

Journal of Materials Chemistry C

Accepted Manuscript



This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this *Accepted Manuscript* with the edited and formatted *Advance Article* as soon as it is available.

You can find more information about *Accepted Manuscripts* in the [Information for Authors](#).

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard [Terms & Conditions](#) and the [Ethical guidelines](#) still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.



ARTICLE

A single emitting layer white OLED based on exciplex interface emission

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

www.rsc.org/

E. Angioni,^a M. Chapran,^b K. Ivaniuk,^b N. Kostiv,^b V. Cherpak,^b P. Stakhira,^{b,*} A. Lazauskas,^c S. Tamulevičius,^c D. Volyniuk,^d N. J. Findlay,^a T. Tuttle,^a J. V. Grazulevicius^{d,*} and P. J. Skabara^{a,*}

A new triaryl molecule based on a benzene-benzothiadiazole-benzene core has been applied in a WOLED device. This very simple molecule emits from a combination of emissive states (exciton/electromer/exciplep/electroplex) to give white light with CIE coordinates of (0.38, 0.45) and a colour temperature of 4500K.

Introduction

During the last two decades, the swift development of new and high performing inorganic and organic emissive materials has brought to the market competitive and efficient solid state lighting (SSL) devices. They are predicted to become the next generation of general illumination systems.¹ Inorganic white light emitting diodes (LEDs) are commercially used nowadays for general illumination purposes (*e.g.* automotive lighting, indoor and outdoor lighting).² White organic light emitting diodes (WOLEDs) instead are principally used as low-cost alternatives for back-lights in flat panel displays.³ They present several advantages compared to their inorganic counterparts, such as lower cost, ease of processability and facile tuning of their properties by chemical modifications.^{3, 4} Furthermore, they can be fabricated as flexible panels with wide viewing angles and a superior white colour balance.⁵ White SSL can be achieved using different approaches, but commercially successful methods include: (I) combining a blue (*ca.* 460 nm) LED and a yellow phosphor pumped from the blue light of the LED;⁶⁻⁸ (II) combining a single chip emitting UV light which is absorbed in the LED package by three phosphors (red, green and blue) and re-emitted as a broad spectrum of white light^{9,10} or (III) three different LED chips, each emitting a different wavelength [red, green and blue (RGB)] in order to simulate the RGB colour model.⁹⁻¹⁵ The design of these configurations is not straightforward and to achieve competitive efficiencies

and luminosity the use of hybrid (inorganic-organic) solutions has been explored.^{11-13, 16} Furthermore, white light produced in this fashion is difficult to fine-tune, in particular when a mixture of several emitters is used.¹⁷ In order to simplify the device architecture, several strategies have been used to fabricate single molecular layer WOLEDs.⁵ Heagy *et al.* employed *N*-aryl-2,3-naphthalimides with low symmetry that exhibited efficient panchromatic emission.^{18, 19} Liu *et al.* observed white solid-state luminescence as well as electroluminescence from the controlled protonation of a molecular blue fluorophore,²⁰ while Chou *et al.* showed white-light emission in a single excited-state-intramolecular-proton-transfer (ESIPT) system by fine-tuning the energetics of the excited state.²¹ White-light emission can also be achieved from intermolecular interactions or from the formation of complexes, *e.g.* excimers or exciplexes.^{17, 22-26} Excimers are possible in single component organic materials due to the resonance interaction of a molecular exciton with a neighbour non-excited molecule,²⁷⁻²⁹ whereas exciplexes can be formed in a bi- or multi-component molecular solid when the formation of bimolecular excited states is facilitated by electron transfer between the donor and acceptor components.^{30, 31} In 2002 Wang and co-workers reported a highly efficient white device based on the exciplex between a boron complex and *N,N'*-di(1-naphthyl)-*N,N'*-diphenylbenzidine (NPB).³² Qiu and co-workers observed exciplex type white-light emission from the interface of a bilayer electroluminescent device consisting of a new electron transport material, anthracene-9,10-diylbis(diphenylphosphine oxide) (DPPA) and *N,N'*-bis(naphthalen-1-yl)-*N,N'*-bis(phenyl)benzidine (NPB)²² as the hole-transporting layer. Recently, Cherpak *et al.* reported a new approach for the fabrication of a WOLED that consists of the combination of the blue phosphorescence emission from the iridium (III) bis[4,6-difluorophenyl]-pyridinato-*N,C2'*-picolinate (**Flrpic**) complex and the highly efficient delayed fluorescent emission from the exciplex formed at the interface between the star-shaped hole transporting material tri(9-hexylcarbazol-3-yl)amine and

^a WestCHEM, Department of Pure and Applied Chemistry, University of Strathclyde, Glasgow, G1 1XL, UK. E-mail: peter.skabara@strath.ac.uk

^b Lviv Polytechnic National University, S. Bandera 12, 79013 Lviv, Ukraine.

