Final Report

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Solid-State Lighting Project Number: DE-EE0007076

Integrated Plastic Substrates for OLED Lighting

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Executive Summary

OLED lighting has immense potential as aesthetically pleasing, energy-efficient general illumination. Unlike other light sources, such as incandescents, fluorescents, and inorganic LEDs, OLEDs naturally emit over a large-area surface. They are glare free, do not need to be shaded, and are cool to the touch, requiring no heatsink. The best efficiencies and lifetimes reported are on par with or better than current forms of illumination. However, the cost for OLED lighting remains high – so much so that these products are not market competitive and there is very low consumer demand. We believe that flexible, plastic-based devices will highlight the advantages of aesthetically-pleasing OLED lighting systems while paving the way for lowering both materials and manufacturing costs. These flexible devices require new development in substrate and support technology, which was the focus of the work reported here.

The project team, led by Sinovia Technologies, has developed integrated plastic substrates to serve as supports for flexible OLED lighting. The substrates created in this project would enable large-area, flexible devices and are specified to perform three functions. They include a barrier to protect the OLED from moisture and oxygen-related degradation, a smooth, highly conductive transparent electrode to enable large-area device operation, and a light scattering layer to improve emission efficiency.

Through the course of this project, integrated substrates were fabricated, characterized, evaluated for manufacturing feasibility and cost, and used in white OLED demonstrations to test their impact on flexible OLED lighting. Our integrated substrates meet or exceed the DOE specifications for barrier performance in water vapor and oxygen transport rates, as well as the transparency and conductivity of the anode film. We find that these integrated substrates can be manufactured in a completely roll-to-roll, high throughput process and have developed and demonstrated manufacturing methods that can produce thousands of feet of material without defects. We have evaluated the materials and manufacturing costs of these films at scale and find that they meet the current and future cost targets for bringing down the cost of OLED lighting while enabling future roll-to-roll manufacturing of the complete device. And finally, we have demonstrated that the inherent light-scattering properties of our films enhance white OLED emission efficiency from 20% to 50% depending on the metric. This work has shown that these substrates can be created, manufactured, and will perform as needed to enable flexible OLED lighting to enter the marketplace.

Milestones and Accomplishments

Table 1 shows our milestone chart for this project. All proposed milestones were accomplished through the course of the project period.

Table 1. Project milestones and accomplishment verification.

Milestone	Description	Verification / Data
1	Small-scale, 5" – 8" wide, integrated substrate that has sheet resistance of 2 - 9 ohm/sq. and meets DOE barrier film requirements.	Integrated substrates of this dimension have been fabricated and tested with sheet resistances under 9 ohms/sq and a WVTR of 2.88×10^{-6} g/m^2*day at 23°C and 50% relative humidity. Integration of electrode does not affect barrier properties.
2	Complete substrate haze vs. nanowire diameter study for substrates with 1 ohm/sq. resistivity and OLED-compatible surface roughness.	Transmission >60% with haze >30% at a sheet resistance of 3.5 ohms/sq, with average roughness <3 nm and RMS roughness <5 nm using 200-nm diameter nanowires.
3	Optimize substrate haze for OLED efficiency.	OLEDs have been successfully fabricated using 100-nm and 200-nm nanowire electrodes. Devices made on integrated substrates show a 1.2 – 1.5X increase in current, luminous, and external quantum efficiencies compared to ITO / glass controls. Lifetimes, at 12,000 hours, are 80% of ITO / glass controls.
4	Proof of ability to manufacture integrated substrate at or under DOE cost target of \$95/m^2.	Barrier film is already being produced at scale in single pass through Aegis machine. Using current scale materials costs and 60% yield, total assembled integrated substrate cost is \$86/m^2. With higher yield and anticipated material cost reductions, we anticipate costs reducing to \$43 - \$48/m^2 in the future.
5	Roll-to-roll produced 10" wide haze-optimized integrated substrates with uniformity deviations under 5%.	12"-wide roll-to-roll integrated substrates produced in-house with less than 5% variation in sheet resistance and transmission. Visual non-uniformities were present but did not affect small-scale OLED emission pattern. Metering roll coating produced over 1000 linear feet of material at 12" wide without any visible defects from nanowire aggregation.
6	150-mm by 150-mm square white OLEDs on integrated substrates that show lifetime comparable to OLEDs fabricated using ITO on glass (>10,000 hours). Improved white OLED efficiency, out-coupling (1.5 – 2X), and angular color stability.	Due to availability, we fabricated 222.25 mm by 47.6 mm white OLEDs. This rectangular shape allowed us to assess the effectiveness of our low-resistance anode. These devices showed performance equal to the small-scale devices, with improved performance over the ITO control. They had 89% emission uniformity.

These milestones served as a guide for the overall project direction, with the overarching aims of first creating and characterizing our substrate, then developing and proving our manufacturing methods, and finally demonstrating its usefulness in flexible OLEDs.

Milestone 1: Small-Scale Integration

Figure 1 shows the layered structure of our integrated substrate. It is based on the combination of the Vitriflex "triad" barrier film layers and Sinovia's composite transparent conductive film. On top of the sputtered reactive and diffusive layers, Vitriflex uses a top seal polymer to enhance the barrier's oxygen and moisture blocking properties as well as to enhance the barrier's mechanical stability. Sinovia's conductive films are comprised of a two-dimensional network of silver nanowires embedded into the surface of a polymer. Through the course of our work, we found that the Vitriflex barrier polymer can be used as the layer into which the silver nanowires could be embedded. This creates the most efficient integrated structure for manufacturing as well as performance.

