

The Limits to Organic Photovoltaic Cell Efficiency

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Abstract

We consider the fundamental limits to organic solar cell efficiency, and the schemes that have been used to overcome many of these limitations. In particular, the use of double and bulk heterojunctions, as well as tandem cells employing materials with high exciton diffusion lengths, is discussed. We show that in the last few years, a combination of strategies has led to a power conversion efficiency of $\eta_P = 5.7\%$ (under AM 1.5 G simulated solar radiation at 1 sun intensity) for tandem cells based on small-molecular-weight materials, suggesting that even higher efficiencies are possible. We conclude by considering the ultimate power conversion efficiency that is expected from organic thin-film solar cells.

Keywords: organic photovoltaics, solar cell efficiency.

The Energy Conversion Process

To determine the limits to the power conversion efficiency of organic solar cells, we begin by referring to the optical-to-electrical conversion process detailed in Figure 1. The internal quantum efficiency, η_{IQE} , is the product of four efficiencies,¹ each corresponding to a step in the charge generation process:

$$\eta_{IQE} = \eta_A \eta_{ED} \eta_{CT} \eta_{CC}, \quad (1)$$

where η_A is the absorption efficiency of light within the active region of the solar cell; η_{ED} is the exciton diffusion efficiency to a dissociation site; η_{CT} is the charge transfer efficiency, which is the efficiency for dissociation of an exciton into a free electron and hole pair at that site; and η_{CC} is the charge collection efficiency. Taking into consideration the optical losses that occur on coupling light in the device active region, we arrive at the external quantum efficiency:

$$\eta_{EQE} = (1 - R) \eta_{IQE}, \quad (2)$$

where R is the reflectivity of the substrate-air interface. Finally, the power conversion efficiency of the cell is given by

$$\eta_P = \frac{V_{OC} J_{SC} FF}{P_{inc}}, \quad (3)$$

where FF is the fill factor, V_{OC} is the open-circuit voltage, J_{SC} is the short-circuit current density, and P_{inc} is the incident power density.

Organic photovoltaic (PV) cells confront several limitations that are apparent by examining Equations 1–3. First, there is an inherent tradeoff between the absorption and the exciton diffusion efficiencies. That is, the exciton diffusion length, L_D , is typically much less than the optical absorption length, $1/\alpha$. For an absorbing organic layer of thickness d , we have

$$\eta_A = (1 - e^{-\alpha d}), \quad (4)$$

whereas, assuming that the arrival of excitons at a dissociation site is independent of electric fields or other extrinsic conditions, the diffusion efficiency is given by

$$\eta_{ED} = e^{-d/L_D}. \quad (5)$$

In general, it has been found that the charge collection and charge transfer efficiencies at organic donor/acceptor (DA) interfaces commonly used in thin-film molecular organic semiconductor PV cells approach 100%, in which case the internal quantum efficiency is determined by the product $\eta_A \eta_{ED}$. This is shown in Figure 2 as a function of d/L_D for $\alpha L_D = 0.1, 0.2,$ and $1.0,$

the former two values being more characteristic of organic semiconductors either cast from solution (in the case of polymers) or grown from the vapor phase (as in the case of small-molecular-weight materials). Under the conditions of $\alpha L_D \ll 1$, the thickness of the active organic region should be such that $d \sim L_D$. However, as L_D decreases, this leads to a decreasing absorption efficiency, ultimately leading to $\eta_{IQE} \sim 0.1$ for materials commonly employed in thin-film bilayer cells based on molecular organic semiconductors. Indeed, η_{IQE} is very close to the value obtained by C.W. Tang² in the earliest demonstration of such a cell based on the DA materials combination of copper phthalocyanine (CuPc) and 3,4,9,10-perylenetetracarboxylic-bis-imidazole (PTCBI), where $L_D < 5$ nm and $1/\alpha > 200$ nm (corresponding to $\alpha L_D = 0.025$), consistent with the results in Figure 2. The situation corresponding to $L_D \ll 1/\alpha$, which is characteristic of almost all organic materials used in PV cells, forms an *exciton diffusion bottleneck*, whereby photogenerated excitons cannot reach a DA interface prior to dissociation into free carriers, ultimately limiting cell efficiency.

