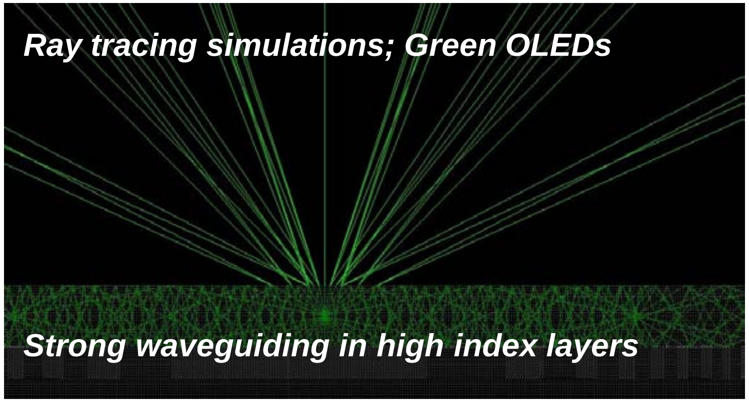
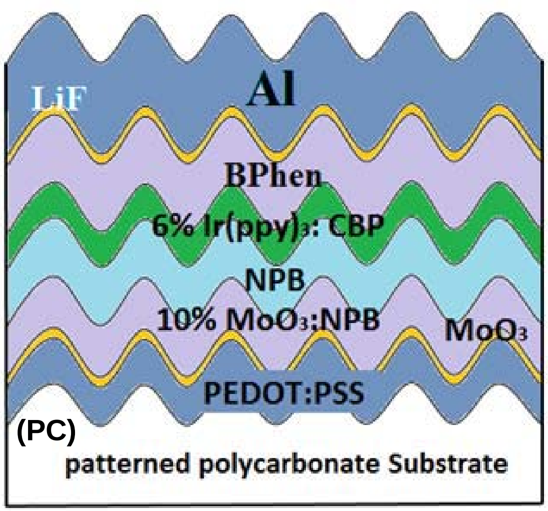
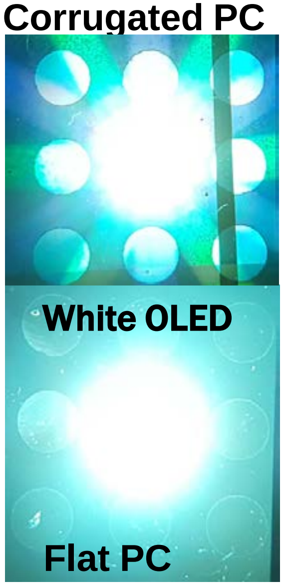
U.S. DEPARTMENT OF
ENERGY
Office of
ENERGY EFFICIENCY &
RENEWABLE ENERGY

Enhanced Light Extraction from Low Cost White OLEDs Fabricated on Novel Patterned Substrates


Ray tracing simulations; Green OLEDs

Strong waveguiding in high index layers





<u>Performing Organizations</u>: Iowa State University , MicroContinuum, Inc., Trovato Mfg., Inc.

<u>PI</u> : Ruth Shinar, *Senior Scientist, Microelectronics Research Center;*
Adjunct Professor, Department of Electrical and Computer Engineering
rshinar@iastate.edu

Project Summary

Timeline:

Start date: Aug. 1, 2016

Planned end date: September 30, 2018

Key Milestones

Milestone 1, August-September 2017: **Demonstrated a fully integrated substrate with internal & external extraction features and metal mesh/anode.**

Milestone 2, April 2017- Jan. 2018: **Demonstrated outcoupling enhancement for phosphorescent OLEDs on patterned plastic substrates and agreement between theory & experiment.**

Key Partners:

Iowa State University (ISU)
MicroContinuum, Inc. (MCI)
Trovato Mfg., Inc. (TMI)

Budget:

Total Project to Date:

- DOE: $933,858.96
- Cost Share: $276,707

Total Project:

- DOE: $1,318,903
- Cost Share: $331,607

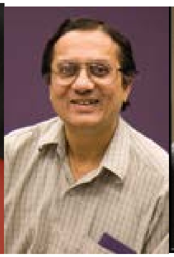
Project Outcome:

- Focus: developing cost-effective, integrated patterned plastic substrates for enhancing OLED light outcoupling η_{out}: demonstrated fully integrated substrates.
- η_{out} ≤20% is hindered by external & internal light trapping and loss to surface plasmons.
- High throughput, inexpensive patterned plastic substrates minimize losses; obtained 1.4x – 2x EQE enhancements relative to OLEDs on flat plastic (larger enhancements relative to glass/ITO OLEDs).
- Efforts to approach 2020 DOE MYPP η_{out}= 70% ongoing via substrate choice & integration design.

Team

R. Shinar

J. Shinar



R. Biswas

D. Slafer

T. Trovato

Iowa State University
Theory + OLED
fabrication &
characterization

MicroContinuum
Integrates
substrates

Trovato Mfg.
Advanced facility
OLED fabrication

ISU graduate students

Clockwise from top left: E. Manna, R. Kaudal, C. Hippola, B. Chen, A. Peer

ISU Team: Veteran forefront researchers in experimental and computational organic photonics & electronics; developed efficient solution-processed small molecule & ITO-free OLEDs, combinatorial near-UV-to-red microcavity OLEDs & outcoupling enhancing approaches; conducted pioneering studies of OLED exciton & polaron dynamics, and pioneered OLEDs + thin film organic & hybrid photodetectors in biochemical analyses. **The team collaborated for years and coauthored many publications.**

MicroContinuum, Inc.: Extensive experience in developing complex R2R (nano)structures on flexible substrates. Developed R2R sputter coater and actively participated in R&D Programs w/Fortune 500 companies, national labs, universities, & government agencies.

Trovato Mfg. Inc.: Expertise in technical & experimental support for OLED design & manufacturing deposition tools, including computer-controlled 8-, 12-, & 16-source thermal evaporators.