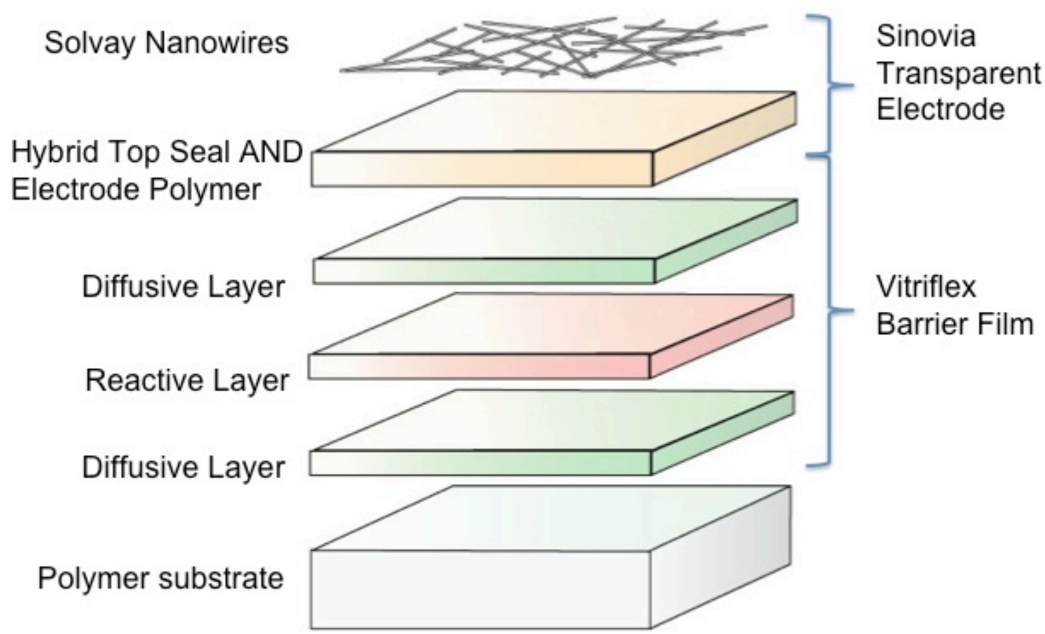


Figure 1. Integrated substrate structure.

While doing the initial integration of these two thin-film technologies, the first goals were to preserve the performance of the individual components when they are integrated and to ensure that the integration process did not damage either the barrier or the conductive film. While we found that the transparent conductive film was not affected at all by being deposited onto the barrier and that our coating process wasn't damaging the barrier, the water vapor transport rate (WVTR) performance of the integrated substrate depended strongly on the thickness of the polymer layer between the sputtered Vitriflex barrier and the embedded nanowires. If the polymer layer was too thick, moisture could ingress from the sides and increase the water vapor transport rate. Table 2 shows initial experiments in small-scale integration with different polymer thicknesses and how they affect WVTR.

Table 2. Water vapor transport rate dependence on polymer thickness.

Electrode	**WVTR**	
	mg/m^2*day @40°C/100%RH	g/m^2*day @23°C/50%RH
Control	2.06	9.89x10^{-05}
P1/P2/AgNW	12.97	6.23x10^{-04}
Control	2.1	1.01x10^{-04}
P1(thick)/AgNW	6.6	3.17x10^{-04}
Control	0.24	1.15x10^{-05}
P1(thin)/AgNW	0.28	1.34x10^{-05}

The target for this work was to produce an integrated substrate with WVTR in the 10^{-6} g/m^2*day order of magnitude to extend the OLED lifetime. In our initial testing of three-layer sputtered barriers, we were able to get close to this value, but did not attain it, so we experimented with adding sputtered layers to the barrier to improve performance. We added one additional reactive and diffusive layer each to create five-layer barrier films and compared them to the three-layer films that we were using previously. Figure 2 shows a schematic of the five-layer structure.

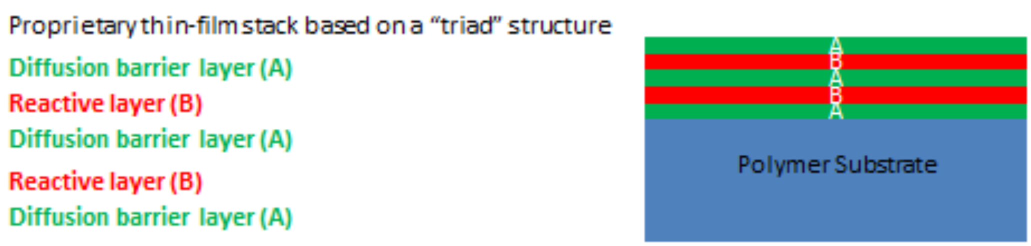


Figure 2. Five-layer barrier structure with diffusive and reactive layers.

Through additional testing, we found that while we were able to reach the 10^{-6} target using the three-layer barrier, we were able to attain even lower WVTR values and more reliably reach 10^{-6} g/m^2*day using the five-layer films. Most of our three-layer tests reached the values in the low 10^{-5} order of magnitude. We suspect that as Vitriflex improves their process, the three-layer films will become more reliable, as those films will be advantageous for their lower cost in the future.

All of the results shown in Table 3 had optimized polymer thicknesses. And all of the barrier films tested preserved the sheet resistance and the transparency of the Sinovia composite films before and after WVTR testing, a promising first step toward understanding the long-term stability of our substrates.

Table 3. Integrated substrate WVTR performance reaching 10^{-6} g/m^2*day using three- and five-layer barrier films.

Layers	**WVTR**		**Test Hours**
	mg/m^2*day @40C/100%RH	g/m^2*day @23C/50%RH	
3	0.20	9.61×10^{-6}	92
5	0.11	5.28×10^{-6}	164
5	0.06	2.88×10^{-6}	168

Milestone 2: Haze and Surface Roughness

The next goal was to control the physical and optical properties of the integrated substrates by understanding optical haze and surface roughness. OLED substrates require low surface roughness due to the thin active layers. Sinovia composite transparent conductive films solve this issue by embedding the conductive nanowires into the surface of the underlying polymer such that the roughness in the network is projected away from the OLED layers. Our goal was to understand how nanowire diameter affects substrate haze and consequently light out-coupling from the OLED while maintaining nanometer-smooth conductive surfaces. In our integrated substrate, the nanowires sit at the interface between the high index organic layers and the lower index substrate layers (Figure 3), in the optimal position to disrupt the total internal reflection that normally occurs at this interface. Higher haze (more light scattering) should more greatly improve OLED efficiency.

