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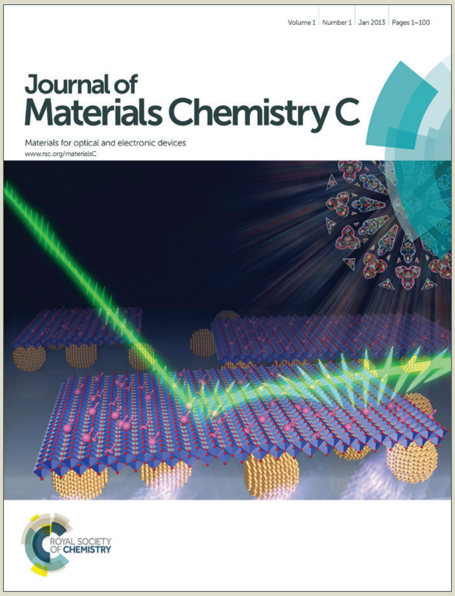

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ARTICLE

On the Selection of a Host Compound for Efficient Host-Guest Light-Emitting Electrochemical Cells

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

DOI: 10.1039/x0xx00000x

www.rsc.org/

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The light-emitting electrochemical cell (LEC) is characterized by its electrochemical doping operation that facilitates advantages as regards to device fabrication and functionality, but it currently suffers from that the efficiency at significant luminance is not very high. A viable solution to this setback could be the implementation of a host-guest active material, where the majority host transports the electronic charge and the guest is a triplet emitter that features an appropriate energy structure for facile exciton transfer and trapping as well as efficient light emission. Here, we demonstrate that an additional critical property of a functional host-guest LEC is that the host can be electrochemically p- and n-type doped, as can be deduced from screening studies on open planar devices and with cyclic voltammetry. LEC devices based on hosts that do not fulfill this fundamental criterion are shown to suffer from low luminance and poor efficiency, whereas host-guest LECs, based on a host material capable of electrochemical doping, exhibit a much improved luminance and efficiency, with the efficiency being well retained also at high luminance values.

Introduction

The light-emitting electrochemical cell (LEC) is a technology under rapid development that features important application advantages in comparison to competing emissive technologies, as exemplified by recent demonstrations of an all-ambient solution-based processing fit for low-cost roll-to-roll manufacturing,[1-5] the realization of stretchable devices,[6, 7] and the fabrication of light-emission films directly on complex-shaped surfaces[8, 9]. These attractive opportunities are effectuated by that the LEC is insensitive to the work function of the electrodes and to the thickness of the constituent materials, which in turn are traceable to its characteristic *in-situ* electrochemical doping operation.[10, 11] The active material of an LEC comprises a blend of luminescent organic semiconductor (OSC) and mobile ions, and when a voltage is applied between the two charge-injecting electrodes, these ions redistribute to allow for electrochemical doping of the OSC; p-type doping takes place at the positive anode and n-type doping takes place at the negative cathode. With time, these doped regions grow in size so that a light-emitting p-n junction doping structure forms in the active material. At the p-n junction, subsequently injected electrons and holes (often termed polarons) recombine to form excitons, which can decay under the emission of photons.

Although the performance of LECs has improved steadily over the last few years, it is clear that a further enhancement of the energy-conversion efficiency at high light output is necessary if the LEC is going to become competitive in high-end applications.[12, 13] A resolution to this challenge needs to address the following issues: (i) During electric driving, ≈75 % of the excitons are formed in a triplet state and ≈25 % in the singlet state in an OSC, and only the singlet excitons are emissive in a fluorescent OSC. (ii) A phosphorescent OSC can harvest also the triplet excitons for light-emission via inter-system crossing, but the characteristic long lifetime of the triplet state can instead result in long-range triplet migration and associated triplet-triplet or triplet-polaron light-quenching reactions.[14, 15] (iii) Before exiting the LEC device structure, the photons formed in the p-n junction need to pass through (at least) one doped layer; and if this layer is based on the same material as the emitter severe self-absorption can result, particularly if a thick active material is employed.[16, 17]

In organic light-emitting diodes, which also utilize a luminescent OSC for the light-emission process and which are structurally similar to the LEC, the above challenge has been successfully resolved through the implementation of a host-guest system, where (a significant portion of) the electronic transport takes place on a majority host and the emission originates from a minority guest. With the guest being a triplet emitter, all excitons can be transformed into photons. If the guest further is dispersed within the host on a molecular level, triplet diffusion and the related triplet-triplet and triplet-polaron annihilation will be effectively suppressed. Finally, with the majority host being a high energy-gap material and the minority guest being a comparatively low energy-gap

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Electronic Supplementary Information (ESI) available: [AFM images, PL data, characteristics of reference devices and energy structure of the electrolyte]. See DOI: 10.1039/x0xx00000x

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material, issues with self-absorption[16, 17] and back transfer to the host[18] will also be minimized.