TMI's OLEDs on flexible substrates

Challenge

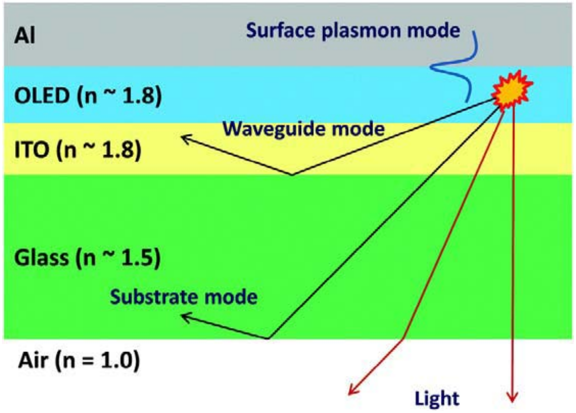
PROBLEM: η_{out} only ~20% - major impediment for solid state lighting

Issues: Photons

(1) Trapped in the substrate due to total internal reflection at substrate/air interface,

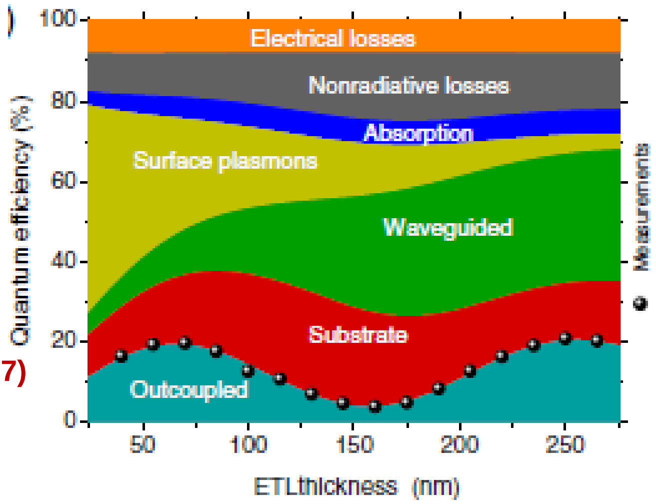
(2) waveguided in the high index organic + anode layers,

(3) lost to surface plasmons at organic/metal electrode interface.



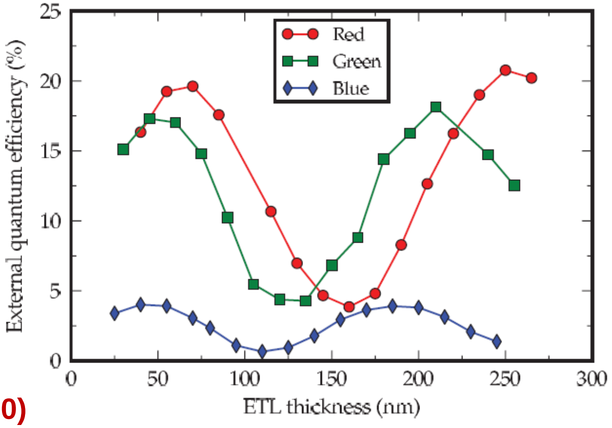
F. Zhao et al. *Mater. Chem. Frontiers* (2017)

Red bottom emitting OLED



R. Meerheim et al. *Appl. Phys. Lett*. 97, 253305 (2010)

Furno et al. *PRB* 85, 115205 (2012)



Goal: Develop low cost method to approach the DOE 2020 MYPP target outcoupling factor of 70%

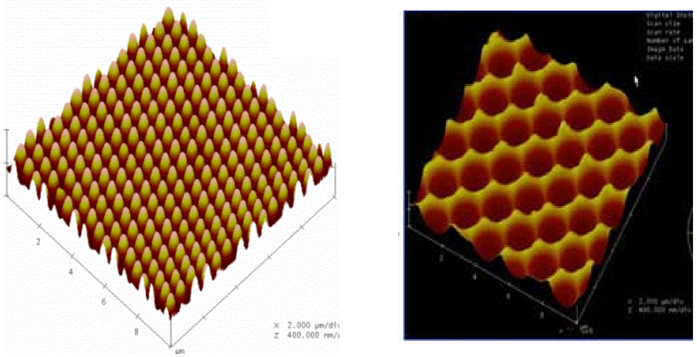
Approach

- **Increase η_{out} by disrupting internal waveguiding & plasmon losses using low-cost corrugated integrated substrates**
- **Test simple patterned OLEDs**
- **Use simulations to support substrate and OLED designs**

MCI's versatile fabrication methods

(1) Direct nanopattern formation

- No patterned layer/substrate interface →
  No peel off & index mismatch losses
- Room temperature production →
  No thermal distortion or degradation
  Reduced production time
- Rapid pattern changes & inexpensive tooling;
  Tunable pattern amplitude w/o expensive new templates



AFM of corrugated polycarbonate (PC) and polyethylene terephthalate (PET)/polymer substrates (10 μm full scale)

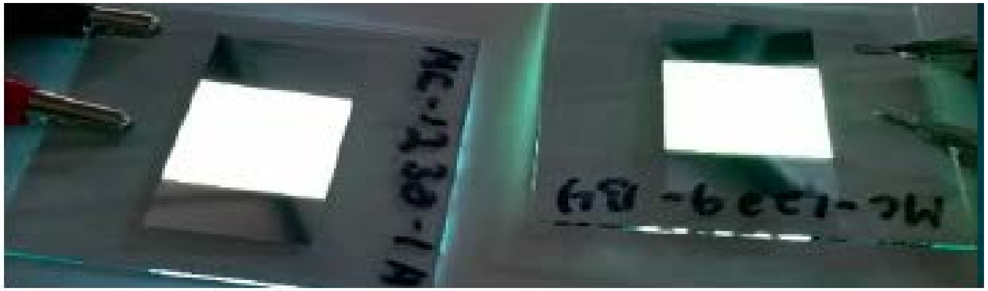
(2) Two layer formulation

- Pattern and substrate independent optimization, adjusting the refractive index by material choice → ability to add high index particles

OLED Fabrication, Characterization, and Modeling

- At ISU & TMI- fabrication on integrated patterned & flat substrates; characterization: structural, chemical, & optoelectronic.

