



Synthesis of acridone-naphthylamine derivative and its thermally-activated delayed fluorescence studies for application in OLEDs

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Abstract. Acridone (acceptor) and naphthylamine (donor) based Donor-Acceptor-Donor (D-A-D) compound (**1**) was synthesised, characterised and its thermally-activated delayed fluorescence (TADF) properties were studied in detail. Compound **1** is fluorescent and emits in the green region (550 nm). The energy gap between the ground and the lowest excited singlet (S_1) state is estimated to be 2.55 eV. The energy gap between the CT singlet and triplet states (ΔE_{ST}) was found to be ~ 0.3 eV. Small $\Delta E_{S_1-T_1}$ is one of the important criteria for TADF to take place in a molecule and thus detailed photophysics has been studied. Transient lifetime measurements showed an increase in the fluorescence lifetime (τ) on purging with N_2 , as compared with that in air-saturated solution, indicating the involvement of the triplet state in emission. Emission at 550 nm was also observed with a delay of 100 μs which corresponded to the delayed fluorescence in **1**. The lifetime of TADF was found to be 176 μs . Applications of TADF materials in organic light-emitting devices (OLEDs) has gotten attention as TADF materials utilise the triplet excitons which helps in increasing internal quantum efficiency of device. Air-saturated based on **1** were fabricated and their intensity was found to be nearly as high as 17,000 Cd/m^2 at 25 mA/cm^2 which was comparable to many of the known TADF emitters.

Keywords. Acridone; thermally activated delayed fluorescence; organic light-emitting devices.

1. Introduction

As the demand for cost-competitive and renewable energy options grows, considerable attention is being placed on the development of new materials for low energy consuming devices. Organic semiconductors offer a range of low-cost device applications, for example, large-area displays, organic light-emitting devices (OLED) and solar cell.^{1–8} Organic chemistry offers a great advantage in designing a large variety of materials of similar chemical structures with tunable photophysical properties by derivatization and this has made possible the development of more efficient organic semiconductors for solid-state devices

including OLEDs. Typically, under electrical excitation, fluorescent molecule generates up to 25% singlet and 75% non-emissive triplets excitons which results in only 25% internal quantum efficiency (IQE). The non-emissive triplet excitons are deactivated *via* non-radiative processes generating heat.^{9–12} Thus, significant research is being pursued to utilize the triplet excitons improving the IQE at room temperature. Electroluminescence in OLEDs arises from the singlet state in fluorescent materials, while that is from the triplet state in phosphorescent emitters. It has been shown that 100% internal efficiency of OLEDs can be obtained when noble metal (e.g., Pd, Pt, Ir, etc.) based phosphorescent materials are used as an emitter.^{13,14}

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Development of efficient and cost-effective noble metal-containing phosphorescent materials for the entire visible region is still challenging and limits their commercial production. Recently, it has been shown that metal-free organic molecules capable of showing thermally activated delayed fluorescence (TADF) can be used to access both the singlet and triplet excitons for emission. To enhance the efficiency of OLEDs, TADF enabled molecules are considered as alternative to phosphors.^{8,10,15–19} Materials having small energy difference between the lowest singlet (S_1) and triplet (T_1) excited states promote reverse intersystem crossing (RISC) at room temperature and thus populate the S_1 state. Such emission from S_1 as a result of RISC is reported as thermally activated delayed fluorescence (TADF). TADF capable materials have potential to populate the singlet by using the triplet states, therefore they are considered as better emissive materials for OLEDs.^{20,21} It is envisaged that with TADF materials, 100% internal efficiency (theoretically) can be obtained in OLEDs as is the case with phosphors. High fluorescence quantum yield, short lifetime of the S_1 state and small ΔE_{ST} (~ 0.3 eV) are the important criteria for organic molecules to act as TADF emitter. The main challenge in designing a TADF molecule is to ensure a small energy gap between the lowest excited singlet and triplet states (ΔE_{ST}). TADF molecules, in which an electron donor (D) and acceptor (A) moieties are covalently linked, have been reported to have small ΔE_{ST} . Such materials show intramolecular charge transfer (ICT) character, which results in very small overlap between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO). Small or no overlap between the HOMO and LUMO leads to decrease in electronic exchange energy, thus helping to obtain small energy gap between the singlet and triplet states.²²