A set of rather sporadic reports on the employment of host-guest systems in LEC devices exists in the scientific literature, and a few representative examples are listed below. Pei and co-workers introduced the concept in 2002 when they reported an LEC comprising a polyfluorene derivate as the host and a triplet-emitting Ir complex as the guest, and although this device featured red emission from the minority guest it also suffered from a very low luminance.[19] Malliaras *et al.* employed an orange-emitting ionic Ru compound as the host and a red-emitting ionic Os compound as the guest, and showed that the emission color could be tuned through the concentration of the guest.[20] Su and co-workers utilized a green-emitting Ir complex as the host and an orange-emitting Ir complex as the guest for the attainment of an impressive power conversion efficacy of 37 lm/W, but only reported data up to a low luminance level of <80 cd/m^2 (at which the efficiency had dropped to approximately half of the peak value).[21] The latter two systems shared another drawback in that the employment of a majority host based on a rare metal will translate into a high material cost. Su and Wong addressed this issue when they reported a host-guest LEC with a cationic terfluorene derivative as the host and a red-emitting Ir complex as the guest, but again only data at a very low luminance of ~1 cd/m^2 were reported.[22] Recently, Bolink and co-workers reported a blue-emitting host-guest LEC, comprising a two-component host system and an Ir complex as the guest, which featured a relatively high current efficacy of 5 cd/A at a significant luminance of 420 cd/m^2 during pulsed-current driving.[23]

Although some promising results indeed have originated from these studies, it is striking that none up-to-this-point has addressed the specific requirements that the characteristic LEC mode, *i.e.* the *in-situ* electrochemical doping of the OSC, put on the host-guest system. Through a systematic study on three different host-guest LECs featuring efficient energy-transfer process to the guest, we show that the capacity for reversible p- *and* n-type electrochemical doping of the host compound is a key enabling factor for achieving a high device performance. We further report that such a functional host-guest LEC can emit red light with a current efficacy of 3.5 cd/A at a luminance of 200 cd/m^2, and importantly that the current efficacy is well maintained at 2.5 cd/A at a high luminance of 2000 cd/m^2. The same device also features a relatively fast turn-on time of 6 s to a luminance of 100 cd/m^2 during galvanostatic driving at 77 mA/cm^2.

Experimental

Materials and characterization

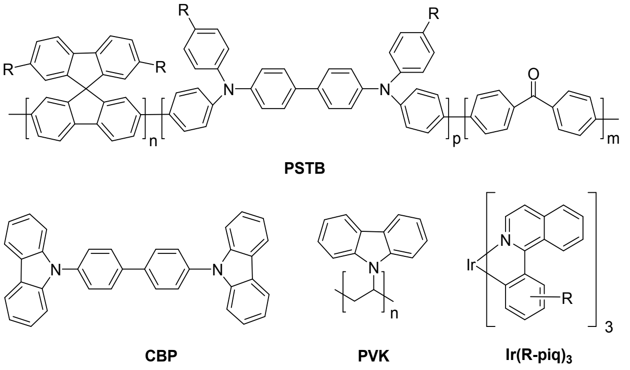

Figure 1. Chemical structure of the host materials, PSTB, CBP and PVK, and the guest compound Ir(R-piq)$_3$.

All of the investigated materials were used as received. The chemical structure of the three commercially available host materials, poly(9-vinylcarbazole) (PVK, Sigma-Aldrich), 4,4-bis(9-carbazolyl)biphenyl (CBP, Sigma-Aldrich) and a copolymer with spirobifluorene, tetra-N-phenylbenzidine, and benzophenone as the backbone building blocks (here termed PSTB, acquired from Merck), as well as the guest compound tris[1-(substituent-phenyl)-isoquinoline]iridium (III) (Ir(R-piq)$_3$, acquired from Merck), are displayed in Fig. 1. The ion-transporter is hydroxyl-capped trimethylolpropane ethoxylate (TMPE-OH, M_w = 450 g/mol, Sigma-Aldrich)[24] or poly(ethylene oxide) (PEO, M_w = 5,000,000 g/mol, Sigma-Aldrich), and the salt is $LiCF_3SO_3$ (Sigma-Aldrich) or KCF_3SO_3 (Sigma-Aldrich).