Fluorescent WOLEDs fabricated at TMI

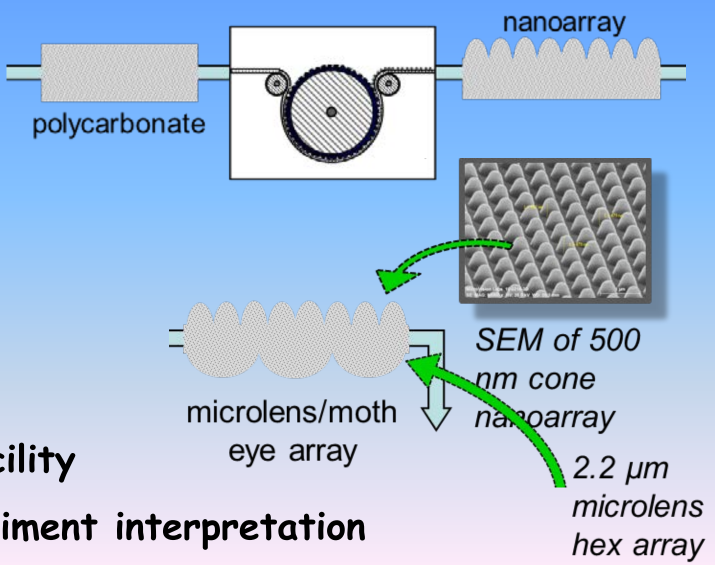


Modelling : Integrated substrates and OLED designs - predictive simulations & interpretation support

Computational Approaches: scattering matrix simulations (in Fourier space) and real space finite difference time domain (FDTD) simulations (Lumerical)

Most systematic modeling and experiments on light outcoupling from OLEDs on corrugated substrates

Approach Advantages



Substrate manufacturing

- Cost-effective R2R, simple direct room temperature molding,
- Large area; scalable for high volume & rapid production
- Versatile, amenable for a variety of structures & substrates
- Multi-level structures: nano-patterns + embedded metal mesh/transparent conductor and microlens array

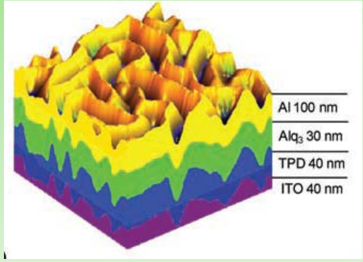
OLED Fabrication, Characterization, & Simulations

- Broad characterization & testing at ISU & advanced TMI facility
- Predictive modeling reduces trial & error, and supports experiment interpretation

A feasible approach to improve OLED performance while lowering costs

Other Approaches:

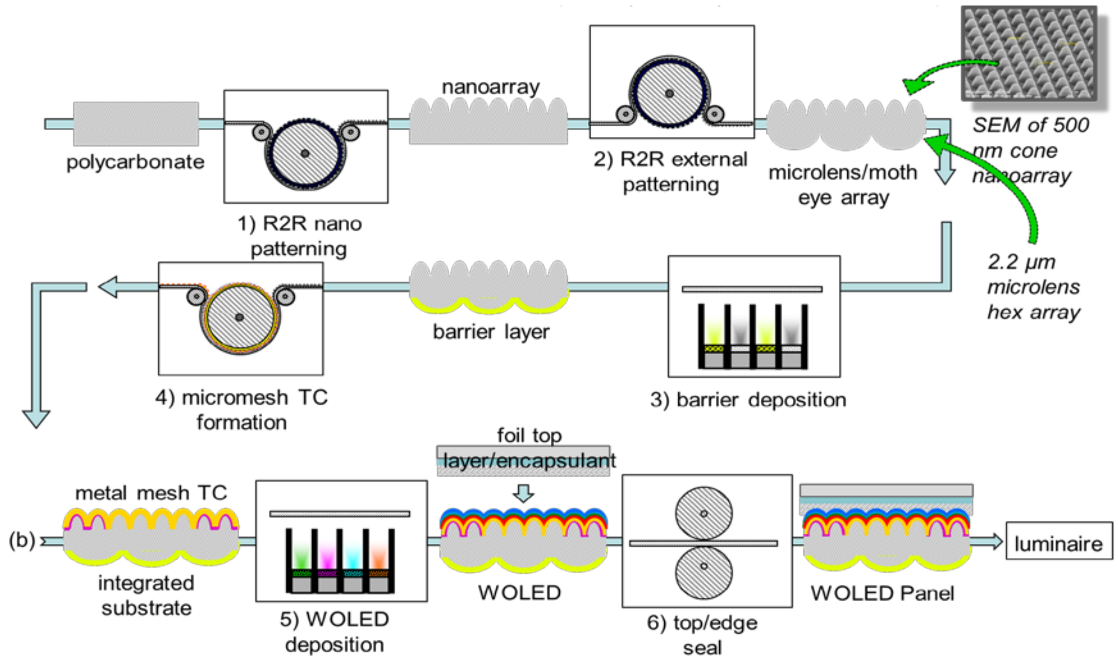
- Costly: expensive substrates, especially for scaling
- May raise environmental issues
- Scattering films → viewing angle – dependent spectra
- Complex designs → more prone to shorts



Koo et al. Adv. Mater. 2011

Impact

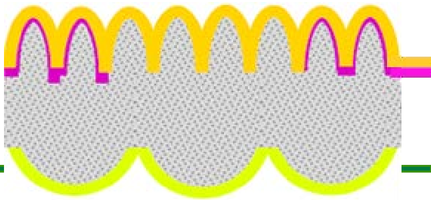
- Achieving our project's goal, i.e., approaching 2020 MYPP DOE target η_{out} = 70%, will become possible with a cost-effective method that is scalable for high volume and rapid production.
- DOE/EERE is currently investing in developing different approaches for enhancing η_{out}. Our approach, utilizing MCI's R2R process, refrains from expensive substrates and costly complex procedures that are less amenable to high throughput scaling.
- Continued substrate optimization will enable MCI to work with existing OLED companies and later with TMI to fabricate efficient, low cost OLEDs for various applications.



