Project End Report

Project Title: Recovery Act: Low Cost Integrated Substrate for OLED Lighting Development

Period of Performance: April 1, 2010 to September 30, 2012

Recipient: PPG Industries, Inc.

Award Number: DE-EE0003209

Date of Report: December 26, 2012

Period Covered by Report: April 1, 2010 to September 30, 2012

Working Partners: Universal Display Corporation

Cost-Sharing Partners: Universal Display Corporation

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Project Objective:

The primary objective of this project was to develop and demonstrate a new, low-cost, integrated glass substrate product based on large-scale glass coating production for use in manufacturing organic light-emitting diode (OLED) solid-state lighting devices that meets or exceeds anticipated future competitive performance and cost requirements. The integrated substrate (**Figure 1**) consists of a low-cost float glass substrate, coated with an anode film layer that achieves optimal performance, and integrated light out-coupling layer(s) that increase the efficacy of OLED devices.

Figure 1: Schematic of an integrated float glass based substrate for OLED lighting developed by PPG Industries.

The economics of this integrated glass substrate product manufactured by large-area coating methods are anticipated to have the potential to achieve the DOE price targets of less than \$36/m² by the end of 2013.

With further commercialization development, the technology is anticipated to ultimately achieve the performance goals and cost target of less than \$26/m² by 2015.

Budget Overview:

Total Award Value: \$2,140,040.00

Total Federal Share of Award: \$1,672,072.00

Required Cost Share: \$467,968 (\$268,168.75 PPG + \$199,799.25 UDC)

Federal Funds Received: \$1,672,072.00

Actual Cost Share: \$810,412.52 (\$391,322.80 PPG + \$419,089.72 UDC)

Figure 2: Project Spend vs Plan

	Total		Government Share	Cost Share	
	Plan	Actual	Plan/Actual	Plan	Actual
PPG	\$1,269,473	\$1,439,160	\$1,047,837	\$221,636	\$391,323
UDC	\$870,567	\$1,043,324	\$624,235	\$246,332	\$ 419,090
Total	\$2,140,040	\$2,482,484	\$1,672,072	\$467,968	\$810,413

Table 1: Project Spend vs Plan

Figure 3: Government Share Spend vs Plan

Technical Details:

Substrate

The objective of this task was to check the compatibility and determine the suitability of PPG's float glass substrates with UDC's device fabrication equipment and practices. The normal practice at UDC is to use 152.1mm x 152.1mm x 0.7mm glass with high dimensional tolerance for OLED processing. Early testing needed to be performed to determine the capability of UDC's OLED manufacturing equipment to process thicker glass. PPG established an internal high tolerance glass cutting capability and prepared raw PPG float glass samples of different thicknesses (2, 3.2 and 4.0 mm) for testing processing compatibility at UDC. With some processing modifications and new holding fixtures, UDC determined that a glass thickness of 2.0mm would be compatible with their OLED fabrication equipment. For development purposes, it was decided to use PPG Solarphire® PV float glass instead of the Clear float glass since it is readily available in the 2.0mm thickness and has higher transmittance (**Figure 4**). Higher transmittance or lower absorbance is also important as it increases the probability of light in the substrate waveguide mode to escape in case of scattering centers being used in light extraction layers.

Figure 4: A comparison of the light transmittance between 2.0mm Clear float glass substrate and Solarphire® (low-Fe float) glass substrates.

Atomic force microscopy (AFM) measurements were done on uncoated 2.0mm Solarphire® PV glass to get baseline roughness measurements for the uncoated glass. The RMS roughness ranges were found to be approximately between 3 to 6 Å (results in **Table 2**), which are typical for glass and were determined to be optimal for anode fabrication.

	Area Measured (μm^2)	RMS Roughness (nm)	R_a Roughness (nm)
Air Side	4.0	0.302	0.212
	25.0	0.448	0.289
Tin Side	4.0	0.407	0.254
	25.0	0.543	0.320

Table 2: RMS and Average roughness values for 2.0mm Solarphire® PV glass.

In addition, accelerated lifetime tests were performed to compare the life-times and voltage variation of devices fabricated on float glass substrates with and without sodium barrier layers. No differences were found and it was concluded that no barrier layers need to be used for OLED lighting device fabrication. It is important to note that this result may be OLED fabricator process-dependent since a process involving prolonged exposure of float glass substrates (e.g. photoresist baking prior to exposure for anode patterning) to elevated temperatures may require the use of sodium barrier layer between the glass substrate and anode layers.

In conclusion, PPG demonstrated that, through careful selection and proper handling, float glass substrates could displace expensive borosilicate or double side polished glass substrates. This result by itself has the potential to significantly reduce the cost of the substrate.

Anode

In this task, four alternate anode materials to ITO were evaluated using pilot-scale MSVD and CVD processes. The coatings were characterized, performance was compared to ITO electrodes, and low-cost fabrication methods were developed. Anode coating development efforts were also focused on optimizing the optical, electrical, and surface quality to make them suitable for OLED device fabrication at UDC. Surface defect density, anode-to-substrate adhesion, and mechanical durability were evaluated; stability during post-heating and chemical exposure expected during OLED processing was also tested.

Table 3 shows a summary of properties that were achieved for the various candidate anode materials evaluated. Also shown for comparison are results for commercial ITO deposited on 0.7 mm double side polished soda-lime glass as a control substrate, which UDC routinely uses for OLED processing.

Coating Type	Sheet Resistance ($\Omega/\square$)	Transmission @ 550nm	RMS Roughness (nm)
Control	18	83	3
Anode 1	10	85	14
Anode 2	8.5	84	6
Anode 3	21	84	1
Anode 4	7.3	89	0.5
Requirement	10	85	2

Table 3: Electrical, optical, and surface roughness properties measured for candidate anode materials chosen for evaluation in the Benchmarking Task. Sheet resistance was measured using a four-point probe. Transmission was measured using PerkinElmer Lamda 9 and Hunterlab UltraScan PRO spectrophotometers. Roughness was determined using a Digital Instrument Dimension 3100 Atomic Force Microscope.

Sheet resistance and transmission values (@550nm & average visible transmittance (not shown)) were comparable or better than that of ITO. In certain cases (Anode 3 & 4) much smoother films were achieved. Anode 1, however, was much rougher as compared to ITO films and strategies to address this drawback are discussed later in this section.

Work function measurements were performed using an AC-2 photoelectron spectrometer before and after UV exposure and compared to commercially available ITO anodes. The results are summarized in **Table 4**.

Coating Type	Before UV Exposure (eV)	After UV Exposure (eV)
ITO- Control	4.7	5.2
Anode 1	4.63	4.96
Anode 2	4.50	5.08
Anode 3	4.81	5.40
Anode 4	4.8	5.33
Requirement	-	>5.0

Table 4: Work-function comparisons for PPG anodes performed using an AC-2 photoelectron spectrometer. Please note that the ITO anode is a commercially available product used by Plextronics, Inc. for device fabrication.

It can clearly be seen that the work function for all the anode materials was comparable to the ITO anode after UV exposure.

