Building Technologies Solid State Lighting (SSL) Program
FINAL REPORT

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Approach to Low-Cost High-Efficiency OLED Lighting

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DISCLAIMER

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1. EXECUTIVE SUMMARY

The objective of the project is to develop an integrated substrate which organic light emitting diode (OLED) panel developers will be able to employ the integrated substrate to fabricate OLED devices with performance and projected cost meeting the MYPP targets of the Solid State Lighting Program of the Department of Energy. The project will optimize the composition and processing conditions of the integrated substrate for OLED light extraction efficiency and overall performance. The process will be further developed for scale up to a low-cost process and fabrication of prototype samples. The encapsulation of flexible OLEDs based on this integrated substrate will also be investigated using commercial flexible barrier films.

Conventional ITO/glass substrates have been the most dominant transparent conductive electrodes for OLED fabrication; however, owing to its increasing material cost, high deposition temperature, and brittle nature of oxide thin films, approaches to develop potential ITO/glass alternatives have gathered general interest. In this two-year program, UCLA together with Polyradiant Corp. planned to develop an integrated plastic substrate with high efficiency to a maturity level at which OLED lighting panel developers will be able to employ the integrated substrate to fabricate OLED devices with performance and projected cost meeting the MYPP targets of the SSL program.

The majority of this project was directed toward the development of a fabrication process of a flexible substrate for light extraction using refractive index matching and light scattering techniques. Materials chosen for this study are silver nanowires and carbon nanotubes as the conductors, barium strontium titanate nanoparticles as the light scattering material, and high index polymers as the index matching layer. The composition and nanostructure of the integrated substrates were refined for the combined properties showing:

- Enhanced OLED efficacy performance by a factor of ~2 (up to 2.7) over the ITO/glass reference device
- Comparable figure-of-merit sheet resistance (~15Ω/sq at 92% transmittance) to ITO/glass.
- Similarly low surface roughness (Ra < 2nm) compared to high quality ITO/glass
- Enhanced mechanical flexibility (bendable to 1 mm radius of curvature)
- Almost no change in emission spectrum (color) over a wide range of observation angles (up to 70 degrees)

Furthermore, the integrated substrates are fabricated based on cost effective solution processes, and the projected cost of the integrated substrate to include the anode, light extraction architecture, and barrier substrate is ≤ $2.5 per klm, a target set forth by the SSL program such that the overall cost of the lighting panels can be lowered to $10 per klm.

Altogether, these properties show great promise for the integrated substrates to be adaptable by OLED panel developers. This project has developed a solid foundation for flexible plastic substrates for OLEDs in light applications. Further developments in roll-to-roll processes and scaling for large panel sizes will move this technology from development to commercialization in the future and contribute to energy conservations.

2. METRICS

The work plan for this project was separated into four main tasks. Task 1 focused on refining the composition and nanostructure of the integrated substrate for low sheet resistance, low surface roughness, and large enhancement of light extraction efficiency. Task 2 focused on the fabrication of OLEDs on the integrated substrates, the optimization of the device structure for high external quantum efficiency (EQE), low driving voltage, long lifetime, and the comparison of enhancement factor to ITO/glass reference devices. Task 3 focused on the upscaling of the integrated substrates with prototypes samples sent to OLEDWorks for the fabrication of the state-of-the-art OLED lighting devices. Task 4 focused on integrating commercially available flexible barrier films to fabricate encapsulated flexible OLED and to evaluate the device performance and stability.

2.1 Accomplishments

- Demonstrated integrated substrates with >1 square inch area, 5 Ohm/sq sheet resistance, average surface profile peak-valley heights (Ra) at ~2 nm, and 150% enhancement of light extraction.
- Demonstrated prototype integrated substrate with 8"x11" area with < 10 ohm/sq sheet resistance, < 2 nm surface roughness (Ra).
- Demonstrated white OLED on an integrated substrate with CRI of 83 using a fluorescence dye in the integrated substrate with a power efficacy of 81 lm/W at ~40% EQE at 1,000 cd/m^2.
- Demonstrated white OLED on an integrated substrate with a power efficacy of 107 lm/W and EQE of ~ 49% at 1,000 cd/m^2.
- Delivered prototype substrate samples to OLEDWorks (OLED developer) for OLED fabrication with results showing an enhancement factor of 1.9X at power efficacy of 50.1 lm/W and EQE of 85.7%.
- Demonstrated flexible OLED on commercial 3M barrier films with a power efficacy of 81 lm/W at 1,000 cd/m^2.
- Demonstrated flexible OLED on willow glass as a barrier film with a power efficacy of 100.8 lm/W at 1,000 cd/m^2.

