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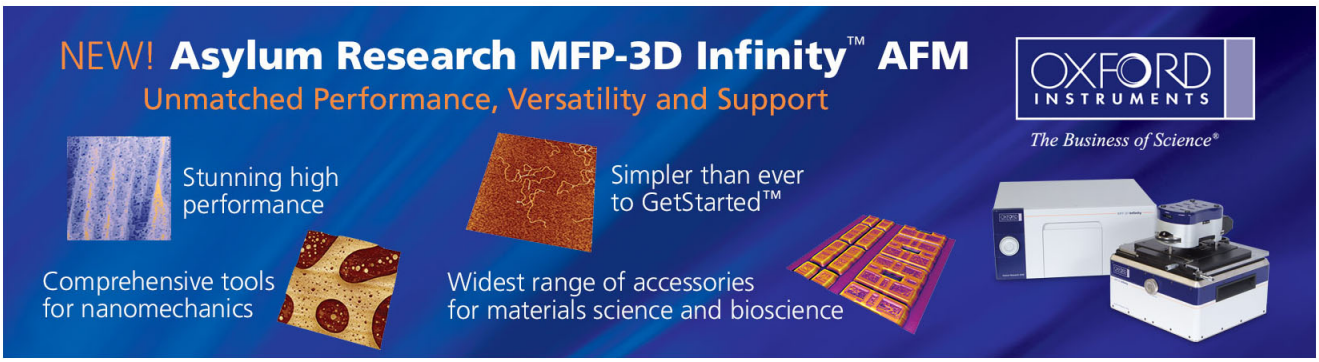

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Doped red organic electroluminescent devices based on a cohost emitter system

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We have developed an efficient red emitter in organic light-emitting diodes based on the fluorescent dye 4-(dicyanomethylene)-2-*t-butyl*-6-(1,1,7,7-tetra-methyljulolidyl-9-enyl)-4*H*-pyran doped in a cohost emitter system of 5,6,11,12-tetraphenylnathacene/*tris*(8-hydroxyquinolinato)aluminum which achieved an electroluminescence (EL) efficiency of 4.4 cd/A and 2.1 lm/W at 20 mA/cm^2 and 6.8 V, with a near saturated Commission Internationale d'Eclairage coordinates ($x=0.65$, $y=0.35$). This cohost emitter system has the advantage of alleviating the current-induced fluorescence quenching often encountered in red organic EL devices and greatly improves the EL efficiency over a wide range of drive current conditions. From accelerated degradation tests, a device half-life of about 33 800 h can be projected in this system at an initial device luminance of 100 cd/m^2. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1635986]

An archetypical red dopant used for many of today's organic light-emitting diodes (OLEDs) displays on the market is 4-(dicyanomethylene)-2-*t-butyl*-6-(1,1,7,7-tetra-methyljulolidyl-9-enyl)-4*H*-pyran, better known as DCJTB.[1] Despite the fact that it is an excellent red fluorescent dye with solution photoluminescence $\lambda_{max}\sim630$ nm and a quantum efficiency $\eta>90\%$, the optimally doped electroluminescence (EL) in *tris*(8-hydroxyquinolinato)aluminum (Alq_3) produces an orange emission that is often contaminated by the residual green emission from the host Alq_3. Although the color saturation of DCJTB can be improved by a high level of doping, its luminance is greatly compromised due to the onset of concentration quenching. As a result, red color approaching Commission Internationale d'Eclairage (CIE) coordinates ($x=0.65$, $y=0.35$) can only be obtained at dopant concentration of higher than ~4% when the luminance has dropped well below its peak. By adding 5% of 5,6,11,12-tetraphenylnathacene (rubrene) as a red-*emitting assist dopant* with 2% DCJTB in Alq_3, Hamada and co-workers at Sanyo[2] were able to achieve a luminance efficiency of 2.1 cd/A with CIE $x,y=[0.64,0.35]$. Subsequently, the Sanyo/Kodak team discovered[3] that by adding 6% of *N,N′-bis*(1-naphthyl)-*N,N′*-diphenyl-1,1′-biphenyl-4,4′ diamine (NPB) as hole-trapping dopant to the above emitting system simultaneously, its efficiency could be improved to 2.8 cd/A at 20 mA/cm^2 and a red chromaticity coordinate of CIE $x,y=[0.65,0.34]$ was also obtained. The encapsulated device structure disclosed in that report showed a remarkable projected operational half-life of 8000 h with a starting luminance of 550 cd/m^2. It is believed that this EL performance is by far one of the best for red fluorescent dye-based OLED emitters.

