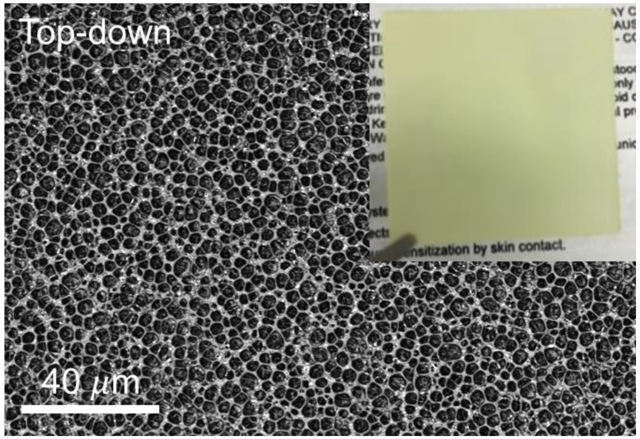
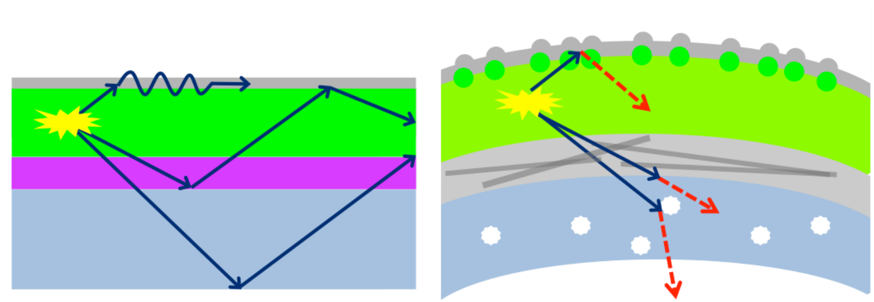
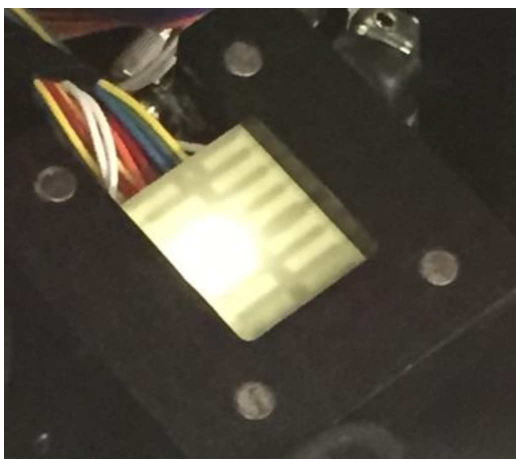
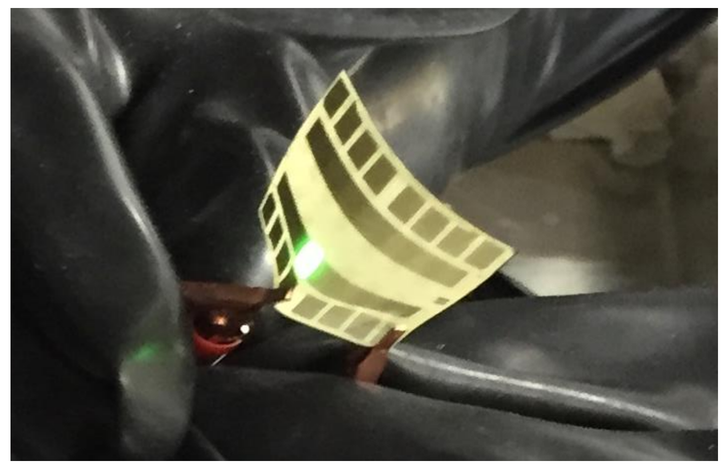
ITO-free white OLEDs on flexible substrates with enhanced light outcoupling

2016 Building Technologies Office Peer Review









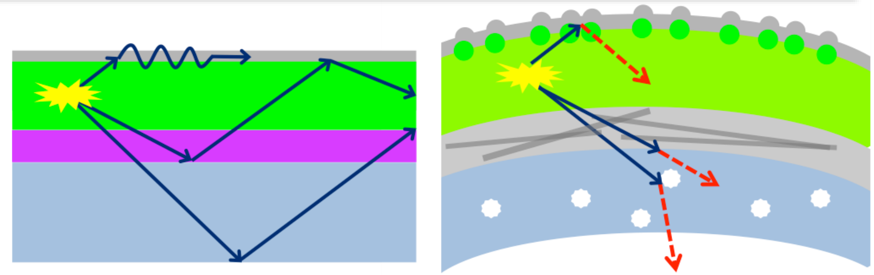
U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

Barry P. Rand
Princeton University
brand@princeton.edu

ITO-free white OLEDs on flexible substrates with enhanced light outcoupling

PIs: Prof. Barry P. Rand, Prof. Craig B. Arnold and Prof. Stephen Y. Chou
Princeton University

Our proposed approach *promises to unlock trapped photons* in state-of-the-art WOLED structures *on flexible substrates*.



Conventional Proposed approach

The keys to our approach are:
1) the use of scattering voids embedded within the flexible substrate which are intrinsically absorption-free, an aspect not achievable with high-index scattering centers reported previously.
2) The elimination of high-index and brittle ITO, replaced with low index electrodes that will not trap light. Specifically, we will utilize ultrathin metals, a nanoimprinted mesh or Ag nanowires.
3) The creation of structures that allow for surface plasmon mode scattering into radiant modes such that SPP losses may be mitigated. Key here is to achieve this without frustrating the structure of the WOLED itself. We will achieve this feat with gentle lithography on top of the sensitive organic layers, and also with the aid of a plasmonic cavity, whereby 'dark' plasmonic modes at the back electrode are coupled to 'bright' plasmonic modes of the mesh electrode.

Ultimately, *these approaches individually cannot meet our objectives*, thus the take-home message is that we will integrate these various approaches into one WOLED structure meeting the following aggressive targets: 1) 3x light extraction enhancement vs. a conventional structure, 2) 1 cm^2 device on a flexible substrate, and 3) a WOLED with 130 lm/W, CRI > 80, EQE = 65% at lighting-relevant brightness

This innovative progress will impact strongly, by helping to *establish U.S. dominance in the OLED SSL domain* via proper care for intellectual property and technology transfer and licensing.

This project has a total budget of $1,276,902, of which $1,021,242 come from EERE and $255,660 are cost share funding from Princeton University.

Purpose and Objectives

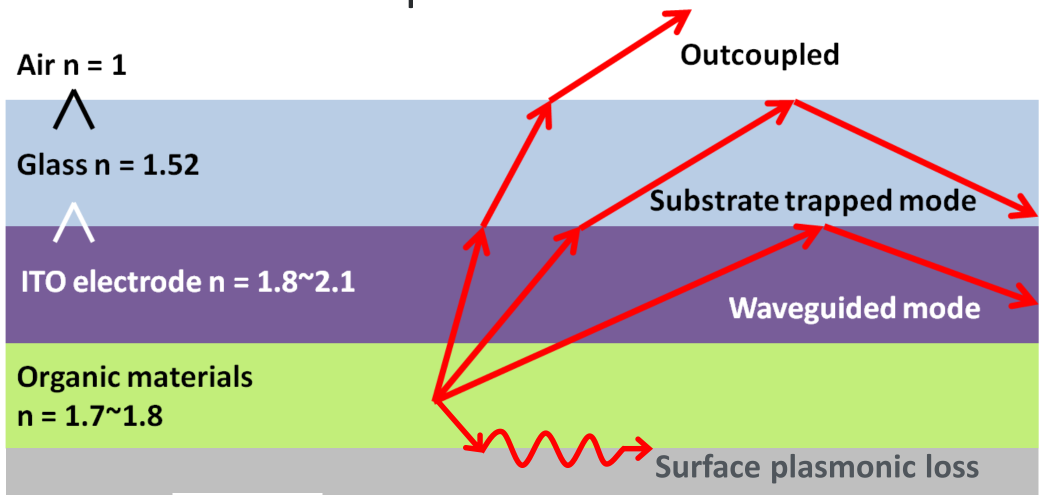
Problem Statement: The DOE has set aggressive targets for solid state lighting efficacy in its MYPP, aiming for >200 lm/W. For OLED lighting, the 2020 target is 140 lm/W. The **major obstacles to efficient OLED SSL sources are light outcoupling and lifetime.**

