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Efficient hybrid white organic light-emitting diodes with extremely long lifetime: the effect of n-type interlayer

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The effect of n-type interlayer in hybrid white organic light-emitting diodes (WOLEDs) has been systematically investigated by using various n-type materials. A new finding, that the triplet energy rather than electron mobility or hole-blocking ability of interlayer plays a more positive role in the performance of hybrid WOLEDs, is demonstrated. Based on the new finding, a more efficient n-type interlayer bis[2-(2-hydroxyphenyl)-pyridine] beryllium has been employed to realize a high-performance hybrid WOLED. The resulting device (without n-doping technology) exhibits low voltages (i.e., 2.8 V for 1 cd/m^2, 3.9 V for 100 cd/m^2) and low efficiency roll-off (i.e., 11.5 cd/A at 100 cd/m^2 and 11.2 cd/A at 1000 cd/m^2). At the display-relevant luminance of 100 cd/m^2, a total power efficiency of 16.0 lm/W, a color rendering index of 73 and an extremely long lifetime of 12596265 h are obtained. Such superior results not only comprehensively indicate that the n-type materials are effective interlayers to develop high-performance hybrid WOLEDs but also demonstrate a significant step towards real commercialization in WOLEDs.

White organic light-emitting diodes (WOLEDs) are now approaching mainstream display markets and also being aggressively explored for the next-generation lighting applications due to their extraordinary characteristics, such as high efficiency, fast switching and flexibility[1–7]. Generally, three types of WOLEDs are created according to the employed emitting materials, including all-phosphorescent WOLEDs, all-fluorescent WOLEDs and hybrid WOLEDs which are based on hybrid (fluorescent (F) and phosphorescent (P)) emitters schemes[8–10]. Among them, the utilization of P emitters is desirable since they can allow for a conversion of up to 100% of injected charges into emitted photons (both singlet and triplet excitons are harvested), resulting in a theoretical internal quantum efficiency of unity[1]. However, no proper blue P material can be obtained in terms of lifetime and color-stability until now, limiting the development of all-phosphor devices[11]. To loosen this bottleneck, researchers have devoted their attention to pursuing hybrid WOLEDs, which combine F blue emitters with P green-red/orange emitters to furnish white emission, due to their merits, high efficiency, stable color and long lifetime[8].

For hybrid WOLEDs, the exchange energy losses (0.5–1.0 eV) originating from intersystem crossing from the host singlet into a blue phosphor triplet state are eliminated by dint of a blue fluorophore, improving the efficiency[8]. Meanwhile, with the smart device engineering, both singlet and triplet excitons are harvested along independent channels and thus nearly resonant energy transfer (ET) from the conductive host to dopants for both singlet and triplet energy (T_1) can be realized[8]. In the last few years, the efficiency of hybrid WOLEDs has experienced a step-by-step increase and now can exhibit a maximum power efficiency (PE) of 58.4 lm/W[12], which has already exceeded some of the best all-phosphorescent devices[3]. Besides, the color-stability limitations exist in all-phosphorescent WOLEDs has been greatly overcome by effective hybrid WOLEDs structures, such as using bipolar interlayer (IL) switch to manage carrier charges balance[13,14], adopting multifunctional dopants to reduce charges mobility[15] and preparing ultrathin emitting layers (EMLs) to produce white light[16]. However, while it is usually considered that hybrid WOLEDs have the potential of long lifetime since they comprise F blue emitters, no hybrid WOLED with long lifetime has been reported in the revealed literature so far, which also limits the further development of hybrid WOLEDs. To alleviate this difficulty, it is undoubtedly that an urgent endeavor is needed, although only rare effort has been taken to the lifetime of hybrid WOLEDs.

