Materials for Polymer Solar Cells: Achievements and Challenges

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Bulk Heterojunction Solar Cell

- **Anode (e.g., ITO)**
- **Interfacial Layer**
- **Active Layer**
- **Cathode (e.g., Ca/Al)**
- **Anode (e.g., ITO)**
I. Past: what we have accomplished? Rational design of conjugated polymers (or molecular engineering)
   a) Backbone
   b) Side chains
   c) Substituents

II. Current: where we stand now?
   a) “F” impact
   b) Orientation at the D/A interface

III. Future: what we could do or should do?
   a) $J_{sc}$
   b) $V_{oc}$
   c) Stability
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Three Constituents of Conjugated Polymers

- Side chains
- Backbone repeating unit
- Substituent
I. Rational Design of Conjugated Polymers

a) Backbone:
   • Weak Donor – Strong Acceptor

b) Side Chains
   • Position
   • Shape and size

c) Substituents
   • The curious case of fluorine
a) The Backbone: Balancing the $V_{oc}$ and $J_{sc}$


**“Weak Donor – Strong Acceptor” Polymers**

<table>
<thead>
<tr>
<th>Polymer</th>
<th>HOMO (eV)</th>
<th>LUMO (eV)</th>
<th>$E_{\text{gap}}$ (opt)</th>
<th>$V_{\text{oc}}$ (V)</th>
<th>$J_{\text{sc}}$ (mA/cm$^2$)</th>
<th>FF</th>
<th>$\eta$ %</th>
<th>Refer.</th>
</tr>
</thead>
<tbody>
<tr>
<td>PBnDT-DTffBT</td>
<td>−5.54</td>
<td>−3.33</td>
<td>1.7</td>
<td>0.89</td>
<td>12.8</td>
<td>0.62</td>
<td>7.2</td>
<td><em>ACIE 2011, 50, 2995.</em></td>
</tr>
<tr>
<td>PBnDT-FTAZ</td>
<td>−5.36</td>
<td>−3.36</td>
<td>2.0</td>
<td>0.79</td>
<td>12.45</td>
<td>0.72</td>
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<td><em>JACS 2011, 133, 4625.</em></td>
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<td>PBnDT-DTPyT</td>
<td>−5.47</td>
<td>−3.44</td>
<td>1.51</td>
<td>0.85</td>
<td>12.78</td>
<td>0.58</td>
<td>6.3</td>
<td><em>ACIE 2010, 49, 7992.</em></td>
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<td>PNDT-DTPyT</td>
<td>−5.36</td>
<td>−3.42</td>
<td>1.53</td>
<td>0.71</td>
<td>14.2</td>
<td>0.62</td>
<td>6.2</td>
<td><em>ACIE 2010, 49, 7992l</em></td>
</tr>
</tbody>
</table>

*among "the hottest research of 2011“ by Thomson Reuters*
b) The Side Chains: Position Matters!
Better Solubility Leads to High $M_w$

$M_n = 30$ kg/mol  
$M_w = 92$ kg/mol

$M_n = 37$ kg/mol  
$M_w = 84$ kg/mol

$M_n = 27$ kg/mol  
$M_w = 54$ kg/mol

It Works!

Polymer | Polymer: PCBM | Thickness (nm) | $V_{oc}$ (V) | $J_{sc}$ (mA/cm²) | FF | $\eta$ (%) | Mobility (cm²/V·s)
---|---|---|---|---|---|---|---
PBTD-4DTBT (3% diiodooctane) | 1:1 | 95 | 0.67 | 6.38 | 50.79% | **2.17%** | $1.60 \times 10^{-5}$
PBTD-4DTBT | 1:1 | 100 | 0.75 | 5.92 | 41.27% | 1.83% | $9.20 \times 10^{-6}$
PBTD-3DTBT | 1:1 | 85 | 0.89 | 0.94 | 24.74% | 0.21% |
PBTD-DTsolBT | 1:1 | 80 | 0.43 | 0.12 | 26.35% | 0.01% |
PBTD-DTBT | 1:1 | 65 | 0.55 | 3.53 | 36.8% | **0.72%** | $3.94 \times 10^{-6}$
PBTD-BT | 1:3 | 55 | 0.72 | 2.06 | 42% | **0.60%** | $4.21 \times 10^{-6}$
b) The Side Chains: Size and Branching are NOT “Trivial”