Thin films for the absorption (UV-3100 spectrophotometer, Shimadzu) and photoluminescence (PL) measurements (FP-6500 spectrofluorometer, JASCO) were spin-coated onto carefully cleaned quartz substrates. The optical energy gap was determined from the absorption spectrum as the intercept of the linear extrapolation of the tangent at half peak height on the low-energy side with the baseline. The surface morphology was measured with atomic force microscopy (AFM, MultiMode SPM microscope equipped with a Nanoscope IV Controller, Veeco Metrology). Cyclic voltammetry (CV) was carried out with an Autolab PGSTAT302 potentiostat using the GPES software. The host material was coated onto the Au-coated surface of a glass substrate to form the working electrode, a Pt rod was the counter electrode, a Ag wire was the quasi-reference electrode, and 0.1 M tetrabutylammonium hexafluorophosphate ($TBAPF_6$, Fluka) in anhydrous CH_3CN was the electrolyte. Directly after each CV scan, a calibration scan was run with a small amount of ferrocene/ferrocenium ion (Fc/Fc^+) added to the electrolyte. All CV potentials are reported *vs.* Fc/Fc^+. The reduction/oxidation onset potentials are defined as the intersection of the baseline with the tangent of the current at the half-peak-height. CV sample preparations and measurements were performed under N_2 atmosphere in a glovebox ($[O_2]$ < 1 ppm, $[H_2O]$ < 0.5 ppm).

Device fabrication and characterization

The planar bilayer LECs were fabricated as follows. Au electrodes with an inter-electrode gap of 300 µm were deposited onto carefully cleaned glass substrates by thermal

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evaporation (at $p < 5\times10^{-4}$ Pa) through a shadow mask. The bi-layer was deposited by first spin-coating (at 2000 rpm for 60 s) the host material dissolved in THF, being PSTB (10 mg/ml), CBP (20 mg/ml) or PVK (15 mg/ml), and then spin-coating (at 800 rpm for 60 s) the {PEO+KCF_3SO_3} electrolyte (10 mg/ml in acetonitrile). The mass ratio of the electrolyte constituents was PEO:KCF_3SO_3 = 1.35:0.5, and the dry thickness of the host and the electrolyte layers were 100 nm and 180 nm respectively, as established with a stylus profilometer (Dektak). The planar bilayer LECs were characterized in an optical-access vacuum chamber at $p < 5\times10^{-5}$ Pa. A computer-controlled source-measure unit (Keithley 2400) sourced the voltage and measured the current. The photographs of the doping process and the light emission were recorded in a dark room under UV illumination (λ = 365 nm) through the optical window of the vacuum chamber, using a digital camera (Canon EOS 50D) equipped with a macro lens (65 mm, F/2.8).

The sandwich-cell LECs were fabricated by sequentially spin-coating first a planarizing layer of poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate) (PEDOT-PSS, Clevios P VP AI 4083, Heraeus) at 4000 rpm for 60 s and then the {Host:Ir(R-piq)$_3$:TMPE-OH:$LiCF_3SO_3$} active material at 2000 rpm for 60 s onto carefully cleaned indium-tin-oxide (ITO) coated glass substrates (20 Ω/square, Thin Film Devices, US). The mass ratio of the active-material constituents was Host:Ir(R-piq)$_3$:TMPE-OH:$LiCF_3SO_3$ = 1:0.05:0.10:0.03. The dry thickness of the PEDOT-PSS and the active material layers was 40 nm and 100 nm, respectively. A set of four Al cathodes was deposited on top of the active material film by thermal evaporation at $p < 5\times10^{-4}$ Pa. The light-emission area, as defined by the size one such cathode, was 0.85×0.15 cm^2. The LEC devices were characterized using a computer-controlled source-measure unit (Agilent U2722A) and a calibrated photodiode, equipped with an eye-response filter (Hamamatsu Photonics), connected to a data acquisition card (National Instruments USB-6009) *via* a current-to-voltage amplifier. The electroluminescence (EL) spectra were recorded using a calibrated fiber-optic spectrometer (USB2000, Ocean Optics), and the Commission Internationale de l'Eclairage (CIE) coordinates were calculated using the SpectraWin software. All of the above procedures, except for the cleaning of the substrates, were carried out in two interconnected N_2-filled glove boxes ($[O_2] < 1$ ppm, $[H_2O] < 0.5$ ppm).