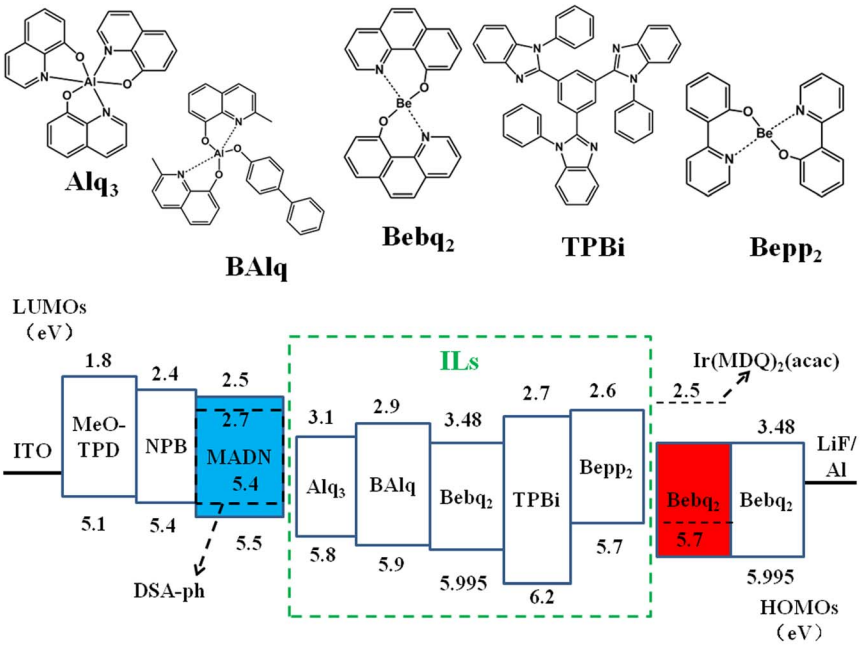

Figure 1 | Top: Chemical structure of ILs. Bottom: Proposed energy-level diagram of hybrid WOLEDs.

On the other hand, it is noted that a key feature of designing hybrid WOLEDs is the use of suitable ILs, locating between the F emitter and the P emitter[8,12–20]. ILs can not only prevent Förster ET from the F blue emitters to the red-green/orange P emitters, but also eliminate the nonradiative Dexter ET between the two layers[8]. As a result, both singlet and triplet excitons are managed via ILs. In fact, a large number of effective ILs have been reported, such as 4,4-N,N-dicarbazolebiphenyl (CBP)[8,12], TCTA: bis[2-(2-hydroxyphenyl)-pyridine] beryllium ($Bepp_2$)[13,14], N,N′-di(naphthalene-1-yl)-N, N′-diphenyl-benzidine (NPB)[15,16] and TCTA: 2,2′,2″-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi)[17,18]. However, it is noted that most of publications are focused on the bipolar or p-type ILs, only negligible attention was paid on the n-type ILs[19,20]. Moreover, no detailed investigation has been documented on the effect of n-type ILs in hybrid WOLEDs.

In this paper, we have systematically investigated the influence of n-type ILs by using four n-type materials, including tris(8-hydroxyquinoline) aluminum (Alq_3), bis-(2-methyl-8-quinolinolate)-4-(phenylphenolato)aluminum (BAlq), bis(10-hydroxybenzo[h] quinolinato)beryllium complex ($Bebq_2$) and TPBi. It is demonstrated a new finding that the T_1 rather than electron mobility or hole-blocking ability of n-type ILs plays the crucial role in device performances. Based on the new finding, a more efficient n-type IL $Bepp_2$ is employed to realize a high-performance hybrid WOLED. The resultant device (without n-doping technology) exhibits low voltages (i.e., 2.8 V for 1 cd/m^2, 3.9 V for 100 cd/m^2) and low efficiency roll-off (i.e., 11.5 cd/A at 100 cd/m^2 and 11.2 cd/A at 1000 cd/m^2). At the display-relevant luminance of 100 cd/m^2, a total PE of 16.0 lm/W, a color rendering index (CRI) of 73 and an extremely long lifetime of 12596265 h are obtained. Such superior results not only comprehensively indicate that the n-type materials are decent ILs to develop high-performance hybrid WOLEDs but also demonstrate a significant step towards real commercialization in WOLEDs.

