Quantum Dot Light Enhancement Substrate for OLED Solid-State Lighting

Final Report

Award No. DE-EE0000628

DoE Round V, Core Technology Project:
Program Area of Interest 5:
OLED Encapsulation and Substrates

Sean Evans, NETL Project Manager

James M. Perkins, Ph.D. (Project Leader),
Matthew Stevenson, Gagan Mahan

Co-Principal Investigators:

Seth Coe-Sullivan, Ph.D.
617.607.9701; SCoe-Sullivan@QDVision.com
&
Peter Kazlas, Ph.D.
617.607.9717; PKazlas@QDVision.com

Primary Performing Organization:
QD Vision, Inc.
313 Pleasant Street
Watertown, MA 02472

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Executive Summary

With DOE Award No. DE-EE00000628, QD Vision developed and demonstrated a cost-competitive solution for increasing the light extraction efficiency of OLEDs with efficient and stable color rendering index (CRI) for solid state lighting (SSL). Solution processable quantum dot (QD) films were integrated into OLED ITO-glass substrates to generate tunable white emission from blue emitting OLED) devices as well as outcouple light from the ITO film. This QD light-enhancement substrate (QD-LED) technology demonstrated a 60% increase in OLED forward light out-coupling, a value which increases to 76% when considering total increase in multi-directional light output. The objective for the first year was an 80% increase in light output.

Comparison of Accomplishments Against Objectives

The summary below shows our progress against project objectives:

Specification	Objective	Progress
QD-LES Performance		
Enhancement to Extraction Efficiency	80%	60% (front face)* 76% (all light)
QD Film Down Conversion Efficiency	60%	65-75%
CRI	85	80-91
CCT [K]	3000-5500	2350-7600

*Measured without microlens array and without accounting for backside light

Project Activities Summary

In this final report, we summarize our progress against DOE Award No. DE-EE0000628, Quantum Dot Light Enhancement Substrate (QD-LES) for organic light emitting devices (OLED) Solid-State Lighting. The following sections review the program goals, activities, technical approaches, and progress against milestones.

Solution processable QD films were integrated into the QD light-enhancement substrate substrates to both generate tunable white emission from blue emitting OLED devices as well as outcouple light from the ITO film. The substrate also includes means to outcouple the generated white light from the glass layer. This QD light-enhancement substrate (QD-LES) technology simultaneously increased OLED light out-coupling, improved external quantum efficiencies, while providing >80 CRI white light that is readily tunable and inherently stable for any diffuse lighting application.

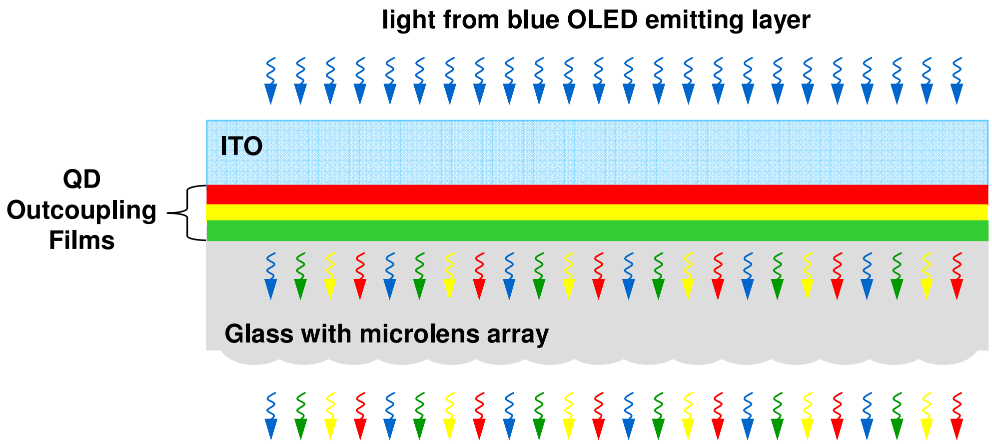

Figure 1: : Initial concept of quantum dot light enhancement substrate (QD-LES) incorporating multi-color QD films in a high refractive index material between an ITO film and glass.

