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Synthesis and characterization of oxadisilole-fused acridines, dioxatrisilole-fused acridines and benzo[*b*]acridines

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ABSTRACT

Oxadisilole-fused acridines, dioxatrisilole-fused acridines and benzo[*b*]acridines were synthesized through nucleophilic additions and aromatization reactions of arynes with 2-aminoaryl ketones or 2-aminoaryl aldehydes in good yields at room temperature. The photophysical, redox and thermal properties of these compounds were characterized. These compounds show potential applications as strong deep-blue or green emitters for OLED because of high fluorescence quantum yields and good thermal stabilities.

1. Introduction

Acridines are one of the important classes of nitrogen containing heterocyclic compounds. Their synthesis and application have been widely studied since had been first extracted by Carl Gräbe and Heinrich Caro in 1871.[1] Due to acridines' planar tricyclic aromatic molecules, which possess the ability to intercalate tightly but reversibly to DNA helical structure,[2] these compounds are one of the most successful classes of bioactive agents and widely utilized as antimalarial,[3] antiprotozoal,[4] antibacterial[5] and antileishmanial.[6] In addition, acridines, including 9-phenyl acridines, exhibit anticancer activities by inducing apoptosis in human cancer cell lines.[2a,7] Meanwhile, acridine derivatives are of pharmaceutical interest as an antiseptic in ophthalmic surgery.[8] Such an important class of nitrogen heterocycles also have several significant properties such as pigment and dye properties,[9] photochemical/physical properties[10] and electrochemical properties.[11] They have been used in the construction of new fluorescent probes that emit light at longer wavelengths in order to inhibit unwanted absorption, autofluorescence and scattering by neighbouring biological tissue.[12] They are also used as an active medium for dye lasers.[13] Iridium complexes with acridines have also been used as triplet state, light emitting materials for applications in OLEDs.[14]

In the early years, the methods for synthesize acridines required rather harsh conditions (high temperatures,[15] strongly acidic media,[16] valued catalyst,[17] uncommon reagents or methods[18]). In recent years, several of reports proved that acridines have been synthesized from benzyne and 2-aminobenzophenones. In 2005, Kim et al.[19] used 5-arylthianthrenium perchlorates as benzyne precursors reacted with 2-aminobenzophenone to acquire acridines. This methodology appears to be a really progress in the scope and yields, except the using of no commercial source for Kim's benzyne precursor and highly explosive salt. In 1960s,[20] 2-aminobenzophenones have been reported having good reaction with dimethyl acetylenedicarboxylate to form stable aromatic compounds. Thus, Larock et al.[21] tried their aryne with 2-aminobenzophenones to find a milder and more efficient way to synthesize acridines. Recently, we reported the synthesis of heterocyclic compounds from our arynes generated from benzobisoxadisilole and 2,3-naphthoxadisilole.[22] In this paper, we report our findings on the synthesis of oxadisilole-fused acridines **5a**–**i**, dioxatrisilole-fused acridines **6a**–**i** and benzo[*b*]acridines **10a**–**g** via nucleophilic additions and aromatization reactions of arynes **3** or **9** with 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** in good yields at room temperature. The benzyne **3** or naphthyne **9** was generated from benzobis(oxadisilole) **1** or 2,3-naphthoxadisilole **7**, respectively (Schemes 1 and 2).[23] The photophysical, redox and thermal properties of these previously unknown **5a**–**i**, **6a**–**i** or **10a**–**g** were characterized. The possible mechanism for the formation of products was also suggested.

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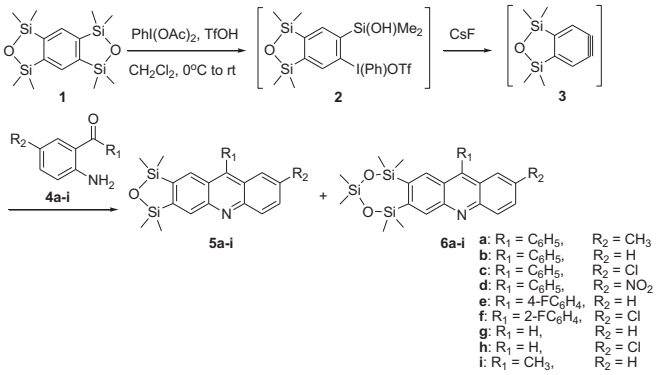
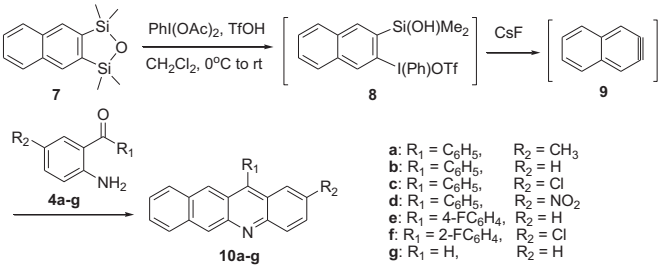


Scheme 1. Syntheses of oxadisilole-fused acridines **5a**–**i** and dioxatrisilole-fused acridines **6a**–**i**.



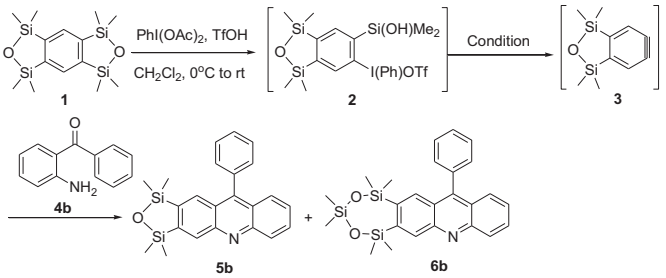
Scheme 2. Syntheses of benzo[*b*]acridines **10a**–**g**.

2. Results and discussion

The syntheses of oxadisilole-fused acridines **5a**–**i**, dioxatrisilole-fused acridines **6a**–**i** or benzo[*b*]acridines **10a**–**g** are outlined in Schemes 1 and 2. Arynes **3** and **9** were generated from benzobis(oxadisilole) **1** and 2,3-naphthoxadisilole **7**, respectively, through our previously reported phenyliodination fluoride induced desilylation protocol.[23] Trapping benzyne **3** and naphthyne **9** at room temperature with 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** by nucleophilic additions and aromatization reactions afforded oxadisilole-fused acridine derivatives **5a**–**i**, dioxatrisilole-fused acridine derivatives **6a**–**i** and benzo[*b*]acridine derivatives **10a**–**g** in good yields.

We optimized the procedure by starting with the reaction of 2-amino benzophenone **4b** and oxadisilole-fused benzyne **3** generated in situ from benzobis(oxadisilole) **1**. Phenyliodination of **1** (1.0 equiv) took place readily at room temperature in dichloromethane with a 1:2 mixture of phenyliodium diacetate ($PhI(OAc)_2$) and trifluoromethanesulfonic acid (TfOH) (Table 1). Without isolation of the ring-opened iodination intermediate **2**, the solvent (CH_2Cl_2) was removed under reduced pressure, benzyne **3** could be generated in situ upon treatment with a cesium fluoride (CsF) (1.0 equiv) in acetonitrile (CH_3CN). Trapping experiments were carried out with 2-amino benzophenone **4b** (0.4 equiv) at 0 °C to form oxadisilole-fused acridine derivative **5b** and dioxatrisilole-fused acridine derivative **6b** in overall 70% yield (Table 1, entry 1). When the reaction temperature was changed from 0 °C to room temperature, 40 °C, 60 °C, or reflux, the yield of desired products (**5b** and **6b**) was changed from 70% to 84%, 58%, 67% or 48%, respectively (Table 1, entries 2–5). At room temperature, using 2.0 equiv or 3.0 equiv of CsF as fluoride source, the isolated yield of products (**5b** and **6b**) were 58% or 76%, respectively (Table 1, entries 6 and 7). Using 1.0 equiv of potassium fluoride (KF) or tetrabutylammonium fluoride (TBAF) in CH_3CN at room temperature, the isolated yield of mixture products (**5b** and **6b**) were 71% or 41%, respectively (Table 1, entries 8 and 9). When the reaction was carried out in various solvents, such as toluene, THF or DCM at room temperature, the results are tabulated in Table 1 (Entries 10–12). The optimal conditions were obtained when the reaction of oxadisilole-fused benzyne **3** and **4b** (0.4 equiv) was carried out in CH_3CN at room temperature, and CsF (1.0 equiv) as fluoride source (Table 1, entry 2).