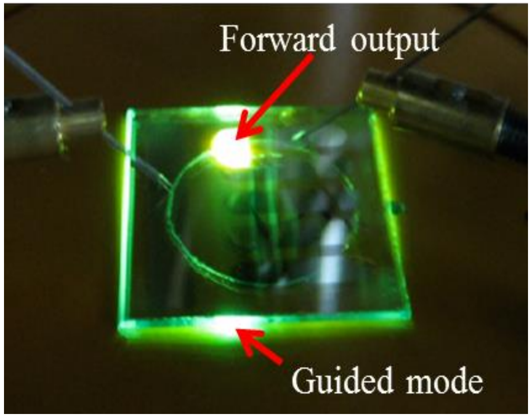
Target Market and Audience: The potential market for solid-state lighting is significant, with worldwide sales totaling $14.4 billion in 2012, a 25% growth over the previous year (according to Strategies Unlimited). Similar levels of growth are projected in the coming years, meaning that SSL will begin to occupy a larger and larger share of the overall lighting market. On the cost savings side, in 2012 SSL was responsible for saving 71 tBtu, roughly $675 million in energy expenditures.

Impact of Project: We target to improve outcoupling of OLED lighting. Furthermore, we do this with processing and materials that are upscalable and amenable to low-cost production of SSL luminaires on flexible substrates.

OLED outcoupling analysis

- Conventional bottom emitting OLEDs face multiple challenges when it comes to getting all of the photons out
  - SPP losses
  - Waveguided
  - Substrate trapped
  - Parasitic absorption





η_{out} **remains 20~30%**

Significant bottleneck

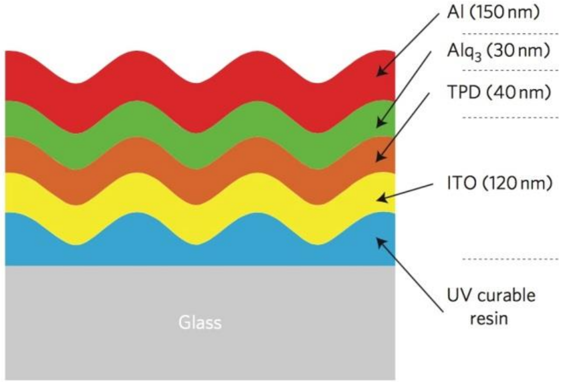
U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

Strategies for maximizing η_{out} of OLEDs

1) Minimize surface plasmonic loss mode

* Calculation based on *Phys*. *Rev*. *B* **85**, 115205

Plasmonic loss minimized by:
1) using thick transport layers
2) introducing corrugation



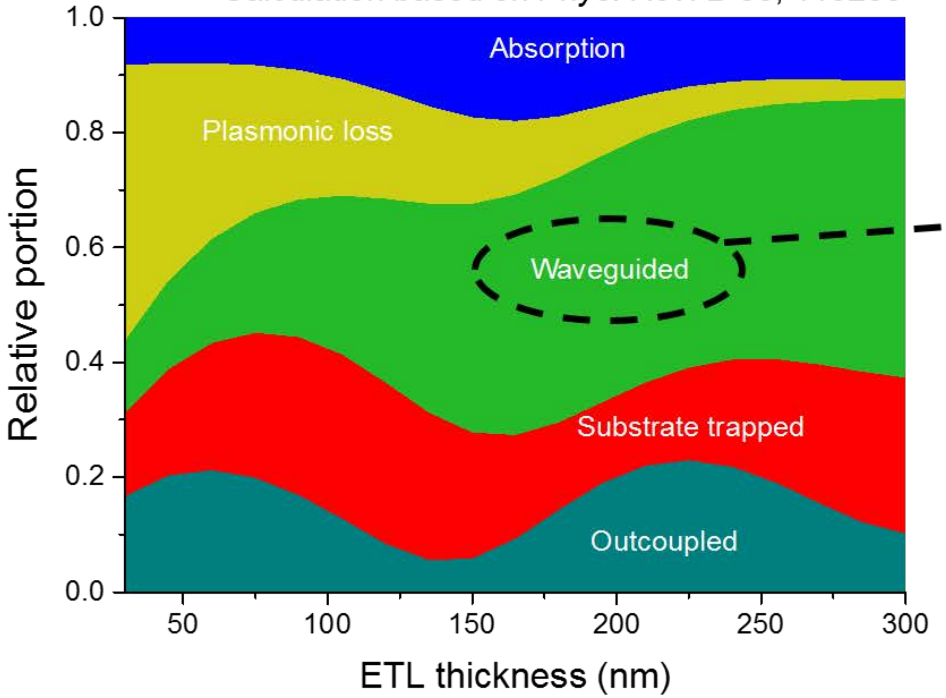
W.H. Koo et al, *Nat*. *Photon*. 4 (2010)

U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

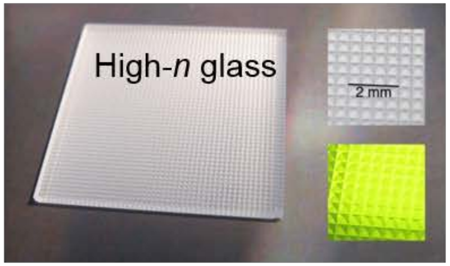
Strategies for maximizing η_{out} of OLEDs

1) Minimize surface plasmonic loss mode
2) Use high-index substrates to convert waveguided mode

