Organic Electronics: Motivations, Status and Promises

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Organic electronics

- **Organic electronics**: field of materials science concerning the design, synthesis, characterization, and application of organic carbon-based small molecules or polymers that show desirable electronic properties such as energy gap and conductivity.
- **Thin film** technology, for flexible, large area, low cost, (opto)electronic applications
Organic electronics today

- Light emission; lighting; displays
- Light harvesting
- Flexible, large area electronics; logic; memory
- Sensing; bio-interfaces
- Field effect transistor
- Memory devices
- Photovoltaic cell
- Photodetectors
Outline

➢ Organic semiconductors
  • Quasi infinite choice of materials and energy levels
  • Basic considerations of electronic structure
  • Key enabling properties

➢ Light emission
  • Organic Light Emitting Diode: origin and evolution
  • Fundamental improvements
  • OLED displays and lighting

➢ Light harvesting
  • Organic Photovoltaic cell: key principles and materials
  • The non-fullerene acceptor revolution

➢ Electronics on plastics
  • Organic Field Effect Transistor: key principles and materials
  • Applications
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Organic semiconductors (OSC): quasi infinite space

S. Barlow et al., Handbook of Conducting Polymer (2018)
OSC devices: charge injection / extraction

- OSCs have extremely low intrinsic carrier densities and are essentially insulators.
- OSC devices function via charge carrier injection and extraction.
- Energy levels for carrier (electrons and holes) injection and transport, i.e. the frontier orbitals, are of paramount importance.

Standard generic OLED structure:

- Cathode
  - Electron transport layer
  - Blocking layer
  - Emission layer
- Hole transport layer
- Hole injection layer
- Anode

Lowest unoccupied molecular orbital (LUMO)

Low work function cathode

High work function anode

Band offset

Highest occupied molecular orbital (HOMO)
Conjugation and frontier energy levels in OSCs

**Benzene**

**Conjugated system has connected p-orbitals**, conventionally represented with alternating single and multiple (e.g. double) bonds

- **delocalization** of the electrons across all adjacent parallel aligned p–orbitals
- Increase stability; lower overall energy of the molecule

**Small molecules**

**Polymers**

**Pentacene**

**P(NDI$_2$OD-T$_2$)**

Energy gap: 1.5 – 3.5 eV
Mind the gap! What are we talking about?


**Single-particle (transport) gap vs. optical gap**

**Inorganic SC; 3D perovskites; ....**
- Exciton binding energy 10-20 meV
- Single particle gap ~ optical gap

**Organic SC; 2D perovskites (n=1, 2); ....**
- Exciton binding energy 0.3-1 eV
- Single particle gap > optical gap

\[
E_{B(\text{exciton})} = E_{g(\text{single particle})} - E_{g(\text{optical})}
\]

Measurement of single-particle (transport) levels

**Inverse Photoelectron Spectroscopy (IPES)**

- Binding Energy
  - $E_{\text{vacuum}}$
  - $E_{\text{Fermi}}$
  - $E_{\text{Vac}}$

- Occupied states
  - HOMO
  - LUMO

- Unoccupied states

- Measurement of single-particle (transport) levels:
  - He I, He II
  - $h\nu = 21.2, 40.8$ eV

**Ultraviolet Photoelectron Spectroscopy (UPS)**

- $e^-$
- Binding Energy
  - $E_{\text{Fermi}}$
  - $E_{\text{Vac}}$

- 5-15 eV
Composite UPS/IPES - complete electronic structure of ZnPc

Example of a film of the organic molecule zinc phthalocyanide (ZnPc)

OSCs: Key enabling properties

➢ Unlimited choice of organic (semiconductor) materials
   • Chemistry control ➔ molecular structure ➔ energy levels, photon absorption/emission, charge carrier transport, etc.

