Microwave-Assisted Multicomponent Reactions: Rapid and Regioselective Formation of New Extended Angular Fused Aza-Heterocycles

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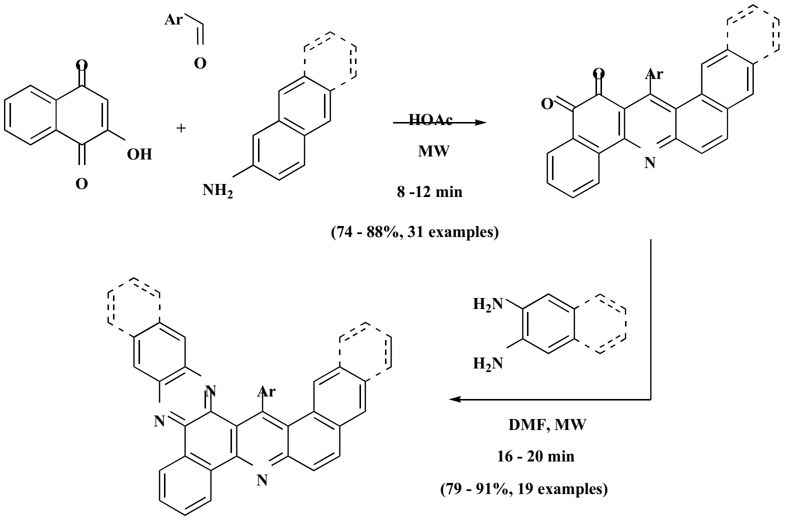
Abstract: A new multicomponent domino reaction for rapid and regioselective synthesis of highly functionalized benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-diones has been established. The reaction can be conducted by using readily available and inexpensive substrates under microwave irradiation. The procedures are facile, avoiding time-consuming and costly syntheses, tedious work-up and purifications of precursors as well as protection/deprotection of functional groups. This method is much more efficient due to short reaction times and easy work up. The resulting naphthoacridines have been readily converted into benzoquinoxaline-fused benzoquinoline analogues by treating with benzene-1,2-diamine under microwave irradiation. The structural assignment has been ambiguously confirmed by X-ray analysis. A new mechanism has been proposed for this new multicomponent domino process.



Keywords: Benzoquinoxaline-fused benzoquinolines, microwave-assisted synthesis, multicomponent reaction, naphthoacridines, regioselectivity.

INTRODUCTION

The charge-transport materials can be used in organic electronic devices such as organic light-emitting diodes (OLED), lasers, photovoltaic cells, photodetectors, active and passive electronic devices, and memories [1]. The extended angular fused aza-heterocycles (V-type fused aza-heterocycles) exhibit important photophysics properties, which are widely applied in the charge-transport materials due to their strong skeleton rigidity and large π-conjugation systems. With large π-conjugation systems, benzacridine derivatives, especially benzophenazine-fused benzacridine, possess interesting photophysical properties such as the presence of intramolecular electron-transfer state of a high energy and long lifetime, which have been tested and applied as an efficient photocatalyst in modeling the photosynthetic reactions [2]. Furthermore, the linear fuzed acridones, i.e. the oxidized derivatives of acridines, are well-known organic pigments and have been extensively studied for their self-assembling [3] photoconductive, doping dye emitter [4] and fluorescent [5] properties in numerous devices stemmed from materials chemistry.

Dihydroacridines with an 1,4-DHPs parent nucleus are well known as therapeutic agents [6]. Due to their interesting biological activities such as antimalarial and antitumor, they have immense utility in pharmaceutical industry [7]. Therefore, this class of compounds has been the focus of much recent research [8], and has led to intensive interest in the synthesis of several drugs based on them [9]. However, their derivatives having further extended conjugated systems such as the benzophenazine-fused benzacridine derivatives have been seldom reported so far. Therefore, a simple, efficient and direct synthetic route for this important class of heterocycles is strongly desired.

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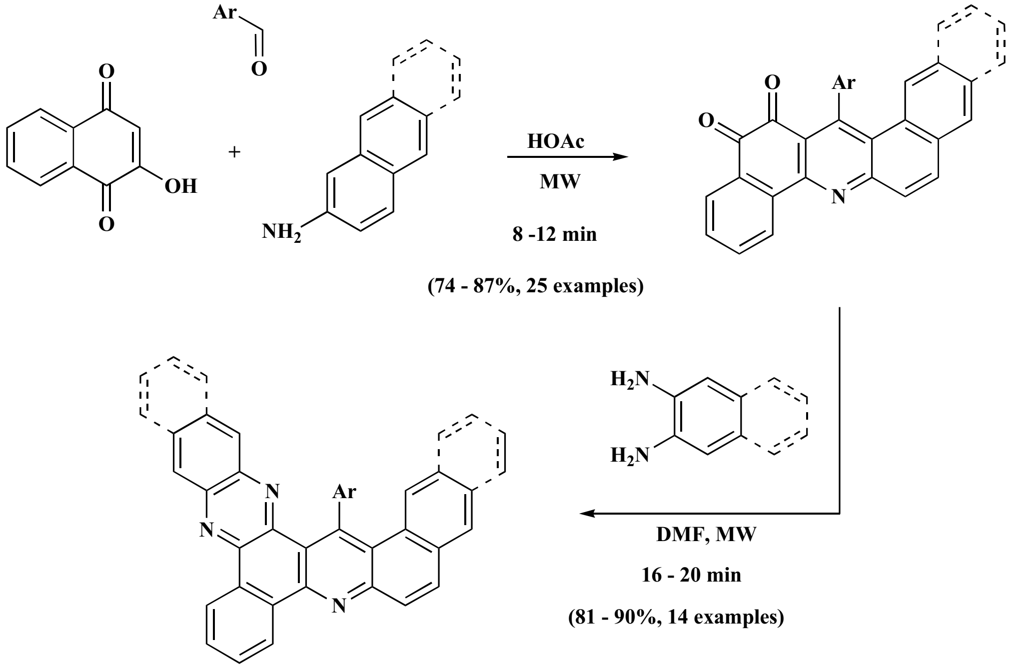


Scheme 1. The domino synthesis of benzophenazine-fused benzacridines.

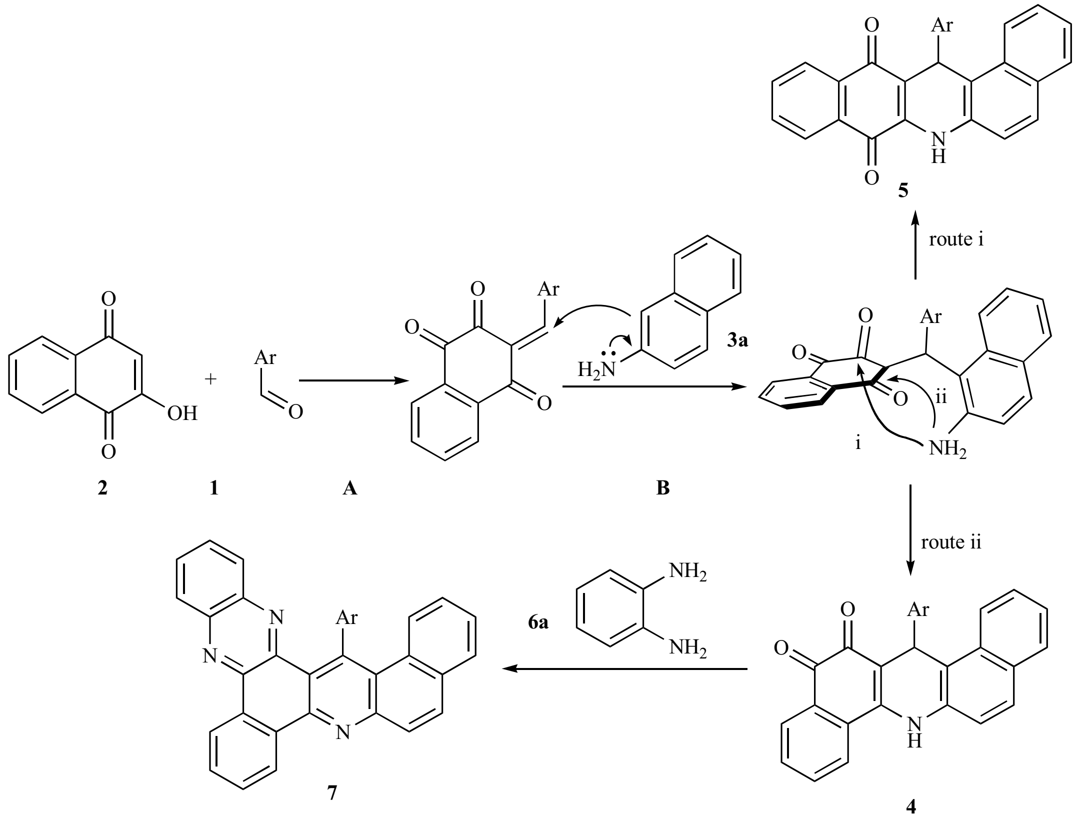
In the past several years, our group has developed various multicomponent domino reactions that can provide easy access to functionalized multiple heterocycles [10-12]. Among these reactions is a four-component domino process for concise synthesis of multi-functionalized quinazoline derivatives [10a]. Furthermore, we have also found that when aliphatic aldehydes were employed to replace their aromatic counterparts, the reaction proceeded along another pathway leading to the formation of multi-functionalized tricyclo[6.2.2.0^{1,6}]dodecanes [10b].