Results and Discussion

The emission from an efficient host-guest LEC should preferably originate solely from the minority guest, and the funnelling of emissive excitons to the guest can result from a variety of processes. Singlet excitons formed on the majority host can be non-radiatively transferred to the minority guest by Förster resonance energy transfer (FRET), and the efficiency of the FRET process is dependent on a good spectral overlap of the photoluminescence (PL) spectrum of the donor (*i.e.* the host) and the absorption spectrum of the acceptor (*i.e.* the guest), and on the distance between the host and the guest.[25] In order to investigate whether the first criterion is fulfilled in the host-guest systems under study, the PL spectra of the three hosts and the absorption spectrum of the guest are displayed in Fig. 2(a). All three host compounds feature a relatively broad PL band in the high-energy visible region, with the PL peak at 370 nm for CBP (solid blue squares), at 410 nm for PVK (open red circles), and at 450 nm for PSTB (solid green triangles). The Ir(R-piq)$_3$ guest (black line) displays strong absorption in the near-UV range, but also a pronounced absorption band in the visible region (peak at 455 nm); the former is assigned to a ligand-to-ligand π-π* transition and the latter to a metal-to-ligand charge-transfer (MLCT) transition.[26] Importantly, the PL spectrum of all three host compounds features a significant overlap with the absorption of the guest compound, thus fulfilling the spectral-overlap criterion for efficient FRET.

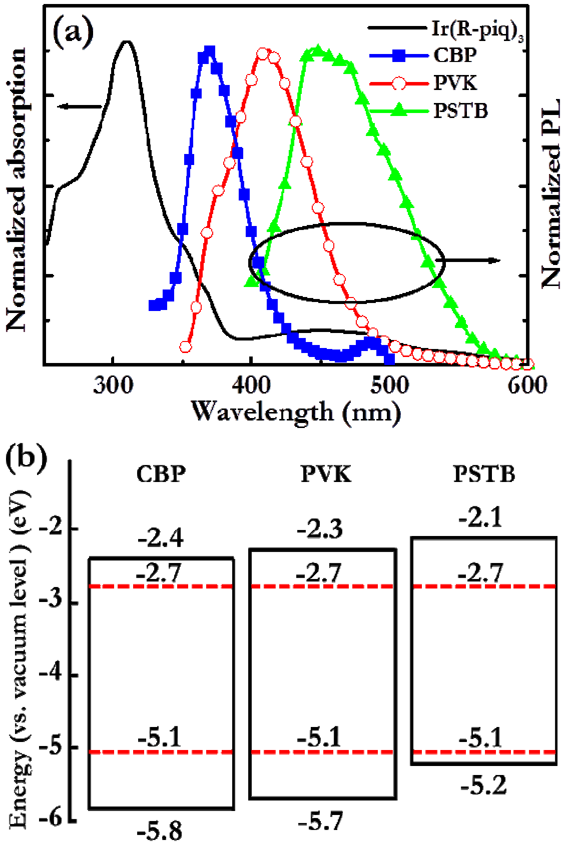

Figure 2. (a) The PL spectra of the three host materials, CBP, PVK and PSTB, and the absorption spectrum of the Ir(R-piq)$_3$ guest compound (black line). (b) The energy structure of the three host materials (solid black lines) and the guest compound (dashed red line).