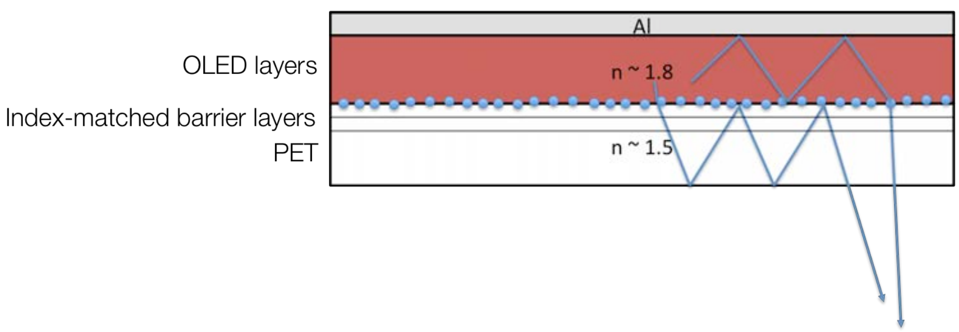


Figure 3. Schematic of how nanowires (blue dots) disrupt total internal reflection at the refractive index interface between OLED and support in our integrated substrates and scatter light that out that would otherwise be trapped.

Solvay USA synthesized nanowires at various diameters so that we could fabricate integrated substrates using them and test the resulting optical properties and surface roughness. Solvay's synthesis process was advantageous for this work as they have very fine control over nanowire diameter via changing physical synthesis parameters such as reaction dwell time and temperature. Figure 4 shows optical microscope images of examples of these nanowires with 45 nm diameters on the left and 200 nm diameters on the right, imaged at the same magnification scale. This represents the lower and upper limits of the diameters that we explored as part of this study, with intermediate diameters including 70 nm, 90 nm, and 100 nm tested as well. Because these nanowires behaved differently in suspension depending on their diameter, Sinovia had to change the coating formulations to ensure optimal nanowire morphology in coated films.

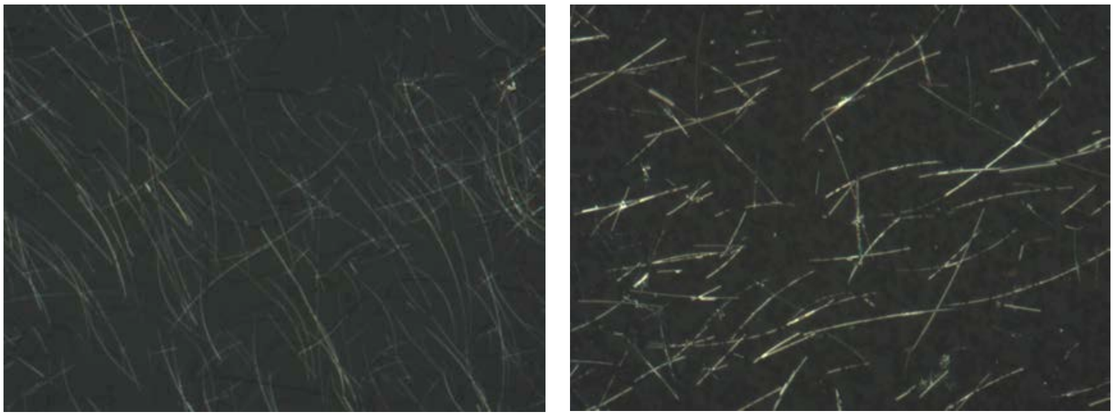


Figure 4. Optical micrographs of silver nanowires synthesized by Solvay's synthesis team with diameters of 45 nm (left) and 200 nm (right) imaged at the same scale.

After formulating the nanowires for coating, the Sinovia team fabricated composite transparent conductive films, measuring optical and electrical properties to understand the dependence on nanowire diameter. Results of this study are found in Table 4, showing total transmission values of the integrated substrates, including surface reflections.

Table 4. Haze values corresponding to nanowire diameter and sheet resistance.

Nanowire Diameter	Sheet Resistance	Transmission	Haze
45 nm	5 ohms/sq	77 %	12 %
90 nm	5 ohms/sq	75 %	20 %
200 nm	3.5 ohms/sq	72 %	34 %
200 nm	1 ohm/sq	69 %	46 %

It is worth noting that there are two factors upon which haze is dependent: nanowire diameter and density, which directly translates to sheet resistance. The more nanowires on the surface, the lower the sheet resistance and the higher the haze, as is evident from the two entries in Table 4 that utilized 200 nm diameter nanowires. But at similar nanowire densities (similar sheet resistance) this study shows that larger diameter nanowires produce more haze. It is also clear from this study that our transparent conductive films on our integrated substrates meet the target values for this project, at 1 – 5 ohms/sq with total transmission over 60% and haze between 12% and 46% (30% haze being our milestone target). All of these transparent conductive films were fabricated using Vitriflex barriers that had measured WVTR values in the 10^{-5} to 10^{-6} g/m^2*day range, demonstrating the full combined performance of these substrates.

We examined the surfaces of our transparent conductive films using both scanning electron microscopy (SEM) and atomic force microscopy (AFM). SEM allowed us to see large areas of our films and understand the morphology of our coated nanowires. SEM micrographs of a typical transparent conductive film surface are shown in Figure 5, with a flat image on the left and an image taken at an oblique angle on the right.