Table 1
Reaction of 2-amino benzophenone **4b** (0.4 equiv) with benzyne **3** generated from benzobis(oxadisilole) **1** (1.0 equiv) using different fluoride sources, solvent and temperature



Entry	Conditions	Yield[a] of **5b**+**6b** %
1	CsF (1.0 equiv), CH_3CN, 0 °C	70
2	CsF (1.0 equiv), CH_3CN, r.t	84
3	CsF (1.0 equiv), CH_3CN, 40 °C	58
4	CsF (1.0 equiv), CH_3CN, 60 °C	67
5	CsF (1.0 equiv), CH_3CN, reflux	48
6	CsF (2.0 equiv), CH_3CN, rt.	58
7	CsF (3.0 equiv), CH_3CN, rt.	76
8	KF (1.0 equiv), CH_3CN, rt.	71
9	TBAF (1.0 equiv), CH_3CN, rt.	41
10	CsF (1.0 equiv), Toluene, rt.	35
11	CsF (1.0 equiv), THF, rt.	72
12	CsF (1.0 equiv), DCM, rt.	77

[a] Isolated yield.

We further studied the nucleophilic additions and aromatization reactions of benzyne **3** generated in situ from benzobis(oxadisilole) **1** with 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** at room temperature. The oxadisilole-fused acridines **5a**–**i** and dioxatrisilole-fused acridines **6a**–**i** were formed in 30%–89% total yields (Scheme 1 and Table 2, entries 1–9).

As shown in Table 2, the reaction of 2-amino benzophenone **4b** (R_1=C_6H_5, R_2=H) or 2-amino acetophenone (R_1=CH_3, R_2=H) **4i** with **3** gave higher total yields (Table 2, entry 2 or 9). The reaction of 2-amino benzophenone **4d** (R_1=C_6H_5, R_2=NO_2) or 2-amino benzaldehyde (R_1=H, R_2=H) **4g** with **3** gave lower total yields (Table 2, entry 4 or 7). The ratio of **5** and **6** was determined by ^{1}H NMR on the mixture of **5** and **6**. The oxadisilole-fused acridines **5a**–**i** was the major product.

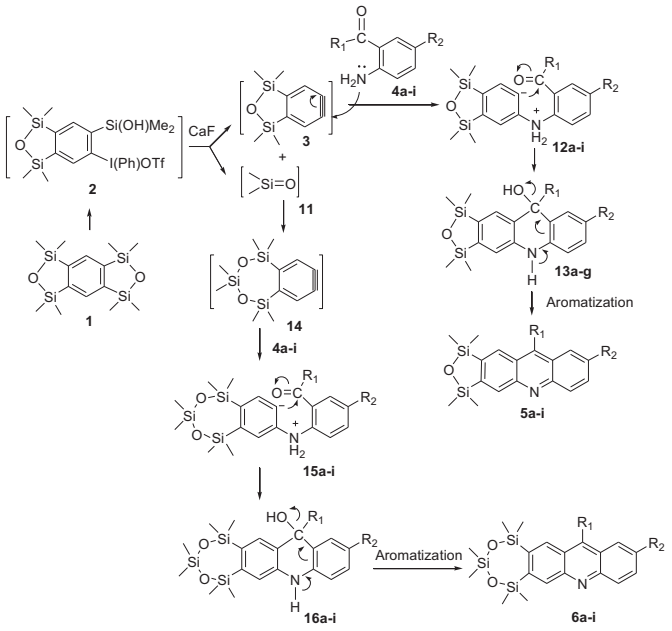
Table 2
Syntheses of **5a**–**i** and **6a**–**i** from **4a**–**i** with benzyne **3** generated from benzobis(oxadisilole) **1** via Scheme 1

Entry	Reactant	R_1	R_2	Products **5** and **6**	Yield[a,b] of **5**+**6** (**5**:**6**) %
1	**4a**	C_6H_5	CH_3	**5a**, **6a**	57 (34:23)
2	**4b**	C_6H_5	H	**5b**, **6b**	84 (44:40)
3	**4c**	C_6H_5	Cl	**5c**, **6c**	45 (28:17)
4	**4d**	C_6H_5	NO_2	**5d**, **6d**	35 (22:13)
5	**4e**	4-FC_6H_4	H	**5e**, **6e**	62 (44:18)
6	**4f**	2-FC_6H_4	Cl	**5f**, **6f**	55 (34:21)
7	**4g**	H	H	**5g**, **6g**	30 (17:13)
8	**4h**	H	Cl	**5h**, **6h**	56 (43:13)
9	**4i**	CH_3	H	**5i**, **6i**	89 (60:29)

[a] Isolated yield.
[b] The ratio of **5** and **6** was determined by ^{1}H NMR on the mixture of **5** and **6**.

During this study, we found that the mixture of oxadisilole-fused acridines **5a**–**i** and dioxatrisilole-fused acridines **6a**–**i** were synthesized via the two-step nucleophilic addition and

aromatization reaction of benzyne **3** with 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** at room temperature. One possible mechanistic pathway is shown in Scheme 3. When benzyne **3** is generated from benzobis(oxadisilole) **1** by a phenyliodination/fluoride induced desilylation protocol, dimethylsilanone **11** is possibly generated by decomposition involving the conversion of an initial reactive silicon-containing intermediate into a silanone. Subsequent insertion of dimethylsilanone **11** into the Si–O single bond of oxadisilole-fused benzyne **3** leads to dioxatrisilole-fused benzyne **14**,[24] the dioxatrisilole-fused acridines **6a**–**i** were formed via the nucleophilic addition and aromatization reaction of benzyne **14** with 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** at room temperature.



Scheme 3. Possible mechanism for the formation of oxadisilole-fused acridines **5a**–**i** and dioxatrisilole-fused acridines **6a**–**i**.

Our attention was then turned to the nucleophilic additions and aromatization reactions of naphthyne **9** generated in situ from the 2,3-naphthoxadisilole **7** with 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**g**. To our delight, 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**g** successfully participated in the nucleophilic additions and aromatization reactions with naphthyne **9** at room temperature to afford the corresponding benzo[*b*]acridine derivatives **10a**–**g** in 47–75% total yields (Scheme 2 and Table 3, entries 1–7). As shown from the yields, the reaction of 2-amino benzaldehyde **4g** with naphthyne **9** gave lower yields of **10g** (Scheme 2, Table 3, entry 7). We have tried the reaction of naphthyne **9** with **4i** (R_1=CH_3, R_2=H), but no product was obtained in this process.

Table 3
Syntheses of **10a**–**g** from **4a**–**g** with naphthyne **9** generated from 2,3-naphthoxadisilole **7** via Scheme 2

Entry	Reactant	R_1	R_2	Product	Yeild[a] of **10a**–**g** %
1	**4a**	C_6H_5	CH_3	**10a**	47
2	**4b**	C_6H_5	H	**10b**	68
3	**4c**	C_6H_5	Cl	**10c**	59
4	**4d**	C_6H_5	NO_2	**10d**	53
5	**4e**	4-FC_6H_4	H	**10e**	75
6	**4f**	2-FC_6H_4	Cl	**10f**	62
7	**4g**	H	H	**10g**	42

[a] Isolated yield.

The structures of compounds **5a**–**i**, **6a**–**i** and **10a**–**g** were established by ^1H and ^{13}C NMR spectroscopies, IR spectroscopy, MS and high-resolution (HR) MS, which are in good agreement with the expected structures.

The absorption spectra of **5a**–**f**, **6a**–**f** and **10a**–**f** were measured in CH_2Cl_2 and the spectra data are summarized in Table 4. The absorption spectra of **5a**–**f**, **6a**–**f** and **10a**–**f** show the characteristic strong β-band absorption at approximately 268 nm for **5a**–**f**, 270 nm for **6a**–**f** and 285 nm for **10a**–**f**, with the long wavelength absorption bands located between 368 and 380 nm for **5a**–**f**, 368 and 376 nm for **6a**–**f**, and 399 and 402 nm for **10a**–**f** (Figs. 1–3). A slight red shift is observed for the absorption maximum of **10a**–**f** compared with **5a**–**f** or **6a**–**f** due to extended conjugation in the latter compounds **10a**–**f**. A substituent effect is also observed in the **5a**–**f** and **6a**–**f** series, with electron-donating groups (R_2=CH_3) in **5a** or **6a** and electron-withdrawing groups (R_2=Cl or NO_2) in **5c,d,f** or **6c,d,f** producing a slight red shift in the absorption with respect to electronically neutral groups (R_2=H) in **5b,e** or **6b,e**. In addition, **10a**–**f** series show similar wavelength in the absorption.

