Manufacturing Process for OLED Integrated Substrate
Final Report

March 31, 2017

Period of Performance: August 1, 2013 to December 30, 2016
Work Performed Under Agreement:
DE-EE0006261

Work Partner: Universal Display Corporation

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Notes: *Glass Division of PPG Industries, the original project recipient, was acquired by Vitro Flat Glass LLC on October 1st, 2016. In the final stages, completion of the project was conducted in Vitro's facilities. In addition to PPG, Vitro and PPG/Vitro names were used for clarification.*

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A. Project Summary:

The main objective of this project was to develop low cost integrated substrates for rigid OLED solid state lighting application in the manufacturing scale. The integrated substrates could include the combinations of soda lime glass substrate with light extraction layers and anode layer (i.e., Transparent Conductive Oxide, TCO). Over the 3^+ year's course of project, the scope of work was revised to focus on the development of glass substrates with internal light extraction (IEL) layer. A manufacturing scale float glass on-line particle embedding process capable of producing IEL glass substrates with less than 1.7mm thickness and larger than 500mm x 400mm size was demonstrated. The Gen 2 display glass substrates (470mm x370mm) were used in the OLED manufacturing process for fabrication OLED lighting panels in single pixel devices as large as 120.5mm x 120.5mm. The measured light extraction efficiency (calculated as external quantum efficiency, EQE) for on-line produced IEL samples (>50%) met initial goal of the project in 2013.

B. Project Objective:

The primary objective of this project was to demonstrate manufacturing processes for technologies that will enable commercialization of a large-area and low-cost “integrated substrate” product for rigid OLED SSL lighting. The integrated substrate product could consist of a low cost, float glass substrate combined with a transparent conductive anode film layer, and/or light out-coupling (internal and external extraction layers) structures. In combination, these design elements would enable an integrated substrate meeting or exceeding 2015 performance targets for cost ($60/m^2), extraction efficiency (50%) and sheet resistance (<10 ohm/sq).

In a previous DOE-funded project (DE-EE0003209), PPG successfully demonstrated viable low-cost alternatives to Indium Tin Oxide (ITO) anodes and light extraction layers on float glass substrate for large-area OLED lighting panels. Extraction efficiencies of nearly 40% were obtained and further optimization of the design elements is expected to lead to further increases. In the previous work, the anode and light extraction layer technologies were developed on laboratory-scale equipment and demonstrated using large area OLED devices fabricated by Universal Display Corporation (UDC).

As a next step, PPG built on these anode and extraction layer capabilities and utilized its existing low-cost, coated glass manufacturing footprint. PPG partnered with Universal Display Corporation and with additional support from OLEDWorks to fabricate and characterize devices on the integrated substrates for manufacturing process optimization studies.

At the completion of the program the following goals were achieved:

- Manufacturing demonstration of a large-area embedded particle Internal Extraction Layer (IEL)
- Validation of textured glass External Extraction Layer (EEL) with IEL
- Quality control measures identified using on-line and off-line analysis tools to achieve the desired variation control and product uniformity
- Production and supply of integrated substrate to OLED lighting panel manufacturers

C. Technical Approach:

PPG scaled-up selective technologies to its manufacturing lines and developed a plan to commercialize a large-area and low-cost “integrated substrate” product for rigid OLED SSL lighting. The integrated substrate products consisted of a float glass substrate in combination with a transparent conductive anode film layer and light out-coupling (internal with or without external extraction layers) structures. Quality control measures of manufacturing processes were identified using on-line and off-line analysis tools to achieve the desired variation control and product uniformity of the large-area integrated substrate which is critical for device consistency and materials waste minimization thus reducing the overall cost of production.

In the first budget period, PPG worked on developing high throughput manufacturing processes for the embedded particle internal extraction layer (IEL) and anode application on float glass. The substrate thickness was less than 1.8mm to be consistent with market requests. Key milestones included achieving desired embedding characteristics (e.g., particle density, depth, and surface roughness), and acceptable anode properties (e.g., sheet resistance, transmission, and surface roughness). During the first budget period, improvements in large-area device yields and performance were investigated through the use of modified anodes based on chemical vapor deposition (CVD) as well as anodes applied via a sputtering process (e.g., magnetron sputtered vacuum deposition (MSVD)). Additionally, external extraction layers were evaluated in combination with the IEL and anode.

Based upon the results obtained in Budget Period 1 (BP 1), PPG improved the IEL, anode, and made need/no-need decision on EEL fabrication processes in the second budget period (BP 2). Product commercialization was targeted in the second stage of the project. Throughout both phases of the project, OLED devices were fabricated and evaluated by our partner (i.e., UDC) and a number were fabricated in an OLED manufacturing production line at OLEDWorks, Inc.

D. Planned Milestones

Original planned milestones are listed in Table 1. Highlighted milestones including FTO anode development and evaluation, and HIL evaluation were switched to sputtered anode development and evaluation with approval from DOE. Revised program management plans were submitted twice in December 2014 and July 2016 for milestones changes and requesting no cost project extensions. The final performance ending period was extended from July 2015 to December 2016 due to organization changes.

The commercialization plan was presented to DOE in the continuation meeting at the end of Budget period I (instead of in the Budget period II).

Table 1 Planned Milestones and Success Criteria

Budget Period	Milestones	Success criteria
I	Program Management Plan	Develop Program Management Plan and update annually or as otherwise needed to reflect significant changes. Initial PMP submitted 11/2013.

I	Short slot coater for IEL deposition	Online particle deposition demonstrated using short slot coater- (200-400 nm particles at a depth of ~2 microns, assuming glass being produced at 2.5mm). Completed 2/2014.
I	Long slot coater for IEL deposition	Online particle deposition demonstrated using long slot coater (200-400 nm particles at a depth of ~2 microns, assuming glass being produced at 2.5mm). Completed 6/2014.
I	IEL Evaluation in OLED devices	27 (3" x 3") green OLED devices assembled and evaluated for IEL extraction efficiency. Completed 7/2014.
I	Manufacturing process for FTO anode on 2.5 mm glass substrate	CVD coating process established with 10 Ohms/Sq. sheet resistance and a surface roughness of < 10 nm for a high throughput manufacturing process on 144" wide, 2.5 mm thick glass substrate with +/- 5% cross ribbon uniformity. Completed 6/2014.
I	FTO Evaluation in OLED devices	18 (2mm x 2mm) green OLED devices assembled and evaluated for anode performance. Completed 7/2014.
I	HIL Evaluation in OLED devices	18 (3" x 3") green, 18 (6" x 6") green, and 8 (6" x 6") white OLED devices assembled and the combination of IEL, anode and HIL will be assessed for device performance. Completed 7/2014.
I	Budget Period 1 OLED devices on integrated substrate	27 (3" x 3") green and 10 (6" x 6") white OLED devices assembled and the combination of IEL, anode, HIL and EEL will be evaluated. Completed 7/2014.
II	Commercialization Plan	Develop plan to commercialize integrated substrate based on market needs and requirements. Date 11/2014.
II	IEL manufacturing process on thin glass	Online particle deposition demonstrated using long slot coater (200-400 nm particles at a depth of ~2 microns, assuming glass being produced at 1.8mm). Date TBD.
II	Manufacturing process for FTO anode on thin glass substrate	Manufacturing process transferred to thin glass substrate (assuming glass being produced at 1.8 mm) and the CVD coating achieves 10 Ohms/Sq. sheet resistance on 144" wide with +/- 5% cross ribbon uniformity ribbon. Date TBD.
II	Final OLED Devices	Six prototype (6" x 6") white OLED devices on float glass-based integrated substrate with panel efficacy of 86-125 lm/W, luminous emittance of 6000-10,000 lm/m^2, and panel life (LT 70) of 25,000-50,000 hours will be characterized and delivered to the DOE. Date TBD.

E. Budget Period I&II Accomplishments: Progress vs. Milestones

Table 2 Milestones and Success Criteria for Phase I (a)

Budget Period	Milestones	Success Criteria	Milestone date or Start/End	Milestone Status
I	Program management plan	Develop program management plan and update annually or as otherwise needed to reflect significant changes.		Initial PMP submitted 11/2013. Revised PMP submitted 12/2014. Revised SOPO and PMP submitted 7/2016.
I	FTO anode (Rsheet, surface roughness)	CVD coating process established with 10 Ohms/Sq. sheet resistance for a high throughput manufacturing process on 144" wide, 2.5 mm thick glass substrate with +/- 5% cross ribbon uniformity.	8/13-3/15	On-line trial tested in 4/14 on 2.5mm glass. Equipment problem occurred. Oxide smoothening layer tested in 4/14 on 4.0mm glass. **(FTO work stopped and changed to sputtered anode)**
I	FTO Evaluation in OLED devices	20 (3" x 3") white OLED devices assembled and evaluated for anode performance.	10/13-3/15	**(FTO work stopped and changed to sputtered anode)**
I	HIL Evaluation in OLED devices	20 (3" x 3") white OLED devices assembled and the combination of IEL, anode and HIL will be assessed for device performance.	7/14-11/14	**(HIL work stopped and changed to sputtered anode)**
I	Sputtered anode	MSVD process established with 10 Ohms/Sq. sheet resistance and 85% transmittance for a high throughput manufacturing process	12/14-7/15	Process developed with metal and ITO layers. 10 Ohms/sq. and 85% transmittance achieved. High throughput process determined.
I	Sputtered anode evaluation	18 (3"x3") green OLED devices assembled and evaluated for anode performance.	7/15	34 (6"x6") submitted for green OLED evaluation with the combination of anode and IEL. Evaluation continued to project BP2.