^c Institute of Materials Science, Kaunas University of Technology, Baršausko str. 59, LT-51423 Kaunas, Lithuania.

^d Department of Polymer Chemistry and Technology, Kaunas University of Technology, Radvilenu Plentas 19, LT-50254 Kaunas, Lithuania.

† Footnotes relating to the title and/or authors should appear here.

Electronic Supplementary Information (ESI) available: [experimental procedures, cyclic voltammetry, spectroscopy data, AFM, modelling, TGA]. See DOI: 10.1039/x0xx00000x

Frpic.²⁶ In this work, an OLED based on the new compound dimethyl 4,4'-(benzo[1,2,5]thiadiazole-4,7-diyl)bis(3-methoxybenzoate) (**1**) has been fabricated and its performance studied.

Results and discussion

Synthesis and opto-electronic properties

Compound **1** was synthesised from commercially available 4,7-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzo[c][1,2,5]thiadiazole *via* Suzuki-Miyaura cross-coupling with methyl 4-iodo-3-methoxybenzoate³³ (**2**) (Fig. 1a). In the UV-Vis absorption spectrum of **1** (10^{-5} M in dichloromethane) (Fig. 1b) the typical absorption band of the benzothiadiazole (BT) core³⁴⁻³⁶ is blue shifted to 295 nm ($19300 \text{ M}^{-1} \text{ cm}^{-1}$) and its vibronic fine structure is visible. In a similar fashion, the less intense band, attributable to the interaction between the peripheral benzene rings and the core (HOMO-LUMO transition, Table S1, ESI) is blue shifted and centered at 366 nm ($9500 \text{ M}^{-1} \text{ cm}^{-1}$). This is likely due to the non-planarity of **1**, contrary to some planar 4,7-di-substituted benzothiadiazole analogous compounds.³⁶⁻³⁸ From the longest wavelength absorption edge of the UV-Vis absorption spectrum (Fig. 1) it was possible to calculate an optical HOMO-LUMO gap of 3.0 eV. Photoluminescence measurements were performed for

a dilute dichloromethane solution of **1** (10^{-6} M) at 300K (excitation at 366 nm) and for a dilute THF solution of **1** (10^{-6} M) at 300 and 77K (excitation at 330 nm) showing emission maxima at 472, 487 and 449 nm, respectively. Furthermore, photoluminescence quantum yields (PLQYs) of *ca.* 50% and *ca.* 6% were recorded at 300K using an excitation wavelength of 366 nm for the dilute dichloromethane solution of **1** (10^{-6} M) and for the encapsulated powder of **1** (pressed against two glass substrates), respectively. The red shift with decreasing polarity of solvent is likely due to the donor-acceptor character of **1**. In the excited state one expects a large dipole moment due to charge transfer and thus a high sensitivity to solvent polarity. Instead, as the temperature of THF is decreased to 77K, the emission spectrum of **1** is observed from a less relaxed excited state resulted in a shift of the fluorescence maxima to shorter wavelengths.³⁹⁻⁴¹

Cyclic voltammograms were recorded for a dilute solution of **1** in dichloromethane using tetrabutylammonium hexafluorophosphate as the supporting electrolyte. They show a reversible reduction ($-1.99/-1.87$ V) and three irreversible oxidations (1.36, 1.50, 1.63 V) (Fig. S1, ESI). The reduction process can be attributed to the semi-reversible reduction of the BT unit, known to be an electron acceptor,⁴² whereas the anodic waves can be attributed to the oxidation processes likely localised on the methoxy groups as revealed in similar 4,7-bis-substituted BT compounds.^{43, 44} The electrochemical HOMO and LUMO levels were calculated from the onset of the first oxidation wave (-6.0 eV), and reduction wave (-2.9 eV), respectively (the data were referenced to ferrocene, which has a HOMO of -4.8 eV).⁴⁵ The electrochemical HOMO-LUMO gap was therefore determined to be 3.1 eV. With respect to the HOMO energy determined by CV (-6.0 eV), a very close value of 5.9 eV was also calculated as the HOMO level from the photoelectron emission spectrum of the solid state film obtained by vacuum deposition onto an indium tin oxide (ITO) coated glass substrate (Fig. S2, ESI). The electrochemical HOMO and LUMO energies were used for the development of the device conception and in the following energy diagrams.^{41, 46}

