

FINAL SCINETIFIC/TECHNICAL REPORT

1. Award Information and Team Membership:

DOE Award Number: DE-FC26-06NT42859

Name of Recipient: University of North Texas

Project Title: Multi-Faceted Scientific Strategies Toward Better Solid-State Lighting of Phosphorescent OLEDs

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*: *Worked part-time. All others worked as full-time team members.*

2. Authorized Distribution Limitation Notices: None

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3. Executive Summary:

This project has advanced solid-state lighting (SSL) by utilizing new phosphorescent systems for use in organic light-emitting diodes (OLEDs). The technical approach was two-fold:

- a) Targeted synthesis and screening of emitters designed to exhibit *phosphorescence with maximized brightness in the solid state.*
- b) Construction and optimizing the performance of monochromatic and white OLEDs from the best new emitters to improve performance metrics versus the state of the art.

The phosphorescent systems were screened candidates among a large variety of recently-synthesized and newly-designed molecular and macromolecular metal-organic phosphors. The emitters and devices have been optimized to maximize light emission and color metrics, improve the long-term durability of emitters and devices, and reduce the manufacturing cost both by simplifying the process flow and by seeking less expensive device components than common ones. The project succeeded in all these goals upon comparison of the best materials and devices investigated vs. the state of the art of the technology.

4. Comparison of Actual Accomplishments with Original Goals and Objectives:

The original goals and objectives were as follows: "The expected result is to demonstrate white and monochrome OLEDs from materials in this project with device performance metrics that exceed those of Ir-ppy based OLEDs and that are consistent with the MYP of DOE-SSL."

The actual accomplishments for devices based on one preferred embodiment phosphorescent material developed in the project, $\text{Pt}(\text{ptp})_2 = \text{bis}[3,5\text{-bis}(2\text{-pyridyl})\text{-}1,2,4\text{-triazolato}]$ platinum(II), after extensive screening are summarized as follows:

- a) Attaining a turquoise-blue OLED with power efficiency maximum of 61 lm/W using our 5%-doped $\text{Pt}(\text{ptp})_2$ devices versus 41 lm/W for analogous turquoise-blue baseline devices using FIrpic. Optimization has been finished so we met and exceeded the milestone goal. This performance constitutes a new world record for a turquoise-blue emitter.
- b) Attaining a yellow-orange OLED with power efficiency maximum of 80 lm/W and near 100% IQE (21% EQE) using neat $\text{Pt}(\text{ptp})_2$ devices versus 45.5 lm/W and 16% EQE for the highest performance baseline devices using Ir(ppy)₃. This performance constitutes a new world record for neat OLEDs.
- c) Attaining a warm-white OLED with power efficiency maxima of 45-50 lm/W and near 100% IQE using a single phosphor by gradient doping. There is no precedent in the literature for a WOLED from a single Ir-ppy phosphor to run as a baseline. This performance constitutes a new world record for a single emitter-based white OLED and is projected to reach total efficacy of at least 90-100 lm/W with 2x out-coupling enhancement as a conservative estimate among current technology protocols to attain such an enhancement.
- d) Regarding white color metrics, we have achieved white OLEDs within the warm-white CCT range and up to 82 CRI in both single-phosphor devices and single phosphor plus single fluorophore devices.
- e) The project identified a road map toward white OLEDs with power efficiency that will exceed 150 lm/W (with out-coupling enhancement) from a single emitter with a simple doping-free device architecture.

5. Summary of Project Activities:

5.1. Screening of Phosphors:

The project focused on screening neat (as opposed to doped) emitting materials so as to ameliorate common quenching mechanism and phenomena in OLEDs. The problems include triplet-triplet annihilation, the culprit behind low wall-plug efficiencies at high current/voltage/brightness, as it pertains to doped thin films of the first ever breakthrough phosphorescent OLED employing Pt(octaethylporphyrin), “PtOEP” (Forrest; Thompson et al., *Nature* **1998**, 395, 151). While we presented some synthetic and photoluminescence screening data suggesting a possible remedy to overcome triplet-triplet annihilation and the consequent self-quenching in OLEDs based on PtOEP to be pursued in the future, we focused more on tackling the extreme situation represented by neat materials of phosphors. The matter is addressed here for the Pt(otp)₂ phosphorescent complex that we have successfully screened as a preferred embodiment in OLEDs during this project following extensive examination of a large variety of synthetic targets. The self-quenching issue limited the maximum doping level to attain optimum OLED efficacy has been reported to be ~ 6% for PtOEP (Forrest et al., *Nature* **1998**, 395, 151) and fac-Ir(ppy)₃ (Forrest; Thompson et al., *APL* **1999**, 75, 4) complexes, whereas the PL and EL data here show that we can obtain very high efficiency for thin films that have ~ 50-100% concentrations of Pt(otp)₂ due to its “self-sensitization” and the insignificance of triplet-triplet annihilation in this material. Slow triplet relaxation of the emitter can form a “bottleneck” in electrophosphorescent OLED performance at high brightness levels that are practical for SSL. A long exciton lifetime (> 10 μs) in OLEDs leads to saturation of emissive sites and consequent decreased quantum efficiency at high currents and low dopant concentrations. While the problem can be ameliorated by increasing the doping concentration of the phosphor, doing so beyond 6-10% cannot lead to improvement in PtOEP and Ir-ppy based OLEDs due to the self-quenching problem whereby the proximity of molecules to one another leads to bimolecular quenching. Since Ir-ppy complexes in common OLED hosts such as CBP exhibit triplet lifetimes on the order of ~ 1 μs (Forrest; Thompson et al., *APL* **1999**, 75, 4; *APL* **2000**, 77, 904), two orders of magnitude shorter than the ~ 100 μs lifetimes for PtOEP, the saturation issue is much less severe but remains significant especially at high current/voltage/brightness and high doping levels. For example, the power efficiency of 6% Ir(ppy)₃:CBP devices drops by 76%, from a maximum of 31 lm/W near turn-on to 7.5 lm/W at 1000 cd/m²; meanwhile, increases in dopant concentration to 12%-doped and 100%-neat films lead to rather significant drops in the peak EQE by 62% and 91%, respectively, i.e. from EQE = 8% for the 6%-doped film to EQE = 3% for the 12%-doped film and EQE = 0.75% for the neat film (Forrest; Thompson et al., *APL* **1999**, 75, 4). The PL data in this section and EL data in subsequent sections show that our breakthrough Pt(otp)₂ material is a “self-sensitizing fast phosphor” that solves both the triplet lifetime and self-quenching problems that have hampered the performance of electrophosphorescent OLEDs at practical brightness levels for SSL applications up to now. In addition to self-sensitization and overcoming triplet-triplet annihilation, another advantage offered by neat or highly-doped films of the phosphor in OLEDs is circumventing exchange energy loss, hence decreasing the device drive voltage.

The screening data in this section pertain to three forms of neat Pt(otp)₂ that have different structural and PL properties in the single crystal or bulk powder form (Fig. 1). The spectral data of the three forms shown in Fig. 1 (top-to-bottom) are rather similar to those exhibited by lightly-doped (~ 5% Pt(otp)₂:CBP), heavily-doped (≥ ~40% Pt(otp)₂:CBP), and neat thin films of Pt(otp)₂, respectively. Fig. 1 shows that the orange form exhibits the brightest emission and shortest radiative lifetime among the three forms of Pt(otp)₂. Triplet-triplet annihilation is negligible in this

form because of two reasons: (1) The triplet lifetime of 92 ns is unusually short for a brightly-phosphorescent solid material at ambient temperature, rendering the material a *“fast phosphor”*. (2) The stacking of molecules is maximized because the molecules perfectly overlap with one another. This overlap *causes* the bright orange emission of this form, *turning the “self-quenching” problem common in other materials into “self-sensitization” in this neat form of Pt(otp)₂*. As the overlap between the Pt(otp)₂ molecules decreases gradually on going from the orange/orange-emitting form (bottom) to the bright-yellow/yellow-emitting form (middle) and then to the pale-yellow/turquoise-green emitting form (top), the emission becomes less bright with a longer lifetimes. This trend accentuates the self-sensitization phenomenon as it manifests exactly the opposite trend known for most luminescent materials, including PtOEP and Ir-ppy complexes, for which lesser degree of overlap between molecules is desirable for their PL or EL while it is desirable to maximize such intermolecular interactions in Pt(otp)₂. Given the fact that the emission in the limiting orange form is metal-centered, the phonon energy associated with the Pt···Pt interaction is much less than that involving ligand-centered or metal-ligand charge transfer emissions for which the phonon energies are typically an order of magnitude higher, making them more susceptible to quenching by non-radiative multiphonon relaxation processes to the ground state while such processes are inhibited for the *Pt-Pt excimeric emissions characteristic of the stacked forms of Pt(otp)₂.

