

Polymer-based solar cells

A significant fraction of the cost of solar panels comes from the photoactive materials and sophisticated, energy-intensive processing technologies. Recently, it has been shown that the inorganic components can be replaced by semiconducting polymers capable of achieving reasonably high power conversion efficiencies. These polymers are inexpensive to synthesize and can be solution-processed in a roll-to-roll fashion with high throughput. Inherently poor polymer properties, such as low exciton diffusion lengths and low mobilities, can be overcome by nanoscale morphology. We discuss polymer-based solar cells, paying particular attention to device design and potential improvements.

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Conjugated polymers are excellent candidates for use in low-cost electronics and photovoltaics (PV)¹. Polymer-based solar cells have reached power conversion efficiencies of 5% in recent reports²⁻⁶. Deposition of organics by screen printing, doctor blading, inkjet printing, and spray deposition is possible because these materials can be made soluble. These techniques are required for the high-throughput roll-to-roll processing that will drive the cost of polymer-based PV down to a point where it can compete with current grid electricity. Additionally, these deposition techniques all take place at low temperature, which allows devices to be fabricated on plastic substrates for flexible devices. In addition to the inherent economics of high-throughput manufacturing, light weight and flexibility are qualities claimed to offer a reduction in the price of PV panels by reducing installation costs. Flexible PV also opens up niche markets like portable power generation and aesthetic PV in building design.

We review current state-of-the-art techniques for making efficient polymer-based PV devices. We discuss the basic device operation, materials requirements, and current technical challenges in making more efficient solar cells. We hope this article will serve as a guide to new researchers in the field who plan to develop better material systems and optimize devices to push organic photovoltaic power conversion efficiencies above 10%.

Device operation and materials challenges

Fig. 1 shows examples of common materials used in state-of-the-art polymer-based PV cells. These and other conjugated molecules are electronically active because of their highly polarizable π -systems, which are hybridized orbitals based on the constituent p atomic orbitals. The π - π^* optical transitions are strong (absorption coefficients greater than $\sim 10^5 \text{ cm}^{-1}$), typically fall in the visible, and can be tuned synthetically through molecular design. Charge carrier mobilities are

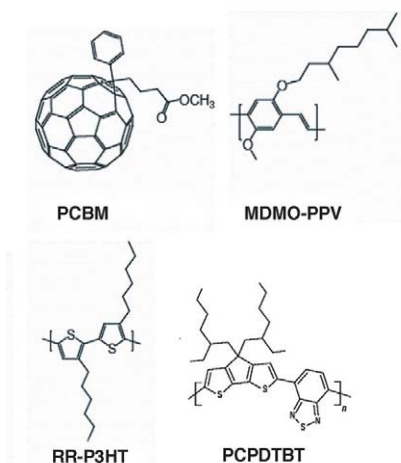


Fig. 1 Common materials used in polymer photovoltaics. From left to right, PCBM: (6,6)-phenyl-C₆₁-butyric acid methyl ester; MDMO-PPV: poly(2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylene-vinylene); RR-P3HT: regioregular poly(3-hexylthiophene); PCPDTBT: poly[2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta[2,1-b;3,4-b']-dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)].

usually in the range of 10^{-7} – 10^{-3} cm²/V.s, when measured under conditions relevant to solar cell operation.

In reaching power conversion efficiencies of 5%, researchers have had to develop novel device architectures (Fig. 2) to address challenges inherent in using polymeric semiconductors. In all these architectures, many steps are involved between photon absorption and electricity generation, all of which must be efficient to make an efficient device.

On photon absorption, an electron is excited from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO). This electron-hole pair then relaxes with a binding energy between 0.1–1.4 eV, and is known as an exciton^{7–9}. This is in contrast to inorganic materials, where the exciton binding energy is only a few millielectron volts. The binding energy is larger in organic semiconductors because electron and hole wavefunctions are localized and the dielectric constants are low ($\epsilon \sim 3$ –4), enhancing the Coulomb attraction between the electron and hole. The bound excitons must migrate to an interface where there is a sufficient chemical potential energy drop to drive dissociation into an electron-hole pair that spans the interface across the *donor* (material with low electron affinity) and *acceptor* (material with high electron affinity) (Fig. 2a). The simplest device that promotes exciton separation is a planar heterojunction (Fig. 2b) sandwiched between a transparent conductor such as indium-tin-oxide (ITO) coated with poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) or fluorinated tin oxide (FTO), and a reflecting metal (usually Al or Ag)¹⁰. The charge pair formed after exciton dissociation, a *geminate pair*, is often still Coulombically bound and must be dissociated by an internal field^{11–15}.