2.2 Major Milestones Summary

Milestone	Description	Verification Process	Anticipated Completion
Year 1			
1.1	Integrated substrate obtained with >1 square inch area each, 10 Ohm/sq sheet resistance, average surface profile peak-valley heights (Ra) ~2 nm, and 100% enhancement of light extraction.	Resistance and roughness directly measured. Light extraction determined from OLED	Q2 (Completed on time)
1.2	Integrated substrate obtained with >1 square inch area each, 5 Ohm/sq sheet resistance, average surface profile	Resistance and roughness directly measured. Light	Q4 (Completed on time)

	peak-valley heights (Ra) ~2 nm, and 150% enhancement of light extraction.	extraction determined from OLED	
2.1	White OLED demonstrated on integrated substrate with CRI >75, 40% EQE and 80 lm/W at 1000 cd/m^2.	Measurement based on current, voltage, and calibrated light intensity	Q4 (Completed on time)
3.1	Prototype integrated substrate fabricated with 8"x11" area, 10 Ohm/sq sheet resistance, and 2 nm surface roughness, and samples delivered to OLED developers.	Sample will have been delivered to OLED developers	Q3 (Completed on time)
Year 2			
1.3	Multi-layer structured OLED, comprising at least a hole-transporting layer, an emissive layer, an electron-transport/injection layer, and the integrated substrate, demonstrated with 50% EQE, or 2X that of the control device on ITO/glass.	Evaporated OLED showing 2X EQE than control on ITO/glass	Q5 (Completed on time)
2.2	White OLED demonstrated with CRI >80, 60% EQE and 150 lm/W efficacy at 1000 cd/m^2.	Measurement based on current, voltage, and calibrated light intensity	Q8 (90% Completion)
4.1	A flexible OLED based on a barrier substrate with 80 lm/W efficacy at 1000 cd/m^2. The barrier substrate is based on a commercial barrier film developed specifically for flexible OLED. The flexible OLED does not reveal any crack formation in the barrier layer that could lower the barrier property.	Measurement based on current, voltage, and calibrated light intensity	Q6 (Completed on time)
4.2	A flexible white OLED based on a barrier substrate with CRI >80 and 150 lm/W efficacy at 1000 cd/m^2. The barrier substrate is based on a commercial barrier film developed specifically for flexible OLED. The flexible OLED does not reveal any crack formation in the barrier layers that could lower the barrier property.	Measurement based on current, voltage, and calibrated light intensity	Q8 (90% Completion)

3. BACKGROUND

The core basis of this project is to develop a flexible integrated substrate with enhanced light extraction. The primary motivation arises from the shortcoming of convention indium tin oxide (ITO) in that only a small fraction of the light generated in the device (~ 20-25%) can escape due to total internal reflection, especially limited by the waveguide mode due to the high refractive index of ITO (n ~ 1.9) which can trap up to 20-30% of the generated light (Figure 1a). Additions of external extraction structures can partially overcome inefficiencies of conventional ITO but often adds significant costs to manufacturing processes of the substrates. Furthermore, the unique OLED feature to offer flexibility will enable completely new design options for various applications. ITO is innately brittle and warrants a flexible replacement.

A silver nanowire (AgNW) based integrated substrate can alleviate both of these shortcomings. First, the replacement of ITO with silver nanowires removes the highest refractive index layer in the OLED stack and reduces the trapped light in the waveguide mode at the ITO/substrate interface. Second, silver nanowires can withstand mechanical deformation due to bending and sliding. By embedding silver nanowires in a polymer matrix, the surface roughness of the integrated substrate can be reduced to be suitable for OLEDs. Moreover, a solution-based light scattering layer made of nanoparticles provides a low-cost alternative for light extraction from the substrate mode (Figure 1b).