This limitation on η_{IQE} results in a power conversion efficiency of only 1% for bilayer cells—a value that was not convincingly exceeded in either polymer or molecular organic cells until the 21st century. Recently, the exciton diffusion bottleneck has been reduced, thereby increasing organic PV cell efficiency, by applying several strategies that have influenced the device architecture. The most effective strategies include:

1. Employing a double heterostructure,³ thereby increasing both $1/\alpha$ and L_D ;
2. Employing materials with long-range order,⁴ thereby increasing L_D ;
3. Employing a bulk^{5,6} or mixed heterojunction^{7,8} between the donor and acceptor materials to increase $1/\alpha$ without decreasing L_D ; and
4. Employing light-trapping schemes³ or multi-heterojunction (tandem) cells^{9,10} to increase the optical path length within the thin film, thereby increasing η_A .

For the remainder of this article, we will briefly discuss each of these schemes, particularly as they have been applied to small-molecular-weight organic thin films. We will show that a combination of strategies has led to $\eta_P = 5.7\%$ (under AM 1.5 G simulated solar radiation at 1 sun intensity*)¹¹ in only the last few years, sug-

*Note that efficiency measurements based on spectra obtained from laboratory solar simulators can differ from those that adhere more closely to the terrestrial solar spectrum. See, for example, the introductory article by Shaheen et al. in this issue.

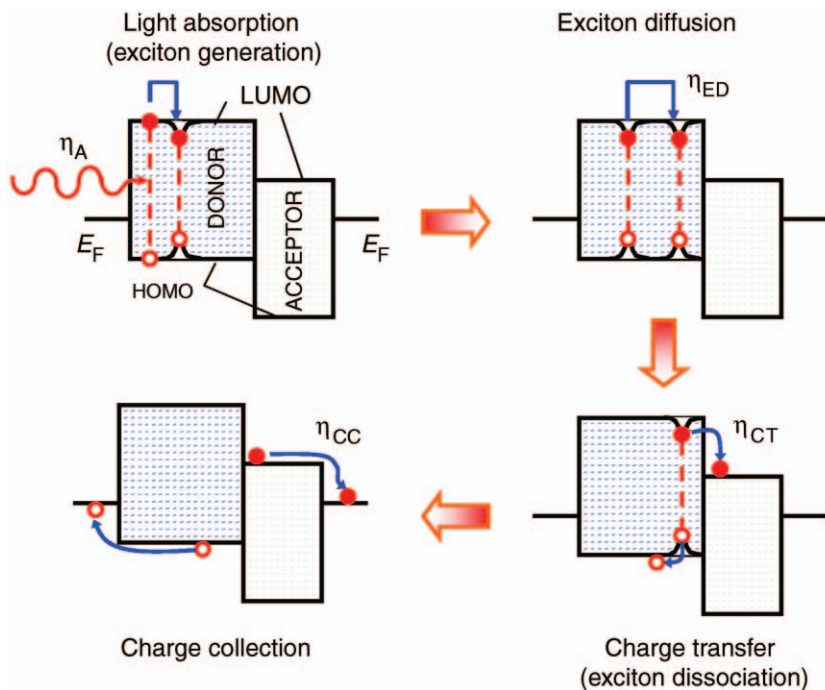


Figure 1. Steps in the photocurrent generation process. The horizontal lines to the right and left of each illustration correspond to the Fermi energies (E_F) of the cathode and anode contacts, respectively. The boxes correspond to the acceptor (left) and donor (right) energy gaps, respectively. Here, LUMO is the lowest unoccupied molecular orbital, and HOMO is the highest occupied molecular orbital of the organic film. Red dots are electrons and red circles are holes, with dashed lines drawn between them to represent excitons. Also, the dip in the energy levels in the vicinity of the exciton qualitatively depicts its binding energy ($\sim 0.5\text{--}1$ eV), placing this quasi-particle at an energy somewhat below that of the HOMO–LUMO gap energy. η_A is the absorption efficiency of light within the active region of the solar cell, η_{ED} is the exciton diffusion efficiency to a dissociation site, η_{CC} is the charge collection efficiency, and η_{CT} is the charge transfer efficiency.

gesting that even higher efficiencies are possible. This article will conclude by considering the ultimate power conversion efficiency that is expected from organic thin-film solar cells.