But, for passive as well as active matrix full color display applications, the DCJTB-doped red emitter of an OLED is still in need of improvement in order to save power consumption and to meet the needs of system requirement.[4] Furthermore, Kodak/Sanyo's three-dopant process involving DCJTB, rubrene, and NPB is also in need of simplification, and the long standing problem of efficiency loss of DCJTB-doped emitters at high drive current conditions[5] remains unsolved and problematical for integrated circuit driver design and system integration. In this letter, we report a cohost emitter (CHE) system, in which the luminance yield of DCJTB-doped emitter can be boosted to 4.4 cd/A and more importantly, we will show that in using this CHE system, the luminance efficiency of DCJTB-doped emitter is unaffected by the drive current in a wide range of doping concentrations and further improve the device reliability.

The CHE consists of a mixture of rubrene and Alq_3 doped with 2% DCJTB. The CF_x, NPB, and Alq_3 were used as the hole injection material,[6] hole transport, and electron transport material, respectively. After a routine cleaning procedure, the indium-tin-oxide (ITO)-coated glass was loaded on the grounded electrode of a parallel-plate plasma reactor, pretreated by oxygen plasma, and then coated with a polymerized fluorocarbon film. Polymerization was carried out with CHF_3 using a low-frequency (13.56 MHz) power generator. The pressure was 270 mTorr. The rf power was 100 W for lasting 15 s. Devices were fabricated under the vacuum of about 10^{-6} Torr in a thin-film evaporation coater following a published protocol.[7] A multilayer structure of NPB/CHE/Alq_3/LiF/Al was deposited on the substrate by resistive heating with a thickness of 120, 30, 50, 1, and 200

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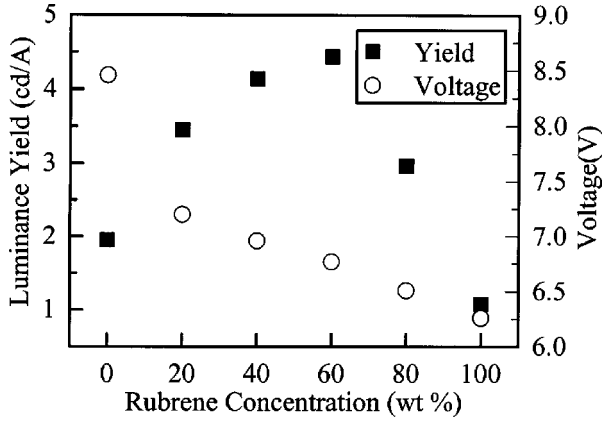

FIG. 1. Efficiency and drive voltage dependency on rubrene concentration in Alq_3 of 2% DCJTB-doped devices.

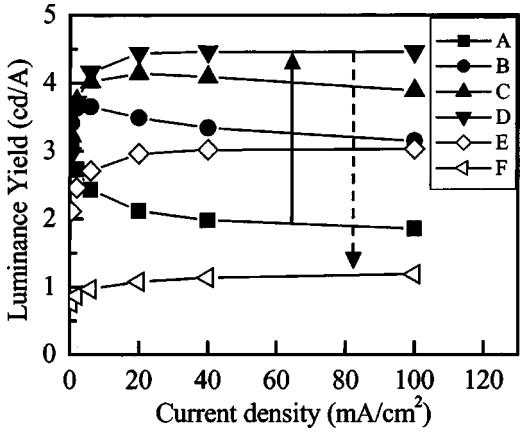

FIG. 2. Luminance efficiency vs current density.