Table 4
Summary of optical measurements and thermal properties of **5a**–**f**, **6a**–**f** and **10a**–**f**

Compound	λ^{abs}_{max}[a,b] [nm] (ε_{max} $10^4/M^{-1}$ cm^{-1})	λ^{em}_{max}[a,c] [nm]	Φ_{FL}[a,d]	T_m [°C]	T_{dec}[e] [°C]
5a	370(3.48)	504	0.27	127	320
5b	368(1.08)	502	0.47	100	292
5c	376(0.78)	505	0.34	169	329
5d	376(0.51)	521	0.30	201	346
5e	368(1.56)	503	0.39	148	240
5f	380(0.72)	498	0.56	153	324
6a	371(1.11)	502	0.16	169	317
6b	368(2.02)	501	0.42	174	326
6c	373(1.35)	504	0.29	180	353
6d	376(0.79)	520	0.44	172	326
6e	368(1.25)	500	0.30	153	309
6f	376(1.40)	501	0.46	170	341
10a	400(0.78)	521	0.47	145	270
10b	399(1.13)	525	0.40	221	322
10c	402(1.69)	527	0.38	189	335
10d	400(0.27)	602	0.56	262	292
10e	400(1.29)	524	0.48	191	384
10f	402(2.88)	526	0.46	243	324

[a] Measured in CH_2Cl_2.
[b] Excited at the absorption maximum.
[c] Excited at the emission maximum.
[d] Using quinine in 0.1 N H_2SO_4 (Φ_{365}=0.54) as a standard for the oxadisilole-fused acridines **5a**–**f**, dioxatrisilole-fused acridines **6a**–**f** or the benzo[*b*]acridines **10a**–**f**.
[e] Determined by thermal gravimetric analyzer with a heating rate of 10 °C min^{-1} under N_2.

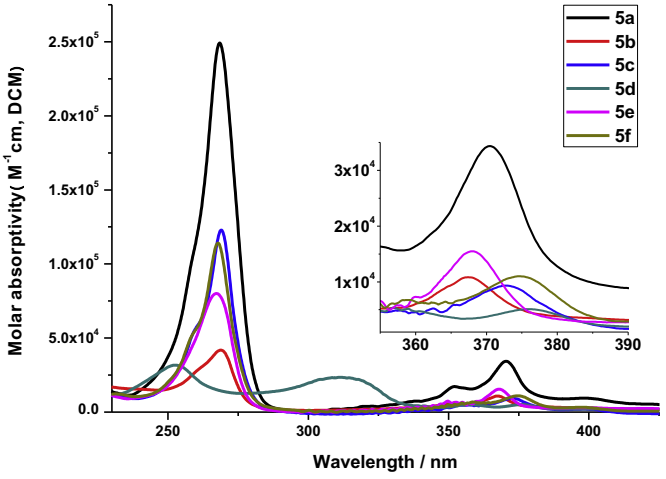


Fig. 1. Absorption spectra of **5a**–**f** in CH_2Cl_2.

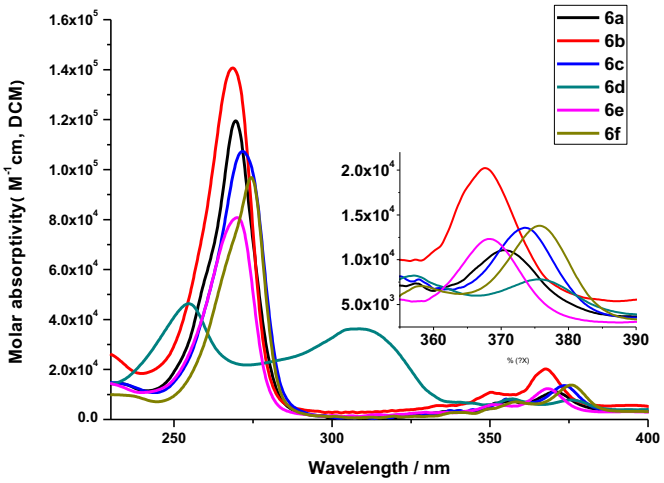


Fig. 2. Absorption spectra of **6a**–**f** in CH_2Cl_2.

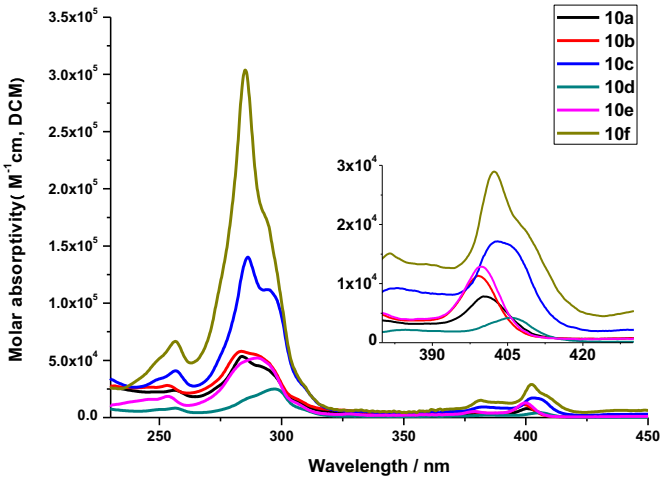


Fig. 3. Absorption spectra of **10a**–**f** in CH_2Cl_2.

The photoluminescence spectra of **5a**–**f**, **5a**–**f** and **10a**–**f** in CH_2Cl_2 show strong deep-blue or green emitters with maxima in the range of 498–521 nm for series **5a**–**f**, 500–520 nm for series **6a**–**f** and 521–602 nm for series **10a**–**f** (Figs. 4–6, Table 4). A substituent effect was also found for the maximum emission in **5a**–**f**, **6a**–**f** and **10a**–**f**, that is, by using the electron-withdrawing groups substituents in **5d**, **6d** or **10d** (R_2=NO_2), gives a red shift of 19 nm (**5d**) compared to **5b** (R_2=H), 19 nm (**6d**) compared to **6b** (R_2=H), and 77 nm (**10d**) compared to **10b** (R_2=H). The weaker electron-donating (R_2=CH_3) or electron-withdrawing substituents (R_2=Cl) in **5a**,**c**,**f**, **6a**,**c**,**f** or **10a**,**c**,**f** compared to electronically neutral substituents (R_2=H) in **5b**, **6b** or **10b**, show similar emission wavelength. Compounds with substituent effect in the **5a**–**f**, **6a**–**f** and **10a**–**f** series in emission spectra, the nitro group showed an obvious red shift in emission spectra. However, other substituent groups exhibited similar results in wavelength photoluminescence. The fluorescence quantum yields (Φ) measured in CH_2Cl_2 by using quinine as reference for **5a**–**f**, **6a**–**f** and **10a**–**f** are in the range of 27–56% for **5a**–**f**, 16–46% for **6a**–**f** and 38–56% for **10a**–**f**, respectively. Thus, molecules **5a**–**f**, **6a**–**f** and **10a**–**f** show potential applications as strong deep-blue or green emitters for organic light-emitting diode (OLED). In addition, the thermal properties of these three series compounds were determined by thermal gravimetric analysis (TGA). In general, they show good thermal stability, ranging from 240 to 384 °C. TGA results were determined by tangent of the curve to the baseline at the onset of 5% weight loss.

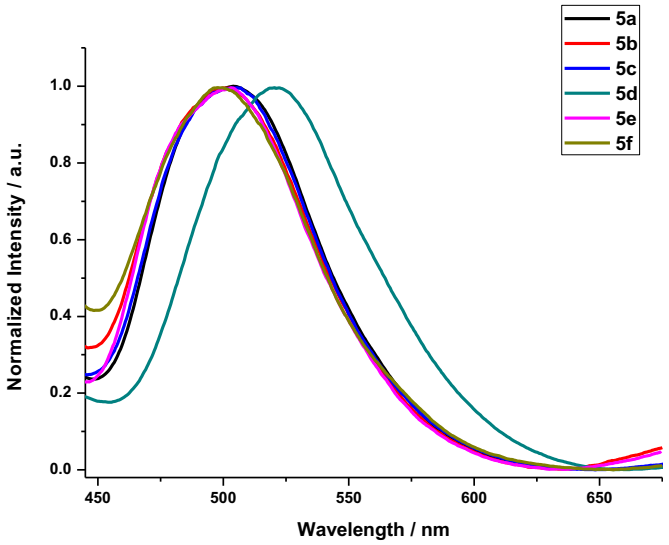


Fig. 4. Emission spectra of **5a**–**f** in CH_2Cl_2.