Double Heterojunctions and Increasing the Exciton Diffusion Length

In addition to the exciton diffusion bottleneck, bilayer PV cells suffer from two other problems that significantly limit their power conversion efficiencies. First, deposition of the cathode metal onto the acceptor layer introduces damage (typically in the form of deep trap levels), thereby reducing the exciton diffusion length in that layer. Second, the incident optical electric field vanishes at the surface of the highly conducting electrode. Since the active layer thicknesses must typically be $d < \lambda/4n \sim 100$ nm, where λ is the incident light wavelength and n is the spatially weighted average refractive index of the thin-film cell, the optical field intensity is less than its maximum value at the heterojunction

where photoinduced charge transfer is most efficient.

To circumvent these problems, the double heterostructure organic PV cell was introduced,³ as illustrated schematically in Figure 3 (inset). Here, a transparent exciton blocking layer (EBL) is interposed between the acceptor layer and the metal cathode. This EBL displaces the active DA interface such that it is centered near where the optical field intensity is at a maximum, thereby maximizing absorption. Furthermore, as this particular layer is photoelectrically “inert” (in that it does not absorb light), it can absorb the damage introduced during cathode metal deposition without that damage resulting in exciton quenching. Due to the larger energy gap (and hence transparency) of the EBL as compared with that of the acceptor layer, the electrons are confined close to the DA heterointerface, thus preventing them from migrating to the damaged region near the cathode. On the contrary, the damage to bathocuproine (BCP) EBLs has been shown to result in the sufficiently high electron conductivity

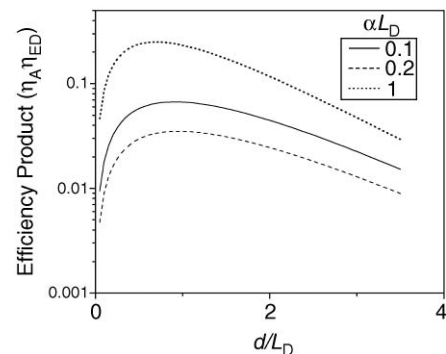


Figure 2. The product of adsorption efficiency (η_A) and the exciton diffusion efficiency (η_{ED}) illustrating the dependence of the “exciton diffusion bottleneck” on the ratio of layer thickness (d) to exciton diffusion length, L_D . Here, α is the optical absorption length within a particular layer.

required to extract separated charges from the DA interface.¹

Employing a double heterostructure allows for the use of very thin active regions without a reduction in η_{EQE} . In the first demonstration based on the archetype CuPc/PTCBI system, an ~ 8 -nm-thick BCP layer was used as the EBL while the active DA layers were each only ~ 10 nm thick. Remarkably, η_{EQE} slightly increased (see Figure 3), even though η_A was reduced by more than a factor of 2.5, providing evidence for exciton confinement in the DA interface region by the EBL. Thinning the active layers has the additional benefit of substantially reducing cell series resistance. This increases the fill factor, as well as allowing for efficient response even at very high incident light intensities (>10 suns), thereby allowing these thin-film cells to be used with solar concentrators and other light-trapping schemes.³

Finally, combining the double heterostructure architecture with materials with long diffusion lengths has resulted in a significant increase in efficiency⁴ over that first reported by Tang.² For example, replacing the acceptor, PTCBI, with C_{60} leads to an increase in the exciton diffusion length from 3 nm to 40 nm. Many of the advantageous properties of C_{60} arise from its spherical symmetry: it can pack tightly to form highly conductive films with excellent orbital overlap between adjacent molecules, thereby improving both the electron and exciton diffusion efficiencies, and the intersystem crossing resulting from the large orbital angular momentum inherent in the π -electron system converts all excited states to triplets with their correspondingly long diffusion lengths. Using C_{60} in an