Table 3 Milestones and Success Criteria for Phase I (b)

Budget Period	Milestones	Success Criteria	Milestone date or Start/End	Milestone Status
I	Medium slot coater for IEL deposition	Online particle deposition demonstrated using medium slot coater- (200-400 nm particles at a depth of ~2 microns, assuming glass being produced at 2.5mm).	2/14	Coater system completed; Process demonstrated for complete particle embedment on glass thickness 6mm to 2.3mm.
I	Long slot coater for IEL deposition	Online particle deposition demonstrated using long slot coater (200-400 nm particles at a depth of ~2 microns, assuming glass being produced at 2.5 mm)	7/15	Coater converted to long slot and tested on 2.1mm glass with targeted particle properties and surface smooth quality.
I	Budget Period 1 OLED devices	20 (3" x 3") OLED devices assembled and evaluated for IEL, EEL extraction efficiency, and anode performance	7/15	13 samples with duplicate evaluated at UDC; Additional 7 samples with duplicate evaluated at UDC. 14 samples with duplicated evaluated at OLEDWorks.
I	Commercialization plan	Developed plan to commercialize integrated substrate based on market needs and requirements	7/15	Reported in the continuation meeting July, 2015.

Table 4 Milestones and Success Criteria for Phase II

Budget Period	Milestones	Success Criteria	Milestone date or Start/End	Milestone Status
II	Large area manufacturing process for sputtered anode on >2.5mm glass substrate in OLED devices	Large area sputtered anode coating process established with 10Ω/□ sheet resistance, transmittance>85% for a high throughput manufacturing process.	4/16	The long slot coater tested on 1.7mm glass with targeted particle properties and surface smooth quality.
II	Budget period 2 OLED devices on integrated substrate	10(3"x3") white OLED devices assembled and the combination of IEL, anode, and EEL will be evaluated.	4/16	More than 70 green and white OLED devices evaluated on IEL and anode with pixel size ranged from 2mm2 to 25 cm2. The size of glass substrates included 2.5"x2.5" and 6"x6". Light extraction performance and device life time evaluated.
II	Final OLED Devices	Six prototype devices on float glass based integrated substrate with panel efficacy of 86-125 lm/W, luminous emittance of 6000-10,000 lm/m² and panel life (LT 70) of 25,000-50,000 hours will be characterized and delivered to the DOE.	12/16	Prequalification of PPG IEL glass substrate in OLED manufacturing process completed. Large area devices produced.

F. Deliverables:

The status of deliverables is highlighted in the table below.

Table 5 Deliverables Associated with Various Project Tasks

Task	Deliverable	Status
1.0 Program Management Plan	Project Management Plan Document	Completed
2.0 On-line IEL manufacturing	Report of results	Completed, reported
3.0 Fluorine Doped Tin Oxide Anode Manufacturing Development	Report of results	Completed, reported
4.0 Sputtered Anode Manufacturing Development	Report of results	Completed, reported.
5.0 Determination of Efficiency of Textured Glass EEL	Report of results	Completed, reported.
6.0 OLED Panel Fabrication and Characterization	Report of results	Reported
7.0 Commercialization Plan	Report plan	Reported
8.0 Manufacturing Process Improvements	Report of results	Completed, reported.
9.0 Large-Area Sputtered Anode Process Development	Compare to Task 4.0 and report results	Completed, reported. Sputtered anode material developed and tested with OLED. Laser etching process evaluated.
10.0 Textured Glass EEL Process Development	To be decided if required	Completed. Textured glass EEL developed and evaluated with OLED. Third party scattering type EEL used in final devices.
11.0 Large-Area OLED Panel Manufacturing	Six prototype devices on float glass based integrated substrate	Completed. Gen 2 (370mm x 470mm) IEL glass substrates used in producing large area OLED panels (120.5mm x 120.5mm) in OLED manufacturing processes.

Note: Updated based on the revised PMP.

G. Technical Detail – Budget Period I:

<u>Task 1 –Program Management Plan (PMP)</u>

The first PMP was submitted in November, 2013, and a revision was submitted in December 2014. The proposed statement of project objectives (SOPO) incorporating our changes in scope from fabrication of the anode by CVD to MSVD was accepted by the DOE. Acceptance by the DOE of the proposed changes to the project including moving the date "go/no go" recommendation from April to July, 2015 was granted on March 24th, 2015.

BP2 continuation was approved on July 31st, 2015 with budget period from August 1st, 2015 to July 31st, 2016. The SOPO and PMP were revised to focus the development works on IEL development.

A five month no cost extension was approved on August 1st, 2016 for completion of OLED large panel fabrication with an end date of December 30th, 2016.

<u>Task 2- On-line IEL Manufacturing</u>

Equipment Design:

Embedding high index oxide particles in the glass substrate to redirect the light is the main concept of internal light extraction. It can be achieved by applying uniform distributed fine particles onto the surface of molten glass and allowing particles to be settled into the glass. Thus, a smooth surface of glass substrate with particles embedded in desired depth can be obtained. Shown in Figure 1 is an on-line IEL manufacturing process schematic. Since particle embedding tool is located in a molten glass chamber, to keep process equipment functional the tool systems need to be maintained at constant low temperature.

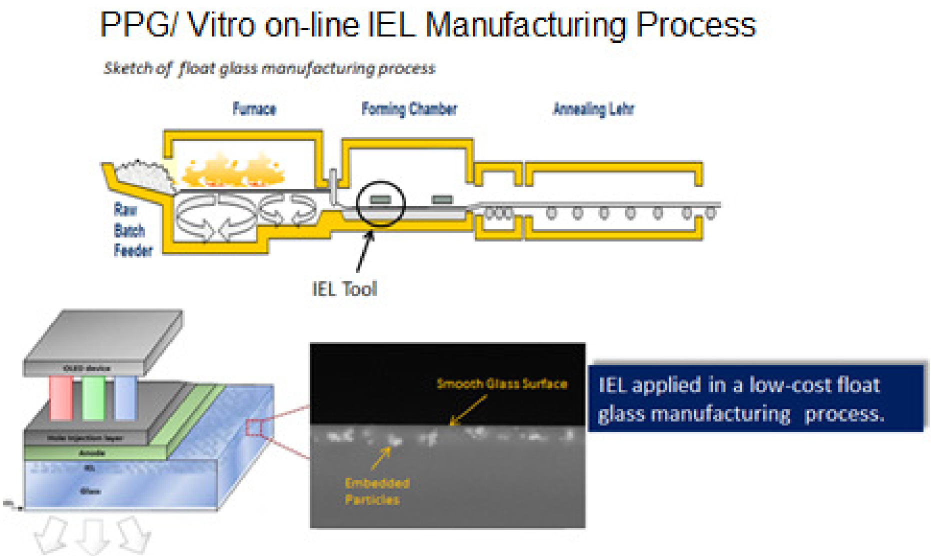


Figure 1 A schematic of on-line IEL manufacturing process at PPG/Vitro.

There were two particle embedding processes used in this study: (1) introduced uniform dispersed extra fine particles onto molten glass, (2) provided a precursor in vapor phase and in-situ generation of ultrafine particles for deposition onto molten glass. In the process#1 (i.e., powder spray process), a powder feeder system was used to control feedrate and powder dispersion. In the process#2, the particles were generated in-situ.

Demonstration of On-line IEL Manufacturing:

(1) Powder spray process

A high refractive index particulate oxide material was used in this study. Powder feedrate was investigated from 23 g/min to 145 g/min. The amount of particles incorporated in the glass, surface smoothness, and particle depth of penetration in the glass were characterized. Results (see Table 6) showed low particle incorporation, high surface roughness, and too deep of particle depth of penetration. Note that the targeted haze is 50%, surface roughness is less than 2nm, and depth of particle penetration is in 1µm range.

Table 6 Optical Property, Surface Roughness, and Depth of Particle Embedding for IEL Samples Produced Using Powder Spray Process

IEL sample	Feedrate (g/min)	Haze (%)	Transmittance (%)	R_q Roughness (nm)	R_a Roughness (nm)	Depth of Penetration (µm)
1	23	1	89	33	15	22
2	68	0.6	90	13	8	45
3	110	4.5	83	470	276	42
4	145	7.4	79	424	256	50

Haze and transmittance were measured using BYK Gardner Haze-gard plus; roughness was determined using Veeco Wyko NT3300 Optical Profilometer; depth of penetration was measured from cross-sectional Scanning Electron Microscope (SEM) images.

(2) In-situ particle generation and embedding process

High index particles were produced in-situ and embedded into the glass surface. The critical properties used as evaluation criteria were:

- Particles incorporation in glass and resulting light scattering as measured by the haze level
- Surface roughness
- Depth of particle penetration in glass
- Uniformity of particle distribution across glass substrate

In Budget Period I, significant improvement on incorporating particles in glass was made (see Figure 2). A couple order of magnitudes improvement was made in every about 3 month's periods. The resulted haze is higher than the targeted 50% haze level for the samples made in October/ November 2014.