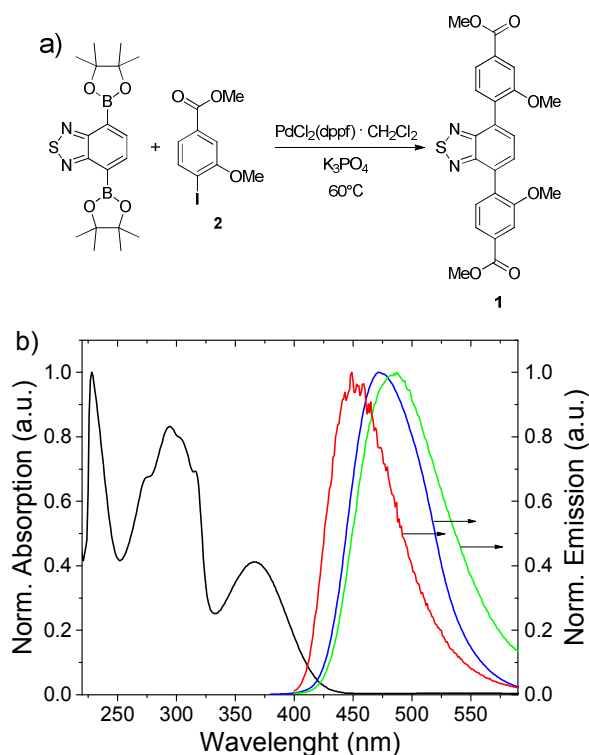


Fig. 1 (a) Synthesis of compound **1** (58% yield). (b) UV-Vis absorption spectrum (black) of a solution of **1** in dichloromethane (10^{-5} M) and the emission spectrum of the solution of **1** in dichloromethane (blue) and tetrahydrofuran (10^{-6} M) at different temperatures (300K, green; 77K, red). The solutions were excited at 366 nm (dichloromethane) and at 330 nm (tetrahydrofuran).

Morphology and charge transport properties

AFM measurements were performed on vacuum deposited layers of **1** at different deposition rates on glass substrates: (a) $< 0.1 \text{ \AA s}^{-1}$, (b) 1.5 \AA s^{-1} and (c) 10 \AA s^{-1} (Fig. S3, ESI). They show random oriented pillar/needle-shaped surfaces with the mean height of the peaks (18.41 nm, 20.07 nm and 25.53 nm) and the root mean square roughness (9.38 nm, 10.62 nm and 15.99 nm) increasing with the deposition rate. The surfaces are dominated by peaks with a skewness (*RSk*) of 2.25, 1.63 and 1.29, respectively, and they have a leptokurtic distribution of the morphological features with a kurtosis (*RKu*) of 11.10, 7.58 and 5.20, respectively. The X-ray diffraction patterns recorded on the samples deposited at (a) $< 0.1 \text{ \AA s}^{-1}$, (b) 1.5 \AA s^{-1} and (c) 10 \AA s^{-1} (Fig. S4, ESI) show a completely amorphous structure for **1**, independent of the deposition rate.

The space-charge-limited current (SCLC) technique was used to evaluate the charge-transporting properties of **1**, as SCLC can

be used to measure the charge carrier mobility in thin films of low molar mass compounds.⁴⁷ Hole-only and electron-only devices were prepared using 4,4',4''-tris[3-methylphenyl(phenyl)amino]triphenylamine (*m*-MTDATA) as the hole-injecting/electron-blocking layer and 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi) as the electron-injecting/hole-blocking layer. The two devices were fabricated by successive deposition onto a pre-cleaned indium tin oxide (ITO) coated glass substrate under a vacuum of 10^{-6} Torr, with architectures of: ITO/*m*-MTDATA(20 nm)/**1**(60 nm)/*m*-MTDATA(20 nm)/Al(60 nm) (hole-only) and ITO/TPBi(20 nm)/**1**(60 nm)/TPBi (20 nm)/Ca(10 nm)/Al(60 nm) (electron only). The hole mobility of **1** ($7.6 \times 10^{-7} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $9.6 \times 10^5 \text{ V cm}^{-1}$) has a higher value than the electron mobility ($1.7 \times 10^{-7} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $9.6 \times 10^5 \text{ V cm}^{-1}$; Fig. S5, ESI). The fitted parameters μ_0 and γ are $2.6 \times 10^{-8} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $3.4 \times 10^{-3} \text{ cm V}^{-1}$, and $2.8 \times 10^{-9} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $4.2 \times 10^{-3} \text{ cm V}^{-1}$ for holes and electrons, respectively (Fig. S6, ESI). Despite the low charge mobility, **1** can be used successfully for the preparation of emitting layers, due to the nano-scale thicknesses required for the OLED fabrication.⁴⁸

Device fabrication and exciplex emission

An electroluminescent device based on **1** was fabricated. Due to the morphology, the low mobility and the energetically deep HOMO level of **1**, *N,N'*-bis(3-methylphenyl)-*N,N'*-diphenylbenzidine (TPD) was used as hole-transporting material to promote a cross-interaction between the excess of electrons from the LUMO of **1** and the excess of holes from the HOMO of TPD with the aim of inducing the formation of a dimeric excited state (*e.g.* exciplex). 2,9-Dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) was used as an electron-transporting/hole-blocking layer in order to increase the injection of electrons from the calcium anode and to balance the amount of electrons and holes that combine in the emissive layer. Additionally, to promote hole and electron mobility, CuI was used as a hole-injecting material.