Push-pull molecules based on anthraquinone, acridone, quinacridone, etc., having singlet and triplet states with strong charge-transfer character, have been reported to possess small ΔE_{ST} and were explored for TADF properties.^{16,23–26} Recently, we reported the aromatic amine (cyclic as well as acyclic) substituted acridone derivatives and their photophysical properties.^{16,27} In our earlier results carbazole (cyclic amine) substituted acridone derivatives showed TADF properties. In this article, we report the design and synthesis of acridone derivative substituted with acyclic bulky 2,2'-dinaphthylamine, (2,7-bis-(N,N' -di-2-naphthyl)-10-methylacridone, **1**). In compound **1**, acridone core was chosen as electron acceptor and dinaphthylamine as electron donor. Our earlier results

with acyclic amine substituted acridone showed promising results for organic electronics materials. We further modified the design and chose naphthyl amine as a bulky substituent which can restrict the rotation of amine and is expected to bring non-planarity in the molecule. Non-planar materials having strong electron acceptor and donating moieties are expected to show charge transfer property, one of the important criterion for TADF materials. Photophysical properties of **1** were studied in detail which shows that it possesses TADF character. OLEDs were also fabricated which showed green emission with a light intensity of 17,000 Cd/m² at a current density of 25 mA/cm².

2. Experimental

2.1 Materials and instruments

Reagents and solvents were purchased from Sigma Aldrich or S. D. Fine Chemicals and used as received. Solvents were dried, wherever anhydrous solvents were required. For absorption and emission, solvents used were of spectroscopic grades. ¹H NMR were recorded on a Varian 600 MHz spectrometer. Mass spectra was recorded using Bruker MALDI-TOF. Cyclic Voltammetry was performed on CH Instrument (620D). Three electrode cell was employed with a glassy carbon working electrode, Ag/AgCl (non-aqueous) reference electrode and Pt wire counter electrode. The measurements were performed in anhydrous acetonitrile with tetrabutylammonium hexafluorophosphate as supporting electrolyte at a scan rate of 100 mV/s. UV-visible spectra were recorded using a Shimadzu 1800 from 800 nm to 200 nm. Steady-state fluorescence spectrophotometer were recorded on a Horiba Fluoromax-4 spectrofluorometer, with sample excited at 410 nm. A diode laser-based time-correlated single-photon counting (TCSPC) spectrometer (IBH, U.K.) was used to obtain the excited-state lifetimes. The sample was excited using a 406 nm laser to obtain the lifetime. TADF lifetime was obtained on an Agilent spectrofluorometer, with emission recorded after a delay time (100–400 μ s). Emission peak intensity was plotted against the delay time and fitted using equation $I = I_0 e^{-t/\tau}$. Wherever required, deconvolution was performed on origin 9 software using nonlinear curve fit operation. Gaussian function and Levenberg-Marquardt Iteration Algorithm was used to fit the multi peaks. For OLED fabrication, films were grown by spin coating (using a Holmarc HO-TH-05 spin coater) at 2000 rpm for 40 s. Vacuum deposition was carried out at a base pressure of 2×10^{-6} mbar. ITO coated substrate (15–25 Ω /sq, Sigma Aldrich) was etched into desired pattern. Substrate cleaning was done with soap solution, propanol and trichloroethylene (TCE) vapours in stepwise manner. UV treatment was given before using

them in device fabrication. The organic layer of **1** was thermally evaporated in vacuum giving a film of ~50 nm over a layer of PEDOT:PSS which was spin-coated (~50 nm thick) and a layer of *N,N'*-Bis(naphthalen-1-yl)-*N,N'*-bis(phenyl)benzidine (NPD; ~40 nm). The thickness of PEDOT:PSS and NPD were measured by surface profilometer (DEKTAK). NPD, bathophenanthroline (Bphen; ~25 nm), LiF (~1 nm), and Al (~160 nm) were all thermally evaporated. The geometry of the most optimized device was ITO/(PEDOT:PSS/ NPD/1/Bphen/LiF/Al).

2.2 Synthesis

2,7-Dibromo-10-methylacridone was synthesized according to literature.