Under the Anode Development task, alternate large area methods to manufacture anode coatings at low-cost and improved properties were developed. **Table 5** summarizes the results achieved for three of the candidate anode materials. For Anode 1 an alternate lower cost deposition technique were explored. This process led to smoother films with some compromise in electrical and optical properties. Separately, process improvements were made to achieve better quality Anode 3 & 4 films.

Coating Type	Sheet Resistance ($\Omega/\square$)	Transmission @ 550nm	RMS Roughness (nm)
Anode 1	10	85	14
Anode 1 (Alternate Process)	16	83	6
Anode 3	21	84	1
Anode 3	17	89	1
Anode 4	7.3	89	0.5
Anode 4	5.5	90	0.5

Table 5: Electrical, optical and surface roughness properties achieved for three candidate anode materials by testing alternate low-cost deposition techniques and process improvements under the Anode Development task. The roughness of Anode 1 deposited using the second lower-cost fabrication technique could not be determined conclusively.

To test these anodes, green PHOLEDs (2mm x 2mm pixels) were fabricated at UDC for all PPG candidate anode materials deposited on 2.0mm Solarphire® PV glass. **Figure 5** shows a schematic of UDC's green PHOLED structure. **Figure 6** is picture of a powered green PHOLED device fabricated on a PPG anode-coated substrate.

Figure 5: Schematic of UDC's green PHOLED structure fabricated on 2.0 mm Solarphire® PV float glass substrates coated with different candidate anode materials and deposition processes. (ETL: Electron Transport Layer, BL: Blocking Layer, EML: Emissive Layer, HTL: Hole Transport Layer, HIL: Hole Injection Layer)

Figure 6: A powered 2mm x 2mm green PHOLED device fabricated on a PPG anode coated substrate.

Anode coated substrates were processed and tested according to standard procedures used at UDC. The samples were first cleaned and visually inspected. Following this, photolithography was used to pattern the anode coating into a series of stripes. A polyimide grid was then deposited onto the anode, defining a 2mm x 2mm pixel area within each anode stripe. After oxygen-plasma and UV ozone pretreatment, samples were transferred to a vacuum thermal evaporation (VTE) system, where organic and metal cathode layers were deposited. To ensure a fair comparison, a well-characterized standard UDC PHOLED structure was used to evaluate all new anode coated substrates and the same PHOLED was fabricated on commercial ITO-coated 0.7mm polished soda-lime glass substrates as controls. After PHOLED deposition, encapsulation was performed in a glove box. A PR-705 spectroradiometer was used to measure current density-voltage-luminance (JVL) response and electroluminescence (EL) spectra. A 20-inch integrating sphere from Labsphere and an imaging sphere from Radiant Imaging were used to measure total light output. Accelerated life testing was measured at room temperature using high power ($J=40\text{mA}/\text{cm}^2$) OLED operating conditions. Also, the HTL thickness was adjusted for PPG anode coated

substrates to give optimal color and efficiency. **Table 6** shows data from one such device optimization task performed for Anode 2.

Anode	HTL	1931 CIE		$\lambda_{\max}$ [nm]	FWHM [nm]	At 1000 nits			
		x	y			Voltage [V]	LE [cd/A]	EQE [%]	PE [lm/W]
Anode 2	1100	0.327	0.631	530	62	4.9	55.3	15.0	35.2
	1000	0.322	0.634	528	58	5.0	54.2	14.9	34.3
	900	0.320	0.631	524	56	4.9	52.9	14.9	33.7
	800	0.321	0.629	522	56	4.9	50.9	14.4	32.8
	700	0.327	0.620	522	60	5.0	47.4	13.6	30.1
	600	0.341	0.608	522	68	4.9	45.2	13.1	29.2
	500	0.345	0.605	522	72	4.8	46.9	13.5	30.5
	400	0.356	0.598	522	80	4.9	44.7	12.8	28.9
	300	0.360	0.599	524	84	4.8	44.6	12.5	29.3
Control	300	0.346	0.614	528	74	4.8	58.1	15.9	37.9

Table 6: Table comparing EL and JVL data obtained from green PHOLED devices optimization task for Anode 2. The results of optimized devices on Anode 2 and control substrate are highlighted.

Results for devices fabricated on PPG substrates coated with candidate anode layers were promising and comparable to the commercial control substrate. Based on cost-benefit analysis Anode 1 and Anode 4 were downselected. Anode 1 is based on a CVD process, while Anode 4 is based on a MSVD process.

Table 7 is a summary of relevant results from EL, JVL, and lifetime tests.

Anode	HTL	1931 CIE				At 1000 nits				At 40 mA/cm ²	
	Thickness (nm)	x	y	$\lambda_{\max}$ (nm)	FWHM (nm)	Voltage (V)	LE (cd/A)	EQE (%)	PE (lm/W)	L ₀ (nits)	LT _{80%} (h)
Commercial ITO	30	0.346	0.614	528	74	4.8	58.1	15.9	37.9	18,582	236
Anode 4	110	0.315	0.643	528	62	5.3	85.7	22.3	51.2	26,158	320
Anode 1	100	0.333	0.622	526	68	5	44.7	12.3	27.8	14,324	305

Table 7: Summary of the EL, JVL, and life-time tests obtained from green PHOLED devices UDC fabricated on PPG 2.0mm Solarphire® PV float glass substrates coated by PPG with downselected anode materials. For comparison, data is shown for identical devices fabricated with a commercial ITO control substrate.

As can be seen from the results, devices fabricated on Anode 4 had better efficiency than the devices fabricated on standard ITO electrodes (external quantum efficiency (EQE) of 22.3% as opposed to 15.9%) at a luminance of 1000 nits. It is pertinent to point out that due to optical cavity effects arising from a multi-layered electrode structure, light is directed preferentially in the forward direction. It could be construed that this may result in higher efficiency because of the Lambertian intensity distribution assumed in the calculations. As a cross-check, a direct measurement was performed to determine the efficiency of the OLED devices fabricated on Anode 4 electrodes using an integrating sphere. These results also showed equal or better efficiency performance than the devices fabricated on ITO anodes. Another advantage of using this electrode is reflected in the lifetimes which are much longer for green PHOLED devices (LT₈₀ (Time required to reach 80% of initial luminance) of 320 hours as opposed to 236 hours for devices fabricated on ITO anodes).

From **Table 7**, it can be seen that devices fabricated on Anode 1 resulted in lower performance as compared to the control ITO anodes. The primary reason for this was determined to be the higher surface roughness ($R_{\text{rms}} \sim 8\text{-}14\text{nm}$) of these anodes resulting in current leakage and affecting the performance of the devices. As a result of anode surface roughness, some of the devices fabricated on these anodes showed “dead spots” which may explain the lower efficiency.

The surface roughness of these anodes was improved from 14nm to 8nm by varying the deposition chemistry and other parameters. The laboratory-scale surface roughness improvement results were also successfully demonstrated on a manufacturing line. Recent results on the pilot-scale CVD coater indicate that there is the potential for even further improvement of the surface roughness of these anode layers.