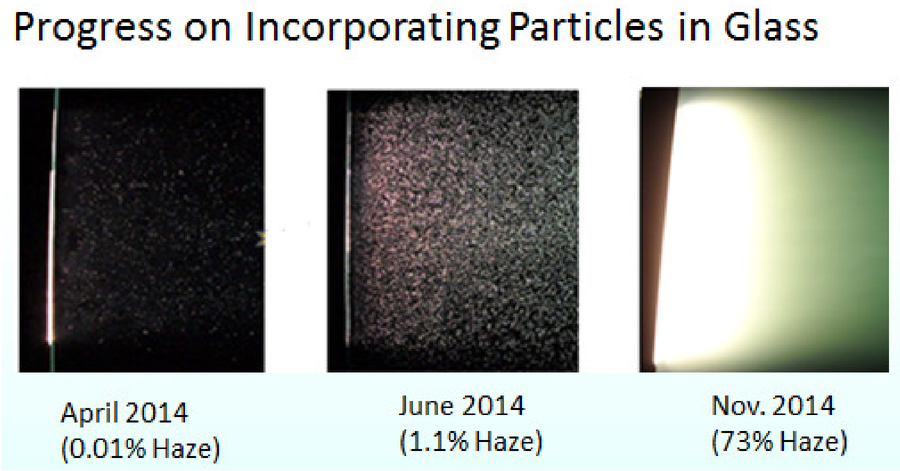


Figure 2 Progress on on-line IEL development toward target requirements. Photos of typical IEL samples illuminated with a 6” edge light from left side of the samples shown light scattering of particles. Higher particle number density resulted in higher scattered light intensity.

Completely embedded particle IEL structures having light scattering controllably and consistently at or above our target level (50%) were achieved in glass substrates as thin as 2.5mm with online IEL process (see Figure 3). Examination of IEL samples using SEM showed uniform distribution of embedding depth and particle size across the substrate. Shown in Figure 4 are a planar view SEM and a cross-sectional view SEM for a typical IEL sample. It can be seen that particles are fully embedded resulting in a smooth surface

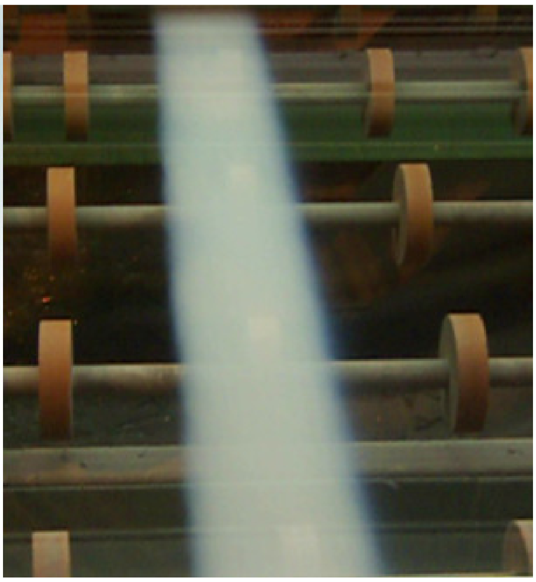


Figure 3 Glass substrate with IEL produced in PPG/Vitro on-line process.

morphology. Analysis using optical profilometry analysis showed <2nm surface roughness, and cross sectional SEM showed ~1µm depth of particle embedment.

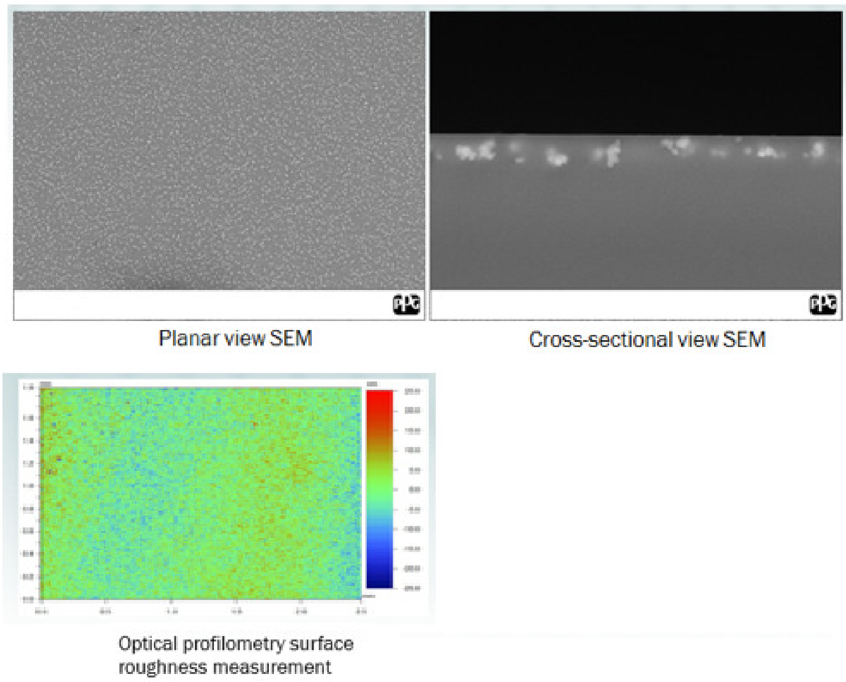


Figure 4 Characterization of the on-line produced particle embedded glass substrate using Scattering Electron Microscopy (SEM) and optical profilometry.

Three different locations in the glass forming chamber were investigated for the effect of glass viscosity and residence time on particle embedment. Comparing with downstream locations, glass temperature is higher (thus lower glass viscosity) at the upstream location. Since it is far away from the cut-off temperature for particle embedding, the residence time for embedding particles is longer. Table 7 showed the effect of glass viscosity and residence time on particle embedment. Operated embedding tool at upstream locations, particles were embedded completely and the surface was smooth. The particles had longer time to settled into glass resulted in deeper depth of penetration. Note that depth of penetration was measured for the particles with deepest distance under the surface. As shown in Figure 4, particles are distributed in the range from the surface to the depth of penetration. Also, the produced particles were crystalline and with high temperature phase (thus, higher index of refraction). Operated embedding tool at the downstream location (e.g., location C), particles were partially embedded and the surface was rough. The produced particles were also crystalline, but including a lower temperature phase.

Table 7 Effect of Glass Viscosity and Residence Time on Particle Embedment

Tool Tested Location#	Relative Bath Temperature	Relative Glass Viscosity	Relative Residence Time++	Complete Particle Embedment	R_q Roughness (nm)	Depth of Penetration (µm)
A	high	low	long	yes	2	1.2
B	medium	medium	medium	yes	1	0.8
C	low	high	short	no	70	0.2**

*#: Tool location A is at upstream of glass forming chamber, B and C are at the downstream of glass forming chamber. ++: Residence time is defined as the time allowing for the particles to be settled into the glass. Thus, a cut off temperature (or glass viscosity) was selected. Residence time depends on the distance to the cut off temperature location and line speed. Typical residence time is in minutes range. **: At this location, the particles were partially embedded. Some particles are on top of the glass surface.*

After the Mt. Zion, IL glass facility was divested in 2014, the IEL process was transferred to another manufacturing facility at Carlisle, PA. Complete particles embedment on 3mm and 2.1mm glass was achieved in the first two trials demonstrating the successful transfer of the process.

At the Carlisle facility, development of the particle embedment process on thinner glass (<1.8mm) at higher line speed and wider particle embedding band continued. Figure 5 showed on-line produced IEL glass using different width slots at the particle embedding tool demonstrating increased width of particle embedding band with that tool modification.

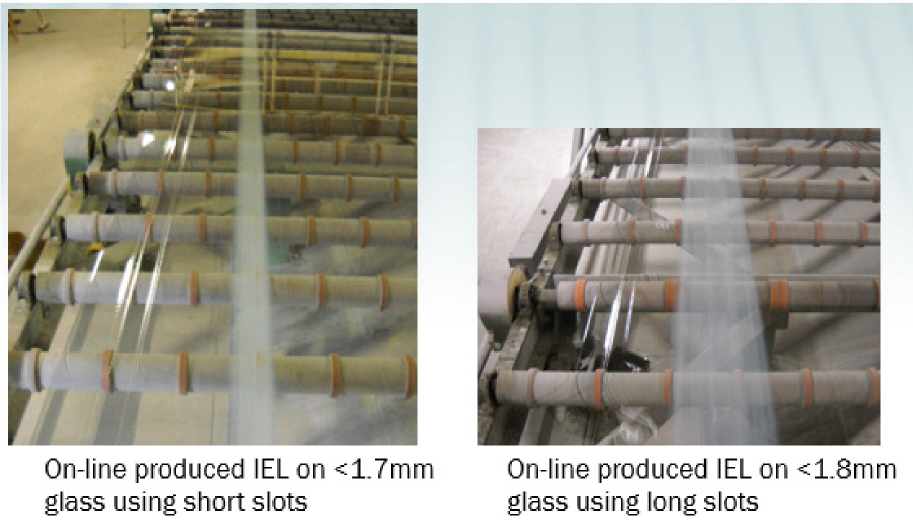


Figure 5 On-line produced IEL glass substrates.

The effect of line speed in embedding particles was studied in the on-line float glass manufacturing process. The line speed that more than 2 orders of magnitude higher than the process in the lab was used to produce 2.0mm thin glass. Since starting of this project in 2013, complete particle embedment in glass substrate from 6mm to 1.7mm was demonstrated. Figure 6 showed high line speeds were used in making thin glass substrates. The embedded particle concentrations were higher than desired as indicated by the measured haze >60%.