Expected Benefits for OLED Solid-State Lighting

The expected benefits of our current approach are summarized in Figure 2 next to DOE's current and target efficiency values for OLED-based solid-state lighting. The efficacy of a light source is given by the product of the electrical-to-optical conversion efficiency ($\eta_{e\text{-}o} = P_o/P_e$) and the luminous efficacy of radiation (LER),

$$\text{Efficacy [lm/W]} = \text{LER [lm/W}_{(optical)}] \ \eta_{e\text{-}o} [\%],$$

where P_o and P_e are the optical output power of the light source and the electrical input power (voltage times current). $\eta_{e\text{-}o}$ is directly proportional to the internal quantum efficiency (IQE), extraction efficiency, $\eta_{e,}$ and inversely proportional to the driving voltage. In light of the equation above, our current DOE program directly addresses improving OLED lamp efficacy and reliability in the following ways:

- The IQE of the blue OLED can be optimized for blue light generation.
- $\eta_{e\text{-}o}$ >40% is possible by using a combination of internal and external outcoupling layers.
- Higher LER values > 350 lm/W are possible by leveraging the narrow band emission of green-yellow-red QDs in combination with bluer, broader OLED emission; current OLED LER values are limited to 350 lm/W due the inherently broad emission of OLEDs resulting in lost photons outside the visible spectra.
- High CRI due to customizable narrow QD emission profiles.
- Higher operational lifetimes are possible due to the single emitter architecture, reducing the color drift failure mode common in mixed emitter and tandem OLED devices
- Improved color stability as a function of drive current by using a single blue emitting OLED

- Simpler construction due the single emitter OLED architecture and solution-processable QD-LES.

If we assume an LER of 350 lm/W, an average blue PhOLED wavelength of 490 nm, an operating voltage of 3.3 V, an IQE of 100% and an extraction efficiency of 40%, the QD-LES-based PhOLED would have an efficacy ranging 100-110 lm/W (electrical-optical power conversion of approximately 31%) in line with DOE goals for OLED lighting for 2010-2011 timeframe. With further improvements in LER and extraction efficiency OLED lamp efficacies can exceed 150 lm/W using our approach. In addition, lifetime improvements can be expected, since the stable downconverters will essentially eliminate color shift over time, causing lifetime to be limited solely by the blue OLED, not the color shift experienced by all mixed-emitter OLED systems.