* Calculation based on *Phys. Rev. B* **85**, 115205



Waveguided mode merged with substrate trapped mode

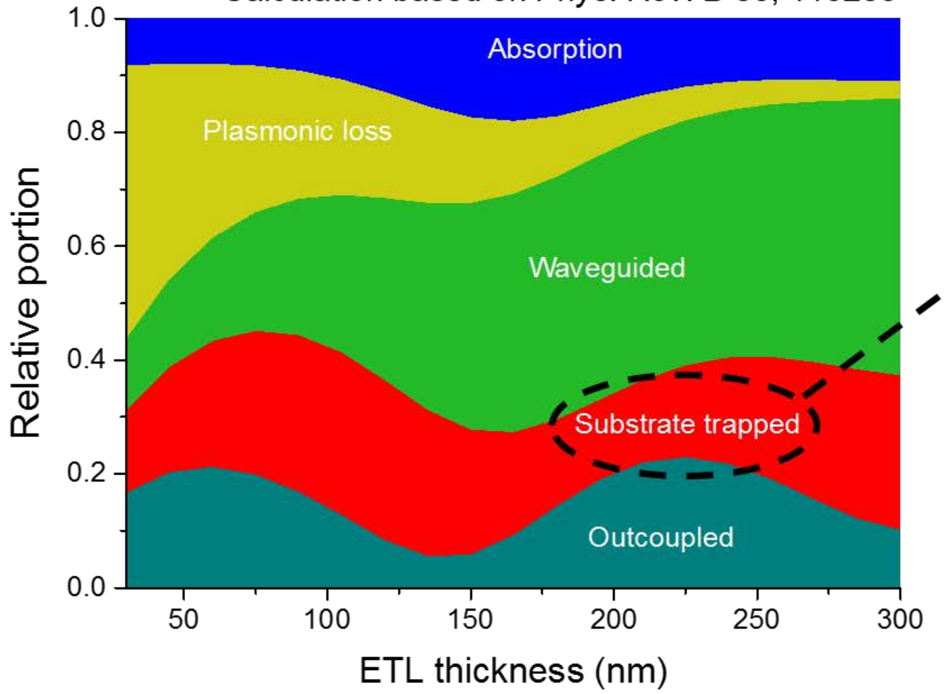


S. Reineke et al, *Nature* 459 (2009)

Strategies for maximizing η_{out} of OLEDs

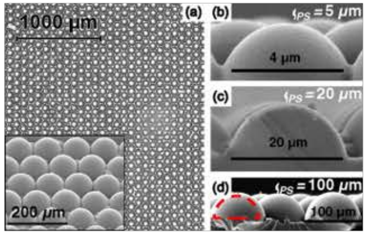
1) Minimize surface plasmonic loss mode
2) Use high-index substrates to convert waveguided mode
3) Use external extraction layers to recover substrate mode

* Calculation based on *Phys. Rev. B* **85**, 115205



- Half-sphere lens,

- Microlens array,



S.-H. Eom et al, *Org. Electron*. 12 (2011)

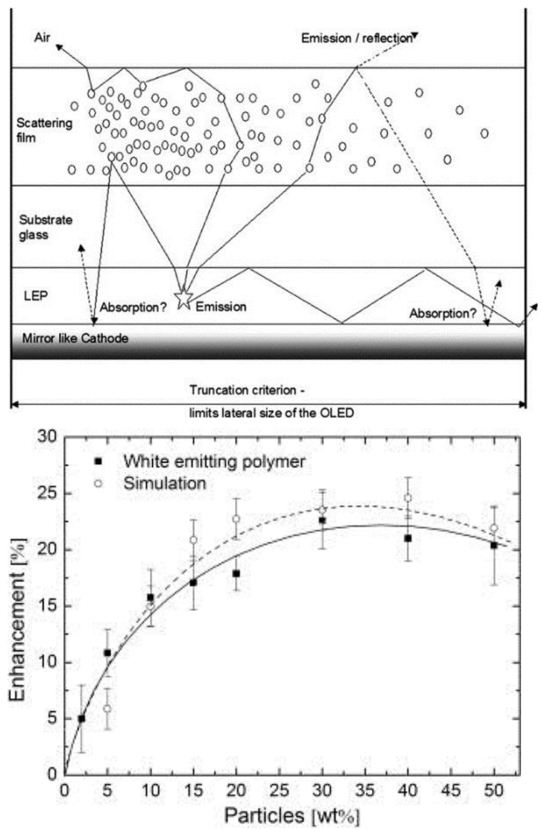
- **Scattering films**

Scattering films as extraction layers

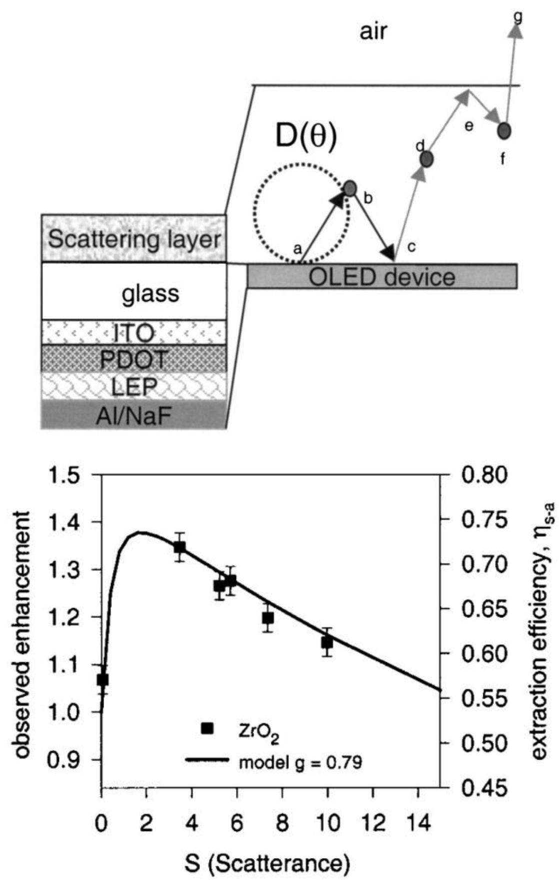
A straightforward way to address the substrate-trapped mode:

Scattering particles embedded in a clear host medium

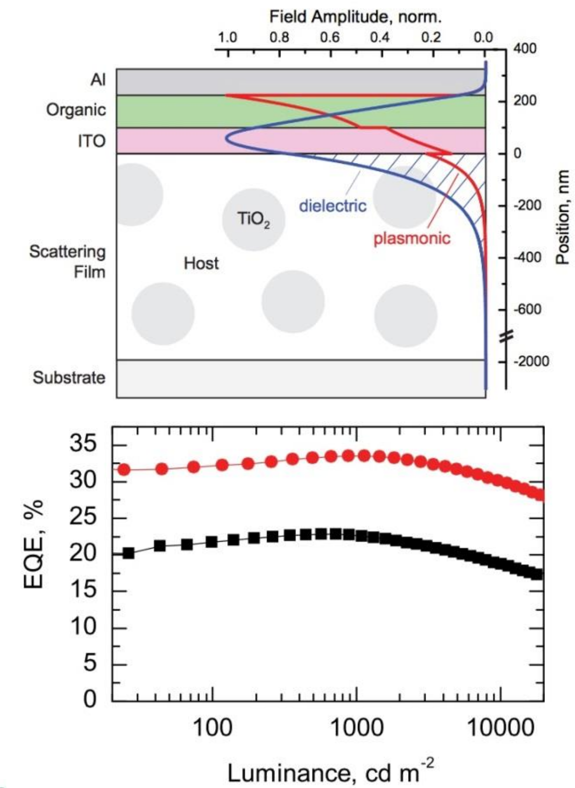
Polymer microspheres in acrylate
R. Bathelt et al, *Org. Electron.* 8 (2007)



ZrO_2 powder in PDMS
J.J. Shiang et al, *J. Appl. Phys.* 95 (2004)



TiO_2 NPs in polymer film
H.-W. Chang et al, *J. Appl. Phys.* 113 (2013)

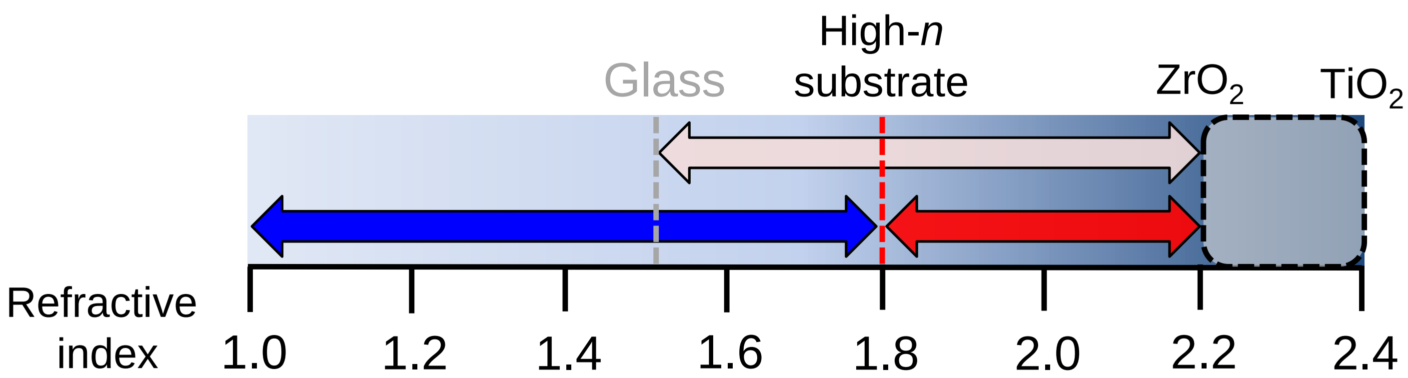


Scattering films as extraction layers

A straightforward way to address the substrate-trapped mode:

Scattering particles embedded in a clear host medium

What if we use high-index (n ~ 1.8) substrates
to merge waveguided mode with substrate-trapped mode?