During our continuous efforts on searching for useful multicomponent domino reactions [10-12], we now report a new regioselective multicomponent domino annulation which provides an easy access to benzo[*h*]naphtho[2,3-*a*]acridines and their derivatives. This reaction was achieved by reacting aldehydes, aromatic amines and 2-hydroxy-1,4-naphthoquinone under microwave irradiation (MW) in the absence of strong acids or metal catalysts/promoters (Scheme **1**). The resulting benzo[*h*]naphtho[2,3-*a*]acridines has been reacted with benzene-1,2-diamine to give polycyclic heteroaromatics, benzophenazine-fused benzacridine. To the best of our knowledge, an efficient domino approach to extended angular fused aza-heterocycle, such as heptacyclic, cctocyclic or nonacyclic-fused benzo[*h*]naphtho [2,3-*a*]acridines, has not been well documented. In this paper, we would like to report the new reaction for their syntheses by controlling reaction substrates and sequence.

RESULTS AND DISCUSSION

It has been reported that when a mixture of aldehyde, 2-hydroxy-1,4-naphthoquinone and naphthalen-2-amine was stirred in [bmim]BF$_4$ at room temperature, a cycloadduct dibenzo[a,i]acridine-1,6-diones **5** were produced in high yields [13]. After analyzing reaction mechanism, we reasoned that this reaction may have two different routes to the final products **4** or **5**: route *i* to the prodcut **5**, and route *ii* to **4**. If the product **5** with *para*-diketone unit was provided, the reaction between product **5** and benzene-1,2-diamine did not further take place. On the contrary, the product **4** with *ortho*-dicarbonyl groups if provided can react with benzene-1,2-diamine to generate fused aza-heterocycles with big π-conjugate system. Based on the above analyse, we employed 4-chlorobenzaldehyde **1a** to react with 2-hydroxy-1,4-naphthoquinone **2** and naphthalen-2-amine **3a** in HOAc under microwave heating. After filtration, a red solid was obtained in 70% chemical yield. When the synthsized red solid was subjected with benzene-1,2-diamine, the yellow precipitates were observed. All of the analytical data showed that a benzene-1,2-diamine unit was introduced in the final product, and a novel heptacyclic benzophenazine fused benzoquinoline **7a** was produced in high yield (Scheme **1**). This product has been fully characterized by ^{1}H NMR, HRMS and IR spectral analysis. Furthermore, the structure of **7g** has been unambiguously determined by X-ray structural analysis as shown in Fig. (**1**). Thus, instead of compound **5**, the structure of red solid described above was dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione **4** established on this experimental results. This surprising result is of value to us not only because we are interested in the design of the new regioseclective domino reaction but also because we were unable to find examples of other methods allowing for such convenient synthesis in related literature.

Next, we choose the most appropriate solvent for the reaction of 4-chlorobenzaldehyde **1a** with 2-hydroxy-1,4-naphthoquinone **2** and naphthalen-2-amine **3a**. This heterocyclization reaction was examined at 100°C using water, EtOH, glycol, *N*,*N*-dimethylformamide (DMF) and acetic acid (AcOH) (Table **1**) as the solvent. As shown in Table **1**, the use of acetic acid at 120°C allowed the direct conversion of 2-hydroxy-1,4-naphthoquinone **2** into the corresponding dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione **4a** in a yield of 83% under microwave irradiation condition



Scheme 2. Synthesis of products **4** or **5**.

(Table **1**, entry 5). Other polar solvents, such as ethanol, ethylene glycol, *N*,*N*-dimethylformamide, and water, resulted in moderate to poor to modest yields of 40%-65% (Table **1**, entries 1-4). The reaction proceeded rapidly to completion within a few minutes at 120°C. Increasing reaction temperature to 140°C did not improve chemical yields.

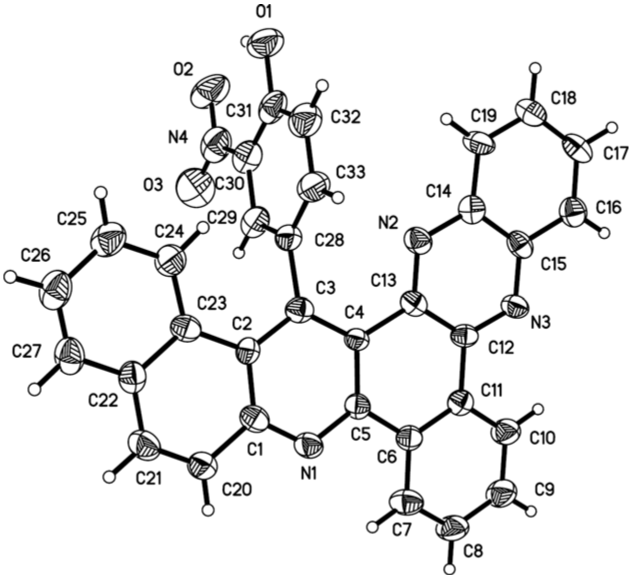


Fig. (1). ORTEP drawing of **7g**.

Table 1. Conditions Optimization for the Synthesis of 4a

Entry	Solvent	T/°C	Time (min)	Yield/%[a]
1	Water	120	8	40
2	EtOH	120	8	60
3	Glycol	120	8	56
4	DMF	120	8	65
5	AcOH	120	8	83

[a]Isolated yield.

Encouraged by the above interesting results, we utilized various aromatic aldehydes with different substituents under the same reaction conditions. The results are shown in Table **2**. From these results, we could see that all of the reactions proceeded smoothly to afford the corresponding dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione **4** in good yields (75-83%). Aromatic aldehydes carrying either electron-donating or electron-withdrawing substituents showed similar reactivity and reacted efficiently to yield the desired product. Moreover, heterocyclic aldehydes such as thiophene-2-carbaldehyde (Table **2**, entry 14) also displayed a high reactivity under this standard condition. Bedsides naphthalen-2-amine (**3a**) substrate, anthracen-2-amine (**3b**) was also proven to be suitable for the reaction with 2-hydroxy-1,4-naphthoquinone (**2**) and sixteen arylaldehydes bearing electron-withdrawing or electron-donating groups to give the corresponding benzo[*h*]naphtho[2,3-*a*]acridine-

Table 2. The Three-Component Synthesis of Products 4a-4y

Entry	4[a]	Ar =	3	Time [b]	Yield [c]
1	**4a**	4-Chlorophenyl (**1a**)	**3a**	8	83
2	**4b**	2-Chlorophenyl(**1b**)	**3a**	8	82
3	**4c**	2,4-Dichlorophenyl(**1c**)	**3a**	7	77
4	**4d**	4-Fluorophenyl(**1d**)	**3a**	8	83
5	**4e**	4-Bromophenyl(**1e**)	**3a**	7	82
6	**4f**	4-Nitrophenyl(**1f**)	**3a**	10	80
7	**4g**	Phenyl(**1g**)	**3a**	8	82
8	**4h**	4-Tolyl(**1h**)	**3a**	8	79
9	**4i**	4-Methoxyphenyl(**1i**)	**3a**	8	81
10	**4j**	3,4-Dimethoxyphenyl(**1j**)	**3a**	10	81
11	**4k**	3,4,5-Trimethoxyphenyl(**1k**)	**3a**	10	75
12	**4l**	4-Hydroxy-3-nitrophenyl(**1l**)	**3a**	10	79
13	**4m**	Benzo[*d*][1, 3]dioxol-5-yl(**1m**)	**3a**	10	81
14	**4n**	Thien-2-yl(**1n**)	**3a**	10	76
15	**4o**	2-Chlorophenyl(**1b**)	**3b**	10	85
16	**4p**	2,4-Dichlorophenyl(**1c**)	**3b**	10	78
17	**4q**	4-Fluorophenyl(**1d**)	**3b**	8	83
18	**4r**	4-Bromophenyl(**1e**)	**3b**	8	87
19	**4s**	4-Nitrophenyl(**1f**)	**3b**	8	80
20	**4t**	4-Tolyl(**1h**)	**3b**	10	81
21	**4u**	4-Methoxyphenyl(**1i**)	**3b**	10	82
22	**4v**	4-Dimethylaminophenyl(**1o**)	**3b**	10	75
23	**4w**	3,4-Dimethoxyphenyl(**1j**)	**3b**	10	79
24	**4x**	3,4,5-Trimethoxyphenyl(**1k**)	**3b**	10	74
25	**4y**	Benzo[*d*][1, 3]dioxol-5-yl(**1m**)	**3b**	10	78

[a] Reagents and conditions: HOAc (1.5 mL), 120°C, MW; [b] min; [c] Isolated yield.