The electroluminescent device (Fig. S7, ESI) was fabricated by successive deposition onto a pre-cleaned indium tin oxide (ITO) coated glass substrate under a vacuum of 10^{-5} Torr using specific deposition rates for the different layers: ITO/CuI (8 nm, 0.1 nm s^{-1})/TPD (10 nm, 0.2 nm s^{-1})/**1** (100 nm, 1 nm s^{-1})/BCP (30 nm, 0.2 nm s^{-1})/Ca (7 nm, 0.1 nm s^{-1})/Al (100 nm, 0.1 nm s^{-1}). The active area of the obtained device was 6 mm^2 and additional passivation was not applied. The fast deposition rate (10 \AA s^{-1}) was adopted in the device fabrication process in order to facilitate the creation of an interaction with the adjacent layers. The electroluminescence (EL) spectrum of the device recorded with an applied voltage of 15 V (Fig. 2a) reveals four superimposed different emission bands. The origins of these bands are shown in Fig. S8. The higher energy emission band (494 nm) is characteristic of the pure fluorescence emission of **1**. This is supported by the luminescence decay time measurements of the spin coated film of **1** (at 488 nm, *ca.* 10^{-2} M in THF) being on a nanosecond scale (Fig. 2b) and from the single emission band centred at 490 nm in its photoluminescence spectrum (Fig. 2a).⁴⁹ The

main band at *ca.* 580 nm originates from the exciplex at the interface between **1** and the hole-transporting material TPD. In fact, the luminescence decay time measurements (at 580 nm) of the composite layer prepared by spin coating a solution of **1** and TPD (*ca.* 0.01M in THF) onto a clean quartz substrate shows a combination of 17 ns (41%) and 61 ns (59%) decay times (Fig. 2b), due to the presence of both the exciton fluorescence emission and the exciplex emission at the interface between TPD and **1**.^{26, 49, 50} When a direct bias is applied, due to high energy barriers between TPD and **1** ($\text{HOMO}^{\text{TPD}} - \text{HOMO}^{\text{1}} = 0.5 \text{ eV}$; $\text{LUMO}^{\text{TPD}} - \text{LUMO}^{\text{1}} = 0.6 \text{ eV}$), electrons and holes accumulate at the interface between the two layers and, due to non-planar conformations, the electronic overlap of donor (TPD) and acceptor (**1**) molecules is efficient for exciplex formation at the interface. In fact, the twisted nature of **1** makes self-stacking less likely, increasing the probability of a hetero-interaction with an adjoining molecule. The long-wavelength shoulder at 635 nm in the EL spectra can be assigned to the typical (classic) electroplex interaction that is associated with the exciplex emission.^{49, 51} The band at *ca.* 520 nm can be attributed to an electromer interaction, that appears upon the injection of electrons and holes under an electric field effect in **1**.^{52, 53} The formation of an electromer is

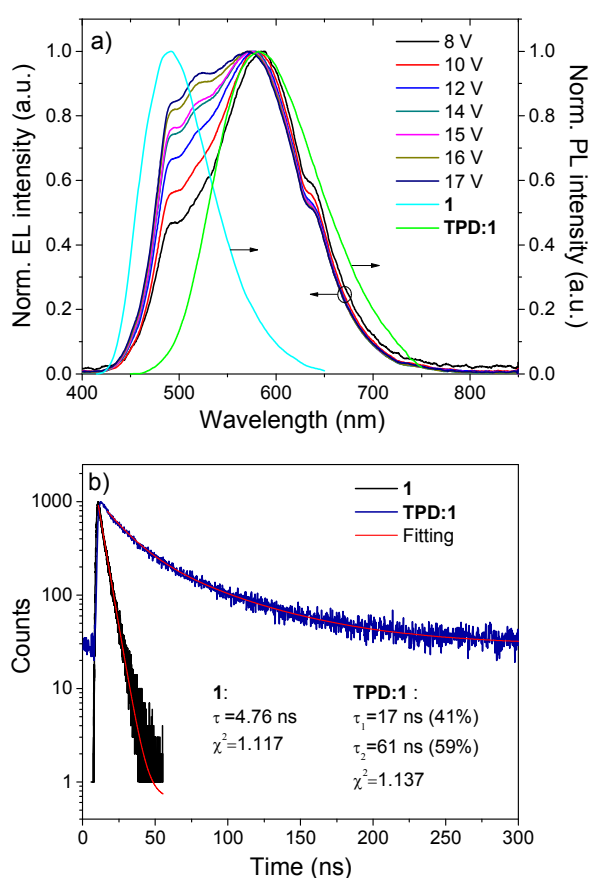


Fig. 2 (a) Electroluminescence spectra of the OLED at selected applied voltages and photoluminescence spectra of the thin films of **1** and of the mixture of TPD with **1** (1:1)