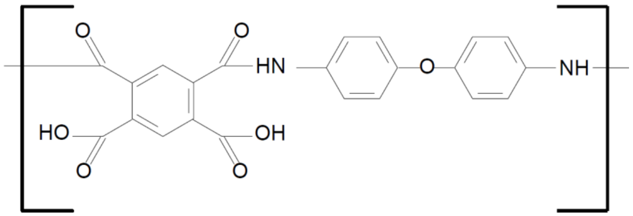
Index contrast between oxide scatterers and the substrate reduces as n_{sub} gets higher :

Utilizing voids (n = 1) as non-absorbing scattering centers in high-index, low-cost plastic substrates

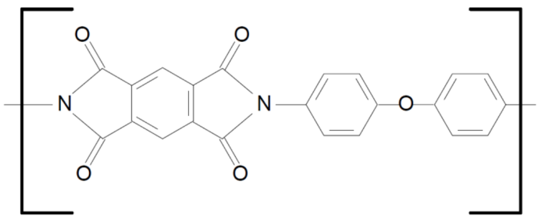
Polyimide as a high-index host medium

T.-W. Koh et al, *ACS Photonics* **2**, 1366 (2015)

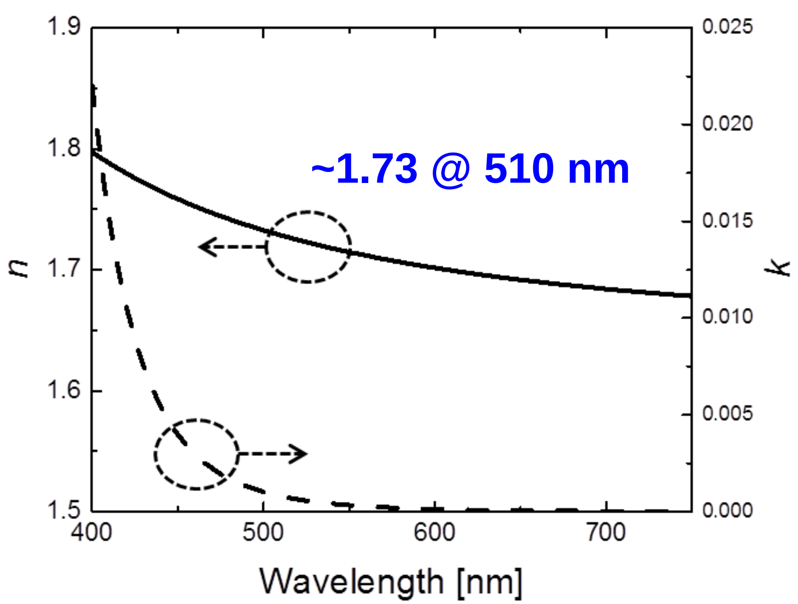
Kapton® polyimide

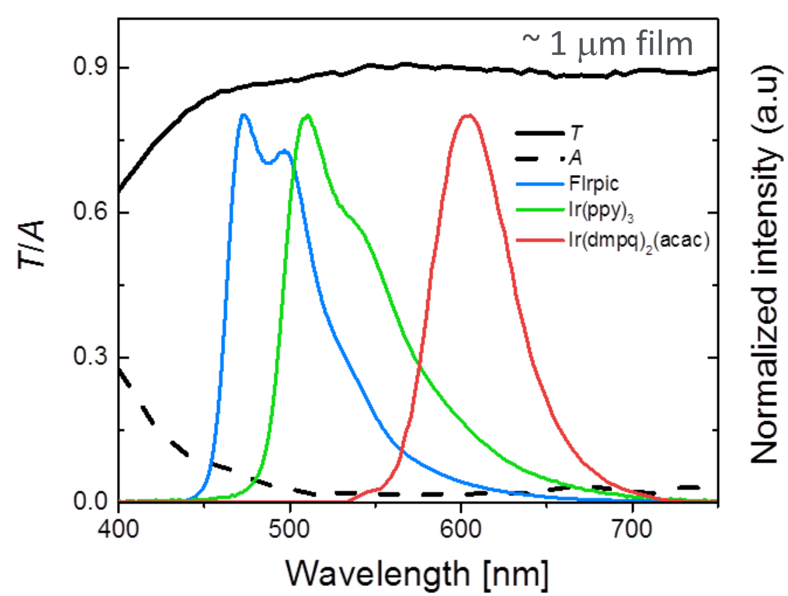


: polyamic acid (PAA) monomer



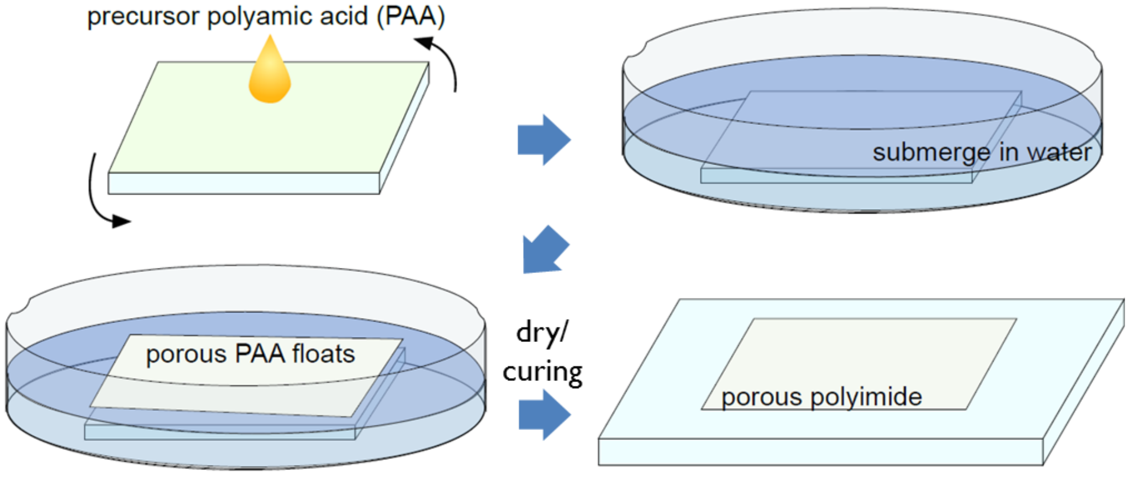
: Kapton® polyimide (PI) after imidization @ 360 °C