➢ Processibility: near room temperature film formation via vacuum or solution processing; printing; etc.
   • enables large area coverage
   • use of ultra-thin, flexible substrates (plastic) ➔ ultra-thin, flexible electronics

➢ Ability to stack/mix unlike materials
   • no constraint of lattice matching, no thermal budget ➔ unmatched flexibility in device engineering
   • Dopability (electrical; optical)

➢ Remarkable opto-electronic characteristics
   • Strong photo-emission and -absorption ➔ excellent conditions for efficient LEDs, solar cells, photodetectors
Unlimited choice of organic (semiconductor) materials
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OSCs: key enabling properties
Ionization energy (IE) and electron affinity (EA)

Molecular levels relevant to transport (via inter-molecular hopping) in the film and across interfaces
OSC: Key enabling properties

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Inorganic SC interfaces: impact of dangling bonds

- Inorganic semiconductor interfaces include defects and/or dangling bonds that give rise to electronically active electronic gap states.
  - Fermi level pinning, internal band bending, recombination centers, etc.
- Paramount importance of lattice/thermal matching; heterojunctions dominated by interface states
OSC interfaces: far more tolerant

- Closed-shell molecular structure and weak van der Waals intermolecular bonds: **no dangling bonds**
- Possible to stack heterogeneous structures, leading to unique opportunities for innovation in material, device structure design and processes!

**OLED structure**

**Property is a key enabler of organic electronics;** allows unmatched flexibility in mixing and stacking different organic materials, leading to complex functional, optimized structures for OLEDs, OPV cells, etc.
Enormous flexibility in device design

Ex.: Introduction of an electron blocker

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The organic light emitting diode (OLED)

The organic light emitting diode (OLED) was invented by C.W. Tang et al. in 1987. The structure includes ITO/glass, α-NPD, Alq₃, Mg:Ag (40:1), and a thickness of 1000 Å. The electron–hole recombination occurs in the electroluminescent layer.

Organic electroluminescent diodes

C. W. Tang and S. A. VanSlyke
Research Laboratories, Corporate Research Group, Eastman Kodak Company, Rochester, New York 14650

(Received 12 May 1987; accepted for publication 20 July 1987)

A novel electroluminescent device is constructed using organic materials as the emitting elements. The diode has a double-layer structure of organic thin films, prepared by vapor deposition. Efficient injection of holes and electrons is provided from an indium-tin-oxide...
Numerous fundamental and technological advances have contributed to the development of a mature technology over the past 30 years:

- High efficiency R, G, B emitters
- Electron and hole transport / blocking materials with proper energetics
- **Fundamental improvements in internal quantum efficiency**
- Doping of transport layers to improve conductivity
- Technological development of light out-coupling
- Encapsulation
- ..............
High efficiency **phosphorescent** OLED

OLED external quantum efficiency = \( \eta_{ext} = \eta_{int}\eta_{out} = \gamma \chi_{ST}\eta_{PL}\eta_{out} \)

- **\( \eta_{int} \)** = internal quantum efficiency
- **\( \eta_{out} \)** = outcoupling efficiency
- **\( \chi_{ST} \)** = spin formation ratio
- **\( \eta_{PL} \)** = quantum yield of the molecule

**Problem:** random formation of singlet and triplet excitons due to pairing of uncorrelated spins of injected electrons and holes. Statistically, only 25% of singlet excitons for 75% of triplet excitons, limiting considerably the efficiency of fluorescent OLEDs, which only utilize singlets

**Late 1990’s:** introduction of emissive phosphorescent dopants containing heavy metal atoms (Pt, Ir, Ru, Re), whose spin orbit coupling promotes singlet/triplet mixing nearly 100% energy transfer from both singlet and triplet states (M. Baldo et al., *Nature* 395, 151 (1998))