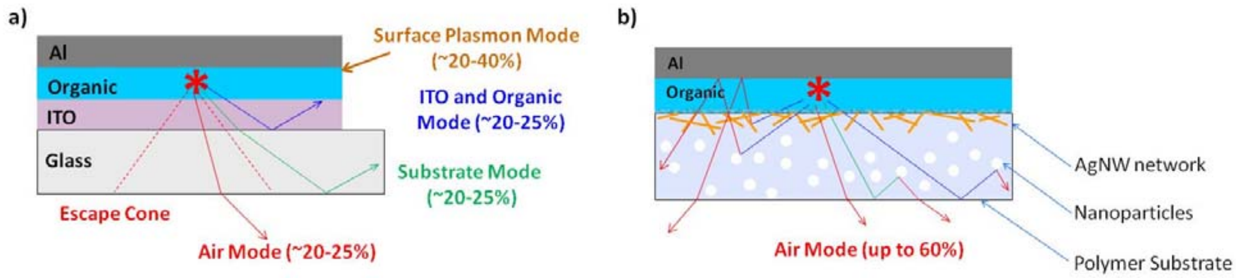

Figure 1: a) Schematic diagram of the mode distributions for OLED fabricated on an ITO/glass substrate. b) Schematic diagram of the mechanisms of light extraction for the integrated substrate.

4. WORK SUMMARY

Tasks in Year 1 consists of the development of the fabrication method of the integrated substrates, including the optimization of the silver nanowire coating, the light scattering layer, and the transfer process, performance testing of the integrated substrates used for fabrication of white OLED, and upscaling of the integrated substrate to send samples to an OLED developer. Tasks in Year 2 consists of further optimization of the integrated substrate structure to further enhance light outcoupling, continual testing of the integrated substrate with an OLED developer, and testing of the integrated substrate fabricated on a barrier substrate.

4.1 Year 1

4.1.1 Developing the fundamentals for integrated substrate fabrication

The fundamental of the fabrication process of the integrated substrate was developed in Year 1. AgNWs were selected as the conductive material because of the large length-to-diameter aspect ratios and the ability to maintain their conductivity during/after mechanical deformation. Improved AgNW inks were developed using surfactants for better wetting and leveling control and coated using a Meyer-rod drawdown machine to coat AgNW films down to a sheet resistance of 5 ohm/sq with > 8”x11” area (Figure 2). A proprietary monomer solution was developed to disperse barium strontium titanate nanoparticles within a polymer matrix for efficient light scattering and was demonstrated to transfer coated AgNW films after UV polymerization resulting in surface roughness < 2 nm.