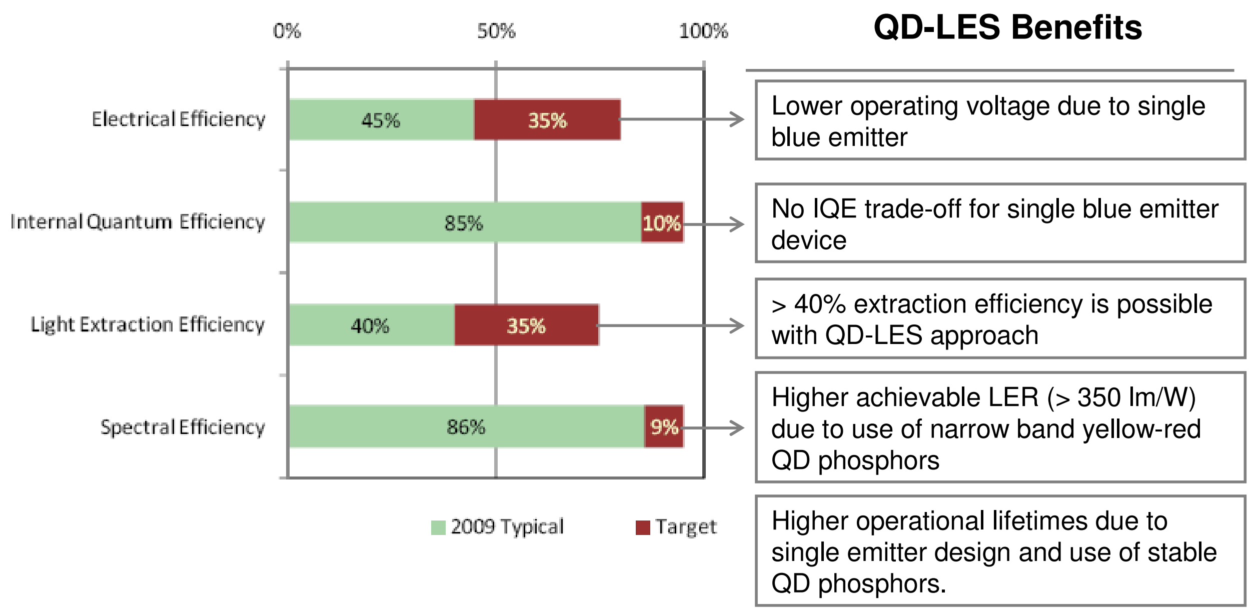

Figure 2: Current OLED panel losses and DOE targets[1] mapped to QD-LES benefits.

Project Objectives

The objective of the proposed effort was to develop and demonstrate a next generation OLED-based solid state lighting solution with both excellent CRI>90 and >80% improvement in external quantum efficiency using QD-based light enhancement substrates. Specific technical objectives for the proposed program included:

(a) Develop high refractive index QD-LES materials and solution deposition processes.
(b) Demonstrate an 80% improvement in extraction efficiency of OLED devices incorporating QD-LES by the end of Year 1.
(c) Optimize QD-LES for maximum extraction efficiency and color stability.

[1] Multi-Year Program Plan Solid-State Lighting Research and Development, US Department of Energy (March 2010).

The Year 1 critical path project milestones for the proposed effort are given in the table below.

Table 1: Critical path project milestones

Specification	Year 1
QD-LES Performance	
Enhancement to Extraction Efficiency *	**80%**
QD Film Down Conversion Efficiency	**60%**
CRI	**85%**
CCT [K]	**3000-5500**

* Including external outcoupling layer (e.g., microlens array)

Specific technical objectives for this program include:

(a) Model and simulate waveguide modes in OLED architecture to optimize optical design for QD films;

(b) Utilize our high quantum yield (QY) RoHS compliant QD emitter set for achieving predetermined luminescent color performance and >90 CRI;

(c) Develop QD-LES materials for solution deposition processes;

(d) Demonstrate an 80% improvement in extraction efficiency of a OLED device incorporating a QD-LES; and

(e) Optimize QD-LES for maximum extraction efficiency and color stability.

Technical Approach and Program Schedule

In Year 1, we studied and optimized at the component level; development emphasis focused on the following:

Tailored organic and QD emission spectrum to >85 CRI white light. QD layers were integrated into the substrate to down-convert blue to fill out the full white spectrum of light.

Improved outcoupling efficiency by 80%. QD light-enhancement substrate was used to break the waveguiding modes present and pull extra photons out of a control OLED device.

Table 2: Technical approach for key technology development areas

Technical Area	Year 1 Model and Demonstrate
QD Films	Investigate and down-select high refractive index host Formulate host-scatterer-QD system Devise a process for depositing multiple QD films
QD-LES Device Stack Optical Design, Modeling and Testing	Identify OLED baseline device stack Model and simulate waveguide modes for OLED-QD-LES stack to ensure maximum light extraction and high CRI Demonstrate proof-of-principle device and identify performance gaps

QD-LES Development Progress

In order to improve the extraction efficiency of the QD-LES device, development proceeded on several fronts. This section will deal with improvements made to the QD-LES with and without color conversion over the last year.