Another approach that was explored is the use of a solution coated- HIL film to circumvent the surface roughness these anodes. PPG has tested this concept in partnership with another OLED fabricator. In preliminary studies, it has been shown that Anode 1 can be effectively planarized by using the solution coated-HIL films. **Figure 7** shows an optical micrograph of a powered 3mm x 3mm orange PHOLED pixel that was fabricated on Anode 2 using an aqueous HIL. No “dead spots” were observed and the device yield was much higher as compared to those fabricated without a solution coated-HIL film.

Figure 7: An optical micrograph of a powered green PHOLED pixel fabricated on Anode II showing dead spots and an orange PHOLED pixel fabricated on Anode II using a solution coated-HIL film.

The device performance results are summarized in **Table 8**. It should be noted that even without optimizing the HIL for the Anode 2, operating voltages, efficiencies and lifetimes for the devices fabricated Anode 1 were observed to be better than those fabricated on the commercial ITO anode.

Anode	At 1000 nits					
	Voltage (V)	Current Density (mA/cm ²)	Luminous Efficiency (Cd/A)	Power Efficiency (Lm/W)	EQE (%)	LT50 (hrs)
Commercial ITO	2.8	10.3	9.7	10.9	4.2	1736
Anode 1	2.79	8.9	11.3	12.7	5.2	1932

Table 8: Performance comparison of the orange PHOLED devices fabricated on Anode 1 and commercial ITO anode.

In conclusion, this program developed and demonstrated two low-cost alternates to a conventional ITO anode, both of which are conducive to OLED device fabrication.

Light Extraction Layers

External Extraction Layers (EELs)

A number of scalable EEL technologies, based on the concepts of light scattering and changing the exit angle of the light at the substrate/air interface, were evaluated under this project.. These technologies were

selected on the basis of scalability and compatibility with glass manufacturing. For screening purposes, a bench-top system was developed (**Figure 8**) to qualitatively compare the external extraction layers.

Figure 8: A schematic of a table top system developed to screen the external extraction layers. The detector was an integrating sphere with an ability to measure the intensity in the visible spectrum. The area under the intensity-wavelength curve was integrated and compared for a qualitative comparison of light extraction efficiency.

Screening was also performed by fabricating large area green PHOLED (**Figure 5**) pixels (~0.6cm x 5cm) on PPG's integrated float glass substrates. For confirmation of results large area white PHOLED pixels were fabricated. A schematic of the white device is shown in **Figure 9**. Photographs of powered large area green and white PHOLED devices fabricated for extraction enhancement measurements are shown in **Figure 10**. The total emissive area for this design was 19.11 cm² on a 35 cm² substrate. Extraction measurements were performed using a 20" integrating sphere at a constant device current density of 2mA/cm² (corresponding to approx. 1000 nits). Optimized devices (by varying the HTL thickness) were constructed on substrates with the light extraction layers and control substrates. Angular variation in color with the angle of emission and spectral dependence were also studied for various LEL films.

Figure 9: A schematic of the white PHOLED device constructed on PPG's float-glass substrates.

Figure 10: A powered large area (a) green and (b) white “vertical blind” PHOLED device fabricated by UDC on PPG’s Float Glass-based integrated substrate for measurement of extraction enhancement from light extraction layers.

Sol-Gel EEL

Pyrolytic sol-gel spray coatings were evaluated as a candidate EEL technology due to its compatibility with the large-scale float glass manufacturing process. The process was developed on a TamGlass system that allowed the uniform coating of 12”x12” substrates. Process parameters such as glass temperature, spray time, and spray gun pressure were optimized to achieve various combinations of haze/scattering and transmission levels. Durability tests were done to ascertain the compatibility of these EELs with UDC’s device fabrication process, which identified some issues. The device fabrication process was modified to accommodate the EEL layers. Initial small scale (2mm x 2mm) devices showed enhancement factors of 1.28x. However, the results could not be confirmed with large area devices. The process for fabricating large area devices is slightly different and more demanding than the smaller devices and therefore magnified the durability issues in these layers. This was due to delamination of the sol-gel films because of high-temperature patterning photoresist baking step followed by exposure of the films to the ITO acid etch solution. An example of a failed sol-gel coating is shown in **Figure 11**.

Figure 11: An optical micrograph of a delaminated sol-gel EEL layer following exposure to the ITO etchant.

Various approaches were taken to address the issue. In the first approach, the sol-gel film preparation was followed by a post-deposition annealing step to improve the adhesion of the films. The second approach involved altering the photoresist baking temperature and ITO etching parameters. Lastly, a clean-room adhesive tape was tested to as an encapsulant to protect the sol-gel EEL films from the ITO etchant. However, only limited success was achieved and it was decided to concentrate the project's efforts on other approaches.

Textured Glass EEL

Scalable textured glass EELs and fabrication technologies were also investigated and developed under this program. This concept was based on the change of the light exit angle at the substrate/air interface. PPG evaluated chemical etching and mechanical abrasion as two scalable techniques to achieve texturing of glass surfaces. It was demonstrated that comparable optical properties could be achieved using both methods. Additionally, light extraction factors were found to be similar for both types of EELs, measured using large-area green PHOLED devices.

Chemical Etching: A chemical etching process was developed for 2.0mm thick glass with lateral dimensions of 24"x48". Process parameters were optimized to achieve three different kinds of textures. Analytical measurements were performed to characterize the optical and morphological properties for these textures. **Table 9** shows the roughness values for these textures measured using a contact profilometer.

Finishes	R_a (μm)	R_q (μm)	R_t (μm)
Type I	2.59	3.49	22.89
Type II	1.85	2.29	17.29
Type III	1.63	2.04	14.14

Table 9: Roughness values measured using a contact profilometer for three different acid etched textures.

Large-area green PHOLED devices were fabricated and extraction efficiency enhancement measurements were performed. **Table 10** shows the color and enhancement variation for various acid etched EELs.

Sample Type	Transmission (%)	Haze (%)	CIE x	CIE y	Output (Lumens)	Enhancement
Control- No EEL	85	0	0.331	0.617	7.09	1.00x
Type I	84.1	100	0.337	0.611	8.55	1.21x
Type II	83.3	78	0.338	0.609	7.82	1.10x
Type III	84.8	56	0.339	0.608	7.58	1.07x

Table 10: Efficacy enhancement for three types of acid etched EEL samples measured using large-area green PHOLED devices.

Type I textured glass demonstrated the best extraction performance. A large-area white PHOLED device was subsequently fabricated on similar 2.0mm float glass based EEL substrate and characterized and the results are shown in **Table 11(a)**. The hole transport layer (HTL) was optimized to achieve the best device performance. The optimum HTL thickness was found to be 165nm. All white devices had a correlated color temperature (CCT) of 3400K. **Table 11(b)** shows the characteristics of the device at 1000nits. A power efficiency of 48 lm/W was achieved.

Sample Type	CIE x	CIE y	Output (Lumens)	Enhancement
Control (No EEL)	0.412	0.409	5.77	1.00x
EEL	0.412	0.404	7.3	1.27x
Control + Diffuser	0.423	0.406	7.54	1.31x

(a)

	HTL	1931 CIE		At 1000 nits		
Sample	Thickness (nm)	x	y	Voltage (V)	Output (Lm)	PE (lm/W)
EEL	165	0.408	0.404	4	6	48

(b)

Table 11: Summary of the light extraction measurements performed using the white PHOLED devices.