R2R production of integrated OLED substrates. PC: directly imprinted on both sides + an external barrier. (b) OLED deposition & encapsulation steps. (not to scale)

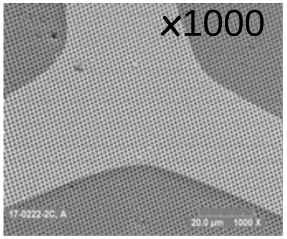
Progress

Schematics of fully integrated plastic substrate/anode



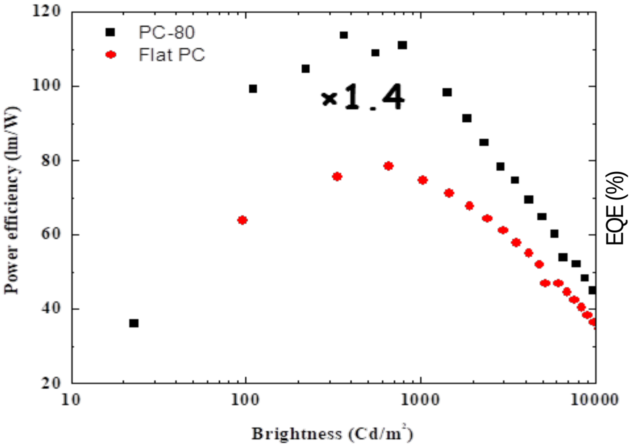
Mid-stage project: demonstrated fully integrated substrates, increased η_{out} & *theory-experiment agreement*

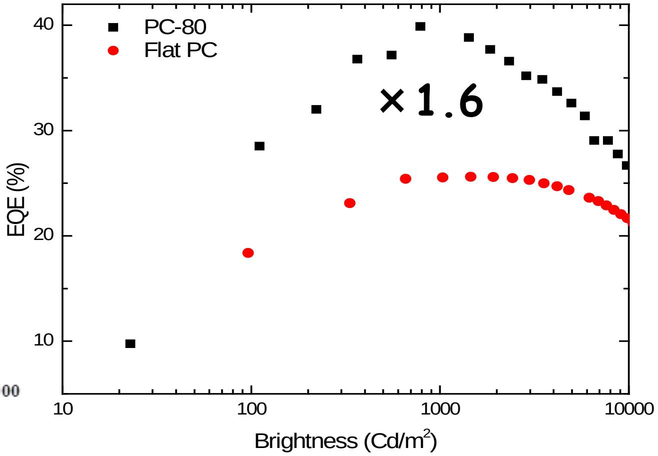
Integrated substrate; buried Cu grid/ITO

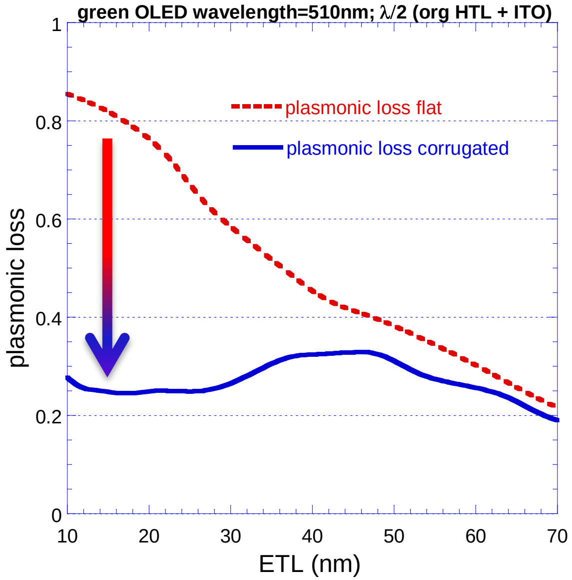


Low corrugation height: *h* ~80 nm, *a* ~750 nm

Green OLEDs: EQE:40%





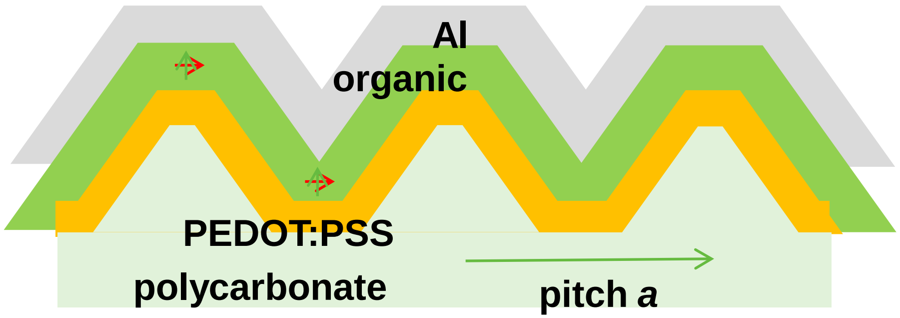


The enhancement is due to disrupting both the surface plasmon & waveguide modes

Preliminary Simulations: *h*=50 nm, *a*=750 nm
Strong reduction in plasmon loss for thin ETL at low *h*

PC-80/PEDOT:PSS/HAT-CN (5 nm)/10% MoOx:TAPC (60 nm)/TAPC (20 nm)/6% $Ir(ppy)_3$:mCP (20 nm)/TmPyPb (20 nm)/20% CsF:TmPyPb (40 nm)/LiF (1 nm)/Al (100 nm)

Large corrugation height: Cathode *overlaps* anode; two dipole emitter positions shown

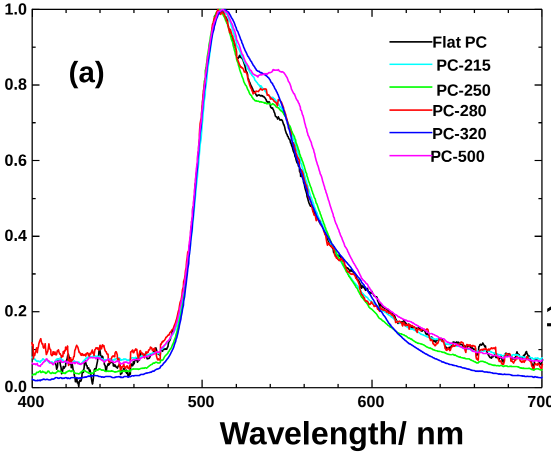


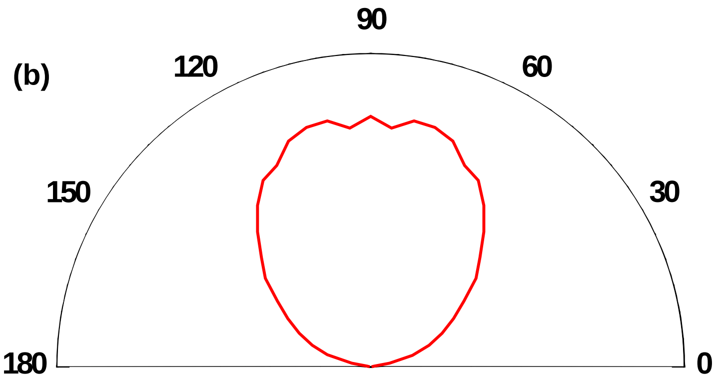
Schematic of pattern (not to scale)

