

Pure White Organic Light-Emitting Diode with Lifetime Approaching the Longevity of Yellow Emitter

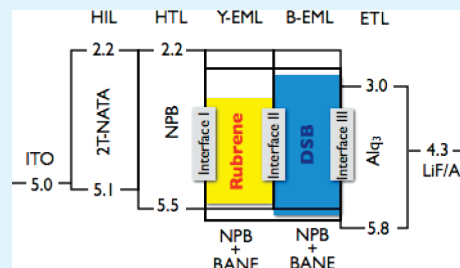
Jwo-Huei Jou,^{*,†} Shih-Ming Shen,[†] and Yung-Cheng Tsai[‡]

[†]Department of Materials Science and Engineering, National Tsing Hua University, Hsin-Chu, Taiwan 30013, Republic of China

[‡]Tetrahedron Technology Corporation, Chu-nan, Taiwan 35059, Republic of China

ABSTRACT: A high-efficiency pure white organic light-emitting diode was fabricated with lifetime approaching that of the low-excitation-energy (yellow) emitter containing counterpart, or six times that of the deep-blue counterpart. The white device was composed of two emission layers with mixed hosts of different compositions. They were respectively doped with yellow rubrene and deep-blue 4,4'-bis-[4-{N,N,N',N'-tetrakis-(4-fluoro-diphenylamino)-phenyl}-vinyl]-biphenyl. The resulting efficiency was 6.0 lm/W (12.4 cd/A) at 20 mA/cm². The long device lifetime may be attributed to the double mixed-host architecture employed that effectively dispersed the injected carriers into three different recombination zones and consequently diluted the damaging effect arising from the accumulated charge from un-recombined carriers, hence leading to a markedly improved lifespan.

KEYWORDS: high-efficiency, pure white, organic light-emitting diode, mixed-host, long lifetime, recombination zones



1. INTRODUCTION

Organic light-emitting diodes (OLEDs), particularly white OLEDs, have attracted increasing attention because of their potential advantages as flat-panel displays and for solid-state lighting applications.^{1–4} Despite significant progress in improving the efficiency of OLEDs in recent years (Figure 1),^{5–29} their operating lifetime remains a fundamental problem that limits commercial applications, such as televisions and lighting panels. Numerous approaches for improving the lifetime of monochromatic OLEDs have been proposed.^{30–33} These include the use of an aggregative additive, a mixed host, or a gradient host to expand the emission zone and dilute the degradation products.^{34–37} Increasing the morphological stability of the materials used prevents the damage caused by Joule heating.^{38,39} Introducing a hole-injection layer (HIL) or modifying the hole-transport layer (HTL) with a trace amount of hole-trapping dopant reduces the transport of holes to the electron-transport layer (ETL), thus preventing the formation of undesired cationic species.^{23,40} For white OLEDs, only limited studies on prolonging their lifetime as well as improving their efficiency have been reported.^{41–43} Moreover, little is known regarding the dependency of a white OLED's lifetime on its constituent blue and yellow or red, green, and blue emitters.

Typically, the lifetime of a white OLED is greatly limited by the lifetime of its high-excitation-energy component, such as blue color, which is the shortest among all emitters.⁴⁴ As a result, the lifetime of a white OLED is usually much shorter than that of its low-excitation-energy constituent of longer lifetime, such as the yellow or red emitter. For the previously reported long-lifetime, high-efficiency white OLED, for example, its lifetime was only 70% that of its counterpart yellow device.⁴² Hence, it is crucial to devise a white OLED with a lifetime closely approaching that of the constituent yellow or red emitters.

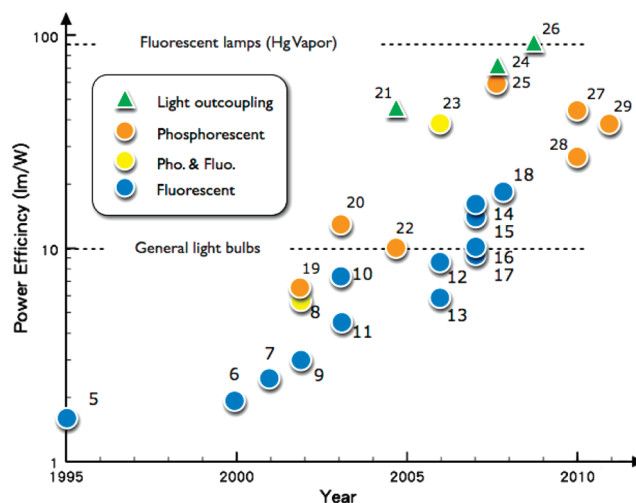


Figure 1. Latest efficacy records of white OLEDs since 1995. Remarkable progress has been made upon using innovative device architectures and light outcoupling techniques as well as highly electroluminescence active dyes, especially those of electrophosphorescence.