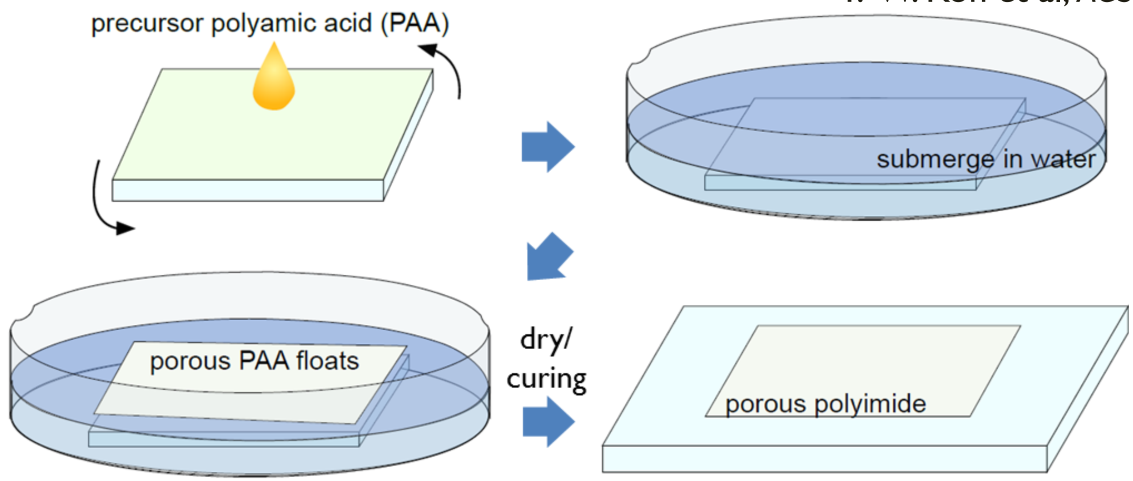
Phase inversion technique to introduce voids

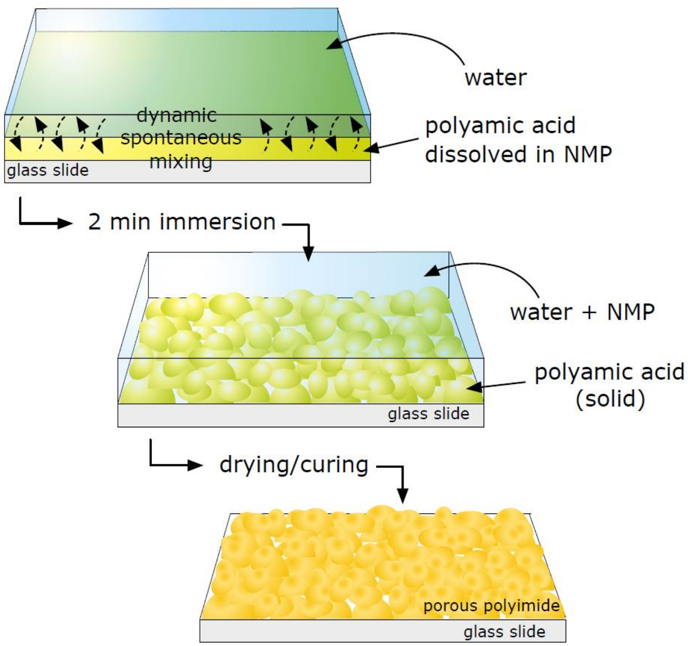
T.-W. Koh et al, *ACS Photonics* **2**, 1366 (2015)



Phase inversion technique to introduce voids

T.-W. Koh et al, *ACS Photonics* **2**, 1366 (2015)





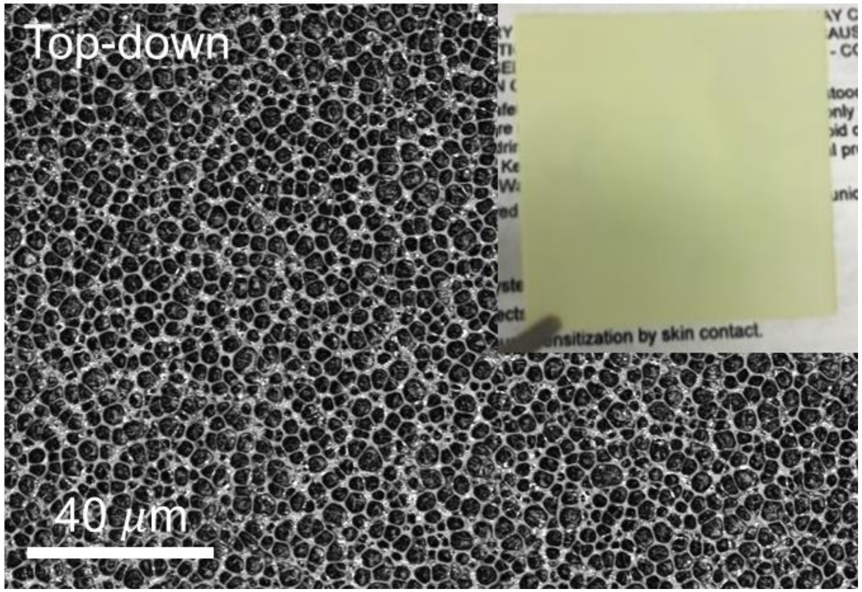
* Phase inversion process:

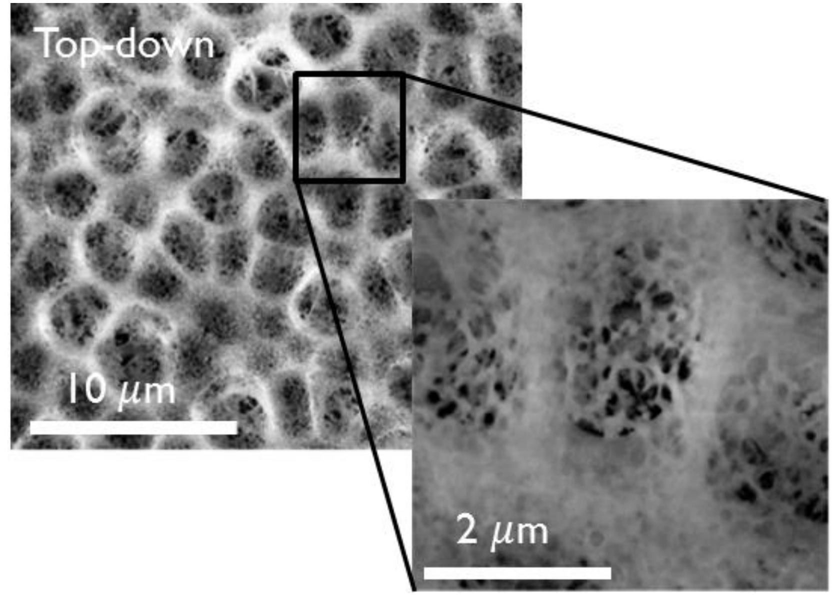
1) NMP (N-methyl-2-pyrollidone)
is miscible with water

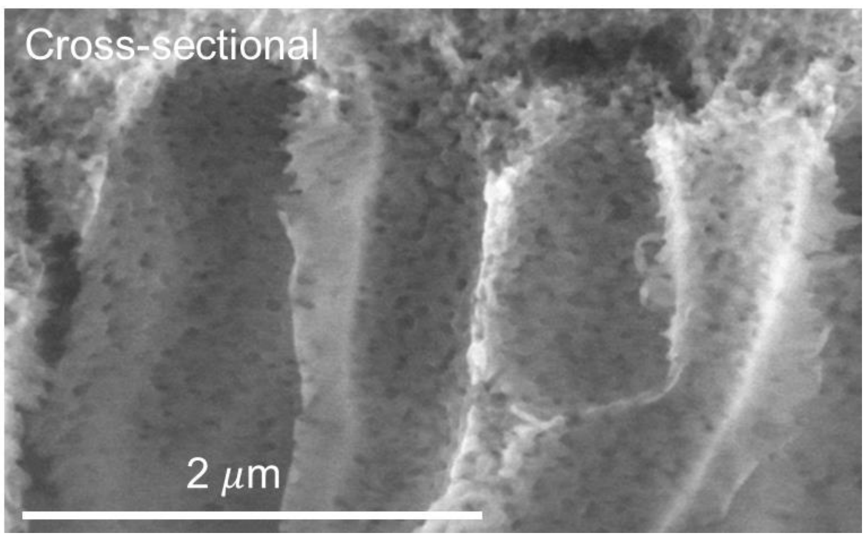
2) PAA dissolves in NMP, but not in water