QD-LES Metrology

For this program, we carefully developed and applied various measurement techniques to assess our QD-LES performance as presented at the peer review. The following methods were applied:

- Measurement of EQE (air mode) using air-coupled photodetector[2]
- Measurement of EQE (air + substrate mode) using gel-coupled photodetector[4]
- Down conversion efficiency (solid-state PL quantum efficiency) using an integrating sphere[3]
- Angular irradiance measurement (air mode)
- Angular irradiance measurement using half-ball lens (air+ glass mode)[6]
- Total front face photon measurement using an integrating sphere.

[2] Forrest, S.R., Bradley, D.C., Thompson, M.E., Advanced Materials, 2003, 15, No. 13, pp 1043-1048.
[3] J.C. De Mello, H. F. Wittman, R. H. Friend ,Advanced Materials 9(3):230 (1997).
[6] Madigan, C.F., Lu, M.H., Sturm, J.S., Applied Physics Letters, 2000, 76, pp1650-1652

QD-LES Down Conversion

We successfully produced white light from a blue emitter through use of the QD-LES, achieving a CRI of 91 at a 4800K color point, meeting our objective.

The angular dependence of the light was also measured and the resulting color shift is seen Figure 3, below. This figure shows that the color of the QD-LES device is largely independent of viewing angle.

CIE x and y Values vs. Angle for OLED on QD-LES structure

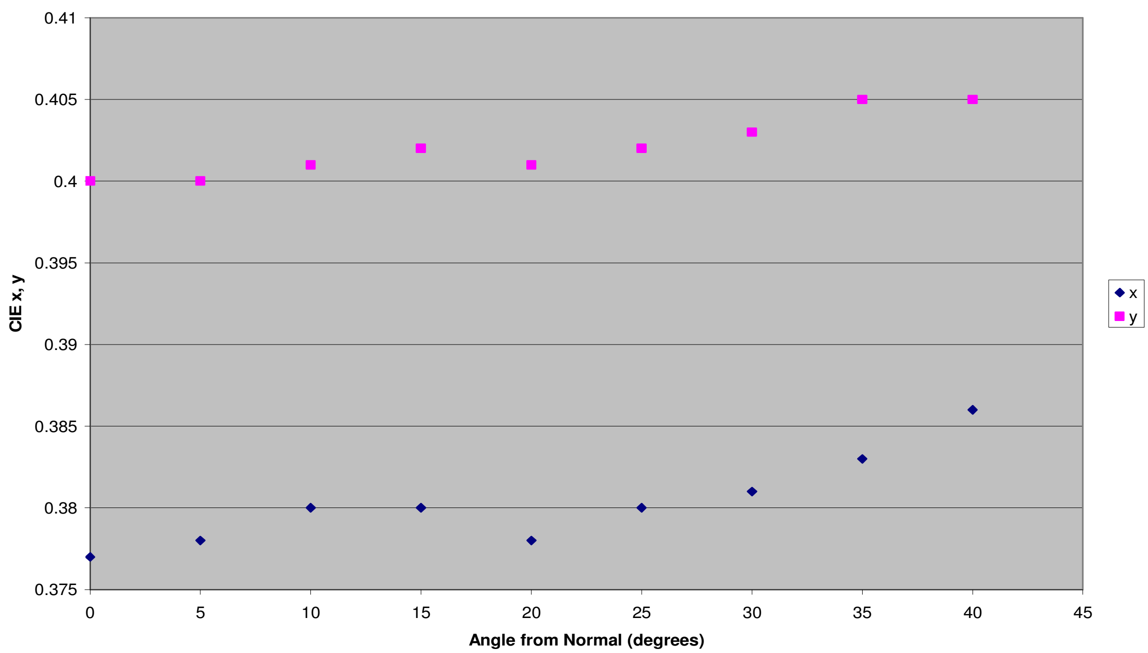

Figure 3: Angular Dependence of Color for QD-LES Device

QD-LES improved outcoupling without color conversion

The QD-LES structure has been developed to extract light in the high index modes of the OLED before the light within those modes is absorbed in the ITO and organic films. The refractive index of this film allows the high index modes to couple into the film, where they can then be extracted into external and substrate modes.