Results and discussion

Figure 1 depicts the configuration of WOLEDs: ITO/MeO-TPD: F4-TCNQ (100 nm, 4%)/NPB (20 nm)/MADN: DSA-ph(20 nm, 7%)/ILs (0 or 3 nm)/$Bebq_2$: $Ir(MDQ)_2(acac)$ (9 nm, 5%)/$Bebq_2$ (25 nm)/LiF (1 nm)/Al (200 nm), where ITO is indium tin oxide, F4-TCNQ is tetrafluoro-tetracyanoqino dimethane, MeO-TPD is N, N, N′,N′-tetrakis(4-methoxyphenyl)-benzidine, DSA-ph is p-bis(p-N,N-diphenyl-aminostyryl) benzene, MADN is 2-methyl-9,10-di(2-naphthyl)anthracene and $Ir(MDQ)_2(acac)$ is iridium(III)bis(2-methyldibenzo[f,h]quinoxaline)(acetylacetonate). Device W1, W2, W3, W4, W5 and W6 correspond to no, Alq_3, BAlq, $Bebq_2$, TPBi and $Bepp_2$ IL, respectively.

To appropriately investigate the effects of n-type ILs in hybrid WOLEDs, several strategies are employed. First, to simplify structures, we utilized two complementary colors (blue/red) to give off white emission, although no two-color (blue/red) hybrid WOLEDs with n-type ILs has been reported. However, it should be pointed out that blue-orange color complement or three color system can also be used to study the effect of n-type ILs, which is expected to achieve similar results. Besides, it is noted that the first successful hybrid WOLEDs needs somewhat complicated structures which comprise the stacked F-IL-P-P-IL-F emitters[8], we use the P-IL-F structures which only need double EMLs (one F EML and one P EML), leading to simplified fabrication processes and reduced cost. Next, $Bebq_2$ is selected as an electron transport layer (ETL) since its electron mobility is as high as $\sim 10^{-4}$ cm^2/(V s)[21], which can effectively enhance the electron injection and then make the carriers balanced. $Bebq_2$ is also used as the host of red emitter, which can not only prevent reverse energy transfer from the dopants to the host as well as confining triplet excitons in the EML since the T_1 of $Bebq_2$ (2.25 eV)[22] is higher than that of $Ir(MDQ)_2(acac)$ (2.0 eV)[13], but also eliminate the structural heterogeneity exist between ETL and EML, boosting the efficiency and lifetime[23]. Finally, since the triplet ET (Dexter transfer) requires spatial overlap of the donor/accepter molecular orbitals and only occur significantly within a range of 1–2 nm[17], 3 nm thick ILs are used to prevent this ET. Whereas, device A without IL is developed for comparison, which is expected to deeply understand the effect of n-type ILs. Based on these considerations, we studied the impact of n-type ILs in hybrid WOLEDs.

As illustrated in figure 2a, upon using different ILs, the electroluminescent (EL) spectra are dramatically affected. At a typical current density of 20 mA/cm^2, the CIE coordinates of device W1, W2, W3, W4 and W5 are (0.194, 0.312), (0.194, 0.313), (0.197, 0.327), (0.194, 0.335) and (0.232, 0.323), respectively. An obvious red emission is generated in W5 while others only show negligible red colors. Hence, we cannot classify device W1, W2, W3 and W4 into

WOLEDs because their colors are far away from the white equivalent-energy point of (0.333, 0.333).