Here, we report a high-efficiency white OLED with a long lifetime, approaching the longevity of the counterpart yellow device or six times that of the deep-blue one. The white device comprises two complementary colors in double emissive layers. The first one emits yellow light obtained by doping 1 wt.% rubrene in a mixed host consisting of 50 wt % N,N'-diphenyl-N, N'-bis-(1-naphthyl)-1,1'-biphenyl-4,4'-diamine (NPB) and 50 wt.%

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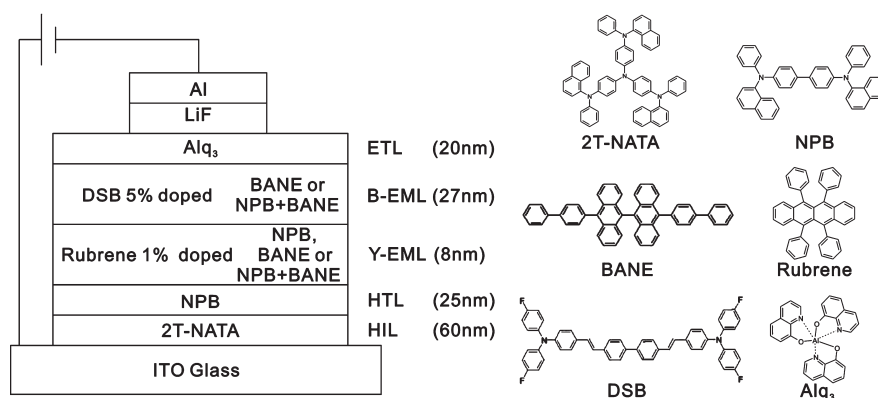


Figure 2. Schematic illustrations of the structures of the white OLEDs and the organic molecules studied.

10,10'-bis-biphenyl-4-yl-[9,9']bianthracenyl (BANE). The second layer emits deep-blue light by doping 5 wt.% 4,4'-bis-[4-{N,N,N',N'-tetrakis-(4-fluoro-diphenylamino)-phenyl}-vinyl]-biphenyl (DSB) (10) in a mixed host consisting of 5 wt.% NPB and 95 wt.% BANE. Besides its long lifetime, the resultant device exhibits a high efficiency, 6.0 lm/W and 12.4 cd/A at 20 mA/cm², emits a pure white light, Commission International de L'Eclairage (CIE) coordinates (0.308, 0.322), and has excellent color stability, the chromatic variation being (0.002, 0.002) for brightnesses between 100 and 10 000 cd/m².

2. EXPERIMENTAL METHODS

Device Fabrication. Figure 2 shows the device architectures of the white OLEDs studied and the chemical structures of the organic materials used. Each device comprises 10 Ω/sq indium tin oxide glass, a 60 nm HIL of 4,4',4''-tris(N-(2-naphthyl)-N-phenylamino)-triphenylamine, a 25 nm HTL of NPB, an 8 nm yellow emissive layer (Y-EML), a 27 nm blue emissive layer (B-EML), a 20 nm ETL of tris-(8-hydroxyquinoline) aluminum (Alq₃), a 1 nm lithium fluoride layer, and a 150 nm thick aluminum cathode. Five white devices with different host systems for the yellow/blue emissive layers, such as NPB/BANE or BANE/BANE for white device I or II, respectively, were investigated. A 50 wt % BANE and 50 wt % NPB mixed host is introduced as the yellow emissive layer in white device III. Furthermore, in white device IV-1 or IV-2 with double mixed-host layers, 5 wt % or 10 wt % NPB is mixed with BANE as the blue emissive layer, respectively. Two monochromatic devices, blue and yellow, were also prepared for comparison. The blue device was composed of a 35 nm single emissive layer with 5 wt % DSB doped in BANE, and the yellow one had a 35 nm single emissive layer with 1 wt % rubrene doped in BANE. The resulting electroluminescent (EL) characteristics are summarized in Table 1.