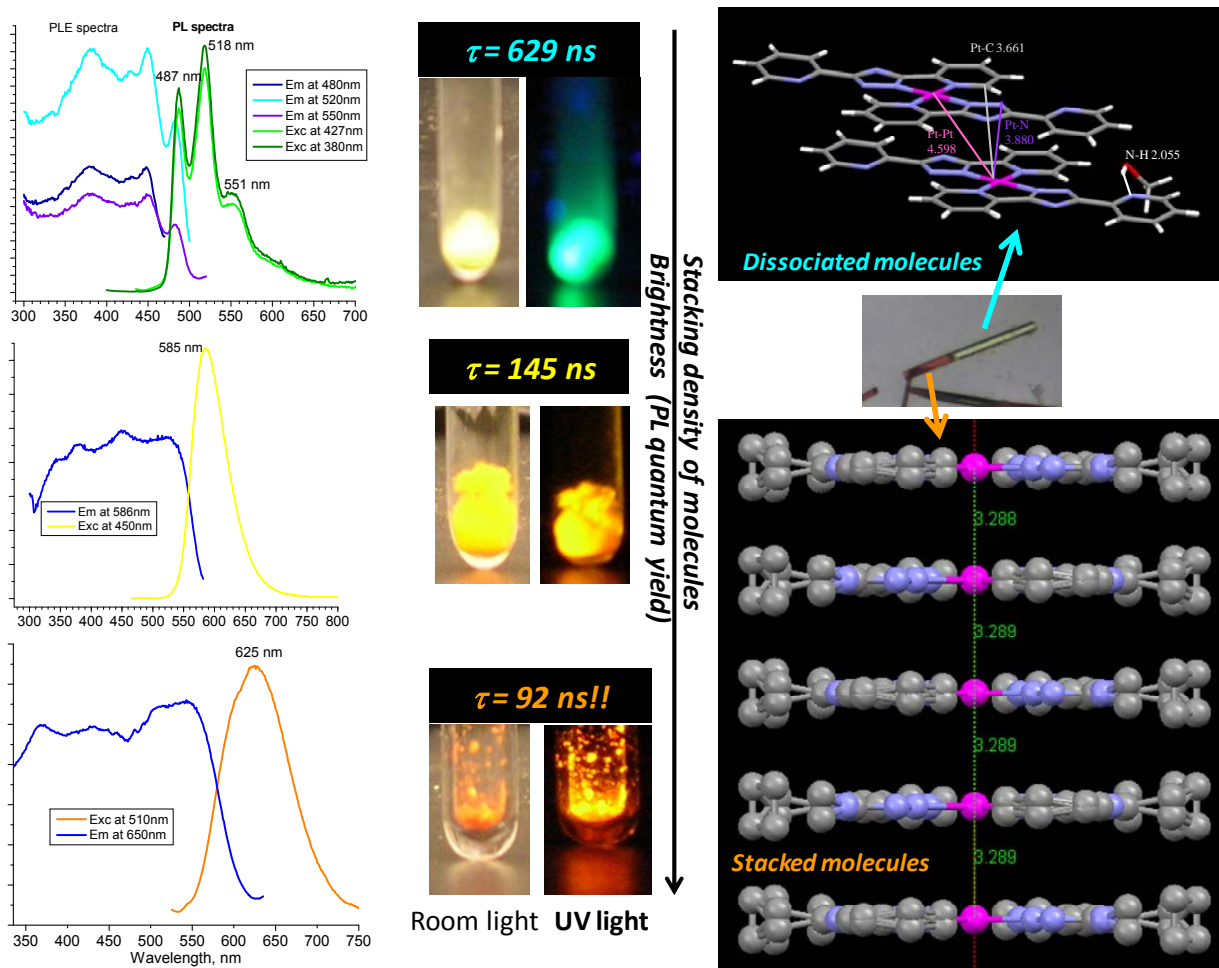


Fig. 1. PL and structural data for three forms of Pt(otp)₂ with differing stacking density of molecules.

Further PL screening evidence to the self-sensitization phenomenon has been collected by spectroscopic studies of Pt(ppy)₂ in solution. Thus, quantitative measurements have shown that upon increasing the total Pt(ppy)₂ concentration from 10⁻⁵ M to 10⁻³ M in dichloromethane/methanol, the peak area of the emission peak (normalized to solution absorption under the respective excitation wavelength) increases by a factor of 2.38, representing a 138% self-sensitization due to the association of the phosphor in solution. The spectral data are shown in Fig. 2. Qualitatively, proceeding from solution to the solid state enhances the self-sensitization effect even further because only the more efficient extended excimer emissions are seen for neat thin films (see Task 2 data).

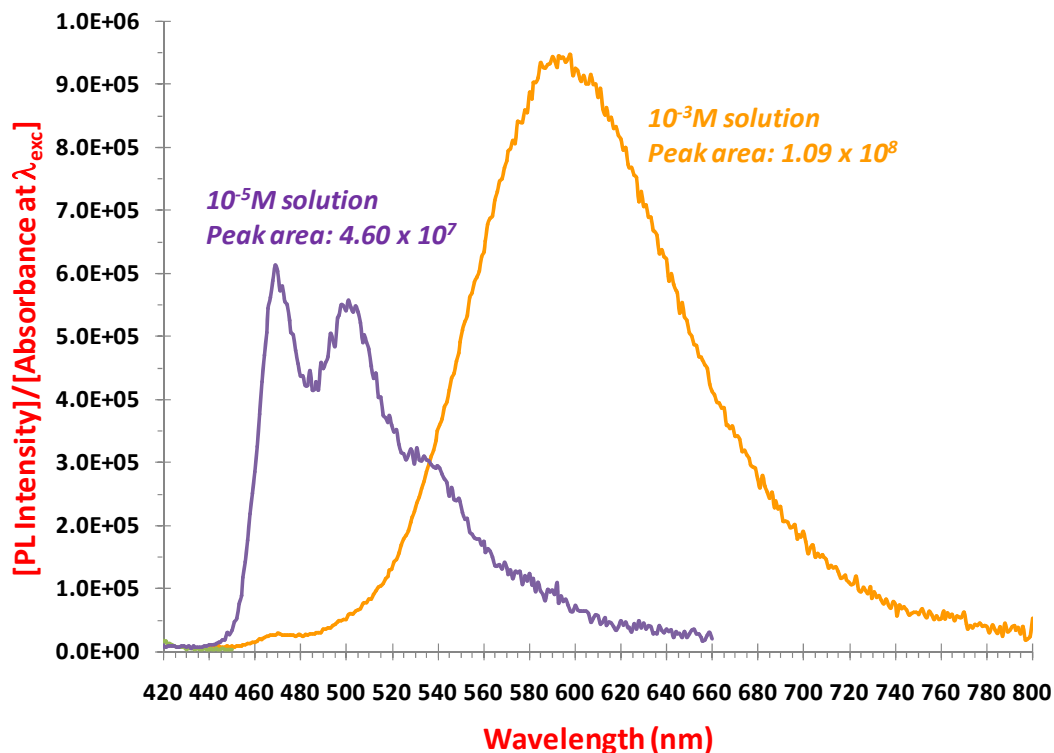


Fig. 2. PL data for solutions of Pt(ppy)₂ in dichloromethane/methanol with 10⁻³ M and 10⁻⁵ M total concentration.

5.2. Monochromatic and White OLED Device Fabrication and Testing:

Figs. 3-6 and Tables 1-3 summarize the results that prove the self-sensitization concept *via* EL while Fig. 7 illustrates a proof-of-concept example showing how these results can be utilized to design a high-CRI WOLED with no roll-off from a single emitter. These data illustrate our approach in detail while further optimization is described subsequently in less detail.