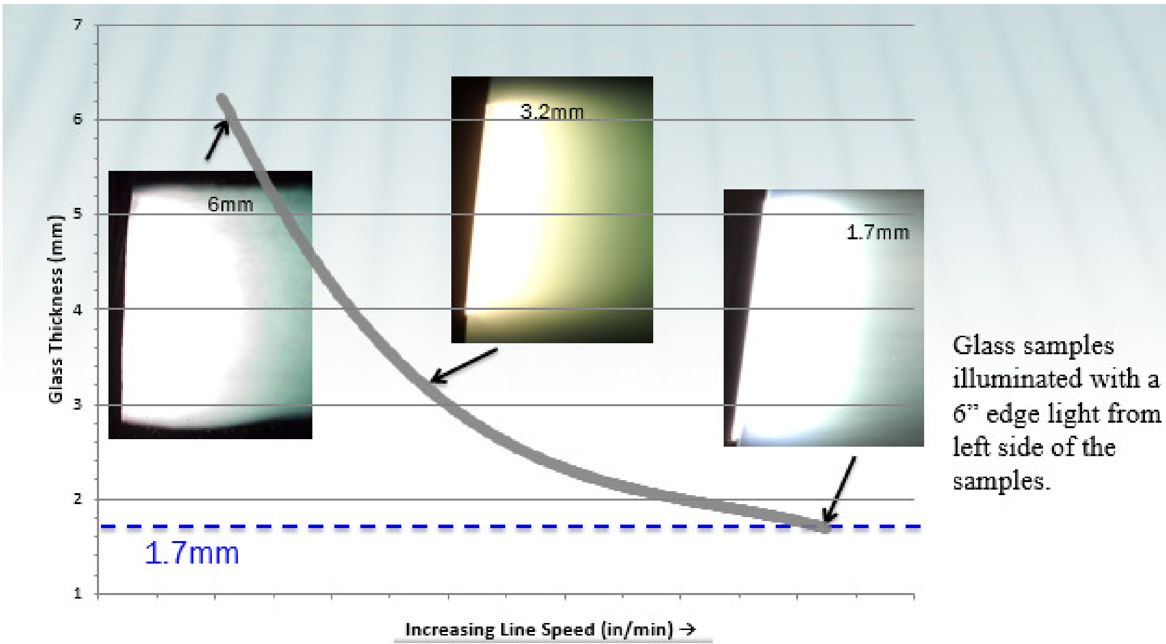


Figure 6 Thin glass was produced at more challenge high line speed. Complete particle embedment was achieved for glass thickness ranged from 6mm to 1.7mm. Note the uniform light scattering when illuminated with light from left side of glass edge.

Uniform distribution of particles in the glass substrate is one of the key requirements for commercialization. The non-uniformity could be cracking lines as shown in Figure 7 (left photo). They were produced due to stretching and compression of the surface during the glass forming process. We have optimized our particle embedding process to avoid generation of cracking lines on the glass substrates. The photo on right side of Figure 7 showed uniform distribution of particles in the glass sample.

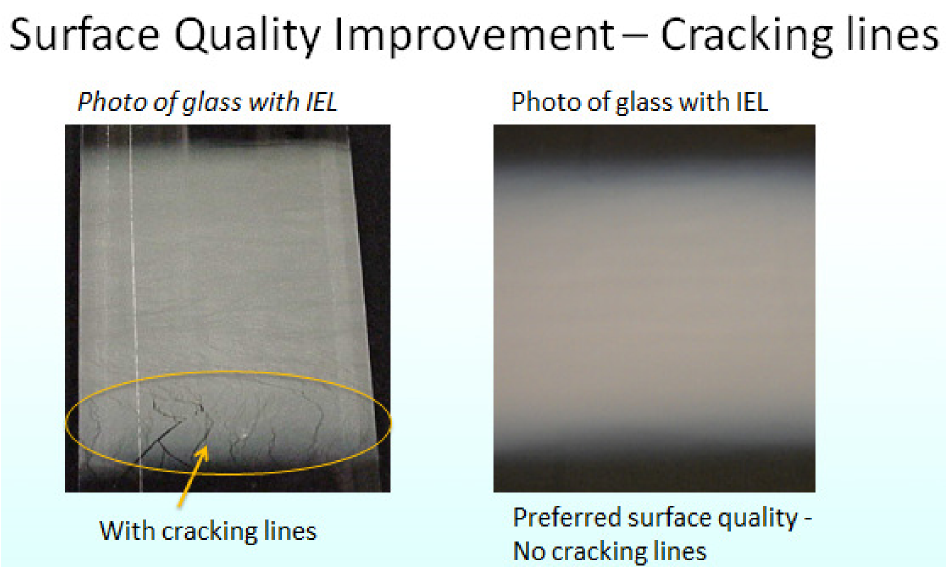


Figure 7 Surface quality was improved through process parameters optimization.

OLED Fabrication and IEL Evaluation:

The first ever OLED devices fabricated using an on-line produced IEL sample were fabricated at OLEDWorks. The OLED device was white emitting with a tandem structure and the substrates were 3.2mm thick soda-lime glass that were 2.5” x 2.5” in size. The measured light redirection (haze) was 40% for the IEL glass samples. The same thickness and dimensions of clear glass samples were used for as controls. Industry standard indium tin oxide was used as transparent conductive anode layer. Two different sizes of pixels (3 cm^2 and 0.1 cm^2) of devices were built on each glass substrates. Light extraction enhancement was compared EQE at 10 mA/cm^2 between the IEL sample and the control. The photo on right side of Figure 8(b) is an OLED device made using a 3.2mm substrate with 40% light scattering layer produced on-line. Comparing with the device without IEL, light extraction enhancement can be seen. For additional comparison, an OLED device made from previous DOE funded project with off-line produced IEL (see photo on right side of Figure 8(a)) is attached. The initial data showed 10% light extraction enhancement for this first OLED device produced with an IEL fabricated with an on-line process.

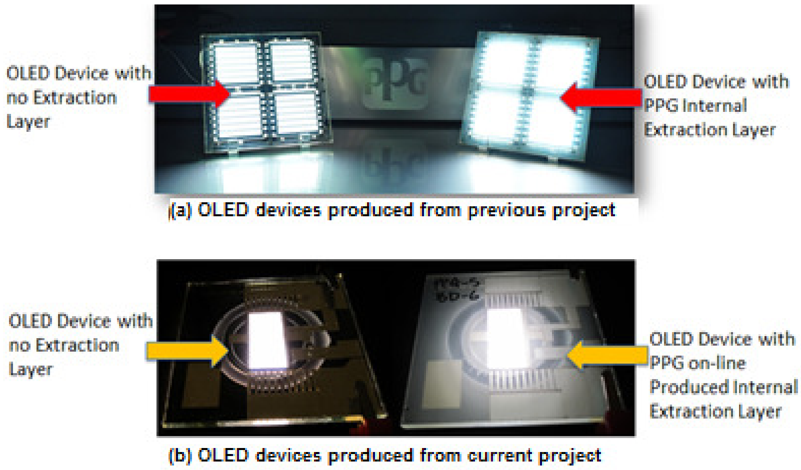


Figure 8 OLED light extraction evaluation of (top, a) off-line produced IEL substrate and (bottom, b) PPG on-line produced IEL substrate.

Task 3 - Fluorine Doped Tin Oxide (FTO) Anode Manufacturing Development

A Chemical Vapor Deposition (CVD) process was used for high throughput manufacturing process of FTO anode. Other than meeting sheet resistance less than 10 Ω/□ requirements, emphasis was placed on reducing the surface roughness and matching deposition rate to the speed of glass being produced. SEM image of a typical FTO anode material (see Figure 9) showed crystalline structure with rough surface.

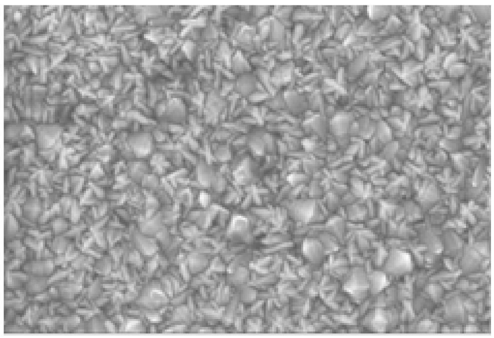


Figure 9 SEM planar view of a typical FTO anode material.

Testing of FTO anode deposition on-line with high line speed showed good sheet resistance (i.e., 10 Ω/□) with above-target surface roughness (i.e., 18nm R_q). To obtain smooth surface, a new approach was tested by deposition an oxide smoothening layer on the top of FTO. This is a similar idea of using Hole Injection Layer (HIL) to smoothen a rough surface on transparent conductive oxide (TCO) layer. The effect of smoothening layer thickness on surface roughness was investigated. Shown in Table 8 is a summary of the results. With the addition of smoothening layer, surface roughness was improved 40% to 50%. However,

R_{pv} values were still higher than the target 20nm value which would have high potential to cause short circuiting on OLED device.

Table 8 Effect of Adding Smoothening Layer on FTO Surface Roughness

Smoothening Layer Thickness (nm)	R_q Roughness (nm)	R_a Roughness (nm)	R_{pv} [#] Roughness (nm)	% Improvement on R_{pv} Roughness
0 (FTO only)	**18**	**14**	**162**	-
22	11	8	86	43
48	9	7	77	54
78	11	8	90	48

Roughness was determined using a Digital Instrument Dimension 3100 Atomic Force Microscope. #: R_{pv} is the maximum peak to valley value.

Since surface quality of the sputtered anode material is better than FTO anode material, the SOPO and PMP was revised with Task 4 to replace FTO anode and HIL coating tasks. This revised SOPO and PMP was approved by the DOE in July 2015.

