Making Heterojunction Solar Cell Structure with Two Doner Polymer Materials and the Effect of Annealing Temperature

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Abstract—The organic solar cells have narrower absorption spectrum than inorganic ones, making the task of widening such narrow absorption spectrum of the organic solar cell extremely important for increasing the efficiency of organic solar cells. There has been studies on creating a tandem structure with donor materials with complementary absorption spectrums to maximize the absorption spectrum, but this method results in a complex manufacturing process, because tandem solar cells (whether serialized or parallelized) required intermediate layers. One of the proposed solutions is manufacturing solar cells with heterojunction structure, called a ternary solar cell. In this study, a simple heterojunction ternary solar cell was made using P3HT and PTB7 (two polymer materials with stepped energy levels) as donor materials, and PC61BM (a fullerene conductor) as a common acceptor. P3HT and PTB7 each have an absorption peak of 550nm and 650nm, capable of absorbing both relatively short and long wavelengths of light. Therefore, this structure increased the absorption spectrum and the photo current compared to when only one material was used. The P3HT:PC61BM solar cell had JSC of 8.473mA/cm2, PCE of 2.97%, and the PTB7:PC61BM solar cell had JSC of 10.96mA/cm2, PCE of 3.77%, but the P3HT:PTB7:PC61BM ternary structure had JSC of 11.949mA/cm2, PCE of 4.02%. Also, the current-voltage characteristics of the P3HT:PTB7:PC61BM ternary solar cell was measured under annealing temperatures from 80°C to 140°C to find the optimal annealing temperature for the ternary solar cell. The optimal annealing temperature was found to be 90°C, which is closer to the 80°C of PTB7 (which is weaker to heat), which confirmed that if two materials with different heat stability were used in a ternary structure, the one with the lower annealing temperature restricted the total efficiency.

Index Terms—Absorption spectrum, fullerene derivatives, Heterojunction structure (ternary solar cell), organic solar cell, polymer materials with stepped energy levels, optimal annealing temperature.

I. INTRODUCTION

Solar energy has become crucially important as the need for a clean, renewable energy rose. The viability of solar cells first became apparent in 1839 when a French physicist called E. Becquerel discovered the photovoltaic effect with electrodes. [1] The first demonstration of a solar cell happened in 1954 when D.M. Chapin and Bell laboratories demonstrated the world's first solar cell with 5% efficiency. [2] Solar cells utilize the energy of the photons to generate charge. When a photon hits a semiconductor with a small energy band gap, the electron can jump the band gap, creating two charges: an electron and an electron hole. These two charges are directed into the negative and positive electrodes respectively, where the charges generate electricity [3].

The first generation solar cells were inorganic solar cells, which used inorganic semiconductors including monocrystalline silicon cells, polycrystalline silicon cells, amorphous silicon cells, and hybrid silicon cells. [4] Since its advent in Bell Laboratories in the 1950s, inorganic silicon based solar cells have improved dramatically. A number of techniques such as quantum dot, III-V compound semiconductor, and thin silicon films have been developed to improve the efficiency of inorganic solar cells. [5] Meanwhile, there have been recent advances in research concerning solar cells using organic semiconductors. Organic solar cells are made of organic semiconductors that are constituted by conjugated pi bonded molecules or polymers. Researchers on the organic solar cells have concluded that organic solar cells may be a viable alternative to inorganic semiconductors because organic solar cells have strong advantages over inorganic solar cells in terms of efficiency, manufacturing cost, and integration into other products. [6] The first organic solar cell was demonstrated in 1980s by Tang of Kodak using low polymer compound semi-conductors. [7] These organic solar cells show promise of developing solar cells with low manufacturing costs using methods such as low-temperature manufacturing, solution manufacturing, and printing manufacturing. Also, the physical property of organic semiconductors and solar cells of being lightweight and flexible compared to their inorganic counterparts makes organic solar cells advantageous to inorganic solar cells [8].

While organic solar cells have many advantages to inorganic solar cells, organic semiconductors have relatively large energy band gaps compared to their inorganic counterparts, and can only absorb part of the solar spectrum. Therefore, the efficiency of the organic solar cells is much lower than that of the inorganic solar cells. Because of this, to make organic solar cells viable, there is a need to increase the efficiency of organic solar cells [9].

Several possible solutions to this problem have been suggested, and one of these solutions is using two different organic semiconductors with complimentary absorption spectrums in a tandem structure. [10] This method has shown some success in improving the efficiency of organic solar cells, but it is mostly considered not to be viable because of the complicated lamination process. The two optically active layers have to be laminated in turn, which results in a complicated and expensive production process [11].

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In this paper, the ternary solar cell structure has been developed as an alternative to a tandem structure. With the structure, two high polymer donor materials and one acceptor is used to form a bulk hetero junction structure that should have the same effect as a parallel structure. To determine the factor annealing temperature plays on the efficiency of the structure, two high polymer donor materials and a high polymer fullerene conductor with stepped energy levels were used to create an organic solar cell.