After dissociation, each charge carrier must be transported through the device to the appropriate contact while avoiding traps and recombination. These charges are transported primarily by drift caused

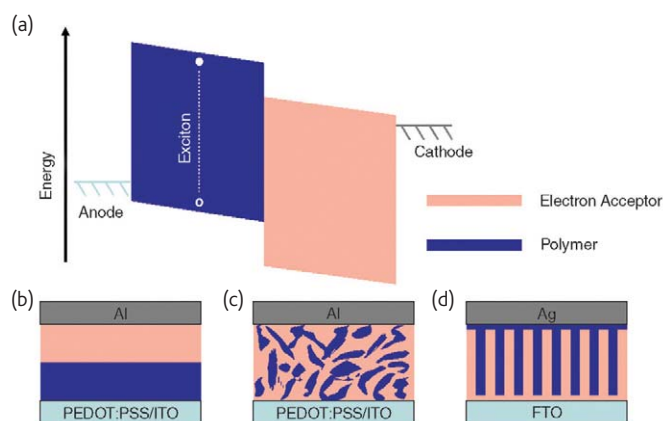


Fig. 2 Polymeric solar cell heterojunctions. (a) Energy diagram of a heterojunction with an exciton in the polymer phase. The first heterojunction was a bilayer (b), but this architecture is limited as the active layer must be thin in order to utilize all excitons. In order to have every exciton separated, a heterojunction must exist within an exciton diffusion length. This can be achieved in a bulk heterojunction (c) or in an ordered heterojunction (d).

by the built-in field, though diffusion can play a large role. The current that reaches the contacts with no applied field is known as the short-circuit current, J_{sc} , and the maximum potential generated by the device is known as the open-circuit voltage, V_{oc} . Of course, for the current to do work, it must be generated with some potential. The ratio between the maximum power generated and the product of J_{sc} and V_{oc} is known as the fill factor, and is related to the quality of the device.

The efficiency of the planar heterojunction device is limited by the exciton diffusion length. This is the distance over which excitons travel before undergoing recombination and is ~ 3 – 10 nm in most organic semiconductors^{16–18}. Excitons formed at a location further from the heterojunction than the exciton diffusion length have a lower probability of being harvested. The active volume of this type of solar cell is thus limited to a very thin region close to the interface, which is not enough to adsorb most of the solar radiation flux. In order to overcome this limitation, researchers have moved to nanostructuring the materials such that the domain size is on the order of twice the diffusion length.

The bulk heterojunction (BHJ) (Fig. 2c) is the most successful device architecture for polymeric photovoltaics because exciton harvesting is made near-perfect by creating a highly folded architecture such that all excitons are formed near a heterojunction. BHJs are formed by spin-casting the polymer and an electron acceptor from a common solvent. Common acceptors used in polymer BHJs are fullerenes^{2–6,19–27}, polymers^{28–31}, and *n*-type inorganic nanoparticles^{32–36}. The bulk of the literature has focused on blends of rr-P3HT or MDMO-PPV and the C₆₀ derivative, PCBM, and these systems will be discussed in detail.

Another important polymeric PV technology stems from replacing PCBM with a mesostructured *n*-type inorganic semiconductor (Fig. 2d). Most successful organic/inorganic hybrid cells use transparent metal

oxides, such as TiO₂ or ZnO, as the *n*-type semiconductor because of their high electron mobility, transparency, and ease of processing.

All the mechanisms mentioned above put strict requirements on the materials and architectures for efficient polymer photovoltaics:

1. Domain sizes of the donor and acceptor must be small compared with the distance excitons migrate to allow efficient exciton harvesting before natural exciton decay. This is often <10 nm and is the reason that BHJ architectures are necessary;
2. The electrochemical potential drop at the donor-acceptor interface must be sufficient to dissociate excitons. This is still poorly understood but exciton splitting has been seen with drops at the donor-acceptor interface as low as 0.3 eV;
3. Since the geminate pair must be split before back transfer of the electron from the acceptor to the donor, the carrier transport rate must be fast compared with the back-transfer rate. While little work has been done on modifying the interfacial back-transfer rate, the hopping rate is proportional to carrier mobility. Simple calculations show that mobilities should be 10⁻⁴ cm²/V.s or higher for efficient geminate separation³⁷. This process also imposes geometric restrictions. Highly folded, disordered geometries should lead to lower probabilities of geminate pair separation. Consequently, ordered nanostructures are ideal for splitting geminate pairs;
4. Both the electron and hole mobilities should be high enough (>10⁻⁴ cm²/V.s) and/or balanced to prevent the build up of space charge, which can hinder further extraction of charge carriers³⁷; and
5. Mobilities should be high enough to extract carriers as they are transported through the device before they undergo recombination, but not so high that the diffusive dark current limits *V*_{oc}. While still under study, mobilities of 10⁻⁴–10⁻² cm²/V.s are probably optimal for device thicknesses of 200–500 nm. Higher mobilities will require changes in device design to minimize dark current.

These restrictions are necessary to ensure high photon-to-electron conversion efficiencies with low voltage dependence (which leads to high fill factors). High photocurrents require light absorption over the whole solar spectrum and this has led to the development of some low bandgap polymers, as will be discussed below. The highest photocurrents achieved under one-sun illumination in polymer PV are still only ~15 mA/cm², compared with over 40 mA/cm² for single-crystal Si cells, leaving much room for progress. Finally, minimizing losses in chemical potential in the above processes is necessary to drive *V*_{oc} to its maximum. In P3HT:PCBM solar cells, 1.9 eV photons are absorbed but a *V*_{oc} of only 0.6 V is usually measured. The other 1.3 eV is lost. Doubling of efficiencies should easily be achievable by simply reducing this energy drop to the minimum necessary.

Bulk heterojunctions

Fig. 3 shows how the efficiency of BHJ schemes has progressed over the last 15 years. Most of these reported efficiencies remain uncertified by the National Renewable Energy Laboratory (NREL), but some have

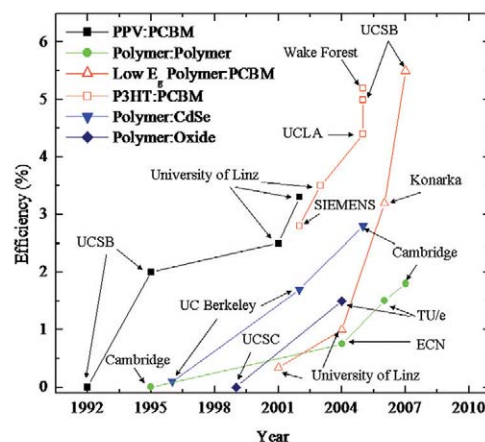


Fig. 3 Reported efficiencies of various BHJ technologies over time. The references in chronological order are: PPV:PCBM^{19–22}, polymer:polymer^{28–31}, polymer:CdSe^{32–34}, polymer:oxide^{35,36}, low *E*_g polymer:PCBM^{6,23,25}, and P3HT:PCBM^{2–5,26,27}. The abbreviations are: University of California, Santa Barbara (UCSB); University of California, Santa Cruz (UCSC); University of California, Berkeley (UC Berkeley); University of California, Los Angeles (UCLA); Technische Universiteit Eindhoven (TU/e).

been corroborated by several groups. The key to making efficient polymer blend solar cells, is to ensure that the two materials are intermixed at a length scale less than the exciton diffusion length. That means every exciton formed in the polymer can reach an interface with the electron acceptor to undergo charge transfer. The film morphology also has to enable charge-carrier transport in the two different phases such that the charges can reach the contacts and be extracted. We discuss important innovations related to PCBM-based BHJs. Though this is specific to PCBM-based PV, the implications are general to all BHJs.

The original BHJs containing PCBM employed polyphenylene vinylenes (PPV) as the electron donor. In PPV:PCBM blends, the choice of solvent and blending ratio greatly affects solar cell performance^{21,38,39}. When toluene is used as the common solvent, PCBM forms domains that are over 100 nm in size. These are effectively dead zones in the solar cell as they do not contribute to the photocurrent^{38,39}. The phase segregation is made finer by switching to chlorobenzene, a good solvent for both PPV and PCBM²¹. Another problem with the PPV system is that the hole mobility in PPV is ten times lower than the electron mobility in PCBM. This leads to a buildup of holes and a field that opposes the built-in field – a phenomenon known as space-charge limited photocurrent (SCLP)^{40,41}. Researchers have found that the hole mobility increases upon PCBM loading, which is attributed to increased ordering in the PPV phase⁴¹. Using a blend containing 80% PCBM sandwiched between the transparent contact and LiF/Al yields an efficiency of ~3.5%²². In order to achieve a higher efficiency, the hole mobility needs to be increased further.