15,16(5*H*,14*H*)-dione in good yields (74-87%) and excellent selectivities (Table **1**, entries 15-25). For all these cases, the reaction proceeded rapidly to completion within 8-10 min. The present protocol provides a new, straightforward, and effective pathway to construct benzo[*h*]naphtho[2,3-*a*]acridine derivatives.

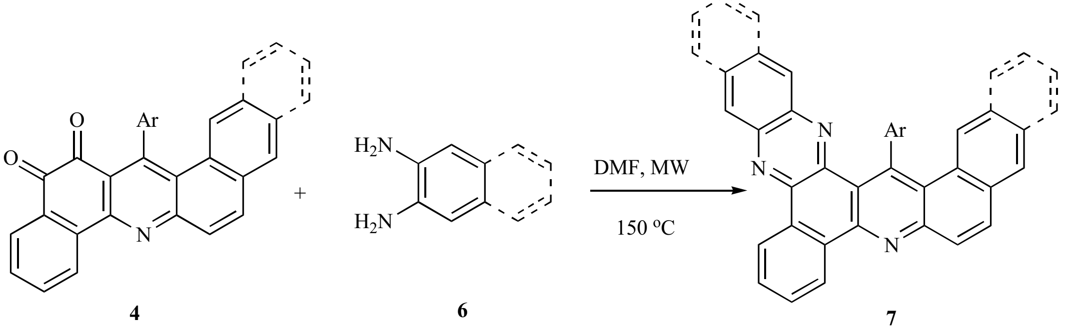
Table 3. The Synthesis of Products 7

Entry	Product	Diamine	4	Time/Min	Yield/%[b]
1	**7a**	**6a**	**4a**	20	90
2	**7b**	**6a**	**4b**	18	89
3	**7c**	**6a**	**4h**	18	81
4	**7d**	**6a**	**4i**	16	82
5	**7e**	**6a**	**4j**	20	89
6	**7f**	**6a**	**4k**	20	82
7	**7g**	**6a**	**4l**	20	89
8	**7h**	**6a**	**4m**	20	86
9	**7i**	**6a**	**4q**	18	87
10	**7j**	**6a**	**4r**	16	83
11	**7k**	**6a**	**4p**	18	82
12	**7l**	**6a**	**4w**	20	88
13	**7m**	**6b**	**4r**	20	84
14	**7n**	**6b**	**4x**	20	81

[a] Reagents and conditions: DMF (2.0 mL), 150°C, MW; [b] Isolated yield.

To study the applicational scope of this reaction, benzo[*h*]naphtho[2,3-*a*]acridine- 15,16(5*H*,14*H*)-dione **4** were further employed to react with benzene-1,2-diamine **6a** under microwave irradiation. After several solvents were screened, DMF was found to be the most suitable solvent for this condensation to afford quinoxaline-fused benzo[*h*]naphtho[2,3-*a*]acridine **7** in excellent yields (81-90%) (Table **3**) (Scheme **3**). The new extended angular fused aza-heterocycles, such as cctocyclic and nonacyclic-fused benzo[*h*]naphtho[2,3-*a*]acridines, were generated in high yields when the naphthalene-2,3-diamine **6b** was used as an *ortho*-diamine component under similar condition. This conversion can be finished within 20 min in this solvent at 150°C. It should be mentioned that the resulting quinoxaline-containing multiheterocycles would be endowed with many significant pharmacological properties, such as antiproliferatives, antimalarial, antitumor, antiamoebic, anti-Mycobacterium tuberculosis agents, and CB2 receptor antagonists [14].

Besides a high efficiency in the formation of multiple bonds as a domino process, this reaction has the following advantages:



Scheme 3. Thesynthesis ofquinoxaline-fused benzo[*h*]naphtho[2,3-*a*]acridine.

(1) the starting materials are readily available and the reagents are very cheap; (2) the reaction proceeds smoothly under very mild conditions without introducing strong acid, base or metal catalyst; (3) the environmentally friendly process in which water is the major by-product without additional use of organic solvent during reaction process; (4) the convenient work-up which only needs simple filtration since the products directly precipitate out after the reaction is finished [15, 16] and when its mixtures are diluted with cold water; and (5) the high regioselectivity in which the reactions generated naphthoacridine with *ortho*-diketone unit that serve as important building blocks. *The novelty of the present multicomponent domino reaction is shown by the fact that multiple chemical bonds' breaking and forming were simultaneously achieved in an intermolecular manner and in a one-pot operation.*

On the basis of all the above results, a possible mechanism has been proposed for the formations of naphthoacridines and its derivatives as shown in Scheme **4**. The formation of **4** involves a ring closure cascade process that consists of initial condensation, intermolecular Michael addition (**A** to **B**), intramolecular nucleophilic cyclization (**B** to **C**) and dehydration (**C** to **D**) (Scheme **4**). The intermediate **B** favors formation of intramolecular hydrogen bond between carbonyl group (position a) and ortho-hydroxyl group (position b), in which enolization of hydroxyl group was further enhanced. During this process, the carbonyl group (position c) would be easily attracted by the amino group (-NH_2) to give intermediate **C** which is then converted into the naphthoacridines *via* dehydration and isomerization steps [10f]. The synthesizing naphthoacridines was further subjected with *ortho*-diamines to give final extended angular fused aza-heterocycles **7** *via* dehydration and dehydrogenation processes.

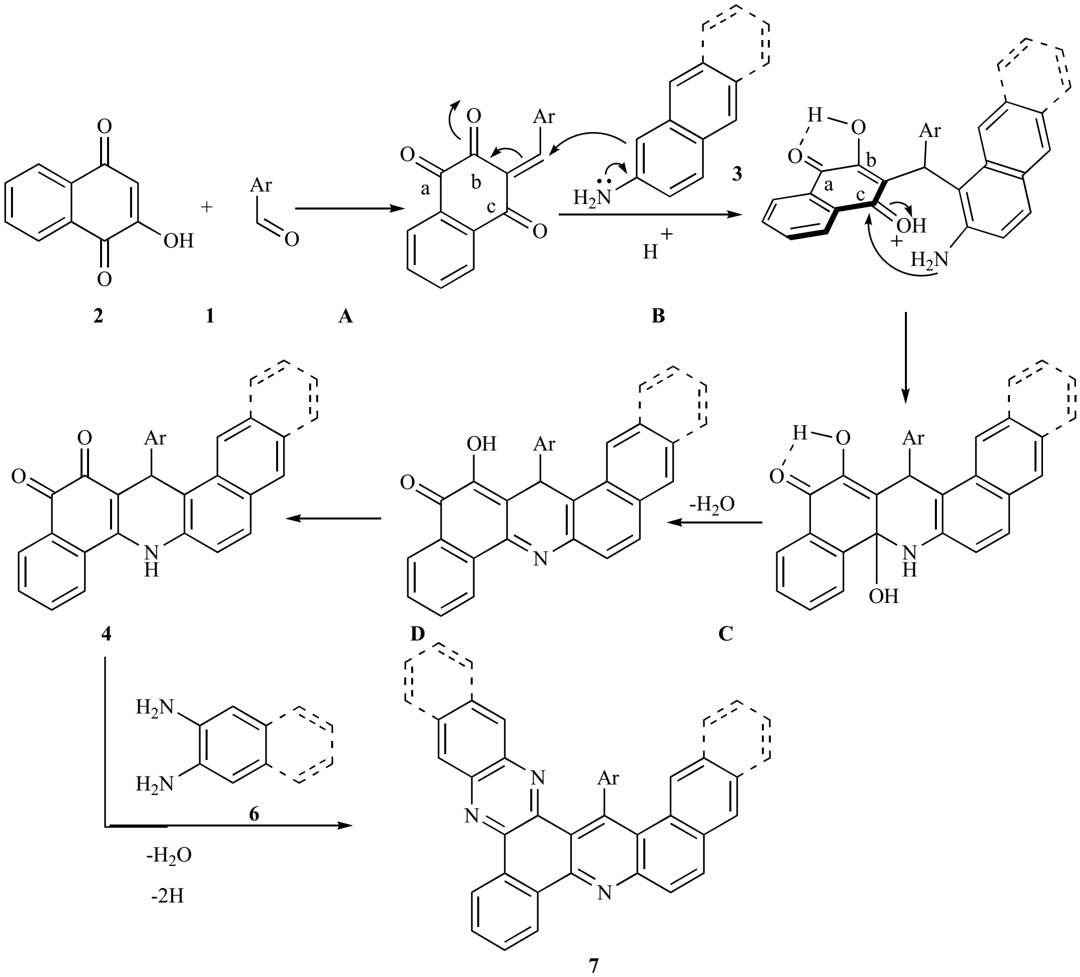
CONCLUSIONS

In summary, a new multicomponent domino reaction have been established to afford benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-diones that serve as versatile building blocks. The reactions showed high regioselectivity and a broad scopes of substrates which can employ a wide range of common commercial starting materials. A new mechanism has been proposed to explain the reaction process and regioselectivity. The resulting benzoquinoline products have been successfully converted into benzoquinoxaline-fused benzoquinolines by reacting with benzene-1,2-diamine under microwave irradiation.