The external quantum efficiencies (EQEs) of QD-LES structures without color conversion were measured and compared to that of a normal OLED devices. The EQE was also compared to the quantum efficiency of the combined external and substrate mode light within the structure. The combined external and substrate mode light was measured by optically coupling the photo-diode to the substrate using an index matching gel. The resulting measurements showed the QD-LES structure without color conversion had a 40% improvement in EQE over the standard device. The efficiency of the substrate and external mode of the QD-LES structure without color conversion device was measured to have a 130% more light than the external mode of a standard device. This can be compared to the 95% improvement observed in a normal OLED device.

Care was taken to make sure the IV characteristics were very similar when measuring the QD-LES devices to rule out the effect of shorts or device leakage.

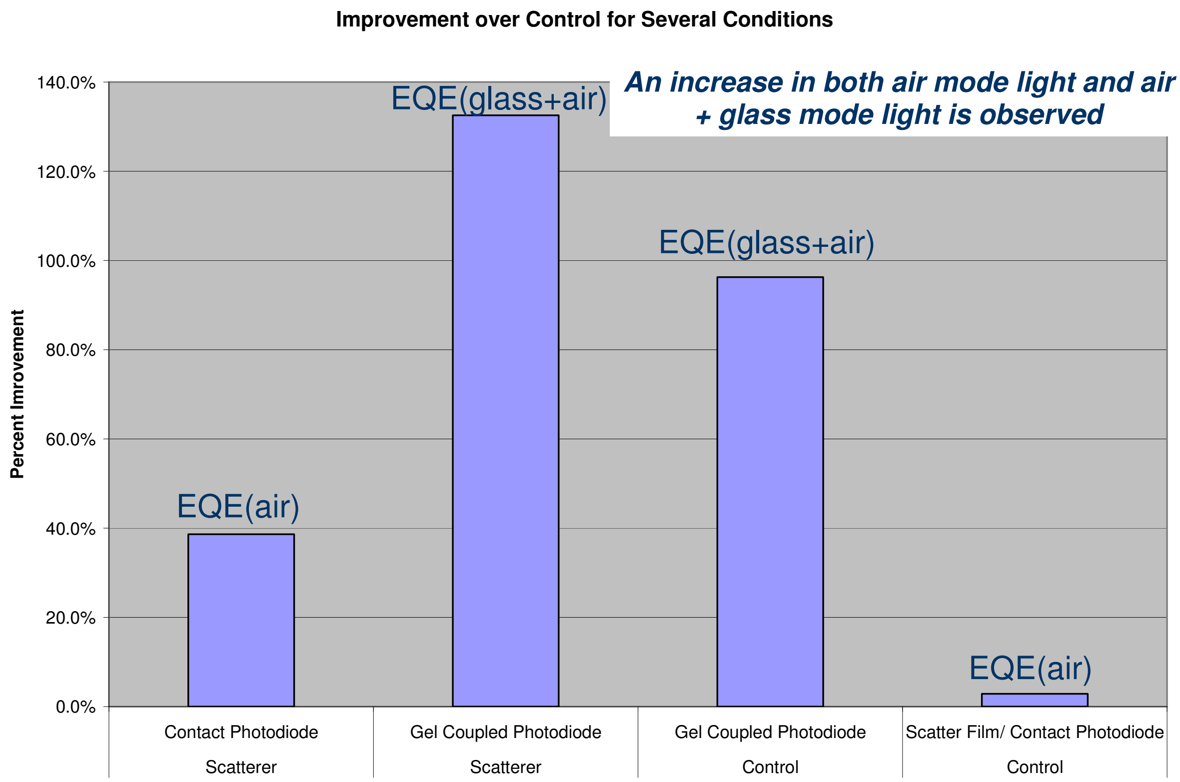

Figure 4: EQE comparison for QD-LES and control substrates.

Multiple internal outcoupling films were measured. Calibrated front face luminance measurements were also taken. The results are seen in the below figure and show a 60% increase in luminance for the case of the QD-LES type A (without color conversion).