**Development of the phosphorescent OLED (PHOLED)**
High efficiency OLED: lifetime performance

<table>
<thead>
<tr>
<th>Color</th>
<th>PhOLEDs 1931 CIE coordinates</th>
<th>$T_{50}^*$ [h]</th>
<th>Fluorescent OLED 1931 CIE coordinates</th>
<th>$T_{50}^*$ [h]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Red</td>
<td>(0.64, 0.36)</td>
<td>900 000</td>
<td>(0.67, 0.33)</td>
<td>160 000</td>
</tr>
<tr>
<td>Green</td>
<td>(0.31, 0.63)</td>
<td>400 000</td>
<td>(0.31, 0.63)</td>
<td>200 000</td>
</tr>
<tr>
<td>Blue</td>
<td>&lt;100</td>
<td></td>
<td>(0.14, 0.12)</td>
<td>11 000</td>
</tr>
</tbody>
</table>

*T$_{50}$: time of decay to 50% of initial brightness

High efficiency OLED from delayed fluorescence

LED external quantum efficiency = $\eta_{ext} = \eta_{int}\eta_{out} = \gamma\chi_{ST}\eta_{PL}\eta_{out}$

- $\eta_{int}$ = internal quantum efficiency
- $\eta_{out}$ = outcoupling efficiency
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- $\eta_{PL}$ = quantum yield of the molecule

• **Problem:** random formation of singlet and triplet excitons due to pairing of uncorrelated spins of injected electrons and holes. Statistically, only 25% of singlet excitons for 75% of triplet excitons, limiting considerably the efficiency of fluorescent OLEDs, which only utilize singlets

• **2012:** introduction of a class of metal-free organic electroluminescent molecules in which the energy gap between singlet and triplet is minimized (10s meV) thereby promoting efficient spin up-conversion from the non-radiative triplet states to radiative singlet states

  (H. Uoyama et al., *Nature* 492, 234 (2012))

• Development of **OLEDs based on Thermally Activated Delayed Fluorescence (TADF)**
OLED displays are here to stay!

Flexible displays (Visionox, China)

LG grand display (Incheon Airport, S. Korea)

Apple

Samsung

Google
Active matrix (AM) OLED market share

OLED Display Forecasts 2017-2030 by Value $ Billions

“Flexible, Printed OLED Displays 2020-2030: Forecasts, Markets, Technologies”
By Raghu Das and Xiaoxi He
Lighting, evolved

• compared to 6-8% today, solid-state lighting is expected to comprise 86% of installations by 2035

• energy savings equivalent to 45 million US homes today

• cumulative energy savings represent $630B

• thin-film solid-state lighting is poised to contribute to this mix

Courtesy of Barry Rand, Princeton
White organic light emitting diode (WOLED)

Three phosphor WOLED blended in a single thin (9 nm) wide energy gap host (UGH2)

Triple phosphor / triple host WOLED
Sun et al., Org. Electron. 9, 994 (2008)
Organic lighting