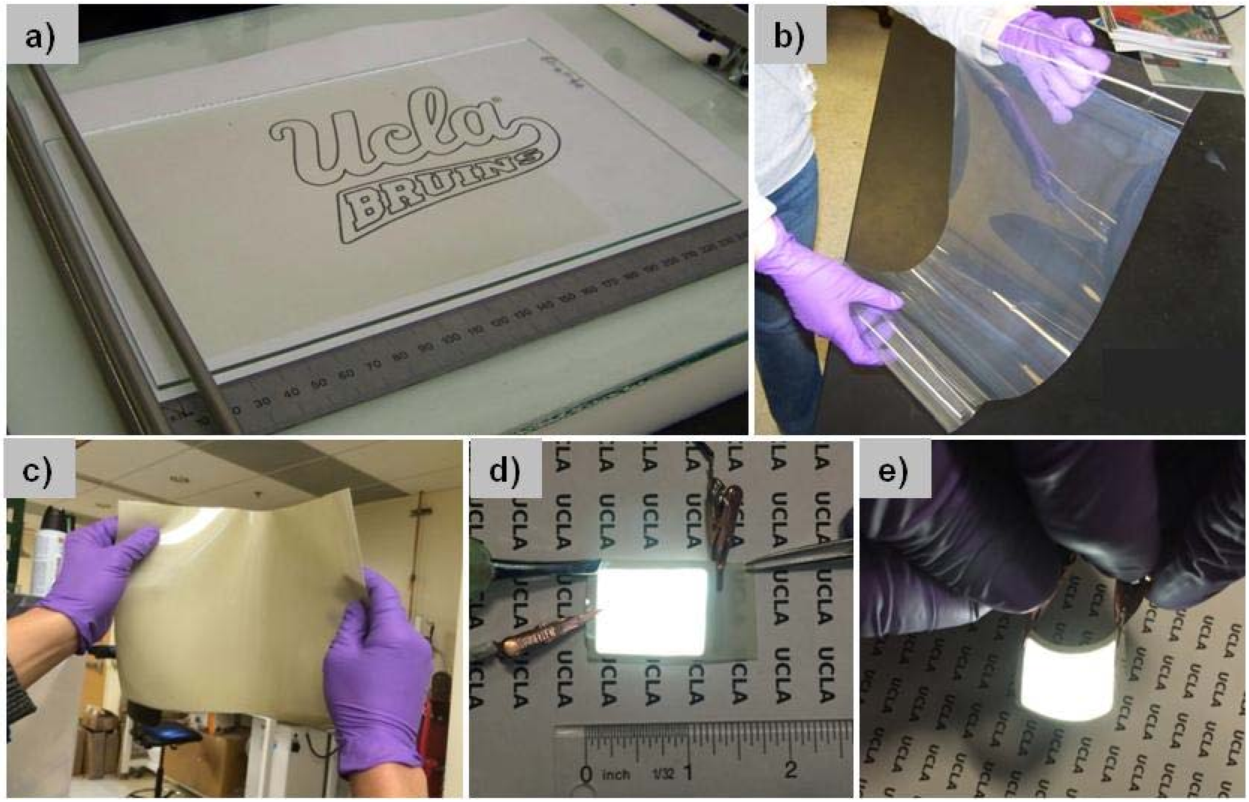

Figure 2: (a) AgNW coating setup using a Meyer-rod drawdown machine. (b,c) Integrated substrate with AgNWs embedded in a polymer matrix. (d,e) Solution-processed white OLED fabricated on an integrated substrate.

4.1.2 Device Performance Testing at UCLA

Initial testing of the integrated substrates was completed using solution processed white OLEDs fabricated with a sandwich structure of substrate / anode / PEDOT:PSS / Emissive Layer (from Cambridge Display Technology) / CsF / Al on both ITO/glass and integrated substrates. The integrated substrates consist of a layer of AgNWs embedded in the light scattering layer. Images of the fabricated devices are shown in Figure 2. The integrated substrates have demonstrated more than 2X enhancement in comparison to the ITO/glass reference. Current efficiency (CE), power efficacy (PE), and EQE results are shown in Table 1.

Evaporated small molecular white OLEDs were fabricated with a sandwich structure of substrate / anode / HTL / Yellow Phosphorescent Emitter / Blue Fluorescent Emitter / ETL / CsF / Al on both ITO/glass and integrated substrates. Similar to the solution processed OLEDs, the devices fabricated on the integrated substrates have also demonstrated more than 2X enhancement in comparison to the ITO/glass reference. CE, PE, and EQE of the fabricated devices are shown in Figure 3. The CRI of these devices is only ~50 due to the use of only two chromophores in the device structure; further work was carried out to improve the CRI in Year 2.

Table 1. Performance of Solution-processed White OLED Devices

	Current Efficiency (cd/m^2)				Power Efficacy (lm/W)				EQE (%)			
	Max	@ 1200 cd/m^2	@ 6900 cd/m^2	@ 11000 cd/m^2	Max	@ 1200 cd/m^2	@ 6900 cd/m^2	@ 11000 cd/m^2	Max	@ 1200 cd/m^2	@ 6900 cd/m^2	@ 11000 cd/m^2
ITO/Glass	18.1	18.1	17.4	17.0	18.8	16.3	12.2	10.7	9.8	9.8	9.4	9.2
Integrated Substrate	39.6	39.2	36.6	36.4	40.7	35.6	20.9	19.0	21.1	20.9	19.5	19.4