Table 3 QD-LES (without color conversion) Performance

Sample	Luminance	Normalized to control
Control	284	100%
Type A	457	161%
Control	313	100%
Type B	345	110%

Measured at 300 μA

The relative irradiance as a function of angle was measured to look at the change in the angular dependence of light between the QD-LES (without color conversion) and normal substrates. This measurement was also performed using a half ball lens index matched to the substrate to enable the comparison the total amount of light in the substrate mode in each case.

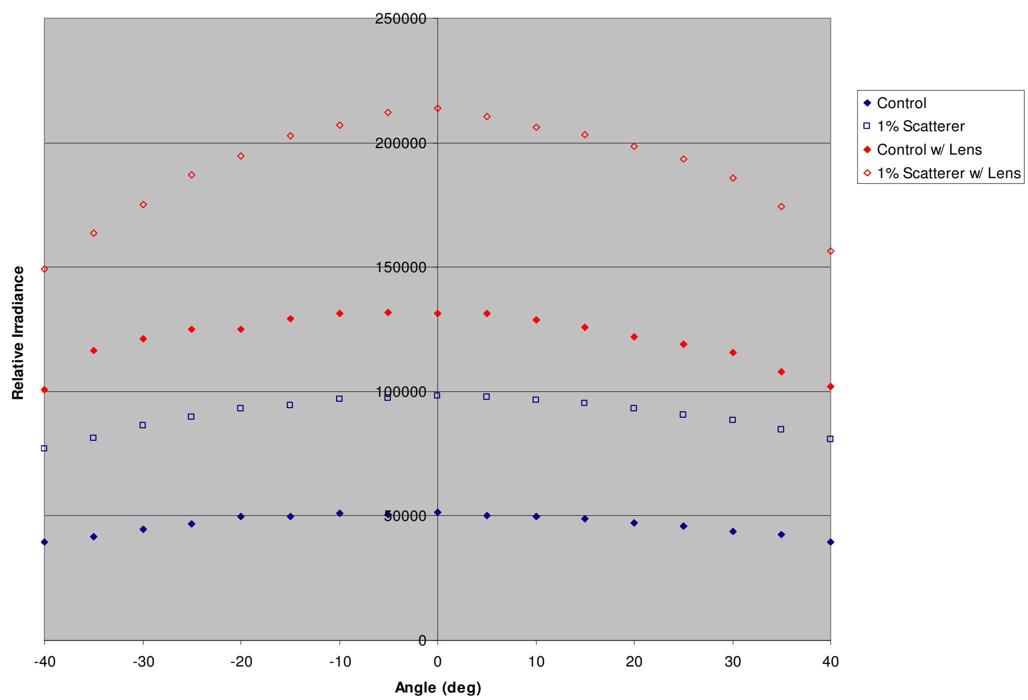

Figure 5: Irradiance as a function of angle for control and QD-LES(without color conversion) **with and without half-ball lens**

The measurement was performed between -40 and 40 degree from normal. The above graph shows the relative magnitudes of light as a function of angle for light in external modes as well as light in both external and substrate modes. This clearly shows that the there is more light in the substrate mode of the device on a QD-LES structure (without color conversion), indicating that light is indeed being moved from the high-index modes to the substrate mode with this design.

Microlens Array (MLA)

The extraction efficiency was measured using various MLAs, as summarized in the chart below.