w/w). (b) Decay fluorescence plots measured for the thin films of **1** (at 488 nm) and of the mixture of **TPD** with **1** (at 580 nm).

observed when a pair of trapped carriers recombines under an electronic interaction and its emission is red-shifted with respect to fluorescence.⁵¹ Therefore, the white emission of the device originates from the combination of the exciplex/electroplex emission at the interface between **TPD** and **1** and from the exciton/electromer emission of **1**. This is possibly due to the thickness of the emissive layer (100 nm) that permits emission from all the excited states simultaneously. The EL spectra at different applied voltages were recorded (Fig. 2a) and they reveal four different emission bands, with relative intensities that depend on the applied voltage. The maxima of the higher energy emission band (494 nm) and of the electromer emission (520 nm) stay constant with increasing applied voltage, whilst the relative intensities increase. The main emission maximum is blue-shifted from 588 to 570 nm when the applied voltage is increased from 8 to 17 V. Conversely, both the position and the intensity of the shoulder at 635 nm remains effectively unchanged at the different applied voltages. Despite these behaviours, the colour quality of the device is maintained at all applied voltages. The CIE coordinates change only slightly from (0.42, 0.44) when the EL is recorded at 8 V to (0.37, 0.44) at 17 V (Fig. S9 and Table S2, ESI), due mainly to the increased relative intensity of the EL and of the electromer transition of **1**. However, at applied voltages higher than 15 V, the device tends to quickly degrade and the characteristics recorded are not maintained. For this reason the following characteristics of the device were recorded up to a maximum applied voltage of 15 V.

The current density–voltage characteristics and luminance–voltage characteristics (Table S3–S4, ESI) indicate an OLED turn on voltage of 5.8 V, which corresponds to an electroluminescence of 1.4 cd m⁻². The device exhibits a maximum current efficiency of 6.5 cd A⁻¹ and a maximum brightness of 5219 cd m⁻² (at 15 V) (Fig. S10–S12, ESI). At maximum brightness an external quantum efficiency of 2.39% and power efficiency of 2.60 Lm W⁻¹ were recorded. The white light emitted from the device at 15 V has Commission Internationale d’Eclairage (CIE 1931) coordinates of (0.38, 0.45) (Fig. S9), with a colour temperature of 4500K; values similar to those of commercial fluorescent tubes.^{13, 54, 55} The relatively low brightness and current efficiency of the single-layer OLED can be explained by the presence of electroplex emission that naturally results in the decrease of OLED efficiency.⁵⁶ On the other hand, the presence of the electroplex-type excited states makes it possible to expand the EL spectrum towards the red region and to obtain a white OLED with a simplified structure.⁵⁶

Computational results

To explain the PL and EL spectra observed experimentally, quantum chemical calculations were performed for **1**, **TPD** and **1:TPD** complexes using density functional theory (DFT) and its time resolved counterpart (TD-DFT).^{57, 58} In order to choose the

most suitable level of theory capable of simulating the vertical transitions associated with the absorption spectra, a benchmark was performed (Table S5, ESI) and, as a result, the PBE0/6-311G(d,p) level of theory was used for all the calculations. Initially eight different geometric isomers of the **TPD:1** complexes (a–h) were optimised (Fig. S13, ESI), with isomer **a** showing the total energy minima and hence was chosen as the starting geometry for the following calculation (Table S6, ESI). The ground state geometries for **1**, **TPD** and the complex **TPD:1** were optimised (Fig. S13–14, ESI) and analysis of the vibrational frequencies revealed the location of the

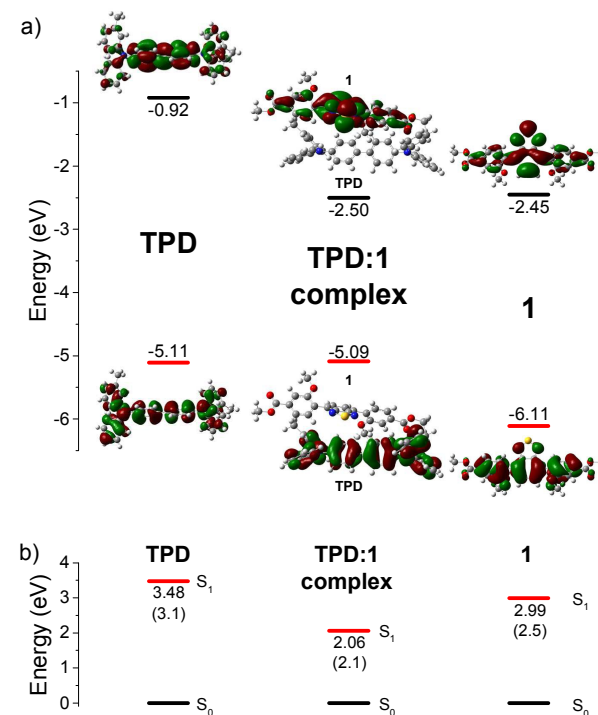


Fig. 3 a) Kohn-Sham molecular orbital diagram and orbital graphical representations (HOMO and LUMO) of **TPD**, **1** and of the complex **TPD:1** calculated at the PBE0/6-311G(d,p) level of theory (isosurface 0.02). b) First singlet excited state energies calculated at the PBE0/6-311G(d,p) level of theory for **TPD**, **1** and for the complex **TPD:1**; the figures in parentheses are the energy levels determined experimentally.

sought-for energy minimum. The HOMO and LUMO orbitals are widely delocalised on the π backbone for **1** and **TPD**, with their energy gaps agreeing qualitatively with the experimental data (Fig. 3a). The **TPD:1** complex shows a LUMO localised on **1** and a HOMO localised on the **TPD** molecule. The reorganisation energies for electrons (λ_e) and holes (λ_h) were calculated theoretically⁵⁹ for **1**. The values obtained for $\lambda_e = 0.425$ eV and $\lambda_h = 0.384$ eV agree qualitatively with the experimental observations, as the reorganisation energy for the electrons is slightly higher than the corresponding value for holes. Note that the lower the λ value, the higher the charge-transport rate.⁵⁹ The first 20 singlet vertical transitions were calculated in the vacuum (Table S1, S7, S8, ESI) using the TD-DFT/PBE0/6-311G(d,p) level of theory. The energies of the first singlet excited state (S₁) of **1**, **TPD** and of the **TPD:1**

complex are reported in Fig. 3b. They agree qualitatively, by the levels of relative disposition, with the experimental PL emission maxima of thin films of the mixture of TPD:1 and compound 1, and from the literature data for the film of TPD.⁶⁰ The recombination process of an electron-hole pair with the electron located on the LUMO of 1 and the hole located on the HOMO of TPD should then be responsible for the EL maximum at ca. 580 nm for the device and explains the exciplex nature of this transition. Here the exciplex is considered as an excited state complex that is formed by an electronically excited state donor molecule (or acceptor) with a complementary acceptor molecule (or donor) in their ground state.⁶¹

Conclusions

A WOLED containing the small and simple organic molecule 1 was fabricated. The resulting electroluminescence was shown to be a combination of the exciton/electromer emission of 1 and of the exciplex/electroplex emission at the heterojunction between 1 and TPD. The device has a maximum current efficiency of 6.5 cd/A, a maximum brightness of 5219 cd/m² and good colour quality with C.I.E. 1931 coordinates (0.38, 0.45) at 15 V and a colour temperature of 4500K.

Acknowledgements

The authors would like to thank the UK Engineering and Physical Sciences Research Council (Grant No: EP/I012591/1), the European Commission (Marie Curie Action of FP7, Grant No: PIRSES-GA-2013-612670), the University of Strathclyde, the Lviv Polytechnic National University and the Kaunas University of Technology for the financial support. We thank the EPSRC UK National mass spectrometry facility for the HRMS measurements. PJS thanks the Royal Society for a Wolfson Research Merit Award.

References

1. Y. Takei, *Sci. Technol. Trends-Quarterly Rev.*, 2009, **32**, 59.
2. M. Bessho and K. Shimizu, *Electron. Comm. Jpn.*, 2012, **95**, 1.
3. J. Kalinowski, V. Fattori, M. Cocchi and J. A. G. Williams, *Coord. Chem. Rev.*, 2011, **255**, 2401.
4. J. Roncali, *Macromol. Rapid Commun.*, 2007, **28**, 1761.
5. S. Mukherjee and P. Thilagar, *Dyes Pigm.*, 2014, **110**, 2.
6. K. Bando, K. Sakano, Y. Noguchi and Y. Shimizu, *J. Light & Vis. Env.*, 1998, **22**, 2.
7. R.-J. Xie, N. Hirotsaki, M. Mitomo, Y. Yamamoto, T. Suehiro and K. Sakuma, *J. Phys. Chem. B*, 2004, **108**, 12027.
8. R.-J. Xie, N. Hirotsaki, K. Sakuma, Y. Yamamoto and M. Mitomo, *Appl. Phys. Lett.*, 2004, **84**, 5404.
9. J. K. Sheu, S. J. Chang, C. H. Kuo, Y. K. Su, L. W. Wu, Y. C. Lin, W. C. Lai, J. M. Tsai, G. C. Chi and R. K. Wu, *IEEE Photon. Technol. Lett.*, 2003, **15**, 18.
10. W.-J. Yang, L. Luo, T.-M. Chen and N.-S. Wang, *Chem. Mater.*, 2005, **17**, 3883.
11. K. T. Kamtekar, A. P. Monkman and M. R. Bryce, *Adv. Mater.*, 2010, **22**, 572.
12. C. Tang, X.-D. Liu, F. Liu, X.-L. Wang, H. Xu and W. Huang, *Macromol. Chem. Phys.*, 2013, **214**, 314.
13. M. C. Gather, A. Köhnen and K. Meerholz, *Adv. Mater.*, 2011, **23**, 233.
14. N. Thejo Kalyani and S. J. Dhoble, *Renew. Sust. Energ. Rev.*, 2012, **16**, 2696.
15. I. Moreno and U. Contreras, *Opt. Express*, 2007, **15**, 3607.
16. N. J. Findlay, J. Bruckbauer, A. R. Inigo, B. Breig, S. Arumugam, D. J. Wallis, R. W. Martin and P. J. Skabara, *Adv. Mater.*, 2014, **26**, 7290.
17. G. Giro, M. Cocchi, J. Kalinowski, P. Di Marco and V. Fattori, *Chem. Phys. Lett.*, 2000, **318**, 137.
18. P. Nandhikonda and M. D. Heagy, *Chem. Commun.*, 2010, **46**, 8002.
19. P. Nandhikonda and M. D. Heagy, *Org. Lett.*, 2010, **12**, 4796.
20. D. Liu, Z. Zhang, H. Zhang and Y. Wang, *Chem. Commun.*, 2013, **49**, 10001.
21. K.-C. Tang, M.-J. Chang, T.-Y. Lin, H.-A. Pan, T.-C. Fang, K.-Y. Chen, W.-Y. Hung, Y.-H. Hsu and P.-T. Chou, *J. Am. Chem. Soc.*, 2011, **133**, 17738.
22. Y. Zhao, L. Duan, X. Zhang, D. Zhang, J. Qiao, G. Dong, L. Wang and Y. Qiu, *RSC Adv.*, 2013, **3**, 21453.
23. J. Karpiuk, E. Karolak and J. Nowacki, *Phys. Chem. Chem. Phys.*, 2010, **12**, 8804.
24. V. Jankus, P. Data, D. Graves, C. McGuinness, J. Santos, M. R. Bryce, F. B. Dias and A. P. Monkman, *Adv. Funct. Mater.*, 2014, **24**, 6178.
25. T. Ishisone, S. Seo, Y. Nonaka, T. Kawata and N. Ohsawa, *J. Soc. Inf. Display*, 2014, **22**, 404.
26. V. Cherpak, P. Stakhira, B. Minaev, G. Baryshnikov, E. Stromylo, I. Helzhynskyy, M. Chapran, D. Volyniuk, Z. Hotra, A. Dabulienė, A. Tomkeviciene, L. Voznyak and J. V. Grazulevicius, *ACS Appl. Mater. Interfaces*, 2015, **7**, 1219.
27. D. Thirion, M. Romain, J. I. Rault-Berthelot and C. Poriel, *J. Mater. Chem.*, 2012, **22**, 7149.