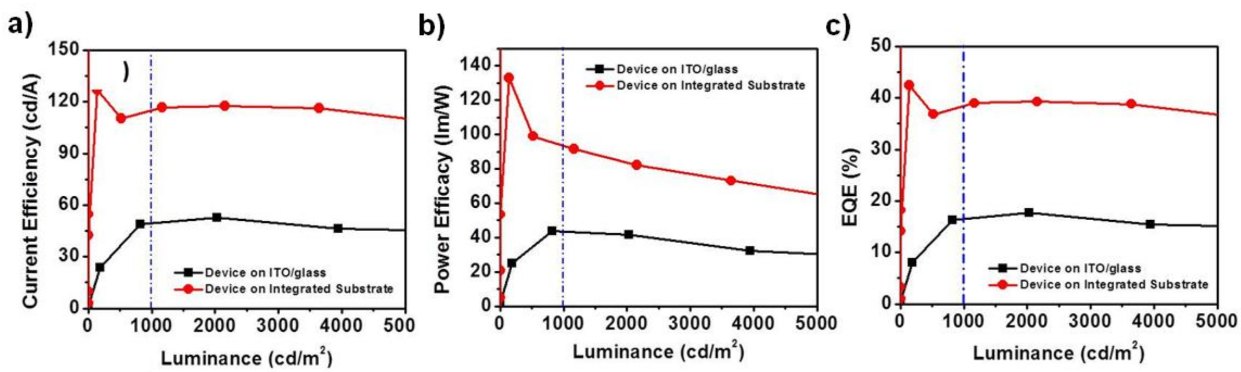

Figure 3: Device performance of evaporated white OLEDs.

4.1.3 Device Performance Testing with OLED Developer

Integrated substrates were fabricated at 2.5"x2.5" substrate size and sent to OLEDWorks for device fabrication and evaluation. A proprietary short reduction layer and a standard 2-stack white OLED was coated on the integrated substrates. Among the tested integrated substrate samples, the best OLED device with an area of 3.0 cm^2 showed a CE of 153.0 cd/A, PE of 62.0 lm/W, EQE of 77.0% at a CRI of 91. A typical behavior of a white OLED fabricated on ITO/glass reference without external light extraction is color shifting with angle as shown in the spectral curve in Figure 4. In contrast, the diffuse scattering nature of our light extraction technique allows white OLED fabricated on the integrated plastic substrates to demonstrate nearly no spectral shift with respect to observation angles.

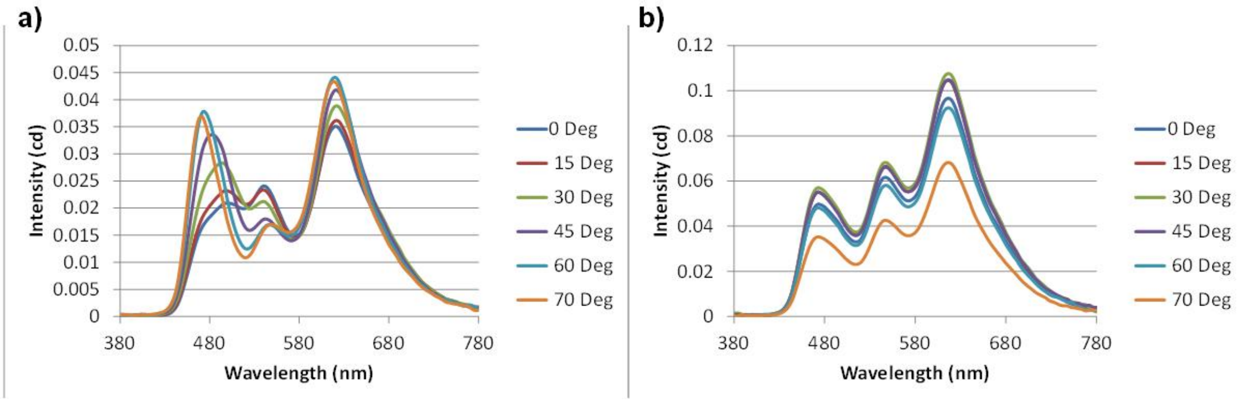

Figure 4: Spectral curves of the fabricated white OLED on ITO/glass substrate (a) and integrated substrate (b) measured at different angles from 0 to 70 degrees.

4.1 Year 2

4.2.1 Improving Integrated Substrate by Modifying Substrate Structure

To increase the CRI of the evaporated small molecule white OLED devices fabricated at UCLA, an additional chromophore was added in the integrated substrate to down convert photons for color tuning. A fluorescence dye, FBtF, was developed, synthesized, and characterized at UCLA. The fluorescence dye was dispersed in the monomer solution of the integrating substrate and UV cured along with the light scattering layer. White OLED fabricated on the integrating substrate containing FBtF achieved a CRI of ~ 83 while maintaining ~ 2X light extraction (Figure 5). The versatility of the fabrication process allows for easy modifications to the constituents within the integrated substrate. This advantage can also be readily adopted by OLED panel developers for additional control in specific spectral power distribution based on the demands of their applications.