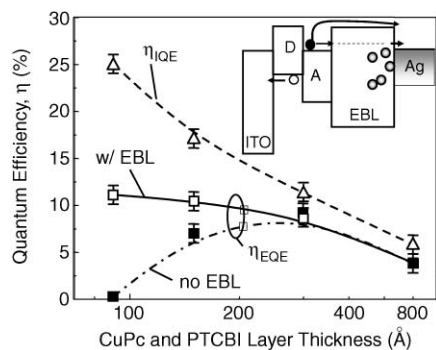


Figure 3. Quantum efficiency versus layer thickness for a bilayer CuPc/PTCBI cell with and without an exciton blocking layer (EBL). Both the external (η_{EQE}) and internal (η_{IQE}) quantum efficiencies are shown. Inset shows the energy-level scheme of a double heterojunction photovoltaic cell. The dashed line indicates defect levels that allow for conduction of electrons (solid circle) to the cathode (Ag). The shaded circles represent Ag atoms or clusters that may have diffused into the wide-energy-gap (and therefore transparent) EBL during the deposition process. Here, D and A represent the donor and acceptor materials, respectively, and ITO is the transparent indium tin oxide anode.

otherwise conventional double heterojunction cell has resulted in efficiencies⁴ of $\eta_p = 3.6\%$, increasing to 4.2% at >1 sun illumination intensity¹² (simulated AM 1.5 G spectrum) in low-series-resistance structures.

The calculated relationship between internal quantum efficiency and active layer thickness for different values of diffusion length, along with values obtained for representative small-molecular-weight bilayer PV cells, is shown in Figure 4, along with several experimental results. Clearly, employing materials with a large L_D results in a peak in η_{IQE} at larger layer thicknesses, with the C_{60} -based device corresponding to $L_D = 20$ nm.

Bulk and Mixed Heterojunctions

An alternative approach to overcoming the exciton diffusion bottleneck is to form a heterojunction (HJ) between the donor and acceptor materials with a very large surface area, as shown in Figure 5a. In this case, by entangling the regions containing the two constituents, a region is formed whereby photons can be absorbed over a very long distance, creating all excitons within a diffusion length of a DA interface where photoinduced charge transfer can occur. The first demonstration of such a "bulk" heterojunction employed a blend

of a polymer and a fullerene that formed an entangled network extending across the device. This approach has resulted in a significant improvement in power conversion efficiency over that obtained using a simple planar structure.^{6,13}

To achieve a bulk HJ based on small-molecular-weight materials is more difficult, since the phase separation must occur in the solid rather than the liquid phase, as in the case of polymers. Simply mixing materials by coevaporation of the donor and acceptor source molecules onto a substrate can result in a significant decrease in charge carrier mobility, as has been observed by the co-deposition of the archetype DA pair, CuPc and PTCBI.⁵ The reduction in carrier mobility (and, hence, increase in cell series resistance) results from the planar shape of the molecules. In a pure thin film, both CuPc and PTCBI form orderly stacks, allowing charge transport along the overlapping π -electron systems in the stacking direction. When deposited into a mixture, the stacks are disrupted, introducing charge trapping and scattering during transport to the electrodes. Hence, mixed-layer devices typically have very low (<0.1%) power conversion efficiencies.

The mixed thin film, however, is not an equilibrium structure. Thus, phase segregation into an entangled HJ can be achieved by annealing the mixture at high temperatures. For example, Peumans and co-workers⁵ have shown that annealing CuPc/PTCBI mixed-layer double HJ cells at >2000°C for ~10 min can result in internal crystallization of the film in the bulk HJ structure shown in Figure 5a. In this

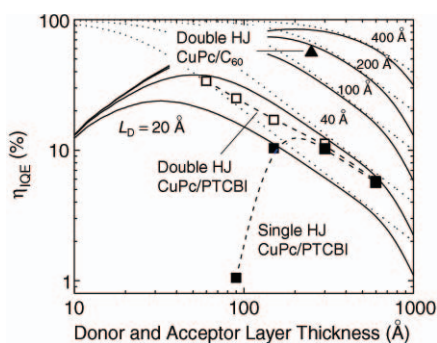


Figure 4. Calculated internal quantum efficiency versus donor and acceptor layer thicknesses. The solid curves are for several different exciton diffusion lengths, L_D , and the dotted curves (labeled 20 Å, 40 Å, 100 Å, 200 Å, and 400 Å) are L_D values for simple exciton diffusion, neglecting the optical boundary conditions. Experimental data points for several different cell configurations are also shown.