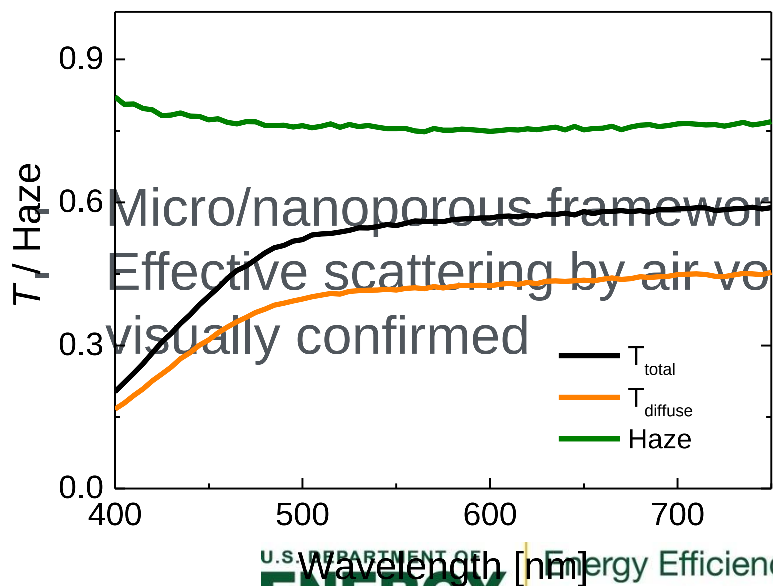
Dynamic, spontaneous void formation

Characterization of the porous PI films





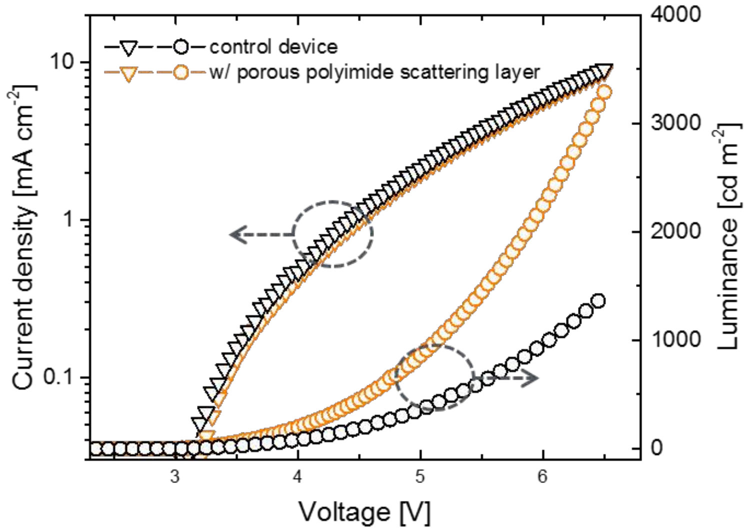


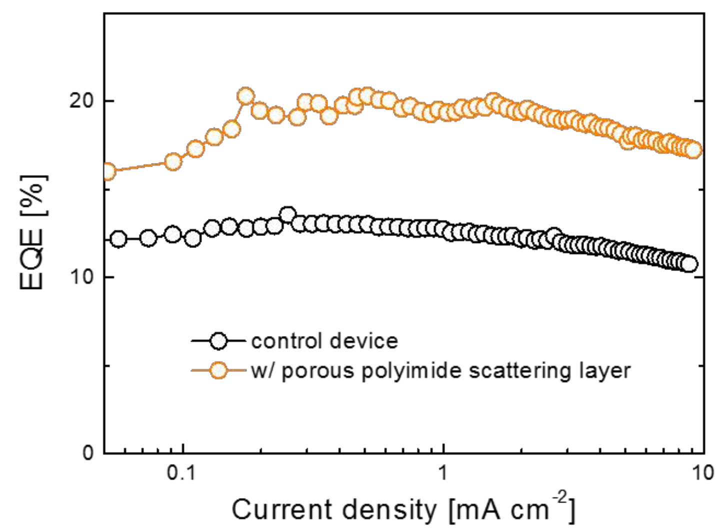


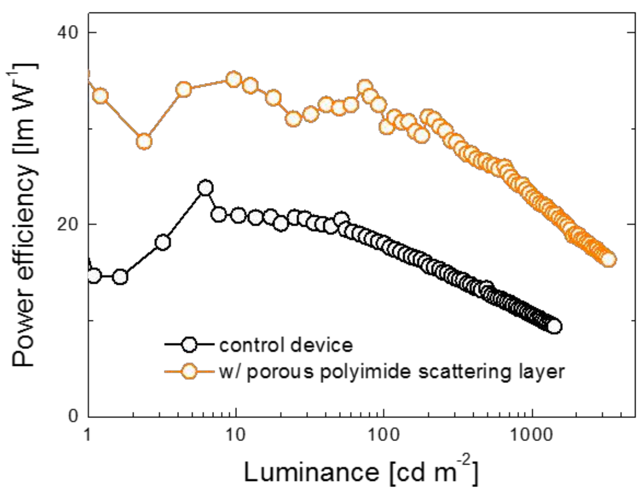
- Micro/nanoporous framework
- Effective scattering by air voids visually confirmed

T.-W. Koh et al, *ACS Photonics* **2**, 1366 (2015)