A comparison between the performance of the device fabricated on a substrate with and without EEL determined the extraction enhancement to be around 1.27x. There was no significant color shift due to the textured glass EEL. Extraction measurements performed using a standard diffuser sheet resulted in an enhancement factor of 1.31x, which is similar to the textured glass substrate. UDC had previously achieved extraction numbers of around 1.4x using this sheet in combination with a 0.7mm thick glass substrate. In order to determine if the glass substrate thickness made an impact on the extraction factor, several measurements were performed using an integrating sphere and diffuser sheet mounted on glass substrates with varying thickness. The trend is shown in **Figure 12** and shows an improvement in light extraction factor with decreasing thickness.

Figure 12: Variation of the extraction factor with glass thickness for a diffuser EEL film. The measurements were performed for varying thickness of clear glass and using an integrating sphere setup that has been previously discussed.

Angle from Normal	Luminance [nits]	CIE 1931 (x, y)	L / Lmax
0	1219	(0.322, 0.627)	1.000
10	1263	(0.323, 0.626)	1.036
20	1246	(0.326, 0.623)	1.022
30	1225	(0.331, 0.618)	1.005
40	1251	(0.338, 0.612)	1.026
50	1299	(0.342, 0.610)	1.065
60	1364	(0.342, 0.612)	1.119
70	1408	(0.337, 0.618)	1.155
80	1352	(0.332, 0.622)	1.109

Table 12: Luminance and color variation measured for a large-area green PHOLED device fabricated on a type I EEL. The luminance and color do not vary much with angle.

This light scattering EEL approach is favorable because not only it is a low-cost alternative but because of other advantages such as no absorption, high durability, and compatibility with OLED device fabrication process. This EEL when incorporated in the luminaire will improve the spatial uniformity of light/no glare (**Table 12**) and mask the appearance of thick gridlines.

Mechanical Abrasion: In an effort to explore low-cost EEL manufacturing routes, the mechanical texturing of glass was explored as an alternative to acid etching. Mechanical abrasion of glass was demonstrated using abrader equipment from Timesavers, Inc. (**Figure 13**) on 36"x36" pieces of 2.0mm

thick glass using a high throughput process. Designed experiments were performed to achieve varying levels of haze (scattering) and to identify suitable abrasive belts and brushes.

Figure 13: A picture of the Timesavers equipment used for mechanical abrasion of glass to generate EEL for OLED lighting applications.

It was demonstrated that optical characteristics similar to those of acid etched samples could be achieved using a mechanical texturing process. **Table 13** is a comparison of the optical properties that were achieved using the two techniques. The results were verified with large area device fabrication on mechanically textured samples.

	Chemical Etching		Mechanical Abrasion	
	Transmission (%)	Haze (%)	Transmission (%)	Haze (%)
Type I	92.4	57.4	92.3	57.7
Type II	88.8	77.6	87.3	80.4
Type III	82	99.1	81.1	92.9

Table 13: Comparison of transmission and haze achieved on 2.0mm float glass substrates using scalable chemical etching and mechanical abrasion techniques. Various sample types correspond to different processing parameters.

Internal Extraction Layers (IELs)

PPG explored several technologies, including flame spray-based embedded oxide particles and sol-gel coating, to develop an IEL layer. These technologies were selected based on cost effectiveness, scalability and time-to-market criteria. Standard large area green and white PHOLED devices, as described in the previous section, were fabricated to test and measure the light extraction factors for these IELs.

Embedded Particle IEL:

The objective of this activity was to develop an in-glass particulate-based scattering layer to be used underneath the anode as an internal light extraction feature. The enhancement of the light extraction is largely dependent on the choice of particulate material, the refractive index and absorption coefficient of the oxide particles, the size and size distribution of the incorporated particles, the depth and concentration of the particle incorporation, and the final haze level. The scattering particles were embedded beneath the glass surface by an atmospheric flame spray process. In principle, the flame spraying can be performed on a tin bath float line as the glass itself is produced. Also, since these particles are embedded inside the

glass surface while the glass is still viscous, the glass surface can be expected to remain very smooth and suitable for anode material deposition.

This project employed a laboratory-scale conveyor furnace, wherein the particulate-based scattering layer was sprayed on the glass substrate at a temperature of 600°C to 800°C. The particulate precursor solution (for example, precursors for alumina) was diluted in water or other organic solvents and fed into the flame directed at the glass surface. The combustion of the precursors and solvents resulted in the decomposition of the precursors to oxide nanoparticles. As the glass substrate translated underneath the flame burner, oxide particles were incorporated into the glass surface.

(a)

(b)

Figure 14: Field emission Scanning Electron Microscopy (SEM) images of a (a) cross-section and (b) surface of an embedded oxide nanoparticle IEL sample.

Figure 14 (a) shows a cross-section SEM image of one such embedded oxide particle sample. A wide distribution of oxide particles (few hundred nanometers in diameter) were noticed in the samples. The typical impingement depth was 1-2 of microns. **Figure 14 (b)** is a surface image of the same sample. For the development of this IEL, different materials were tested and one was downselected based on the ease of deposition and the ability to achieve a wide combination of haze/scattering and transmission levels. Haze levels of up to 100% were demonstrated for high transmission levels. **Figure 15** shows the variation of haze with wavelength for one particular sample.

Figure 15: Haze dependence on the light wavelength for an embedded oxide particle IEL substrate.

The initial samples produced by using a lab setup had high levels of surface roughness and waviness of the glass surface. AFM measurements yielded the RMS roughness of the samples in the range of 100-200nm.

Additional work was done to understand the defects in the nanoparticle samples using energy-dispersive X-ray (EDX) spectroscopy. **Figure 16** shows SEM images of the surface of one IEL sample containing some defects. Two sources of contamination were determined through SEM imaging & elemental mapping: nanoparticle agglomerates and alumina fibers. Nanoparticle agglomerates were a result of the process itself, with the furnace insulation being a source of alumina fibers. The nanoparticle agglomerates are a few microns in diameter whereas the alumina fibers are around a few hundred microns in length.

(a)

(b)

Figure 16: Plan view SEM images of the surface of an embedded oxide particle IEL sample showing the surface defects due to (a) nanoparticle agglomerates and (b) alumina fibers from furnace insulation.

Initial attempts to fabricated devices had low yields because of these defects. Also, there were dead areas observed in some of the devices as shown in **Figure 17**.

Figure 17: An image of a powered green PHOLED device on an embedded particle IEL sample showing dead areas.

Based on these results, significant efforts were made to decrease the defect density and surface quality. Thicker glass substrates were used for subsequent deposition in order to reduce surface waviness. However, that added a time consuming grinding and polishing step. Modifications were made to the exhaust system of the nanoparticle coating unit. This largely eliminated the particle agglomerates and also prevented the furnace lining material from being deposited on the glass surface. The samples were analyzed for particle distribution, defect size and density. It was determined that the nanoparticle distribution remained the same. **Figure 18** shows the distribution of particles (size greater than $0.36\mu\text{m}$) on the samples fabricated under similar process conditions before and after the modifications. No agglomerates larger than 1 micron were observed.