Simulation Results

- Large *h* - strong diffraction
- Internal waveguide, surface plasmon, and possibly substrate modes are extracted into air
- *a* ~500-750 nm and *h* ~200-400 nm enhance OLEDs
- Experimentally: optimal *h* ~300 nm; *a* ~750 nm
- Preliminary simulations: η_{out} ~50-60%

Optical Attributes





Ideal Lambertian Distribution

Angular Dependence with Patterned PC

<u>PC-320</u>:
EQE = 36%

~x1.6 relative to flat PC (~22%);

x2 relative to glass/ITO (~18%)

PC-320/PEDOT:PSS/MoO_x(1 nm)/10% MoO_x:NPB (22.5 nm)/NPB (22.5 nm)/6% $Ir(ppy)_3$:CBP (11 nm)/BPhen (40 nm)/LiF (1nm)/Al (100 nm)

Enhanced Green OLEDs (non-encapsulated devices)

Device Structure: Anode/HAT-CN (5 nm)/10% MoOx:TAPC (120 nm)/TAPC (20 nm)/6% $Ir(ppy)_3$:mCP (20 nm)/TmPyPb (20 nm)/20% CsF:TmPyPb (40 nm)/LiF (1 nm)/Al(100 nm)

Anodes:

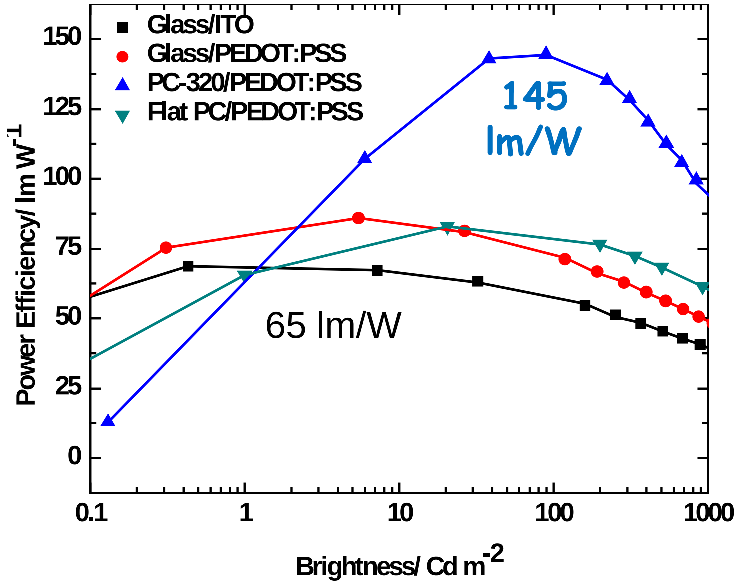
1. Glass/ITO
2. Glass/PEDOT:PSS
3. PC-320/PEDOT:PSS

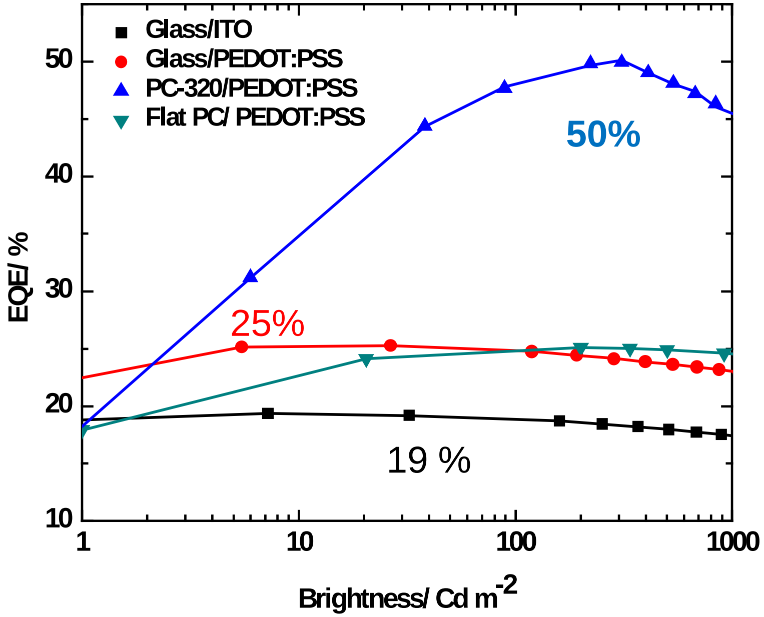
(*h* ~320 nm; *a* ~750 nm)

4. Flat PC/PEDOT:PSS

EQE = 50%; $\eta_{out} \geq 50\%$

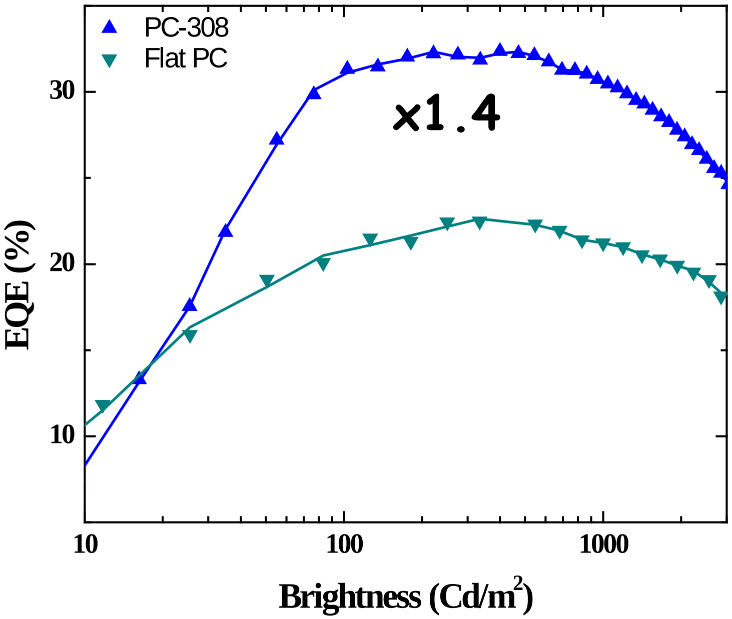
×2.6 relative to glass/ITO





Blue OLEDs

Device structure: PC-308/PEDOT:PSS/HAT-CN (5 nm)/10% MoOx:TAPC (60 nm)/TAPC (20 nm)/8% Firpic:mCP (20 nm)/TmPyPb (20 nm)/20% CsF:TmPyPb (40 nm)/LiF (1 nm)/Al (100 nm)