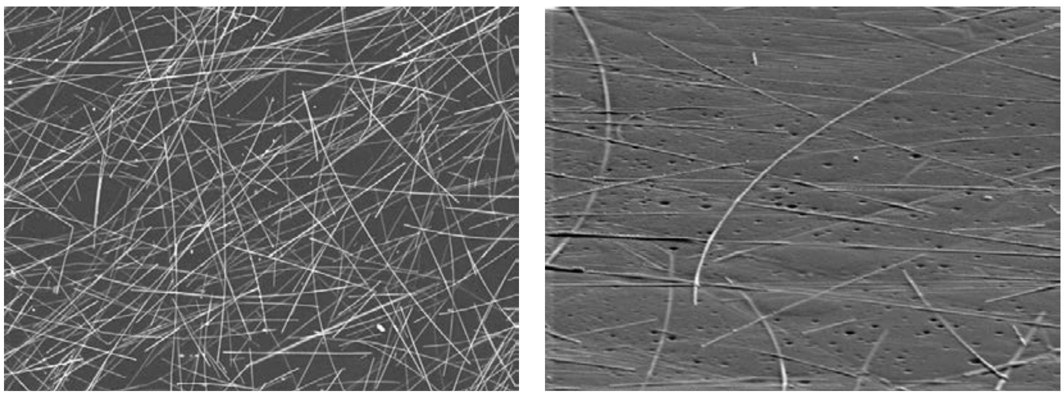
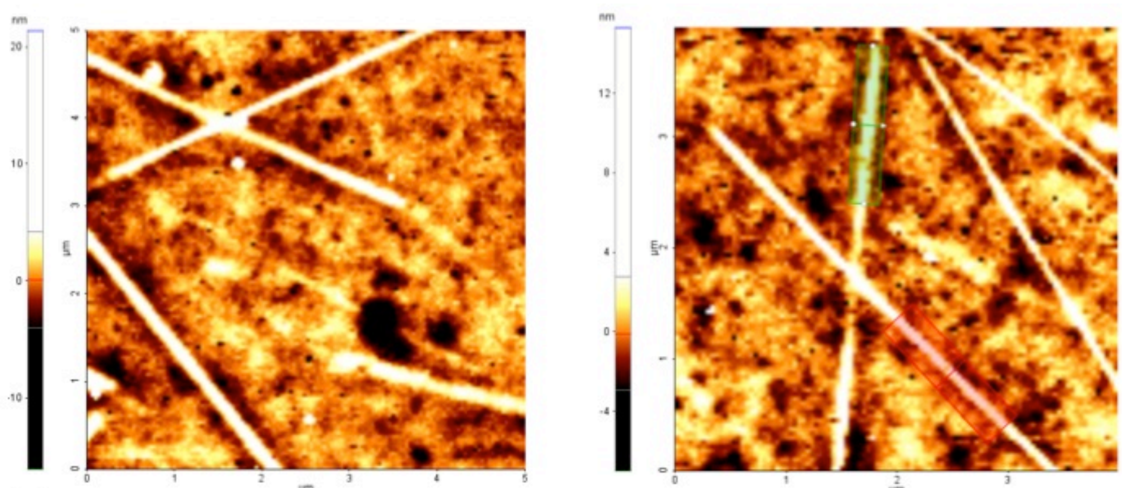


Figure 5. SEM micrographs of integrated substrate transparent composite electrode fabricated with 90-nm diameter nanowires. Left: flat image showing nanowire density and morphology. Right: angled image showing embedding and low surface roughness.

The flat image shows that our nanowires are evenly distributed across the surface of the film in random directions, which is essential for isotropic performance. The nanowires are also straight, as opposed to curled tightly around themselves, which lowers the nanowire percolation threshold and gives us better transparency for a given conductivity value. This optimal morphology is achieved through our nanowire coating formulations. The angled image shows how our nanowires are embedded into the surface of the polymer over their entire lengths, with the polymer filling the gaps between them, reducing surface roughness. But while SEM is good for an overview of these films and for locating any defective areas that may be present, it cannot give us quantitative values for surface roughness.

To get quantitative data on the surface roughness of our integrated substrates, we used AFM. In fact, AFM, as a mechanical method, is the only means of accurately acquiring this data, as optical methods cannot account for the changes in material that are present in this surface composite. We report here two representative data sets to illustrate the ability of our electrode structure to create smooth, OLED-compatible surfaces at sheet resistances under 10 ohms/sq. For electrodes fabricated using 100-nm diameter nanowires, disregarding film defects, the typical peak-valley roughness was around 30 nm, the average roughness was around 4 nm, and the RMS roughness was <10 nm. The nanowires protruded between 1 and 10 nm above the polymer surface. For electrodes fabricated using 200 nm diameter nanowires, the peak-valley roughness was around 30 nm, the average roughness was around 2 nm, and the RMS roughness was <5 nm. The nanowires protruded between 3 and 10 nm above the polymer surface. Thus there was not much difference in the roughness characteristics between 100-nm diameter nanowires and 200- nm diameter nanowires, showing that our electrode structure is effective at yielding a smooth, OLED-compatible surface regardless of nanowire diameter. These metrics exceeded our milestone RMS roughness target of <15 nm.



Left

Region	Min(nm)	Max(nm)	Mid(nm)	Mean(nm)	Rpv(nm)	Rq(nm)	Ra(nm)	Rz(nm)	Rsk	Rku
Whole	-16.101	21.384	2.642	0.077	37.485	2.098	1.396	30.773	-1.454	11.997

Right

Region	Min(nm)	Max(nm)	Mid(nm)	Mean(nm)	Rpv(nm)	Rq(nm)	Ra(nm)	Rz(nm)	Rsk	Rku
Whole	-6.996	15.317	4.160	-0.092	22.313	1.465	1.047	16.490	-1.020	8.146

Line	Min(nm)	Max(nm)	Mid(nm)	Mean(nm)	Rpv(nm)	Rq(nm)	Ra(nm)	Rz(nm)	Rsk	Rku
Red	-1.191	3.143	0.976	0.120	4.334	1.539	1.255	N/A	-1.222	2.720
Green	**-1.571**	**1.826**	**0.128**	**0.158**	**3.397**	**1.183**	**1.014**	**N/A**	**0.011**	**1.544**

Figure 6. AFM topographical data for composite electrode structures fabricated using 200 nm diameter silver nanowires. Region data is shown for both images. In the left image, peak-valley roughness is 37.485 nm, RMS roughness is 2.098 nm, and average roughness is 1.396 nm. In the right image, peak-valley roughness is 22.313 nm, RMS roughness is 1.465 nm, and average roughness is 1.047 nm. Line data is also shown for the right image, which follows the lines across the nanowires shown, and calculates roughness in the defined regions. These measurements identify the height of the nanowires relative to the polymer surface by the peak-valley roughness, which is 4.334 nm in the red area and 3.397 nm in the green area.