Fig. 3 and Table 1 show EL spectra and device performance metrics for OLEDs based on Pt(ppy)₂ versus doping level (3-100%) in an unoptimized device structure. Color tuning between blue and orange is achieved by controlling the dopant concentration. The performance metrics summarized in Table 1 are consistent with the normal behavior in electrophosphorescent OLEDs because the best performance is achieved for devices with 5-12% doping level, similar to reported devices based on Ir(III) phenylpyridine and Pt(II) porphyrin complexes such as Ir(ppy)₃ and PtOEP, respectively. Below 5% incomplete energy transfer takes place, resulting in

CBP emission. A first look at the EL data at > 12% doping level suggests a trend that is contrary to the PL trends in the previous section, as Table 1 data seem more consistent with self quenching as opposed to self sensitization at higher dopant concentrations. However, Fig. 3 clearly shows that devices with the higher doping levels exhibit emission due to the ETL material NPB; the effect is maximized for the neat film device in which the NPB emission is quite significant. (Indeed, the CIE coordinates and CRI are close to those of white light sources for the 100% device due to the combination of the broad orange emission of Pt(otp)₂ and the deep blue emission of NPB.) This suggests excess of electrons in devices that contain significant amounts of the Pt(otp)₂ dopant, necessitating the addition of an electron-blocking layer in order to achieve a better charge balance. When this was done, the trend became consistent with the PL data and we obtained efficient devices for highly-doped (Fig. 4 and Table 2) and neat emissive layer devices (Figs. 5-6 and Table 3) to prove the self-sensitization effect via EL.

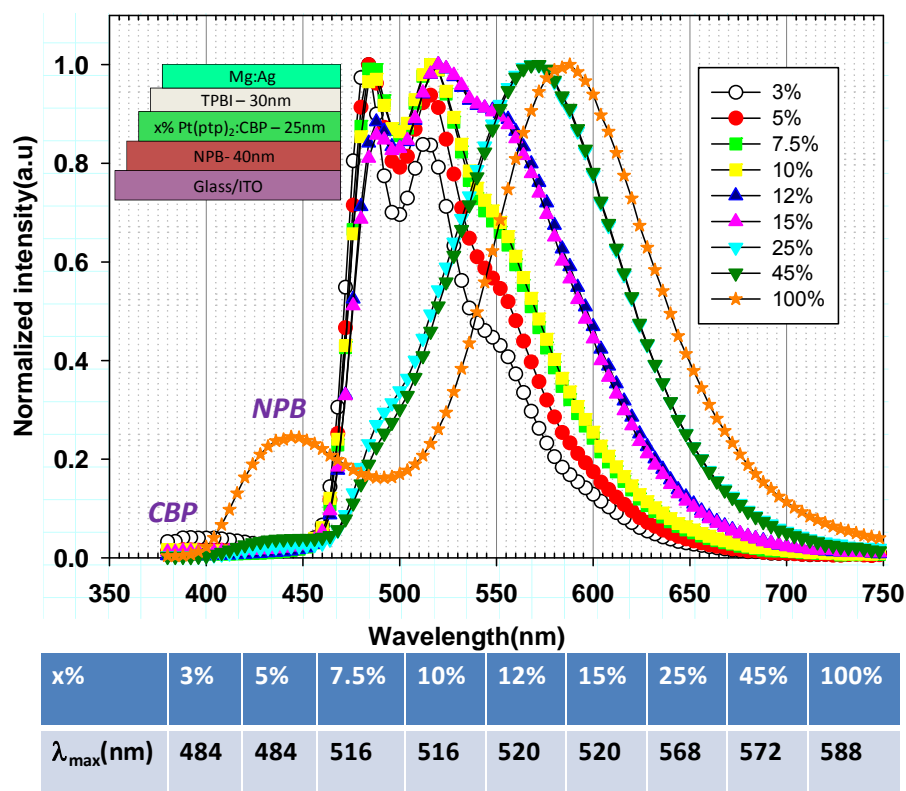


Fig. 3. EL spectra for unoptimized OLEDs based on Pt(otp)₂ showing color tuning by controlling the doping level.

Table 1. Performance metrics for the OLEDs shown in Fig. 3. Device efficacies and color coordinates are reported at peak performance and at 1000 cd/m².

x%	PE _{peak} (lm/W)	PE ₁₀₀₀ (lm/W)	EOE _{peak} (%)	EOE ₁₀₀₀ (%)	CIE _{peak} (x, y)	CIE ₁₀₀₀ (x, y)	CRI _{peak}	CRI ₁₀₀₀	V _T (V)
3%	6.32 ± 0.72	2.47 ± 0.06	3.92 ± 0.20	2.46 ± 0.07	(0.229, 0.496)	(0.225, 0.472)	26	27	5.0
5%	<u>9.83 ± 0.02</u>	5.81 ± 0.04	<u>6.55 ± 0.03</u>	<u>5.32 ± 0.04</u>	(0.248, 0.517)	(0.245, 0.514)	27	30	4.0
7.5%	8.07 ± 0.02	<u>7.29 ± 0.03</u>	4.91 ± 0.02	4.89 ± 0.03	(0.274, 0.526)	(0.272, 0.526)	32	32	4.2
10%	7.16 ± 0.4	4.27 ± 0.03	4.03 ± 0.04	3.53 ± 0.03	(0.276, 0.513)	(0.268, 0.509)	38	35	4.4
12%	7.63 ± 0.21	7.22 ± 0.16	4.84 ± 0.14	4.78 ± 0.11	(0.334, 0.533)	(0.328, 0.532)	45	40	4.2
15%	6.72 ± 0.16	4.26 ± 0.12	2.98 ± 0.05	2.82 ± 0.07	(0.333, 0.536)	(0.326, 0.533)	45	41	4.2
25%	1.72 ± 0.15	0.87 ± 0.14	0.84 ± 0.08	0.69 ± 0.07	(0.469, 0.508)	(0.418, 0.518)	47	51	4.4
45%	2.38 ± 0.15	1.13 ± 0.14	1.02 ± 0.08	0.79 ± 0.07	(0.462, 0.509)	(0.436, 0.510)	51	56	4.0
65%	2.28 ± 0.22	1.17 ± 0.20	1.69 ± 0.23	1.09 ± 0.22	(0.491, 0.491)	(0.450, 0.484)	50	53	4.2
100%	3.18 ± 0.07	1.15 ± 0.23	1.44 ± 0.18	0.82 ± 0.13	(0.537, 0.454)	(0.464, 0.424)	60	<u>68</u>	<u>3.2</u>

Fig. 4 shows that modifying the baseline structure by adding a 10-nm thin film of *N,N'*-dicarbazolyl-3,5-benzene (mCP) as an electron- and exciton-blocking layer results in EL only from the Pt(otp)₂ with complete absence of any emission from the ETL and host materials, suggesting charge balance and exciton confinement in the emissive layer. Table 2 show that the higher-doped devices actually have a better performance than the lower-doped devices, accentuating the self-sensitization phenomenon and absence of self-quenching. Remarkably, the performance of the best devices in Table 2 is an order of magnitude higher in this device structure than in the analogous structure that does not contain the ultrathin mCP layer; compare with Table 1 data for x = 45 and 65%.

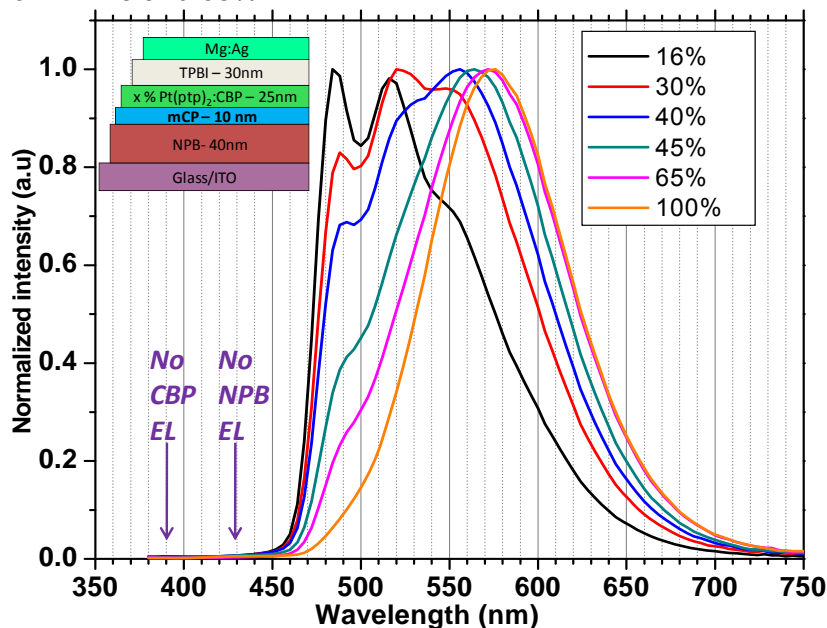


Fig. 4. EL spectra for OLEDs based on Pt(otp)₂ that are partially-optimized by adding the exciton- and electron-blocking material mCP to modify the baseline device structure in Fig. 3. Note the absence of EL from NPB or CBP due to the better charge balance and exciton

confinement in this device structure. The improvement in performance metrics is seen upon comparing Table 2 vs. Table 1 data.