Long branched side chains weaken intermolecular interactions, leading to an increased $V_{oc}$ but a much lower $J_{sc}$

## Quantitative Analysis

<table>
<thead>
<tr>
<th>Polymer</th>
<th>$J_s$ (mA/cm²)</th>
<th>$J_{so}$ (mA/cm²)</th>
<th>n</th>
<th>$\frac{n k T}{q} \ln\left(\frac{J_{sc}}{J_{so}}\right)$</th>
<th>HOMO (eV)</th>
<th>$\frac{\Delta E_{DA}}{2q}$</th>
<th>$V_{oc}$ (V) Cal</th>
<th>$V_{oc}$ (V) Exp</th>
<th>$J_{sc}$ Cal (mA/cm²)</th>
<th>$J_{sc}$ Exp (mA/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C10,6–C8</td>
<td>1.80 E–04</td>
<td>148</td>
<td>2.16</td>
<td>−0.16</td>
<td>−5.32</td>
<td>0.76</td>
<td>0.60</td>
<td>0.59</td>
<td>7.05</td>
<td>7.98</td>
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<tr>
<td>C10,6–C6,2</td>
<td>8.09 E–03</td>
<td>3.38</td>
<td>4.90</td>
<td>0.06</td>
<td>−5.33</td>
<td>0.77</td>
<td>0.83</td>
<td>0.81</td>
<td>5.02</td>
<td>5.62</td>
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<tr>
<td>C8–C8</td>
<td>2.32 E–02</td>
<td>399</td>
<td>2.64</td>
<td>−0.28</td>
<td>−5.13</td>
<td>0.67</td>
<td>0.39</td>
<td>0.41</td>
<td>9.75</td>
<td>6.97</td>
</tr>
<tr>
<td>C8–C12</td>
<td>3.78 E–04</td>
<td>254</td>
<td>2.12</td>
<td>−0.21</td>
<td>−5.27</td>
<td>0.74</td>
<td>0.53</td>
<td>0.52</td>
<td>5.22</td>
<td>5.88</td>
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<tr>
<td>C8–C6,2</td>
<td>7.42 E–03</td>
<td>68.8</td>
<td>3.17</td>
<td>−0.15</td>
<td>−5.30</td>
<td>0.75</td>
<td>0.60</td>
<td>0.59</td>
<td>10.04</td>
<td>10.93</td>
</tr>
<tr>
<td>C6,2–C6,2</td>
<td>4.69 E–03</td>
<td>22.6</td>
<td>3.51</td>
<td>−0.07</td>
<td>−5.34</td>
<td>0.77</td>
<td>0.70</td>
<td>0.69</td>
<td>9.58</td>
<td>10.67</td>
</tr>
</tbody>
</table>

\[ J_s = J_{so} \exp\left(\frac{-\Delta E_{DA}}{2nkT}\right) \]

\[ V_{oc} \approx \frac{n k T}{q} \ln\left(\frac{J_{sc}}{J_{so}}\right) + \frac{\Delta E_{DA}}{2q} \]
c) The Substituents: Fine-Tuning


I. Past: what we have accomplished? Rational design of conjugated polymers (or molecular engineering)
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II. Current: where we stand now?
   a) “F” impact
   b) Orientation at the D/A interface

III. Future: what we could do or should do?
   a) \( J_{sc} \)
   b) \( V_{oc} \)
   c) Stability
a) Understanding the F impact