Milestone 3: Small-Scale White OLEDs

Once we had OLED-compatible-smooth surfaces on our integrated substrates, the next step was to fabricate OLEDs and compare them to control devices to understand how our integrated substrate performed. We did this in collaboration with OLEDWorks in Rochester, New York. From our very first OLED sample test, the devices fabricated on flexible integrated substrates out-performed the control devices fabricated on rigid glass and ITO in all efficiency metrics. Through the course of multiple tests we saw 1.2X – 1.5X efficiency increases over the ITO/glass control devices and lifetimes of more than 12,000 hours, which is 80% of the standard control device lifetime. Our lifetime measurements were corroborated by two different tests. The first accelerated lifetime test was a fade test, conducted at a high current density of 10 mA/cm^2 and a high initial brightness of approximately 5000 nits. Our extrapolated data from this test yielded a lifetime to 70% brightness (LT70) of 12,000 hours, assuming normal lighting operation at 2500 nits. This gives us an idea of the electrical performance over time and is the limiting factor in our lifetime studies. The second was an accelerated environmental lifetime test at 85°C and 85% relative humidity, which tested the barrier's ability to preserve the OLED. There were no differences in the emission pattern of our devices before and after the lifetime test, indicating that dark spots from moisture damage were not an issue. This means that the barrier properly protected the OLED for its accelerated lifetime of over 500 hours in the environmental chamber, which corresponds to 50,000 hours under normal operating conditions. This is an extremely good result for the barrier. While our overall lifetime exceeded our milestone goal of 10,000-hour LT70, as it is only 80% of the control device lifetime this is an area that we believe we can improve upon in the future. Images of our white OLEDs compared to the ITO/glass controls can be seen in Figure 7.

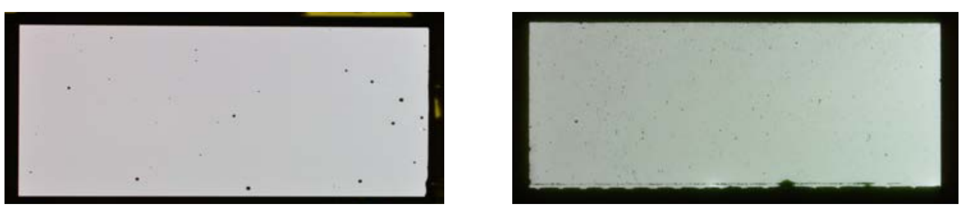


Figure 7. White OLEDs made using: left: ITO/glass control; right: integrated substrate.

Milestones 4 and 5: Scale Manufacturing Cost Targets and Uniformity

Once we had proven that our integrated substrates could be effective in white OLEDs and improve efficiency performance, the next step was to prove that they could be manufactured at scale and at a cost that is reasonable for the market to bear.

Vitriflex barrier films are fabricated in a single-pass sputtering chamber designed for this purpose. And while the performance can have some variation, this process is fairly well developed. The primary manufacturing step that we were investigating is the integration between the barrier, the polymer, and the nanowire conductive surface. Open questions included whether a fully roll-to-roll process would not damage the barrier, whether we could reliably coat the polymer at the thicknesses required to improve barrier performance, and whether our nanowires could be coated over long lengths and times without creating defects from aggregation.

Coating this monomer was a fairly standard process. The material itself was not complex to work with, although the sensitivity of the WVTR performance to the polymer thickness led us to finely tune the coating parameters to obtain uniformly thick films under 4 microns thick. In addition to monomer dilution, which controls viscosity, two coating parameters were important for this step: uniformity and line speed. Our line speed had two possible limiting factors at play: the coating method itself and UV lamp curing power. We ran a speed series to find our maximum speed. It turned out that we were limited by the coating method itself. At speeds over 75 feet per minute the coating cylinder started to spit material out of the pan. On a larger coater with a larger pan, this would not occur at this speed (the maximum speed would be increased) but this is the issue that limits the possible speed of this process. For this process, we were able to hit our topcoat thickness target with very good coating and lamination uniformity. Based on various thickness measurements both down and across the web, our average thickness was 4 microns, exactly within our target range for improving barrier performance. The standard deviation of our measurements was 0.24 microns and the total range was 0.6 microns. Testing WVTR on these films confirmed that this process can produce polymer of the correct thickness to improve barrier properties on the large scale.

As anticipated, coating the nanowires at scale was a far more difficult process. Sinovia, unlike others who work with silver nanowire coatings, works with formulation rather than coating parameters to tune the film's conductivity and transparency. To reach the high conductivity values required by OLED lighting, this meant that we had to use highly concentrated silver suspensions. And this adds complexity to our process and eliminates the possibility of using certain coating methods because silver nanowires are prone to aggregation.

At Carestream Contract Manufacturing's Oregon plant, the Sinovia team worked with their technicians on coating our silver nanowire suspension at scale. We tried a variety of methods to see if we could get a scaled industrial process to work with our materials. After trying various methods, we realized that while bladed coating methods are effective for many materials, they cause more issues than they solve when trying to meter suspensions with high aspect ratio particles. Thus our final successes came using a bladeless coating method that uses the rolls themselves to meter the wet thickness of our suspension.

By adjusting the roll speeds from even with the web speed (1X) to 1.2X, 1.4X, and 1.5X, we were able to create films with very good performance: 8 ohm/sq at 81% transparency with under 5% variation in these characteristics. The films were uniform on the macroscale to the eye without any downweb streaking, and had proper microscale morphology. But the most important result of this work is that we were able to coat thousands of feet of release liner with nanowire suspension without aggregates or other defects forming. This is the indication to us that we will be able to use this manufacturing method to coat our nanowire suspension with high performance at the commercial scale.