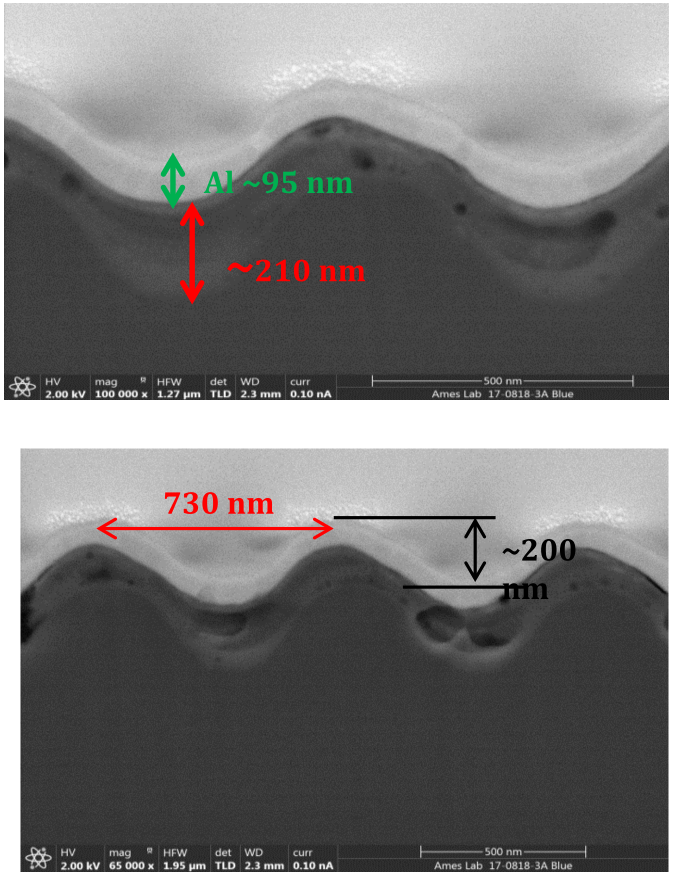


Luminous efficiency x1.4: 80 vs 57 cd/A

EQE x1.4: 32.5% vs 22.5%

Power efficiency x1.25, 50 vs 40 lm/W

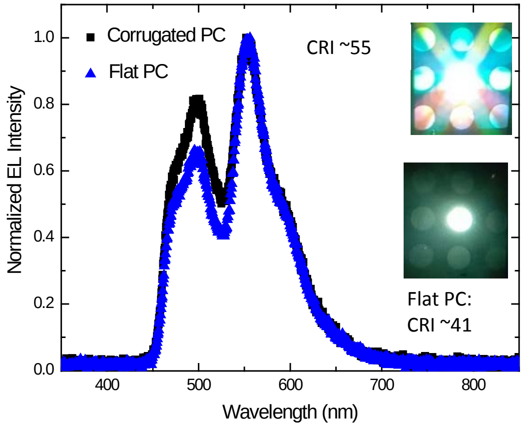
FIB Analysis

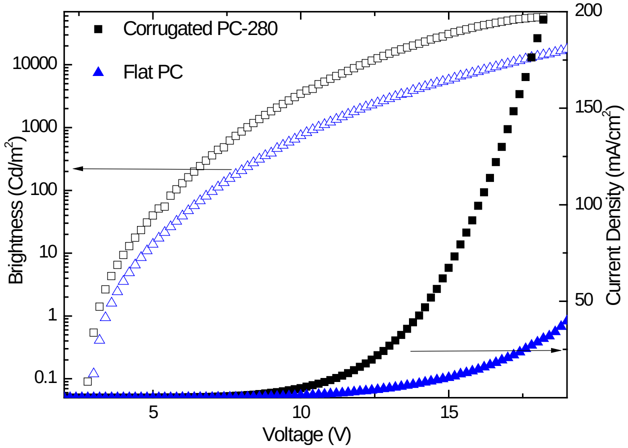


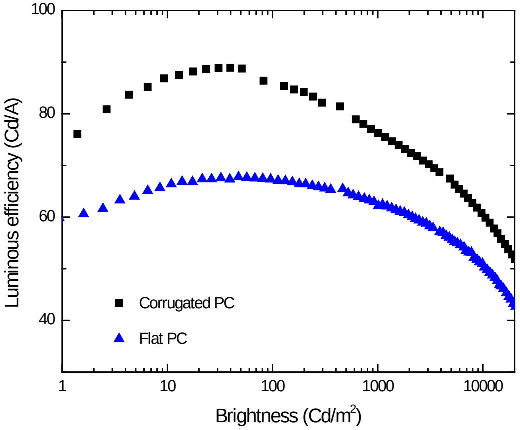
- Total FIB-measured thickness: ~305 nm
- Device thickness: ~295 nm; ~265 nm, measured with thickness monitor + ~30 nm PEDOT:PSS thickness
- Corrugation height at Al ~200 nm
- Substrate: *h* ~308 nm

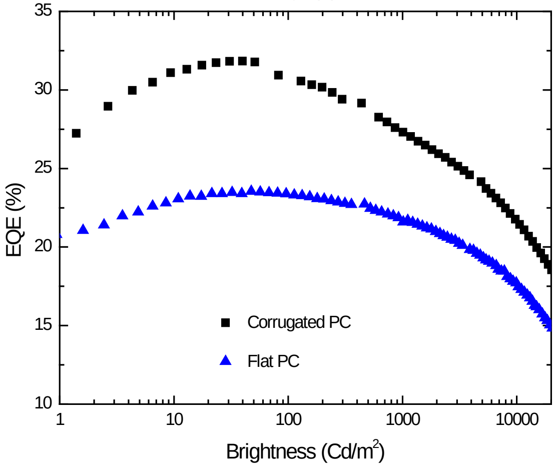
White OLEDs

Device structure: PC-280/PEDOT:PSS (~20 nm)/HAT-CN (5 nm)/10% MoOx:TAPC (120 nm)/TAPC (20 nm)/8% FIrpic:mCP (19 nm)/6% PO-01:mCP (1 nm)/TmPyPb (20 nm)/20% CsF:TmPyPb (40 nm)/LiF(1 nm)/Al (100 nm)