- 3 Polymers with varied F substitution
- Minimized other variables
- 3 different thicknesses to vary transport length
- Observe trends in $J_{sc}$, $V_{oc}$ and $FF$

$X_1=H, X_2=H$: PBnDT-DTBT
$X_1=H, X_2=F$: PBnDT-DTfBT
$X_1=F, X_2=F$: PBnDT-DTffBT
$R_1 = 3$-butynonyl
$R_2 = 2$-ethylhexyl
UV-Vis Absorption

Absorption Coefficient (cm$^{-1}$)

Wavelength (nm)

DTBT
DTfBT
DTffBT

Normalized Fluorescence (A.U.)
PV Properties at Three Thicknesses

- **(a)**: Dependence of $V_{oc}$ (V) on Thickness (nm) for DTBT, DTfBT, and DTffBT.
- **(b)**: Dependence of $J_{sc}$ (mA/cm²) on Thickness (nm) for DTBT, DTfBT, and DTffBT.
- **(c)**: Dependence of FF (%) on Thickness (nm) for DTBT, DTfBT, and DTffBT.
- **(d)**: Dependence of $\eta$ (%) on Thickness (nm) for DTBT, DTfBT, and DTffBT.
(i) More F, Suppressed Bimolecular Recombination

\[ J_{\text{photo}} = \beta \left( P_{\text{light}} \right)^\alpha \]
“True” Conformation of DTBT, etc.
(ii) More Fluorines, Large $\Delta \mu_{ge}$: hindered geminate?

<table>
<thead>
<tr>
<th>Repeat unit</th>
<th>$\mu_g$ (Debye)</th>
<th>$\mu_e$ (Debye)</th>
<th>$\Delta \mu_{ge}$ (Debye)</th>
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</thead>
<tbody>
<tr>
<td>BnDT-DTBT</td>
<td>1.54</td>
<td>12.10</td>
<td>11.20</td>
</tr>
<tr>
<td>BnDT-DTfBT</td>
<td>1.49</td>
<td>14.10</td>
<td>15.18</td>
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<tr>
<td>BnDT-DTffBT</td>
<td>0.75</td>
<td>15.50</td>
<td>16.02</td>
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</tbody>
</table>
(iii) F Substitution Improves the Structural Order

In collaboration with John Tumbleston, Harald Ade
b) Two Different Types of “Edge on” vs. “Face on”

Courtesy of John Tumbleston and Harald Ade
Orientation can be engineered through Materials Chemistry, and choice of solvent for processing.

Courtesy of John Tumbleston and Harald Ade.
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     a) $J_{sc}$
     b) $V_{oc}$
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a) Can We Get Higher $J_{sc}$?

\[ \text{Band gap (eV)} \]

\[ J_{sc} \text{ (mA/cm}^2\text{)} \]

