ArH), 6.97 (t, *J* = 8.8 Hz, 2 H, ArH), 5.49 (s, 1 H, CH), 2.55 (d, *J* = 16.4 Hz, 1 H, CH), 2.42 (d, *J* = 16.8 Hz, 1 H, CH), 2.20 (d, *J* = 16.0 Hz, 1 H, CH), 2.00 (d, *J* = 16.0 Hz, 1 H, CH), 1.04 (s, 3 H, CH_3), 0.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 198.0, 193.4, 161.1, 159.2, 151.9, 147.7, 146.9, 136.9, 129.8 (d, $J_{C,F}$ = 8.0 Hz), 129.6 (d, $J_{C,F}$ = 8.0 Hz), 129.4, 128.2, 123.2, 122.1, 117.2 (d, $J_{C,F}$ = 10.0 Hz), 116.5, 116.3, 115.0, 114.8, 105.8, 53.8, 47.3, 32.6, 32.5, 28.3, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}FN_3ONa$ [M + Na]$^+$: 384.1488; found: 384.1497.

11-(4-Chlorophenyl)-8,8-dimethyl-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6i)

Yellow solid; yield: 321.2 mg (85%); mp 271–273 °C.

IR (KBr): 3394, 1618, 1590, 1488, 1385, 1253, 1155, 1029, 845, 767, 607, 578, 497 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.99 (s, 1 H, NH), 9.58 (s, 1 H, NH), 8.02 (s, 1 H, ArH), 7.36 (d, *J* = 8.8 Hz, 1 H, ArH), 7.33 (d, *J* = 8.4 Hz, 2 H, ArH), 7.21 (d, *J* = 8.8 Hz, 2 H, ArH), 7.12 (d, *J* = 8.8 Hz, 1 H, ArH), 5.49 (s, 1 H, CH), 2.55 (d, *J* = 16.8 Hz, 1 H, CH), 2.42 (d, *J* = 16.8 Hz, 1 H, CH), 2.21 (d, *J* = 16.0 Hz, 1 H, CH), 2.00 (d, *J* = 16.0 Hz, 1 H, CH), 1.04 (s, 3 H, CH_3), 0.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.2, 151.9, 147.2, 137.9, 131.9, 130.6, 129.8, 129.6, 129.4, 128.2, 122.1, 117.3, 115.0, 110.0, 105.6, 50.7, 38.1, 32.6, 29.7, 28.3, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}ClN_3ONa$ [M + Na]$^+$: 400.1193; found: 400.1209.

11-(3,4-Dichlorophenyl)-8,8-dimethyl-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6j)

Yellow solid; yield: 338.1 mg (82%); mp 201–203 °C.

IR (KBr): 3415, 1590, 1486, 1384, 1264, 1232, 1158, 1080, 1030, 932, 803, 604 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.01 (s, 1 H, NH), 9.59 (s, 1 H, NH), 8.09 (s, 1 H, ArH), 7.58 (d, *J* = 2.0 Hz, 1 H, ArH), 7.42 (d, *J* = 8.4 Hz, 1 H, ArH), 7.38 (d, *J* = 8.8 Hz, 1 H, ArH), 7.25 (dd, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1 H, ArH), 7.12 (d, *J* = 8.8 Hz, 1 H, ArH), 5.53 (s, 1 H, CH), 2.55 (d, *J* = 16.8 Hz, 1 H, CH), 2.43 (d, *J* = 16.4 Hz, 1 H, CH), 2.22 (d, *J* = 16.0 Hz, 1 H, CH), 2.01 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 152.2, 149.2, 132.2, 131.8, 130.9, 130.6, 130.0, 129.8, 129.4, 128.6, 128.5, 128.3, 122.1, 117.3, 114.3, 105.2, 50.6, 38.0, 32.6, 29.6, 28.3, 26.9.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{19}Cl_2N_3ONa$ [M + Na]$^+$: 434.0803; found: 434.0827.