Task 4 - Sputtered Anode Manufacturing Development

A Magnetron Sputtered Vacuum Deposition (MSVD) process was used for as alternate high throughput anode manufacturing process. A thin layer metal material is used for electrical conductivity. The metal layer is sufficient thin for high optical transmission while still providing low sheet resistance. In general, a sputtered anode material has many advantages over a CVD anode material including a surface that is much smoother than CVD produced anode material. Table 9 showed key property comparisons between sputtered anode and CVD anode material. The material cost for off-line sputtered anode is expected to be comparable to the total cost of on-line CVD anode plus off-line HIL. Furthermore, based on our experience the production yield is expected to be better for the sputtered anode process. Thus, using the alternate sputtered anode the cost of integrated substrate can meet DOE MYPP $60/m^2 performance target for 2015.

Table 9 Key Property Comparisons – Sputtered Anode vs. CVD Anode

Metric	Sputtered[#] Anode	CVD[**] Anode
Sheet Resistance (<10 Ω/□)	5 Ω/□	10 Ω/□
Visible Transmission (>85%)	85%	80%
Work Function (>5eV)	5.3ev	5ev
Surface Roughness (R_q<2nm, R_{pv}<20nm)	Rq ~0.5nm R_{pv} ~10nm	Rq >8nm R_{pv} >30nm
Low-Cost Processing	Yes	Yes

#: PPG propriety metal based anode material
***: FTO material*

For the anode component of the integrated substrate, we have developed a low cost high performance metal containing material to replace the single ITO layer anode that is industry standard. The initial test at UDC with this anode found that it is not compatible with their chemical patterning and etch process. Thus, an alternative laser etching patterning process was used. The produced samples were evaluated for OLED performance and durability at UDC (see results in Task 9 section).

Task 5 - Determination of efficiency of Textured Glass EEL

Scattering type External Extraction Layer (EEL) was produced using a surface textured patterning equipment. Designed experiments were performed on the machine to study the haze as a function of machine and process variables using 24”X12” pieces of 2.5mm glass substrate. Samples with haze level ranged from 2% to 69% were produced (see Figure 10). The combination of IEL+EEL was also studied. An on-line produced 2.5mm IEL substrate with 59% measured haze was used for EEL processing on the opposite size. Table 10 is a summary of haze measurement for the produced samples. It showed that Insignificant haze increased with low haze level EEL. EEL and IEL+EEL samples will be characterized and sent to partner organization for OLED device fabrication and testing.

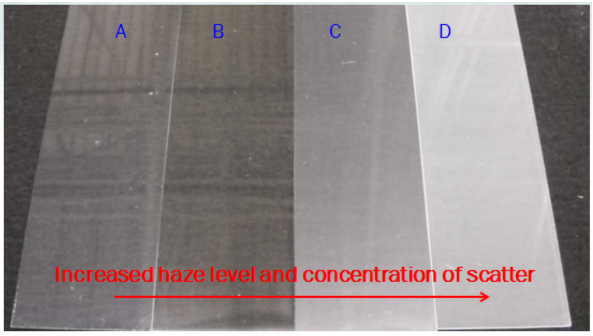


Figure 10 Clear glass processed with EEL in different haze level.

Table 10 %Haze for EEL, IEL, and IEL+EEL Samples

Sample #	EEL	IEL	IEL+EEL
A	2	59	60
B	8	59	60
C	12	59	65
D	68	59	87

The results from UDC on green OLED showed enhanced light extraction on both IEL and EEL samples. Although the measured EQE is low (12% to 16%), the photos of the OLED showed

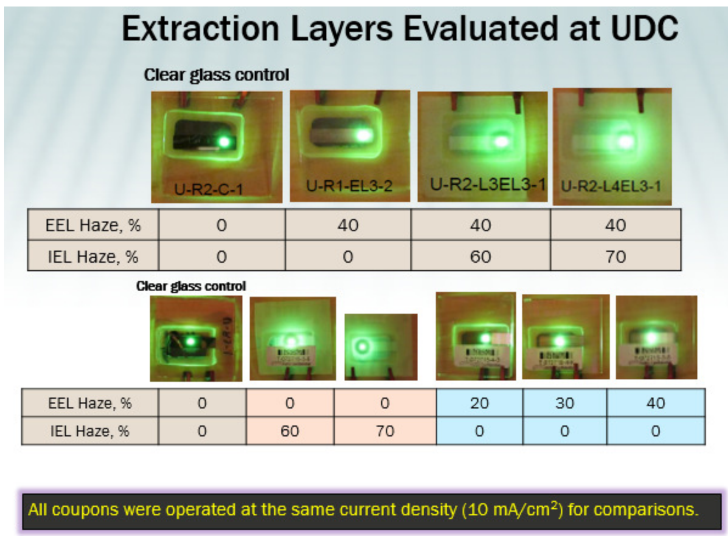


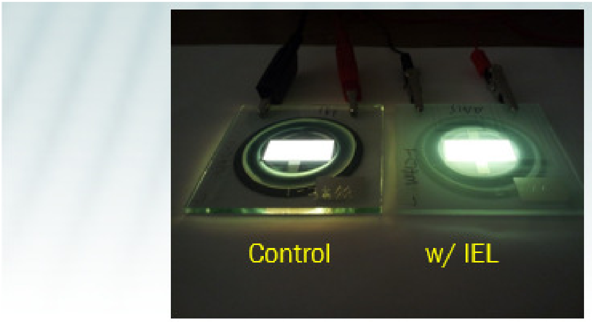
Figure 11 2.5mm glass samples with IEL and EEL evaluated with green OLED at UDC.

more light extraction on high haze IEL, EEL, and the combination of IEL+ EEL (see Figure 11). Note that a low EQE OLED device structure was used for these devices.

<u>Task 6: OLED Panel Fabrication and Characterization</u>

Initial on-line produced IEL samples were fabricated at OLEDWorks, Inc. for white emitting OLED (see the report in Task 2 section). In addition, matrix of integrated substrate configurations including individual elements of IEL and EEL along with combinations of each of these elements was evaluated (see the report in Task 5 section).

Figure 12 showed x1.25 light extraction enhancement for an IEL sample with 60% haze. This, combined with a high EQE OLED device structure enabled an EQE of 50% for the IEL-only (i.e., no EEL) sample.



IEL Haze, %	0	60
EQE, %	40	50

Figure 12 Anode laser etched 2.5mm glass sample with IEL evaluated at OLEDWorks.

<u>Task 7: Commercialization Plan</u>

A commercialization plan was presented to DOE in the continuation meeting on July 16^{th}, 2015 at PPG. Due to involving with business sensitive information, no details of that plan will be included in this report.

H. Technical Detail – Budget Period II

<u>Task 8: Manufacturing Process Improvements</u>

8.1 Manufacturing Process Improvements:

Uniformity and surface striation were improved continuously during this work by (1) moving the particle embedding tool to a region with less shrinkage while still allowing for complete embedment of the particles, (2) modifying the design of the tool, and (3) adjusting flow conditions. Thus, the effect of dynamic of glass forming process on stretching of glass was minimized, and a uniform environment for improved particle formation/deposition was provided. The effect of high line speed was also investigated. Both visual and tactile inspection of the produced samples was used as initial measures of the surface roughness. The sample generated at the new locations showed optimum depth of particle penetration and <5nm surface roughness (R_q).

Sand dumping experiments were conducted on-line to understand and minimize the effect of bath dynamics in the glass forming chamber on IEL surface quality. An optimal insertion location and orientation for the particle embedding tool was obtained and used following this experiment.

Uniform of chemistry flow through the feed slot is important in the generation of uniform particle embedding. A room-temperature physical model was constructed for this study (see Figure 13). The learnings from this investigation were used in the December 2016 on-line trials.

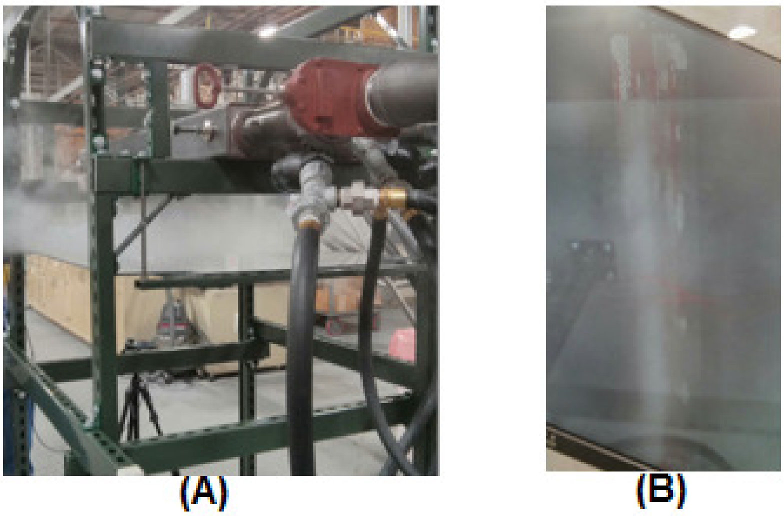


Figure 13 Cold model testing of flow patterns through a particle embedding tool. In the study, smoke generated using propylene glycol was doped in the flows (shown in A). Flow uniformity through the feed slot was monitored (as shown in B).

In addition to the physical flow modeling, out-of-bath testing of the flow uniformity through the feed slot was conducted by generating a flame through the slot for uniformity observation. A wide uniform flame was obtained after optimizing the slot width uniformity and back distributor plates. In addition CFD modeling was used to guide investigation of slot geometry designs for improved uniformity.

Gen 2 sized IEL glass substrates (470mm x 370mm) were produced in September, and Gen 2.5 (500mm x 400mm) substrates were achieved during the December 2016 trials for OLED qualification and evaluation. Figure 14 shows the online produced IEL glass substrates used in the large area OLED panel manufacturing.