EXPERIMENTAL SECTION

General

Microwave irradiation was carried out with Biotage from Personal Chemistry, Uppsala, Sweden. Melting points were



Scheme 4. The reasonable mechanism for the formation of products **4** and **7**.

determined in open capillaries and were uncorrected. IR spectra were taken on a FT-IR-Tensor 27 spectrometer in KBr pellets and reported in cm^{-1}. ^{1}H NMR spectra were measured on a Bruker DPX 400 MHz spectrometer in $CDCl_3$ or DMSO-d_6 (100 MHz, ^{13}C NMR) with chemical shift (δ) given in ppm relative to TMS as internal standard. HRMS (ESI) was determined by using microTOF-QⅡ HRMS/MS instrument (BRUKER). X-Ray crystallographic analysis was performed with a Siemens SMART CCD and a Siemens P4 diffractometer.

Typical procedure for the synthesis of benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-diones **4** (**4b** as an example): In a 10-mL Initiator reaction vial, 2-hydroxy-1,4-naphthoquinone **2** (1.0 mmol), 2-chlorobenzaldehyde **1b** (1.0 mmol), naphthalen-2-amine **3** (1.0 mmol) and AcOH (1.5 mL) were mixed and then capped. The mixture was irradiated for a given time at 120°C under microwave irradiation (initial power 100 W and maximum power 250 W). Upon completion, monitored by TLC, the reaction mixture was cooled to room temperature and washed with 2 mL 95% EtOH, filtered to give the crude products, which were further purified by recrystallization from 95% EtOH to afford the desired pure **4b**.

Typical procedure for the synthesis of benzo[*c*]phenazine[2,1-*b*]quinolines **7** (**7a** as an example): In a 10-mL Initiator reaction vial, 14-(4-Chlorophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione **4a**, benzene-1,2-diamine **6a** (1.0 mmol) and DMF (1.5 mL) were mixed and then capped. The mixture was irradiated for a given time at 150°C under microwave irradiation. Upon completion, monitored by TLC, the reaction mixture was cooled to room temperature and washed with 2 mL 95% EtOH, filtered to give the crude products, which were further purified by recrystallization from 1:1 mixture of MeOH and DMF to afford the desired pure **7a**.

14-(4-Chlorophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4a)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3401.2, 3050.6, 1589.2, 1515.2, 1500.3, 1402.6, 1051.4, 822.0, 801.8, 758.3, 670.5, 587.7; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.52 (s, 1H, NH), 8.47 (d, J = 8.0 Hz, 1H, ArH), 8.05-8.00 (m, 2H, ArH), 7.96-7.85 (m, 4H, ArH), 7.70 (t, J = 7.6 Hz, 1H, ArH), 7.53-7.49 (m, 1H, ArH), 7.45-7.41 (m, 1H, ArH), 7.35-7.33 (m, 2H, ArH), 7.24-7.22 (m, 2H, ArH), 6.05 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{17}ClNO_2$: 422.0943 [M+H]$^+$, found: 422.0950.

14-(2-Chlorophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4b)

m.p.: 278-280°C; IR (KBr, ν, cm^{-1}): 3305.6, 3048.7, 1580.1, 1520.4, 1489.6, 1400.9, 1060.8, 811.1, 758.3, 625.6; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.52 (s, 1H, NH), 8.52 (d, J = 8.0 Hz, 1H, ArH), 8.24 (d, J = 8.4 Hz, 1H, ArH), 8.02-8.00 (m, 1H, ArH), 7.97-7.91 (m, 2H, ArH), 7.89-7.84 (m, 2H, ArH), 7.73-7.70 (m, 1H, ArH), 7.54-7.50 (m, 1H, ArH), 7.43-7.41 (m, 2H, ArH), 7.30-7.28 (m, 1H, ArH), 7.15-7.11 (m, 1H, ArH), 7.08-7.04 (m, 1H, ArH), 6.25 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{17}ClNO_2$: 422.0943 [M+H]$^+$, found: 422.0955.

14-(2,4-Dichlorophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4c)

m.p.: 269-271°C; IR (KBr, ν, cm^{-1}): 3285.5, 3049.5, 1580.1, 1513.6, 1475.3, 1386.6, 1041.2, 905.3, 802.1, 667.8; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.50 (s, 1H, NH), 8.51 (d, J = 8.0 Hz, 1H, ArH), 8.16 (d, J = 8.8 Hz, 1H, ArH), 8.02-8.00 (m, 1H, ArH), 7.96-7.92 (m, 2H, ArH), 7.89 (d, J = 8.0 Hz, 1H, ArH), 7.85-7.83 (m, 1H, ArH), 7.71 (t, J = 7.6 Hz, 1H, ArH), 7.54-7.50 (m, 1H, ArH), 7.44-7.40 (m, 3H, ArH), 7.22 (dd, J_1 = 8.4 Hz, J_2 = 2.4 Hz, 1H, ArH), 6.22 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{16}Cl_2NO_2$: 456.0553 [M+H]$^+$, found: 456.0560.

14-(4-Fluorophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4d)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3320.1, 3051.5, 1603.4, 1523.9, 1508.0, 1345.5, 1052.4, 826.0, 839.6, 762.7, 676.7, 556.8; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.51 (s, 1H, NH), 8.46 (d, J = 8.0 Hz, 1H, ArH), 8.05 (d, J = 8.4 Hz, 1H, ArH), 8.00 (d, J = 8.0 Hz, 1H, ArH), 7.95-7.84 (m, 4H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.52-7.49 (m, 1H, ArH), 7.44-7.40 (m, 1H, ArH), 7.36-7.33 (m, 2H, ArH), 6.99 (t, J = 7.2 Hz, 2H, ArH), 6.05 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{17}FNO_2$: 406.1238 [M+H]$^+$, found: 406.1257.

14-(4-Bromophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4e)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3310.6, 3049.5, 1580.1, 1513.6, 1475.3, 1386.6, 1041.2, 905.3, 802.1, 667.8; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.51 (s, 1H, NH), 8.46 (d, J = 7.6 Hz, 1H, ArH), 8.03-8.01 (m, 2H, ArH), 7.96-7.89 (m, 3H, ArH), 7.70 (t, J = 7.6 Hz, 1H, ArH), 7.52-7.48 (m, 1H, ArH), 7.44-7.40 (m, 1H, ArH), 7.38-7.35 (m, 2H, ArH), 7.29-7.27 (m, 2H, ArH), 6.03 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{17}BrNO_2$: 466.0438 [M+H]$^+$, found: 466.0453.

14-(4-Nitrophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4f)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3288.4, 3052.7, 1608.9, 1501.4, 1420.5, 1333.9, 1315.0, 1260.4, 1199.2, 1172.5, 1120.0, 789.7; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.60 (s, 1H, NH), 8.50 (d, J = 7.6 Hz, 1H, ArH), 8.07-7.88 (m, 8H, ArH), 7.71 (t, J = 7.2 Hz, 1H, ArH), 7.62 (d, J = 8.4 Hz, 2H, ArH), 7.53-7.49 (m, 1H, ArH), 7.45-7.41 (m, 1H, ArH), 6.20 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{17}N_2O_4$: 433.1183 [M+H]$^+$, found: 433.1201.

14-Phenyldibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4g)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3305, 3050, 1698, 1541, 1524, 1347, 1218, 1118, 1053, 726; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.49 (s, 1H, NH), 8.46 (d, J = 8.0 Hz, 1H, ArH), 8.06 (d, J = 8.4 Hz, 1H, ArH), 8.00 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H, ArH), 7.95-7.84 (m, 4H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.52-7.50 (m, 1H, ArH), 7.41 (t, J = 7.2 Hz, 1H, ArH), 7.34 (s, 1H, ArH), 7.32 (s, 1H, ArH), 7.18-7.14 (m, 2H, ArH), 7.03 (t, J = 7.2 Hz, 1H, ArH), 6.04 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{18}NO_2$: 388.1333 [M+H]$^+$, found: 388.1362.

14-*p*-Tolyldibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4h)

m.p.: 289-291°C; IR (KBr, ν, cm^{-1}): 3355.1, 3051.6, 2867.8, 1681.3, 1561.0, 1524.4, 1460.9, 1416.5, 1259.8, 1038.1, 739.8; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.46 (s, 1H, NH), 8.45 (d, J = 8.0 Hz, 1H, ArH), 8.04 (d, J = 8.4 Hz, 1H, ArH), 7.99 (d, J = 7.6 Hz, 1H, ArH), 7.93-7.83 (m, 4H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.51-7.47 (m, 1H, ArH), 7.43-7.39 (m, 1H, ArH), 7.20-7.18 (m, 2H, ArH), 6.97-6.94 (m, 2H, ArH), 5.99 (s, 1H, CH) 2.11 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{28}H_{20}NO_2$: 402.1489 [M+H]$^+$, found: 402.1499.