rr-P3HT has among the highest reported field-effect transistor mobilities for a conjugated polymer because of the semicrystalline nature of its spin-cast films⁴². Moreover, the morphology, and hence the mobility, of pure rr-P3HT and blended rr-P3HT:PCBM films is highly

dependent on casting conditions. For example, the hole mobility in blends spun from chlorobenzene is slightly lower than the electron mobility, but for films less than 200 nm-thick, SCLP is not a problem. However, attempts to make the active layer thicker in order to absorb more sunlight result in SCLP^{43,44}. Using optical fields and spacers along with optimized annealing yields 100 nm films with efficiencies around 5%^{2,4,5}. However, further increases in mobility are required to enable the use of thicker films and increase absorption.

Decreasing the evaporation rate of the solvent by changing from chlorobenzene to *ortho*-dichlorobenzene greatly enhances the power conversion efficiency of the device. This is because the hole mobility in the rr-P3HT phase is increased by more than a factor of ten over films spun from chlorobenzene^{3,44}. Even with this increase, the device thickness is still limited to around 300 nm or 400 nm. The large bandgap of P3HT (1.9 eV) limits the short circuit current to ~15 mA/cm². A smaller bandgap polymer would absorb more of the solar spectrum and generate a higher photocurrent.

Small bandgap polymers for solar cells have been synthesized since 2001²³. These polymers have lower optical density and mobility than PPV and P3HT, as well as an unoptimized nanostructure. Several researchers have tried to overcome these problems by replacing the PCBM with PC₇₁BM, which has an optical density twice that of PCBM and complements the absorption spectra of the polymers well. This approach yields efficiencies ~3.2%, which is still below PPV- or P3HT-based solar cells. Recently however, Heeger and Bazan *et al.*⁶ at UCSB have improved the efficiency of blends based on PCPDTBT and PC₇₁BM by gaining control of the morphology. This control is gained by mixing 1,8-octanedithiol into the solution, and yields an efficiency of 5.5%. Further control of the nanomorphology should extend this efficiency to 7%, which puts polymer-based solar cells one step closer to being competitive with inorganic technologies.

The ultimate efficiency of the BHJ will eventually be limited by the random network that is formed through the rapid drying of the solution. For example, variations in the side-chain length of the polymer, weight percentage of materials, and solvent choice can lead to vastly different morphologies^{45,46}, which can severely affect exciton harvesting. Phase segregation kinetics during casting is a complicated process that may produce dead ends and isolated domains (Fig. 2c) that trap charge carriers and prevent them from being extracted.

Structuring one of the components into vertically aligned rods or pores with a diameter less than the exciton diffusion length would alleviate this problem. Ordered organic–organic solar cells have yet to yield an efficient device. However, replacing the disordered electron acceptor with an ordered nanostructured oxide is a viable route.

Ordered heterojunctions

Nanostructured oxides can be fabricated by several techniques^{47–52} with precise control of the continuous nanostructure. Titania templates can be made having continuous pores with a size matching the

exciton diffusion length of a polymer, allowing the possibility of complete exciton harvesting and charge collection. Ordered heterojunctions are generally made by infiltrating polymers into nanostructured oxides. Nanostructured TiO₂ is an attractive approach to develop extremely efficient organic photovoltaics. Titania is both abundant, nontoxic, and has been well-studied by the dye-sensitized solar cell community^{53,54}.

The nanostructure in Fig. 2d is considered ideal because it has small, straight pores and is thick enough to absorb most of the sunlight. The pore radius should be slightly less than the exciton diffusion length to enable maximum exciton harvesting. The thickness of the nanostructure should be 300–500 nm so that the infiltrated polymer can absorb most of the incident light. The pores (or channels) should be straight to provide the most direct path to the anode and cathode.

Holes traverse the entire thickness of the structure through the low-mobility polymer. Therefore, thicker films may lead to high series resistance, recombination losses, and SCLP, resulting in low charge collection efficiencies. Recall that morphology affects the mobility greatly. In fact, it is possible to align the polymers in the pores to increase the mobility⁵⁵, and that the above problems should be alleviated through further control of the morphology.