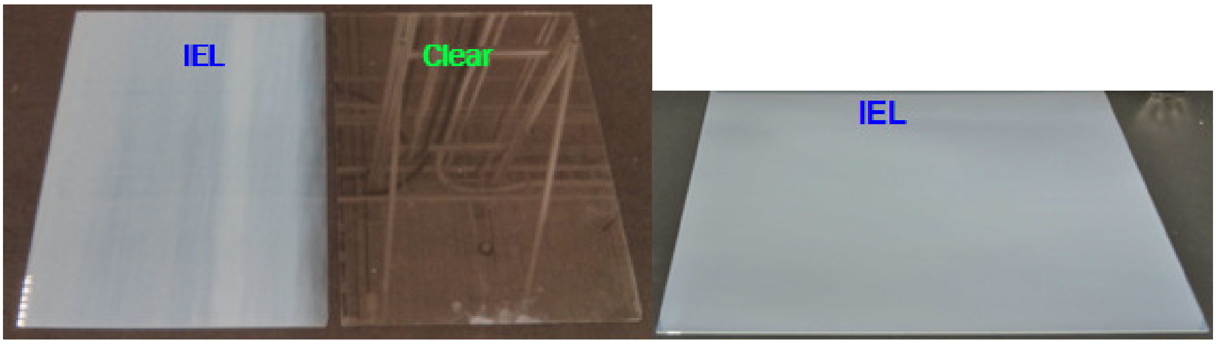
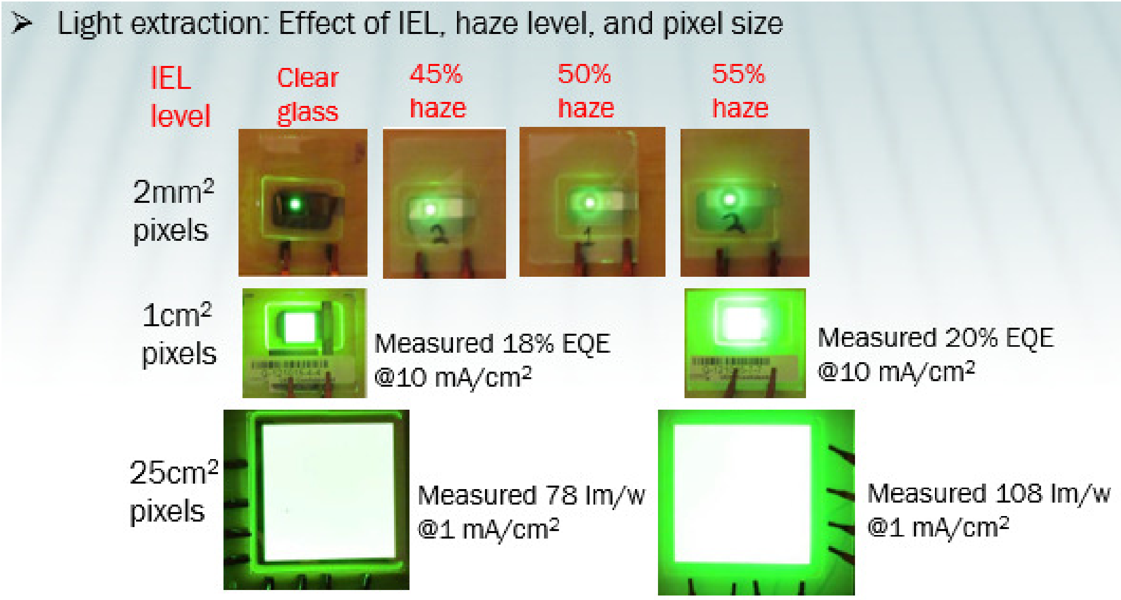


Figure 14 Photos of glass substrates prepared for OLED panel fabrication. The measured haze is ~40% for the IEL samples. (Left photos) Gen 2 (470mmx370mm) size, 2mm glass thickness, (Right photo) Gen 2.5 (500mm x400mm) size, 1.7mm glass thickness.

8.2 Device Manufacturing and Characterization:

1.7mm IEL glass samples with standard ITO anode coating were evaluated at UDC with green OLED at 2 mm^2, 1 cm^2, and 25 cm^2 pixel size. The light extraction performance and device % yield are encouraging (see Figure 15). The device lifetime measurement on 2mm^2 pixel OLED showed 15-20,000 hrs $LT_{80\%}$ and 50-70,000 hrs $LT_{50\%}$. These lifetimes are comparable to the UDC clear glass control substrate (i.e., the IEL does not impact the lifetime of the device), and meet our targeted project goal.



➢ Yield (%): 2mm^2 ,1cm^2, and 25 cm^2 pixel size

			Yield (%) (range)			
Pixel size	Anode coating	Pattering	Clear glass	45% haze IEL	50% haze IEL	55% haze IEL
2mm^2	ITO	Litho-graphy	76 (63/88)	41 (31/50)	63 (38/88)	91 (88/94)
1cm^2	ITO	Litho-graphy	90 (89/100)			84 (78/89)
25 cm^2	ITO	Litho-graphy	50 (25/75)			88 75/100)

Figure 15 1.7mm thick IEL glass samples evaluated at UDC with green OLED.

IEL glass samples were also provided to OLEDWorks for OLED cavity optimization. The targeted efficacy is 70% to meet DOE goals in 2020. 3 cm^2 white OLED pixels were processed on 2.5” x2.5” glass substrates. The results showed that the measured angle EQE was 50.5% for an IEL sample with about 0.5µm depth of particle penetration and 40% haze, and was 55.2% with an additional scattering type EEL film applied (provided by OLEDWorks).

OLEDWorks reported a color shifting to low wavelength (i.e., yellow-greenish) with IEL. However, an accurate white color was regained by adjusting red dopant concentration in the OLED device.

We have been working with OLEDWorks in validating our glass substrates in their OLED manufacturing process. The 3 phases of the validation process are (1) checking for any

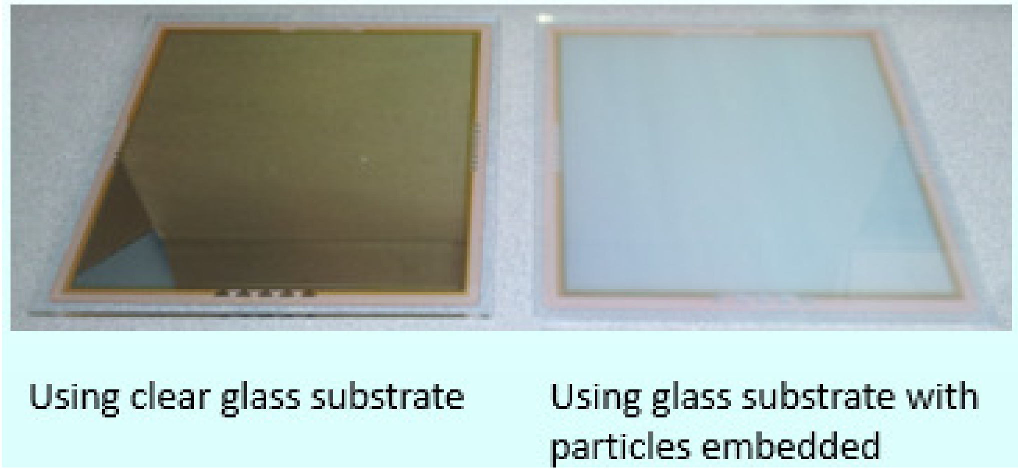


Figure 16 The produced white light OLED panels (120.5mm x 120.5mm) in Phase 2 manufacturing process qualification (left photo - using clear glass; right photo - using the on-line produced IEL glass).

process compatibility issues using 2mm thick Gen 2 size glass substrates, (2) OLED panel fabrication in their manufacturing process with anode coating, (3) repeat step 2 with full width Gen 2 size IEL and produce 120.5mmx120.5mm finished OLED panels with characterizations. The first phase was completed in March, and the second phase was completed in August. Figure 16 showed the produced devices. The anode film quality was good, and anode patterning was successful. Although the efficiency gain due to internal light extraction was not able to evaluated, the panels that functioned performed about as expected with the given white OLED stack. The third phase was started in September using the on-line produced IEL glass substrates with clear glass as control for comparisons.

8.3 Feedback Control Methodology:

On-line quality checking system was being developed with step-by-step approaches: (1) monitoring deposition uniformity, (2) measuring light scattering to correlate for haze level or particle concentration, (3) measuring back light scattering to correlate for surface defect, (4) implementing large area on-line system for feedback control. Due to the limited timing and higher priority on IEL development, the first two steps in this task were tested by the end of the project.

A line light source and detector combination was developed in the laboratory for haze level correlation measurement (see Figure 17). For this lab system in still condition, the particle size information can be obtained. However, when investigating the effect of line speed (i.e., with movement of the sample at speeds comparable to that of the manufacturing line), the particle size information became unavailable even when shorter wavelength of light sources were used (see Figure 18).

The developed monitoring system was tested on-line during the September and December 2016 IEL trials. Figure 19 showed the system installed in the downstream of on-line system for measuring light scattering through the particles embedded area.