14-(4-Methoxyphenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4i)

m.p.: 291-293°C; IR (KBr, ν, cm^{-1}): 3356.1, 3050.9, 1603.5, 1522.9, 1543.0, 1343.3, 1240.4, 1032.5, 841.5, 768.7, 753.2, 676.8; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.47 (s, 1H, NH), 8.45 (d, J = 8.0 Hz, 1H, ArH), 8.05 (d, J = 8.4 Hz, 1H, ArH), 7.99 (d, J = 7.6 Hz, 1H, ArH), 7.93-7.83 (m, 4H, ArH), 7.68 (t, J = 7.6 Hz, 1H, ArH), 7.51-7.48 (m, 1H, ArH), 7.43-7.39 (m, 1H, ArH), 7.23-7.21 (m, 2H, ArH), 6.73-6.71 (m, 2H, ArH), 5.98 (s, 1H, CH) 3.56 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{28}H_{20}NO_3$: 418.1438 [M+H]$^+$, found: 418.1460.

14-(3,4-Dimethoxyphenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4j)

m.p.: 271-273°C; IR (KBr, ν, cm^{-1}): 3401.3 3049.1, 2950.0, 1606.8, 1545.5, 1522.6, 1507.6, 1344.7, 1239.1, 1032.9, 840.8, 761.2, 753.4; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.48 (s, 1H, NH), 8.44 (d, J = 8.0 Hz, 1H, ArH), 8.09 (d, J = 8.4 Hz, 1H, ArH), 8.00 (d, J = 7.6 Hz, 1H, ArH), 7.94-7.88 (m, 3H, ArH), 7.87 (d, J = 8.4 Hz, 1H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.52-7.49 (m, 1H, ArH), 7.44-7.40 (m, 1H, ArH), 7.07 (d, J = 1.6 Hz, 1H, ArH), 6.69 (d, J = 8.4 Hz, 1H, ArH), 6.63-6.60 (m, 1H, ArH), 5.99 (s, 1H, CH) 3.66 (s, 3H, CH_3), 3.58 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{29}H_{22}NO_4$: 448.1544 [M+H]$^+$, found: 448.1558.

14-(3,4,5-Trimethoxyphenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4k)

m.p.: 282-284°C; IR (KBr, ν, cm^{-1}): 3403.2, 3038.1, 2940.7, 1589.5, 1523.6, 1528.2, 1507.7, 1346.3, 1238.2, 1039.1, 840.0, 767.6, 757.4; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.53 (s, 1H, NH), 8.45 (d, J = 7.6 Hz, 1H, ArH), 8.13 (d, J = 8.4 Hz, 1H, ArH), 8.01 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H, ArH), 7.96-7.90 (m, 3H, ArH), 7.84 (d, J = 8.8 Hz, 1H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.55-7.51 (m, 1H, ArH), 7.45-7.42 (m, 1H, ArH), 6.59 (s, 2H, ArH), 6.01 (s, 1H, CH) 3.61 (s, 6H, CH_3), 3.51 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{30}H_{24}NO_5$: 478.1649 [M+H]$^+$, found: 478.1674.

14-(4-Hydroxy-3-nitrophenyl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4l)

m.p.: 298-300°C; IR (KBr, ν, cm^{-1}): 3430.6, 3360.6, 3054.7, 1622.9, 1523.1, 1418.6, 1345.7, 1320.9, 1268.4, 1236.3, 1191.1, 1134.0, 765.9; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.75 (s, 1H, OH), 10.45 (s, 1H, NH), 8.46 (d, J = 8.8 Hz, 1H, ArH), 8.05 (d, J = 8.4 Hz, 1H, ArH), 8.01-7.90 (m, 4H, ArH), 7.86 (d, J = 8.8 Hz, 1H, ArH), 7.79 (d, J = 2.0 Hz, 1H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.54-7.49 (m, 2H, ArH), 7.43 (t, J = 7.6 Hz, 1H, ArH), 6.97(d, J = 8.4 Hz, 1H, ArH), 6.05 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{27}H_{17}N_2O_5$: 449.1132 [M+H]$^+$, found: 449.1160.

14-(Benzo[*d*][1,3]dioxol-5-yl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4m)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3388.6, 3023.1, 2940.6, 1611.8, 1534.7, 1583.5, 1525.6, 1356.6, 1224.7, 1023.4, 841.3, 768.2, 754.1; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.47 (s, 1H, NH), 8.45 (d, J = 8.0 Hz, 1H, ArH), 8.07 (d, J = 8.0 Hz, 1H, ArH), 8.01 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H, ArH), 7.94-7.89 (m, 3H, ArH), 7.84 (d, J = 8.8 Hz, 1H, ArH), 7.69 (t, J = 7.6 Hz, 1H, ArH), 7.54-7.49 (m, 1H, ArH), 7.45-7.41 (m, 1H, ArH), 6.88 (d, J = 2.0 Hz, 1H, ArH), 6.73-6.67 (m, 2H, ArH), 5.86 (s, 1H, CH) 5.85 (d, J = 8.0 Hz, 2H, CH_2). HRMS (ESI): m/z calcd for $C_{28}H_{18}NO_4$: 432.1231 [M+H]$^+$, found: 432.1253.

14-(Thiophen-2-yl)dibenzo[*a*,*h*]acridine-12,13(7*H*,14*H*)-dione (4n)

m.p.: 269-271°C; IR (KBr, ν, cm^{-1}): 3326.2, 3045.3, 1698.6, 1580.7, 1500.5, 1491.8, 1451.9, 1440.4, 1402.9, 1230.4, 1116.6, 877.9; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.61 (s, 1H, NH), 8.45 (d, J = 7.6 Hz, 1H, ArH), 8.11 (d, J = 8.8 Hz, 1H, ArH), 8.03 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H, ArH), 7.97-7.91 (m, 3H, ArH), 7.82 (d, J = 8.8 Hz, 1H, ArH), 7.71 (t, J = 7.6 Hz, 1H, ArH), 7.59-7.55 (m, 1H, ArH), 7.48-7.45 (m, 1H, ArH), 7.16 (dd, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H, ArH), 6.77-6.74 (m, 2H, ArH), 6.35 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{25}H_{16}NO_2S$: 394.0897 [M+H]$^+$, found: 394.0919.

14-(2-Chlorophenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-dione (4o)

m.p.: 283-285°C; IR (KBr, ν, cm^{-1}): 3320, 3051, 1670, 1603, 1580, 1564, 1502, 1458, 1449, 1252, 1170, 873, 725; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.51 (s, 1H, NH), 8.84 (s, 1H, ArH), 8.54-8.52 (m, 2H, ArH), 8.09 (d, J = 8.8 Hz, 1H, ArH), 8.03-7.93 (m, 4H, ArH), 7.86 (d, J = 9.2 Hz, 1H, ArH), 7.71 (t, J = 7.2 Hz, 1H, ArH), 7.54-7.46 (m, 3H, ArH), 7.30 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 2H, ArH), 7.13-7.10 (m, 2H, ArH), 7.05-7.01 (m, 2H, ArH), 6.37 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{31}H_{19}ClNO_2$: 472.1099 [M+H]$^+$, found: 472.1118.

14-(2,4-Dichlorophenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-dione (4p)

m.p.: 276-278°C; IR (KBr, ν, cm^{-1}): 3314, 3071, 1698, 1609, 1588, 1557, 1504, 1469, 1460, 1257, 1232, 877, 739; ^{1}H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.53 (s, 1H, NH), 8.77 (s, 1H, ArH), 8.53-8.52 (m, 2H, ArH), 8.10 (d, J = 9.2 Hz, 1H, ArH), 8.04-7.93 (m, 4H, ArH), 7.87 (d, J = 9.2 Hz, 1H, ArH), 7.72 (t, J = 7.6 Hz, 1H, ArH), 7.53-7.49 (m, 3H, ArH), 7.45 (d, J = 1.6 Hz, 1H, ArH), 7.21 (dd, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1H, ArH), 6.35 (s, 1H, CH). HRMS (ESI):

m/z calcd for $C_{31}H_{18}Cl_2NO_2$: 506.0710 $[M+H]^+$, found: 506.0729.

14-(4-Fluorophenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16 (5*H*,14*H*)-dione (4q)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3318, 3051, 1694, 1585, 1504, 1461, 1257, 1232, 889; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.54 (s, 1H, NH), 8.70 (s, 1H, ArH), 8.55 (s, 1H, ArH), 8.49 (d, *J* = 8.0 Hz, 1H, ArH), 8.16-8.11 (m, 2H, ArH), 8.05-8.01 (m, 2H, ArH), 7.93 (t, *J* = 7.2 Hz, 1H, ArH), 7.89 (d, *J* = 9.2 Hz, 1H, ArH), 7.71 (t, *J* = 7.6 Hz, 1H, ArH), 7.51-7.47 (m, 4H, ArH), 6.98 (t, *J* = 8.8 Hz, 2H, ArH), 6.23 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{31}H_{19}FNO_2$: 456.1395 $[M+H]^+$, found: 456.1416.