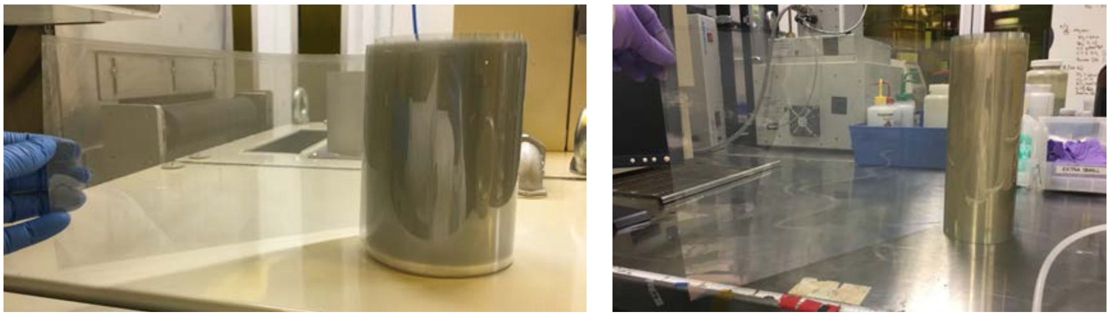


Figure 8. Photographs of 5”-wide and 12”-wide integrated substrates fabricated using only roll-to-roll methods.

Based on our scale coating trials, we worked with Carestream to produce an estimate of manufacturing costs at scale, to ensure that our costs align with market requirements. We assumed at scale that we would use Carestream’s production line at 58” wide rather than the pilot coater at 12” wide. Our model did not take into account any modifications to equipment that might need to occur, only how much it would cost to run the process once set up. However if we were to go into production with Carestream, there would not be significant capital investment required. It also did not take into account slitting and packaging. Our estimate used the same coating speeds that we used on the small coater, translated to scale on the large coater. We modeled the same two-step process developed on the pilot coater and assumed at scale that we are looking at 100,000 – 500,000 meters of material production per year. The main drivers of manufacturing cost are volume and yield. Our first estimate, one for the near future, estimates yield at 60%. Our second, for a more experienced process, estimates yield at 80%. Our third takes into account reduced materials costs in the future, keeping yield at 80%. And our fourth improves yield to 90%. As shown in table 5, even our current low yield gives us a cost under the DOE target of $95/m^2$.

Table 5. Cost estimates for integrated substrate at scale, current and future.

	Cost at 60% yield	Cost at 80% yield	Future at 80% yield	Future at 90% yield
Substrate / Barrier	$58/m^2$	$43.75/m^2$	$37.50/m^2$	$33.30/m^2$
Topcoat	$6.68/m^2$	$5/m^2$	$1.25/m^2$	$1.11/m^2$
Silver Nanowires	$10/m^2$	$7.5/m^2$	$4.50/m^2$	$4/m^2$
Release Film	$4.18/m^2$	$3.13/m^2$	$2.50/m^2$	$2.22/m^2$
Coating / Assembly	$6.50/m^2$	$2.50/m^2$	$2.50/m^2$	$2.50/m^2$
Total Cost	**$85.83/m^2$**	**$61.88/m^2$**	**$48.25/m^2$**	**$43.13/m^2$**

Materials costs for our estimate are based on current pricing for barrier, barrier topcoat, and release film. For future costs, the most significant reduction comes from materials yield. In addition, we assume modest materials cost reductions for the two future projections. We anticipate that the largest cost reduction will come from the topcoat, as this material is also not currently at scale. Production costs for that material will lower dramatically as those economies of scale increase. Thus, we are confident that we can meet DOE cost targets for our process and our materials now and in the future.

Milestone 6: Large-Scale OLED Emission Uniformity

With our scaled integrated substrate we first repeated our small white OLED testing to gauge both performance and uniformity. Figure 9 shows images of white OLEDs fabricated on our roll-to-roll integrated substrate. Emission appeared uniform and performance surpassed that of the glass / ITO control devices in the same manner as in previous testing. The metrics of current efficiency (cd/A), luminance efficiency (lm/W), external quantum efficiency (EQE %), and external quantum efficiency over all angles (Angle EQE %) and their corresponding increase percentages over the control devices are shown in Table 6. As a note, we were comparing our transparent conductive films on our integrated substrates with sheet resistance of 7 ohms/sq and transparency of 83% to ITO with sheet resistance of 15 ohms/sq and transparency of 85%. As the transparency of our film was lower, we concluded that the efficiency increase was due to increased out-coupling, brought about by the haze in our films, allowing light to escape in spite of apparent lower optical transmission.

Figure 9. OLEDs made using integrated substrate fabricated roll-to-roll in-house on 12”- wide web. While subtle visible non-uniformities were present in the nanowire coating, visible variation in resulting emission pattern is not present.

Table 6. Efficiency data for white OLEDs fabricated on roll-to-roll coated 12”-wide integrated substrate, including percentage increases over control device. All devices on integrated substrates out-performed the controls in all efficiency metrics. Each efficiency metric saw maximum increases between 22% and 52%.

Sample	cd/A, % Inc.	lm/W, % Inc.	EQE, % Inc.	Angle EQE, % Inc.
Control	52.7, N/A	24.1, N/A	27.3, N/A	27.8, N/A
Int. 1	72.8, 38%	29.2, 21%	30.7, 12%	32.3, 16%
Int. 2	80.3, 52%	29.7, 23%	32.0, 17%	30.3, 9%
Int. 3	71.5, 36%	29.0, 20%	32.8, 20%	35.3, 27%
Int. 4	71.6, 36%	29.8, 24%	30.6, 12%	32.8, 20%
Int. 5	77.4, 47%	31.2, 29%	33.3, 22%	35.0, 26%

Following the successful roll-to-roll coating of our substrates and successful small-scale OLED testing, our next step was to fabricate large-scale white OLEDs and test them for efficiency and uniformity. When we fabricated our small-scale devices, we found that our device uniformity was improved by adding a PEDOT layer on top of our anode and by eliminating OLEDWorks's short reduction layer. As our goal was to reproduce the promising results of our small-scale devices, we faced two primary challenges: patterning our large-scale anodes and applying PEDOT to large-scale substrates in a uniform manner.