Porous PI layers on white OLEDs



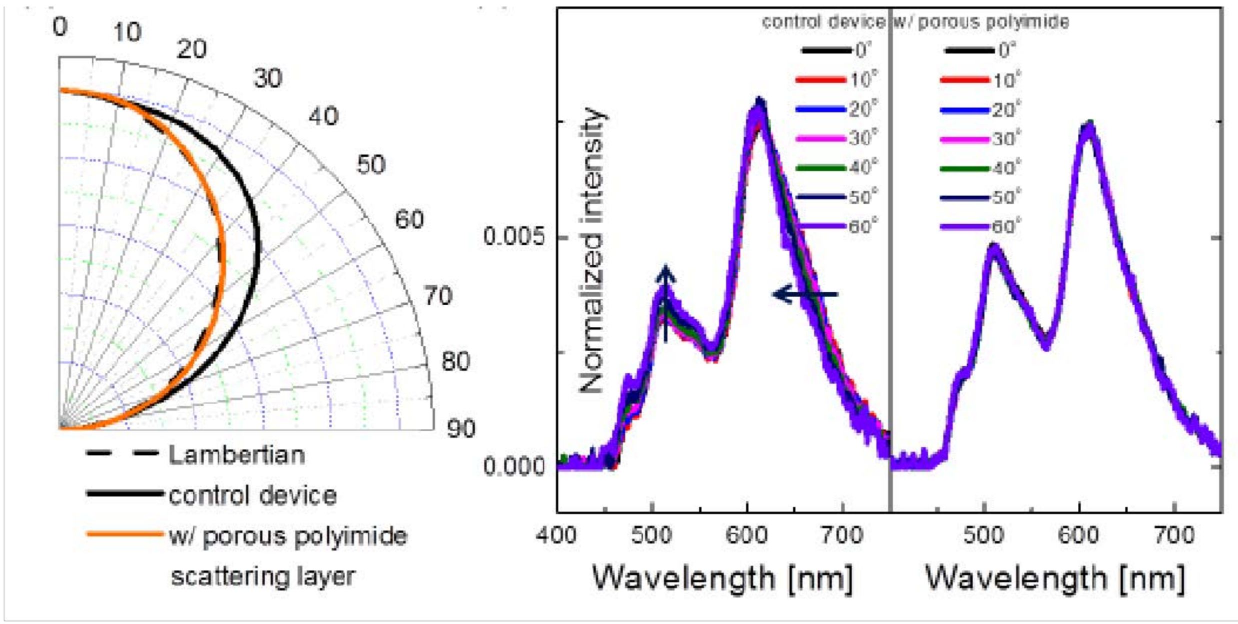


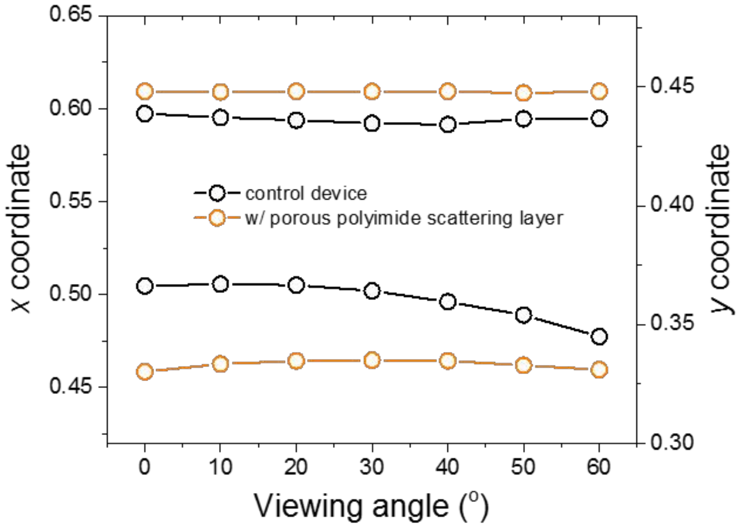


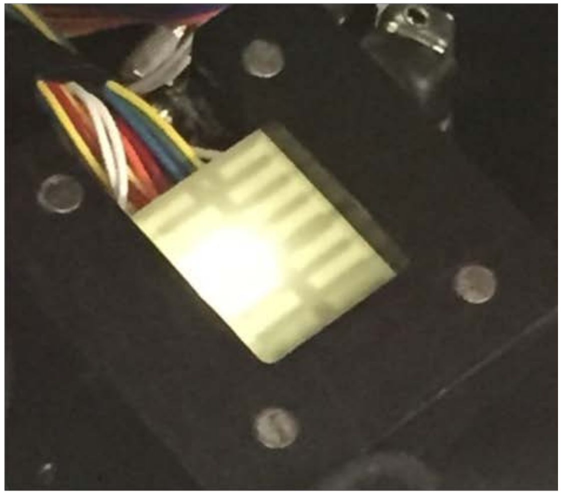
	EQE @ J = 3 mA/cm²	Power efficiency @ L = 100 cd/m²
control	11.9%	18.0 lm/W
with porous PI	19.0%	32.1 lm/W
	1.60x	**1.78x**

Porous PI layers on white OLEDs

T.-W. Koh et al, *ACS Photonics* **2**, 1366 (2015)

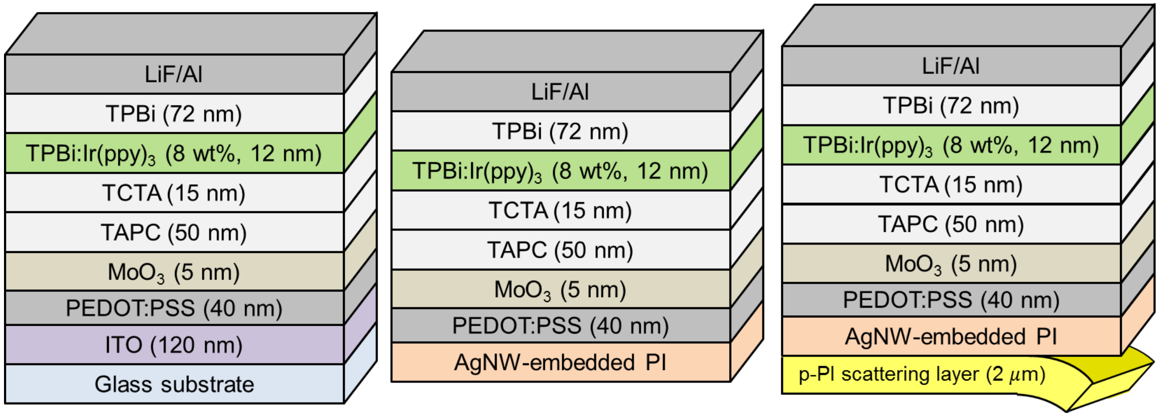


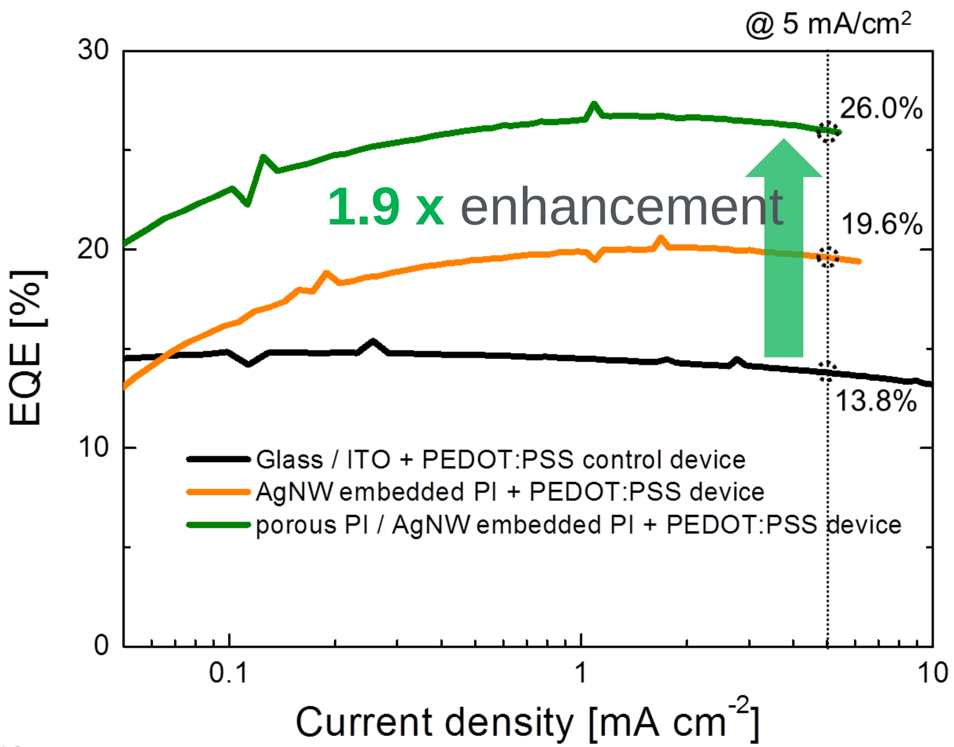


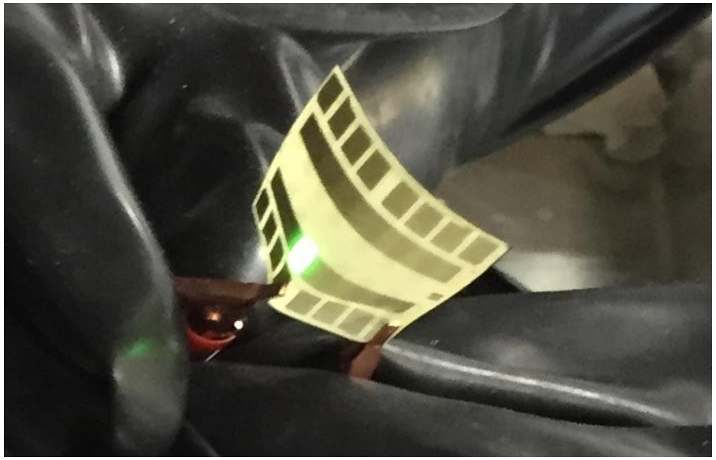


OLEDs on PI substrates + porous PI

J.A. Spechler et al, *Adv, Funct. Mater.* **25**, 7428 (2015)