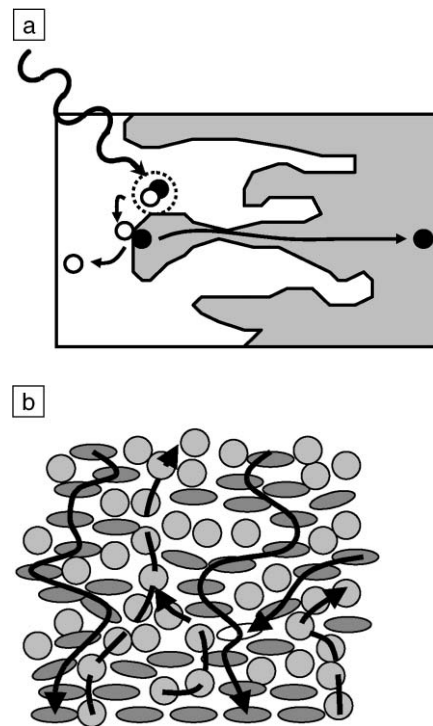


Figure 5. (a) A bulk heterojunction consisting of an entangled region of donor and acceptor materials, shown as open and shaded areas. Excitons are indicated by a close pairing of an electron (solid circle) and hole (open circle) enclosed with a dotted line. (b) A mixed heterojunction shown on the microscopic scale, with percolating conducting pathways formed by stacks of the planar CuPc molecules, as well as by the close-packed spherical C_{60} molecules.

case, the solar power conversion efficiency rises from 1% for a simple planar HJ, to nearly 1.5% under 1 sun, AM 1.5 G simulated solar radiation. However, the fill factor of the annealed cell is only $FF = 0.31$, which is somewhat lower than a planar HJ, where $FF = 0.52$. This reduction indicates that the phase separation has resulted in resistive bottlenecks and cul-de-sacs where the free charge is trapped prior to collection at the electrodes, which in many cases may present a significant limitation to the efficiency achievable using this approach.

A considerably different situation applies for CuPc/ C_{60} mixtures. In this case, the spherically symmetric C_{60} apparently does not disrupt the CuPc stacking to the same extent as in similar mixtures with the planar PTCBI molecule. Hence, CuPc/ C_{60} mixed cells do not exhibit an improvement on annealing. On the contrary, the as-deposited mixed cell itself has a high

efficiency, equal to that of a planar cell containing the same materials.⁸ Coupled with the fact that “annealing” the CuPc/C₆₀ mixed PV cell has no significant effect on the device efficiency, this suggests that the deposited mixture is an equilibrium phase, where percolating “bulk heterojunction-like” conduction paths are already formed during deposition, as shown in Figure 5b.

Further benefits can be realized using a combination of a planar and a mixed layer within the same double heterostructure, known as the hybrid planar-mixed heterojunction (PM-HJ). This structure takes maximum advantage of both the charge conduction properties of homogenous layers of an organic film, and the exciton separation properties of films consisting of a mixture of the donor and acceptor molecular species.¹⁴ Here, in a single device structure, the homogeneous donor and acceptor layers form a sandwich, with the mixed layer as the filling. Analysis of carrier transport and charge separation in the PM-HJ cell has shown¹⁴ that the thicknesses of the homogeneous layers are optimized when they are on the scale of L_D , whereas the mixed-layer thickness should be on the order of the carrier collection length (L_c)—i.e., the distance a free carrier can diffuse within a layer prior to recombination with its opposite charge. The total thickness of the device then becomes the sum of the thicknesses of these three characteristic lengths, resulting in increased optical absorption over that of a somewhat thinner bilayer heterojunction.

A recent demonstration of a PM-HJ device consisted of a combination of the donor, CuPc, and the acceptor, C₆₀. Since the diffusion lengths as well as the carrier collection lengths are all in the range of 20–40 nm, the device active region thickness increased to ~50–60 nm. The efficiency¹⁴ measured under 1 sun illumination and AM 1.5 G simulated solar radiation was¹⁴ $\eta_p = 5.0\%$ with a high fill factor (>0.6), indicating the clear benefits of the PM-HJ structure.