The ITO substrate was etched by lithography process to define the active area of the device which is 0.04 mm². All of the organic and metal layers were sequentially deposited at 10⁻⁶ Torr by thermal evaporation onto pre-cleaned and oxygen plasma pre-treated ITO glass. The devices were then encapsulated in a glass-to-glass-based epoxy-sealed package with desiccant incorporated into the non-active area.

Measurement. The current-voltage-luminance characteristics, the EL spectra and lifetimes of the devices were obtained using a programmable Keithley 2400 electrometer and a Minolta CS-1000S spectrophotometer. The photoluminescence spectra (PL) were measured using a Hitachi F-4500 fluorescence spectrophotometer. The ultraviolet visible (UV-vis) absorption spectra were measured using a Hitachi U-3010 UV-vis spectrophotometer. The highest occupied molecular orbital (HOMO) energy levels of the organic materials studied were calculated from their oxidation potentials measured by a cyclic

voltammetry,⁴⁵ while the corresponding lowest unoccupied molecular orbital (LUMO) energy levels were estimated based on their HOMO energy levels and the lowest-energy absorption edge of the UV-vis absorption spectra.

3. RESULT AND DISCUSSION

Figure 3 shows the effect of the architecture on the lifetime of the studied devices driven under constant current with an initial brightness of 5000 cd/m² at room temperature. The lifetime of the pure-yellow device was 484 h, seven times that of the pure-blue counterpart, 69 h. Interestingly, the lifetimes of the white OLEDs varied markedly with their architecture. The lifetime of device I was 83 h, whereas that of device II was 258 h. Both these white devices employed only a single host in the two emissive layers. The lifetime of device I was improved to 351 h by using a mixed host of 50% NPB and 50% BANE in the yellow emissive layer (device III). Surprisingly, the lifetime was further increased to 451 h when a mixed host of 5% NPB and 95% BANE was used in lieu of 100% BANE in the blue emissive layer (device IV-1). This lifetime was 93% of that of the pure-yellow counterpart.

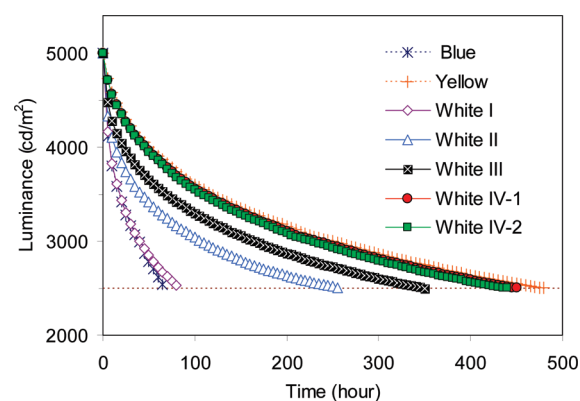
There are two possible reasons why the lifetime improvement was so marked. First, in device IV-1 or IV-2, the carriers were widely dispersed in the vicinity of the three interfaces, thus preventing damage by excessive charge accumulation at the emissive interface.^{30,31} However, for Device III there were only two interfaces to disperse the carriers and only one for Device I or II. As the energy-level diagrams of the white OLEDs are shown in Figure 4, the large barriers at the interface between NPB and BANE, 0.2 and 0.4 eV, respectively, to the transport of holes and electrons explain why the carriers would mostly accumulate at interface II for device I or interface I for device II. As device III employed a 50 wt % BANE and 50 wt % NPB mixed host for the yellow emissive layer, some holes could be transported freely from the HTL to interface II via the NPB in the mixed host. Some electrons could be transported freely from the blue emissive layer to interface I via the BANE in the mixed host. On the introduction of an NPB co-host in the blue emissive layer of device IV-1 or IV-2, some holes that originally accumulated at interface II could be transported freely to interface III. Hence, device IV-1 or IV-2, with double mixed-host layers, most effectively dispersed the carriers into interfaces I, II, and III, thus diluting any undesired degradation products generated during operation.³⁴

