Improving Fluorescent Blue OLEDs Through Advanced Development Approaches in Materials and Device Design

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Abstract

Since 1985, Idemitsu (Idemitsu Kosan Co., Ltd.) has been continuously developing OLED materials centered on fluorescent blue. In addition to materials, performance improvements through device structures have also been pursued. This report reviews Idemitsu's development of fluorescent blue technology and introduces the latest advancements such as Dual EML (stacked emitting layer with high T1 blue host and low T1 blue host), Dual HTL (stacked hole transport layer with high-n HTL and low-n HTL), and new blue dopant with narrow FWHM (Full Width at Half Maximum). Additionally, we report on the outcomes of material property prediction using machine learning, which contributed to our recent developments.

Author Keywords

OLED; Organic light emitting diode; Fluorescence; Blue; Triplet-triplet fusion; TTF; Bilayer EML; Dual EML; Top emission; Dual HTL; Blue dopant;

1. Introduction

Recently, OLED displays have been increasingly adopted across a wide range of applications, from small devices like smartphones to medium-sized devices like tablets and large devices like TVs. In particular, the integration of generative AI in smartphones and IT applications is expected to increase power consumption, necessitating OLED displays with even lower power consumption. For TVs, reducing power consumption through higher efficiency is also essential to increase the market share of OLED TVs.

Phosphorescent and TADF emission devices can theoretically achieve higher internal quantum efficiencies than fluorescent emission devices. While material development has improved device performance, no sufficiently long lifetime blue phosphorescent or TADF materials have been found to date. Therefore, achieving high-efficiency blue devices with practical lifetimes requires focusing on fluorescent blue.

Since 1985, Idemitsu has been engaged in OLED research, primarily developing fluorescent materials. Idemitsu's strengths lie not only in material design but also in comprehensive device design technologies, including device structure optimization and optical design to improve light out-coupling efficiency. In this report, we review Idemitsu's efforts in fluorescent blue technology and introduce the latest advancements. The performance of the fluorescent blue device used in Idemitsu's 1997 world-first full-color OLED display [5], when converted to top-emission structure efficiency by optical simulation, could show Blue Index (BI, cd/A/y) efficiency of 41. However, the latest technology has achieved a BI of 350. Henceforth, top-emission efficiency will be expressed as BI in this report. Idemitsu has also focused on constructing efficient material development environments, recently introducing material property prediction tools using machine learning. The prediction accuracy of these tools will also be reported.

2. Results and Discussions

2-1. Fluorescent blue materials

In the early stages of OLED development, Idemitsu developed distyrylarylene as a simple fluorescent blue emitter [1][2]. Subsequently, we found materials that improved performance by doping blue emitters into blue hosts, enhancing emission efficiency and FWHM by doping styrylamine into a distyrylarylene blue host [3]. In 1997, we reported the world's first full-color OLED display [4][5], using blue host (BH) IDE120 and blue dopant (BD) IDE102, which exhibited light blue. When converted to the current top-emission structure by optical simulation, the device performance could show CIEy 0.173 and BI of only 41. Later, we succeeded in developing a deep blue dopant, reporting styrylamine IDE105 in 2001 [6]. Subsequently, we developed highly durable BHs such as BH-215 that is the original of current main BH series [7] and continued the development of BD [9][10], device structures that confine triplets to enhance Triplet-Triplet-Fusion (TTF) [11], and Dual EML structures to further improve TTF efficiency [16].

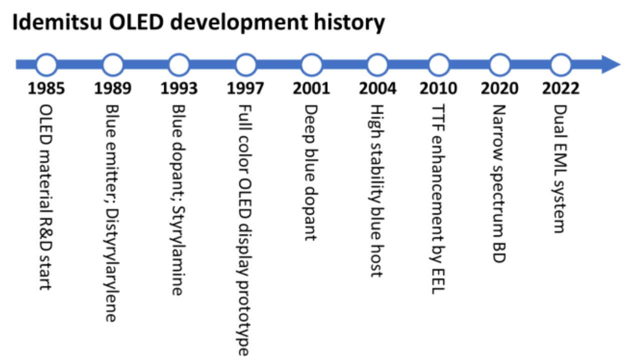

Figure 1. Idemitsu's OLED development history.