14-(4-Bromophenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16 (5*H*,14*H*)-dione (4r)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3330, 2912, 1585, 1504, 1482, 1469, 1462, 1255, 1011, 911, 778; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.56 (s, 1H, NH), 8.69 (s, 1H, ArH), 8.56 (s, 1H, ArH), 8.49 (d, *J* = 8.0 Hz, 1H, ArH), 8.16-8.12 (m, 2H, ArH), 8.05-8.01 (m, 2H, ArH), 7.94 (t, *J* = 7.2 Hz, 1H, ArH), 7.88 (d, *J* = 8.8 Hz, 1H, ArH), 7.71 (t, *J* = 7.2 Hz, 1H, ArH), 7.50-7.49 (m, 2H, ArH), 7.42 (d, *J* = 8.4 Hz, 2H, ArH), 7.36 (d, *J* = 8.4 Hz, 2H, ArH), 6.20 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{31}H_{19}BrNO_2$: 516.0594 $[M+H]^+$, found: 516.0611.

14-(4-Nitrophenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16 (5*H*,14*H*)-dione (4s)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3313, 3044, 1698, 1608, 1588, 1557, 1504, 1479, 1469, 1460, 1257, 877, 738, 631; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.62 (s, 1H, NH), 8.70 (s, 1H, ArH), 8.56 (s, 1H, ArH), 8.51 (d, *J* = 7.6 Hz, 1H, ArH), 8.16-8.14 (m, 2H, ArH), 8.06-8.02 (m, 4H, ArH), 7.97-7.90 (m, 2H, ArH), 7.76 (d, *J* = 8.8 Hz, 2H, ArH), 7.72 (t, *J* = 7.6 Hz, 1H, ArH), 7.52-7.46 (m, 2H, ArH), 6.37 (s, 1H, CH). HRMS (ESI): m/z calcd for $C_{31}H_{19}N_2O_4$: 483.1340 $[M+H]^+$, found: 483.1364.

14-*p*-Tolylbenzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-dione (4t)

m.p.: 292-294°C; IR (KBr, ν, cm^{-1}): 3335, 3048, 1585, 1504, 1481, 1470, 1461, 1424, 1255, 870, 737; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.50 (s, 1H, NH), 8.68 (s, 1H, ArH), 8.54 (s, 1H, ArH), 8.47 (d, *J* = 8.0 Hz, 1H, ArH), 8.14-8.09 (m, 2H, ArH), 7.95-7.91 (m, 1H, ArH), 7.87 (d, *J* = 8.8 Hz, 1H, ArH), 7.70 (t, *J* = 7.2 Hz, 1H, ArH), 7.50-7.47 (m, 2H, ArH), 7.33 (d, *J* = 8.0 Hz, 2H, ArH), 6.95 (d, *J* = 8.0 Hz, 2H, ArH), 6.17 (s, 1H, CH), 2.09 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{32}H_{22}NO_2$: 452.1646 $[M+H]^+$, found: 452.1671.

14-(4-Methoxyphenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15, 16(5*H*,14*H*)-dione (4u)

m.p.: 289-291°C; IR (KBr, ν, cm^{-1}): 3324, 1586, 1504, 1480, 1469, 1460, 1417, 1256, 1177, 1029, 870, 707; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.49 (s, 1H, NH), 8.69 (s, 1H, ArH), 8.53 (s, 1H, ArH), 8.47 (d, *J* = 8.0 Hz, 1H, ArH), 8.14-8.08 (m, 2H, ArH), 8.03-8.00 (m, 2H, ArH), 7.94-7.90 (m, 1H, ArH), 7.87 (d, *J* = 9.2 Hz, 1H, ArH), 7.69 (t, *J* = 7.6 Hz, 1H, ArH), 7.50-7.47 (m, 2H, ArH), 7.35 (d, *J* = 8.4 Hz, 2H, ArH), 6.71 (d, *J* = 8.8 Hz, 2H, ArH), 6.16 (s, 1H, CH), 3.58 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{32}H_{22}NO_3$: 468.1595 $[M+H]^+$, found: 468.1611.

14-(4-(Dimethylamino)phenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-dione (4v)

m.p.: 286-288°C; IR (KBr, ν, cm^{-1}): 3306, 3061, 1692, 1608, 1585, 1502, 1480, 1460, 1257, 874, 812, 738; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.45 (s, 1H, NH), 8.69 (s, 1H, ArH), 8.53 (s, 1H, ArH), 8.46 (d, *J* = 8.0 Hz, 1H, ArH), 8.13-8.07 (m, 2H, ArH), 8.04-8.00 (m, 2H, ArH), 7.92 (t, *J* = 7.6 Hz, 1H, ArH), 7.85 (d, *J* = 9.2 Hz, 1H, ArH), 7.69 (t, *J* = 7.6 Hz, 1H, ArH), 7.51-7.45 (m, 2H, ArH), 7.24 (d, *J* = 8.8 Hz, 2H, ArH), 6.50 (d, *J* = 8.8 Hz, 2H, ArH), 6.09 (s, 1H, CH), 2.70 (s, 6H, CH_3). HRMS (ESI): m/z calcd for $C_{33}H_{25}N_2O_2$: 481.1911 $[M+H]^+$, found: 481.1929.

14-(3,4-Dimethoxyphenyl)benzo[*h*]naphtho[2,3-*a*]acridine- 15, 16(5*H*,14*H*)-dione (4w)

m.p.: 274-276°C; IR (KBr, ν, cm^{-1}): 3327, 3062, 2930, 1692, 1587, 1504, 1481, 1470, 1462, 1264, 1238, 1140, 1029, 871, 737; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.50 (s, 1H, NH), 8.76 (s, 1H, ArH), 8.54 (s, 1H, ArH), 8.47 (d, *J* = 7.6 Hz, 1H, ArH), 8.16 (d, *J* = 7.2 Hz, 1H, ArH), 8.10 (d, *J* = 8.8 Hz, 1H, ArH), 8.04-8.01 (m, 2H, ArH), 7.94-7.91 (m, 1H, ArH), 7.87 (d, *J* = 8.8 Hz, 1H, ArH), 7.72-7.68 (m, 1H, ArH), 7.50-7.47 (m, 2H, ArH), 7.29 (d, *J* = 2.0 Hz, 1H, ArH), 6.74-6.67 (m, 2H, ArH), 6.53 (s, 1H, CH), 3.72 (s, 3H, CH_3), 3.55 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{33}H_{24}NO_4$: 498.1700 $[M+H]^+$, found: 498.1721.

14-(3,4,5-Trimethoxyphenyl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-dione (4x)

m.p.: 290-293°C; IR (KBr, ν, cm^{-1}): 3321, 3047, 1692, 1587, 1503, 1481, 1469, 1461, 1417, 1251, 1124, 871; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.55 (s, 1H, NH), 8.82 (s, 1H, ArH), 8.55 (s, 1H, ArH), 8.48 (d, *J* = 8.0 Hz, 1H, ArH), 8.21 (d, *J* = 8.4 Hz, 1H, ArH), 8.11 (d, *J* = 9.2 Hz, 1H, ArH), 8.03 (m, 2H, ArH), 7.94-7.86 (m, 2H, ArH), 7.70 (t, *J* = 7.6 Hz, 1H, ArH), 7.53-7.46 (m, 2H, ArH), 6.77 (s, 2H, ArH), 6.18 (s, 1H, CH), 3.64 (s, 6H, CH_3), 3.48 (s, 3H, CH_3). HRMS (ESI): m/z calcd for $C_{34}H_{26}NO_5$: 528.1806 $[M+H]^+$, found: 528.1819.

14-(Benzo[*d*][1,3]dioxol-5-yl)benzo[*h*]naphtho[2,3-*a*]acridine-15,16(5*H*,14*H*)-dione (4y)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3320, 3006, 2920, 1684, 1580, 1524, 1477, 1465, 1455, 1259, 1235, 1144, 1010, 872, 739; ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 10.50 (s, 1H, NH), 8.72 (s, 1H, ArH), 8.54 (s, 1H, ArH), 8.47 (d, *J* = 7.6 Hz, 1H, ArH), 8.17-8.15 (m, 1H, ArH), 8.12 (d, *J* = 9.2 Hz, 1H, ArH), 8.05-8.01 (m, 2H, ArH), 7.94-7.90 (m, 1H, ArH), 7.87 (d, *J* = 9.2 Hz, 1H, ArH), 7.70 (t, *J* = 7.6 Hz, 1H, ArH), 7.51-7.48 (m, 2H, ArH), 7.04 (d, *J* = 1.6 Hz, 1H, ArH), 6.86-6.84 (m, 1H, ArH), 6.68 (d, *J* = 8.0 Hz, 1H, ArH), 6.15 (s, 1H, CH), 5.83 (d, *J* = 10.0 Hz, 1H, CH_2). HRMS (ESI): m/z calcd for $C_{32}H_{20}NO_4$: 482.1387 $[M+H]^+$, found: 482.1398.