Tandem Cells

One significant limitation of almost all organic compounds is their relatively narrow absorption spectra. Hence, it is not feasible to absorb the entire solar spectrum using a single DA pair forming the organic PV cell. Furthermore, the open-circuit voltage that is produced by most organic DA heterojunction cells is small (typically ranging between 0.4 V and 0.8 V). One particularly powerful means of circumventing these shortcomings is the use of a multicell series stack of PV cells, each optimized to absorb in a different region of the solar spectrum. In this so-called tandem

architecture (Figure 6), currents generated due to absorption within each subcell flow in series to the opposing contacts. In a series configuration, the photocurrent flowing in the tandem cell is limited by the smallest current generated by a particular subcell in the stack. Furthermore, the open-circuit voltage is equal to the sum of the open-circuit voltages of the subcells.^{9,10}

The example tandem cell shown in Figure 6 consists of two PM-HJ cells in series.¹¹ To allow the charges that flow between the subcells (as opposed to those that flow to the electrodes) to recombine within the device interior, the tandem cell requires sites that serve to attract carriers of opposite charge. One means to generate such sites is to evaporate ultrasmall (~5 Å diameter) metal particles onto the surface of the first cell prior to depositing the layers for the second subcell. This nanocluster layer is sufficiently thin to efficiently provide recombination centers, yet not thick enough to absorb light on its way to the back cell, nearest the reflecting cathode.

With the plane of polarization of the incident light oriented along the particle axis, separation of free charge on the metal surface results in an instantaneous oscillating dipole, or “surface plasmon.”¹¹ This excitation re-radiates the light, whose peak

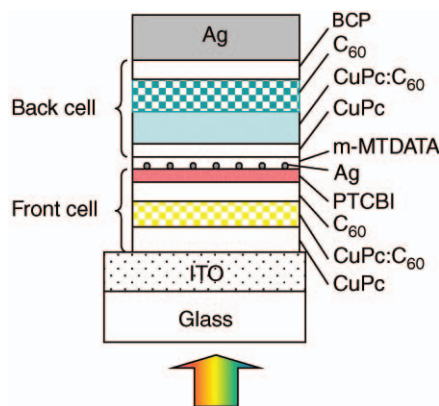


Figure 6. A bilayer tandem photovoltaic cell consisting of a stack of planar mixed double heterojunction subcells. The cell nearest the Ag cathode (the back cell) is rich in C₆₀, which absorbs in the blue spectral region, whereas the front cell is rich in CuPc, which absorbs preferentially in the red and yellow spectral regions. Hence, the cell is designed to have a maximum overlap between a given layer and the part of the solar spectrum which it optimally absorbs. Such a tandem cell, including the Ag nanocluster layer that enhances absorption via surface plasmon effects, has demonstrated power conversion efficiencies of 5.7% under 1 sun, AM 1.5 G simulated illumination. (See Reference 11.)

intensity lies ~10 nm from the particle, thereby localizing the radiation almost ideally at the nearby DA interface.¹ This concentrates the light within L_D of the active heterojunction, circumventing the exciton diffusion bottleneck. Indeed, in the first successful CuPc/PTCBI tandem cells, the Ag nanocluster layer introduced a ~20% increase in efficiency beyond that expected due to simple absorption in the multilayer cell, achieving $\eta_p = 2.5\%$ for a double-layer tandem¹⁹ (versus $\eta_p \sim 1.9\%$, as expected due to the increased cell thickness and, hence, increased absorption).

In the case of the cell in Figure 6, the front cell was designed to preferentially absorb red light by making it rich in CuPc, whereas the back cell preferentially absorbed blue solar radiation due to the larger total thickness of C₆₀ in that region.¹¹ By this means, the efficiency of the tandem cell is optimized to respond to the broadest possible span of the solar spectrum using only the standard materials of CuPc, C₆₀, and PTCBI. It was found that an efficiency of $\eta_p = 5.7\%$ (1 sun illumination and AM 1.5 G simulated solar radiation)¹¹ was obtained with a fill factor equal to that of a single junction cell (>0.6) and $V_{OC} = 1.02$ V, corresponding to a doubling of V_{OC} compared with that of a single-element CuPc/C₆₀ cell.