**a) Choosing appropriate structural units in D-A polymers**

**b) Increasing Full Width Half Maximum (FWHM)**

**c) Making n-type material absorb**

**d) Improving EQE**
<table>
<thead>
<tr>
<th>Classification</th>
<th>Effic. (%)</th>
<th>Area (cm²)</th>
<th>J(SC) (mA/cm²)</th>
<th>FF (%)</th>
<th>Test centre (and date)</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silicon</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si (crystalline)</td>
<td>25.0 ± 0.5</td>
<td>4.00</td>
<td>0.706</td>
<td>82.8</td>
<td>Sandia (3/99)</td>
<td>UNSW PERL [18]</td>
</tr>
<tr>
<td>Si (multicrystalline)</td>
<td>20.4 ± 0.5</td>
<td>1.002</td>
<td>0.664</td>
<td>80.9</td>
<td>NREL (5/04)</td>
<td>FhG-ISE [19]</td>
</tr>
<tr>
<td>Si (thin film transfer)</td>
<td>19.1 ± 0.4</td>
<td>3.983</td>
<td>0.650</td>
<td>77.6</td>
<td>ISFH (43-μm thick)</td>
<td></td>
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<tr>
<td>Si (thin film submodule)</td>
<td>10.5 ± 0.3</td>
<td>94.0</td>
<td>0.492</td>
<td>72.1</td>
<td>FhG-ISE (8/07)</td>
<td>CSG Solar (1-2 μm on glass; 20 cells) [21]</td>
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<tr>
<td>III-V cells</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>GaAs (thin film)</td>
<td>28.3 ± 0.8</td>
<td>0.9944</td>
<td>1.107</td>
<td>29.47</td>
<td>NREL (8/11)</td>
<td>Alta devices [3]</td>
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<tr>
<td>GaAs (multicrystalline)</td>
<td>18.4 ± 0.5</td>
<td>4.011</td>
<td>0.994</td>
<td>23.2</td>
<td>NREL (11/95)</td>
<td>RTI, Ge substrate [22]</td>
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<tr>
<td>InP (crystalline)</td>
<td>22.1 ± 0.7</td>
<td>4.02</td>
<td>0.878</td>
<td>29.5</td>
<td>NREL (4/90)</td>
<td>Spire, epitaxial [23]</td>
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<tr>
<td>Thin film chalcogenide</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CIGS (cell)</td>
<td>19.6 ± 0.6</td>
<td>0.996</td>
<td>0.713</td>
<td>34.8</td>
<td>NREL (4/09)</td>
<td>NREL, CIGS on glass [24]</td>
</tr>
<tr>
<td>CIGS (submodule)</td>
<td>17.4 ± 0.5</td>
<td>15.993</td>
<td>0.6815</td>
<td>33.84</td>
<td>FhG-ISE (10/11)</td>
<td>Solibro, four serial cells [4]</td>
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<tr>
<td>CdTe (cell)</td>
<td>16.7 ± 0.5</td>
<td>1.032</td>
<td>0.845</td>
<td>26.1</td>
<td>NREL (9/01)</td>
<td>NREL, mesa on glass [25]</td>
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<tr>
<td>Amorphous/nanocrystalline Si</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>Si (amorphous)</td>
<td>10.1 ± 0.3</td>
<td>1.036</td>
<td>0.886</td>
<td>16.75</td>
<td>NREL (7/09)</td>
<td>Oerlikon Solar Lab, Neuchatel [26]</td>
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<tr>
<td>Si (nanocrystalline)</td>
<td>10.1 ± 0.2</td>
<td>1.199</td>
<td>0.539</td>
<td>24.4</td>
<td>JQA (12/97)</td>
<td>Kaneka (2 μm on glass) [27]</td>
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<td>Photochemical</td>
<td></td>
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<td></td>
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<tr>
<td>Dye sensitised</td>
<td>11.0 ± 0.3</td>
<td>1.007</td>
<td>0.714</td>
<td>21.93</td>
<td>70.3</td>
<td>AIST (9/11)</td>
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<tr>
<td>Dye sensitised (submodule)</td>
<td>9.9 ± 0.4</td>
<td>17.11</td>
<td>0.719</td>
<td>19.4</td>
<td>71.4</td>
<td>AIST (8/10)</td>
</tr>
<tr>
<td>Organic</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Organic thin film</td>
<td>10.0 ± 0.3</td>
<td>1.021</td>
<td>0.899</td>
<td>16.75</td>
<td>66.1</td>
<td>AIST (10/11)</td>
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<tr>
<td>Organic (submodule)</td>
<td>4.2 ± 0.2</td>
<td>294.5</td>
<td>0.714</td>
<td>12.26</td>
<td>47.7</td>
<td>AIST (9/11)</td>
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<tr>
<td>Multijunction devices</td>
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<td></td>
</tr>
<tr>
<td>GaInP/GaInAs/Ge</td>
<td>34.1 ± 1.2</td>
<td>30.17</td>
<td>2.691</td>
<td>14.7</td>
<td>86.0</td>
<td>AZUR (monolithic) [8]</td>
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<tr>
<td>a-Si/nc-Si/nc-Si (thin film)</td>
<td>12.4 ± 0.7</td>
<td>1.050</td>
<td>1.936</td>
<td>8.96</td>
<td>71.5</td>
<td>United Solar [29]</td>
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<tr>
<td>a-Si/nc-Si (thin film cell)</td>
<td>12.3 ± 0.3</td>
<td>0.962</td>
<td>1.365</td>
<td>12.93</td>
<td>69.4</td>
<td>AIST (7/11)</td>
</tr>
<tr>
<td>a-Si/nc-Si (thin film submodule)</td>
<td>11.7 ± 0.4</td>
<td>14.23</td>
<td>5.462</td>
<td>2.99</td>
<td>71.3</td>
<td>AIST (9/04)</td>
</tr>
</tbody>
</table>
**Tandem vs. PBHJ**