2-2. Triplet-Triplet-Fusion (TTF)

TTF emission occurs when triplet excitons with long lifetimes collide, with some excitons converting to singlets, transferring energy from BH to BD, and ultimately emitting from BD. Improving TTF efficiency is key to enhancing emission efficiency of fluorescent devices. Besides optimizing the carrier balance of holes and electrons injected into the emission layer, Idemitsu reported in 2010 at SID that inserting an efficiency-enhancement layer (EEL) with high T1 level between the electron transport layer (ETL) and emission layer (EML) can suppress triplet exciton diffusion to the ETL, increasing triplet exciton density and TTF efficiency by up to 28% [11][12].

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2-3. Dual EML (Dual Emission Layer)

Recently, Idemitsu invented the Dual EML structure, achieving further TTF efficiency improvements by suppressing triplet quenching by carriers using a two-layer EML [16]. Dual EML is a technology that improves exciton generation efficiency of fluorescent devices. While theoretically possible to exceed 25% exciton generation efficiency by utilizing TTF to achieve 40%, increasing TTF efficiency requires high triplet concentrations to promote triplet collisions. In conventional fluorescent blue devices, recombination mainly occurs at the electron blocking layer (EBL) and EML interface because of using electron transport BH, where TTF also occurs. However, due to the long excitation lifetime of triplets, they are prone to be quenched by surrounding carriers (Triplet-Polaron-Quench; TPQ), reducing actual emission efficiency below theoretical TTF limits. Dual EML separates recombination and TTF regions by introducing an emission layer structure with a high T1 level BH1 layer on the hole transport layer (HTL) side and a low T1 level BH2 layer on the ETL side, suppressing TPQ and increasing internal emission efficiency, enhancing emission efficiency by 10%. (Figure 2)

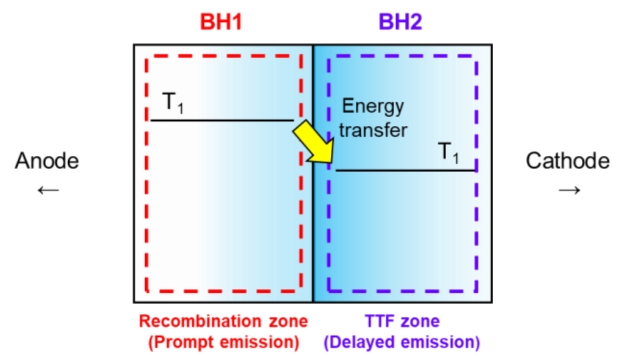

Figure 2. Concept of Dual EML [16].

Idemitsu will present detailed analyses of the Dual EML mechanism, deuteration effects on Dual EML materials, and high-efficiency Dual EML devices using the latest materials in another report at SID2025.

2-4. light-outcoupling

While optical interference-based light extraction improvements are widely implemented in top-emission structures, Idemitsu has utilized optical simulations to analyze emission devices and improve light out-coupling efficiency [8][12]. BD orientation related to molecular shape and other factors also impacts light out-coupling efficiency, correlating with emission efficiency [13][14]. Particularly, the emission spectrum shape of BD significantly influences light out-coupling efficiency in top-emission devices. Narrowing the emission spectrum increases overlap with the optical amplification wavelength band, enhancing light extraction.