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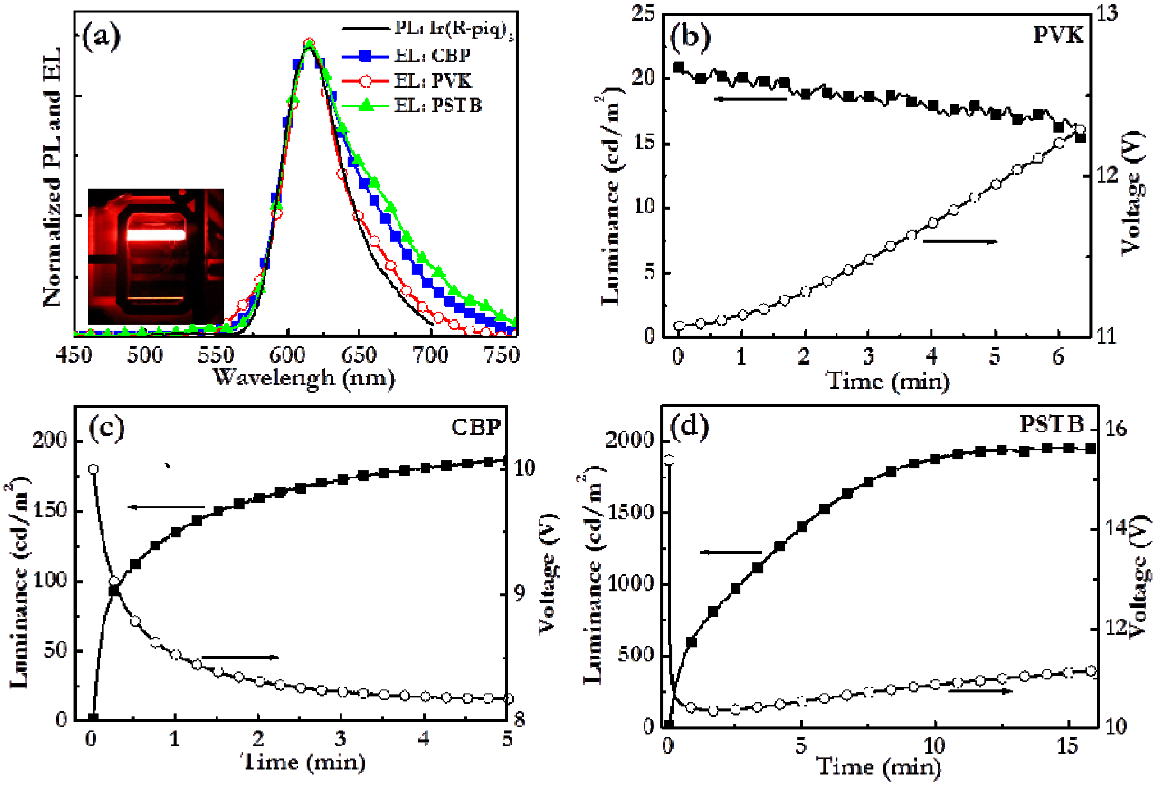

Figure 3. (a) The PL spectrum of the Ir(R-piq)$_3$ guest compound (black line), and the EL spectra from sandwich-cell LECs with the following configuration: ITO/PEDOT-PSS/Host:Ir(R-piq)$_3$:TMPE-OH:LiCF$_3$SO$_3$/Al, with the host being identified in the inset. Inset: Photograph of the red emission from a sandwich-cell LEC with host = PSTB. (b-d) The temporal response of the sandwich cells with the host being (b) PVK, (c) CBP, (d) PSTB. The devices were driven at *j* = 77 mA/cm^2.

Although the host and guest do not need to be in *direct* contact for FRET to take place, the efficiency of the process is still dependent on them being in close proximity as the transfer rate is inversely proportional to the sixth power of the host-guest distance. Moreover, the triplet excitons formed on the host are commonly transferred to the guest by the Dexter mechanism, and since this is a direct electron-exchange process, it depends on that the wave functions of the host and guest overlap and that the two species accordingly are in direct contact.[27] Finally, triplet-triplet annihilation can be a significant loss mechanism in triplet-emitting systems, but this reaction can be effectively suppressed if the low-energy triplet-emitting guests are isolated from each other within a higher-energy host matrix, so that the triplet exciton is locked upon one single guest molecule during its lifetime.

Thus, it is clear that a functional host-guest system is characterized by that the guest molecule is dispersed in the host matrix on the molecular level. We have therefore probed the surface morphology of 100-nm-thin active-material films comprising the different host-guest systems with AFM (see Fig. S1 in ESI), and we observe a smooth surface morphology, with a root-mean-square surface roughness of ≈ 0.5 nm, for all three host-guest system. This indicates that the host and guest are well blended, but more conclusive evidence is provided by the photoluminescence (PL) data presented in Fig. S2 in the ESI. We observe PL emission solely from the guest compound in all three systems, but note that the photoluminescence quantum yield (PLQY) is different, with the PLQY being highest for the PSTB host and lowest with the PVK host. This demonstrates that the host-guest energy transfer is efficient in all three systems, but that triplet-triplet quenching is most prominent with PVK as the host and least of a problem with PSTB. Our interpretation of these findings is that the guest is relatively well-dispersed in the host in all three systems, but that some intermolecular guest-guest interactions do exist, and that these are most of an issue with PVK as the host and least problematic with PSTB being the host.

The singlet and triplet excitons can also be formed directly on the guest compound via charge trapping, and the energy structures (with respect to the vacuum level, VL) of the host materials (solid black lines) and the guest compound (dashed red line), as established with CV and using the equation $E_{VL} = -e\cdot(4.8\ V + V_{Fc/Fc+})$, are presented in Fig. 2(b). It is notable that the LUMO level of the PVK host could not be measured with CV, and that its value therefore was estimated by adding its optical energy gap, as gleaned from an absorption measurement, to its measured HOMO level. Importantly, as both the HOMO and LUMO levels of the guest compound are

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positioned within the energy gap of all three host compounds, it is possible for an exciton to form on the guest compound through both hole and electron trapping.