Figure 18: Distribution of particles (with $D_{\text{circ}} > 0.36\mu\text{m}$) for samples fabricated under similar conditions before and after the coater modifications.

Figure 19 shows a representative image for defect density comparison for before and after samples. The defect density showed dramatic decrease from ~49 to ~2 features in the same area.

Figure 19: Defect density in sample (a) before and (b) after coater modifications. For this comparison a defect was defined as a particle having a $D_{\text{circ}} > 1 \mu\text{m}$. The above images were taken at 1000x.

As mentioned earlier, another source of defects was the alumina fibers from the furnace insulation. Different tubing materials were evaluated in an effort to improve the defect densities. **Table 14** shows the comparison between the two samples made before and after tubing material change.

Sample	# fibers/cm ²
Before	4.9 ± 3.02
After	11.6 ± 1.91

Table 14: Defect density (from alumina fibers) comparison on the samples before and after coater modifications. Images were taken at 40X to determine the fiber density and 9cm² areas were examined. SEM was used as a cross-check.

Green PHOLED devices were constructed on the improved substrates. **Table 15** shows extraction enhancement results for IELs having various combinations of haze and transmission levels. Extraction factors of around 1.24x were achieved for sample having a haze and transmission levels of around 60%.

Sample	Transmission (%)	Haze (%)	Extraction Factor
I	36.8	100	1.05x
II	60	87	1.13x
III	63.5	62.7	1.24x

Table 15: Extraction factors for various embedded particle IEL samples having different haze and transmission levels. These were determined by fabricating and characterizing large area green PHOLED samples.

Subsequently, 6"x6" white PHOLED panels were fabricated. **Figure 20** is a photograph of the lit 6"x6" white PHOLED devices fabricated on the control and IEL plates. It can be clearly seen that the device fabricated on the IEL plate is much brighter, even around the edges.

Figure 20: Image of the lit white PHOLED 6''x6'' devices on control and embedded oxide particle IEL plates.

Figure 21: Electroluminescence spectra for white PHOLED devices fabricated on a control and embedded particle IEL substrate.

Figure 21 shows the EL spectra for a white PHOLED device fabricated on a control and embedded particle IEL substrate. It can clearly be seen that the enhancement is uniform across all wavelengths.

Tables 16(a) & (b) summarize the white PHOLED device data for two IEL plates. Extraction factors of 1.26x and 1.31x were achieved for these IEL substrates. Additionally, measurements were performed using UDC's 12mm thick acrylic block as an external extraction layer in combination with IEL plate that increased the extraction factor from 1.31x to 1.73x.

Sample Type	Transmission (%)	Haze (%)	Enhancement
Sample I	69.3	33.2	1.26x
Sample II	73.3	29.5	1.31x

(a)

Substrate	At J=2mA/cm ²							
	Voltage (V)	PE (lm/W)	1931 x	1931 y	CCT [K]	Δuv	Extraction Factor	Extraction Factor with 12mm acrylic block
Control	4.12	36.8	0.41	0.436	3722	0.02	NA	1.45x
IEL	3.98	48.3	0.399	0.43	3920	0.02	1.31x	1.73x

(b)

Table 16: (a) Enhancement factors measured using an integrating sphere for white PHOLED devices fabricated on two different embedded particle IEL substrates; (b) IEL substrates

Extraction factors of 1.31x were achieved for embedded particle IEL substrates, but it should be mentioned that not all combinations of transmission and haze substrates could be tested. In other words, the embedded particle IEL was not optimized because of the high roughness and defect densities present in the embedded IEL substrates, which resulted in low device yields. Towards the conclusion of the program, alternate strategies were identified to address this problem. Preliminary tests were done using two different kind of coatings (which were solution-deposited but could also be deposited using a large area aerosol spray method). A 100 nm coating having a refractive index of 1.95 resulted in RMS roughness improvement from 65nm to 26nm. A 1 micron coating having a refractive index of 1.55 resulted in RMS roughness improvement from 65nm to less than 1nm as illustrated in **Figure 22**.

Figure 22: Plan view SEM images of the embedded oxide particle IEL substrate as fabricated, after deposition of a 100nm coating (n=1.95) and after deposition of a 1 micron coating (n=1.55).

Due to time and budget constraints, these devices could not be fabricated for extraction enhancement factor calculations.

Sol-gel IEL:

Sol-gel deposition technology was investigated as an alternative means of be used underneath the anode as an internal light extraction feature (**Figure 23**).

Figure 23: A schematic of a sol-gel based scattering layer IEL.

As with the embedded particle IEL, the sol-gel IEL layer provides the opportunity to produce a layer of scattering particles by which the light extraction enhancement can be controlled by choice of particulate material and by the size distribution, refractive index, and absorption coefficient of the particulates. Note the similarity between **Figures 14 (a)** and **23**; both illustrate one concept of scattering particles located near the surface of the substrate.

Previous work had demonstrated that defect free and smooth films could be obtained using a sol-gel approach which is an advantage over embedded particle IEL. Some of the other advantages include no surface waviness (due to its not being a high temperature process), better control of agglomerates, and a wide range of haze and transmission values, as shown in **Figure 24**.

To demonstrate the feasibility of this approach, a number of commercially available materials were assessed as potential scattering particles, and the material that provided the smoothest coating surface when dispersed in a commercially available sol-gel coating was chosen for a device fabrication trial. A series of sol-gel coatings spanning the 10% to 40% haze region (**Figure 24**) were produced.

Figure 24: Typical Haze and Transmission values for sol-gel IEL coating layers.

Samples of the sol-gel IEL coating were subjected to the thermal durability screening test (which replicated the thermal cycling of the substrates in an OLED device fabrication) and passed with no change

in optics, adhesion, or hardness. A more comprehensive durability test matrix was constructed based on detailed fabrication process information provided by UDC, and augmented with some related coating tests that may provide more rapid screening results for these types of processes.

By selecting an alternate scattering material and making minor modification to the dispersion and spin-coat application processes, good quality coatings with over 80% haze were obtained. A second set of sample substrates with sol-gel scattering IEL layers exhibiting 40% to 60% haze were prepared for device fabrication. The samples are in process at the time of this report preparation. Samples from this set were also provided for the qualitative extraction efficiency comparison (section below), with very encouraging results.

Figure 25: Comparison of two sol-gel IEL coatings on PPG Solarphire® PV float glass, at approximately 10% and 80% haze. The samples are spin coat-applied 6''x6'' substrates.

The sol-gel IEL coatings were applied by spin coating to 2 mm thick PPG Solarphire® PV float glass samples ranging in size from 2'' x 2'' for initial screening experiments to 6'' x 6'' for test device fabrication. Scale up to commercial manufacturing production is anticipated to be achievable by scale up of the spin coating technique for sample sizes having a diagonal of approximately 20''. Beyond that, commercial slot/die, curtain, meniscus, and spray technology should allow for the coating of any desired part size by a rapid continuous process.