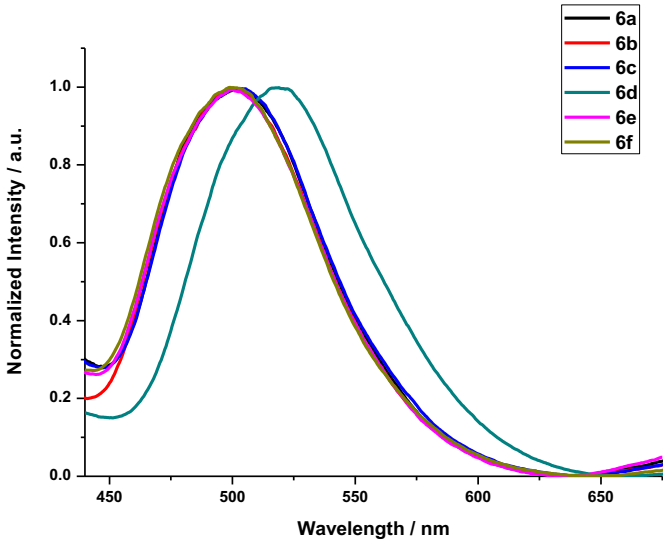


Fig. 5. Emission spectra of **6a**–**f** in CH_2Cl_2.

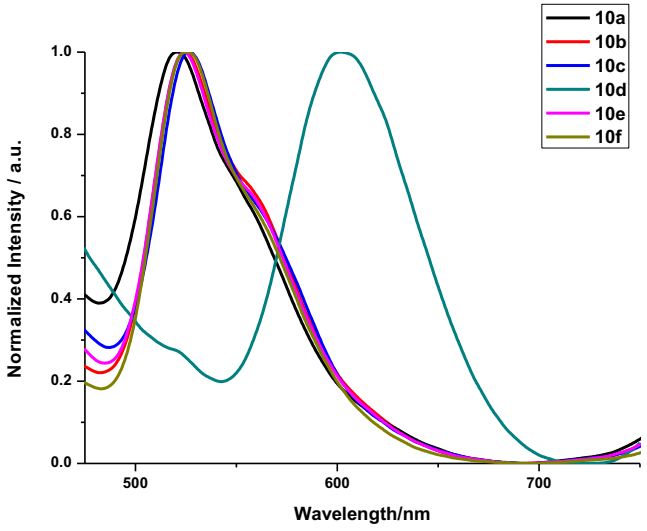


Fig. 6. Emission spectra of **10a**–**f** in CH_2Cl_2.

The redox properties as well as the HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital) energy levels of **5a**–**f**, **6a**–**f** and **10a**–**f** are tabulated in Table 5. The redox properties of the **5a**–**f**, **6a**–**f** and **10a**–**f** series were studied by cyclic voltammetry, which was carried out in a three-electrode cell setup with 0.1 M of Bu_4NPF_6 as a supporting electrolyte in CH_3CN. All of the potentials reported are referenced to Fc/Fc^+ as standard. In contrast to the optical properties, the oxidation potential (*E*ox) lies in the range of 0.97–1.44 V for **5a**–**f**, 1.03–1.23 V for **6a**–**f** and 0.97–1.40 V for **10a**–**f**. The HOMO energy values were calculated by the equation of Janietz or Schmidt,[25] and the LUMO energy levels were obtained by subtraction of the optical band gap from the HOMO. Molecules within the same series, that is, **5a**–**f** or **6a**–**f**, show similar HOMO energies. In addition, the band gaps within the same series are also very similar. Tendencies are apparent that the band gap values of compounds **10a**–**f** are smaller than compounds **5a**–**f** and **6a**–**f**. This is mainly contributed to their enhanced conjugation, which lower the energy levels between HOMO and LUMO.

Table 5
Summary of oxidation potential, HOMO and LUMO energy levels of **5a**–**f**, **6a**–**f** and **10a**–**f**

Compound	E_{ox}[a], V	HOMO[b], eV	Band gap[c], eV	LUMO[d], eV
5a	1.44	−6.17	3.50	−2.67
5b	1.33	−6.06	3.49	−2.57
5c	1.03	−5.76	3.49	−2.27
5d	1.02	−5.75	3.54	−2.21
5e	1.09	−5.82	3.52	−2.30
5f	0.97	−5.70	3.44	−2.26
6a	1.22	−5.95	3.49	−2.46
6b	1.03	−5.76	3.35	−2.41
6c	1.21	−5.94	3.44	−2.50
6d	1.06	−5.79	3.54	−2.25
6e	1.10	−5.83	3.35	−2.48
6f	1.23	−5.96	3.44	−2.47
10a	1.21	−5.94	2.95	−2.99
10b	1.32	−6.05	3.17	−2.88
10c	1.40	−6.13	2.76	−3.37
10d	0.97	−5.70	3.49	−2.21
10e	1.33	−6.06	2.88	−3.18
10f	1.00	−5.73	2.76	−2.97

[a] *E*ox was estimated by CV in CH_3CN with a platinum disk electrode as the working electrode, a platinum wire as the counter electrode and SCE as the reference electrode with an agar salt bridge connecting the compound solution and ferrocene as an external standard, and 0.1 M Bu_4NPF_6 at a scan rate of 100 mV/s, $E_{1/2}(Fc/Fc^+)$=0.49 V versus SCE, calculated with ferrocene (4.8 eV vs vacuum).
[b] HOMO=*E*ox−[$E_{1/2}(Fc/Fc^+)$]+4.8.
[c] Estimated from the absorption edge in CH_2Cl_2.
[d] LUMO=HOMO+Optical band gap.

3. Conclusions

Three unknown series of oxadisilole-fused acridines **5a**–**i**, dioxatrisilole-fused acridines **6a**–**i** and benzo[*b*]acridines **10a**–**g** have been synthesized, respectively, by the nuclephilic additions and aromatization reactions of 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** with arynes **3** or **9** generated in situ from benzobis(oxadisilole) **1** or 2,3-naphthoxadisilole **7** in good yields at room temperature. This simple and efficient synthetic method could offer great opportunities for the syntheses of important heterocyclic compounds. The photophysical, redox and thermal properties of oxadisilole-fused acridines **5a**–**f**, dioxatrisilole-fused acridines **6a**–**f** and benzo[*b*]acridines **10a**–**f** have been characterized. These compounds show potential applications as strong deep-blue or green emitters for OLED, because of their high fluorescence quantum yields and thermal stabilities.

4. Experimental section

4.1. General methods

Purification was effected by silica gel column chromatography (200–300 mesh silica gel) using mixtures of reagent-grade EtOAc/petroleum ether (PE, 60–80 °C) as eluents. NMR spectra were recorded with a Bruker DRX-500 NMR spectrometer at 500 MHz for ^1H and at 125 MHz for ^{13}C with $CDCl_3$ as solvent. Chemical shifts are reported in parts per million on the δ scale relative to the residual resonance of $CHCl_3$ (δ=7.26 ppm for ^1H and 77.00 ppm for the central peak of the triplet in ^{13}C). Coupling constants (*J*) are reported in Hertz. IR spectra were recorded with an FTIR spectrometer in KBr discs. Low-resolution mass spectra were recorded with an Agilent spectrometer in EI or API-ES mode are reported as *m*/*z* values. High-resolution mass spectra were recorded with a Waters Micromass GCT instrument. The fluorescence quantum yields in solution were determined by the dilution method using quinine in 1.0 N H_2SO_4 (Φ_{365}=0.54) as standard with a LS-55 spectrometer. Thermal stabilities were determined by thermal gravimetric analyzer with heating rate of 10 °C min^{-1} under N_2.