A high index polymer layer (n ~ 1.7) was introduced to the integrated substrate for microcavity tuning to further enhance light extraction. The high index polymer layer was used to embedded the AgNW conductive electrode and is positioned on top of the light scattering layer (Figure 6). A white OLED fabricated on this substrate showed further improvement in light extraction, achieving up to a PE of ~ 107 lm/W and an EQE of 49%. In comparison with the ITO/glass reference device, these devices demonstrated ~ 2.7X in enhancement and are believed to be benefited from both internal and external extraction modes.

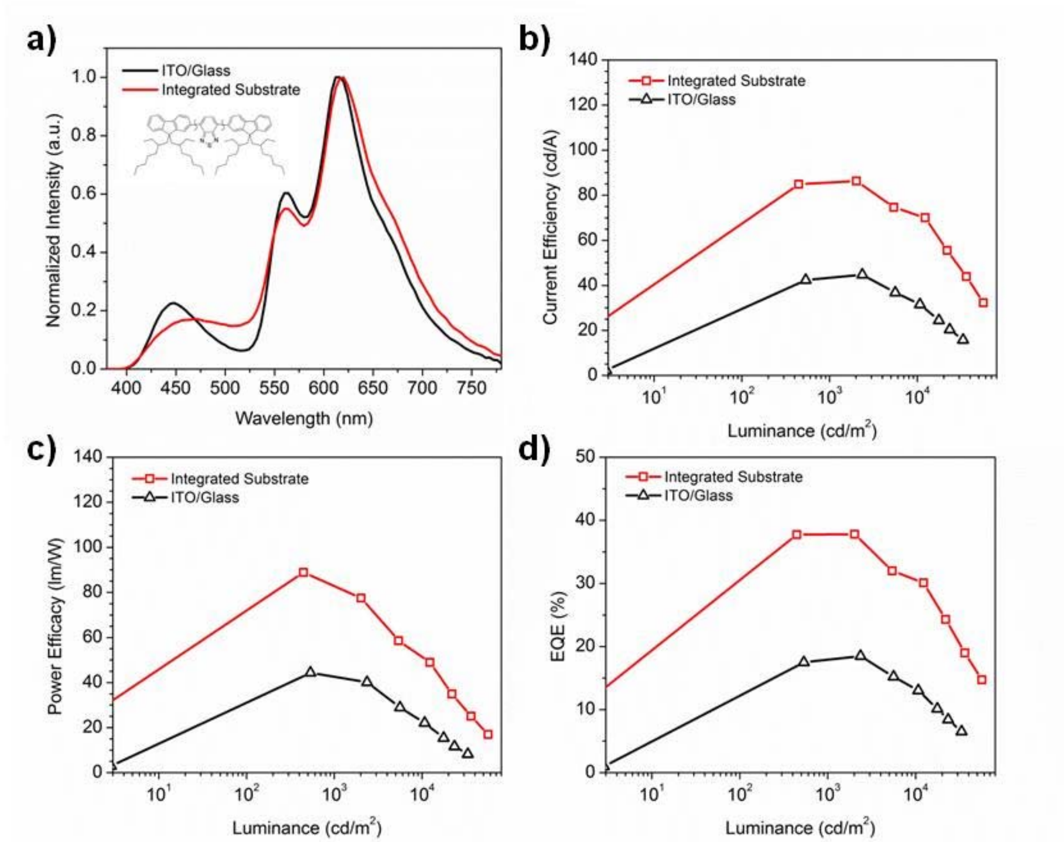

Figure 5: (a) Spectral curve of white OLEDs with improved CRI (Inset shows molecular structure of FBtF. (b, c, d) Device performance showing CE, PE, and EQE vs. luminance.