The overall efficiency of the host-to-guest energy transfer, independent on whether it originates in FRET, Dexter transfer, or charge trapping, can conveniently be evaluated by measuring the electroluminescence (EL) spectrum from a device based on the system-under-study. Fig. 3(a) presents the normalized EL spectrum of ITO/PEDOT-PSS/{Host:Ir(R-piq)$_3$:TMPE-OH:LiCF$_3$SO$_3$}/Al LEC sandwich cells, with the host being CBP (solid blue squares), PVK (open red circles), or PSTB (solid green triangles). All three devices display similar EL spectra, with a slight variation in the magnitude of the low-energy tail. The emission peak is invariably positioned at 615 nm and the CIE coordinates are (0.65, 0.33), *i.e.,* very close to the standard red point of (0.66, 0.33), as defined by National Television System Committee (NTSC). A photograph of the red emission from the device with host = PSTB is displayed in the inset of Fig. 3(a). Notably these EL spectra are highly reminiscent of the PL spectrum of the Ir(R-piq)$_3$ guest (black line), and distinctly different from the PL spectrum of the corresponding host (see Fig. 2a). Thus, it is clear that the energy transfer from the host to the guest is effectively complete in all three systems, which further supports the notion that the guest is relatively well-dispersed in the host matrix.

Although all three host-guest systems exhibit efficient energy transfer, other qualitative and quantitative measures of the device performance deviate distinctly. The device with PVK as the host material, as presented in Fig. 3(b), feature a low peak luminance of 21 cd/m^2 and a modest maximum current efficacy (CE) of 0.027 cd/A during galvanostatic driving at a current density of j = 77 mA/cm^2. Moreover, the device turn-on to peak luminance is “instant” and the observed temporal effects are a decreasing luminance and an increasing voltage. The latter is in sharp contrast with the situation in a functional LEC device, where the voltage is decreasing and the luminance is increasing during the *in-situ* electrochemical doping and p-n junction formation process. The device with CBP as the host does in contrast display these characteristic signs of LEC operation (see Fig. 3c), and both the peak luminance and CE at the same current density are also distinctly improved to 203 cd/m^2 and 0.3 cd/A, respectively. The best performance was however attained with the host being PSTB. This device also displayed the characteristic LEC turn-on behavior and the peak luminance and maximum CE reached 1950 cd/m^2 and 2.5 cd/A, respectively, at j = 77 mA/cm^2. The latter host-guest LEC was also probed at a lower current density of j = 5.8 mA/cm^2 (see Fig. S4a), at which it featured a peak luminance of 198 cd/m^2 and a maximum CE of 3.5 cd/A. The latter corresponds to an external quantum efficiency of 2.7 %. This system is further preferable in that it features a fast turn-on to significant brightness. If we define the turn-on time as the time to reach 100 cd/m^2 at j = 77 mA/cm, a pristine CBP device takes 22 s to turn on whereas the PSTB device is much faster at 6 s. With this definition the PVK device never turned on, as it never reached the required luminance. The device performance data are summarized in Table 1. The PSTB based LEC also featured a longer operational lifetime than the PVK and CBP based LECs (see Fig. S3), although it should be noted that the stability at this point is far below that of host-free state-of-the-art LECs, which can feature operational lifetimes of several thousand hours.[28-31] We tentatively attribute the limited stability of the herein presented host-guest LECs to the high drive voltage, which we anticipate will result in detrimental electrochemical side reactions.

We have also tested the PVK host with another phosphorescent guest in the form of the green-emitting tris(2-phenylpyridine)iridium (III) Ir(ppy)$_3$. It is well-established that the PVK/Ir(ppy)$_3$ system can feature a very high efficiency in OLED devices,[32] but as an LEC material it does not function well. We again measure very modest device characteristics as shown in Fig. S4(b) in the ESI, and importantly we do not observe the characteristic signs of electrochemical doping in the form of increasing luminance or decreasing voltage during galvanostatic driving.

Table 1. Device performance of sandwich-cell host-guest LECs.