2-5. Narrow FWHM and shoulder less BD

In top-emission devices, narrowing the FWHM and reducing shoulder peaks of BD emission spectra are crucial for improving light out-coupling efficiency. At SID2020, Idemitsu reported a BD with a narrow FWHM achieved by suppressing structural relaxation in the excited state and reduced shoulder peaks by introducing particular molecular structures [15]. The newly developed BD-4 has a narrower FWHM and further reduced shoulder peaks compared to BD-3 that was reported at SID2020. Figure 3 shows the PL spectra of films doped with BD-3 and the newly developed BD-4 in the BH. BD-4 exhibits narrower FWHM (from 23nm to 21nm) and reduced shoulder peaks compared to BD-3.

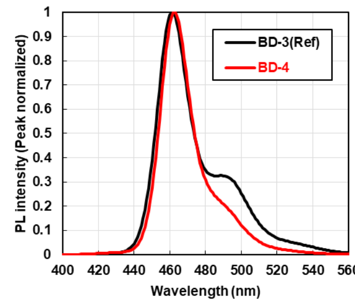

Figure 3. Normalized PL spectrum of BD doped BH films.

Table 1 shows the device characteristics of top-emission devices using BD-3 and BD-4. The improved overlap between the emission spectrum and optical interference wavelength band resulted in higher emission efficiency.

Table 1. Top emission device performance of narrow BD (BD-4). The device structure is Ag/ITO(10)/HTL:p(10:5%)/HTL(114)/EBL(7.5)/BH:BD(20:2%)/HBL(5)/ETL:Liq(25:33%)/Yb(1)/Mg:Ag(15:90%)/Cap(65) (nm). IVL at 10mA/cm^2. LT at 50mA/cm^2, 25°C.

BD	Voltage [V]	CIE		BI [cd/A/y]	LT95 [h]
		x	y		
BD-3(Ref)	3.5	0.141	0.042	213	200
BD-4	3.5	0.141	0.039	265	197

2-6. Dual HTL for light-outcoupling enhancement

(a) Concept of Dual HTL

This report introduces a new light extraction enhancement technique using Dual HTL system. The mechanism involves replacing the typical single thick HTL with a stacked structure of high refractive index HTL near the anode side (HTL1) and low refractive index HTL near the cathode side (HTL2), leveraging the refractive index difference at the HTL1/HTL2 interface to enhance light out-coupling (Figure 4).

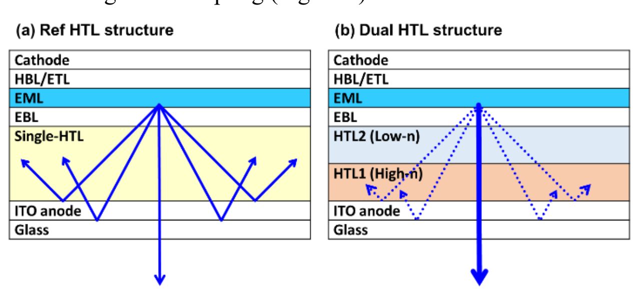

Figure 4. Schematic of the Dual HTL structure for light out-coupling enhancement. (a) shows the conventional HTL device. (b) shows the newly developed Dual HTL device.

Figure 5 shows the wavelength dependence of the refractive indices of High-n HTL and Low-n HTL films measured by ellipsometry. At a typical blue wavelength of 460 nm, High-n HTL shows n2.00, while Low-n HTL shows n1.74.

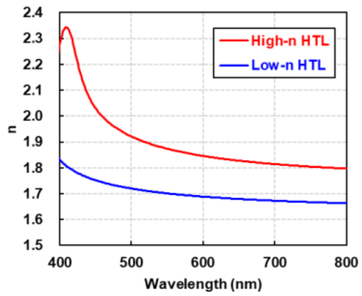

Figure 5. Refractive index (n) of high-n and low-n HTL films for Dual HTL.

Optical simulations of bottom-emission devices using these HTLs indicated higher emission efficiency for the High-n HTL/Low-n HTL stacked Dual HTL device compared to single High-n HTL or Low-n HTL devices, primarily due to reduced optical losses in guided mode within organic and ITO layers (Table 2).