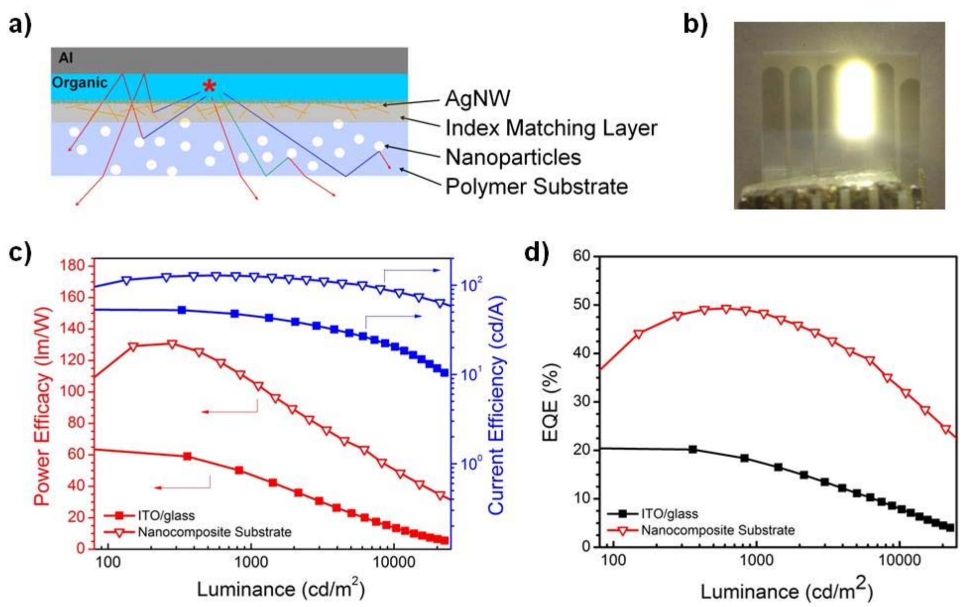

Figure 6: (a) Schematic diagram of integrated substrate with an index matching layer. (b c, d) Image and device performance of a white OLED fabricated on an integrated substrate.

4.2.2 Continual Device Performance Testing with OLED Developer

Additional integrated substrates were sent to OLEDWorks for device fabrication and evaluation. A proprietary short reduction layer, an insulator layer, and a 3-stack white OLED were coated on the integrated substrates. The devices were encapsulated with laminated getter foil. The best OLED device on the integrated substrate with an area of 3.0 cm^2 showed nearly 1.9X enhancement in comparison to ITO/glass reference. Details of results are shown in Table 2. Additionally, microlens as external extraction layer (EEL) were added to the bottom of the devices for comparison. With the addition of the EEL, the EQE of the ITO/glass reference increased from 44.9% to 84.6%. The EQE of the integrated substrate nearly did not change with the EEL layer, indicating that the substrate mode of the device was efficiently extracted by the light scattering technique.

Table 2. Device Performance Results from OLEDWorks

	Without External Extraction layer						With External Extraction layer					
	Voltage (V)	CE (cd/A)	PE (lm/W)	CIEx	CIEy	EQE (%)	Voltage (V)	CE (cd/A)	PE (lm/W)	CIEx	CIEy	EQE (%)
ITO/Glass	9.6	75.5	24.7	0.445	0.342	44.9	9.6	166.0	54.3	0.436	0.377	84.6
Integrated Substrate	11.4	182.0	50.1	0.446	0.418	85.7	11.6	181.0	49.0	0.449	0.419	85.5

4.2.3 Testing of Integrated Substrates on Commercial Barrier Films

Device performance of white OLEDs on integrated substrates fabricated on commercial barrier films was tested. The commercial barrier films chosen for this study were the 3M plastic barrier films and the flexible willow glass from Corning. Integrated substrates were fabricated on both types of barrier films, and the resulting flexible OLEDs did not reveal any crack formation in the barrier layer that could lower the barrier property. Since both types of barrier films are optically transparent, the light extraction enhancement of the resulting substrates was demonstrated to exhibit ~ 2X light enhancement. For the devices fabricated on the Corning Willow glass, the resulting devices achieved a CE of 96.2 cd/A and a PE of 100.8 lm/W (at ~ 2X enhancement to ITO/glass control). This reveals that the efficiency of the OLEDs would decrease by less than 10% even with the addition of a commercial barrier film to the integrated substrate and shows the potential of the integrated substrates for reliable stability from oxygen and moisture while maintaining its high light extraction efficiency.