*4.1.1. General procedure for the preparation of oxadisilole fused acridines **5a**–**i**, dioxatrisilole-fused acridines **6a**–**i** or benzo[b]acridine derivatives **10a**–**g**.* TfOH (0.14 mL, 1.50 mmol) was added by means of a syringe to a stirred solution of $PhI(OAc)_2$, (247 mg, 0.75 mmol) in CH_2Cl_2 (10 mL) at 0 °C under N_2. The mixture was stirred under N_2 at 0 °C for 1 h and at room temperature for 2 h. The clear yellow solution was cooled again to 0 °C, followed by dropwise addition into the cold solution of the benzobis(oxadisilole) **1** (or 2,3-naphthoxadisilole **7**) (0.5 mmol) in CH_2Cl_2 (5 mL) at 0 °C. The mixture was stirred at 0 °C for 0.5 h and at room temperature for 3 h. The clear yellow solution was washed with water (200 mL) and was extracted by CH_2Cl_2 three times. Then the low yellow solid was obtained under reduced pressure and resolved in CH_3CN (5 mL). The solution of 2-aminoaryl ketones or 2-aminoaryl aldehydes **4a**–**i** (0.2 mmol) and CsF (76 mg, 0.5 mmol) in CH_3CN (5 mL) was dropwise added into low yellow solution of **2** (or **8**), The mixture stirred under N_2 at room temperature until **4a**–**i** was disappeared (monitored by TLC). The crude product was purified by column chromatography on silica gel using a gradient of 4–5% EtOAc in PE (60–80 °C) as the eluent to afford products **5a**–**i**, **6a**–**i** and **10a**–**g**.

*4.1.1.1. 2,3-Dioxatrisilole-7-methyl-9-phenyl-acridine (**5a**).* Yield: 27.2 mg, 34%; yellow powder; mp 126–128 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 0.35 (s, 6H, $SiMe_2$), 0.47 (s, 6H, $SiMe_2$), 2.45 (s, 3H, CH_3), 7.40 (s, 1H, Ar–*H*), 7.43–7.45 (m, 2H, Ar–*H*), 7.62–7.66 (m, 4H, Ar–*H*), 7.86 (s, 1H, Ar–*H*), 8.23 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.52 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 1.0, 1.2, 22.0, 124.8, 125.3, 125.5, 128.3, 128.5, 129.2, 129.9, 130.4, 132.7, 133.2, 135.6, 136.0, 143.2, 146.5, 147.7, 147.8, 148.0 ppm; IR (KBr, cm^{-1}) 3440, 2954, 1631, 1252; MS (EI): *m*/*z* (%)=400 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 400.1547. $C_{24}H_{25}NOSi_2$ requires 400.1552.

*4.1.1.2. 2,3-Oxadisilole-9-phenyl-acridine (**5b**).* Yield: 33.9 mg, 44%; yellow powder; mp 99–101 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 0.37 (s, 6H, $SiMe_2$), 0.49 (s, 6H, $SiMe_2$), 7.39–7.44 (m, 1H, Ar–*H*), 7.45–7.47 (m, 2H, Ar–*H*), 7.60–7.64 (m, 3H, Ar–*H*), 7.68 (d, *J*=8.5 Hz, 1H, Ar–*H*), 7.74–7.78 (m, 1H, Ar–*H*), 7.92 (s, 1H, Ar–*H*), 8.28 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.53 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 1.0, 1.2, 125.1, 125.5, 125.7, 126.9, 128.3, 128.4, 129.6, 130.0, 130.1, 130.5, 132.9, 135.9, 143.3, 147.5, 148.2, 148.5, 149.1 ppm; IR (KBr, cm^{-1}) 3433, 3316, 1628, 1248; MS (EI): *m*/*z* (%)= 385 (2, $[M+H]^+$), 196 (100); HRMS (EI): $[M+H]^+$, found: 385.1318. $C_{23}H_{23}NOSi_2$ requires 385.1312.

*4.1.1.3. 2,3-Oxadisilole-7-chloro-9-phenyl-acridine (**5c**).* Yield: 23.5 mg, 28%; light yellow powder; mp 168–170 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.36 (s, 6H, $SiMe_2$), 0.49 (s, 6H, $SiMe_2$), 7.43 (d, J=2.0 Hz, 1H, Ar–*H*), 7.44 (d, J=2.0 Hz, 1H, Ar–*H*), 7.62–7.66 (m, 5H, Ar–*H*), 7.90 (s, 1H, Ar–*H*), 8.21 (d, J=8.5 Hz, 1H, Ar–*H*), 8.51 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.9, 1.1, 125.1, 125.3, 125.7, 128.6, 129.9, 130.3, 131.29, 131.33, 131.6, 132.9, 135.2, 144.3, 146.6, 147.3, 148.5, 148.7 ppm; IR (KBr, cm^{-1}) 3442, 2955, 1613, 1251; MS (EI): m/z (%)=419 (63, $[M+H]^+$), 404 (100); HRMS (EI): $[M+H]^+$, found: 419.0928. $C_{23}H_{22}ClNOSi_2$ requires 419.0927.

*4.1.1.4. 2,3-Oxadisilole-7-nitro-9-phenyl-acridine (**5d**).* Yield: 18.9 mg, 22%; luminous yellow powder; mp 200–201 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.37 (s, 6H, $SiMe_2$), 0.48 (s, 6H, $SiMe_2$), 7.46–7.48 (m, 2H, Ar–*H*), 7.68–7.69 (m, 3H, Ar–*H*), 7.94 (s, 1H, Ar–*H*), 8.34 (d, J=9.5 Hz, 1H, Ar–*H*), 8.46 (dd, J=9.5, 2.5 Hz, 1H, Ar–*H*), 8.51 (s, 1H, Ar–*H*), 8.68 (d, J=2.5 Hz, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.9, 1.1, 122.9, 123.7, 125.0, 125.5, 128.9, 129.3, 130.2, 130.4, 131.7, 133.0, 134.3, 144.8, 145.4, 149.6, 150.3, 151.3, 151.4 ppm; IR (KBr, cm^{-1}) 3440, 2958, 1624, 1339; MS (EI): m/z (%)=430 (55, $[M+H]^+$), 415 (100); HRMS (EI): $[M+H]^+$, found: 430.1169. $C_{23}H_{22}N_2O_3Si_2$ requires 430.1173.

*4.1.1.5. 2,3-Oxadisilole-9-(4-fluoro phenyl)-acridine (**5e**).* Yield: 35.5 mg, 44%; dark yellow powder; mp 147–149 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.37 (s, 6H, $SiMe_2$), 0.48 (s, 6H, $SiMe_2$), 7.32–7.35 (m, 2H, Ar–*H*), 7.42–7.45 (m, 3H, Ar–*H*), 7.65 (d, J=8.5 Hz, 1H, Ar–*H*), 7.75–7.79 (m, 1H, Ar–H), 7.87 (s, 1H, Ar–*H*), 8.27 (d, J=8.5 Hz, 1H, Ar–*H*), 8.52 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 1.0, 1.2, 115.7 (d, J=21.3 Hz), 125.2, 125.6, 125.9, 126.6, 129.7 (d, J=8.8 Hz), 130.2, 131.7 (d, J=3.8 Hz), 132.2 (d, J=7.5 Hz), 132.9, 136.1, 143.7, 146.3, 148.3, 148.4, 149.1, 161.8, 163.8 ppm; ^{19}F NMR (470 MHz, $CDCl_3$): δ −113.1 (s, Ar–*F*) ppm; IR (KBr, cm^{-1}) 3437, 2959, 1626, 1246; MS (EI): m/z (%)=403 (2, $[M+H]^+$), 214 (100); HRMS (EI): $[M+H]^+$, found: 403.1224. $C_{23}H_{22}FNOSi_2$ requires 403.1229.