Table 2. Optical simulation results of the bottom emission devices with single HTL and Dual HTL. The simulated device structure is Glass/ITO(135)/HTL1(27.5)/HTL2(60)/EBL(7.5)/BH:BD-4(20:2%)/HBL(5)/ETL(20)/Al (nm).

Device	HTL1	HTL2	Simulated relative EQE
Single HTL	High-n		100%
Single HTL	Low-n		111%
Dual HTL	High-n	Low-n	119%

(b) Bottom-emission device

We fabricated bottom-emission devices with Dual HTL and evaluated their performance (Table 3). The Dual HTL device showed higher emission efficiency than single HTL of high-n HTL or low-n HTL devices, consistent with optical simulation results.

Table 3. Bottom emission device performance of the single HTL and the Dual HTL devices. The device structure is Glass/ITO(135)/HTL1:p(10:5%)/HTL1(37.5)/HTL2(40)/EBL(7.5)/BH:BD-4(20:2%)/HBL(5)/ETL:Liq(25:33%)/Yb(1)/Al(80) (nm). IVL at 10mA/cm^2. LT at 50mA/cm^2, 25°C.

Device	HTL1	HTL2	Voltage [V]	EQE [%]	Relative EQE	LT95 [h]
Single HTL	High-n		3.3	11.4	100%	266
Single HTL	Low-n		3.7	12.0	105%	132
Dual HTL	High-n	Low-n	3.3	14.0	120%	213

We conducted transient electroluminescence (EL) measurements on these devices to calculate the delayed emission efficiency (TTF EQE) derived from TTF and the prompt emission efficiency (prompt EQE) derived from singlet states, using the delayed emission ratio (TTF ratio). The results revealed that the prompt EQE for the Dual HTL device increased compared to the Single HTL device, and the extent of this increase was generally consistent with that obtained from the optical simulations (Table 4). Consequently, it can be inferred that variations in prompt efficiency correspond to changes in light out-coupling efficiency.

Table 4. Efficiency and transient EL measurement results of the single HTL and the Dual HTL BE devices. TTF ratio represents the proportion of total EQE that is attributed to TTF EQE.

Device	HTL1	HTL2	Total EQE [%]	TTF ratio [%]	TTF EQE [%]	Prompt EQE [%]	Relative prompt EQE
Single HTL	High-n		11.4	30.6	3.5	7.9	100%
Single HTL	Low-n		12.0	33.5	4.0	8.0	101%
Dual HTL	High-n	Low-n	14.0	33.6	4.7	9.3	118%

(d) Top-emission device

The Dual HTL structure was also applied to top-emission devices. Table 5 and Table 6 show the results compared to single HTL devices. Dual HTL devices exhibited higher emission efficiency and increased prompt efficiency (prompt BI), indicating enhanced light out-coupling efficiency also in top-emission structures.

Table 5. Top emission device performance of the single HTL and the Dual HTL devices. The device structure is Ag/ITO(10)/HTL1:p(10:5%)/HTL1(x)/HTL2(50)/EBL(7.5)/BH:BD-4(20:2%)/HBL(5)/ETL:Liq(25:33%)/Yb(1)/Mg:Ag(15:90%)/Cap(65) (nm)(x=65 for high-n single HTL, x=76 for low-n single HTL, x=69 for Dual HTL). IVL at 10mA/cm^2. LT at 50mA/cm^2, 25°C.

Device	HTL1	HTL2	Voltage [V]	CIE		BI [cd/A/y]	LT95 [h]
				x	y		
Single HTL	High-n		3.5	0.142	0.036	271	267
Single HTL	Low-n		4.0	0.142	0.038	285	77
Dual HTL	High-n	Low-n	3.5	0.139	0.041	295	234

Table 6. Efficiency and transient EL measurement results of the single HTL and the Dual HTL TE devices.