Table 2. Performance metrics for highly-doped OLEDs with the device structure shown in Fig. 4. Device efficacies and color coordinates are reported at peak performance and at 1000 cd/m².

x%	PE _{peak} (lm/W)	PE ₁₀₀₀ (lm/W)	EQE _{peak} (%)	EQE ₁₀₀₀ (%)	CIE _{peak} (x, y)	CIE ₁₀₀₀ (x, y)	CRI _{peak}	CRI ₁₀₀₀	V _T (V)
16%	18.14 ±0.05	11.02±0.04	9.39±0.05	7.83±0.02	(0.285,0.521)	(0.290,0.522)	37	37	4.1
25%	19.12±0.24	9.09±0.08	8.32±0.15	5.64±0.05	(0.322,0.532)	(0.321,0.530)	37	38	<u>3.8</u>
30%	21.30±0.06	14.55±0.08	9.41±0.06	8.39±0.09	(0.342,0.536)	(0.341,0.535)	47	46	<u>3.8</u>
40%	19.70±0.10	12.20±0.10	8.66±0.07	7.22±0.06	(0.376,0.531)	(0.369,0.532)	48	43	<u>3.9</u>
45%	26.30±0.30	19.9±0.19	11.92±0.14	11.07±0.11	(0.417,0.525)	(0.411,0.528)	49	46	<u>3.9</u>
65%	<u>27.80±0.16</u>	<u>21.10±0.12</u>	<u>13.16±0.04</u>	<u>12.40±0.07</u>	(0.446,0.516)	(0.443,0.518)	<u>49</u>	<u>47</u>	<u>3.9</u>

It is important to note the little roll-off in the Table 2 data at 1000 cd/m² in both power efficiency and especially in EQE. Thus, the data herein manifest increase in OLED stability (which roll-off predicts best aside from actual lifetime testing) while simultaneously increasing EQE in the highly-doped devices. This is exactly what we have promised when we submitted the proposal application for this project originally in a literal implementation of the language used in the solicitation. The following is the exact text to this effect reproduced from the introduction of the original proposal application:

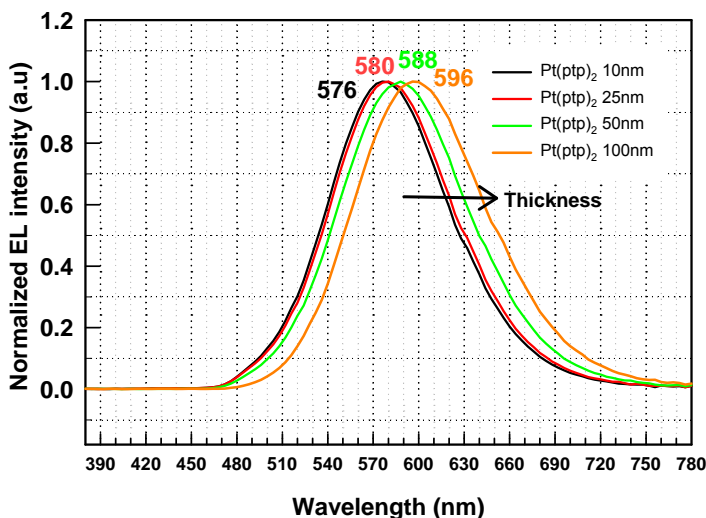
[[We firmly believe that the planned work will make major strides in all research objectives confronting the advancement of OLEDs for practical SSL applications, as identified in Area of Interest 3 of the solicitation. To illustrate, that section of the solicitation states:

“Of special importance under this topic is research that will increase stability while simultaneously increasing IQE.”]]

The ultimate manifestation of the self-sensitization phenomenon is for OLEDs based on neat emissive layers of Pt(otp)₂. Interestingly, Fig. 5 shows that the EL spectra and CRI of such devices are sensitive to the thickness of the neat emissive layer. Although the 50-55 CRI values are not ideal for these light orange OLEDs, they are more than a factor of two better than the CRI values for high pressure sodium so it is possible that they find use in some SSL applications if the performance can be satisfactory (albeit we are addressing the CRI issue; vide infra).

Fig. 6 and Table 3 show that the performance of neat emissive layers of Pt(otp)₂ can be as high as 30 lm/W upon improving the device structure with better ETL and HTL materials. The approach is to replace the materials that sandwich the neat emissive layer by better materials. Thus, we replaced NPB, or both NPB and mCP, by 1,1-bis-(4-bis(4-methyl-phenyl)-amino-phenyl)-cyclohexane (“TPAC”) from the anode side and replaced TPBI by tris(2,4,6-trimethyl-3-(pyridin-3-yl)phenyl)borane (“TPYMB”) from the cathode side. This was done because TPAC can function a triple role (hole transporting, electron blocking, and exciton blocking) based on its HOMO and LUMO energy while TPYMB has superior mobility for electron transport. We have performed multiple device optimizations to systematically introduce these two materials and vary their thickness along with the thickness of the Pt(otp)₂ neat emissive layer. Fig. 6 and Table 3 illustrate some of these optimization efforts. While the work is still ongoing, the available results from partial optimization already amount to a world-record performance for neat emissive

materials. In comparison, neat PtOEP does not exhibit any EL; neat Ir(ppy)₃ exhibits EQE = 0.75% (Forrest; Thompson et al., *APL* **1999**, 75, 4) compared to 15.5% for neat Pt(ptp)₂ (i.e., a factor of 20.7x higher); the highest reported peak power efficiency for neat Ir(III) complexes we are aware of is 7.0 lm/W for neat IrIr₆ (Forrest; Thompson et al., *APL* **2003**, 82, 2422) compared to 30.25 lm/W for Pt(ptp)₂ (i.e., a factor of 4.3x higher). Even higher performance is expected for neat and highly-doped Pt(ptp)₂ OLEDs than what is conveyed in Tables 2 and 3, respectively, upon further optimization of our device structure outlined below.



Thickness Pt(ptp) ₂	λ (nm) FWHM	CRI
10 nm	628 - 532	51
25 nm	632 - 536	51
50 nm	640 - 540	52
100 nm	652 - 552	54

FWHM: Full width at half maximum based on the EL spectra of Pt(ptp)₂ neat films.

Fig. 5. EL spectra for OLEDs based on a neat emissive layer of Pt(ptp)₂ using the same partially-optimized device structure shown in Fig. 4 except for using a neat emissive layer instead of doping. Note the fine tuning of the EL peak maximum and CRI by controlling the thickness of the neat emissive layer.