We needed to scale up our laser patterning capabilities to fabricate large anodes. Our laser system could only pattern a 120 mm by 120 mm square area. So we built a mechanical system (Figure 10, left) that allowed us to tile the laser patterns such that we can isolate and create larger devices. It uses CNC machining placement technology on an aluminum base with a precision translation stage. The challenge in this system was alignment – ensuring that the laser was able to create continuous lines from one tiled area to the next. This was crucial for creating devices that were larger than our original patterning capabilities. The right side of Figure 11 shows a microscope image of this alignment. The horizontal line and upper portion of the vertical line were patterned together with the laser in one position. The laser was then moved and the lower portion of the vertical line was scribed to create a continuous vertical line. While there is a small offset, we estimate that our positioning accuracy is on the order of one micron.

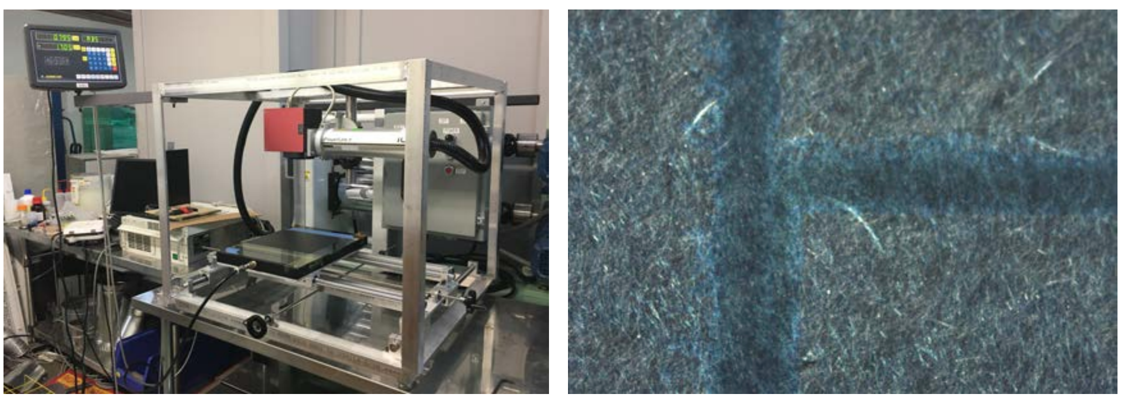


Figure 10. Left: Laser patterning setup to create large-area OLED electrodes. Right: Aligned laser marks. Upper vertical line and horizontal line were drawn first as laser made a right angle. Lower vertical line was subsequently aligned and scribed.

OLEDWorks gave us some options for anode layout, and we wished to maximize the number of devices that we could produce. Their large-scale runs are Gen 2 size, which is 14.6” by 18.5”. As our substrate was 10” wide, it would not be fundamentally possible to cover the entire layout using one piece of our film, and the patterning area was a limitation. Our initial goal for this project was to produce OLEDs that were 6” by 6” square, as this was the large-scale panel that our previous partner, Solvay OLED, was producing. However, OLEDWorks had different panel layouts for their Gen 2 sized runs. One option was 4" by 4" squares. However, this was not as good a test for our low sheet resistance films, as the linear distance was smaller. In addition, this layout was far from optimal with respect to the number of devices we could fabricate, due to how the devices were tiled relative to our size limitations. We opted instead for a higher aspect ratio rectangular device design. This allowed us to better test the limits of our low sheet resistance film with respect to brightness uniformity. And because of how the devices were tiled on the overall layout, this also allowed us to fabricate more complete devices in the same substrate area. So the OLEDs we chose to fabricate were 8.75 inches long by 1.875 inches wide. As they resembled strips or ribbons, they also were nice demonstrations of flexibility when illuminated.

The next challenge was depositing the PEDOT layer. PEDOT (Clevios formulations) are designed to be spun-cast over small areas, and we modified our PEDOT from the as-received formulation. We neutralized the pH from 1.5 to 5.5, as we found this reduced leakage current, we added a solvent that improved wetting on our anode, and then filtered the entire suspension to remove any larger particles or aggregates. In an ideal situation, we would be able to coat our PEDOT roll-to-roll. But we did not have the capability of coating PEDOT roll-to-roll with uniformly controlled thicknesses on the order of 50 nm. So we needed to use spin coating. Unfortunately our lab's spin-coater has a maximum substrate size of a 4" round wafer. Instead we used the large spin-coater Stanford's nanofabrication lab, with a diameter of 10.5 inches, just large enough for our substrates.

Table 7. Representative efficiencies of both small-area and large-area white OLEDs fabricated on our integrated plastic substrates.

Control (cd/A)	Small Devices (cd/A)	Large Devices (cd/A)
52.7	62.1	71.8
	75.6	64.1
	68.4	70.0
	75.4	64.1
	72.6	73.1

As OLEDWorks's Gen 2 production line is slightly different from the small-scale one, there were some additional challenges with respect to process flow. This included some exposure to atmosphere during processing, and our corresponding attempts to limit this by changing the electrical contact method. For this reason, it was not practical or accurate to do lifetime testing on these devices, as they had exposure to atmosphere that was unavoidable. If we were to do this again in the future, we would solve these issues beforehand with more extensive process flow testing, as current processes are designed to only work with glass / ITO. However, as shown in Table 7, the efficiencies of our large white OLEDs very closely mirror those of our small-scale test devices. This shows that we have effectively scaled our process without losing integrated substrate performance or white OLED efficiency. So as it stands, we will need to assume that with the similar initial OLED performance to the small-scale tests that we observed, and with more careful handling that would occur in a fully-developed fabrication process, that the lifetime of our large-scale OLEDs would be the same as our small-scale results, meeting our project goal of over 10,000 hours.