An ideal nanostructure should have as little titania as possible to enable maximum absorption. Since the oxide nanostructure is fabricated first, the interface is amenable to modifications in order to maximize forward electron transfer and minimize recombination, as well as promote polymer wetting. Straight pores with smooth sidewalls would likely allow the polymer chains to align themselves in the vertical direction, thereby promoting exciton diffusion and charge transport. It would be advantageous if the films were also made at low cost on plastic substrates, which would require that the deposition and thermal treatments be done at modest temperatures (<500°C).

Currently, the most efficient ordered nanostructured oxide/polymer device (0.4%)⁵⁶ is significantly worse than BHJ PV. There are many challenges to develop efficient ordered heterojunctions. In the past, fabricating nanostructures with small pore sizes (~10 nm) and high aspect ratios (30) has proven difficult. However, there are now many promising routes to develop ordered mesostructures⁵⁷, embossed nanostructures⁵⁰, nanotubes⁴⁹, and nanowires⁵¹ that could meet requirements. Some possible nanostructures are shown in Fig. 4, but have yet to be successfully incorporated in a polymer solar cell^{49,50}.

There are a variety of methods (i.e. melt infiltration, dip-coating, and polymerization) used to fill pores. However, the polymer mobility, which is crucial in determining fill factor and J_{sc} in semicrystalline polymers, can be reduced inside small pores (~10 nm). There is no general rule to understand the effects of pore size on the mobility of a given polymer, because hole transport is dependent on the specific microstructure of the polymer film. It has been shown that only 68% of excitons are split at the titania/polymer interface inside mesoporous films⁵⁸, possibly because of reduced exciton diffusivity and imperfect

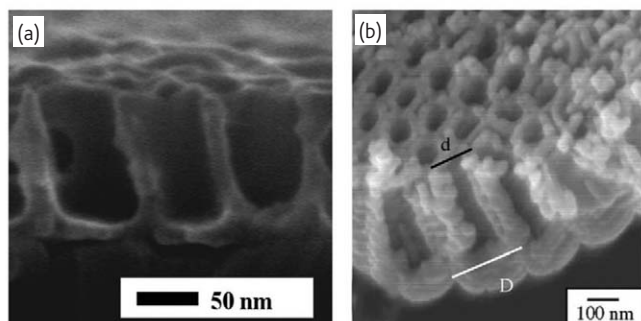


Fig. 4 (a) Cross-sectional image of embossed TiO_2 after calcination. Reprinted with permission from⁵⁰. © 2005 American Chemical Society. (b) Field-emission scanning electron microscopy image of anodized TiO_2 nanotube arrays. Reprinted with permission from⁴⁹. © 2006 Elsevier.

exciton quenching. Fortunately, the nanostructured oxide surface can be modified using dyes, carboxylic, and phosphoric acid groups, which have the potential to improve polymer wetting and charge transfer dynamics. Using interface modifiers, it is possible to completely quench excitons at the TiO_2 /polymer interface^{52,59}.

Förster Resonant Energy Transfer (RET) can increase the effective excitation diffusion lengths of various polymers. RET can be thought of as the transfer of energy from an excited chromophore (exciton) in an energy donor to an energy acceptor. The strength of RET, and consequently the distance over which RET is effective, depends on the photoluminescence efficiency of the donor and the overlap between donor emission and acceptor absorption spectra. RET has been used to increase the exciton diffusion length from 3 nm to 27 nm in solar cells that use a RET absorbing layer on top of titania⁶⁰. Enhanced diffusion lengths have significant implications for nanostructure requirements. Larger domain sizes would suffice for complete exciton harvesting and larger nanostructures would give more room for the polymer chains inside the nanostructure to align themselves.

Nanostructured TiO_2 provides scaffolding with fixed domain sizes with interfaces that can be easily modified – this is the most important reason to develop nanostructured TiO_2 templates. Such a nanostructure enables great flexibility to fine tune ordered nanostructures by independently exploring polymer choices and device architectures. This would allow large improvements in light absorption (e.g. by using a small bandgap polymer), exciton harvesting, charge transport, and V_{oc} (e.g. by reducing the LUMO-LUMO difference between TiO_2 and the polymer by adjusting the polymer side chains or by using interface modifiers). Two avenues for significant progress lie in creating the ideal nanostructure that enables maximum efficiency given a particular TiO_2 /polymer system, and polymer engineering to enhance intrinsic properties such as bandgap, exciton diffusion lengths, energy levels, and mobility. In the case of TiO_2 /P3HT, harvesting all incident photons, perhaps by using nanostructures, coupled with reported V_{oc} values and fill factors, achieves an efficiency of over 5%. Suitable polymer engineering can catapult efficiencies above 10% in the future⁵⁵.