Konika Minolta

Novaled AG
OLED lighting: lifetimes and other performance marks

<table>
<thead>
<tr>
<th>Particulars</th>
<th>OLED</th>
<th>LED</th>
<th>Fluorescent Lamp</th>
<th>Compact Fluorescent Lamp</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lighting technology</td>
<td>Fourth generation lighting system</td>
<td>Fourth generation lighting system (Solid state lighting)</td>
<td>Third generation lighting system (1938)</td>
<td>Third generation lighting system (1976)</td>
</tr>
<tr>
<td>Types</td>
<td>(Solid state lighting)</td>
<td>Point source. Requires optical diffusion to protect occupants from glare by a bright light source. Generates heat. Thermal management system mandatory</td>
<td>Point – Area light source.</td>
<td>Point – Area light source.</td>
</tr>
<tr>
<td>Flexibility</td>
<td>Yes</td>
<td>None. Flexible LED strips are possible.</td>
<td>None</td>
<td>None</td>
</tr>
<tr>
<td>Efficiency</td>
<td>~60–90 lm/W</td>
<td>65–160 lm/W</td>
<td>&gt;80 lm/W</td>
<td>50–70 lm/W</td>
</tr>
<tr>
<td>Color rendering index (CRI)</td>
<td>80–90</td>
<td>&gt;65–95</td>
<td>&gt;62–80</td>
<td>about 80</td>
</tr>
<tr>
<td>Color temperature</td>
<td>2700–5,400 K</td>
<td>2700–10,000 K</td>
<td>2700–6000 K</td>
<td>2700–6000 K</td>
</tr>
<tr>
<td>Mercury</td>
<td>No</td>
<td>No</td>
<td>Yes. Contain toxic mercury</td>
<td>Yes. Contain toxic mercury</td>
</tr>
<tr>
<td>Infrared radiation</td>
<td>No</td>
<td>No</td>
<td>Yes. A small amount of Infrared (&gt;700 nm) radiation</td>
<td>Yes. A small amount of infrared (&gt;700 nm) radiation</td>
</tr>
<tr>
<td>UV radiations</td>
<td>No</td>
<td>No</td>
<td>Yes. Very low levels of UVB (280–315 nm), UVA (315–400 nm) radiations</td>
<td>Yes. Very low levels of UVB (280–315 nm), UVA (315–400 nm) radiations</td>
</tr>
<tr>
<td>Cost</td>
<td>Expensive. Lower cost in the future</td>
<td>Upfront cost relatively higher compared to CFL and fluorescent lamps</td>
<td>Affordable. Widely adopted</td>
<td>Affordable. Widely adopted.</td>
</tr>
<tr>
<td>Dimming</td>
<td>Dimmable when paired with dimming drivers</td>
<td>Very easy to dim</td>
<td>Often not suitable for dimming</td>
<td>CFL bulbs can be dimmed.</td>
</tr>
<tr>
<td>Response time</td>
<td>Much faster about 10 ms (0.01 ms)</td>
<td>Fraction of a second (1/10th of ms)</td>
<td>Slight delay on turn-on and turn off.</td>
<td>Slight delay on turn-on and turn off.</td>
</tr>
<tr>
<td><strong>Life time</strong></td>
<td>5000–10,000 h. Expecting lifespan of 10,000 h at 100% brightness or 40,000 h at 25% brightness</td>
<td>50,000 to 100,000 h</td>
<td>7000–15,000 h</td>
<td>Rated service life of 6000–15,000 h</td>
</tr>
<tr>
<td>Acoustic noise</td>
<td>None</td>
<td>None</td>
<td>Ballast can make a buzzing sound</td>
<td>Buzzing sound is caused by a hum in the ballast</td>
</tr>
</tbody>
</table>
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- Fundamental improvements
- OLED displays and lighting

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- The non-fullerene acceptor revolution

Electronics on plastics
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- Applications
Two-layer organic photovoltaic cell

C. W. Tang  
*Research Laboratories, Eastman Kodak Company, Rochester, New York 14650*

APL 48, 183 (1986)  
(Received 28 August 1985; accepted for publication 31 October 1985)

A thin-film, two-layer organic photovoltaic cell has been fabricated from copper phthalocyanine and a perylene tetracarboxylic derivative. A power conversion efficiency of about 1% has been achieved under simulated AM2 illumination. A small feature of the device is that the...
Excitons: inorganic vs. organic PV cell

**Inorganic SC**
- **Wannier exciton** (typical of inorganic semiconductors)
- **Exciton binding energy** < \( kT \)
- \( \rightarrow \) dissociation in both diffusion and space charge regions

**Organic**
- **Frenkel exciton** (typical of organic materials)
- **Exciton binding energy** >> \( kT \)
- \( \rightarrow \) requires a donor/acceptor (D/A) type of structure

**Inorganic SC Picture**
- CONDUCTION BAND
- VALENCE BAND
- GROUND STATE
- WANNIER EXCITON
- Binding energy \(~10\text{meV}\)
- Radius \(~100\text{Å}\)

**Organic Picture**
- MOLECULAR PICTURE
- GROUND STATE
- FRENKEL EXCITON
- Binding energy \(~1\text{eV}\)
- Radius \(~10\text{Å}\)
Photocurrent generation at an organic heterojunction

\[ \eta_{EQE} = \eta_A \times \eta_{ED} \times \eta_{CT} \times \eta_{CC} \]