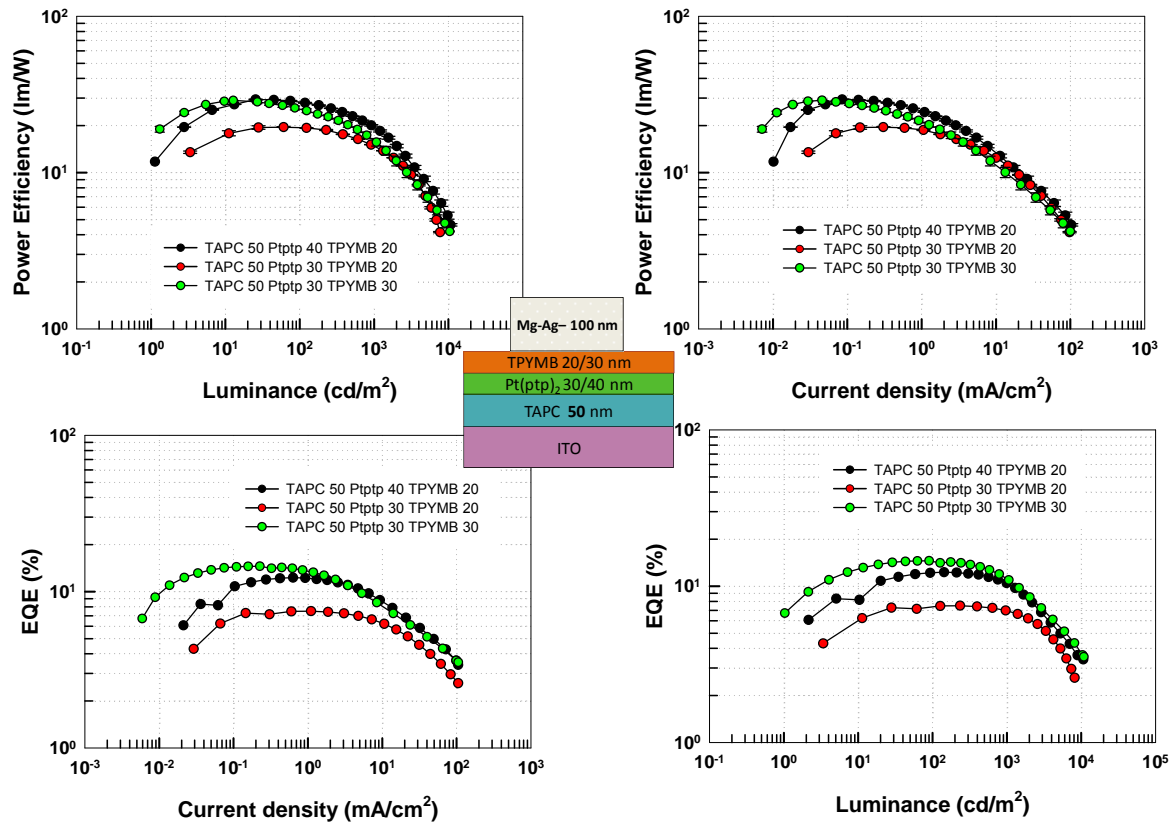


Fig. 6. Plots of EL metrics for OLEDs based on a 30-nm neat emissive layer of $Pt(ppy)_2$ upon partial optimization of the device structure shown in Fig. 3 by replacing the ETL material from TPBI to TPYMB and the HTL material from NPB to TAPC. The order-of-magnitude improvement in performance metrics is seen upon comparing Table 3 data vs. the corresponding data in the last row of Table 1.

Table 3. Performance metrics for OLEDs with a neat emissive layer using the device structure shown in Fig. 6 by partial optimization of the thicknesses of TAPC, TPYMB, and Pt(ptp)₂. Device efficacies and color coordinates are reported at peak performance and at 1000 cd/m².

Device ID	PE _{peak} (lm/W)	PE ₁₀₀₀ (lm/W)	EQE _{peak} (%)	EQE ₁₀₀₀ (%)	CIE _{peak} (x, y)	CIE ₁₀₀₀ (x, y)	LE _{peak} (cd/A)	LE ₁₀₀₀ (cd/A)	V _T (V)
TAPC 60 Ptptp 30 TPYMB 30	28.63 ± 1.95 (30.25)	12.85 ± 0.55 (14.23)	13.97	10.3	(0.50,0.48)	(0.50,0.49)	(38.21)	(27.8)	3.0
TAPC 50 Ptptp 30 TPYMB 30	29.00 ± 0.10 (29.25)	15.65 ± 1.50 (16.30)	14.53	10.95	(0.50,0.48)	(0.50,0.49)	(40)	(31)	2.8
TAPC 40 Ptptp 30 TPYMB 30	26.67 ± 1.10 (28.24)	16.1 ± 0.50 (17.1)	10.48	9.15	(0.50,0.48)	(0.49,0.49)	(29.5)	(26.2)	2.6
TAPC 30 Ptptp 30 TPYMB 30	23.37 ± 1.35 (26.47)	17.03 ± 1.0 (17.80)	9.41	8.64	(0.50,0.48)	(0.49,0.49)	(26.7)	(24.9)	2.5
TAPC 50 Ptptp 30 TPYMB 20	19.43 ± 1.04 (20.16)	15.1 ± 0.10 (15.18)	7.5	7.0	(0.50,0.48)	(0.50,0.49)	(21.5)	(20.1)	2.5
TAPC 50 Ptptp 30 TPYMB 30	29.00 ± 0.10 (29.25)	15.65 ± 1.5 (16.30)	14.53	10.95	(0.50,0.48)	(0.50,0.49)	(40)	(31)	2.8
RUN 2 for 50/30/30	27.55 ± 2.10 (29.5)	16.00 ± 1.52 (17.6)	14.50	10.8	(0.50,0.48)	(0.50,0.49)	(37.35)	(30.2)	2.8
TAPC 50 Ptptp 30 TPYMB 40	25.10 ± 2.1 (26.8)	9.1 ± 0.60 (9.3)	15.5	7.9	(0.50,0.48)	(0.50,0.49)	(44.9)	(23)	3.8
TAPC 50 Ptptp 40 TPYMB 20	29.40 ± 0.25 (29.57)	19.20 ± 0.50 (19.76)	12.30	10.48	(0.50,0.48)	(0.50,0.48)	(34.42)	(31)	2.4
TAPC 50 Ptptp 30 TPYMB 30	29.00 ± 0.10 (29.25)	15.65 ± 1.5 (16.3)	14.53	10.95	(0.50,0.48)	(0.50,0.49)	(40)	(31)	2.8

In order to attain white light, the orange emission of the highly-doped or neat devices with near 30 lm/W performance can be combined with either a deep-blue emission to attain good CIE coordinates (cool WOLED approach) or blue-green emission to attain high CRI (warm WOLED approach). Detailed results from this ongoing effort will be communicated in subsequent reports. To illustrate the latter approach, Fig. 7 shows that we have been able to combine the orange emission from a 100-nm neat Pt(ptp)₂ layer with the blue-green emission from a second emissive layer consisting of a 5% Pt(ptp)₂:CBP lightly-doped film to attain a WOLED with a high CRI (up to 82). Note that no roll-off is obtained in the EQE in this WOLED made from a single emitter.

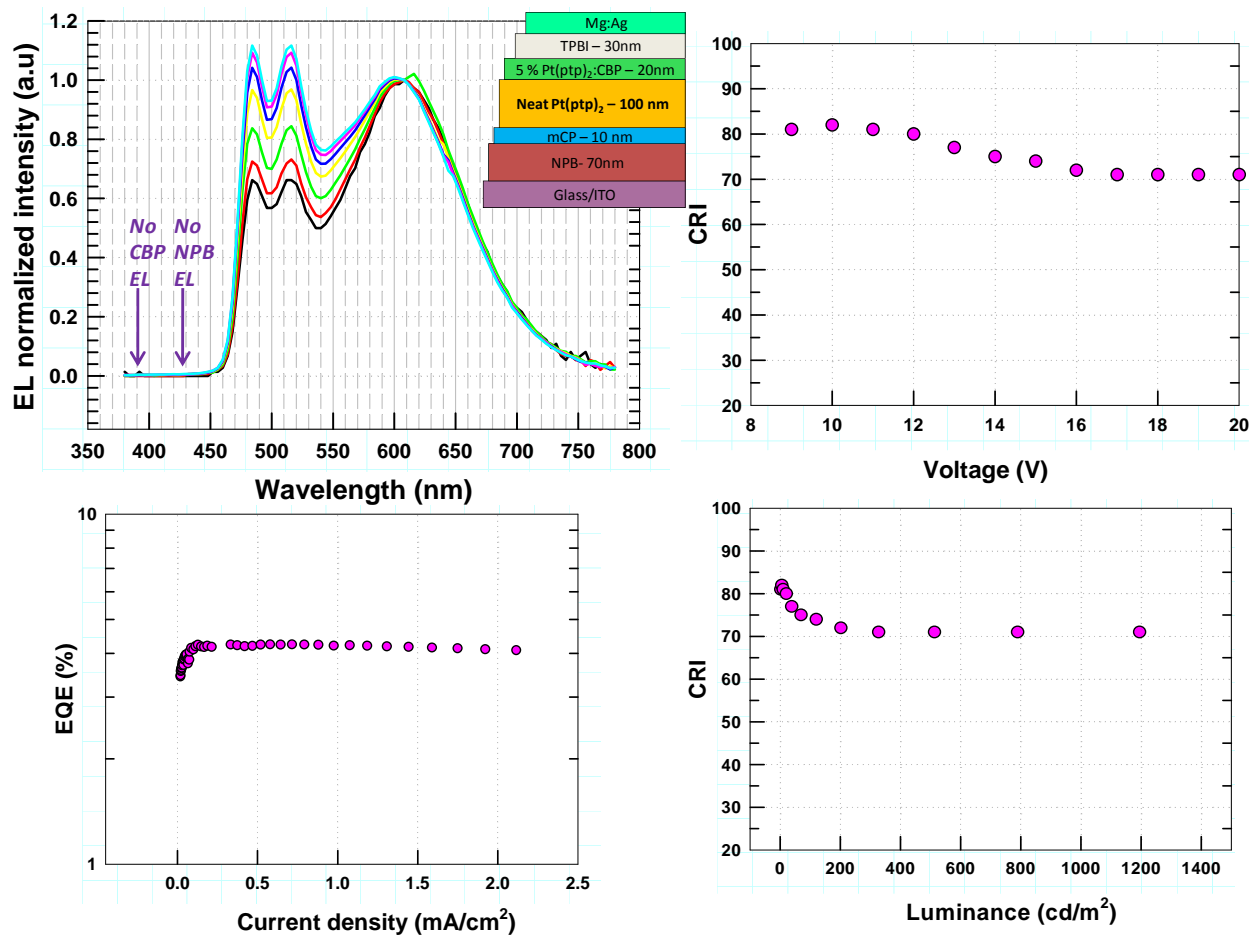


Fig. 7. Plots of EL metrics for WOLEDs based on a single emitter, $Pt(otp)_2$, in two emissive layers: a 100-nm neat layer and a 5%-doped layer in CBP.