nm for NPB, CHE, Alq_3, LiF, and Al, respectively.[8] All devices were hermetically sealed prior to testing. The active area of the EL device, defined by the overlap of the ITO and the cathode electrodes, was 0.09 cm^2. The current–voltage–luminance characteristics of the devices were measured with a diode array rapid scan system using a Photo Research PR650 spectrophotometer and a computer-controlled programmable dc source. The lifetime measurements were performed in a glove box at a constant current density.

In this study, the key element of the red OLEDs is the ''CHE'' which consists of the mixture of bipolar transport host (rubrene) and organic metal chelate (Alq_3) doped with 2% DCJTB. By tuning the relative ratio of the rubrene/Alq_3 in the cohost matrix, we predict that the balanced carrier injection and transport can be achieved and further improve the device efficiency. Figure 1 shows the plots of luminance yield (cd/A) and drive voltage with respect to rubrene concentration (in wt %) of a series of 2%-doped DCJTB devices. The detailed EL performance of these devices is listed in Table I. It is found that the device drive voltage decreases steadily with respect to increasing rubrene concentration while the luminance efficiency tops out at ~60% and then begins to decrease sharply to 100% rubrene. The best EL performance is found in device **D**, where the device was driven with a dc current density of 20 mA/cm^2 and 6.77 V which achieved a luminance efficiency of 4.44 cd/A and a power efficiency of 2.09 lm/W with a color coordinate of $CIE_{x,y}$=[0.646,0.351]. This efficiency is extremely high for an electrofluorescent red OLED.

Figure 2 shows the plot of luminance efficiency versus drive current density for all cohost-doped DCJTB (2%) devices. It is clear from the profile that only as the cohosted matrix of rubrene/Alq_3 ratio reaches 60/40, can the *cd/A* response be obtained independent of drive current density. This means that the usual quenching phenomenon of the DCJTB-doped Alq_3 device[9] at high drive current is effectively suppressed by the presence of a large proportion of the bipolar transport rubrene in the cohost mixture. This also suggests that the presence of large amount of rubrene in the DCJTB-doped device can also remove the excess hole that is produced at high current density and thus reduce the propensity for the formation of $[Alq_3]^{+\bullet}$ which is a well known quenching species that can also lead to device instability.[10] A further increase of rubrene did not significantly change the profile of the ''flat'' response in Fig. 2 except that the overall luminance efficiency dropped precipitously. This could be due to the luminescence quenching of cohost rubrene at high concentration and a separate phenomenon, which is independent of carrier density. Additional evidence for this rationalization came from the observation that at cohost rubrene/Alq_3 ratio >60/40, the emissive color of 2% DCJTB-doped device became increasingly more yellow [as in (100/0): $CIE_{x,y}$=0.58, 0.41] with a lot of contamination of emission due to rubrene.

Since the CHE system can efficiently suppress the intrinsic quenching effect of DCJTB-doped OLEDs, the device operational lifetime should be expected to improve. Indeed,

TABLE I. EL performance of 2% DCJTB-doped in rubrene/Alq_3 CHE system.

Device	Rubrene concentration(%)	Voltage (V)	CIE *x*	CIE *y*	Lum yield (cd/A)	Efficiency (lm/W)
A	0	8.46	0.639	0.357	1.95	0.73
B	20	7.20	0.658	0.340	3.49	1.54
C	40	6.96	0.649	0.348	4.14	1.89
D	60	6.77	0.646	0.351	4.44	2.09
E	80	6.51	0.631	0.365	2.96	1.45
F	100	6.26	0.581	0.406	1.03	0.55