*4.1.1.6. 2,3-Oxadisilole-7-chloro-9-(2-fluoro phenyl)-acridine (**5f**).* Yield: 29.7 mg, 34%; yellow powder; mp 152–154 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.36 (s, 3H, $SiMe_2$), 0.38 (s, 3H, $SiMe_2$), 0.47 (s, 3H, $SiMe_2$), 0.48 (s, 3H, $SiMe_2$), 7.37–7.41 (m, 2H, Ar–*H*), 7.42–7.45 (m, 1H, Ar–*H*), 7.59 (s, 1H, Ar–*H*), 7.62–7.67 (m, 1H, Ar–*H*), 7.70 (dd, J=9.0, 2.5 Hz, 1H, Ar–*H*), 7.82 (s, 1H, Ar–*H*), 8.24 (d, J=9.0 Hz, 1H, Ar–*H*), 8.50 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.9, 1.0, 1.1, 1.2, 116.3 (d, J=21.3 Hz), 122.6 (d, J=17.5 Hz), 124.5 (d, J=3.8 Hz), 124.6, 125.4, 125.9, 129.3, 131.2 (d, J=7.5 Hz), 131.5 (d, J=5.0 Hz), 132.1, 132.4 (d, J=2.5 Hz), 133.0, 140.4, 144.9, 147.3, 148.5, 148.9, 159.1, 161.0 ppm; ^{19}F NMR (470 MHz, $CDCl_3$): δ −112.7 (m, Ar–*F*) ppm; IR (KBr, cm^{-1}) 3442, 2957, 1613, 1251; MS (EI): m/z (%)=437 (67, $[M+H]^+$), 57 (100); HRMS (EI): $[M+H]^+$, found: 437.0834. $C_{23}H_{21}ClFNOSi_2$ requires 437.0837.

*4.1.1.7. 2,3-Oxadisilole-acridine (**5g**).* Yield: 10.5 mg, 17%; yellow powder; mp 79–81 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.47 (s, 6H, $SiMe_2$), 0.48 (s, 6H, $SiMe_2$), 7.56 (t, J=8.0 Hz, 1H, Ar–*H*), 7.79–7.82 (m, 1H, Ar–*H*), 8.03 (d, J=8.0 Hz, 1H, Ar–*H*), 8.25 (s, 1H, Ar–H), 8.28 (d, J=8.0 Hz, 1H, Ar–*H*), 8.49 (s, 1H, Ar–*H*), 8.82 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 1.0, 1.2, 125.9, 126.6, 126.9, 128.3, 129.1, 130.8, 131.7, 132.1, 136.9, 143.4, 148.4, 149.1, 149.3 ppm; IR (KBr, cm^{-1}) 3048, 2957, 1667, 1252; MS (EI): m/z (%)=310 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 310.1078. $C_{17}H_{19}NOSi_2$ requires 310.1076.

*4.1.1.8. 2,3-Oxadisilole-7-chloro-acridine (**5h**).* Yield: 29.5 mg, 43%; dark yellow solid; mp 96–98 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.47 (s, 12H, $SiMe_2$), 7.71 (dd, J=9.0, 2.5 Hz, 1H, Ar–*H*), 8.00 (d, J=2.5 Hz, 1H, Ar–*H*), 8.18 (d, J=9.0 Hz, 1H, Ar–*H*), 8.22 (s, 1H, Ar–*H*), 8.43 (s, 1H, Ar–H), 8.70 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 1.0, 1.2, 126.4, 126.9, 127.0, 131.2, 131.57, 131.60, 131.7, 132.6, 135.4, 144.2, 147.7, 148.9, 149.4 ppm; IR (KBr, cm^{-1}) 3448, 2922, 1614, 1250; MS (EI): m/z (%)=344 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 344.0688. $C_{17}H_{18}ClNOSi_2$ requires 344.0695.

*4.1.1.9. 2,3-Oxadisilole-9-methyl-acridine (**5i**).* Yield: 38.8 mg, 60%; yellow solid; mp 182–184 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.47 (s, 6H, $SiMe_2$), 0.49 (s, 6H, $SiMe_2$), 3.16 (s, 3H, CH_3), 7.52–7.56 (m, 1H, Ar–*H*), 7.74–7.77 (m, 1H, Ar–*H*), 8.23–8.25 (m, 2H, Ar–*H*), 8.468 (s, 1H, Ar–H), 8.473 (s, 1H, Ar–*H*), ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 1.0, 1.3, 13.6, 124.6, 125.5, 125.6, 125.8, 127.7, 130.0, 130.1, 133.2, 143.0, 143.1, 147.8, 148.2, 148.4 ppm; IR (KBr, cm^{-1}) 3422, 2918, 1624, 1252; MS (EI): m/z (%)=323 (67, $[M+H]^+$), 100 (100); HRMS (EI): $[M+H]^+$, found: 323.1162. $C_{18}H_{21}NOSi_2$ requires 323.1165.

*4.1.1.10. 2,3-Dioxatrisilole-7-methyl-9-phenyl-acridine (**6a**).* Yield: 21.8 mg, 23%; yellow powder; mp 168–170 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.13 (s, 6H, $SiMe_2$), 0.300 (s, 6H, $SiMe_2$), 0.55 (s, 6H, $SiMe_2$), 2.46 (s, 3H, CH_3), 7.43 (d, J=2.0 Hz, 1H, Ar–*H*), 7.45 (d, J=2.0 Hz, 1H, Ar–*H*), 7.49 (s, 1H, Ar–H), 7.59–7.65 (m, 4H, Ar–*H*), 7.90 (s, 1H, Ar–*H*), 8.22 (d, J=9.0 Hz, 1H, Ar–*H*), 8.48 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 1.7, 2.1, 22.0, 124.2, 124.9, 125.6, 128.3, 129.3, 130.4, 133.2, 133.5, 135.6, 135.7, 141.3, 146.1, 146.2, 147.3, 148.2 ppm; IR (KBr, cm^{-1}) 3422, 29,601, 16,321, 1257; MS (EI): m/z (%)=474 (14, $[M+H]^+$), 57 (100); HRMS (EI): $[M+H]^+$, found: 474.1735. $C_{26}H_{31}NO_2Si_3$ requires 474.1736.

*4.1.1.11. 2,3-Dioxatrisilole-9-phenyl-acridine (**6b**).* Yield: 36.7 mg, 40%; pale yellow lump crystal; mp 173–174 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.14 (s, 6H, $SiMe_2$), 0.32 (s, 6H, $SiMe_2$), 0.56 (s, 6H, $SiMe_2$), 7.42–7.47 (m, 3H, Ar–*H*), 7.58–7.63 (m, 3H, Ar–*H*), 7.76–7.79 (m, 2H, Ar–*H*), 7.95 (s, 1H, Ar–*H*), 8.28–8.30 (m, 1H, Ar–*H*), 8.49 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 1.7, 2.0, 124.0, 125.6, 125.7, 126.9, 128.3, 128.4, 129.6, 130.2, 130.4, 133.6, 135.4, 136.1, 141.4, 146.8, 147.2, 147.9, 149.3 ppm; IR (KBr, cm^{-1}) 3450, 2961, 1621, 1258; MS (EI): m/z (%)=459 (44, $[M+H]^+$), 444 (100); HRMS (EI): $[M+H]^+$, found: 459.1506. $C_{25}H_{29}NO_2Si_3$ requires 459.1503.

*4.1.1.12. 2,3-Dioxatrisilole-7-chloro-9-phenyl-acridine (**6c**).* Yield: 25.1 mg, 17%; pale yellow lump crystal; mp 179–180 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.14 (s, 6H, $SiMe_2$), 0.31 (s, 6H, $SiMe_2$), 0.56 (s, 6H, $SiMe_2$), 7.44 (d, J=6.5 Hz, 2H, Ar–*H*), 7.60–7.63 (m, 3H, Ar–*H*), 7.68 (dd, J=9.0, 2.0 Hz, 1H, Ar–*H*), 7.73 (d, J=2.0 Hz, 1H, Ar–*H*), 7.92 (s, 1H, Ar–*H*), 8.22 (d, J=9.0 Hz, 1H, Ar–*H*), 8.46 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 1.7, 2.0, 124.2, 125.2, 125.9, 128.5, 128.7, 130.4, 131.4, 131.7, 133.4, 134.8, 136.1, 142.4, 146.3, 147.3, 147.5, 147.9 ppm; IR (KBr, cm^{-1}) 3442, 2960, 1614, 1257; MS (EI): m/z (%)=493 (42, $[M+H]^+$), 478 (100); HRMS (EI): $[M+H]^+$, found: 493.1116. $C_{25}H_{28}ClNO_2Si_3$ requires 493.1118.