**Tandem:**
- Effectively cover solar spectrum
- Avoid thermalization losses
- Complicated fab & costly

**PBHJ:**
- Much broader absorption
- Simple device fab
- Possible energy transfer

Proof of Concept: Selections of Materials

<table>
<thead>
<tr>
<th>Chemical</th>
<th>HOMO (eV)</th>
<th>Band Gap (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TAZ</td>
<td>5.29</td>
<td>1.98</td>
</tr>
<tr>
<td>DTBT</td>
<td>5.40</td>
<td>1.70</td>
</tr>
<tr>
<td>DTffBT</td>
<td>5.54</td>
<td>1.70</td>
</tr>
<tr>
<td>DTPyT</td>
<td>5.45</td>
<td>1.51</td>
</tr>
</tbody>
</table>
Absorption, IPCE and J-V Characteristics

**TAZ:DTBT**

- TAZ sub-cell
- DTBT sub-cell
- Parallel BHJ cell

**DTffBT:DTPyT**

- DTffBT sub-cell
- DTPyT sub-cell
- Parallel BHJ cell

**Current Density (mA/cm²)**

<table>
<thead>
<tr>
<th>Voltage (V)</th>
<th>TAZ sub-cell</th>
<th>DTBT sub-cell</th>
<th>Parallel BHJ cell</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>-12</td>
<td>-12</td>
<td>-12</td>
</tr>
<tr>
<td>0.8</td>
<td>-10</td>
<td>-10</td>
<td>-10</td>
</tr>
<tr>
<td>0.6</td>
<td>-8</td>
<td>-8</td>
<td>-8</td>
</tr>
<tr>
<td>0.4</td>
<td>-6</td>
<td>-6</td>
<td>-6</td>
</tr>
<tr>
<td>0.2</td>
<td>-4</td>
<td>-4</td>
<td>-4</td>
</tr>
<tr>
<td>0.0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

**Voltage (V)**

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Absorption (a.u.)</th>
<th>EQE (%)</th>
<th>Current Density (mA/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>400</td>
<td>0</td>
<td>0</td>
<td>-14</td>
</tr>
<tr>
<td>500</td>
<td>0</td>
<td>0</td>
<td>-12</td>
</tr>
<tr>
<td>600</td>
<td>0</td>
<td>0</td>
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</tr>
<tr>
<td>700</td>
<td>0</td>
<td>0</td>
<td>-8</td>
</tr>
<tr>
<td>800</td>
<td>0</td>
<td>0</td>
<td>-6</td>
</tr>
</tbody>
</table>