Host	Applied current density (mA/cm^2)	Turn-on time (≥ 100 cd/m^2) (s)	CE (@cd/m^2) (cd/A)	PCE (lm/W)	EQE (%)
PVK	77	NA	0.027 (21)	0.008	0.02
CBP	77	22	0.3 (203)	0.1	0.23
PSTB	77	6	2.5 (1950)	0.7	1.9
PSTB	5.8	1500	3.5 (198)	1.4	2.7

In order to investigate and understand why the host materials perform so differently in LEC devices, we have carried out two sets of experiments aimed at establishing the capacity of the host for electrochemical doping. In Fig. 4(a) we present CV traces measured with thin host films as the active material, and in Figs. 4(b-c) we show selected results from a study on open planar LEC devices, with the host material being the electroactive compound. We find that the two “functional” LEC host materials, CBP and PSTB, exhibit reversible or semi-reversible oxidation and reduction processes in CV (*i.e.* a matching back reaction when the scan direction is reversed, with a similar magnitude for the peak current and integrated-charge as in the forward reaction), whereas PVK only features signs of reversibility during oxidation. This implies that it is possible to electrochemically dope CBP and PSTB both p- and n-type, whereas PVK only can be electrochemically doped p-type.

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Further support for this conclusion is provided by the study on open planar LEC devices in a bilayer configuration; see inset in Fig. 4(b) for the schematic device structure.[33] It is well-established that p-type/n-type electrochemical doping of a conjugated compound results in an increase of the conductivity through an increase in the concentration (and mobility) of the holes/electrons.[34] As the current through the bilayer device presented in Fig. 4(b) is observed to increase with time for both the PSTB and CBP based devices, the conclusion is that the conductivity of the host material is increasing with time, and that it accordingly is doped p-type at the anode and n-type at the cathode. The PVK-based device, in contrast, displays a comparatively low and constant current with time, and we attribute this to that the conductivity-enhancing p-type doping event at the anode is balanced by a side-reaction event at the cathode, which produces side products that significantly lowers the current-carrying capacity of the host at the cathode.