18-(4-Chlorophenyl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*] quinoline (7a)

m.p.: 279-281°C; IR (KBr, ν, cm^{-1}): 3051.8, 1603.4, 1523.9, 1508.0, 1345.5, 1052.4, 826.0, 839.6, 762.7, 676.7, 556.8; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.38 (d, *J* = 8.0 Hz, 1H, ArH), 9.19 (d, *J* = 8.0 Hz,1H, ArH), 8.33-8.21 (m, 3H, ArH) , 8.11 (d, *J* = 8.0 Hz, 1H, ArH), 8.01-7.87 (m, 4H, ArH), 7.73 (d, *J* = 8.0 Hz, 2H, ArH), 7.62 (t, *J* = 7.6 Hz, 1H, ArH), 7.55 (d, *J* = 9.2 Hz, 1H, ArH), 7.48 (d, *J* = 8.0Hz, 2H, ArH), 7.30-7.22 (m, 2H, ArH); HRMS (ESI): m/z calcd for $C_{33}H_{18}ClN_3$: 492.1262 $[M+H]^+$, found: 492.1296.

18-(2-Chlorophenyl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7b)

m.p.: 278-280°C; IR (KBr, ν, cm^{-1}): 3047.3, 2887.3, 1522.5, 1489.3, 1446.9, 1346.7, 1242.6, 1228.6, 1041.1, 941.1, 764.4, 676.4; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.54 (d, *J* = 8.0 Hz, 1H, ArH), 9.31 (d, *J* = 7.6 Hz, 1H, ArH), 8.31 (d, *J* = 8.8 Hz, 1H, ArH), 8.22 (d, *J* = 8.4 Hz, 1H, ArH), 8.11 (d, *J* = 8.8 Hz, 1H, ArH), 7.94-7.85 (m, 3H, ArH), 7.79-7.71 (m, 3H, ArH), 7.65 (t, *J* = 7.2 HZ, 1H, ArH), 7.57-7.53 (m, 2H, ArH), 7.51-7.47 (m, 1H, ArH), 7.30-7.27 (m, 2H, ArH), 7.23-7.21 (m, 1H, ArH); HRMS (ESI): m/z calcd for $C_{33}H_{18}ClN_3$: 492.1262 $[M+H]^+$, found: 492.1351.

18-*p*-Tolyl benzo[*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7c)

m.p.: 296-298°C; IR (KBr, ν, cm^{-1}): 3051.6, 2867.8, 1681.3, 1561.0, 1524.4, 1460.9, 1416.5, 1259.8, 1038.1, 739.8; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = (d, *J* = 7.6 Hz, 1H, ArH), 9.28 (d, *J* = 7.2 Hz, 1H, ArH), 8.20 (d, *J* = 8.4 Hz, 1H, ArH), 8.09 (d, *J* = 8.8 Hz, 1H, ArH), 7.93-7.84 (m, 3H, ArH), 7.77-7.73 (m, 1H, ArH), 7.67-7.62 (m, 2H, ArH), 7.51(t, *J* = 7.2 Hz, 1H, ArH), 7.44 (d, *J* = 8.0 Hz, 2H, ArH), 7.29-7.24 (m, 3H, ArH), 7.20-7.16 (m, 1H, ArH), 2.69 (s, 3H, CH3); HRMS (ESI): m/z calcd for $C_{34}H_{21}N_3$: 472.1808 $[M+H]^+$, found: 472.1849.

18-(4-Methoxyphenyl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*] quinoline (7d)

m.p.: 288-290°C; IR (KBr, ν, cm^{-1}): 3050.9, 1603.5, 1522.9, 1543.0, 1343.3, 1240.4, 1032.5, 841.5, 768.7, 753.2, 676.8; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.36 (d, *J* = 8.0 Hz, 1H, ArH), 9.18 (d, *J* = 7.6 Hz, 1H, ArH), 8.29-8.18 (m, 3H, ArH), 8.08 (d, *J* = 7.2 Hz, 1H, ArH), 8.01-7.85 (m, 3H, ArH), 7.83 (t, *J* = 7.2 Hz, 1H, ArH), 7.61-7.57 (m, 2H, ArH), 7.31-7.21 (m, 6H, ArH), 3.99 (s, 3H, CH_3); HRMS (ESI): m/z calcd for $C_{34}H_{21}N_3O$: 488.1757 $[M+H]^+$, found: 488.1792.

18-(3,4-Dimethoxyphenyl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7e)

m.p.: 295-297°C; IR (KBr, ν, cm^{-1}): 3051.5, 2901.7, 1600.7, 1510.3, 1529.9, 1341.4, 1238.6, 1033.5, 838.8, 764.2, 752.6, 677.7; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.36 (d, *J* = 7.6 Hz, 1H, ArH), 9.18(d, *J* = 7.6 Hz, 1H, ArH), 8.29-8.18 (m, 3H, ArH), 8.07 (d, *J* =7.6 Hz,1H, ArH), 8.00-7.82 (m, 4H, ArH),7.66 (d, *J* = 8.4 Hz, 1H, ArH), 7.60 (t, *J* = 7.6 Hz, 1H, ArH), 7.28-7.21 (m, 2H, ArH), 7.22 (d, *J* = 8.0 Hz, 1H, ArH), 7.08 (d, *J* = 1.6 Hz, 1H, ArH), 6.86 (dd, J_1 = 8.0 Hz, J_2 = 1.2 Hz, 1H, ArH), 4.00 (s, 3H, CH_3), 3.56 (s, 3H, CH_3); HRMS (ESI): m/z calcd for $C_{35}H_{23}N_3O_2$: 518.1863 $[M+H]^+$, found: 518.1891.

18-(3,4,5-Trimethoxyphenyl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7f)

m.p.: 290-292°C; IR (KBr, ν, cm^{-1}): 2990.9, 1581.3, 1517.8, 1503.5, 1352.4, 1234.0, 1126.9, 1011.5, 873.2, 767.6, 739.3; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.40 (d, *J* = 8.0 Hz, 1H, ArH), 9.22 (d, *J* = 7.6 Hz, 1H, ArH), 8.32-8.21 (m, 3H, ArH), 8.09 (dd, J_1 = 8.0Hz, J_2 =0.8 Hz, 1H, ArH), 8.01-7.85 (m, 4H, ArH), 7.68 (d, *J* = 9.2 Hz, 1H, ArH), 7.62 (t, *J* = 7.2 Hz, 1H, ArH), 7.39 (d, *J* = 8.4 Hz, 1H, ArH), 7.33-7.29 (m, 1H, ArH), 6.78 (s, 2H, ArH), 3.99 (s, 3H, CH_3), 3.56 (s, 6H, CH_3); HRMS (ESI): m/z calcd for $C_{36}H_{25}N_3O_3$: 548.1969 $[M+H]^+$, found: 548.1989.

18-(4-Hydroxy-3-nitrophenyl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7g)

m.p.: >300°C; IR (KBr, ν, cm^{-1}): 3257, 3021, 1589, 1516, 1501, 1351, 1238, ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.45 (d, *J* = 7.2 Hz, 1H, ArH), 9.28 (d, *J* = 7.6 Hz, 1H, ArH), 8.25-8.22 (m, 2H, ArH), 8.09 (d, *J* = 9.2Hz, 1H, ArH), 8.04 (d, *J* = 2.0 Hz, 1H, ArH), 7.93-7.85 (m, 3H, ArH), 7.81-7.74 (m, 2H, ArH), 7.68 (t, *J* = 6.8 Hz, 1H, ArH), 7.62 (d, *J* = 7.6 Hz, 1H, ArH), 7.57-7.51 (m, 2H, ArH), 7.32 (d, *J* = 8.0 Hz, 1H, ArH), 7.62 (s, 1H, ArH); HRMS (ESI): m/z calcd for $C_{33}H_{18}N_4O_3$: 519.1452 $[M+H]^+$, found: 519.1473.

18-(Benzo[*d*][1,3]dioxol-6-yl)benzo[*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7h)

m.p.: 168-171°C; IR (KBr, ν, cm^{-1}): 3051.8, 1513.6, 1400.2, 1355.7, 1323.6, 1248.9, 1024.9, 861.2, 799.5, 740.1; ^1H NMR (400MHz, $CDCl_3$, 25°C): δ = 9.44 (d, *J* = 8.0 Hz, 1H, ArH), 9.26 (d, *J* = 8.0 Hz, 1H, ArH), 8.23-8.20 (m, 2H, ArH), 8.07 (d, *J* = 8.8 Hz, 1H, ArH), 7.92-7.75 (m, 5H, ArH), 7.71-7.67 (m, 1H, ArH), 7.54 (t, *J* = 7.6 Hz, 1H, ArH), 7.45 (d, *J* = 8.4 Hz, 1H, ArH), 7.29-7.25 (m, 1H, ArH), 7.09 (d, *J* = 8.0 Hz, 1H, ArH), 6.88 (s, 1H, ArH), 6.85 (dd, J_1 = 8.0Hz, J_2 = 1.2 Hz, 1H, ArH), 6.19 (s, 2H, CH_2); HRMS (ESI): m/z calcd for $C_{34}H_{19}N_3O_2$: 502.1550 $[M+H]^+$, found: 502.1589.