Secondly, more power-efficient devices generate less heat upon emission, preventing damage caused by the generation of excess heat during operation.^{30,32} For device I, the efficiency

Table 1. EL Characteristics of the Blue, Yellow, and White OLEDs Studied Here

device ^a	host material		at 20 mA/cm ²			CIE 1931 (x,y) chromatic coordinates			half lifetime at initial 5 000 cd/m ² (h)	CIE variation after aging to half of initial brightness (delta x, delta y)
	yellow BANE : NPB (%)		current efficiency (Cd/A)	Half lifetime (hour)	EQE (%)	at 100cd/m ²				
	NPB (%)	blue BANE : NPB (%)				at 10000 cd/m ²	at 100cd/m ²			
blue		100:0	7.2	6.6	69	5.2	(0.149, 0.178)	(0.149, 0.177)	69	(+0.002, +0.005)
yellow	100:0		7.6	10.5	484	3.7	(0.473, 0.463)	(0.473, 0.462)	484	(−0.001, −0.000)
I	0:100	100:0	7.0	9.9	83	4.5	(0.295, 0.315)	(0.256, 0.276)	83	(−0.003, −0.000)
II	100:0	100:0	7.2	11.4	258	4.4	(0.407, 0.408)	(0.378, 0.382)	258	(+0.009, +0.010)
III	50:50	100:0	6.6	12.0	351	4.9	(0.362, 0.373)	(0.348, 0.360)	351	(+0.003, +0.004)
IV-1	50:50	95:5	6.5	12.4	451	5.6	(0.310, 0.324)	(0.308, 0.322)	451	(+0.009, +0.010)
IV-2	50:50	90:10	6.2	10.6	441	4.8	(0.303, 0.323)	(0.299, 0.318)	441	(+0.009, +0.010)

^aDevices I–IV are white OLEDs.

^a Devices I–IV are white OLEDs.Figure 3. Lifetimes of the blue, yellow, and white OLEDs driven under constant current with an initial brightness of 5000 cd/m² at room temperature.

at 20 mA/cm² was 4.5 lm/W, whereas it was 4.9 lm/W for device II. The efficiencies of devices III and IV-1 were 5.7 and 6.0 lm/W, respectively, the improved efficiency arising from the use of single or double mixed-host layers. Specifically, the recombination zone of devices I and II was localized mainly in the vicinity of one interface of one of the emission layers, and this led to charge imbalance in the other emission layer. In device I, for example, the major recombination zone was localized in the vicinity of interface II of the blue emissive layer, so there were more holes but fewer electrons in the yellow emissive layers. In device II, the major recombination zone was localized in the vicinity of interface I of the yellow emissive layer, and there were fewer holes but more electrons in the blue emissive layers. These charge-imbalance phenomena gave rise to the comparatively poor efficiencies of both devices.

Additionally, the efficiency of devices also relies on energy transfer from host to guest,⁴⁶ which may be revealed by the overlapping area of the UV-Vis absorption spectra of the guest with the photoluminescence (PL) spectra of the host. As shown in Figure 5, the PL peaks of the pure NPB and BANE films were at 438 and 447 nm, respectively, while those of the mixed-host films were broaden and red-shifted. Especially when 50% BANE and 50% NPB were mixed, a largest red-shift to 474 nm was observed. The red-shift may be attributed to the exciplex formation between NPB and BANE.^{47–49} Interestingly, the red-shifted PL spectra of the 50% BANE and 50% NPB mixed host had a greatest overlapping with the UV-Vis absorption spectra of the yellow rubrene, revealing the likeliness of highest energy transfer efficiency. This may explain why the device with mixed host in the yellow emissive layer exhibited higher efficiency. For device III, the introduction of a BANE co-host into the NPB host in the yellow emissive layer provided a path free of energy barriers for electrons to be transported further into the hole-rich but electron-poor yellow-emission zone. For device IV-1, the introduction of 5% NPB co-host into the BANE host in the blue emissive layer also provided a path for holes to be transported further into the electron-rich but hole-poor blue-emission zone. These changes of architecture resulted in a much better charge balance and a lower driving voltage, and hence a higher power efficiency was obtained, as shown in Table 1.