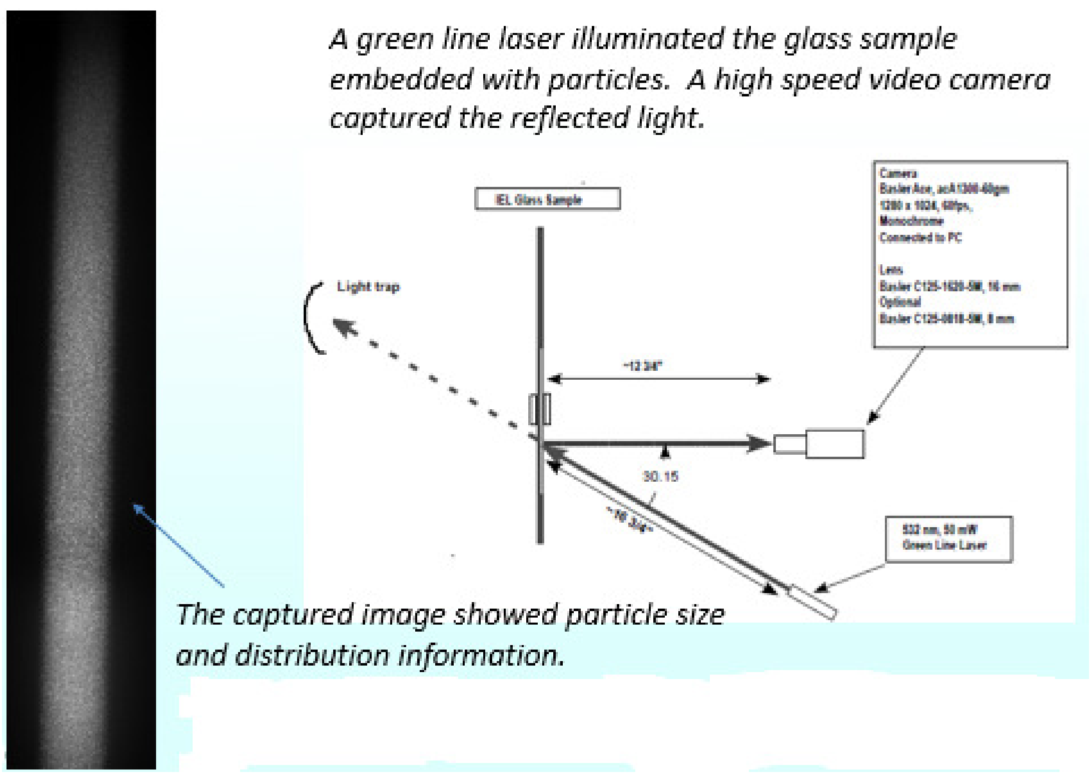


Figure 17 A lab monitoring system. A green line laser was used as light source. A high speed camera was used to capture the images of reflecting lights.

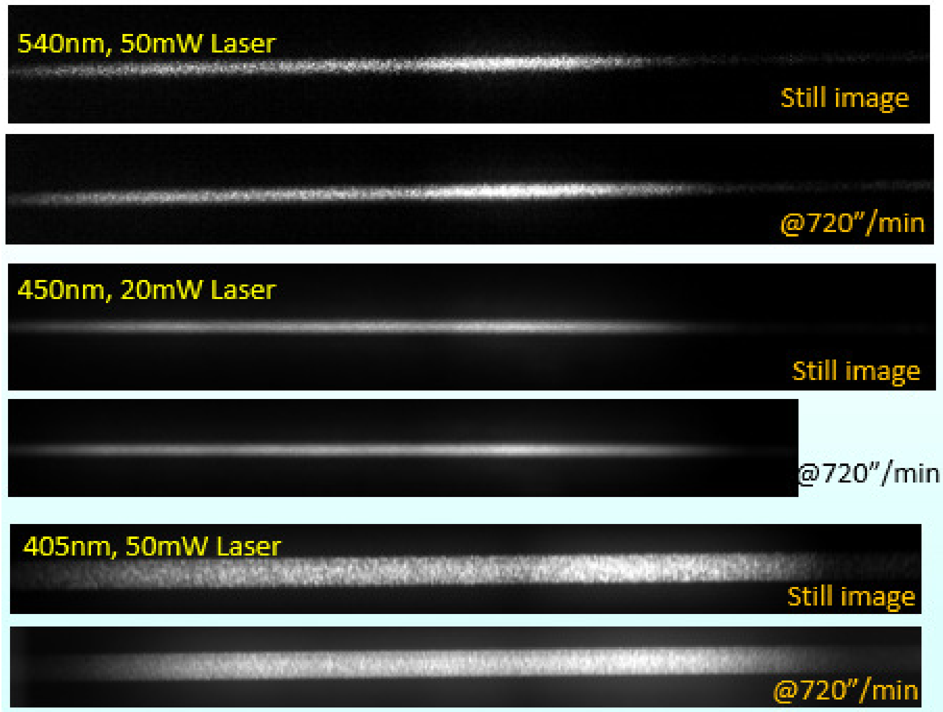


Figure 18 Effect of laser power, wavelength, and line speed on the quality of captured images of reflecting lights.

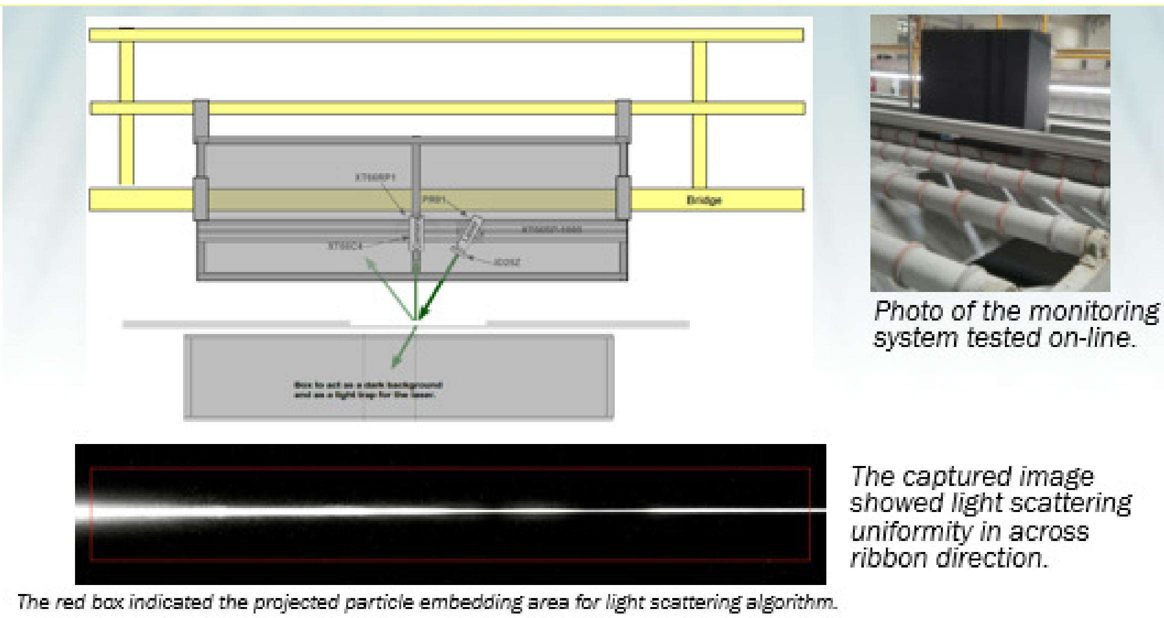
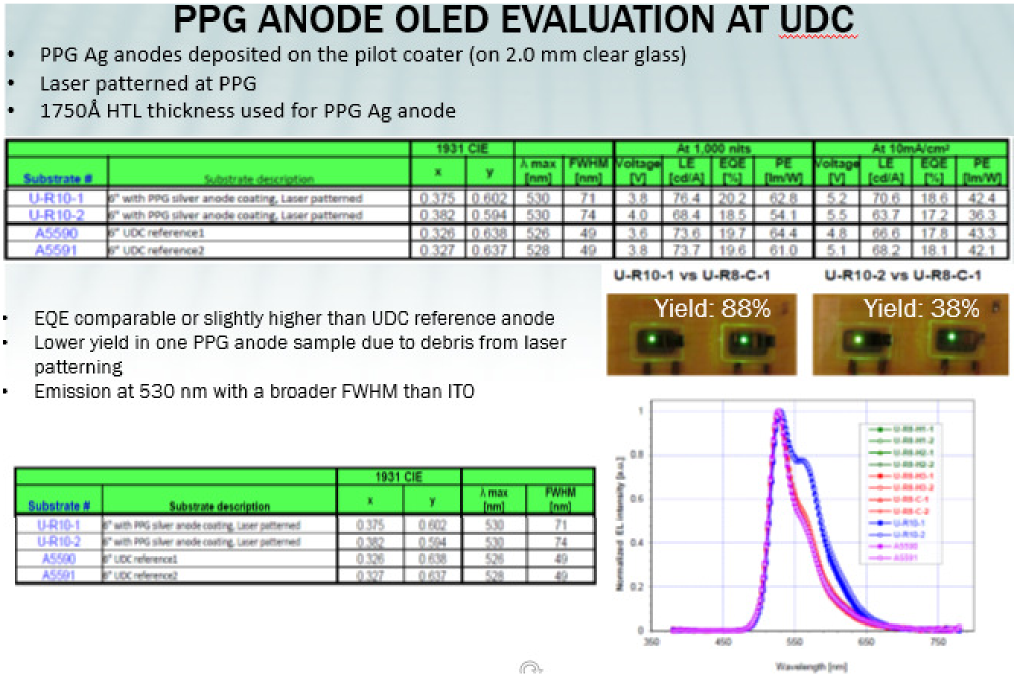


Figure 19 The monitoring system tested on-line.