1: Compound **1** was synthesized by Buchwald-Hartwig amination. 2,7-Dibromo-10-methylacridone (0.075 g, 0.20 mmol) and 2,2'-dinaphthylamine (0.13 g, 0.50 mmol) were dissolved in anhydrous toluene (20 mL) under argon atmosphere. [Pd₂(dba)₃] (5–8 mol%), 2-dicyclohexylphosphino-2',6'-dimethylbiphenyl (SPhos) (10–15 mol%) and sodium-*t*-butoxide (3.1 mmol) were added to the reaction mixture. The reaction mixture was thoroughly stirred under argon atmosphere while the temperature was slowly raised to 100 °C and refluxed for 18 h. Product was extracted using dichloromethane (DCM) and organic layers were washed with water. Crude mixture was purified through column chromatography using 50% DCM in Hexane. Yield: 8% ¹H NMR (CDCl₃, 600 MHz, δ ppm): 8.30 (d, 2H, *J* = 2.4 Hz), 7.74 (m, 8H); 7.63 (d, 1H, *J* = 3.0 Hz); 7.61 (d, 1H, *J* = 2.4 Hz); 7.56 (s, 2H); 7.55 (s, 2H); 7.53 (s, 1H); 7.51 (s, 1H); 7.44 (d, 4H *J* = 2.4 Hz); 7.39 (d, 1H, *J* = 1.8 Hz); 7.38 (d, 2H, *J* = 1.2 Hz); 7.37 (s, 1H, *J* = 1.6 Hz); 7.36 (s, 1H, *J* = 1.6 Hz); 7.35 (d, 2H, *J* = 0.6 Hz); 7.33 (d, 3H, *J* = 2.4 Hz); 7.32 (d, 2H, *J* = 1.8 Hz); 3.95 (s, 3H). MALDI-TOF: *m/z* [M]⁺ calcd. C₅₄H₃₇N₃O, 743.89; found: 742.42.

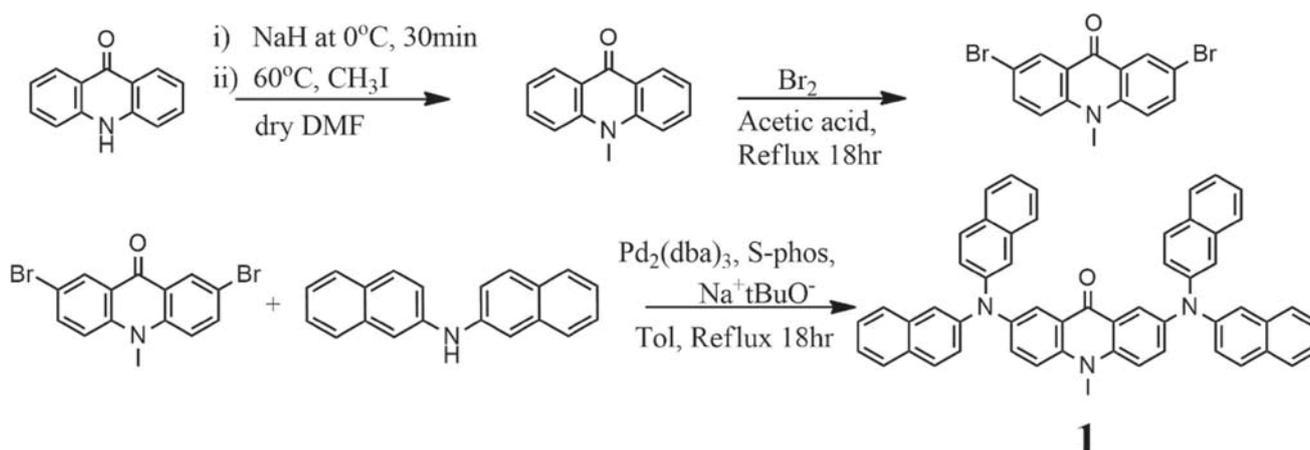
3. Results and Discussions

3.1 Synthesis

Compound **1** was synthesized in three steps, starting from acridone, as shown in Scheme 1. For methylation, 9(10H)-acridone was reacted with iodomethane in the presence of NaH. A light-yellow solid product, methylacridone, was further brominated using molecular bromine in acetic acid. Brominated acridone was then dried, purified and identified as 2,7-dibromo-10-methylacridone as yellow solid.²⁷ The amination of 2,7-dibromo-10-methylacridone and 2,2'-dinaphthylamine was carried using B-H amination as mentioned in the experimental section. Pure **1** was obtained in ~8% yield. Compound **1** was characterised by ¹H-NMR and MALDI-TOF (Supplementary Information).