The devices were further optimized at later project stages to improve performance, color, processing cost, and/or stability metrics by multiple means to attain the best results mentioned in Section 4 above. The corresponding technical data are overviewed briefly as follows:

a) Turquoise-blue OLEDs with record performance: We demonstrated high-efficiency turquoise-blue electrophosphorescence from our bis[3,5-bis(2-pyridyl)-1,2,4-triazolato]platinum(II) ($Pt(otp)_2$) phosphor doped in 4-(diphenylphosphoryl)-*N,N*-diphenylaniline (HM-A1) host developed by the PNNL lighting group. Organic light-emitting diodes (OLEDs) with 5% $Pt(otp)_2$:HM-A1 attain peak power efficiency of 70.6 lm/W, versus 42.8 lm/W for analogous devices employing the standard turquoise-blue phosphor bis[(4,6-difluorophenyl)-pyridinato- N,C^2](picolato)iridium(III) (FIrpic). Devices with x% $Pt(otp)_2$:HM-A1 exhibit blue emission maxima (λ_{max} ~480 nm) with monotonic increase in excimer/monomer intensity ratio at higher doping levels within 1-10%, causing color shift toward green and less charge balance. Fig. 8 and Table 4 summarize the results.



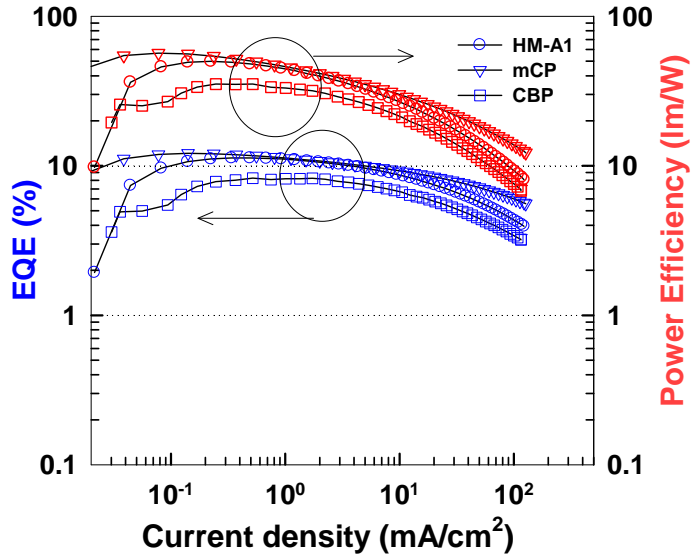


Fig. 8. EQE and PE plots of a 5% Pt(*otp*)₂ doped in various hosts. Device structure: ITO/TAPC 350 Å/Host: 5% Pt(*otp*)₂ 150 Å/PO15 500 Å/LiF-AI.

Table 4. Summary of important parameters for OLEDs with variable doping concentrations of Pt(*otp*)₂ in HM-A1. The subscripts (*peak*) and (1000) refer to the numbers at their peak value and at 1000 cd/m², respectively. The values reported are an average ± standard deviation of 8 devices and the numbers in parentheses are the best-pixel data.

Doping Conc. (%)	1 mA cm ⁻²		EQE _{peak} (%)	EQE ₁₀₀₀ (%)	PE _{peak} (lm W ⁻¹)	PE ₁₀₀₀ (lm W ⁻¹)	λ _{max} (nm)	CIE (x,y)
	Voltage (V)	Luminance (cd m ⁻²)						
1% Pt(<i>otp</i>) ₂	3.94 ± 0.01	358.1 ± 6.0	11.5 ± 1.1 (12.4)	8.4 ± 0.3 (9.0)	37.7 ± 6.8 (44.5)	21.0 ± 0.8 (22.3)	482	(0.19,0.45)
2.5% Pt(<i>otp</i>) ₂	4.13 ± 0.01	568.5 ± 7.5	12.0 ± 0.4 (12.6)	9.7 ± 0.1 (9.9)	59.9 ± 3.5 (65.6)	38.6 ± 0.7 (39.5)	483	(0.22,0.49)
5% Pt(<i>otp</i>) ₂	3.96 ± 0.03	568.4 ± 11.2	11.8 ± 0.6 (13.1)	10.6 ± 0.2 (10.8)	61.2 ± 5.9 (70.6)	40.3 ± 1.2 (41.6)	486	(0.26,0.51)
7.5% Pt(<i>otp</i>) ₂	3.93 ± 0.04	550.2 ± 6.0	11.5 ± 0.2 (12.6)	10.4 ± 0.1 (10.5)	57.8 ± 2.2 (68.1)	40.2 ± 0.7 (41.1)	488	(0.29,0.53)
10% Pt(<i>otp</i>) ₂	3.98 ± 0.03	510.9 ± 21.4	11.1 ± 0.7 (11.6)	9.4 ± 0.5 (9.9)	58.1 ± 3.6 (60.8)	35.2 ± 2.2 (37.9)	490	(0.32,0.54)
5% Flrpic (Control)	4.02 ± 0.02	431.5 ± 12	16.7 ± 0.4 (17.6)	15.4 ± 0.4 (16.0)	40.8 ± 1.7 (42.8)	29.4 ± 0.8 (30.7)	474	(0.16,0.30)

b) Yellow-orange OLEDs with record performance from a neat emissive layer: When we reproduced the same neat devices described in Fig. 6 and Table 3 above but replaced the photospectrometer PR-650 (Photo Research Inc.) detector that we utilized in early stages of the project with a Si photodiode (Hamamatsu – 18 mm x18 mm) that we custom built, the power efficiency was greatly improved by > 50%, from 27 to 41 lm/W. The PR-650 can provide light intensity (luminance) and chromaticity (CIE x,y coordinates, CRI, CCT and EL spectra) simultaneously. However, it suffers from lower sensitivity and longer response times compared to a calibrated Si photodiode. The duration for a normal voltage sweep of 0 to 10 V (0.2 V steps) is >10 min with the PR-650, which affects device efficiencies, compared to merely seconds with a calibrated Si photodiode. The improved results with the calibrated Si photodiode are summarized in Fig. 9a. Further improvement to 79.4 lm/W (80 lm/W for best pixel) was obtained upon using a 100-nm neat film of Pt(otp)₂ that acts as both emissive layer and electron transport layer (due to its n-type semiconducting behavior as we proved in a different NSF-supported project) in combination with mCP as electron-/exciton-blocking layer and TAPC as hole transporting layer (Fig. 9b).