Finally, a number of other approaches were evaluated. These involved alternative host matrices, such as in-house sol-gel matrices and other commercially-available coating resins, for the scattering particles. These matrices yielded cured coatings on sample substrate which conformed to the pattern illustrated in **Figure 24** and passed the critical durability tests for device fabrication processes, thus illustrating the potential generality of this approach.

Qualitative Extraction Efficiency Comparison:

Since the device yield on embedded particle IEL substrates was low, there was a need to measure the extraction enhancement factors, without fabricating the actual devices, so that the appropriate combination of optical properties could be determined and efforts could be focused on fabricating defect-free IEL substrates. Offline extraction enhancement measurements were made for various IEL samples

(embedded oxide particle and sol-gel having varying haze and transmission values). This work was done in partnership with Professor Franky So (University of Florida). The IEL devices were coated with ITO and subsequently an OLED active layer stack was deposited on it as shown in **Figure 26**.

Figure 26: Schematic of the OLED device deposited on top of IEL substrates for offline extraction factor measurements.

A UV lamp was used to activate the OLED layers and luminance at various angles was measured using a spectrometer. A schematic of the measurement equipment is shown in **Figure 27**.

Figure 27: A schematic of the instrument used to measure the extraction factor for IEL substrates.

Figure 28 shows the variation of the measured extraction factors versus angle for a sol-gel (with transmission~80% and haze~42%) and embedded oxide particle (transmission~80% and haze~50%) IEL substrates. The extraction factors were around 2.0x for angles>20 degrees. No measurements were made between 0 and 20 degrees.

Figure 28: Extraction factor versus angle measured for sol-gel and embedded oxide particle IEL substrates using the tool

These offline measurements offer evidence that further improvements in light enhancement factors can be achieved using the embedded particle and sol-gel IEL approaches.

Milestones & Success Criteria:

The status of milestones and success criteria, as decided in the Project Management Plan (PMP), is highlighted in the tables below.

Milestones	Success Criteria	Start/End	Milestone Status
PMP	Submission/Acceptance	4/10-6/10	•Completed
Fabrication & Characterization of Anodes	Achieve S.R.~ 10-80 $\Omega/\square$; Transmission $\geq$ 75%; Roughness 10-40nm; Work Function~ 3-5 eV	7/10-7/10	•Completed
OLED Device Fabrication, Testing & Evaluation	•Devices Tested •Properties and stability of anodes characterized	8/10-8/10	•Completed
Identification of CFG & anode layer issues	•Root cause of potential anode issues identified •Comparative data generated •Potential need for Na barrier assessed •Down-selected at least two anodes for further development	8/10-10/10	•Completed •Completed •Completed

(a)

Milestones	Success Criteria	Start/End	Milestone Status
Optimize anode layer properties	Achieve S.R. ~ 10-20 $\Omega/\square$; Transmission $\geq 80\%$; Roughness ~ 2nm	10/10-9/11	•Completed
Low-cost deposition process development	•Completion of investigation of alternative low-cost large area manufacturing techniques •Achieve anode metrics using the above techniques	2/11-11/11	•Completed
OLED device fabrication & characterization	•Devices Tested •Properties and stability of anodes characterized	Various (Iterative Task)	•Completed
Cost-Benefit Analysis	•Completion of cost-benefit analysis for anode materials & deposition techniques	8/11-10/11	•Completed

(b)

Milestones	Success Criteria	Start/End	Milestone Status
EEL development	•Fabrication & characterization of sol-gel layer & surface texturing EELs •Achieve EEL with light extraction efficiency of 1.25x by 1/11 •Achieve light extraction efficiency of 1.5x by 6/11	8/10-6/11	•Completed •Demonstrated 1.27x on 2.0mm substrate with the potential of 1.4x On 1.0mm substrate
IEL development	•Fabrication & characterization of AR & nanoparticle IELs •IEL with light extraction efficiency of 1.25x by 6/11 •IEL with light extraction efficiency by 1.5x by 11/11	1/11-5/12	•Completed •Demonstrated 1.31x on 2.0mm substrate with embedded IEL •Offline results show >2.0x enhancement achievable
OLED device fabrication & characterization	Devices Tested & extraction efficiency determined	Various (Iterative Task)	•Completed
Cost-Benefit Analysis	Completion of cost-benefit analysis for out-coupling layers	4/11-5/12	•Completed

(c)

Milestones	Success Criteria	Start/End	Milestone Status
Integrated CFG substrate fabrication trials	Fabrication of optimized integrated float glass substrate which has the potential to meet the DOE cost targets	10/11-6/12	•Completed •Device yield low to draw firm conclusions
Subscale SSL demonstration device fabrication & testing	<ul style="list-style-type: none"> •Fabrication & characterization of five 2mmX2mm white PHOLED pixels which meet the DOE performance targets and delivery thereof •Fabrication of 6"x6" white PHOLED on integrated float glass substrate and conversion into a high-luminance output lighting panel 	12/11-8/12	•Completed •Produced 6"x6" white PHOLED panels on IEL substrates demonstrating 48.3 lu/W efficiency
Submission of manufacturing roadmap plan	Completion & submission of manufacturing roadmap plan outlining steps to meet DOE manufacturing cost goals	1/12-4/12	•Completed •Completed/Submitted with final report

(d)

Table 17: Table listing the various milestones, success criteria, timelines and current status for various subtasks associated with (a) Program Management (Task 1) and Benchmarking (Task 2), (b) Anode Development (Task 3), (c) LEL Development (Task 4), and (d) Prototype Device Fabrication (Task 5) and Manufacturing Roadmap Plan (Task 6).

Deliverables:

The status of deliverables is highlighted in the **Table 18** below.

Task	Deliverable	Status
1.0 Program Management Plan	Project Management Plan Document	Complete
2a. Benchmarking	Report of results of Subtasks 2.1 through 2.3	Complete
2b. Benchmarking	Updated timelines of proposed path forward	Complete
3.0 Anode Development	Report of results of Subtasks 3.1 through 3.5	Complete
4.0 LEL Development	Report of results of Subtasks 4.1 through 4.4	Complete
5.0 Prototype Device Fabrication	6"x6" white OLED panels	Complete
6.0 Manufacturing Roadmap Plan	Manufacturing Roadmap Plan	Complete

Table 18: Table listing the deliverables associated with various project tasks.

Manufacturing Road Map:

PPG has aggressively pursued the development of an integrated substrate, including the anode and external and internal extraction layers, under DOE Award DE-EE0003209. PPG believes that savings from in-line high volume processing of float glass substrates can effectively lower the cost of manufacturing of OLEDs for lighting. Savings over existing glass substrates can come from replacement of expensive borosilicate glass, high volume manufacture of anode, light extraction layers on larger substrates, and improving efficiency of manufacturing processes. In this section, manufacturing strategies

are highlighted that are based on the learning from the work done under the integrated substrate development project and have the potential to achieve the cost metrics and performance targets highlighted in the 2009 multi-year program plan (MYPP). The objective of this section is to outline strategies to achieve DoE's integrated substrate cost targets of \$26/m² by 2015.