*4.1.1.13. 2,3-Dioxatrisilole-7-nitro-9-phenyl acridine (**6d**).* Yield: 13.1 mg, 13%; deep yellow solid; mp 171–172 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 0.14 (s, 6H, $SiMe_2$), 0.33 (s, 6H, $SiMe_2$), 0.55 (s, 6H, $SiMe_2$), 7.48–7.49 (m, 2H, Ar–*H*), 7.68–7.69 (m, 3H, Ar–*H*), 7.99 (s, 1H, Ar–*H*), 8.38 (d, J=9.5 Hz, 1H, Ar–*H*), 8.48 (s, 1H, Ar–*H*), 8.50 (s, 1H, Ar–*H*), 8.78 (d, J=2.5 Hz, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 1.6, 1.9, 123.1, 123.8, 124.5, 124.9, 128.8, 129.5, 130.4, 131.5, 133.6, 133.8, 136.0, 143.6, 144.9, 149.7, 149.9, 150.1, 151.1 ppm; IR (KBr, cm^{-1}) 3442, 2959, 1623, 1258; MS (EI): m/z (%)=504 (41, $[M+H]^+$), 489 (100); HRMS (EI): $[M+H]^+$, found: 504.1357. $C_{25}H_{28}N_2O_4Si_3$ requires 504.1359.

*4.1.1.14. 2,3-Dioxatrisilole-9-(4-fluoro phenyl)-acridine (**6e**).* Yield: 17.2 mg, 18%; yellow powder; mp 151–153 °C; ^{1}H NMR (500 MHz,

$CDCl_3$): δ 0.14 (s, 6H, $SiMe_2$), 0.33 (s, 6H, $SiMe_2$), 0.55 (s, 6H, $SiMe_2$), 7.31–7.35 (m, 2H, Ar–*H*), 7.42–7.47 (m, 3H, Ar–*H*), 7.73 (d, *J*=8.5 Hz, 1H, Ar–*H*), 7.77–7.80 (m, 1H, Ar–*H*), 7.91 (s, 1H, Ar–*H*), 8.29 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.48 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 1.7, 2.0, 115.5 (d, *J*=21.3 Hz), 124.1, 125.7, 126.0, 126.6, 129.7, 130.3, 131.3 (d, *J*=3.8 Hz), 132.2 (d, *J*=8.8 Hz), 133.2, 136.1, 141.8, 146.0, 147.0, 147.8, 149.3, 161.8, 163.8 ppm; ^{19}F NMR (470 MHz, $CDCl_3$): δ −112.9 (s, Ar–*F*) ppm; IR (KBr, cm^{-1}) 3438, 2958, 1621, 1258; MS (EI): *m*/*z* (%)=477 (31, $[M+H]^+$), 462 (100); HRMS (EI): $[M+H]^+$, found: 477.1412. $C_{25}H_{28}FNO_2Si_3$ requires 477.1414.

*4.1.1.15. 2,3-Dioxatrisilole-7-chloro-9-(2-fluoro phenyl)-acridine (**6f**).* Yield: 21.5 mg, 21%; pale yellow powder; mp 169–170 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 0.13 (s, 3H, $SiMe_2$), 0.14 (s, 3H, $SiMe_2$), 0.31 (s, 3H, $SiMe_2$), 0.33 (s, 3H, $SiMe_2$), 0.548 (s, 3H, $SiMe_2$), 0.551 (s, 3H, $SiMe_2$), 7.36–7.40 (m, 2H, Ar–*H*), 7.41–7.44 (m, 1H, Ar–*H*), 7.61–7.65 (m, 1H, Ar–*H*), 7.66 (s, 1H, Ar–*H*), 7.71 (dd, *J*=9.0, 2.5 Hz, 1H, Ar–*H*), 7.83 (s, 1H, Ar–*H*), 8.24 (d, *J*=9.0 Hz, 1H, Ar–*H*), 8.46 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.69, 0,71, 1.6, 1.7, 2.0, 116.2 (d, *J*=21.3 Hz), 122.3 (d, *J*=16.3 Hz), 124.3, 124.4 (d, *J*=3.8 Hz), 124.7, 126.1, 128.8, 131.3 (d, *J*=8.8 Hz), 131.6 (d, *J*=3.8 Hz), 132.2, 132.4 (d, *J*=2.5 Hz), 132.8, 136.2, 140.0, 143.0, 147.5, 147.9, 159.0, 161.0 ppm; ^{19}F NMR (470 MHz, $CDCl_3$): δ −112.5 (d, *J*=6.5 Hz, Ar–*F*) ppm; IR (KBr, cm^{-1}) 3446, 2960, 1613, 1256; MS (EI): *m*/*z* (%)=511 (73, $[M+H]^+$), 496 (100); HRMS (EI): $[M+H]^+$, found: 511.1022. $C_{25}H_{27}ClFNO_2Si_3$ requires 511.1020.

*4.1.1.16. 2,3-Dioxatrisilole-acridine (**6g**).* Yield: 10.0 mg, 13%; yellow solid; mp 55–56 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 0.16 (s, 6H, $SiMe_2$), 0.54 (s, 6H, $SiMe_2$), 0.55 (s, 6H, $SiMe_2$), 7.54–7.57 (m, 1H, Ar–*H*), 7.79–7.83 (m, 1H, Ar–*H*), 8.01 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.21 (s, 1H, Ar–*H*), 8.28 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.49 (s, 1H, Ar–*H*), 8.78 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 2.0, 2.03, 125.6, 126.0, 127.1, 128.4, 128.8, 129.0, 130.9, 132.3, 135.0, 142.0, 147.7, 148.0, 149.1 ppm; IR (KBr, cm^{-1}) 3441, 2959, 1623, 1257; MS (EI): *m*/*z* (%)=384 (16, $[M+H]^+$), 321 (100); HRMS (EI): $[M+H]^+$, found: 384.1266. $C_{19}H_{25}NO_2Si_3$ requires 384.1274.

*4.1.1.17. 2,3-Dioxatrisilole-7-chloro-acridine (**6h**).* Yield: 19.2 mg, 13%; dark yellow solid; mp 112–113 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 0.15 (s, 6H, $SiMe_2$), 0.536 (s, 6H, $SiMe_2$), 0.542 (s, 6H, $SiMe_2$),7.71 (dd, *J*=9.0, 2.5 Hz, 1H, Ar–*H*), 7.99 (d, *J*=2.5 Hz, 1H, Ar–*H*), 8.18–8.19 (m, 2H, Ar–*H*), 8.42 (s, 1H, Ar–*H*), 8.66 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.7, 2.00, 2.03, 125.9, 126.4, 127.3, 131.2, 131.7, 131.8, 134.8, 135.0, 135.9, 142.7, 147.8, 147.9, 148.3 ppm; IR (KBr, cm^{-1}) 2957, 2851, 1609, 1255; MS (EI): *m*/*z* (%)=418 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 418.0876. $C_{19}H_{24}ClNO_2Si_3$ requires 418.0889.

*4.1.1.18. 2,3-Dioxatrisilole-9-methyl-acridine (**6i**).* Yield: 31.1 mg, 29%; yellow powder; mp 184–186 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 0.16 (s, 6H, $SiMe_2$), 0.54 (s, 6H, $SiMe_2$), 0.56 (s, 6H, $SiMe_2$), 3.14 (s, 3H, CH_3), 7.55–7.58 (m, 1H, Ar–*H*), 7.76–7.79 (m, 1H, Ar–*H*), 8.23 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.26 (dd, *J*=8.5, 0.5 Hz, 1H, Ar–*H*), 8.42 (d, *J*=0.5 Hz, 1H, Ar–*H*), 8.47 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 0.8, 2.06, 2.10, 13.3, 124.4, 124.6, 125.6, 126.2, 130.0, 130.2, 131.1, 136.8, 141.2, 142.3, 146.5, 147.5, 148.8 ppm; IR (KBr, cm^{-1}) 3423, 2955, 1623, 1251; MS (EI): *m*/*z* (%)=398 (11, $[M+H]^+$), 382 (100); HRMS (EI): $[M+H]^+$, found: 398.1422. $C_{20}H_{27}NO_2Si_3$ requires 398.1424.