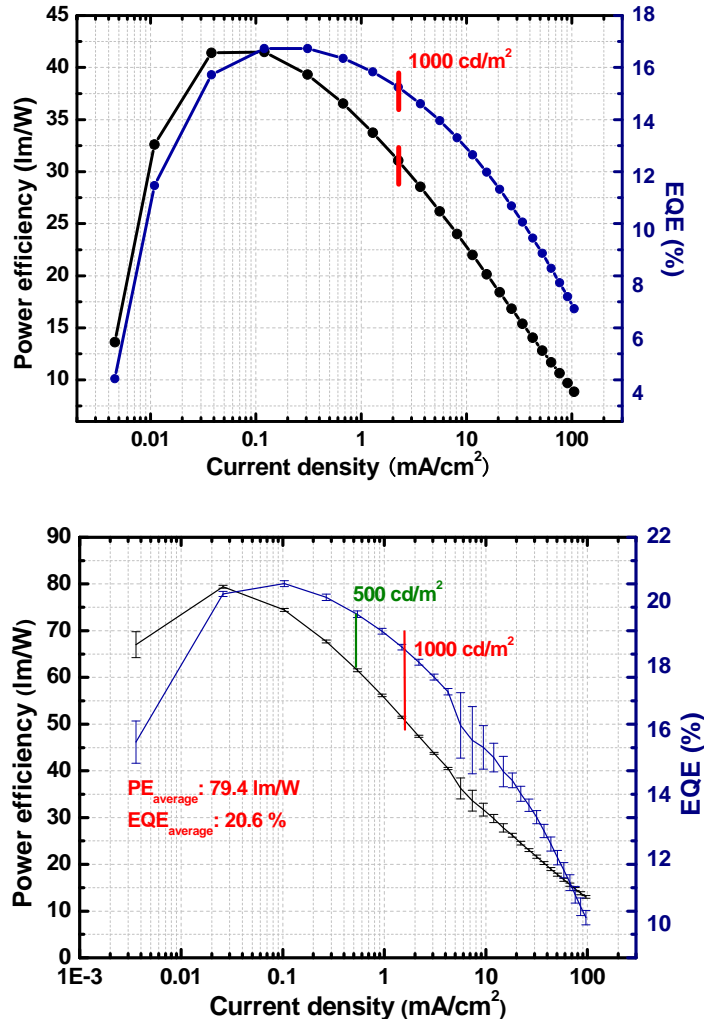


Fig. 9. EQE and PE plots of two neat Pt(otp)₂ devices with device structure as follows:
 (a; top): ITO/TAPC (40 nm)/Pt(otp)₂ (30 nm)/Tpymb (30 nm)/LiF/Al.
 (b; bottom): ITO/TAPC (40 nm)/mCP (10 nm)/Pt(otp)₂ (100 nm)/LiF/Al.

c) Warm-white OLEDs from a single phosphor by a gradient doping device architecture: We demonstrated *single-emitter* warm-white organic light emitting diodes (SWOLEDs) with high efficiency commensurate with that attained by multiple dopants. Broad electroluminescence (EL) spectra have been achieved rendering balanced-white color through simultaneous monomer (turquoise-blue), excimer (green-yellow) and extended excimer (yellow-orange) emissions from low doped ($\leq 5\%$), medium doped ($\sim 10\%$) and neat (100% phosphor with no host) films of $\text{Pt}(\text{ptp})_2$, respectively, in a graded-doping device architecture. We have also demonstrated good control of the recombination zone and its impact on device performance by adjusting the number of emissive layers, their thicknesses and relative positions in the device stack. We have evaluated several simple device structures to optimize efficiency and color of these WOLEDs, keeping in mind low-cost, reliability and high-volume manufacturability. Device optimization has resulted in SWOLEDs with peak power efficiency of $30.4 \pm 1.3 \text{ lm/W}$, EQE of $17.1 \pm 0.1 \%$, and correlated color temperature (CCT) of 3450 K, which is well-within the acceptable range for warm-white according to the MYPF despite the fact that the color rendering index (CRI) is only 62. The devices exhibit negligible roll-off of EQE at lighting brightness (500 cd/m^2 corresponding to 1000 cd/m^2 with 2x out-coupling enhancement as a conservative estimate) maintaining $> 90\%$ of its peak EQE value and remains $> 85\%$ of peak EQE even at double lighting brightness (1000 cd/m^2 corresponding to 2000 cd/m^2 with 2x out-coupling enhancement). Fig. 10 summarizes the results. These data were for devices measured with the PR-650 detector. Based on the aforementioned power efficiency improvement with Si photodiode, we project the gradient-doped devices to attain 45-50 lm/W maximum ($47.3 \pm 2.02 \text{ lm/W}$ with the same improvement ratio), constituting a new world record for a white OLED made with a single phosphorescent emitter. This performance translates to 90-100 lm/W SWOLEDs upon 2x out-coupling enhancement.

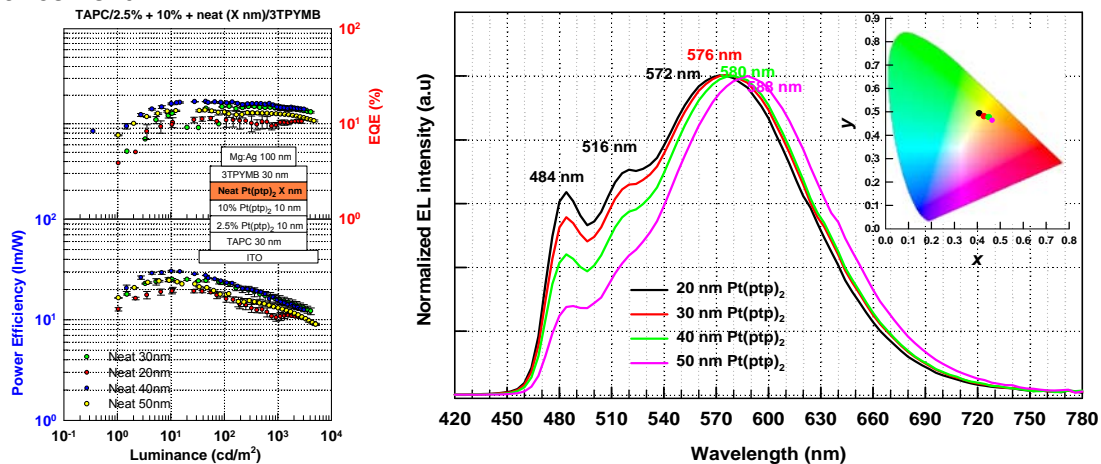
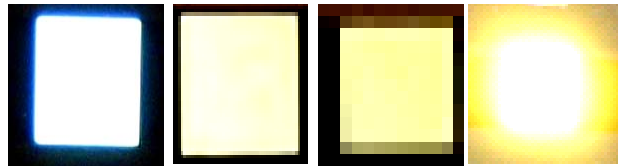


Fig. 10. EQE and PE plots (left) and EL spectra (right) of four gradient-doped SWOLEDs.

d) Tunability of white color metrics: Gradient doping has not been our only strategy to attain balanced white EL color. Other strategies have included: (i) hybrid fluorescent/phosphorescent WOLEDs with up to 82 CRI, some with (0.3, 0.3) CIE coordinates and CCT in the cool-white range, (ii) dual-phosphor doping-free WOLEDs with warm-white CCT and up to 65 CRI, which could be improved at the expense of performance to 75 CRI, and (iii) SWOLEDs consisting of one doped and one neat emissive layer that give rise to warm-white CCT. All these device architectures could attain some degree of tuning each of the three

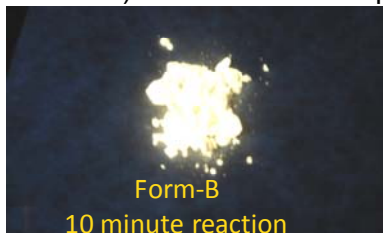
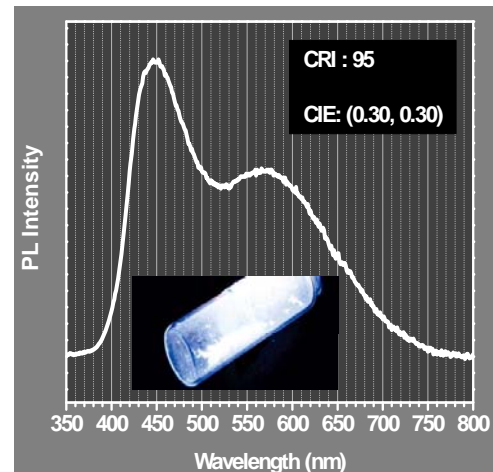


aforementioned color metrics and, in general, we have found that the highest performance is associated with the warmest-white devices.

d) Road map for tomorrow's WOLEDs: This SSL project, upon leverage by another NSF-supported project on electrical characterization, has identified a road map toward attaining white OLEDs with power efficiency that will exceed 150 lm/W (including out-coupling enhancement) at lighting brightness from a single emitter with a simple doping-free device architecture capable of attaining acceptable CRI > 80. Such a road map toward the aforementioned efficiency metric was identified as the goal for next-generation WOLEDs during the OLED panel discussion of the SSL Core technology Workshop in Raleigh, NC (February, 2010). Although this goal is ambitious since it represented nearly 50% improvement of the current technology status at the time (UDC's ~ 100 lm/W WOLEDs), our project team believes that this goal is not only realistic but could be further amended to facilitate the manufacturing aspects by both simplifying the device structure and decreasing material and device production cost without compromise of neither efficacy nor stability. Our road map is based on the aforementioned doping free white and warm-white OLEDs (which we affectionately call "DFW-OLEDs" and "DFW-WOLEDs", respectively, with pun intended to our geographical location). The logic for this road map is as follows:

(i) We have attained doping-free OLEDs with up to 80 lm/W with a single emissive layer consisting of a neat phosphor that also acts as an electron-transport layer due to its n-type behavior that we discovered in the NSF project; see Fig. 9b and discussion thereof in subsection *b*) above. Though these devices fall within the acceptable CCT range so as to merit the "DFW-WOLED" name, they do not exhibit acceptable CRI. The performance of such devices is expected to reach at least 160 lm/W given the aforementioned 2x out-coupling enhancement as a conservative estimate. The slight drop at lighting brightness is expected to be more than offset by reproduction by an OLED manufacturer with better deposition conditions than what we currently have (including more efficient encapsulation and completely *in situ* device deposition without breaking vacuum before cathode deposition like we currently have to do). Thus, new materials that exhibit near 100% PL quantum yield in neat form (like Pt(otp)₂ neat films do) are needed to attain the sought 150 lm/W performance but with acceptable CRI.