Background:

White OLEDs (WOLEDs) are considered a potential high efficiency, low cost, solid state replacement for general lighting. While OLED lighting panels have been available since 2009, commercial offerings have been limited to expensive luminaries for decorative applications and prototyping panel kits. Widespread adoption of OLED-based solid-state lighting sources is constrained by the current high cost of these devices.

Figure 29: A schematic of various components of an OLED device

Table 19 shows the projected costs (from the MYPP developed at the DOE SSL Manufacturing Workshop, Vancouver, WA in June 2009) of sheet-processed OLED lighting panels. Highlighted in the table are the projected costs for the integrated substrate (glass plus electrodes and light extraction; often referred to as the foundation structure and defined in **Figure 29**) on which organic emitting layers of an OLED device are fabricated. Target integrated substrate projected 2014 costs (sum of the substrate, electrode, and light extraction layers costs) at \$26/m² represents 26% of the projected total OLED lighting sheet processed panel cost.

Rigid Sheet SSL Cost Targets							
Stage/Year	Units	2011	2012-13	2014			
Depreciation	\$/m ²	520	200	40			
Labor	\$/m ²	305	45	5			
Other Operations	\$/m ²	70	20	4			
Organic Materials	\$/m ²	30	15	10			
Substrate	\$/m ²	6	6	6			
Electrodes	\$/m ²	20	15	10			
Light Extraction	\$/m ²	20	15	10			
Encapsulation	\$/m ²	10	8	5			
Other Materials	\$/m ²	20	15	10			
Total Direct Costs	\$/m ²	1000	340	100			
Total Direct Costs	\$/klm	330	57	10			

Integrated Substrate Cost Targets			
2011	2012/3	2014/5	
\$46	\$36	\$26	

Table 19: Projected Costs of Sheet Processed OLED Panels as noted in DoE's 2009 SSL MYPP.

Several reasons for the high cost of these integrated substrates are:

- The high cost of borosilicate or double side polished float glass. Borosilicate glass and double side polished glass cost around \$20/m² and \$10/m² respectively. Other substrates based on plastics present major challenges, especially thermal expansion, stability, temperature limits for processing.
- ITO is used as the conventional anode in these substrates and costs roughly \$20/m² before patterning. Although, high indium costs have been highlighted to justify these costs, the real issue is the anode manufacturing process; estimated Indium materials cost \$0.2/m² in sheets sold at \$20/m². Also, fear over scarcity/availability and supply interruptions causes price increases and fluctuations.
- The exact costs of the extraction layers are not known but DoE estimates upwards from \$20/m². Light extraction costs are anticipated to increase as new light extraction techniques, especially for IELs, are developed to meet future MYPP high performance goals.

It is of interest to note that the cost targets were revised in 2011. **Figure 30** shows the current targets for material cost reductions to make this technology commercially viable and competitive with other lighting alternatives. Over 40% of the material cost of the OLED devices is attributed to the dressed substrate component. The target is to drive these costs down from the current \$60/m² to \$52/m² by 2015 and \$25/m² by 2020, while achieving significant improvement in the metrics such as light extraction enhancement. The main difference between the 2009 and 2011 MYPP 2015 cost target arises from the extraction layer component. The 2015 cost target was \$52/m² in 2009 MYPP as opposed to \$26/m² in the 2011 MYPP. This difference, in part, is driven by the change in the performance metric requirement for the extraction layers- 3.5x light extraction enhancement as opposed to 2.0x enhancement targeted in 2009 MYPP.

Figure 30: DOE targets for material costs for OLED lighting panels. (Source: 2011 SSL R&D Manufacturing Roadmap)

Strategies to Mitigate Substrate Costs

PPG has been developing a large-area and low-cost integrated substrate for OLED lighting under this program. This development work was done on pilot-scale manufacturing equipment and technologies that were down-selected and developed to be compatible with established float glass manufacturing technology. PPG believes that by combining these technologies with its existing infrastructure of large-area float glass, online vapor and aerosol spray CVD, and architectural glass large-area vacuum physical vapor deposition (PVD), for manufacture of integrated substrates, the costs could be decreased drastically. Substantial experience has been gained through interactions with lab-scale and commercial OLED device manufacturers that gave important feedback to ascertain the product specifications. Some of the key learnings are highlighted below that could help achieve the short and long-term DoE MYPP cost targets.

- **Substrate:** Significant opportunity for glass cost reduction exists in switching from display glass (\$20/m²) to unpolished float glass substrates (\$6-7/m²). Under this program, it has been demonstrated that through proper selection and pre-treatments, float glass can be utilized as a substrate for OLED lighting applications. The alkali barrier layers may also not be required. PPG operates 4 float glass lines in North America and produces a number of commercial coated glass products. PPG has a deep understanding of the float glass substrate material costs and additional pre-treatment costs and believes that manufacture a quality large-area substrate for OLED lighting applications that meets the cost targets of less than \$7/m² (by 2015) is certainly possible. There may be additional glass cutting costs involved depending on the substrate size requirement of the OLED industry.
- **Anode:** Low resistance and high transmittance ITO coatings are commonly achieved by high deposition temperature processes at about 300°C. The cost and technical challenges for an anode vacuum deposition manufacturing process is to develop an in-line process with the ability to heat glass quickly in vacuum and maintain uniform heating during transparent conductive oxide (TCO) deposition while being transported at high line speed, or to develop a TCO that can be deposited without the need to heat glass during transport in vacuum. Otherwise, post processing will be required to optimize the sheet resistance and optical transmission, increasing process complexity and cost. For all these approaches, TCO coatings need to be stable during OLED fabrication processing. Under this program, PPG has developed two anodes (based on a room-temperature MSVD and online CVD process) that match the performance of the conventional ITO electrode, and which are low-cost alternatives. Since both of the anode technologies

highlighted in **Table 20** are promising, PPG believes that either of these technologies could be used for SSL OLED lighting and meet the current cost targets.

Metric	MSVD Anode	CVD Anode
Sheet Resistance (<20 Ω/□)	~5 Ω/□	~10 Ω/□
Visible Transmission (>85%)	>85%	~80%
Work Function (>5eV)	5.33eV	4.96eV
Surface Roughness (R_{rms} <2nm, R_{pv} <20nm)	R_{rms} ~0.5nm, R_{pv} ~10nm	R_{rms} ~8nm, R_{pv} ~30nm
Reliance on Scarce Materials	Small to None	None
Low-Cost Processing	Yes	Yes

Table 20: Comparison of down-selected MSVD and CVD anode metrics.

The CVD anode (Anode 1) is potentially the cheaper of two approaches but may further development to assess the requirement of a solution deposited HIL to address the roughness issues. The MSVD anode is a inexpensive short term alternative to the conventional ITO anode.