Analysis of the tandem architecture using this particular materials combination has indicated that cells with efficiencies of 6.5% are possible,¹¹ although this has not yet been achieved. Further improvements can be expected using improved coupling of light into the cell active region, and extending the materials combinations to reach into the infrared part of the solar spectrum, perhaps by employing a third subcell in the stack.

The Outer Limits

Figure 7 illustrates the recent progress made in improving the efficiency of organic thin-film photovoltaic cells. Following a long period since the demonstration of the first DA heterojunction by Tang² in 1986, there has recently been a rapid increase in cell efficiency. Many of these latest developments are due to lessons learned from improving the performance of organic light-emitting devices that often employ materials and device structures that have direct application to light detection. As of this writing, the maximum reported efficiency obtained for an organic PV cell in a dual tandem structure is 5.7%. It is reasonable, therefore, to ask what the limits to cell efficiencies might be.

Based on simple physical considerations, the ultimate limitation to the cell efficiencies is the charge separation process at the

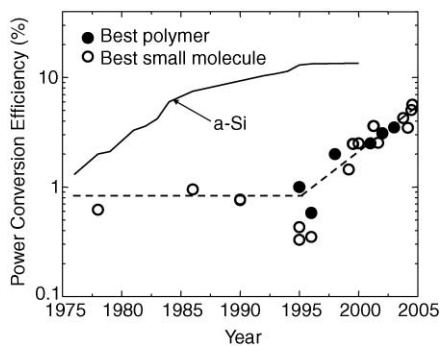


Figure 7. Progress in power conversion efficiencies for both small-molecular-weight and polymer photovoltaic cells over a span of 30 years. Also shown for comparison is the progress made in a more conventional thin-film technology, amorphous Si. Note that the initial slope of a-Si cell progress is similar to what we are experiencing today in organic thin films. (Adapted from Wronski et al., Reference 16.)

heterojunction formed between the donor and acceptor materials. Analysis of this process based on mechanisms of exciton dissociation at the 2D interface suggests that ultimately the fill factor will be limited to¹⁵ 0.6–0.7. Hence, assuming 100% conversion efficiency across the visible spectrum, this sets an *approximate* upper bound to the obtainable efficiency¹⁵ of $\eta_p \approx 20\%$. Yet, there are many hurdles to surmount before even that number can be reasonably approached. We briefly list the challenges that must be overcome to exceed the benchmark 10%, and to approach this “ultimate limit”:

1. The cell must have an efficient response across the entire solar spectrum. Given the limited spectral coverage of organic thin films (see the section on “Tandem Cells”), this almost certainly will require the use of tandem structures, with different materials systems making up the subcells to provide the necessary spectral coverage. To benefit from the stacked structure, however, each subcell must generate an equal current under 1 sun illumination intensity. This will most likely limit the number of cells to three.⁹ More important, this presents a major challenge to the materials scientist to develop DA combinations that have comparable efficiencies across the solar spectrum.

2. The existence of the exciton diffusion bottleneck, coupled to the inherently large series resistance of the organic thin films, requires that the individual subcells in the tandem stack be very thin. Hence, it is essential that the cells be designed to couple most of the light into the active hetero-

junction regions. This will require innovation in the optics of the thin-film devices to result in concentrating the incident radiation directly into the regions that are most photoactive. The use of surface plasmons from metal particles between the subcells is one of many possible avenues available to solving the coupling problem.

3. Cell resistance also presents a problem in transporting the power from its generation site to the load. Creating low-optical-loss, high-conductivity contacts, therefore, is an important and nontrivial challenge that has yet to be addressed in this emerging field.

In spite of the formidable challenges that confront materials and device scientists in developing high-efficiency solar cells, the rewards for success are substantial: organics can potentially transform the cost structure for generating cheap and clean renewable energy from the sun. Their ability to be deposited on lightweight and flexible plastic substrates can allow for ultralow cost manufacturing, while the cells can find applications that are inaccessible to conventional thin-film PV cell technologies. Coupled to the very high potential efficiencies within reach of organic thin-film cells, this technology offers opportunities that are simply too great to ignore.

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