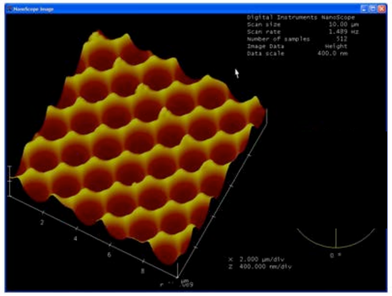
- **Patterned OLED:**
  ***h* ~280 nm; *a* ~730 nm**
- **EQE ~32%; x1.4 vs planar plastic**
- **CRI increases from flat to corrugated**

- **Narrow EL spectrum:**
  only orange and blue emitters

FIB Analysis: White OLEDs

White PhOLED:
PET/ITO/20 nm MoOx/126 nm organics/1 nm LiF/100 nm Al

ITO Anode



Substrate AFM image

Backscattered electron image

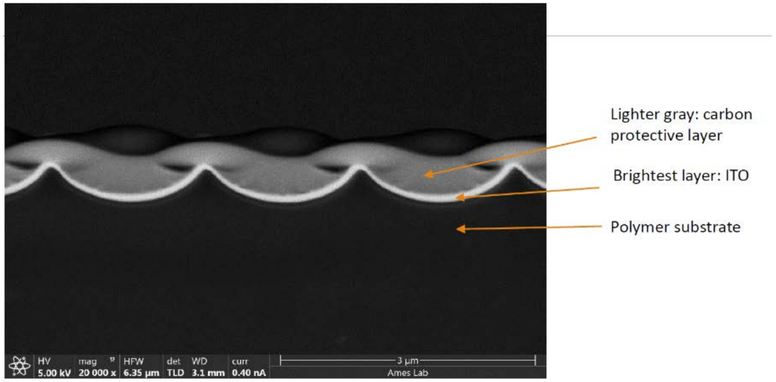
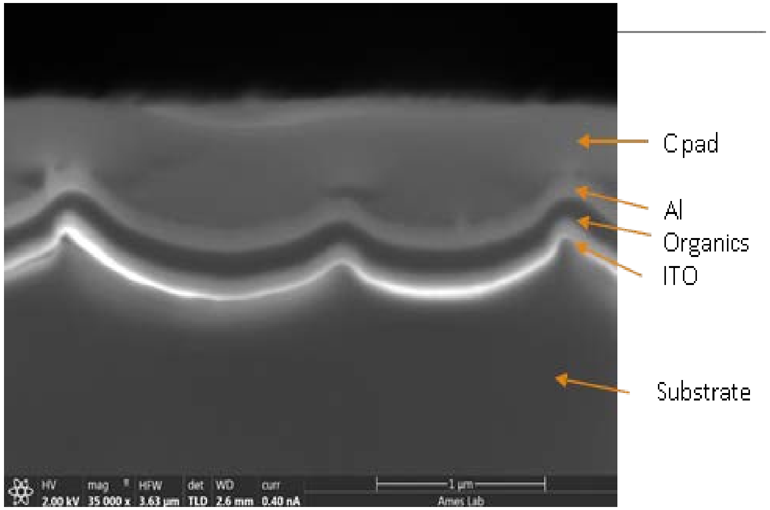
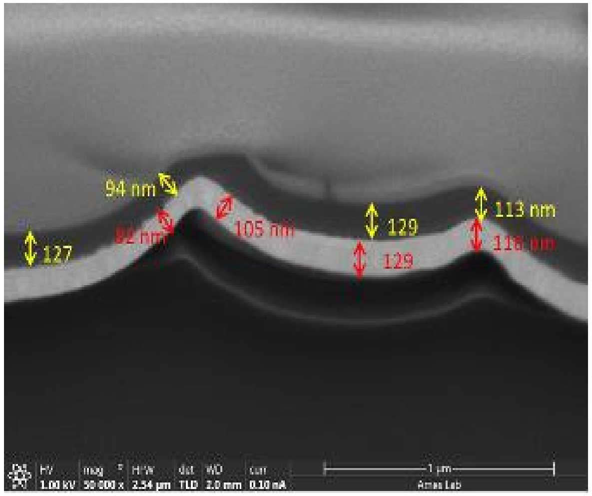
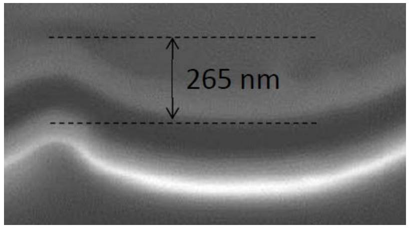


Image of conformal ITO on concave PET/CAB (h ~250 nm; pitch ~2 µm)





Al layer corrugation



ITO (red arrows)

organics (yellow arrows)

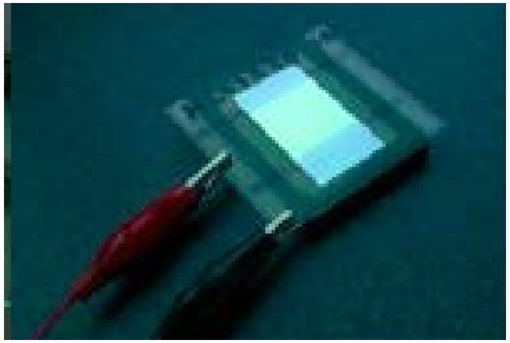
The OLED structure is largely conformal including the Al electrode w/some thickness variations across features