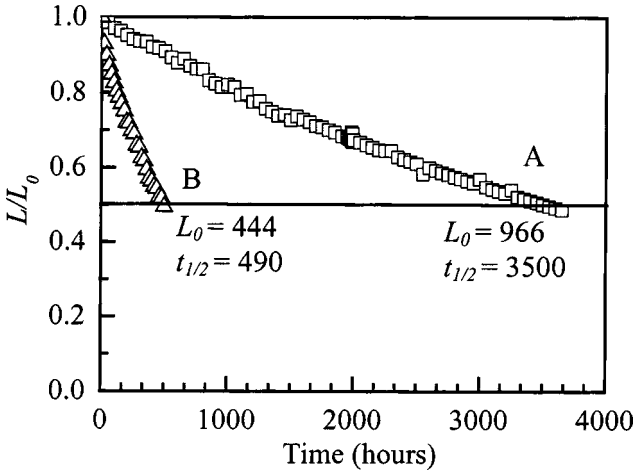

FIG. 3. Luminance (L)/initial luminance (L_0) vs time of 2% DCJTB-doped OLEDs, both driven at 20 mA/cm^2, and the compositions of CHE [rubrene: Alq_3:DCJTB]=(A)[60:40:2] and (B)[100:0:2], respectively.

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results from the device operational stability tests for two 2% DJCTB-doped OLEDs, driven at 20 mA/cm^2, and the compositions of CHE [rubrene:Alq_3:DCJTB]=[60:40:2] and [0:100:2], are shown in Fig. 3, curves A and B, respectively. L_0, measured at the CHE composition, was 966 and 444 cd/m^2, respectively. Assuming the scalable Coulombic degradation,[11] for driving at L_0 of 100 cd/m^2, the half-life ($t_{1/2}$) of the CHE is projected to be 33 810 h.

In conclusion, we have developed an extremely high luminance efficiency for the archetypical DCJTB-doped red emitter at 4.44 cd/A at 20 mA/cm^2 and 6.77 V with a near-saturated $CIE_{x,y}$ red color coordinate of [0.65,0.35]. By doping DCJTB into the cohost matrix emitting system of rubrene/Alq_3, we have also eliminated the problem of luminescence quenching at high drive voltage and achieved a projected operational $t_{1/2}$ lifetime of $>30\,000$ h with initial L_0 of 100 cd/m^2.

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[1] C. H. Chen, C. W. Tang, J. Shi, and K. P. Klubek, Thin Solid Films **363**, 327 (2000).
[2] Y. Hamada, H. Kanno, T. Tsujioka, H. Takahashi, and T. Usuki, Appl. Phys. Lett. **75**, 1682 (1999).
[3] T. K. Hatwar, G. Rajeswaran, J. Shi, Y. Hamada, H. Kano, and H. Takahashi, *Proc. EL'00, Hamamatsu, Japan* (2000), p. 31.
[4] G. Rajeswaran, M. Itoh, M. Boroson, S. Barry, T. K. Hatwar, K. B. Kahen, K. Yoneda, R. Yokoyama, T. Yamada, N. Komiya, H. Kanno, and H. Takahashi, SID Digest **40**, 1 (2000).
[5] R. H. Young, C. W. Tang, and A. P. Marchetti, Appl. Phys. Lett. **80**, 874 (2002).
[6] L. S. Hung, L. R. Zheng, and M. G. Mason, Appl. Phys. Lett. **78**, 673 (2001).
[7] S. A. Van Slyke, C. H. Chen, and C. W. Tang, Appl. Phys. Lett. **69**, 2160 (1996).
[8] C. H. Chen, *Proceedings of the International Meeting on Information Display (IMID 2002)*, Daegu, Korea, 21–23 August 2002.
[9] R. H. Young, C. W. Tang, and A. P. Marchetti, Appl. Phys. Lett. **80**, 874 (2002).
[10] Z. Popovic and H. Aziz, IEEE J. Sel. Top. Quantum Electron. **8**, 362 (2002).
[11] S. A. Van Slyke, C. H. Chen, and C. W. Tang, Appl. Phys. Lett. **69**, 2160 (1996).

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