- \( \eta_A \) \approx 50\% – 100\%
- \( \eta_{ED} \) \approx 10\% – 50\%
- \( \eta_{CT} < 100\%
- \( \eta_{CC} \approx 100\%

Important for exciton dissociation process (although physics should be described in terms of excitonic states)

Maximizing LUMO_A-HOMO_D gap crucial to maintain large \( V_{OC} \)
Origin of the short circuit current, $J_{sc}$

$J_{sc}$ related to:
- number of absorbed photons $\rightarrow$ excitons
- conversion of exciton into $e^-$ and $h^+$
- Efficiency of $e^-/h^+$ transport to, and collection at, electrodes

Necessary to dissociate excitons at the D/A heterojunction

Exciton diffusion length: 10 – 50 nm, depending on material

Planar heterojunction e.g. CuPc/C$_{60}$

- More control on structure
- Simpler; better conduction path
- Amenable to doping
- Trade-off between thickness (photon absorption) and exciton diffusion length

Bulk heterojunction e.g. P3HT:PCBM

- Higher interface area
- Shorter exciton diffusion path
- Internal structure more difficult to control
- Connectivity/conduction path

Exciton diffusion length: $10^{-5}$ – $50$ nm, depending on material

Poly(3-hexylthiophene) (P3HT)

[6,6]-phenyl C$_{61}$ butyric acid methyl ester (PCBM)
Typical OPV materials

Donors
- Pentacene
- SubP
- SubNc
- MePc
- P3HT
- PBTTT-C_{12}
- PCDTBT
- MEH-PPV

Acceptors (OPV acceptors dominated by fullerene derivatives till ~ 2015)
- PTCBI
- C_{60}
- C_{70}
Anatomy of a typical OPV cell

Glass, plastic foil

Carrier extraction/selective layer

Polymer-based (solution processed), small molecule-based (solution or vacuum)

Doped organic films (PEDOT:PSS), metal oxides (TiO$_x$, ZnO, MoO$_x$, etc.); carrier extraction/selective layer

TCOs, thin metals, carbon, polymers, Ag nanowires
Rise of the non-fullerene acceptors (NFA) .... or the rebirth of the OPV field.

Advantages of NFAs over fullerene counterparts

- Stronger absorption in the visible $\rightarrow$ more active participation in current generation
- Much larger variability of electronic properties, $\rightarrow$ ability to tune energy levels by chemical means for better match with the donor
- Better stability of film morphology in bulk heterojunctions.

2015 conception and development of a class of non-fullerene acceptors termed “fused ring electron acceptors (FREA), with ITIC as the benchmark molecule.

Zhong et al., Nat. Comm. 6, 8242 (2015)
Photovoltaic conversion efficiencies vs. t
Organic photovoltaics: niche to main stream application

Heliatek

Konarka (defunct)

infinityPV

ORENge
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The organic field effect transistor (OFET)

- OFET typically operates in accumulation, with the channel right next to the dielectric interface.

The organic field effect transistor (OFET)

- **Small molecules**: best electron and hole mobility well above 5 cm²/V.s
- **Polymers**: hole and electron mobility above 2 cm²/V.s
- All solution processing; Flexible substrates
- @ few μA, OFET life-times above 100,000 hours
- Backplanes with half-million printed OFETs have been realized
- Increasing panoply of electron- and hole-transport semiconductors → complementary n- and p-channel OFETs

Vacuum-deposited **hole**-transport small molecules

- Pentacene
- 6T
- DNTT

Solution-deposited **hole**-transport small molecules

- TIPS-Pentacene

Vacuum-deposited **electron**-transport small molecules

- C₆₀
- PTCDI-C₈H₁₇

Solution-deposited **electron**-transport polymers

- Vacuum-deposited hole-transport small molecules
Flexible electronics: OFET on plastic

Circuits built on < 1 μm tick foil; sustains folding to radius of < 5 μm

Printed circuit on flexible plastic substrate

Ultra-light

Applications in wearable health-care monitoring systems

Bionic skin; sensing; robotics

Courtesy from T. Someya, Univ. of Tokyo.
.... a bright future for organic electronics