Reasons for the above phenomena can be explained as follows. In general, the properties of n-type materials are affected by three key factors in phosphor-based devices, including electron mobility, hole blocking ability and T_1[1,2]. On one hand, since the electron mobilities of Alq_3, BAlq, $Bebq_2$ and TPBi are 10^{-5} $cm^2/(V\,s)$[21], 10^{-5} $cm^2/(V\,s)$[24], 10^{-4} $cm^2/(V\,s)$ and 10^{-4} $cm^2/(V\,s)$[25], respectively, electrons may be easier to pass through the $Bebq_2$ and TPBi IL than Alq_3 and BAlq IL, which leads to much more blue intensity in W4 and W5 than W2 and W3, and stronger red emissions are expected in W2 and W3. On the other hand, since the HOMOs of Alq_3, BAlq, $Bebq_2$ and TPBi are 5.8 eV[21], 5.9 eV[24], 5.995 eV[26] and 6.2 eV[25], respectively, holes may be difficult to pass through the TPBi IL than Alq_3, BAlq and $Bebq_2$ IL, which results in much more red intensity in W2, W3 and W4 than W5. Considering these two factors, device W2, W3 and W4 should exhibit more balanced white emissions than W5. However, contrary to the above reasons, white light can only be observed in W5 and others show negligible red emission. Therefore, it cannot be concluded that the electron mobility and hole blocking ability of the n-type IL are the main factors that affect the device performance. In fact, the T_1, which usually has a great influence on the performance of phosphor-based devices[1], is attributed to the main reason for the above phenomena. Since the triplet energies of $Bebq_2$, Alq_3, BAlq and TPBi are 2.25 eV, 2.03 eV[27], 2.18 eV[27] and 2.74 eV[25], respectively, the low triplet energies of Alq_3 and BAlq IL cannot prevent the diffusion of high-energy triplet excitons which generated in the red emissive regions, leading these excitons to the nonradiative triplet state of ILs and blue EMLs in W2 and W3[17]. W1 without IL cannot eliminate the influence of Dexter ET between the F emitter and the P emitter because Dexter ET can occur within 1 ~ 2 nm, leading to a negligible red emission and hence we cannot obtain a balanced white color[8]. For W4 with the $Bebq_2$ IL, the generated triplet excitons in the red region can still be easily transfer to the blue region due to the similar T_1 of the $Bebq_2$ IL and $Bebq_2$ host, resulting in exciton quenching[17]. However, the mutual quenching can be effectively omitted by introducing the 3 nm TPBi IL owing to the high T_1 (2.74 eV), and hence white light can be generated in W5. Besides, another interesting phenomenon is observed that the thicker TPBi IL is, the less red emission is, as shown in figure 2 inset. The phenomenon can be attributed to the fact that holes are difficult to pass through this IL bridge with increasing thickness (4.5 nm, 6 nm) since the TPBi is an n-type material. However, compared with the conventional work which used complicated methods to adjust the color of OLEDs, it is demonstrated that the color can be tuned by simply changing the n-type IL thickness, indicating a simplified but effective approach[28].

Although W5 exhibits a white emission, its efficiency is not high, as shown in figure 3. The maximum forward-viewing CE and PE are 9.8 cd/A and 10.6 lm/W at 1 cd/m^2, respectively. At 100 cd/m^2, the CE and PE are 8.5 cd/A and 6.5 lm/W, respectively. To verify the new finding that the T_1 rather than electron mobility or hole-blocking ability of n-type ILs plays the crucial role in device performances and to further improve the properties of WOLEDs with n-type ILs, we explored another n-type material $Bepp_2$ as the IL because of its high T_1 (2.6 eV)[14], which can also be expected to avoid the mutual quenching between the F and P emitter. The efficiency of W6 with the $Bepp_2$ IL is remarkably enhanced, as shown in figure 3. The maximum forward-viewing PE of W6 is 12.3 lm/W at 8 cd/m^2. As illumination sources are typically characterized by their total emitted power[8], our device exhibits a maximum total PE of 20.9 lm/W, which remains 16.0 lm/W and 11.9 lm/W at 100 cd/m^2 and 1000 cd/m^2, respectively, higher than the previous hybrid WOLEDs with n-type ILs[19,20]. The enhanced efficiency of W6 may be explained as follows. Although the electron mobility of $Bepp_2$ is similar to TPBi[8], the HOMO of $Bepp_2$ (5.7 eV) is lower than that of TPBi (6.2 eV), which indicates that holes can be more easily transported into the red EML to form excitons and then harvested by $Ir(MDQ)_2(acac)$ which can manage both singlet and triplet excitons, enhancing the efficiency. More importantly, it is noted that the efficiency roll-off is rather low. For example, a maximum forward-viewing CE of 12.1 cd/A (a total CE of 20.6 cd/A) is achived at 8 cd/m^2, which slightly rolls off to 11.5 cd/A at 100 cd/m^2 and 11.2 cd/A at 1000 cd/m^2.