*4.1.1.19. 2-Methyl-12-phenyl-benzo[b]acridine (**10a**).* Yield: 30.1 mg, 47%; red brown powder; mp 144–145 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 2.44 (s, 3H, CH_3), 7.35–7.37 (m, 1H, Ar–*H*), 7.41–7.45 (m, 2H, Ar–*H*), 7.51–7.52 (m, 2H, Ar–*H*), 7.57 (d, *J*=9.5 Hz, 1H, Ar–*H*), 7.66–7.67 (m, 3H, Ar–*H*), 7.82 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.07 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.18 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.27 (s, 1H, Ar–*H*), 8.93 (s, 1H, Ar–H) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 22.1, 124.4, 124.5, 125.5, 126.0, 126.3, 126.7, 128.2, 128.4, 128.5, 129.2, 130.6, 131.3, 133.6, 134.1, 135.0, 136.3, 144.6, 146.4, 149.0 ppm; IR (KBr, cm^{-1}) 3440, 3051, 1648, 1536; MS (EI): *m*/*z* (%)=319 (1, $[M+H]^+$), 57 (100); HRMS (EI): $[M+H]^+$, found: 320.1434. $C_{24}H_{17}N$ requires 320.1435.

*4.1.1.20. 12-Phenyl-benzo[b]acridine (**10b**).* Yield: 41.5 mg, 68%; yellow brown powder; mp 220–222 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 7.33–7.38 (m, 1H, Ar–*H*), 7.43–7.46 (m, 1H, Ar–*H*), 7.51–7.53 (m, 1H, Ar–*H*), 7.65 (s, 1H, Ar–*H*), 7.66 (s, 1H, Ar–*H*), 7.69–7.71 (m, 1H, Ar–*H*), 7.73–7.74 (m, 1H, Ar–*H*), 7.83 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.08 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.25 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.33 (s, 1H, Ar–*H*), 8.94 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 124.3, 124.5, 125.2, 125.5, 126.2, 126.4, 127.0, 128.2, 128.45, 128.51, 128.6, 129.7, 130.3, 130.6, 131.3, 134.3, 136.2, 145.3, 147.6, 150.1 ppm; IR (KBr, cm^{-1}) 3441, 3051, 1681, 1487; MS (EI): *m*/*z* (%)=305 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 305.1204. $C_{23}H_{15}N$ requires 305.1205.

*4.1.1.21. 2-Chloro-12-phenyl-benzo[b]acridine (**10c**).* Yield: 40.0 mg, 59%; deep red powder; mp 188–190 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 7.38–7.40 (m, 1H, Ar–*H*), 7.45–7.48 (m, 1H, Ar–*H*), 7.50 (d, *J*=2.0 Hz, 1H, Ar–*H*), 7.52 (d, *J*=2.0 Hz, 1H, Ar–*H*), 7.63 (d, *J*=8.5 Hz, 1H, Ar–*H*), 7.66–7.69 (m, 4H, Ar–*H*), 7.84 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.07 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.18 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.29 (s, 1H, Ar–*H*), 8.90 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 124.3, 124.5, 125.0, 125.9, 126.2, 126.7, 127.2, 128.3, 128.6, 128.75, 128.77, 130.5, 131.2, 131.5, 131.6, 131.7, 134.5, 135.6, 145.3, 146.8, 148.2 ppm; IR (KBr, cm^{-1}) 3443, 3046, 1681, 1529; MS (EI): *m*/*z* (%)=339 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 339.0815. $C_{23}H_{14}ClN$ requires 339.0816.

*4.1.1.22. 2-Nitro-12-phenyl benzo[b]acridine (**10d**).* Yield: 37.1 mg, 53%; bronzing powder; mp 261–263 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 7.42–7.45 (m, 1H, Ar–*H*), 7.50–7.53 (m, 1H, Ar–*H*), 7.56–7.58 (m, 2H, Ar–*H*), 7.73–7.74 (m, 3H, Ar–*H*), 7.87 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.07 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.27 (d, *J*=10.0 Hz, 1H, Ar–*H*), 8.36–8.38 (m, 2H, Ar–*H*), 8.71 (d, *J*=2.0 Hz, 1H, Ar–*H*), 8.90 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 122.2, 122.8, 124.2, 125.5, 126.4, 127.2, 127.5, 127.7, 128.2, 128.8, 128.9, 129.5, 130.6, 131.7, 132.0, 134.6, 135.6, 144.4, 146.5, 149.8, 152.4 ppm; IR (KBr, cm^{-1}) 3442, 3048, 1619, 1333; MS (EI): *m*/*z* (%)=350 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 350.1055. $C_{23}H_{14}N_2O_2$ requires 350.1058.

*4.1.1.23. 12-(4-Fluoro-phenyl)-benzo[b]acridine (**10e**).* Yield: 48.5 mg, 75%; reddish brown powder; mp 190–192 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 7.37–7.44 (m, 4H, Ar–*H*), 7.48–7.53 (m, 3H, Ar–*H*), 7.69 (d, *J*=8.5 Hz, 1H, Ar–*H*), 7.77–7.80 (m, 1H, Ar–*H*), 7.87 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.10 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.31 (s, 1H, Ar–*H*), 8.35 (d, *J*=8.5 Hz, 1H, Ar–*H*), 9.03 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 115.8 (d, *J*=21.3 Hz), 124.4, 124.6, 125.5, 125.8 (d, *J*=16.3 Hz), 126.6, 126.7, 127.0, 128.3, 128.6, 129.7, 130.4, 131.5, 132.0, 132.3 (d, *J*=8.8 Hz), 134.4, 145.2, 146.5, 150.0, 162.0, 163.9 ppm; ^{19}F NMR (470 MHz, $CDCl_3$): δ −113.1 (s, Ar–*F*) ppm; IR (KBr, cm^{-1}) 3446, 3046, 1601, 1510; MS (EI): *m*/*z* (%)=323 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 323.1110. $C_{23}H_{14}FN$ requires 323.1114.

*4.1.1.24. 2-Chloro-12-(2-fluoro-phenyl)-benzo[b]acridine (**10f**).* Yield: 44.3 mg, 62%; reddish brown powder; mp 242–243 °C; ^1H NMR (500 MHz, $CDCl_3$): δ 7.40–7.50 (m, 5H, Ar–*H*), 7.61 (s, 1H, Ar–*H*), 7.66–7.71 (m, 2H, Ar–*H*), 7.86 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.09 (d, *J*=8.5 Hz, 1H, Ar–*H*), 8.25–8.27 (m, 2H, Ar–*H*), 8.96 (s, 1H, Ar–*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 116.4 (d, *J*=21.3 Hz), 122.9 (d,

J=16.3 Hz), 124.2, 124.5, 124.6 (d, J=3.8 Hz), 124.9, 125.5, 126.2, 126.9, 127.2, 128.3, 128.5, 131.3 (d, J=8.8 Hz), 131.8, 132.0 (d, J=8.8 Hz), 132.5 (d, J=2.5 Hz), 134.7, 141.0, 144.7, 147.8, 159.1, 161.1 ppm; ^{19}F NMR (470 MHz, $CDCl_3$): δ −112.7 (m, Ar−*F*) ppm; IR (KBr, cm^{-1}) 3442, 3068, 1637, 1614; MS (EI): m/z (%)=358 (100, $[M+H]^+$); HRMS (EI): $[M+H]^+$, found: 358.0793. $C_{23}H_{13}ClFN$ requires 358.0798.

*4.1.1.25. Benzo[b]acridine (**10g**).* Yield: 19.3 mg, 42%; brown powder; mp 82–84 °C; ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.62–7.65 (m, 1H, Ar−*H*), 7.67–7.70 (m, 1H, Ar−*H*), 7.73–7.76 (m, 1H, Ar−*H*), 7.83–7.86 (m, 1H, Ar−*H*), 7.92 (d, J=8.5 Hz, 1H, Ar−*H*), 7.98–8.00 (m, 1H, Ar−*H*), 8.06 (d, J=8.5 Hz, 1H, Ar−*H*), 8.12 (d, J=8.5 Hz, 1H, Ar−*H*), 8.32 (d, J=8.5 Hz, 1H, Ar−*H*), 8.77 (d, J=8.5 Hz, 1H, Ar−*H*), 9.46 (s, 1H, Ar−*H*) ppm; ^{13}C NMR (125 MHz, $CDCl_3$): δ 122.9, 124.2, 126.3, 126.5, 127.6, 127.9, 128.3, 128.4, 128.5, 128.9, 129.8, 130.5, 131.1, 131.2, 133.1, 147.5, 148.8 ppm; IR (KBr, cm^{-1}) 3439, 2920, 1615, 1502; MS (EI): m/z (%)=230 (20, $[M+H]^+$), 229 (100); HRMS (EI): $[M+H]^+$, found: 230.0964. $C_{17}H_{11}N$ requires 230.0965.

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Supplementary data

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