11-(4-Bromophenyl)-8,8-dimethyl-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6k)

Yellow solid; yield: 380.1 mg (90%); mp 209–211 °C.

IR (KBr): 3203, 1711, 1589, 1485, 1384, 1232, 1122, 1010, 931, 805, 766, 597 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.99 (s, 1 H, NH), 9.55 (s, 1 H, NH), 8.02 (s, 1 H, ArH), 7.38–7.32 (m, 3 H, ArH), 7.27 (d, *J* = 8.4 Hz, 2 H, ArH), 7.10 (d, *J* = 8.8 Hz, 1 H, ArH), 5.47 (s, 1 H, CH), 2.56 (d, *J* = 9.2 Hz, 1 H, CH), 2.41 (d, *J* = 16.4 Hz, 1 H, CH), 2.21 (d, *J* = 16.0 Hz, 1 H, CH), 1.99 (d, *J* = 16.0 Hz, 1 H, CH), 1.04 (s, 3 H, CH_3), 0.93 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.2, 151.8, 147.7, 132.4, 131.1, 130.2, 129.4, 122.1, 119.1, 117.3, 114.9, 110.0, 105.5, 50.7, 38.2, 32.6, 31.2, 28.3, 27.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{22}H_{20}BrN_3ONa$ [M + Na]$^+$: 444.0687; found: 444.0682.

8,8-Dimethyl-11-(*p*-tolyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6l)

Yellow solid; yield: 300.3 mg (84%); mp 221–223 °C.

IR (KBr): 3412, 1717, 1647, 1559, 1497, 1396, 1267, 1158, 1099, 1030, 754, 577 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.93 (s, 1 H, NH), 9.45 (s, 1 H, NH), 7.96 (s, 1 H, NH), 7.32 (d, *J* = 8.8 Hz, 1 H, ArH), 7.19 (d, *J* = 8.0 Hz, 2 H, ArH), 7.08 (d, *J* = 8.8 Hz, 1 H, ArH), 6.94 (d, *J* = 7.6 Hz, 2 H, ArH), 5.42 (s, 1 H, CH), 2.55 (d, *J* = 16.8 Hz, 1 H, CH), 2.41 (d, *J* = 16.4 Hz, 1 H, CH), 2.20 (d, *J* = 16.0 Hz, 1 H, CH), 2.14 (s, 3 H, CH_3), 1.98 (d, *J* = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.95 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.2, 151.6, 145.5, 134.9, 129.3, 128.8, 127.9, 122.2, 117.2, 115.9, 106.0, 50.8, 38.2, 32.6, 29.8, 27.0, 21.0.

HRMS (ESI-TOF): *m*/*z* calcd for $C_{23}H_{23}N_3ONa$ [M + Na]$^+$: 380.1739; found: 380.1748.

11-(3,4-Dimethylphenyl)-8,8-dimethyl-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6m)

Yellow solid; yield: 297.2 mg (80%); mp >280 °C.

IR (KBr): 3427, 3270, 1587, 1541, 1487, 1420, 1383, 1148, 1031, 883, 799, 586 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.91 (s, 1 H, NH), 9.42 (s, 1 H, NH), 8.04 (s, 1 H, ArH), 7.31 (d, *J* = 8.8 Hz, 1 H, ArH), 7.11 (s, 1 H, ArH), 7.07 (d, *J* = 8.8 Hz, 1 H, ArH), 6.99 (dd, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1 H, ArH), 6.88 (d, *J* = 7.6 Hz, 1 H, ArH), 5.37 (s, 1 H, CH), 2.55 (d, *J* = 16.4 Hz, 1 H, CH),

2.41 (d, J = 16.0 Hz, 1 H, CH), 2.20 (d, J = 16.0 Hz, 1 H, CH), 2.10 (s, 3 H, CH_3), 2.05 (s, 3 H, CH_3), 1.98 (d, J = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.2, 151.6, 146.0, 135.6, 133.7, 129.4, 129.3, 129.2, 125.4, 122.2, 117.2, 116.0, 106.0, 50.8, 40.9, 38.2, 32.6, 29.8, 27.0, 20.0, 19.3.