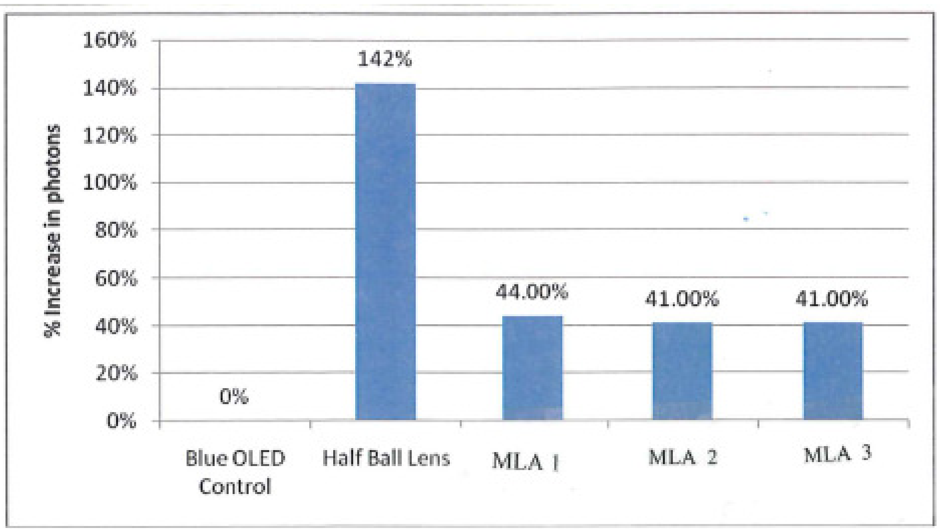

Figure 6: Comparison of light extraction efficiency for different Microlens Arrays over bare OLED control.

The figure above shows a comparison of the light extraction efficiency obtained using different lens types. The Half Ball lens represents the ideal case to show the maximum achievable gain using a lens as an out-coupling layer. We saw a 44% increase in the light extraction efficiency using MLA number 1 and a 41% increase in extraction efficiency using MLA number 2.

QD-LES with Color Conversion Demonstrator Performance

In order to understand the overall performance of the QD-LES system color conversion and MLAs were fabricated and then analyzed. This analysis allowed the source and magnitude of both gains and losses to be understood. All measurements are based on calibrated spectrometer measurements of a device positioned at the entry port of an integrating sphere.

In order to understand the total light output of the QD-LES structure, the integrated photon emission is normalized to that of a control OLED device. Also, the light emitted in the forward and backward direction is compared to the total light output for each case.

The photon emission from a QD-LES system without color conversion shows a 54% improvement in total output due to enhanced outcoupling of modes that were previously lost to absorption in the high-index layers. When a microlens array is applied to the QD-LES structure, the total output rises to 76% more than the control OLED, close to our goal of 80% outcoupling enhancement. A QD-LES structure with color conversion shows a total output of about 17% more than the control OLED, which increases to 25% more than the control OLED with the addition of the microlens array. The output of the QD-LES structure with color conversion and with microlens array is 70% of the output of the QD-LES structure with a microlens array but no Color conversion. This 70% loss can be attributed to the efficiency of the QDs, underlining the importance (and potential) of improving QD efficiency. All in all, these measurements show that using an OLED on QD-LES, we can convert the light from a blue OLED into high CRI white light while maintaining 125% of the photons generated by a control OLED device, even in this un-optimized case.

Here is a summary of the key learnings from this work:

- Our current outcoupling enhancement for QD-LES without color conversion and microlens array is 60% over the control in the forward direction. Accounting for all light raises this to 76%.
- Our current testing method introduces extraneous interfaces into the stack, artificially lowering our system-level results.

High Index Mode Outcoupling Analysis

In order to more fully understand the path to improving the QD-LES outcoupling efficiency, and analysis was undertaken to understand the effect of refractive index on outcoupling from the QD-LES.

To determine the maximum outcoupling improvement possible, if we assume that we have perfect outcoupling from the QD-LES high index hosts, a simple ray optics model can be used. This same model can be used to determine the improvements that might be expected if we increased the QD-LES hosts from 1.6 to a higher index of refraction.

The critical angle for internal reflection as a function of index is simply,

$$\theta_c = \sin^{-1}\left(N_0 / N_{organic} \right),$$

where θ_c is the critical angle, N_0 is the index to which light is extracted, and $N_{organic}$ is the index of the organic material in which light is emitted. We can assume all emitted light at angles greater than this cannot couple into a given material. If one assumes the cathode is an ideal reflector and the light emission is isotropic, the percentage of light (η) that can reach a particular index material without being internally reflected can be written as.

$$\eta = \int_0^{\theta_c} \sin\theta d\theta = 1 - \cos(\theta_c).$$

If we take the index of refraction of the organic material to be 1.75 we can plot the percentage of emitted light that can reach a certain index material as shown in the figure below.