Figure 4a presents the luminance-voltage-current density characteristics of W6, a turn-on voltage is 2.8 V (defined as the voltage at 1 cd/m^2, 0.009 mA/cm^2), which is the lowest value in the P-IL-F based WOLEDs using doped EMLs (without p-i-n or outcoupling structures) in the literature. At 100 cd/m^2, the applied voltage is 3.9 V (0.919 mA/cm^2) and a luminance of 58697 cd/m^2 is obtained at 10.3 V (926.9 mA/cm^2), higher than the previous WOLEDs with n-type ILs[19,20]. Since it is still a challenge for WOLEDs to achieve low driving voltages for practical use (e.g., <3 V for onset and <4 V at 100 cd/m^2 for portable display)[29], our device can alleviate this difficulty. Besides, the normalized EL spectrum is shown in figure 4a inset, a CIE coordinates of (0.277, 0.354) is obtained at 100 cd/m^2, corresponding to a CRI of 73, which is the highest value in two-color (blue/red) WOLEDs with P-IL-F structures.

Finally, since the device lifetime is essential to practical use[30–32], we have measured the lifetime of W6. As shown in figure 4b, a half

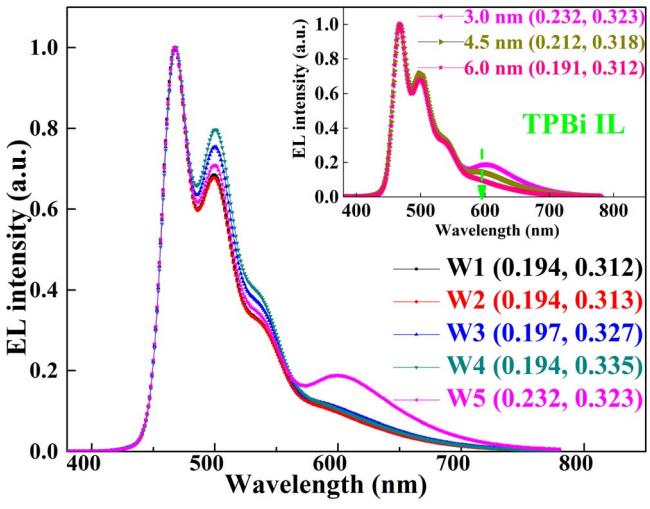

Figure 2 | (a) EL spectra of devices with different ILs at 20 mA/cm^2, Inset: EL spectra of devices with different TPBi IL thickness at 20 mA/cm^2.