Multijunctions

Organic multijunction solar cells are a promising route to increase efficiencies. For III-V compounds and amorphous Si alloys, stacking different absorbers is primarily a means of reducing the thermalization losses of high-energy photons, which significantly limits the theoretical efficiency of a single absorber solar cell. This is also true for organic semiconductors, but stacking cells additionally overcomes the limited bandwidths that are typically not broad enough to absorb the entire spectrum. Stacking a large number of cells is also a possible solution for devices that are too thin to absorb all the light because of short carrier collection lengths or short exciton diffusion lengths⁶¹.

The most common way to stack cells is to connect them in series⁶². For this method, the main requirements are that the cells must be current matched, and the interconnect junction between the cells must be transparent and have low series resistance while making proper contact as anode for one cell and cathode for the other.

Hiramoto *et al.*⁶³ made the first organic tandem solar cell in 1990 by stacking two identical small molecule bilayer devices connected in series by a thin discontinuous Au layer. The layer of metal clusters absorb little light and act as efficient recombination centers. This technique has been used extensively with great success for both polymer and small-molecule tandem cells. Recently, the use of high and low work function transparent conductors to form the interconnect junction⁶⁴ and the use of two different absorbers to cover more of the solar spectrum has been demonstrated by several groups^{65–67}.

The additional complexity of a tandem structure entails a significant amount of device engineering to realize higher efficiencies. Matching the currents while ensuring that photons are not wasted, and that each subcell is operating near its maximum power point is challenging. Xue *et al.*⁶⁵ have shown the importance of engineering the optical interference effects. By exploiting the fact that the absorber layers can be very thin and that the absorption spectrum of each organic is very narrow, they increased the efficiency by putting the wide-gap cell at the back of the device. Engineering the optical effects of interconnect materials with a very different index of refraction (e.g. metals or oxides) may also prove useful.

The additional manufacturing steps for a multijunction cell also need to be considered. Much of the appeal of polymer photovoltaics is the possibility of solution processing in a roll-to-roll fashion. One of the major difficulties for polymer multijunction cells is sequentially depositing multiple layers from solution without dissolving away the underlying films. However, several groups have recently found success by depositing a protective transparent oxide as the first interconnect layer^{68,69}. The oxide serves as the top contact for the bottom cell and an ideal hydrophilic surface for depositing PEDOT:PSS, which serves as the bottom contact for the next cell. Gilot *et al.*⁶⁹ have demonstrated a triple-stack device with a high V_{oc} of 2.19 V using the ZnO/PEDOT:PSS interconnect layer. The ability to stack organic devices in such a manner is a distinct advantage over their inorganic

counterparts, where growing crystalline layers of different materials epitaxially is difficult.

The highest reported efficiency for an organic solar cell (with the exception of the dye-sensitized solar cell) – a respectable 6.5% – is a tandem device that employs solution processing for all of the active and interconnect layers⁶⁸. Even with current materials and understanding, efficiencies in excess of 10% should be readily attainable. However, for efficiencies significantly higher, let alone efficiencies exceeding the single-absorber Shockley-Queisser limit⁷⁰ developing new polymers with complementary absorption spectra and achieving higher V_{oc} in the wide-bandgap layers will be essential.

Summary and outlook

Polymer-based solar cells are approaching power conversion efficiencies that make them competitive with CO₂-producing technologies. The

record efficiencies have been observed in disordered nanostructured heterojunctions but further gains are expected upon optimizing ordered nanostructure architectures. Current challenges are:

1. Creating an ordered nanostructure with the appropriate domain size (<10 nm) and dimensions (300–500 nm thick);
2. Developing small bandgap polymers with absorption edges as low as 1 eV, absorption coefficients larger than 10⁵ cm⁻¹, and charge carrier mobilities higher than 10⁻⁴ cm²/V.s; and
3. Minimizing energy loss at the donor-acceptor interface by tuning energy levels.

Progress on these few basic points is necessary to drive efficiencies higher and the field forward, ultimately to provide the cheap, clean energy technology that is in such desperate demand. Once high efficiency has been obtained, efforts will be needed to be to make the devices stable in sunlight for at least ten years. **mt**

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