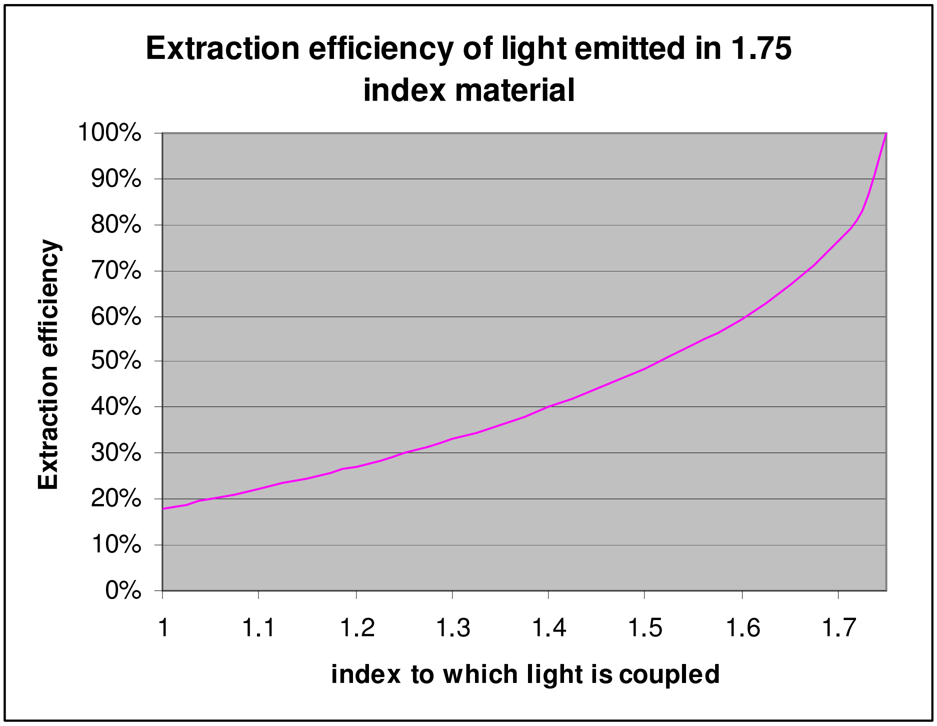

Figure 7: Light extraction efficiency from a 1.75 refractive index material as a function of refractive index of the neighboring medium

This analysis suggests that almost 50% of the emitted light can reach the substrate material which has an index of 1.5. The amount of light that can reach the QD-LES host, which has an index of 1.6, is about 60% of total emitted light. This suggest a 20% increase in light can be achieved in the combined glass and air modes if light is optimally outcoupled from the QD-LES host to these modes. Our EQE measurements show such an increase is achievable with this system.

If we further increase the internal outcoupling film index to 1.7, 76% of emitted light can reach this material. This suggests a 52% increase in light can be achieved over a control device in the combined glass and air modes if light is optimally outcoupled from the QD-LES hosts to these modes. It also suggests that such a change would improve our current outcoupling system by 27%. Furthermore improving the index of fraction in the QD-LES to 1.725 would enable 83% of emitted light to reach the external and substrate modes, improving our current outcoupling system by 38% without changes to the overall system. A perfectly index matched QD-LES could improve outcoupling by 66%.

Table 4. Extraction Efficiency Calculations

Index of Coupling Material	Extraction Efficiency
1.75	100%
1.725	83%
1.7	76%
1.675	71%
1.65	67%
1.625	63%
1.6	59%

Invention Disclosures Developed Under Award

As per DOE Form 2050.11, "Patent Certification", QD Vision has reported the following Confidential Memoranda of Inventions for Disclosures docket numbered QDV 157, QDV 182, QDV 183, and QDV 184.