U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

REFERENCE SLIDES

Project Budget

Project Budget: See below.
Variances: Due to TA assignments relieving the burden of student expenses, we were able to spend more funds on materials and supplies that were needed to complete the project.
Cost to Date: 67% of the budget has been expended thus far.
Additional Funding: No other funding sources

Budget History					
9/10/2014 – Project Year 1 (past)		Project Year 2 - FY 2016 (current)		Total	
DOE	Cost-share	DOE	Cost-share	DOE	Cost-share
$507,000	$126,278	$514,242	$129,382	$1,021,242	$204,195

Milestones, go/no-go decisions, and deliverables

Task #	Task or Subtask Title	Type	Number	Description (+ Go/No-Go Criteria)	Milestone Verification Process	Expected Month
1-3		Meeting		Kick-off meeting	Kick-off meeting held	1
1	Reducing substrate loss	Milestone	M1.1	Computational study of gas bubbles	Team will investigate various parameters including bubble size distribution, refractive index, substrate thickness and bubble density by designing a haze calculation model and a WOLED model	3
2	Reducing waveguiding loss	Milestone	M2.1	Modeling of optimized TCEs for large area applications	Team will investigate various parameters including wire length, diameter, density and junction resistance and their effect on the overall resistance, optical transmission, and power loss.	3
3.2	Corrugated back electrode	Milestone	M3.2.1	Modeling of randomly distributed features	Team will model the effect of randomly distributed feature size and density made by HCL using FEM	3
1.1	Gas bubbles via agitation	Milestone	M1.1.1	Bubbles introduced into substrate via agitation	Team will test multiple methods - shaking, ultrasonic, ball miller to create bubble-embedded plastic substrates for haze > 5% and RMS < 30 nm	6
1.1	Gas bubbles via laser ablation	Milestone	M1.2.1	Bubbles introduced into substrate via laser irradiation	Team will test different types of laser irradiation to introduce bubbles/defects into plastic substrates	6
3.2	Corrugated back electrode	Milestone	M3.2.2	Fabrication of randomly distributed features by HCL	Team will use HCL on top of WOLED layers to only propagate corrugation to the top metal electrode	6
2.1	Ag NW TCEs	Milestone	M2.1.1	Ag NW TCEs on PDMS	Team will spray coat AgNW inks on sacrificial glass substrate and process to reduce junction resistance before casting the PDMS layer on the network. Resistance and transmission properties will be compared to models as well as the results from task 2.2	9
2.2	Metal nanomesh	Milestone	M2.2.1	Nanomesh TCE on PDMS	Team will fabricate nanomesh electrodes on flexible substrates and compare their properties with those on rigid substrates. Electrode properties (T and sheet resistance) on flex to within 90% on rigid.	9
3.1	Plasmonic cavity	Milestone	M3.1.1	PlaCSH WOLED modeling and test devices	Team will model and test WOLEDs inside the PlaCSH cavity so as to be able to predict a sufficient enhancement in advance of M3.1	10

M3.2.2 stopped because of go/no-go M3.1

Milestones, go/no-go decisions, and deliverables

	Task #	Task or Subtask Title	Type	Number	Description (+ Go/No-Go Criteria)	Milestone Verification Process	Expected Month
✓	1.1	Gas bubbles via agitation	Milestone	M1.1.2	Optimized formation of bubbles via agitation	Team will find optimal processing conditions required to meet target properties of scattering substrate, targeting haze > 10% and RMS < 20 nm	11
✓	1.2	Gas bubbles via laser ablation	Milestone	M1.2.2	Optimized formation of bubbles via laser irradiation	Team will find optimal parameters required for laser irradiation that result in optimized scattering substrate targeting haze > 10% and RMS < 20 nm	11
✓	**1**	**Reducing substrate loss**	**Go/no-go decision point**	**M1.2**	**Extraction increase in WOLED > 1.5x**	**Team will fabricate and measure WOLEDs with scattering layers with extraction increase > 1.5x**	**12**
✓	**2**	**Reducing waveguiding loss**	**Go/no-go decision point**	**M2.2**	**Average roughness < 10 nm, FOM > 300, conductivity decrease < 20% after 1000x bending at R_c=1 cm**	**Team will characterize AgNW and MESH TCE for FOM, flexibility, surface roughness, identifying the approach to proceed forward.**	**12**
✓	**3**	**Reducing SPP loss**	**Go/no-go decision point**	**M3.1**	**Extraction increase higher than 1.5x**	**Team will fabricate and test WOLEDs w/ SPP reducing strategies; extraction increase > 1.5x**	**12**
✓	1	Reducing substrate loss	Deliverable	D1	WOLEDs on scattering substrates	Deliverable achieved	12
✓	1-3		Meeting		1st year review meeting	1st year review meeting held	13

Go/no-go summaries:

M1.2	M2.2	M3.1
• Gas bubble introduction via agitation: Go • Gas bubble introduction via laser irradiation: No-go	• Ag NW TCEs: Go • Metal nanomesh: Go	• Plasmonic nanocavity: Go • Corrugated back electrode: No-go

U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

Milestones, go/no-go decisions, and deliverables

Task #	Task or Subtask Title	Type	Number	Description (+ Go/No-Go Criteria)	Milestone Verification Process	Expected Month
4	WOLED extraction approach integration	Deliverable	D2	WOLEDs on flexible scattering substrates	Deliverable achieved	15
2	Reducing waveguiding loss	Milestone	M2.3	Optimized TCE on flexible scattering substrate	Team will combine scattering substrate and TCE to produce an integrated and optimal substrate/TCE structure	18
4.1	WOLEDs w/o substrate and waveguiding loss	Milestone	M4.1.1	WOLED with extraction increase > 2x w/o substrate and waveguiding loss	Team will integrate WOLEDs on scattering plastic flexible substrate with low-index TCEs to demonstrate extraction increase > 2x and angular variation in color $\Delta u'v' < 0.02$	18
4	WOLED extraction approach integration	Deliverable	D3	WOLEDs on optimized TCE on flexible scattering substrates	Deliverable achieved	19
4	WOLED extraction approach integration	Deliverable	D4	WOLEDs on optimized TCE on flexible scattering substrates with reduced SPP losses	Deliverable achieved	24
All		Meeting		Final review meeting	Final review meeting held	24
4.2	WOLEDs w/ full outcoupling strategy integration	Milestone	M4.2.1	$1cm^2$ WOLED w/ light extraction enhancement > 3x, CRI > 80	Team will fully integrate all the results obtained from tasks 1,2 and 3 to demonstrate a WOLED (> $1cm^2$) w/ extraction enhancement >3x compared to baseline, with CRI > 80 and $\Delta u'v' < 0.01$	24

U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy