A Review : Progress in Organic Solar Cells

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ABSTRACT: An organic solar cell (OSC[1]) or plastic solar cell is a type of photovoltaic that uses organic electronics, a branch of electronics that deals with conductive organic polymers or small organic molecules,[2] for light absorption and charge transport to produce electricity from sunlight by the photovoltaic effect. Most organic photovoltaic cells are polymer solar cells.

The molecules used in organic solar cells are solution-processable at high throughput and are cheap, resulting in low production costs to fabricate a large volume.[3] Combined with the flexibility of organic molecules, organic solar cells are potentially cost-effective for photovoltaic applications.[4] Molecular engineering (e.g., changing the length and functional group of polymers) can change the band gap, allowing for electronic tunability. The optical absorption coefficient of organic molecules is high, so a large amount of light can be absorbed with a small amount of materials, usually on the order of hundreds of nanometers. The main disadvantages associated with organic photovoltaic cells are low efficiency, low stability and low strength compared to inorganic photovoltaic cells such as silicon solar cells.

KEYWORDS: organic solar cell, photovoltaic effect, band gap, nanometers, silicon, optical, flexibility, engineering

I. INTRODUCTION

Compared to silicon-based devices, polymer solar cells are lightweight (which is important for small autonomous sensors), potentially disposable and inexpensive to fabricate (sometimes using printed electronics), flexible, customizable on the molecular level and potentially have less adverse environmental impact. Polymer solar cells also have the potential to exhibit transparency, suggesting applications in windows, walls, flexible electronics, etc. An example device . The disadvantages of polymer solar cells are also serious: they offer about 1/3 of the efficiency of hard materials, and experience substantial photochemical degradation.[5]

Polymer solar cells' stability problems,[6] combined with their promise of low costs[7] and potential for increasing efficiencies[8] have made them a popular field in solar cell research. In 2015, polymer solar cells were achieving efficiencies of more than 10% via a tandem structure.[9] In 2010, a new record-breaking efficiency of 19.3% was achieved by Hong Kong Polytechnic University.[10]

A photovoltaic cell is a specialized semiconductor diode that converts light into direct current (DC) electricity. Depending on the band gap of the light-absorbing material, photovoltaic cells can also convert low-energy, infrared (IR) or high-energy, ultraviolet (UV) photons into DC electricity. A common characteristic of both the small molecules and polymers used as the light-absorbing material in photovoltaics is that they all have large conjugated systems. A conjugated system is formed where carbon atoms covalently bond with alternating single and double bonds. These hydrocarbons' electrons pz orbitals delocalize and form a delocalized bonding π orbital with a π* antibonding orbital. The delocalized π orbital is the highest occupied molecular orbital (HOMO), and the π* orbital is the lowest unoccupied molecular orbital (LUMO). In organic semiconductor physics, the HOMO takes the role of the valence band while the LUMO serves as the conduction band. The energy separation between the HOMO and LUMO energy levels is considered the band gap of organic electronic materials and is typically in the range of 1–4 eV.[11]

All light with energy greater than the band gap of the material can be absorbed, though there is a trade-off to reducing the band gap as photons absorbed with energies higher than the band gap will thermally give off their excess energy, resulting in lower voltages and power conversion efficiencies. When these materials absorb a photon, an excited state is created and confined to a molecule or a region of a polymer chain. The excited state can be regarded as an exciton, or an electron-hole pair bound together by electrostatic interactions. In photovoltaic cells, excitons are broken up into free electron-hole pairs by effective fields. The effective fields are set up by creating a heterojunction between two dissimilar materials. In organic photovoltaics, effective fields break up excitons by causing the electron to fall from the conduction band of the absorber to the conduction band of the acceptor molecule. It is necessary that the acceptor material has a conduction band edge that is lower than that of the absorber material.[12][13][14][15]
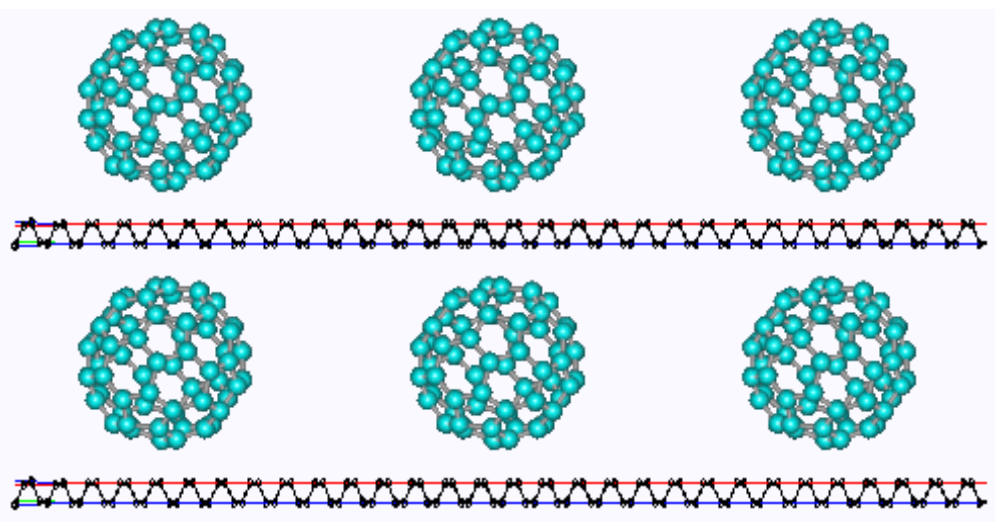

Fig. 1: Polymer chain with diffusing polaron surrounded by fullerene molecules

Polymer solar cells usually consist of an electron- or hole-blocking layer on top of an indium tin oxide (ITO) conductive glass followed by electron donor and an electron acceptor (in the case of bulk heterojunction solar cells), a hole or electron blocking layer, and metal electrode on top. The nature and order of the blocking layers – as well as the nature of the metal electrode – depends on whether the cell follows a regular or an inverted device architecture. In an inverted cell, the electric charges exit the device in the opposite direction as in a normal device because the positive and negative electrodes are reversed. Inverted cells can utilize cathodes out of a more suitable material; inverted OPVs enjoy longer lifetimes than regularly structured OPVs, and they usually show higher efficiencies compared with the conventional counterparts.[16]

In bulk heterojunction polymer solar cells, light generates excitons. Subsequent charge separation in the interface between an electron donor and acceptor blend within the device's active layer. These charges then transport to the device's electrodes where the charges flow outside the cell, perform work and then re-enter the device on the opposite side. The cell's efficiency is limited by several factors, especially non-geminate recombination. Hole mobility leads to faster conduction across the active layer.[17][18]

Organic photovoltaics are made of electron donor and electron acceptor materials rather than semiconductor p-n junctions. The molecules forming the electron donor region of organic PV cells, where exciton electron-hole pairs are generated, are generally conjugated polymers possessing delocalized π electrons that result from carbon p orbital hybridization. These π electrons can be excited by light in or near the visible part of the spectrum from the molecule's highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO),

denoted by a π -$\pi*$ transition. The energy bandgap between these orbitals determines which wavelength(s) of light can be absorbed.

Unlike in an inorganic crystalline PV cell material, with its band structure and delocalized electrons, excitons in organic photovoltaics are strongly bound with an energy between 0.1 and 1.4 eV. This strong binding occurs because electronic wave functions in organic molecules are more localized, and electrostatic attraction can thus keep the electron and hole together as an exciton. The electron and hole can be dissociated by providing an interface across which the chemical potential of electrons decreases. The material that absorbs the photon is the donor, and the material acquiring the electron is called the acceptor. The polymer chain is the donor and the fullerene is the acceptor. Even after dissociation, the electron and hole may still be joined as a "geminate pair", and an electric field is then required to separate them. The electron and hole must be collected at contacts. If charge carrier mobility is insufficient, the carriers will not reach the contacts, and instead recombine at trap sites or remain in the device as undesirable space charges that oppose the flow of new carriers. The latter problem can occur if electron and hole mobilities are not matched. In that case, space-charge limited photocurrent (SCLP) hampers device performance.
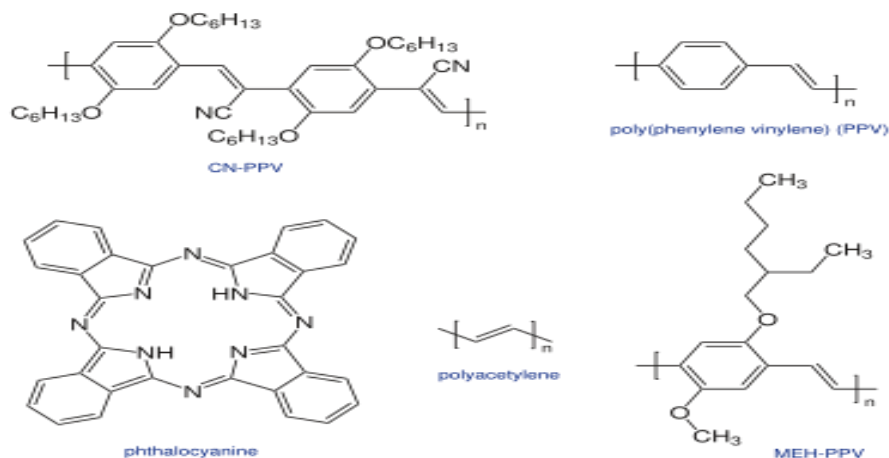

Fig.2. Organic Photovoltaic Materials

Organic photovoltaics can be fabricated with an active polymer and a fullerene-based electron acceptor. Illumination of this system by visible light leads to electron transfer from the polymer to a fullerene molecule. As a result, the formation of a photoinduced quasiparticle, or polaron (P^+), occurs on the polymer chain and the fullerene becomes a radical anion (C^-_{60}). Polarons are highly mobile and can diffuse away. The simplest organic PV device features a planar heterojunction . A film of organic active material (polymer or small molecule), of electron donor or electron acceptor type is sandwiched between contacts. Excitons created in the active material may diffuse before recombining and separate, hole and electron diffusing to its specific collecting electrode. Because charge carriers have diffusion lengths of just 3–10 nm in typical amorphous organic semiconductors, planar cells must be thin, but the thin cells absorb light less well. Bulk heterojunctions (BHJs) address this shortcoming. In a BHJ, a blend of electron donor and acceptor materials is cast as a mixture, which then phase-separates. Regions of each material in the device are separated by only several nanometers, a distance suited for carrier diffusion. BHJs require sensitive control over materials morphology on the nanoscale. Important variables include materials, solvents and the donor-acceptor weight ratio.

The next logical step beyond BHJs are ordered nanomaterials for solar cells, or ordered heterojunctions (OHJs). OHJs minimize the variability associated with BHJs. OHJs are generally hybrids of ordered inorganic materials and organic active regions. For example, a photovoltaic polymer can be deposited into pores in a ceramic such as TiO_2. Since holes still must diffuse the length of the pore through the polymer to a contact, OHJs suffer similar thickness limitations. Mitigating the hole mobility bottleneck is key to further enhancing device performance of OHJ's.

Single layer organic photovoltaic cells are the simplest form. These cells are made by sandwiching a layer of organic electronic materials between two metallic conductors, typically a layer of indium tin oxide (ITO) with high work function and a layer of low work function metal such as Aluminum, Magnesium or Calcium. The difference of work function between the two conductors sets up an electric field in the organic layer. When the organic layer absorbs light, electrons will be excited to the LUMO and leave holes in the HOMO, thereby forming excitons. The potential created by the different work functions helps to split the exciton pairs, pulling electrons to the positive electrode (an electrical conductor used to make contact with a non-metallic part of a circuit) and holes to the negative electrode.[12][13][14]

II. DISCUSSION

Bulk heterojunctions have an absorption layer consisting of a nanoscale blend of donor and acceptor materials. The domain sizes of this blend are on the order of nanometers, allowing for excitons with short lifetimes to reach an interface and dissociate due to the large donor-acceptor interfacial area.[29] However, efficient bulk heterojunctions need to maintain large enough domain sizes to form a percolating network that allows the donor materials to reach the hole transporting electrode 1 and the acceptor materials to reach the electron transporting electrode (Electrode 2). Without this percolating network, charges might be trapped in a donor or acceptor rich domain and undergo recombination. Bulk heterojunctions have an advantage over layered photoactive structures because they can be made thick enough for effective photon absorption without the difficult processing involved in orienting a layered structure while retaining similar level of performances.

Bulk heterojunctions are most commonly created by forming a solution containing the two components, casting (e.g., drop casting and spin coating) and then allowing the two phases to separate, usually with the assistance of an annealing step. The two components will self-assemble into an interpenetrating network connecting the two electrodes.[30] They are normally composed of a conjugated molecule based donor and fullerene based acceptor. The nanostructural morphology of bulk heterojunctions tends to be difficult to control, but is critical to photovoltaic performance.

After the capture of a photon, electrons move to the acceptor domains, then are carried through the device and collected by one electrode, and holes move in the opposite direction and collected at the other side. If the dispersion of the two materials is too fine, it will result in poor charge transfer through the layer.[13][14][19][31]

Most bulk heterojunction cells use two components, although three-component cells have been explored. The third component, a secondary p-type donor polymer, acts to absorb light in a different region of the solar spectrum. This in theory increases the amount of absorbed light. These ternary cells operate through one of three distinct mechanisms: charge transfer, energy transfer or parallel-linkage.

In charge transfer, both donors contribute directly to the generation of free charge carriers. Holes pass through only one donor domain before collection at the anode. In energy transfer, only one donor contributes to the production of holes. The second donor acts solely to absorb light, transferring extra energy to the first donor material. In parallel linkage, both donors produce excitons independently, which then migrate to their respective donor/acceptor interfaces and dissociate.[32] Since its active layer largely determines device efficiency, this component's morphology received much attention.[44]

If one material is more soluble in the solvent than the other, it will deposit first on top of the substrate, causing a concentration gradient through the film. This has been demonstrated for poly-3-hexyl thiophene (P3HT), phenyl-C_{61}-butyric acid methyl ester (PCBM) devices where the PCBM tends to accumulate towards the device's bottom upon spin coating from ODCB solutions.[45] This effect is seen because the more soluble component tends to migrate towards the "solvent rich" phase during the coating procedure, accumulating the more soluble component towards the film's bottom, where the solvent remains longer. The thickness of the generated film affects the phases segregation because the dynamics of crystallization and precipitation are different for more concentrated solutions or faster evaporation rates (needed to build thicker devices). Crystalline P3HT enrichment closer to the hole-collecting electrode can only be achieved for relatively thin (100 nm) P3HT/PCBM layers.[46]

The gradients in the initial morphology are then mainly generated by the solvent evaporation rate and the differences in solubility between the donor and acceptor inside the blend. This dependence on solubility has been clearly demonstrated using fullerene derivatives and P3HT.[47] When using solvents which evaporate at a slower rate (as chlorobenzene (CB) or dichlorobenzene (DCB)) you can get larger degrees of vertical separation or aggregation while solvents that evaporate quicker produce a much less effective vertical separation. Larger solubility gradients should lead to more effective vertical separation while smaller gradients should lead to more homogeneous films. These two effects were verified on P3HT:PCBM solar cells.[48][49]

The solvent evaporation speed as well as posterior solvent vapor or thermal annealing procedures were also studied.[50] Blends such as P3HT:PCBM seem to benefit from thermal annealing procedures, while others, such as PTB7:PCBM, seem to show no benefit.[51] In P3HT the benefit seems to come from an increase of crystallinity of the P3HT phase which is generated through an expulsion of PCBM molecules from within these domains. This has been demonstrated through studies of PCBM miscibility in P3HT as well as domain composition changes as a function of annealing times.[52][53][54]

The above hypothesis based on miscibility does not fully explain the efficiency of the devices as solely pure amorphous phases of either donor or acceptor materials never exist within bulk heterojunction devices. A 2010 paper[55] suggested that current models that assume pure phases and discrete interfaces might fail given the absence of pure amorphous regions. Since current models assume phase separation at interfaces without any consideration for phase purity, the models might need to be changed.

The thermal annealing procedure varies depending on precisely when it is applied. Since vertical species migration is partly determined by the surface tension between the active layer and either air or another layer, annealing before or after the deposition of additional layers (most often the metal cathode) affects the result. In the case of P3HT:PCBM solar cells vertical migration is improved when cells are annealed after the deposition of the metal cathode.

Donor or acceptor accumulation next to the adjacent layers might be beneficial as these accumulations can lead to hole or electron blocking effects which might benefit device performance. In 2009 the difference in vertical distribution on P3HT:PCBM solar cells was shown to cause problems with electron mobility which ends up with the yielding of very poor device efficiencies.[56] Simple changes to device architecture – spin coating a thin layer of PCBM on top of the P3HT – greatly enhance cell reproducibility, by providing reproducible vertical separation between device components. Since higher contact between the PCBM and the cathode is required for better efficiencies, this largely increases device reproducibility.

III. RESULTS

Transparent or semi-transparent PSCs allow for the absorption of low- or high-energy photons outside the visible spectrum, thus optimizing its sunlight harnessing capabilities and covering a broader absorption spectra.[77][78] These types of PSCs are ideal for capturing near-infrared or ultraviolet photons due to its low inherent sensitivity to photons within the visible spectrum. Typical PSCs utilize opaque metal electrodes that limit its transparency, and thus its performance.[77] The absorber layer of PSCs are intrinsically semi-transparent.[79] Thus, one approach to achieving a visibly transparent PSC is to modify the top electrode to make it more transparent. Materials such as ITO, ultra-thin metals, metal grids, graphene, and carbon nanotubes have been used to fabricate semi-transparent top electrodes.[80][81] Yet, the performance of transparent PSCs have shown to be lacking when compared to their opaque electrode PSC counterparts.[82] When the top electrode is made transparent, the cell's ability to trap the electromagnetic field in the absorber layer decreases, resulting in a low PCE. An extensive amount of research is currently being conducted to improve the PCE of such cells.[80] These types of PSCs have been applied to building-integrated photovoltaics, tandem devices, and portable electronics.[77][81][82]

Infrared polymer cells

Infrared cells preferentially absorb light in the infrared range rather than visible wavelengths. A 2010 study developed infrared-transparent PSCs with a CNT film top electrode on the back side and an ITO glass layer on the front side allowing for optical transmittance from both sides of the cell. A ZnO layer was placed on top of the ITO

with a P3HT:PCBM layer being added to the ZnO, thus creating an ITO/ZnO/P3HT:PCBM/CNT (bottom to top) cell. It was observed that the top CNT electrode and bottom ITO electrode both exhibited 80% transmittance within a 500 nm to 2.5 um spectra. The cell itself had an optical transmittance of 80% in the 670 nm to 1.2 um range, 60% in the 1.2 um to 2.5 um range. Conversely, a control cell with an Ag top electrode resulted in no transmittance within this spectra. Additionally, the cell had a relatively low transmittance in the visible region due to the high visible absorbance of the P3HT:PCBM layer. Such cells can be applied to tandem devices and the vertical assembly of PSCs.[77]

As of 2012, infrared cells were nearly 70% transparent to visible light. The cells allegedly can be made in high volume at low cost using solution processing. The cells employ silver nanowire/titanium dioxide composite films as the top electrode, replacing conventional opaque metal electrodes. With this combination, 4% power-conversion efficiency was achieved.[83]

In 2014, near-infrared polymer solar cells based on a copolymer of naphthodithiophene diimide and bithiophene (PNDTI-BT-DT) were fabricated in combination with PTB7 as an electron donor. Both PNDTI-BT-DT and PTB7 formed a crystalline structure in the blend films similar to in the pristine films, leading to the efficient charge generation contributed from both polymers.[84]

Much research has been focused on developing a transparent top electrode for PSCs. However, a 2014 study explored optimizing the active layer of semi-transparent PSCs. The researchers proposed a semi-transparent PSC with enhanced efficiency that utilizes both narrow bandgap polymer donor, PTB7-Th, and non-fullerene acceptor, IHIC. The results of this study showed that the proposed PSC exhibited high transmittance and absorption in the infrared spectrum but low absorption in the visible spectrum. This cell showed to be relatively stable and have a maximum PCE of 9.77%, which, as of 2014, is the highest reported PCE value.[85]

Organic photovoltaics, similar to inorganic photovoltaics, are generally characterized through current-voltage analysis.[86] This analysis provides multiple device metrics values that are used to understand device performance. One of the most crucial metrics is the Power Conversion Efficiency (PCE).
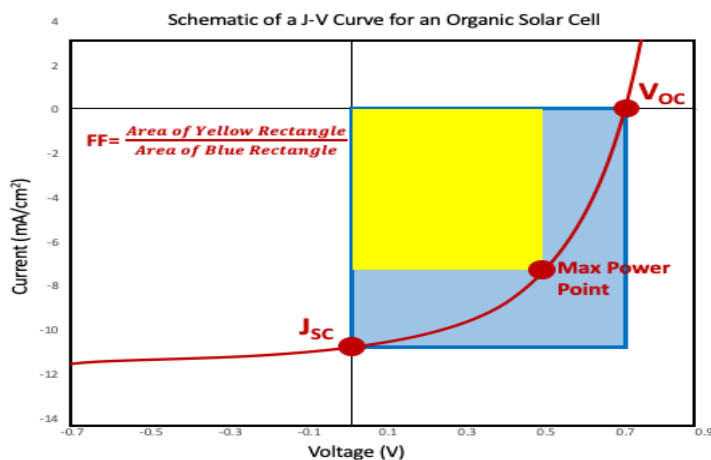

Fig.3 -PCE (η) is proportional to the product of the short-circuit current (J_{SC}), the open-circuit voltage (V_{OC}), and the fill factor (FF), all of which can be determined from a current-voltage curve.

Where P_{in} is the incident solar power.

The short circuit current (Jsc), is the maximum photocurrent generation value.[87] It corresponds to the y-intercept value of standard current-voltage curve in which current is plotted along the y-axis and voltage is plotted along the x-axis. Within organic solar cells, the short circuit current can be impacted by a variety of material factors. These

include the mobility of charge carriers, the optical absorption profile and general energetic driving forces that lead to a more efficient extraction of charge carriers [87]

The open-circuit voltage (Voc) is the voltage when there is no current running through the device.[87] This corresponds to the x-intercept on a current-voltage curve. Within bulk heterojunction organic photovoltaic devices, this value is highly dependent on HOMO and LUMO energy levels and work functions for the active layer materials [87]

Since power is the product of voltage and current, the maximum power point occurs when the product between voltage and current is maximized.

The fill factor, FF, can be thought of as the "squareness" of a current voltage curve.[86] It is the quotient of the maximum power value and the product of the open-circuit voltage and short circuit current.[86] This is shown in the image above as the ratio of the area of the yellow rectangle to the greater blue rectangle. For organic photovoltaics, this fill factor is essentially a measure of how efficiently generated charges are extracted from the device.[87] This can be thought of as a "competition" between charges transporting through the device, and charges that recombine.[87]

A major issue surrounding polymer solar cells is the low Power Conversion Efficiency (PCE) of fabricated cells. In order to be considered commercially viable, PSCs must be able to achieve at least 10–15% efficiency[88]—this is already much lower than inorganic PVs. However, due to the low cost of polymer solar cells, a 10–15% efficiency is commercially viable.

Recent advances in polymer solar cell performance have resulted from compressing the bandgap to enhance short-circuit current while lowering the Highest Occupied Molecular Orbital (HOMO) to increase open-circuit voltage. However, PSCs still suffer from low fill factors (typically below 70%). However, as of 2013, researchers have been able to fabricate PSCs with fill factors of over 75%. Scientists have been able to accomplish via an inverted BHJ and by using nonconventional donor / acceptor combinations.[89]

IV. CONCLUSIONS

One major area of current research is the use of non-fullerene acceptors. While fullerene acceptors have been the standard for most organic photovoltaics due to their compatibility within bulk heterojunction cell designs as well as their good transport properties, they do have some fallbacks that are leading researchers to attempt to find alternatives.[119] Some negatives of fullerene acceptors include their instability, that they are somewhat limited in energy-tunability and they have poor optical absorption.[119] Researchers have developed small molecule acceptors that due to their good energy tunability, can exhibit high open-circuit voltages.[119] Combining a polymer donor (D18) with a small molecule acceptor (Y6), scientists have fabricated organic solar cells in the laboratory giving high efficiencies over 18%.[120] However, there are still major challenges with non-fullerene acceptors, including the low charge carrier mobilities of small molecule acceptors, and that the sheer number of possible molecules is overwhelming for the research community.[119]

A challenge facing the development of organic solar cells utilizing non-fullerene acceptors (NFAs) is the selection of a solvent that has a high boiling point and is environmentally friendly, whereas conventional solvents such as chloroform (CF) tend to exhibit low boiling points and toxicity. Such a solvent is required for further scale-up of organic solar cells, but has also been associated with decreases in PCE due to poor solubility of donor and acceptor materials within the solvent. Appending alkyl chains to NFAs has led to increases in solubility but decreases in molecular packing (π-stacking), which leads to no net impact on PCE. The use of guest assistance has been found to benefit both solubility and molecular packing. A guest molecule named BTO with oligo(ethylene glycol) (OEG) side chains used in conjunction with the NFA Y6 as the acceptor, PM6 as the donor, and paraxylene (PX) as the high-melting-point and sustainable solvent led to an increase in PCE from 11% to over 16%, regarded an acceptable level of efficiency. [121] A further modification that has been successful in the development of cleaner organic photovoltaics is the hot-spin coating of substrates by non-halogenated solvents. It was found that the temperature at which hot-spin coating was operated altered the solution to solid phase evolution of the acceptor-donor blends so that higher temperatures resulted in a higher acceptor concentration in the surface of the substrate. This is because

higher temperatures facilitated decreased aggregation and precipitation, allowing the substrate to retain a higher acceptor concentration. In an experiment, organic solar cells constructed with ternary blends of PM6 donor and Y6-1O and BO-4Cl acceptors and various non-halogenated solvents including o-xylene and toluene exhibited PCE values of over 18%, which are the most efficient organic photovoltaics constructed with non-halogenated solvents, to date. Further morphological analyses showed that the hot-spun OPVs prepared with non-halogenated solvents exhibited similar morphological characteristics to that of OPVs prepared with halogenated solvents. [122]

Small molecules are also being heavily researched to act as donor materials, potentially replacing polymeric donors. Since small molecules do not vary in molecular weights the way polymers do, they would require less purification steps and are less susceptible to macromolecule defects and kinks that can create trap states leading to recombination.[123] Recent research has shown that high-performing small molecular donor structures tend to have planar 2-D structures and can aggregate or self assemble.[123] Sine performance of these devices is highly depended on active layer morphology, present research is continuing to investigate small molecule possibilities, and optimize device morphology through processes such as annealing for various materials.[123]

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