**EQE (%)**

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Absorption (a.u.)</th>
<th>EQE (%)</th>
<th>Current Density (mA/cm²)</th>
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<tbody>
<tr>
<td>400</td>
<td>0</td>
<td>0</td>
<td>-14</td>
</tr>
<tr>
<td>500</td>
<td>0</td>
<td>0</td>
<td>-12</td>
</tr>
<tr>
<td>600</td>
<td>0</td>
<td>0</td>
<td>-10</td>
</tr>
<tr>
<td>700</td>
<td>0</td>
<td>0</td>
<td>-8</td>
</tr>
<tr>
<td>800</td>
<td>0</td>
<td>0</td>
<td>-6</td>
</tr>
</tbody>
</table>
Composition Dependence

**TAZ:DTBT**

![Graph showing EQE (%) vs Wavelength (nm) for TAZ:DTBT and DTBT](image)

- Red: TAZ
- Orange: TAZ:DTBT (0.7:0.3)
- Green: TAZ:DTBT (0.5:0.5)
- Blue: TAZ:DTBT (0.3:0.7)
- Black: DTBT

**J_sc (mA/cm²)** vs **wt. (TAZ) %**

- Red: J_sc
- Blue: V_oc

- Red line: Maximum J_sc
- Blue line: Minimum V_oc

**Wavelength (nm)**: 400, 500, 600, 700, 800

**EQE (%):** 8.0, 8.5, 9.0, 9.5, 10.0, 10.5, 11.0, 11.5

**Voc (V):** 0%, 20%, 40%, 60%, 80%, 100%

**TAZ:DTBT**

**DTffBT:DTPyT**

![Graph showing EQE (%) vs Wavelength (nm) for DTffBT:DTPyT and DTPyT](image)

- Red: DTffBT
- Orange: DTffBT:DTPyT (0.7:0.3)
- Green: DTffBT:DTPyT (0.5:0.5)
- Blue: DTffBT:DTPyT (0.3:0.7)
- Black: DTPyT

**J_sc (mA/cm²)** vs **wt. (DTPyT) %**

- Red: J_sc
- Blue: V_oc

- Red line: Maximum J_sc
- Blue line: Minimum V_oc

**Wavelength (nm)**: 400, 500, 600, 700, 800

**EQE (%):** 0%, 20%, 40%, 60%, 80%, 100%

**Voc (V):** 11.0, 11.5, 12.0, 12.5, 13.0, 13.5, 14.0, 14.5

**wt. (DTPyT) %**: 0%, 20%, 40%, 60%, 80%, 100%
b) Can We Get Higher $V_{oc}$?

- a) Further understanding the origin of $V_{oc}$
- b) Searching for new acceptors
- c) Engineering the fullerene
(a,b) The CT state is significantly lower than any component singlet in the blend. EL emission originates from the CT state only.

(c,d) As the CT state energy approaches the energy of the lowest singlet of both blend materials, EL emission is a mixture between CT state emission and component singlet emission.

\[ V_{OC} \lesssim \frac{E_{opt,\min}}{e} - (0.66 \pm 0.08) \text{eV}. \]
New Acceptor? $V_{oc}$ up to 1.2 V!

Brunetti, Gong, Tong, Heeger, & Wudl

*Figure 3.* Current–voltage characteristics of dimer 4:P3HT devices (1:1.5 weight ratio) under AM 1.5G illumination using Al (circles) and Ba/Al (triangles) cathodes. [40]
c) Stability: Better Polymers?

*Typically, We do Polycondensation – Step Growth*
Step Growth to Chain Growth?

2,5' or head-to-tail (HT) coupling  

2,2' or head-to-head (HH) coupling  

5,5' or tail-to-tail (TT) coupling

HT-HT-HT  
Regioregular (planar backbone)

HT-HH-TT  
Regioirregular (non-planar backbone)
Ni(dppp)Cl₂ is believed to act as an initiator rather than a catalyst.

Recommendations

I. Further improve the Voc, and esp. the Jsc, via (a) new molecules, (b) new device designs

II. Further understanding the loss mechanism, overcome/suppress them via molecular design?

III. Stability: can we precisely control the synthesis of polymers? (Also It would impact all PV parameters)

IV. We need to work together! Small groups, Centers, etc.