- Light Extraction Layers:** A critical component of the integrated substrate is the light extraction layer(s). This component has the potential to impact all four OLED lighting market drivers namely, cost, lifetime, efficiency and brightness. There are numerous examples of technologies that address the substrate, anode, and surface plasmon modes such as micro lenses. However, these technologies suffer from issues such as scalability, durability/compatibility with OLED device manufacturing and costs. Under this program, PPG investigated an in-glass embedded oxide particle approach for light extraction. This approach had no durability or device compatibility issues since the IEL is essentially fused into the glass surface. Offline measurements indicate a potential to achieve over 2.0x enhancement factors. While, for this effort, offline deposition equipment was utilized, PPG's previous experience suggests that this approach can be integrated with float glass manufacturing and is also scalable. Additionally, roughness and substrate waviness issues (as discussed previously in this report) would be mitigated by integrating this technology with the float glass manufacturing process. This approach would require equipment and online process development but has an advantage because it utilizes the existing base of aerosol spray equipment manufacturers. PPG believes that this technology can be integrated with large-area float-glass manufacturing process and total average cycle time (TACT) could be achieved to meet the IEL cost targets. Additionally, this IEL technology may be combined with an appropriate EEL to achieve even better extraction enhancement numbers. PPG has demonstrated two scalable methods to produce EELs which can be integrated with float-glass manufacturing.
- Large-Area Process Development:** In order to ascertain the true manufacturing costs there is a need to develop and demonstrate a large-scale, high-throughput, continuous manufacturing process for all components of integrated substrate. PPG has significant experience in large-area coated glass products and believes that manufacture of large-area substrates could significantly reduce the costs.

For the manufacturing process, real time inspection and process control systems also need to be developed to ensure the quality and uniformity of the large-area substrates. This is essential for lowering the costs associated with excess off-specification production. Development of process

controls plus the application of in-line testing and inspection will allow PPG to produce an integrated substrate which is within tight performance specifications and low in defects. This will in turn allow OLED panel manufacturers to attain higher device yields and avoid the need for binning, indirectly contributing to overall cost reduction.

- **Collaboration with Device Manufacturers:** A critical need for successfully developing a low-cost integrated substrate is to engage with the device manufacturers through all stages of the development process. The need to define product specifications is essential for the OLED substrate. For example, one of the metrics for anode layers is its sheet resistance, which is dependent on the OLED device structure. Earlier engagement with the device manufacturers will enable the earlier definition of the manufacturing steps for the OLED substrate. Another example is the defect size and density requirement in the anode layer. The size requirement is dependent on the thickness of the OLED stack and the defect density will be determined by the pixel area. Feedback from device production runs in the pilot OLED lighting panel manufacturing lines employing float-glass based substrate will help to determine these metrics and lead to an adoption of the right manufacturing tools and technologies for integrated substrate manufacturing.
- **Integrated Substrate Manufacturing Process Flow:** Various process step flows associated with different costs are possible in the manufacturing of an integrated substrate depending on the choice of the production method for the anode coating, either off-line MSVD or on-line CVD. Cost reductions may be realized by reordering the sequence of processing steps upon further manufacturing insight. Examples of the anticipated lowest cost manufacturing process flows are shown in **Figure 31** for high volume production using manufacturing techniques for integrated substrates employing the integration of IEL fabrication with float glass manufacturing process and either MSVD or CVD anode layer.

Figure 31: Two flow diagrams of low-cost manufacturing process using large-area glass manufacturing methods for an integrated substrate based on either a CVD anode (top diagram) or MSVD anode (bottom diagram).

PPG believes that there is disconnect between the current OLED lighting market demand and low-cost float glass manufacturing capabilities- the current OLED substrate demands are relatively low (<1 M sq. ft./year) for the coated glass manufacturing lines which produce a few million square feet of glass per week. As the demand picks up, the unit cost would decrease because of lower fixed cost per unit. Also, production yields would improve with experience and result in cost reduction.

Summary:

PPG pursued the development of an integrated substrate, including the anode, external, and internal extraction layers, under DOE award DE-EE0003209. The objective of PPG's program was to achieve cost reductions by displacing the existing expensive borosilicate or double-side polished float glass substrates and developing alternative electrodes and scalable light extraction layer technologies through focused and short-term applied research. One of the key highlights of the project was proving the feasibility of using PPG's high transmission Solarphire® float glass as a substrate to consistently achieve OLED devices with good performance and high yields.

Under this program, four low-cost alternatives to the Indium Tin Oxide (ITO) anode were investigated using pilot-scale magnetron sputtered vacuum deposition (MSVD) and chemical vapor deposition (CVD) technologies. The anodes were evaluated by fabricating small and large phosphorescent organic light-emitting diode (PHOLED) devices at Universal Display Corporation (UDC). The device performance and life-times comparable to commercially available ITO anodes were demonstrated. A cost-benefit analysis was performed to down-select two anodes for further low-cost process development.

Additionally, PPG developed and evaluated a number of scalable and compatible internal and external extraction layer concepts such as scattering layers on the outside of the glass substrate or between the transparent anode and the glass interface. In one external extraction layer (EEL) approach, sol-gel sprayed pyrolytic coatings were deposited using lab scale equipment by hand or automated spraying of sol-gel solutions on hot glass, followed by optimizing of scattering with minimal absorption. In another EEL approach, PPG tested large-area glass texturing by scratching a glass surface with an abrasive roller and acid etching. Efficacy enhancements of 1.27x were demonstrated using white PHOLED devices for 2.0mm substrates which are at par with the standard diffuser sheets used by OLED manufacturers.

For an internal extraction layer (IEL), PPG tested two concepts combining nanoparticles either in a sol-gel coating inserted between the anode and OLED or anode and glass interface, or incorporated into the internal surface of the glass. Efficacy enhancements of 1.31x were demonstrated using white PHOLED devices for the IEL by itself and factors of 1.73x were attained for an IEL in combination of thick acrylic block as an EEL. Recent offline measurements indicate that, with further optimization, factors over 2.0x could be achieved through an IEL alone.

Patents: No patent applications have been filed to date. Two are expected to be filed in early 2013.

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

- Peer Review, Westlake-TX, January 12, 2011: A. Bhandari, H. Buhay and T. Hart updated on the project progress
- DoE SSL R&D Workshop, San Diego-CA, February 1-3, 2011: A. Bhandari gave an invited talk on "Low-cost Integrated Substrate for OLED lighting" and presented a poster
- Budget Period I Review, Cheswick- PA, March 16, 2011: Budget period I review with the Department of Energy was conducted at PPG
- DoE SSL Manufacturing R&D Workshop, Boston- MA, April 12-13, 2011: H. Buhay co-chaired an OLED track session and presented on "OLED Materials Manufacturing Issues and Priorities: Review of OLED Roundtable Meeting"
- DoE SSL R&D Workshop, Atlanta- GA, January 31- February 2, 2012: A. Bhandari presented a poster on the project progress

- DoE SSL Manufacturing R&D Workshop, San Jose-CA, June 13-14: D.J. O'Shaughnessy co-chaired an OLED track session and presented on "Low Cost Materials Manufacturing"
- DoE SSL R&D Workshop, Long Beach-CA, January 29-31: PPG has been selected to be recognized for contributions to the SSL lighting and is invited to give a presentation