When the mixing ratio of the NPB co-host was increased to 10% (device IV-2), more holes would be transported into the blue emissive layer. After recombining with the holes in the blue

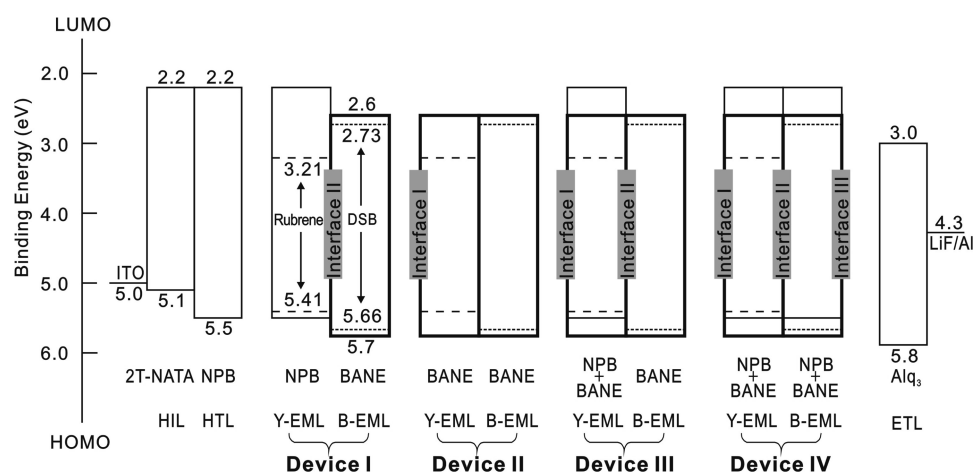


Figure 4. Schematic energy-level diagrams of the white OLEDs. Four types of white OLEDs with different host systems for the yellow/blue emissive layers were studied: NPB/BANE (device I); BANE/BANE (device II); NPB+BANE/BANE (device III); and NPB+BANE/NPB+BANE (device IV-1 & IV-2).

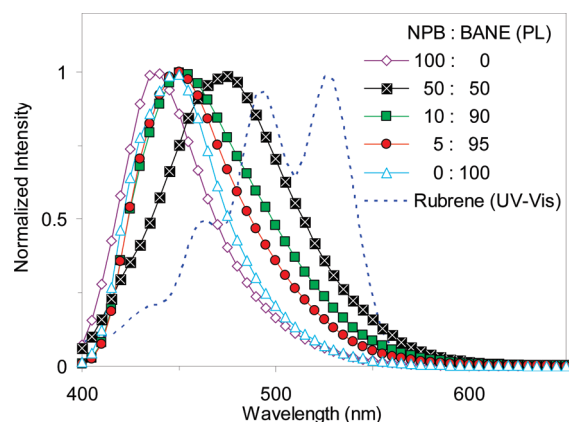


Figure 5. Photoluminescence spectra of the thin films with different host systems and UV-vis absorption spectrum of the guest molecule of rubrene in tetrahydrofuran solution.

emissive layer, fewer electrons would be left to enter further into the yellow emissive layer, wherein charge-imbalance would hence occur or become more seriously. The charge-imbalance would consequently result in a reduction of yellow emission, as revealed by the EL blue shift shown in Figure 6, and a lowering of the device efficiency to 5.4 lm/W, a decrease of 0.6 lm/W. This lower efficiency explains why device IV-2 had a slightly shorter lifetime, 441 h, a decrease of 10 h as compared with device IV-1.

The CIE coordinates of all studied devices are shown in Table 1. Those of device IV-1 and IV-2 shift but very slightly between 100 and 10 000 nits. The reason why the CIE coordinates are stable is because the carriers can be well dispersed in the two different emissive layers by the use of the mixed hosts that provide additional barrier-free transporting channels for the carriers. The CIE variations after aging to half of the initial brightness for all studied devices are also shown in Table 1. The yellow OLED and device I shows little CIE variation, whereas the blue OLED and devices II to V-2 all exhibit slight yellow-shift, which may be attributed to the degradation of the high-excitation-energy deep-blue emitter, DSB.