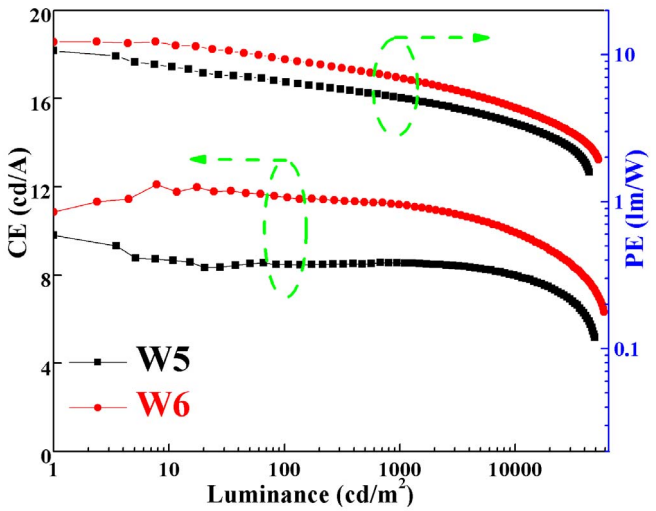

Figure 3 | CE and PE of W5 and W6 as a function of luminance.

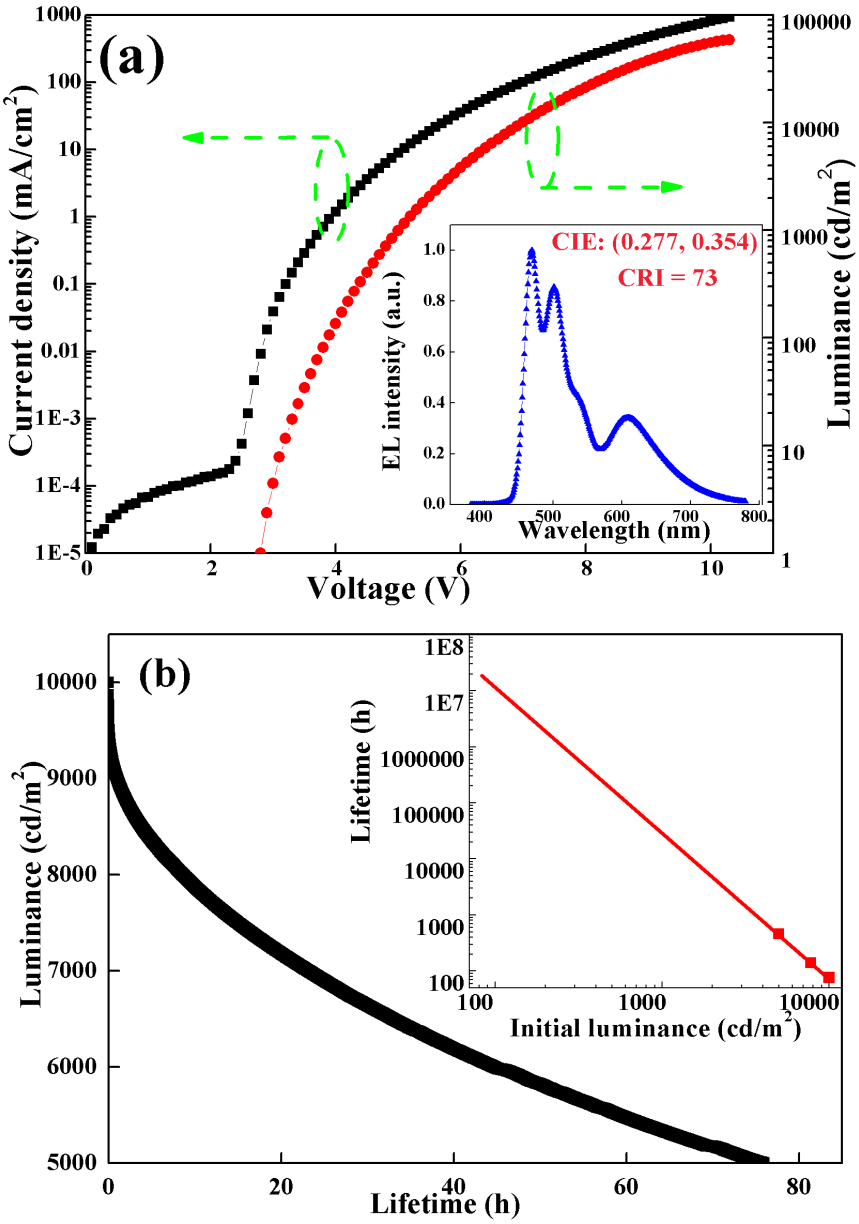

Figure 4 | (a) The luminance-voltage-current density characteristics of W6; inset: the spectrum. (b) Luminance decay curve; inset: the lifetime versus initial luminance relationship.