(ii) Our screening efforts have identified materials besides Pt(otp)₂ that can emit balanced white light effectively in neat form, including one macromolecular material that has attained 95 CRI via photoluminescence (as shown) and other materials that attained varying degrees of warm- and cool-white light (see picture here for three forms of a single neat material whose white PL color quality could be controlled by microwave reaction time). Such materials provide suitable candidates for



SWOLEDs in bi-layer or tri-layer DFW-OLED device architectures similar to those utilized for Pt(ppy)₂ high-performance devices but will also satisfy the CRI color metric. Likewise, we also have screened other materials that are suitable for dual-emissive-layer DFW-WOLEDs that will consist of a neat blue-turquoise phosphor in one layer and orange-red phosphor in another layer. Several such materials have been screened with neat 100% PL quantum yield in neat form, some offering deeper-blue (than neat Irpic or doped Pt(ppy)₂ films) and others deeper-red (than neat Pt(ppy)₂ films) alternative DFW-WOLED emissive materials so as to attain suitable CRI >80 along with the 150 lm/W performance. However, such materials need to be investigated and scrutinized in functioning OLEDs in future projects.

6. Products and Technology Transfer:

6.1. Patent Applications:

1. Omary, M. A., "Bidentate Square Planar Complexes of Triazolates and Uses Thereof" *United State Patent and Trademark Office; U.S. Application Number 61/188,428; Filing Date: August 8, 2008.*
2. Omary, M. A. "Improved Organic Light-Emitting Diodes from Homoleptic Square Planar Complexes" *United State Patent and Trademark Office; U.S. Application Numbers 61/1756,190; Filing Date: May 7, 2009.*
3. Omary, M. A., "Improved Organic Light-Emitting Diodes from Homoleptic Square Planar Complexes" *International Patent Cooperation Treaty Application Number: PCT/US 09/49938; Filing Date: July 8, 2009.*

6.2. Graduate Theses and Dissertations:

1. Bhansali, U., "Fabrication and Optimization of Phosphorescent Organic Light Emitting Diodes for Solid-State Lighting Applications", Ph. D. Dissertation in Materials Science and Engineering, University of Texas-Dallas, 2010.
2. Li, M., "Device Engineering for Enhanced Efficiency from Platinum(II) Phosphorescent OLEDs", Ph. D. Dissertation in Materials Science and Engineering, University of North Texas, 2010.
3. Lin, M.-T., "Transient Electroluminescence Determination of Carrier Mobility, Charge Trapping Effects on the Efficiency of Heavily Doped Organic Light-Emitting Diodes, and Eu(III) Based Red OLEDs", Ph. D. Dissertation in Materials Science and Engineering, University of North Texas, 2010.
4. Chen, W.-H., "Triimine Complexes of Divalent Group 10 Metals for Use in Molecular Electronic Devices", Ph. D. Dissertation in Chemistry, University of North Texas, 2010.
5. Arvapally, R., "Sensitization of Lanthanide and Organic-Based Phosphorescence via Energy Transfer and Heavy-Atom Effects", Ph.D. dissertation, University of North Texas, 2009.
6. Sinha, P. "Phosphorescent Emissions of Coinage Metal-Phosphine Complexes: Theory and Photophysics", Ph.D. dissertation, University of North Texas, 2009.
7. Determan, J. J. "Photophysics and Photochemistry of Copper(I) Phosphine and Collidine Complexes: An Experimental/Theoretical Investigation", Ph.D. dissertation, University of North Texas, 2011.

8. Determan, J. J. "Computational Studies of Bonding and Phosphorescent Properties of Group 12 Oligomers and Exciplexes", M.S. thesis, 2008.

6.3. Publications:

i- Related to Device Fabrication and Testing

1. Li, M.; Chen, W.-S.; Lin, M.-T.; Omary, M. A.; Shepherd, N. D. "Near-white and tunable electrophosphorescence from bis[3,5-bis(2-pyridyl)-1,2,4-triazolato]platinum(II) based organic light emitting diodes", *Org. Electron.* **2009**, *10*, 863-870.
2. Bhansali, U.; Jia, H.; Quevedo-Lopez, M. A.; Gnade, B. E.; Chen, W.-H.; Omary, M. A. "Controlling the carrier recombination zone for improved color stability in a two-dopant fluorophore/phosphor white organic light emitting diode", *Appl. Phys. Lett.* **2009**, *94*, 203501.
3. Bhansali, U.; Polikarpov, E.; Swensen, J. S.; Chen, W.-H.; Jia, H.; Gaspar, D. J.; Gnade, B. E.; Padmaperuma, A. B.; Omary, M. A. "High-efficiency turquoise-blue electrophosphorescence from a Pt(II)-pyridyltriazolate complex in a phosphine oxide host", *Appl. Phys. Lett.* **2009**, *95*, 233304.
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5. Li, M.; Chen, W.-S.; Lin, M.-T.; Oswald, I.; **Omary, M.**; Shepherd, N. D. "High efficiency orange-red phosphorescent organic light emitting diodes based on a Pt(II)-pyridyltriazolate complex from a structure optimized for charge balance and reduced efficiency roll-off", *J. Phys. D: Appl. Phys.* **2011**, *44*, 365103/1-365103/5.
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ii- Related to Material Screening and Characterization

1. Bhansali, U. S.; Quevedo Lopez, M. A.; Jia, H.; Alshareef, H. N.; Cha, D. K.; Kim, M. J.; Gnade, B. E. "Characterization of Organic Thin Films Using Transmission Electron Microscopy and Fourier Transform Infra Red Spectroscopy", *Thin Solid Films* **2009**, *517*, 5825.
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3. Yang, C.; Elbejrani, O.; Palehepitiya Gamage, C. S.; Dias, H. V. R.; Omary, M. A. "Luminescence enhancement and tuning via multiple cooperative supramolecular interactions in an ion-paired multinuclear complex", *Chem. Commun.* **2011**, *47*, 7434-7436 **(INVITED BY THE RSC FOR A SPECIAL THEMATIC ISSUE ON SUPRAMOLECULAR CHEMISTRY)**
4. Satumtira, N. T.; Mahdi, A.; Chehbouni, M.; Elbejrani, O.; Omary, M. A. "Novel Method for Waste Analysis Using a Highly Luminescent Diplatinum (II) Octaphosphite Complex as a Heavy Metal Detector", *Ceramic Transac.* **2011**, *227*, 279-288.

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- b) Freestone, N. P. "MOFs for High-Density Gas Adsorption", *Chemistry and Industry* **2008**, March 10 Applied Highlights; URL: <http://www.entrepreneur.com/tradejournals/article/177363186.html>.
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6.4. Conference Proceedings:

1. Omary, M. A. "Bright phosphors for white and monochrome OLEDs based on transition metal complexes besides Ir(III) phenylpyridines", *SPIE Photonic Devices and Applications - Organic Light Emitting Materials and Devices XIV Symposium*, San Diego, CA, August 1-5, 2010, Paper 7776-8. **(INVITED SPEAKER)**
2. McDougald, R. N., Jr.; Tekarli, S. M.; Cundari, T. R.; Omary, M. A. "Trinuclear metal complexes exhibiting metalloaromaticity", *239th ACS National Meeting*, San Francisco, CA, March 21-25, 2010, INOR-856.

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