Stakeholder Engagement

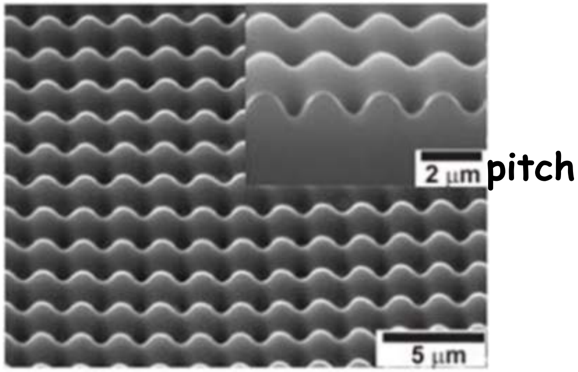
- The project is at ~mid-stage; the approach with R2R fully integrated patterned substrate production is in early development
- MCI's strategy: first transfer the batch-based process to their R2R pilot line, which will ultimately produce medium volume OLED substrates.
- The initial commercialization strategy is to provide enhanced light extracting structures to OLED manufacturers. Their initial customers would include current OLED lighting companies, e.g., Philips/OLEDWorks, Osram, LG, Konica Minolta.
- MCI has identified a specific OLED lighting market opportunity in the auto industry with a major OEM auto parts supplier for the use of OLED in brake lights, side markers, and interior lighting in cars.
- The long-term goal is to expand the R2R line to include (with partners, including TMI) an OLED production module to enable R2R manufacturing of OLED panels.

Remaining Project Work

- **Substrates**: Improve integrated substrate/anode by material choice, design, defect-free fabrication, buried metal mesh/field conductor optimization, conductivity improvement; longer term - barrier integration exploration
- **OLEDs**: Optimize patterned white OLEDs: characterize conformality, optoelectronic, and structural/chemical properties vs substrate design; test effect of added microlens array - all to achieve higher efficiencies @ higher brightness + higher CRI + eventual improved stability
- **Theory**: Obtain guiding maximal achievable enhancement with corrugated integrated substrates to address expectations, including industrial; work closely with substrate & OLED optimization efforts



TMI's encapsulated flat fluorescent WOLED



Optimal microlens array:
h 1.2 μm; 1.6 μm dia.

J-M. Park et al., Opt. Express, 19, A786-A792 (2011)

Acknowledgement: DOE EERE program

Thank You

Performing Organizations:

Iowa State University

MicroContinuum, Inc., Cambridge, MA

Trovato Mfg. Inc., Victor, New York

Ruth Shinar, Senior Scientist Microelectronics Research Center

Adjunct Professor, Department of Computer and Electrical Engineering

Iowa State University, Ames, IA 50011

Tel. 515-294-5898

rshinar@iastate.edu

REFERENCE SLIDES

Project Budget

Project Budget and History

Variances: There were no variances from the original planned budget, but two-months no-cost extension was added to Year 1.

Cost to Date: ~70% of the project budget has been expended to date.

Additional Funding: There are no other funding sources.

Budget History					
Aug. 1, 2016– FY 2017 (past)		FY 2018 (current)		FY 2018 (current) End date – Sept. 30, 2018 (planned)	
DOE	Cost-share	DOE	Cost-share	DOE	Cost-share
$660,798	$227,089	$273,061	$49,618	$385,044	$54, 900

Project Plan and Schedule

Initiation date Aug. 23, 2016;
Planned completion date Sept. 30, 2018
not including a possible no-cost extension

Milestone Summary Table – Year 1

Milestone	Description	Verification Process	~Month	Status
1.1	OLEDs on flexible substrates, without & with metal mesh transparent conductor (MMTC), η_{out} ~ 45%, CRI ~ 80	Measurements of EQE, luminous and power efficiency, EL spectra, CRI , structural and conformality analyses of devices on flat & patterned substrates	6	We achieved reproducible enhancements for phosphorescent OLEDs on corrugated PC substrates without metal mesh. For the green OLED the EQE was 50%, x2 that of a device on flat PC; x2.6 relative to a device on glass/ITO. The highest CRI we observed was ~65. In Year 2 we will first focus on assessing challenges with the integrated substrate (see milestone 1.2)
1.6	Demonstrate agreement between OLED performance and model.	Comparison of measured OLED performance on flat vs patterned flexible substrates vs model predictions.	9	Achieved for initial results. Based on theory, the best corrugation includes convex features with a pitch of ~750 nm for blue emission, and feature height of 350 nm, which is in agreement with experiments. Prediction of optimized ETL thickness is also supported by experiment.
1.2	Successful fabrication of uniform high fidelity plastic substrates without & with MMTC.	Fabrication and evaluation of OLEDs on integrated substrates without and with MMTC	12	Achieved the ability to fabricate a fully integrated enhanced substrates with internal and external extraction features and a buried metal mesh/anode -With the fabrication process in hand, the next challenge is to reduce processing defects, and enable OLED testing on such integrated structures (Year 2)

Project Plan and Schedule (cont.)

Budget Period 2				
Milestone	**Description**	**Verification Process**	**~Month**	**Status**
2.1 & 2.2	Design nanoarrays with η_{out} ~ 60%	Similar to that of Milestone 1.1, including EL angular dependence	15	The best achieved so far is $\eta_{out} \geq 50\%$ for green OLEDs. EL angular dependence was shown to be close to Lambertian. Expected increased η_{out} following ongoing systematic evaluation of the integrated substrates: month ~20
3.1	L70 ≥ 1,000 h at an initial brightness of 1,000 Cd/m^2.	Accelerated lifetime measurements at brightness > 1,000 nits, and extrapolation to 1,000 nits to determine L70 at initial brightness of 1,000 nits.	15	Encapsulation of corrugated devices is being explored. Expected evaluation results: month ~22
2.3	Design nanoarrays with η_{out} = 70%.	Similar to that of Milestone 1.1; determine losses due to waveguiding and plasmonic excitation in new designs	21	Losses due to waveguiding and surface plasmon excitation were determined (theory & experiment). Further increased η_{out} is expected in month ~23
4.1	Low cost OLEDs with η_{out} ~ 70%, CRI ~ 90, and L70 ≥ 1,000 h at an initial brightness of 1,000 Cd/m^2.	Similar to that of Milestone 1.1,	24	

A delay in achieving Year 2 milestones is due to issues with MCI's imprinting tool and ISU's glovebox thermal evaporator repair.

In particular, at the end of Year 1 and during Q1 Year 2 we identified challenges with the multifaceted fabrication of the fully integrated substrates, mostly with the buried metal mesh and ITO. We are hence devoting time to pinpoint the materials/steps that are responsible for these issues, starting with substrates from different sources.

Encapsulation of the corrugated structure is challenging and is being explored.

The general approach & goals are unchanged.