Device	HTL1	HTL2	Total BI [cd/A/y]	TTF ratio [%]	TTF BI [cd/A/y]	Prompt BI [cd/A/y]	Relative prompt BI
Single HTL	High-n		271	29.5	80.0	191.0	100%
Single HTL	Low-n		285	31.4	89.8	195.8	102%
Dual HTL	High-n	Low-n	295	31.3	92.0	202.9	106%

2-7. Combination of Dual EML, Dual HTL, narrow BD

To maximize the potential of fluorescent blue devices, we combined Dual EML, Dual HTL, and narrow FWHM BD to fabricate bottom-emission and top-emission devices. The performance of these devices is shown in Table 7 and Table 8, achieving EQE of 16.3% in bottom-emission and BI of 350 in top-emission, which represents a very high efficiency for fluorescent blue devices to date. Detailed results will be presented by Idemitsu in another report at SID2025.

Table 7. Bottom emission device performance of the combination of Dual EML, narrow BD and Dual HTL. The device structure is Glass/ITO(135)/HTL1:p(10:5%)/High-n HTL(37.5)/Low-n HTL(40)/EBL(7.5)/BH1:BD(5:2%)/BH2:BD-4(15:2%)/HBL(5)/ETL:Liq(25:33%)/Yb(1)/Al(80) (nm). IVL at 10mA/cm^2. LT at 50mA/cm^2, 25°C.

EML	BD	HTL	Voltage [V]	CIE		EQE [%]
				x	y	
Dual	Narrow	Dual	3.5	0.134	0.067	16.3

Table 8. Top emission device performance of the combination of Dual EML, narrow BD and Dual HTL. The device structure is Ag/ITO(10)/HTL1:p(10:5%)/High-n HTL(61)/Low-n HTL(50)/EBL(7.5)/BH1:BD(5:2%)/BH2:BD-4(15:2%)/HBL(5)/ETL:Liq(25:33%)/Yb(1)/Mg:Ag(15:90%)/Cap(65) (nm). IVL at 10mA/cm^2. LT at 50mA/cm^2, 25°C.

EML	BD	HTL	Voltage [V]	CIE		BI [cd/A/y]	LT95 [h]
				x	y		
Dual	Narrow	Dual	3.6	0.139	0.042	350	>200

2-8. Material development acceleration using materials informatics

To efficiently conduct innovative material development, Idemitsu utilized machine learning as a tool to accelerate the development process. For example, the HOMO and refractive index of hole transport materials, which are important parameters for Dual HTL structures, were predicted using machine learning. Figure 6 shows the predicted values for the DFT-calculated HOMO and experimentally measured refractive indices, with R^2 values of 0.95 for the DFT HOMO and 0.84 for the measured refractive indices. This prediction tool enabled faster and more accurate property value predictions, significantly improving development efficiency.

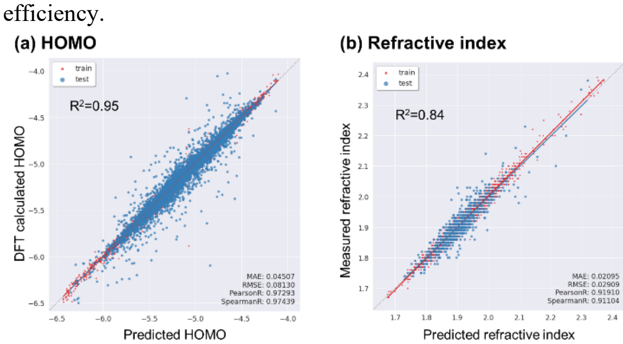

Figure 6. Prediction results of DFT calculated HOMO (a) and measured refractive index (b).

3. Conclusion

Since 1985, Idemitsu has been developing OLED materials, focusing on fluorescent blue. We developed new device structures such as Dual EML and Dual HTL to maximize the potential of stable fluorescent blue materials and succeeded in developing narrow FWHM BD. Combining these materials and device technologies, we achieved over 16% EQE in fluorescent blue devices. We also introduced machine learning tools for material property prediction, achieving high-accuracy predictions in a short time, improving development efficiency.

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