3.2 Photophysical studies

Absorption and emission properties of **1** have been studied in acetonitrile and spectra are shown in Figure 1 and tabulated in Table 1. The absorption spectrum shows multiple bands. Two prominent bands with maxima at ~260 and 345 nm are the characteristics of 2,2'-dinaphthylamine. A broad shoulder in the 400–500 nm region is assigned to the acridone moiety. The emission spectrum of compound **1** shows a maximum at 550 nm. The energy gap between the ground state (S₀) and the first excited singlet state (S₁) is determined from the intersection of emission and excited spectra, which corresponds to 2.55 eV (Figure S3, Supplementary Information). The fluorescence quantum yields (Φ_F) of **1** was determined by using a comparative method using curcumin in acetonitrile as



Scheme 1. Synthesis of acridone-dinaphthylamine derivative, **1**.

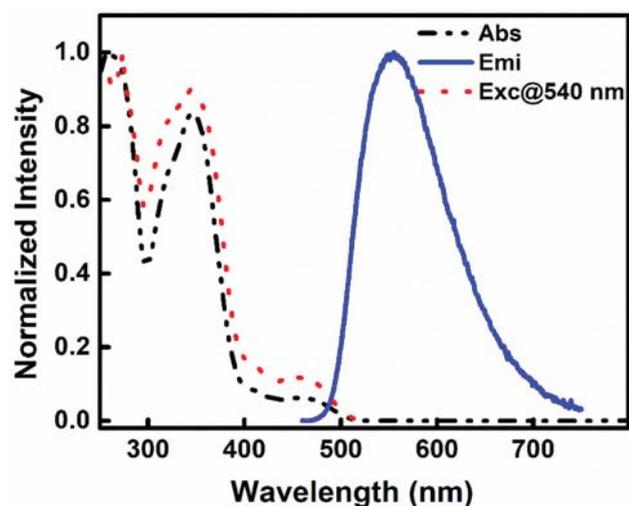


Figure 1. Absorption, emission and excitation spectra of the compound **1** in acetonitrile.

the standard (Q.Y. = 0.075).²⁸ Fluorescence quantum yield was found to be 0.09 for **1**.

In acetonitrile, the broad emission band suggests ICT character of the S_1 state. ICT character of **1** was further investigated by solvatochromic study. The absorption and emission spectra of **1** were recorded in various solvents of different polarities and they have been shown in Figure S4 and S5, respectively (Supplementary Information). While the absorption characteristics (such as width and maximum of the spectrum) of **1**, which is a D-A-D type molecule having 2,2'-dinaphthylamine as the electron donor and 10-methylacridone as the electron acceptor moieties, are independent on solvent polarity, characteristics of the emission spectrum show a strong polarity dependence. The emission spectrum recorded in hexane shows a maximum at 490 nm and a shoulder at 525 nm, which could be assigned to the vibronic features.²⁹ The width of the emission spectrum increases and the emission maximum displays larger bathochromic shift as the polarity of the medium increases. The shift is as large as about 25 nm in acetonitrile as compared to that in hexane. These observations confirm the prediction about the strong ICT character of the excited state of **1**.

To understand the possibility of delayed fluorescence in **1**, it is desirable to know the photophysical

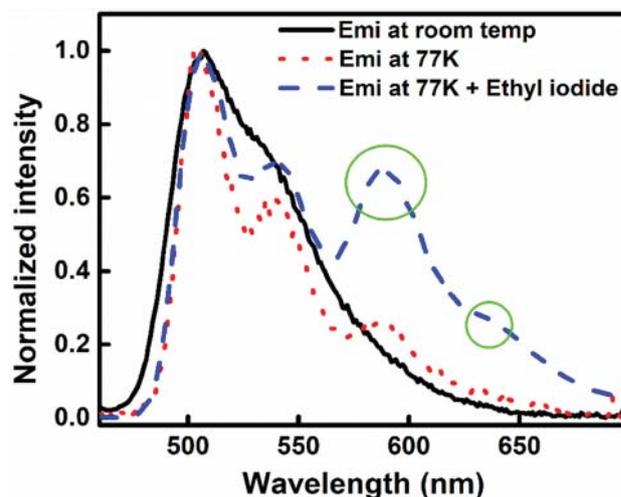


Figure 2. Emission spectra of **1** in methyl THF at room temperature (black), at 77 K (red) and at 77K with ethyl iodide (blue).

properties of the phosphorescent or the triplet energy state of the molecule. Due to the long lifetime (in the range of a few μ s to ms) of the triplet state, the triplet state emission in liquid solution at room temperature is efficiently quenched *via* various non-radiative processes, for example, intermolecular collisional quenching, vibrational relaxations, etc.^{30,31} To eliminate the possibility of the triplet state emission quenching, emission spectrum of **1** was recorded in THF solvent matrix at 77 K. This emission spectrum shows two additional bands with the emission maxima at 590 and 635 nm in addition to those due to fluorescence emission at 500 and 530 nm, which could be considered to be arising from the triplet state (phosphorescence).

The intensity of the phosphorescence band (triplet emission) is found to be smaller as compared to that of singlet emission. Addition of ethyl iodide ($\sim 10\%$ v/v) to the solvent matrix increases the intensities of phosphorescence bands by several folds (Figure 2) due to heavy atom effect. Heavy atoms (I, Pt, Pd, Ru, etc.) are known to enhance the triplet state population.¹⁶ Thereby, it confirms that emission bands at 590 and 635 nm at low temperature are arising from the triplet state(s). The emission spectrum recorded at 77 K in the THF solvent matrix containing ethyl iodide was

Table 1. Photophysical data of **1**.

Comp.	^a λ_{abs} nm	^a λ_{em} (nm)	^a ϕ_{F}	^a τ_{PF} (ns)	^a τ_{DF} (μ s)	^b ΔE_{ST} eV
1	260, 346, 465	550	0.09	11.7 (in air) 18.6 (in N ₂)	70 (in air) 178 (in N ₂)	0.3

^aIn acetonitrile; ^bin Me-THF.

deconvoluted to obtain the characteristic peaks corresponding to the singlet and triplet states (Figure S6, Supplementary Information). The energy levels (singlet and triplet) of **1** were estimated from the onset of fluorescence and phosphorescence peaks. The singlet-triplet energy gap (ΔE_{ST}) was found to be 0.3 eV.

To obtain information on the relaxation of the excited states of **1**, we studied the fluorescence lifetimes using TCSPC technique in solution. A picosecond (ps) diode laser of 375 nm and 75 ps pulse duration was used for excitation. The TCSPC instrument has a time resolution of about 300 ps. Emission was monitored at 525 nm. Fluorescence lifetimes were recovered by fitting the temporal profiles using iterative deconvolution method using multi-exponential fit functions. Fluorescence lifetimes of **1** were measured in a few solvents with air saturation as well as N₂ purged (Figure 3). In all the cases, the fluorescence profiles could be fitted well with single exponential function and the lifetimes thus recovered are tabulated in Table 2, which suggests that lifetimes are nearly independent of the solvent polarity.

Earlier, it was proposed that a short TADF lifetime is beneficial to suppress triplet quenching processes like triplet-triplet annihilation, triplet-polaron annihilation, etc., thereby improving singlet fluorescence efficiency *via* TADF.³² The delayed fluorescence lifetime in TADF molecules is mostly in microseconds-milliseconds scale.^{10,33} Here, we have attempted to evaluate the TADF lifetime of compound **1** in acetonitrile solution. TADF process engages both the singlet and triplet states and thus the presence of molecular oxygen (air) and lifetime scale of these two states will affect the efficiency of the TADF emission. Fluorimetric studies were carried out to find out the TADF lifetime in an air-saturated as well as a nitrogen-purged atmosphere at room temperature. Here, we recorded emission spectra at various delay times ranging from 100 μ s to 400 μ s in air-saturated and nitrogen purged acetonitrile (Figures S7 and 8, Supplementary Information) and the peak intensity of emission (at 550 nm) was plotted against delay time (Figure S9, Supplementary Information). Lifetime was calculated using the equation, $I = I_0 e^{-t/\tau}$. Figure 4

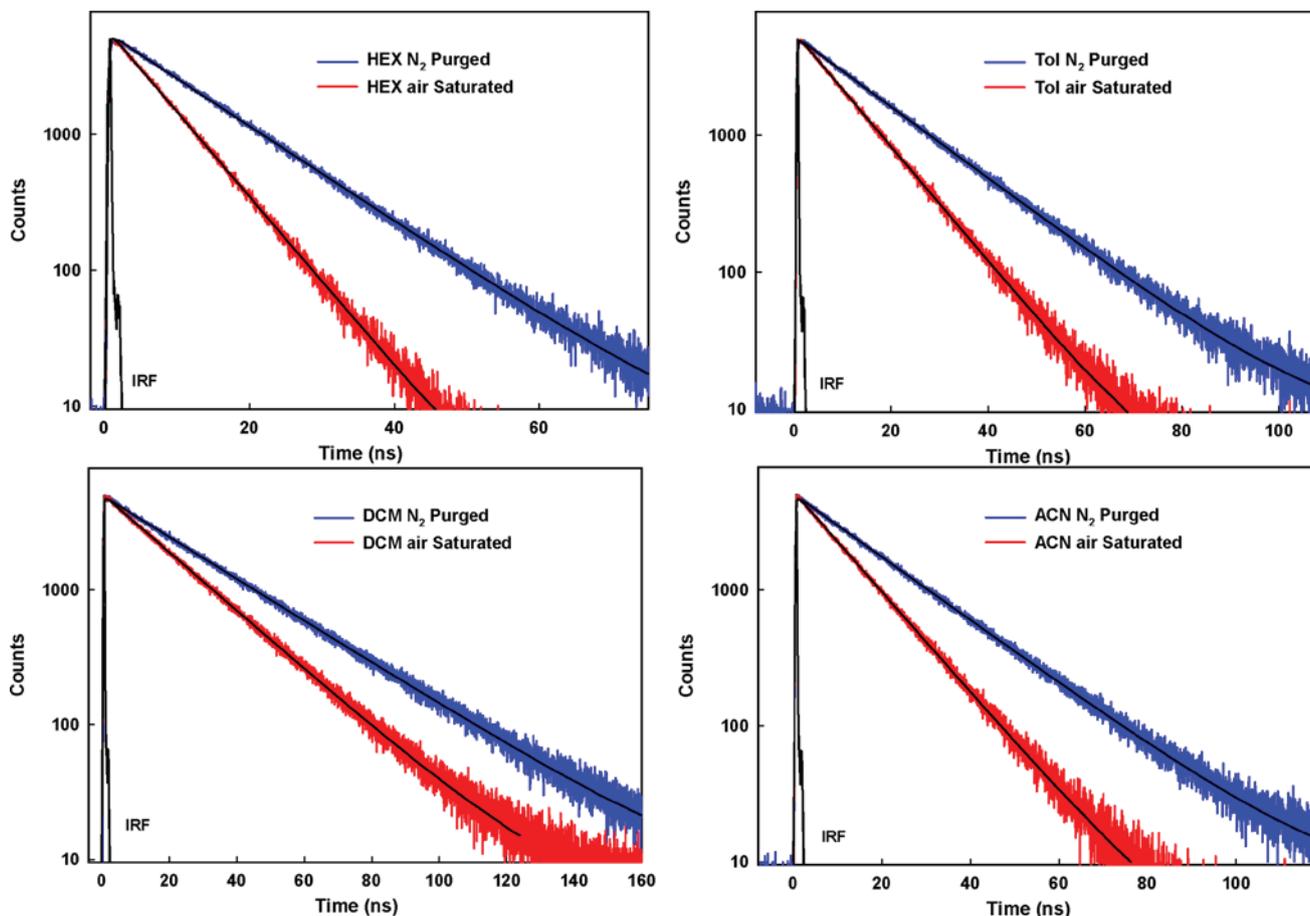
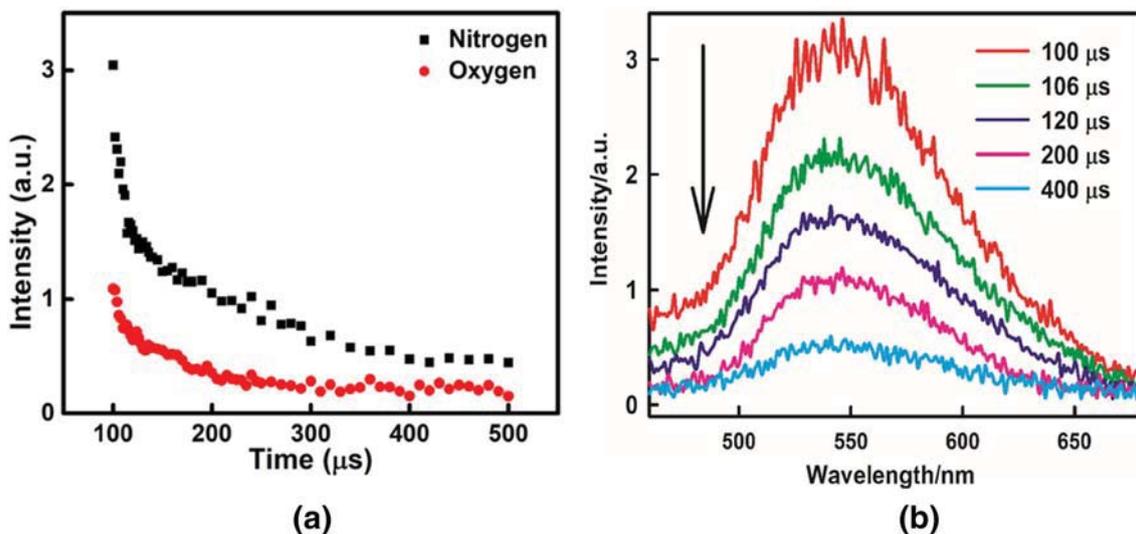


Figure 3. Fluorescence decay profile of **1** in different air saturated and nitrogen purged solvents.

Table 2. Fluorescence lifetime (ns) data of **1** in different solvents.

	Acetonitrile	Hexane	Dichloromethane	Toluene
Air-saturated	11.7	6.9	19.9	10.3
N ₂ purged	18.6	12.4	27.8	16.2

**Figure 4.** (a) Emission decay profile of **1** in acetonitrile at room temperature in air-saturated and nitrogen purged solvent. (b) Emission spectra of **1** recorded after various delays in a nitrogen-purged solvent.

shows the TADF emission and lifetimes of **1** in microseconds. Since delay time was kept much longer than 100 ns, therefore, prompt fluorescence could be ruled out as prompt fluorescence lifetime is about 10 ns only. Also, the emission maximum (550 nm in the same solvent) in the time-resolved emission spectra matches well with that observed in prompt fluorescence of **1**. These observations clearly suggest that the emission process occurring at delay times longer than 100 μ s time domain is due to the TADF process. In N₂ purged solution, emission intensity was seen to increase and the lifetime ($\sim 178 \mu$ s) was also found to be longer than that ($\sim 70 \mu$ s) in air-saturated solution, thus confirming the participation of the triplet state in the emission process.

3.3 Cyclic voltammetry

Cyclic voltammetric studies were carried out to find out the HOMO and LUMO energy levels. E_{HOMO} and E_{LUMO} play an important role in finding suitable electrodes and supporting organic materials while working on the device geometry. The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels of **1** were

estimated using the oxidation potential and optical bandgap. The cyclic voltammetric method was used to find out the oxidation potential of **1**. First oxidation peak was found at 0.65 V. The HOMO energy levels of **1** were calculated using oxidation potentials, taking Fc/Fc^+ (0.33 V) as a standard.³⁴ Using oxidation potential, the HOMO energy of **1** is found to be roughly ~ -5.4 eV. The optical gap was measured from the intersection of excitation and emission spectra and found to be 2.55 eV. Using E_{HOMO} and optical gap, E_{LUMO} was estimated at ~ -2.85 eV.