19-(4-Fluorophenyl)naphtho[2,3-*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7i)

m.p.: 289-291°C; IR (KBr, ν, cm^{-1}): 3054, 1517, 1346, 1318, 1054, 1011. ^1H NMR (400MHz, DMSO-d_6, 25°C): δ = 9.51 (d, *J* = 8.0 Hz, 1H, ArH), 9.32 (d, *J* = 7.6 Hz, 1H, ArH), 8.35 (s, 1H, ArH), 8.23 (d, *J* = 7.6 Hz, 1H, ArH), 8.14 (s, 1H, ArH), 7.99 (d, *J* = 8.0 Hz, 1H, ArH), 7.84-7.77 (m, 3H, ArH), 7.71-7.68 (m, 2H, ArH), 7.59-7.49 (m, 1H, ArH), 7.47-7.42 (m, 6H, ArH), 7.37 (d, *J* = 7.6 Hz, 1H, ArH); HRMS (ESI): m/z calcd for $C_{37}H_{20}FN_3$: 526.1715 $[M+H]^+$, found: 526.1699.

19-(4-Bromophenyl)naphtho[2,3-*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7j)

m.p.: 280-282^{o}C; IR (KBr, ν, cm^{-1}): 3053.2, 1519.3, 1485.4, 1343.8, 1316.1, 1089.5, 1011.8, 955.7, 891.7, 769.0, 742.7; ^{1}H NMR (400MHz, CDCl$_3$, 25^{o}C): δ = 9.49 (d, J = 7.6 Hz, 1H, ArH), 9.30 (d, J = 7.6 Hz, 1H, ArH), 8.34 (s, 1H, ArH), 8.23 (d, J = 8.4 Hz, 1H, ArH), 8.12 (s, 2H, ArH), 8.00-7.97 (m, 2H, ArH), 7.94-7.88 (m, 1H, ArH), 7.87-7.84 (m, 3H, ArH), 7.82-7.77 (m, 1H, ArH), 7.75-7.71 (m, 1H, ArH), 7.58-7.48 (m, 2H, ArH), 7.43 (d, J = 8.0 Hz, 1H, ArH), 7.38-7.34 (m, 3H, ArH); HRMS (ESI): m/z calcd for $C_{37}H_{20}BrN_3$: 586.0913 [M+H]$^+$, found: 586.0912.

19-(2,4-Dichlorophenyl)naphtho[2,3-*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7k)

m.p.: 221-222^{o}C; IR (KBr, ν, cm^{-1}): 3054.6, 1516.4, 1470.8, 1352.3, 1090.6, 1018.2, 890.0, 768.1, 748.9; ^{1}H NMR (400MHz, DCCl$_3$, 25^{o}C): δ = 9.51 (dd, J_1 = 8.0 Hz, J_2 = 1.2 Hz, 1H, ArH), 9.32 (d, J = 7.6 Hz, 1H, ArH), 8.38 (s, 1H, ArH), 8.26 (d, J = 8.4 Hz, 1H, ArH), 8.16-8.13 (m, 2H, ArH), 8.04-8.007.95-7.80 (m, 4H, ArH), 7.77-7.73 (m, 1H, ArH), 7.60-7.48 (m, 4H, ArH), 7.40-7.38 (m, 1H, ArH), 7.30 (d, J = 8.0 Hz, 1H, ArH); HRMS (ESI): m/z calcd for $C_{37}H_{19}Cl_2N_3$: 576.1029 [M+H]$^+$, found: 576.1017.

19-(3,4-Dimethoxyphenyl)naphtho[2,3-*f*](benzo[*c*]phenazine)[2,1-*b*]quinoline (7l)

m.p.: >300^{o}C; IR (KBr, ν, cm^{-1}): 3053.6, 1510.9, 1463.5, 1346.7, 1313.1, 1248.1, 1142.3, 1025.8, 893.1, 766.8, 743.6; ^{1}H NMR (400MHz, DCCl$_3$, 25^{o}C): δ = 9.54 (d, J = 7.2 Hz, 1H, ArH), 9.32 (d, J = 7.2 Hz, 1H, ArH), 8.35 (s, 1H, ArH), 8.23 (d, J = 7.6 Hz, 1H, ArH), 8.15-8.11 (m, 2H, ArH), 7.99 (d, J = 8.4 Hz, 1H, ArH), 7.95-7.85 (m, 2H, ArH), 7.80-7.77 (m, 1H, ArH), 7.69-7.65 (m, 1H, ArH), 7.56-7.52 (m, 1H, ArH), 7.45-7.44 (m, 2H, ArH), 7.41 (d, J = 7.6 Hz, 1H, ArH), 7.20 (d, J = 8.0 Hz, 1H, ArH), 7.11 (d, J = 2.0 Hz, 1H, ArH), 9.92 (dd, J_1 = 8.0 Hz, J_2 = 2.0 Hz, 1H, ArH), 4.19 (s, 3H, CH$_3$), 3.74 (s, 3H, CH$_3$); HRMS (ESI): m/z calcd for $C_{37}H_{22}ClN_3$: 568.2020 [M+H]$^+$, found: 568.2020.

21-(4-Bromophenyl)naphtho[2,3-*f*](dibenzo[*c,j*]phenazine)[2,1-*b*]quinoline (7m)

m.p.: >300^{o}C; IR (KBr, ν, cm^{-1}): 3049.9, 1557.3, 1539.2, 1486.8, 1391.6, 1371.1, 1347.9, 1272.8, 892.7, 882.5, 720.6; ^{1}H NMR (400MHz, DCCl$_3$, 25^{o}C): δ = 9.41 (d, J = 8.0 Hz, 1H, ArH), 9.30 (d, J = 8.0 Hz, 1H, ArH), 8.80 (s, 1H, ArH), 8.35 (s, 1H, ArH), 8.14-8.11 (m, 4H, ArH), 8.08-8.06 (m, 1H, ArH), 8.00 (d, J = 8.0 Hz, 1H, ArH), 7.91-7.86 (m, 4H, ArH), 7.87-7.83 (m, 1H, ArH), 7.58-7.54 (m, 3H, ArH), 7.52-7.45 (m, 2H, ArH), 7.40 d, J = (8.4 Hz, 2H, ArH); HRMS (ESI): m/z calcd for $C_{41}H_{22}BrN_3$: 636.1070 [M+H]$^+$, found: 636.1109.

21-(3,4,5-Trimethoxyphenyl)naphtho[2,3-*f*](dibenzo[*c,j*]phenazine)[2,1-*b*]quinoline (7n)

m.p.: >300^{o}C; IR (KBr, ν, cm^{-1}): 2990.9, 1581.3, 1517.8, 1503.5, 1352.4, 1234.0, 1126.9, 1011.5, 873.2, 767.6, 739.3; ^{1}H NMR (400MHz, DCCl$_3$, 25^{o}C): δ = 9.43 (d, J = 8.0 Hz, 1H, ArH), 9.32 (d, J = 8.0 Hz, 1H, ArH), 8.82 (s, 1H, ArH), 8.36 (s, 1H, ArH), 8.21 (s, 1H, ArH), 8.15-8.13 (m, 2H, ArH), 8.10-8.08 (m, 2H, ArH), 8.03-7.99 (m, 2H, ArH), 7.93-7.83 (m, 2H, ArH), 7.57-7.54 (m, 4H, ArH), 7.47-7.44 (m, 1H, ArH), 6.78 (s, 2H, ArH), 4.34 (s, 3H, CH$_3$), 3.69 (s, 6H, CH$_3$); HRMS (ESI): m/z calcd for $C_{44}H_{29}N_3O_3$: 648.2282 [M+H]$^+$, found: 648.2354.

Crystal data for 7g: $C_{33}H_{18}N_4O_3$, yellow, crystal dimension 0.34 x 0.09 x 0.07 mm, Triclinic, space group P-1, a = 10.2377(9) Å, b = 10.8522(12) Å, c = 11.8273(13) Å, α = 98.8780(10)^{o}, γ = 110.247(2)^{o}, β = 100.8140(10)^{o}, V = 1176.2(2) Å^3, Mr = 518.51, Z = 2, Dc = 1.464 Mg/m^3, λ = 0.71073 Å, μ(Mo K$_\alpha$) = 0.096 mm^{-1}, F(000) = 536, R = 0.0878, wR_2 = 0.1452, S = 1.053, largest diff. Peak and hole: 0.316 and -0.210 e/Å^3.

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CONFLICT OF INTEREST

Declared none.

SUPPLEMENTARY MATERIAL

Supplementary material is available on the publisher's web site along with the published article.

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