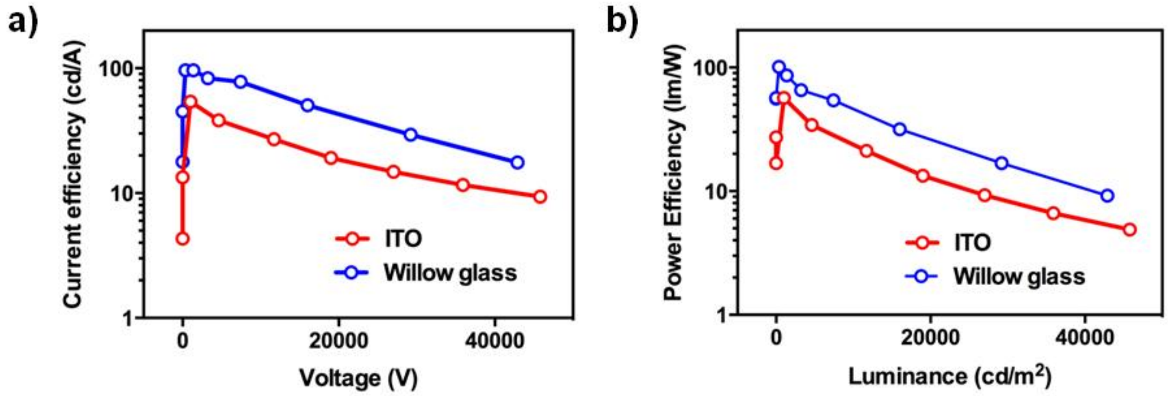

Figure 7: Device performance of a white OLED fabricated on an integrated substrate fabricated on the flexible Corning Willow glass.

5. CONCLUSION

During this two-year program, the flexible integrated substrates have been developed with high efficiency light outcoupling for use in OLED lighting applications. We have demonstrated the development of an integrated plastic substrate technology with 5 ohm/sq sheet resistance, surface roughness less than 2 nm, nearly no color shifting with observation angles, matching or exceeding figures of merit in comparison with the relatively expensive OLED ITO/glass substrates while offering mechanical flexibility for innovative OLED design options. Light extraction has been enhanced to a factor of ~ 2X (up to 2.7X) in reference to ITO/glass control, primarily by extracting the substrate mode and to a lesser degree the waveguide mode. This process has been adopted to convert commercial barrier films and flexible glass substrates into integrated substrates with similarly enhanced OLED performance. The process was also demonstrated with the versatility to integrate additional constituents for coloring tuning.

Currently, this integrated plastic substrate technology is close to maturity for product development. Laboratory results have revealed the potential for efficient light extraction with an architecture that requires low-cost manufacturability; however, the laboratory process has relatively low yield, which could be overcome in a well-controlled clean room environment and on more advanced processing equipments. Going forward, we are procuring mew projects and funding to build up the advanced processing facility for upscaling of the integrated substrates to supply OLED panel developers and manufacturers. Further work will be done to improve the integrated substrate architecture to enhance the internal light extraction. Our commercialization target is to manufacture the integrated substrate at ≤ \$2.5/klm for the fabrication of flexible white OLEDs with 150 lm/W efficacy.

6. Journal Publications And Patents

Patent Application:

- Efficient light extraction of organic light-emitting diodes on a fully solution-processed flexible substrate: UCLA invention disclosure; provisional patent application to be filed.

Publications:

- Chen, D., Zhao, F., Tong, K., Saldanha, G., Liu, C. and Pei, Q. "Mitigation of Electrical Failure of Silver Nanowires under Current Flow and the Application for Long Lifetime Organic Light-Emitting Diodes", Advanced Electronic Materials. DOI: 10.1002/aelm.201600167 (2016).
- Liang, J., Tong, K. and Pei, Q., 2016. A Water-Based Silver-Nanowire Screen-Print Ink for the Fabrication of Stretchable Conductors and Wearable Thin-Film Transistors. Advanced Materials, 22, 5986–5996 (2016).
- Fangchao Zhao, Ying Wei, Hui Xu, T. Ahamad, S. M. Alshehri, Dustin Chen, Qibing Pei, Dongge Ma, "Spatial Exciton Allocation Strategy with Reduced Energy Loss for High-Efficiency Fluorescent/Phosphorescent Hybrid White Organic Light-Emitting Diodes" manuscript submitted for publication.
- K. Tong, et al. "Efficient light extraction of organic light-emitting diodes on a fully solution-processed flexible substrate", manuscript to be published.