Figure 11 shows an image of our 8.75 inches long by 1.875 wide white OLED on the left. As evidenced by the photo, there are a few defective spots that could be due to dust or the aforementioned atmosphere exposure, both of which would be fixed in a production manufacturing process. To test brightness uniformity, and to ensure we met our program target of less than 15% brightness deviation across a lit panel, we did spot brightness testing. The devices were lit and brightness was measured in 9 spots across different areas of the panel, as shown in Figure 12 on the right. The data from a representative device is shown in Table 8. Calculating the deviation from this data gives a uniformity of 89% at 700 nits, surpassing our goal of 85% uniformity, which is not visible to the naked eye.

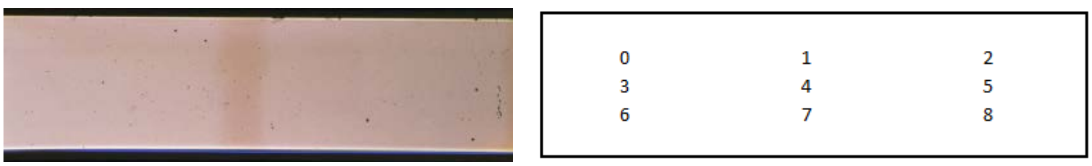


Figure 11. Left: white OLED in collaboration with OLEDWorks, 8.75” by 1.875” using our integrated substrate. Right: testing spots for brightness uniformity.

Table 8. Uniformity data across a large-area white OLED panel tested in the configuration shown in Figure 3.

Spot	Brightness (cd/m^2)
0	595
1	593
2	718
3	641
4	586
5	700
6	590
7	641
8	731

And finally, Figure 12 shows images of our completed devices. As shown, they are bright, flexible, and uniformly emitting. This marks the completion of our final program milestones, showing that our integrated substrates have the promise to effectively enable OLED makers to implement low-cost flexible lighting designs.

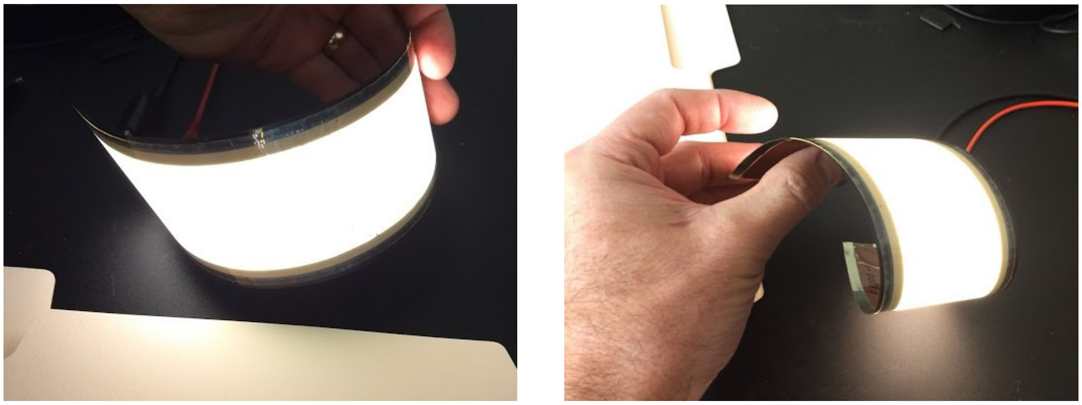


Figure 12. Large-area flexible white OLEDs on integrated substrate.

Products

Conferences and presentations:

- The PI gave a talk covering this project's goals and preliminary results at the OLEDs World Summit in Berkeley, CA, on October 29, 2015.
- The PI presented a poster at the DOE SSL Lighting Conference in Raleigh, NC, Feb 1-4, 2016.
- Dr. George Burkhard gave a talk on roll-to-roll manufacturing at the OLED Stakeholder's Meeting in Rochester, NY, October 18 – 19, 2016.
- The PI presented a poster at the DOE SSL Lighting Conference in Long Beach, CA, January 31 - Feb 2, 2017.
- The PI gave a talk covering the outcomes of this project at the Society for Information Display's Display Week Conference and Symposium, May 23, 2017.

Collaborations Fostered

This program was very beneficial in the area of fostering collaborations and bringing together a multidisciplinary team. Through the course of the project, team members included Sinovia Technologies, Vitriflex, Solvay USA, OLEDWorks, and Carestream Contract Manufacturing. Each collaborative team member brought important materials and experience to the program and contributed knowledge that allowed the team to achieve the project milestones. Going forward, many of these team members will continue to collaborate to research topics in OLED lighting and to bring products to market.

Technologies / Techniques

The two most important technologies and techniques developed through this work are the successful integration of the barrier film with the transparent conductive film while preserving the high performance of both thin-film technologies, and the development of the metering roll technique for scaled coating of silver nanowire films.

In integrating Sinovia composite transparent conductive films with Vitriflex barrier films and observing such high performance from these combined substrates, we have built a more complete plastic-based support solution for flexible OLEDs than has been done in the past. These integrated substrates meet or exceed the DOE's targets for both performance and cost and have the potential to be transformative for the OLED lighting industry. Not only will they enable flexible devices to show performance similar to or better than glass-based rigid ones, having the support in a roll format will pave the way toward the cost reductions that will come with roll-to-roll manufacturing of the OLED stack itself.

In developing the roll-coating method, we have solved the most challenging issue that others have experienced in silver nanowire thin-film manufacturing: how to coat a nanowire suspension developed for high conductivity over long lengths of substrate and long coating times without introducing aggregation defects. Suspensions of silver nanowires, because the particles have such high aspect ratios, have historically been very difficult to coat into a smooth, isotropic thin film without aggregation. Many specialized pieces of coating equipment have been used and many times they have not been effective. While our method itself is not new, using it and modifying it to work with high conductivity silver nanowire suspensions has broken new ground in the area of silver nanowire coating thanks to the collaboration between Sinovia and Carestream. This is extremely important work for manufacturing yield, feasibility, and cost, and the development and understanding of this method will influence all of Sinovia's future products using silver nanowires.