➢ Thirty years of high-level research on the unique physical and (opto)electronic properties of organic semiconductors and their interfaces

➢ Continuous flow of newly synthesized molecules and polymers

➢ Unique processing flexibility offered by organic semiconductors

➢ Ever expanding demand for high-resolution and inexpensive displays, flexible and wearable electronics and sensors, and large area solar energy conversion

➢ With continued innovation and strong link between research and development, the sky is the limit .....
Complexity of the charge separation mechanism

Exciton → charge transfer → charge separation

B. Kippelen & JLB, Energy & Environmental Science 2, 251 (2009)
J. Durrant et al., Chem. Rev. (2009)

Courtesy of J.-L. Brédas, Georgia Tech
Ionization energy (IE) and electron affinity (EA)

Molecular levels relevant to transport (via inter-molecular hopping) in the film and across interfaces

LUMO

HOMO
Some powerful n- and p-dopants

High electron affinity oxidizing molecules
p-dopants

Low ionization energy reducing molecules
n-dopants

Molecular levels relevant to transport (via inter-molecular hopping) in the film and across interfaces
Hopping transport in disordered solids

Energetic distribution due to static and dynamic structural disorder

The jump rate between site \(i\) and \(j\) is described by the Miller-Abrahams relation where

\[
\Delta E_{ij} = E_j - E_i + (field) \cdot r_{ij}
\]

\(E_i\) and \(E_j\) are the carrier energies on sites \(i\) and \(j\)
\(\Gamma\): inter-molecular coupling coefficient, inverse localization length (localization length \(\sim 10^{-8}\) cm)
\(\nu_0\) is the attempt frequency, estimated as \(\sim\) phonon frequency for a thermally activated process \((10^{12}\) s\(^{-1}\))
\(r_{ij}\) is the distance between localized states \(i\) and \(j\)
\(\theta = 1/kT\)

A. Miller and E. Abrahams, Phys. Rev. 120, 745 (1960)
Unlimited choice of semiconductor parameters

**Polyacenes**

- **Benzene**: 2550 Å (4.86 eV)
- **Naphthalene**: 3150 Å (3.93 eV)
- **Anthracene**: 3800 Å (3.2 eV)
- **Naphthacene or Tetracene**: 4800 Å (2.6 eV)
- **Pentacene**: 5800 Å (2.13 eV)

**Contorted hexabenzocoronene (cHBC) + halogenated derivatives**

Where modern organic electronics started

THE 2011 WOLF PRIZE IN CHEMISTRY

Ching W. Tang
University of Rochester

Two-layer organic photovoltaic cell

C. W. Tang
Research Laboratories, Eastman Kodak Company, Rochester, New York 14650

(Received 28 August 1985; accepted for publication 31 October 1985)

A thin-film, two-layer organic photovoltaic cell has been fabricated from copper phthalocyanine and a perylene tetracarboxylic derivative. A power conversion efficiency of about 1% has been achieved under illumination with light of wavelength 620 nm and intensity 1 milliwatt per square centimeter.

Organic electroluminescent diodes

C. W. Tang and S. A. VanSlyke
Research Laboratories, Corporate Research Group, Eastman Kodak Company, Rochester, New York 14650

(Received 12 May 1987; accepted for publication 20 July 1987)

A novel electroluminescent device is constructed using organic materials as the emitting elements. The diode has a double-layer structure of organic thin films, prepared by vapor deposition. Efficient injection of holes and electrons is provided from an indium-tin-oxide...
Characteristics of a hole-channel DNTT OFET

- Recall: in Si MOSFET, threshold voltage defined as the minimum gate-source voltage necessary to bring the SC in strong inversion

- OFETs do not operate in inversion, so strictly speaking, $V_T$ cannot be defined. However, still very useful concept, since it is the minimum gate-source voltage required to obtain an appreciable drain current

- Switch-on voltage $V_{SO}$ is $V_{GS}$ when $I_D$ reaches minimum ($\sim -1$ V here)