ARTICLE

Journal Name

28. J.-Y. Hu, Y.-J. Pu, Y. Yamashita, F. Satoh, S. Kawata, H. Katagiri, H. Sasabe and J. Kido, *J. Mater. Chem. C*, 2013, **1**, 3871.
29. J.-Y. Hu, Y.-J. Pu, G. Nakata, S. Kawata, H. Sasabe and J. Kido, *Chem. Commun.*, 2012, **48**, 8434.
30. M. Sharnoff, *J. Lumin.*, 1971, **4**, 69.
31. S. Yang and M. Jiang, *Chem. Phys. Lett.*, 2009, **484**, 54.
32. Y. Liu, J. Guo, H. Zhang and Y. Wang, *Angew. Chem. Int. Ed.*, 2002, **41**, 182.
33. D. Rankine, A. Avellaneda, M. R. Hill, C. J. Doonan and C. J. Sumbly, *Chem. Commun.*, 2012, **48**, 10328.
34. J.-M. Raimundo, P. Blanchard, H. Brisset, S. Akoudad and J. Roncali, *Chem. Commun.*, 2000, 939.
35. B. A. D. Neto, A. A. M. Lapis, E. N. da Silva Júnior and J. Dupont, *Eur. J. Org. Chem.*, 2013, **2013**, 228.
36. S.-i. Kato, T. Matsumoto, T. Ishi-i, T. Thiemann, M. Shigeiwa, H. Gorohmaru, S. Maeda, Y. Yamashita and S. Mataka, *Chem. Commun.*, 2004, 2342.
37. M. Akhtaruzzaman, N. Kamata, J.-i. Nishida, S. Ando, H. Tada, M. Tomura and Y. Yamashita, *Chem. Commun.*, 2005, 3183.
38. D. Aldakov, M. A. Palacios and P. Anzenbacher, *Chem. Mater.*, 2005, **17**, 5238.
39. W. Rettig, *Angew. Chem. Int. Ed.*, 1986, **25**, 971.
40. J. R. Lakowicz and A. Balter, *Photochem. Photobiol.*, 1982, **36**, 125.
41. G. Weber and F. J. Farris, *Biochemistry*, 1979, **18**, 3075.
42. P. Ledwon, N. Thomson, E. Angioni, N. J. Findlay, P. J. Skabara and W. Domagala, *RSC Adv.*, 2015, **5**, 77303.
43. K. M. Omer, S.-Y. Ku, K.-T. Wong and A. J. Bard, *J. Am. Chem. Soc.*, 2009, **131**, 10733.
44. G. M. Saltan, H. Dinçalp, M. Kiran, C. Zafer and S. Ç. Erbaş, *Mater. Chem. Phys.*, 2015, **163**, 387.
45. J. L. Bredas, R. Silbey, D. S. Boudreaux and R. R. Chance, *J. Am. Chem. Soc.*, 1983, **105**, 6555.
46. J. Sworakowski and J. Ulanski, *Annu. Rep. Prog. Chem., Sect. C: Phys. Chem.*, 2003, **99**, 87.
47. J. C. Blakesley, F. A. Castro, W. Kylberg, G. F. A. Dibb, C. Arantes, R. Valaski, M. Cremona, J. S. Kim and J.-S. Kim, *Org. Electron.*, 2014, **15**, 1263.
48. S. R. Tseng, Y. S. Chen, H. F. Meng, H. C. Lai, C. H. Yeh, S. F. Horng, H. H. Liao and C. S. Hsu, *Synth. Met.*, 2009, **159**, 137.
49. J. Kalinowski, M. Cocchi, D. Virgili, V. Fattori and J. A. G. Williams, *Adv. Mater.*, 2007, **19**, 4000.
50. Y.-H. Lee, T.-C. Wu, C.-W. Liaw, T.-C. Wen, S.-W. Feng, J.-J. Lee, Y.-T. Wu and T.-F. Guo, *Org. Electron.*, 2013, **14**, 1064.
51. J. Kalinowski, *Mater. Sci.-Poland*, 2009, **27**, 735.
52. S. Kwon, K.-R. Wee, C. Pac and S. O. Kang, *Org. Electron.*, 2012, **13**, 645.
53. J. Kalinowski, G. Giro, M. Cocchi, V. Fattori and P. Di Marco, *Appl. Phys. Lett.*, 2000, **76**, 2352.
54. G. M. Farinola and R. Ragni, *Chem. Soc. Rev.*, 2011, **40**, 3467.
55. Q. Wang and D. Ma, *Chem. Soc. Rev.*, 2010, **39**, 2387.
56. G. M. Farinola and R. Ragnar, *Chem. Soc. Rev.*, **2011**, **40**, 3467.
57. R. Improta, V. Barone, G. Scalmani and M. J. Frisch, *J. Chem. Phys.*, 2006, **125**, 054103.
58. F. Furche and R. Ahlrichs, *J. Chem. Phys.*, 2002, **117**, 7433.
59. L.-Y. Zou, A.-M. Ren, J.-K. Feng and X.-Q. Ran, *J. Phys. Org. Chem.*, 2009, **22**, 1104.
60. H. Mattoussi, H. Murata, C. D. Merritt, Y. Iizumi, J. Kido and Z. H. Kafafi, *J. Appl. Phys.*, 1999, **86**, 2642.
61. C.-C. Yang, C.-J. Hsu, P.-T. Chou, H. C. Cheng, Y. O. Su and M.-k. Leung, *J. Phys. Chem. B*, 2010, **114**, 756.

Graphical Abstract