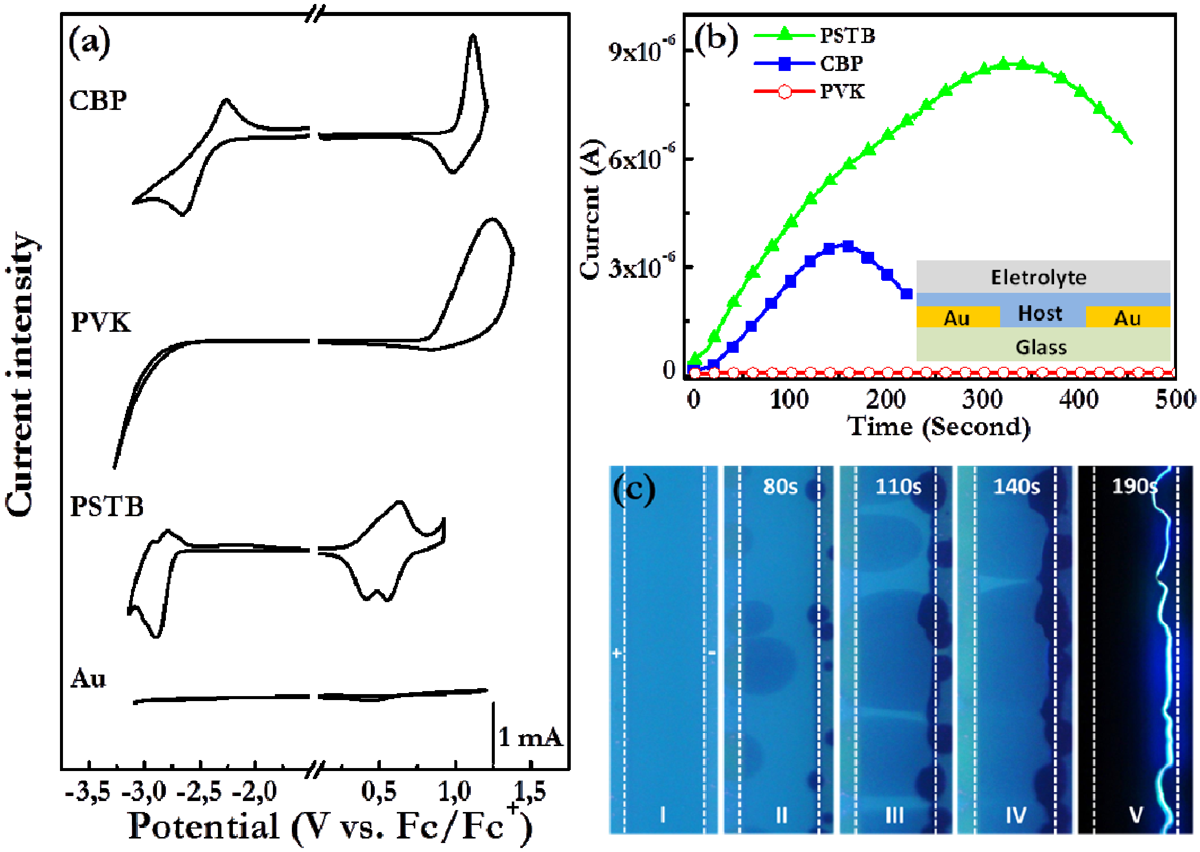

Figure 4. (a) CV traces recorded with a thin film of the host compound as the active material coated on a Au electrode. The electrolyte was 0.1 M TBAPF$_6$ in CH$_3$CN, the counter electrode a Pt rod, and the quasi-reference electrode a Ag wire. The lowest trace was carried out on a bare Au working electrode. (b) The current vs. time recorded for bilayer surface cells, comprising PSTB, CBP, and PVK as the active material. The devices were driven at $V = 5$ V at $T = 360$ K. (c) A time-sequence of photographs of the doping-front propagation and subsequent light emission from a PSTB bi-layer surface cell.

A more visual evidence for the appearance of balanced electrochemical doping in the functional host materials is provided by the simultaneously recorded set of time-lapse photographs of the PSTB based planar device, as displayed in Fig. 4(c). The device was operating in a dark room under UV illumination, because the UV-light will excite the (blue) PL of PSTB, and since the doping process is known to effectively quench the PL.[35] In other words, the dark regions with quenched PL that are observed to form and grow away from the electrode interfaces in photographs II-IV reveal the formation and growth of a p-type doped region at the positive anode and an n-type doped region at the negative cathode. When the two doped regions have made contact in the last photograph V, a light-emitting p-n junction is observed at the meeting point.[36-38]

Finally, we note that the two functional host materials produce rather different performance in the sandwich-cell LECs, with PSTB featuring a better efficiency than CBP (see Table 1 and Figs. 3c and 3d), despite the employment of the same triplet emitter and the energy-transfer process being highly efficient in both systems (see Fig. 2). We tentatively attribute this deviating performance to a combination of two factors: (i) triplet-triplet quenching, which is found to be more prominent

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in the CBP system (see Fig. S2 and related discussion). (ii) The higher HOMO level of PSTB (see Fig. S5), which we anticipate will better suppress undesired electrochemical side reactions involving the electrolyte at the anodic interface.[39,40]

Conclusions

To summarize, in order to develop the LEC technology for high-end applications, it is important to realize devices that can emit with good efficiency at high luminance levels. One potentially viable solution to this end, which has been successfully implemented in OLEDs, involves the utilization of a host-guest system in the active material. Here, we demonstrate that, in addition to the well-established baseline criteria established during OLED development, it is fundamental that the host material in a functional host-guest LEC can allow for both electrochemical p- and n-type doping. This critical electrochemical capacity can be conveniently established by cyclic voltammetry or through studies of open planar devices. We have identified two different host materials that fulfil this electrochemical-doping criterion, and report on a red-emitting LEC device, comprising a copolymer host and a triplet-emitting Ir complex guest, which features a high external quantum efficiency of 1.9 % at a luminance of 2000 cd/m^2.

Acknowledgements

This project is financially supported by the Swedish Foundation for Strategic Research, the Swedish Research Council, the Swedish Energy Agency, Kempestiftelserna, Umeå University, and the Knut and Alice Wallenberg foundation under contract KAW 2012.0083.

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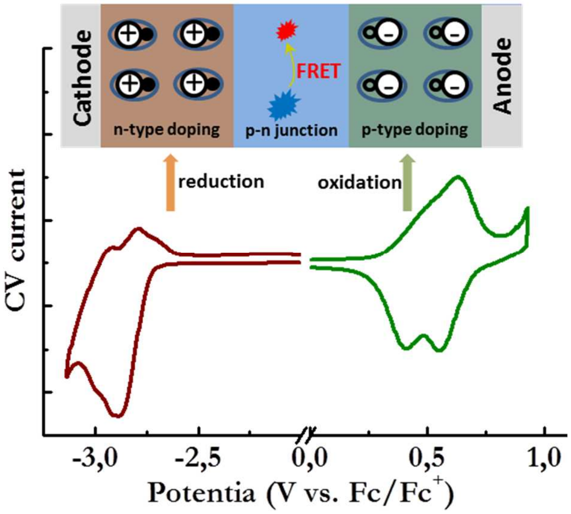
TOC graphic:

Reversible electrochemical doping capacity is a fundamental criterion for an efficient and functional host-guest LEC.



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