Panels a and b in Figure 7, respectively, show the effects of power efficiency and number of interfaces for carriers to be dispersed on the lifetime of resultant white OLEDs. Generally,

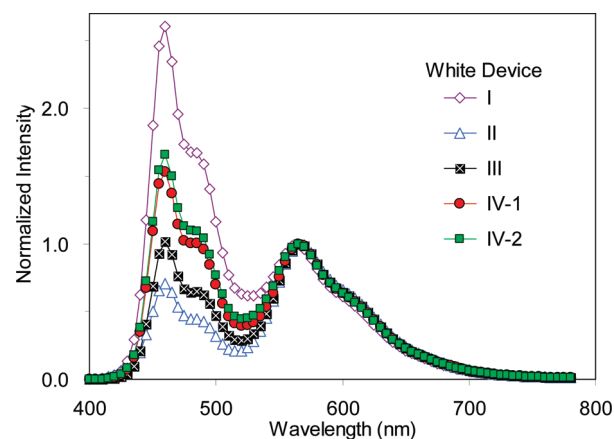


Figure 6. Effect of device architecture on the EL spectra of the resultant white OLEDs at 10 000 cd/m².

device lifetime would increase with the increase of its efficiency.^{32,49} Nevertheless, as shown in Figure 6a, though device III exhibited a power efficiency much higher than that of device IV-2, the lifetime of device III was unexpectedly lower. In contrast, as shown in Figure 6b, the device lifetime increased continuously and substantially with the increase of number of interfaces for carriers to be dispersed. Hence, effective carrier-dispersion seems to be the key factor in resulting in a long device lifetime.

The formation of undesired cationic species, such as Alq₃⁺, was thought to be one fatal factor in shortening the device lifetime.^{30,33} However, the damage caused by the unstable Alq₃ cationic species, if any, was not dominant in the devices studied here. As shown in Figure 5, the introduction of an NPB and BANE mixed host in both emissive layers of devices IV-1 and IV-2 increased the intensity of blue emission, confirming that more holes had been transported to the blue emissive layer. Since the blue emissive layer was adjacent to the ETL (Alq₃), the probability of un-recombined holes entering the Alq₃ was the highest in device IV-1 and IV-2 among the four types of white devices studied here. The generation of Alq₃ cationic species was hence presumably the greatest in devices IV-1 and IV-2. Serious amounts of Alq₃ cationic species cause damage and a relatively

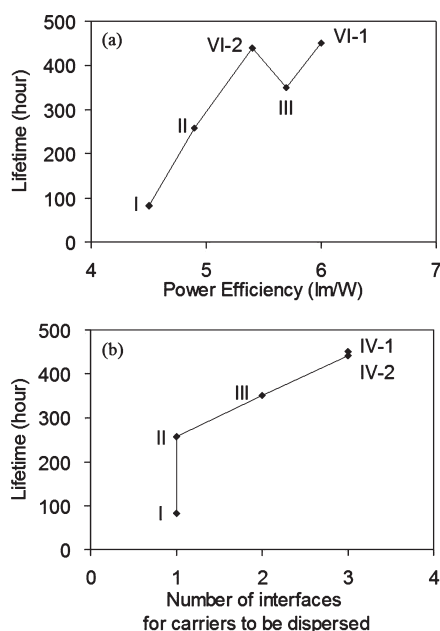


Figure 7. (a) Effects of power efficiency and (b) number of interfaces for carriers to be dispersed on the lifetime of the resultant white OLEDs.

short device lifetime might be expected. However, devices IV-1 and IV-2 exhibited much longer lifetimes than the others, indicating that the damage caused to them by Alq_3^+ was minor.

4. CONCLUSION

In conclusion, we have presented relatively long-lifetime, high-efficiency pure-white OLEDs with double emissive layers composed of mixed hosts. Doping 1 wt % yellow rubrene in a 50% NPB and 50% BANE mixed host and 5 wt % deep-blue DSB in a 5% NPB and 95% BANE mixed host yielded the longest lifetime of 451 h, at an initial brightness of $5,000 \text{ cd/m}^2$. This is almost as long as that of the low-excitation-energy counterpart (yellow device), 484 h, and six times that of the high-excitation-energy one (blue device), 69 h. The resultant efficiency was 6.0 lm/W or 12.4 cd/A at 20 mA/cm^2 with a pure white emission of (0.308, 0.322). This long lifetime was attributable to the double mixed-host architecture, which effectively dispersed the carriers, diluted the damage into the three interfaces.

AUTHOR INFORMATION

Corresponding Author

*E-mail: jjou@mx.nthu.edu.tw.

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