Task 9: Large Area Sputtered Anode Process Development

Metal containing anode coating on clear glass and IEL glass was laser etched and evaluated with green OLED at UDC. The EQE and lifetime performance of PPG anode material were comparable to UDC ITO material and the third party ITO material (see Figure 20). The low device % yield was attributed to the laser etching process and further research is needed to develop this process.



Substrate #	Substrate description	1931 CIE x	1931 CIE y	λ max [nm]	FWHM [nm]	At 1,000 nits Voltage [V]	At 1,000 nits LE [cd/A]	At 1,000 nits EQE [%]	At 1,000 nits PE [lm/W]	At 10mA/cm² Voltage [V]	At 10mA/cm² LE [cd/A]	At 10mA/cm² EQE [%]	At 10mA/cm² PE [lm/W]
U-R10-1	6" with PPG silver anode coating, Laser patterned	0.375	0.602	530	71	3.8	76.4	20.2	62.8	5.2	70.6	18.6	42.4
U-R10-2	6" with PPG silver anode coating, Laser patterned	0.382	0.594	530	74	4.0	68.4	18.5	54.1	5.5	63.7	17.2	36.3
A5590	6" UDC reference1	0.326	0.638	526	49	3.6	73.6	19.7	64.4	4.8	66.6	17.8	43.3
A5591	6" UDC reference2	0.327	0.637	528	49	3.8	73.7	19.6	61.0	5.1	68.2	18.1	42.1

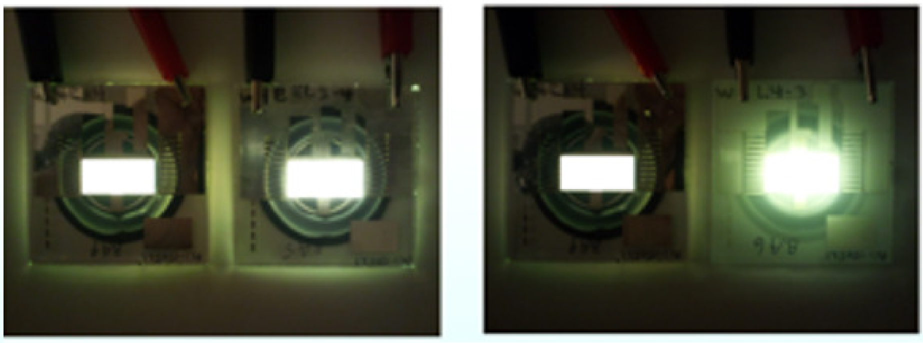
Substrate #	Substrate description	1931 CIE x	1931 CIE y	λ max [nm]	FWHM [nm]
U-R10-1	6" with PPG silver anode coating, Laser patterned	0.375	0.602	530	71
U-R10-2	6" with PPG silver anode coating, Laser patterned	0.382	0.594	530	74
A5590	6" UDC reference1	0.326	0.638	526	49
A5591	6" UDC reference2	0.327	0.637	528	49

Figure 20 PPG/Vitro anode evaluated at UDC.

Task 10: Textured Glass EEL Process Development

Early results from OLEDWorks white devices showed x1.2 light extraction enhancement on EEL (30% EQE) and x1.52 on IEL (38% EQE) as shown in Figure 21. Note the IEL samples were produced on-line and the EEL samples were produced in-house using surface texturing equipment.

Extraction Layers Evaluated at OLEDWorks



Clear Glass Control	EEL at 40% haze	Light Extraction Enhancement
25% EQE	30% EQE	1.20x

Clear Glass Control	IEL at 70% haze	Light Extraction Enhancement
25% EQE	38% EQE	1.52x

White OLED pixels in 3 cm^2 size were operated at the same current density (3 mA/cm^2) for comparisons. Extraction efficiency (in terms of EQE) was measured with angle integrated.

Figure 21 2.5mm glass samples with IEL and EEL evaluated with white OLED at OLEDWorks.

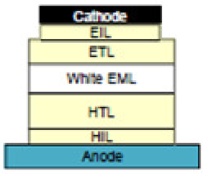
As discussed in the BP2 continuation meeting and approved in the revised SOPO / PMP, we used external EELs in conjunction with our IEL towards meeting the 70% EQE/IQE goal (DOE roadmap 2020 target). The recent data showed 55.2% EQE with the combination of IEL+EEL.

Task 11: Large-area OLED Panel Manufacturing

On-line produced IEL samples were provided to UDC for white OLED panel fabrication and evaluations. The final fabricated pixel size was 25cm^2. UDC developed a white OLED process and fabricated OLED pixels from 2 mm^2, 1 cm^2, and scaling up to 25 cm^2. The results are showed in the Figure 22. A summary of the results are listed as follows.

- x1.6 light extraction enhancement (32%EQE with IEL vs 19.3%EQE without IEL)
- 84 lm/w achieved with an additional EEL (57 lm/w with IEL and 84 lm/w with the combination of IEL+EEL)
- 57,000 hours life time LT_{70} achieved at 500 nits based on the accelerated test results at 40 mA/cm^2

		1931 CIE					At 1,000 nits*			
Substrate #	Substrate description	x	y	CRI	CCT	duv	Voltage [V]	LE [cd/A]	EQE [%]	PE [lm/W]
U-R14-C-7	PPG Clear glass	0.427	0.439	80	3,443	0.015	4.6	46	20	31
U-R14-C-8	PPG Clear glass	0.406	0.455	80	3,915	0.026	4.7	44	20	30
U-R14-C-9	PPG Clear glass	0.395	0.454	78	4,127	0.028	4.7	40	18	27
U-R14-C-10	PPG Clear glass	0.398	0.456	78	4,086	0.028	4.8	43	19	29
U-R14-H1-2	PPG Glass with EIL	0.406	0.442	81	3,852	0.021	4.3	72	32	52
A9432	UDC ref	0.408	0.451	81	3,850	0.024	4.5	43	18	30



*Forward luminance was measured
Devices were assumed Lambertian

Figure 22 J-V-L characteristics of WOLED 25 cm^2 PHOLEDs using PPG/Vitro substrates.

For final deliverables of the project, we have been working with OLEDWorks, LLC to fabricate 120.5mm x 120.5mm white OLED panels using Gen 2 size glass substrates. Vitro provided on-line produced IEL substrate in Gen 2 size; OLEDWorks used the anode coated, patterned (coating and patterning done by a third party) glass substrates in their manufacturing process to fabricate OLED panels and characterize the produced panels. Two type of IEL samples were provided for OLED evaluations. Type 1 samples were approximately 60% covered with IEL with Gen 2 sized substrates, while Type 2 samples were 100% covered with IEL in the OLED processing area. The clear glass samples and Type 1 samples were processed through anode coating, patterning, and OLED coatings with no issues. However, the Type 2 samples had photoresist residue from anode patterning, this resulted in high voltage and low yield on the OLED panels fabricated using these samples. This residue showed as water mark like staining during inspection after patterning and as wipe marks after OLED processing. Since each Gen 2 substrate can be produced 6 OLED panels in 120.5mmx120.5mm size, the Type 1 samples were still be able to produce 3 large area OLED panels from each Gen 2 substrates.

Characterization results for the IEL large panel OLED devices are as follows:

- Yield – 67% for clear, 59% for IEL (Yield losses are due to the combined contributions from IEL quality, anode coating, patterning, and OLED processing)
- EQE enhancement – x1.43 (averaged 52.8% EQE)
- Efficacy enhancement – x1.66 (averaged 29 lm/w)

I. Budget Overview:

Table 11 Project Spend vs Plan

Total Amount of Award	Total	Government Share
Total	$4,704,593	$2,352,296
Budget Period 1	$2,746,193	$1,373,097 (100% spent)

Budget Period 2	$1,945,083	$1,945,083 (100% spent)
Actual Spend* through 9/2016	$4,788,211	$2,352,296 (100% spent)
PPG Overage* through 9/2016	$83,618	$0

**The Actual spend numbers include all actual labor and line time charges, and now include indirect rate adjustments through 2014. Further increases may result when final approved indirect rate adjustments are applied.*

J. Patents:

No patent applications have been filed to date under this DOE funding.

K. Publications/Presentations: Several talks and poster presentations were given during the course of the project

- DOE SSL R&D Workshop, San Diego, CA, May 7, 2014: C. Hung gave an invited talk on" Large area integrated substrate for OLED lighting" and presented a poster on the project progress
- Peer Review, Morgantown, WV, June 11, 2014: C. Hung, J. McCamy, and S. Benton updated on the project progress
- DOE SSL R&D Workshop, San Francisco, CA, January 28, 2015: C. Hung presented a poster on the project progress
- EERE BTO Peer Review, Vienna, VA, April 14, 2015: C. Hung presented updates on the project progress
- Peer Review, Morgantown, WV, June 9, 2015: C. Hung, J. McCamy, M. Arbab, and S. Benton updated on the project progress
- Budget Period I Review, Cheswick, PA, August 29, 2015: Budget period I review and commercial plan presentation were conducted at PPG Glass Business and Discovery Center.
- DOE SSL R&D Workshop, Raleigh, NC, February 2, 2016: C. Hung presented a poster on the project progress
- PPG Cross SBU Group Meeting, Pittsburgh, PA, June 21, 2016: C. Hung presented a poster about the project
- Project Review/DOE Visit, Cheswick, PA, August 29, 2016: Budget period II review was conducted at PPG Glass Business and Discovery Center.
- DOE SSL R&D Workshop, Long Beach, CA, 2017: C. Hung presented a poster on the project progress