3.4 Organic light-emitting device and characterization

To study the potential of **1** as an emissive layer in OLEDs, devices were fabricated and studied. The OLEDs were fabricated using **1** as a neat emitter without doping in any host material as the latter is believed to alter device properties. Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), a well-known hole injecting layer, is used in these studies. *N,N'*-di-(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPD) was used as hole transporting layer (HTL). The most optimized device

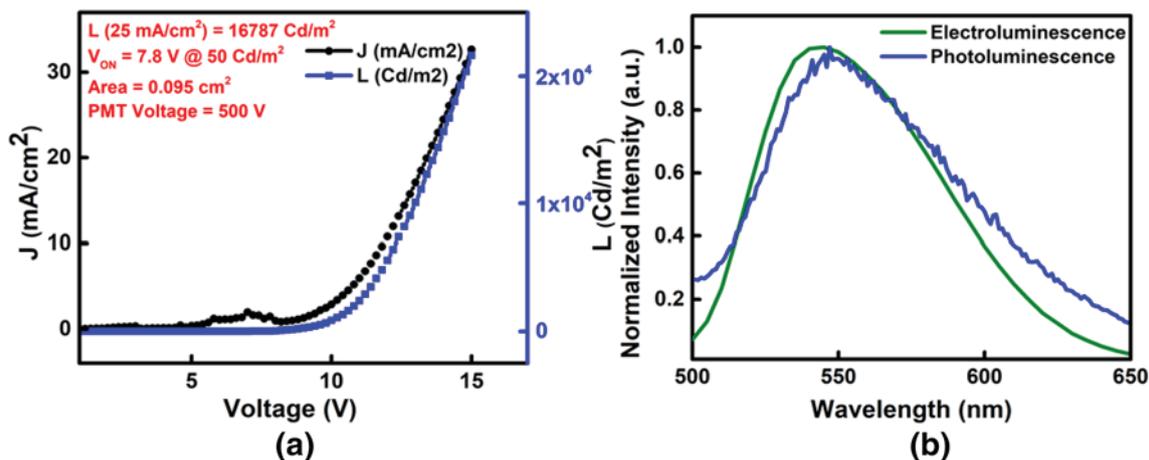


Figure 5. (a) J-V-L characteristics of the OLED device with the geometry ITO/PEDOT:PSS/NPD/1/BPhen/LiF/Al and (b) electroluminescence and photoluminescence (thin film) spectra.

structure used was ITO/PEDOT:PSS/NPD/1/BPhen/LiF/Al. Typically, hole blocking layers (HBL) are most commonly placed after the emissive layer closer to the cathode to confine the carriers and excitons in emissive layer. 4,7-Diphenyl-1,10-phenanthroline (BPhen) was used as the hole blocking layer (HBL) in our devices. Electroluminescence spectra of the devices showed the peak emission nearly at 550 nm and matches with the photoluminescence of **1** (Figure 5). The turn-on voltage, V_{ON} (voltage at which intensity is 50 Cd/m^2) was found to be 7.8 V. A high luminous intensity of $\sim 17000 \text{ Cd/m}^2$ at a current density of 25 mA/cm^2 was observed for the OLED of **1** (Figure 5). High luminous intensity is believed to be because of TADF emission of compound **1**. Earlier reported acridone based compounds showed luminous intensity of $\sim 1000 \text{ Cd/m}^2$ at a current density of 10 mA/cm^2 and 9800 Cd/m^2 at 10 mA/cm^2 ¹⁶.

4. Conclusions

Acridone-amine derivative (**1**) was synthesized by substituting acyclic di-naphthyl amine on acridone. To study its prospects in TADF photophysical studies were carried out in detail. Compound **1** is fluorescent and emits in the green region (550 nm). Phosphorescence spectra was recorded at 77 K to find out the triplet energy level. The energy gap between the singlet and triplet state (ΔE_{ST}) is found to be $\sim 0.3 \text{ eV}$. Emission at 550 nm was also observed with a delay of $100 \mu\text{s}$ which correspond to the delayed fluorescence in **1**. The lifetime of TADF was found to be $176 \mu\text{s}$ in a nitrogen-purged solvent. OLEDs based on **1** were fabricated and their intensity was found to be nearly $17,000 \text{ Cd/m}^2$ at 25 mA/cm^2 .

Supplementary Information (SI)

Spectra related to NMR, mass, absorption, emission and delayed lifetime studies, cyclic voltammogram is available at www.ias.ac.in/chemsci.

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