lifetime of 75.9 h at an initial luminance of 10000 cd/m^2 is obtained. To determine whether or not the lifetime is suitable for use in display luminance, an acceleration factor of 2.61 is obtained by measuring half-lifetimes at various initial luminances. Therefore, the lifetime is extrapolated to be 12596265 h at an initial luminance of 100 cd/m^2[33], as shown in figure 4b inset. Even at an initial luminance of 1000 cd/m^2, the lifetime is as long as 30920 h. To the best of our knowledge, this is the first hybrid WOLED with long lifetime in the revealed literature so far. Since the half lifetimes for TVs applications must be about 100000 h[30], it is obviously seen that our device can satisfy this practical demand. On one hand, although all-fluorescent WOLEDs can exhibit long lifetime over 150000 h at 1000 cd/m^2, the maximum efficiency is only 14.7 cd/A because the triplet excitons cannot be harvested[34]. On the other hand, although all-phosphorescent WOLEDs can exhibit fluorescent tube efficiency, the lifetime should be poor due to the P blue emitter[35]. However, since hybrid WOLEDs can manage both singlet and triplet excitons together with the fact that stable F blue emitters can be obtained, the trade-off between lifetime (12596265 h) and efficiency (20.6 cd/A and 20.9 lm/W) are realized, demonstrating a significant breakthrough in WOLEDs. Previous works have reported that hybrid WOLEDs can achieve high efficiency[12], stable color[13–16], herein, we have reported that hybrid WOLEDs can realize long lifetime, indicating an important step towards real commercialization.

In summary, we have investigated the effects of n-type ILs in hybrid WOLEDs and found that the T_1 rather than electron mobility or hole-blocking ability plays the critical role in device performances. Intrigued by the achieved results, we have used $Bepp_2$ as a more effective IL to obtain a high-performance hybrid WOLED. The resulting device can exhibit low voltage (2.8 V), high efficiency (16.0 lm/W), high CRI (73) and long lifetime (12596265 h) at a practical luminance of 100 cd/m^2. Such presented facts will be beneficial to the design of both materials and device structures for WOLEDs in the emerging display and lighting applications.

Methods

All material layers were thermally deposited without breaking the vacuum at a base pressure of 2×10^{-7} Torr. In the deposition of the doping layers, deposition rates of

both host and guest were controlled by their correspondingly independent quartz crystal oscillators. The devices were encapsulated immediately after preparation under a nitrogen atmosphere using epoxy glue and glass lids. The EL spectra, CIE color coordinates and CRI of packaged devices were obtained by a **Konica Minolta CS2000** spectra system. The emission area of the devices is 3×3 mm^2 as defined by the overlapping area of the anode and cathode. The luminance-current density-voltage characteristics were recorded simultaneously, using a computer-controlled source meter (Keithley 2400) and multimeter (Keithley 2000) with a calibrated silicon photodiode. The lifetime of the devices was tested through a 512 channel OLED testing system (New Vision Opto-Electronic Technology Co., Ltd) at a constant direct current density. All the measurements were carried out at room temperature under ambient conditions.

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Author contributions

B.L. conceived the idea and wrote the paper, L.W., M.X., H.T., J.Z. and L.L. performed OLED device studies, data analysis, and provided detailed insights into the device engineering, H.N. made a detailed discussion and gave some suggestions for revising the manuscript, D.G. performed devices fabrication, J.P. and Y.C. initiated and supervised the project. All authors discussed the results and reviewed the manuscript.

Additional information

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