HRMS (ESI-TOF): m/z calcd for $C_{24}H_{25}N_3ONa$ [M + Na]$^+$: 394.1895; found: 394.1908.

8,8-Dimethyl-11-(3,4,5-trimethoxyphenyl)-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6n)

Yellow solid; yield: 359.8 mg (83%); mp 278–280 °C.

IR (KBr): 3422, 1592, 1487, 1395, 1233, 1122, 1028, 1004, 986, 838, 698 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.94 (s, 1 H, NH), 9.47 (s, 1 H, NH), 8.26 (s, 1 H, ArH), 7.33 (d, J = 8.8 Hz, 1 H, ArH), 7.09 (d, J = 8.8 Hz, 1 H, ArH), 6.65 (s, 2 H, ArH), 5.42 (s, 1 H, CH), 3.67 (s, 6 H, CH_3), 3.54 (s, 3 H, CH_3), 2.59 (d, J = 16.4 Hz, 1 H, CH), 2.44 (d, J = 16.4 Hz, 1 H, CH), 2.24 (d, J = 16.0 Hz, 1 H, CH), 2.03 (d, J = 16.0 Hz, 1 H, CH), 1.07 (s, 3 H, CH_3), 1.03 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.4, 154.2, 152.9, 152.0, 144.2, 136.0, 129.2, 122.2, 117.3, 115.8, 105.7, 105.4, 60.3, 56.2, 50.8, 41.5, 38.7, 32.6, 30.0, 28.3, 26.8.

HRMS (ESI-TOF): m/z calcd for $C_{25}H_{27}N_3O_4Na$ [M + Na]$^+$: 456.1899; found: 456.1911.

11-(Benzo[*d*][1,3]dioxol-5-yl)-8,8-dimethyl-3,6,7,8,9,11-hexahydro-10*H*-pyrazolo[4,3-*a*]acridin-10-one (6o)

Yellow solid; yield: 329.3 mg (85%); mp >280 °C.

IR (KBr): 3448, 3359, 1654, 1578, 1487, 1438, 1243, 1158, 1122, 1034, 920, 858, 793, 585, 454 cm^{-1}.

^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.94 (s, 1 H, NH), 9.47 (s, 1 H, NH), 8.06 (s, 1 H, ArH), 7.33 (d, J = 8.8 Hz, 1 H, ArH), 7.09 (d, J = 8.8 Hz, 1 H, ArH), 6.84 (s, 1 H, ArH), 6.78–6.74 (m, 1 H, ArH), 6.67 (d, J = 8.0 Hz, 1 H, ArH), 5.87 (d, J = 11.2 Hz, 2 H, CH_2), 5.40 (s, 1 H, CH), 2.53 (d, J = 16.4 Hz, 1 H, CH), 2.42 (d, J = 16.8 Hz, 1 H, CH), 2.20 (d, J = 16.0 Hz, 1 H, CH), 2.01 (d, J = 16.0 Hz, 1 H, CH), 1.05 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3).

^{13}C NMR (100 MHz, DMSO-d_6): δ = 193.3, 151.6, 147.3, 145.4, 142.7, 129.3, 122.2, 120.8, 117.2, 115.7, 108.6, 108.0, 106.0, 101.0, 50.8, 38.1, 32.6, 29.7, 27.1.

HRMS (ESI-TOF): m/z calcd for $C_{23}H_{21}N_3O_3Na$ [M + Na]$^+$: 410.1481; found: 410.1496.

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Supporting Information

Supporting information for this article is available online at https://doi.org/10.1055/s-0036-1589124.

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