II. THEORETICAL BACKGROUND

A. The Operating Principle of Solar Cell

Fig. 1 shows the basic structure and operation of an organic solar cell. To generate electricity from photons, first, photons pass through the transparent electrode, and is absorbed in the photoactive layer. (1 of Fig. 1) In an organic solar cell, the photoactive layer is composed of high polymer donors and acceptors that play the same role as the p-type inorganic semiconductor and the n-type inorganic semiconductor, respectively.

Energy bands that are found in inorganic solar cells are reproduced in organic solar cells by using conjugated polymers. From the energy levels found from the conjugations, it is possible to find the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), which play the same role as valence bands and conduction bands in the inorganic solar semiconductor, respectively.

Similar to how the valence bands and conduction bands are used in an inorganic solar cell, when a photon with high energy is absorbed, the energy is used by electrons to jump the energy band gap from HOMO to LUMO. This results in an electron in LUMO, and an electron hole in HOMO.

The electron – electron hole pair form an exciton because of the Coulomb force attraction. (2 of Fig. 1) Extra energy is needed to break the exciton bond, and this energy is provided by electric field that forms because of the difference in electronegativity between the donor and the acceptor.

When the exiton is broken by the electric field, it forms two charges, an electron and an electron hole. (3 and 4 of Fig. 1) Once free, the free electron and the free electron hole go to the electron transport layer (ETL) and the hole transport layer (HTL) which transports them to the anode and the cathode, respectively. (6 of Fig. 1)



Fig. 1. The structure and operation of the organic solar cell.

B. Photovoltaic Parameters

1) Open circuit voltage, VOC

The open circuit voltage is the voltage when the circuit connected to the solar cell is open. When the circuit is open, no current flows in the external circuit connected to the solar cell, but the voltage is at the maximum the solar cell can provide. This open circuit voltage is related to the energy level of the photoactive layer, and has the empirical formula below [12].

$$V_{\rm OC} = \frac{1}{e} \left(\left| \rm HOMO_{\rm Donor} \right| - \left| \rm LUMO_{\rm Acceptor} \right| \right) - 0.3 \rm V$$
 (1)

2) Short circuit current, ISC

The short circuit current is the current when the circuit connected to the solar cell is short-circuited. When the circuit is short, maximum current flows in the external circuit. The short circuit voltage is related to the quantum conversion efficiency of the photoactive layer, and has the empirical formula below.

3) Fill factor

While the theoretical maximum voltage of a solar cell is V_{OC} , because of real life factors such as resistance, the actual maximum voltage is lower than the V_{OC} . Similarly, while the theoretical maximum current of a solar cell is I_{SC} , because of the real life factors such as resistance, the actual maximum current is lower than I_{SC} . The fill factor is a measure of the discrepancy between the theoretical and actual maximum voltage and current, and represents how close the solar cell is to an ideal solar cell. The equation below is used to calculate the fill factor (Fig. 2).

$$FF = \frac{V_{\rm MP}I_{\rm MP}}{V_{\rm OC}I_{\rm SC}} \tag{2}$$

From the fill factor, the energy conversion efficiency is given by the equation below.



C. Ternary Organic Solar Cells

In the experiment, the high polymer donor materials used

are poly (3-hexylthiophene) (P3HT, Ricke Metals), and Thieno[3,4-*b*]-thiophene/benzodithiophene (PTB7, One-Material), and their molecular structure is shown in Fig. 3. The common accepter used was [6,6]-phenyl C61 butyric acid methyl ester ($PC_{61}BM$, Nano-C), also shown in Fig. 3.

 $PC_{61}BM$ is a fullerene derived conductor, and has the molecular structure of a Methyl group attached to C60, as shown in Fig. 3 [13].



As shown in the energy level diagram of Fig. 4, P3HT, PTB7, and $PC_{61}BM$ has stepped energy levels. P3HT has a LUMO energy level of -2.9eV, and a HOMO energy level of -5.1eV, while PTB7 has a LUMO energy level of -3.3eV, and a HOMO energy level of -5.2eV, and PC₆₁BM had a LUMO energy level of -3.7eV, and a HOMO energy level of -6.1eV, which, as shown in Fig. 4, results in a stepped energy level.

P3HT and PTB7, which are the two supplementary donor materials used, have energy band gaps of 2.2eV and 1.9eV, respectively, which results in absorption wavelengths of 560nm and 650nm, respectively. (Fig. 5) Thus, P3HT absorbs photons with relatively short wavelengths, and PTB7 absorbs photons with relatively long wavelengths.

Since the two donor materials have complimentary absorption spectrums, the organic solar cell with a ternary structure utilizing these two materials will have increased efficiency since both photons with relatively high and low wavelengths can be absorbed.

When compared to a tandem solar cell connected either in series or in parallel, the ternary solar cell will act more like a tandem solar cell connected in parallel, because the energy level difference between P3HT and PC₆₁BM is much larger than the energy level difference between P3HT and PTB7 or PTB7 and PC₆₁BM, charges will be transferred from P3HT to PC₆₁BM much more frequently than from P3HT to PTB7. But, because there still will be some charges transferred from P3HT to PTB7, the current of the ternary structure is expected to be less than the current of a parallel tandem solar cell structure.







Fig. 5. Solar Irradiance and the absorption spectrum of P3HT and PTB7.

III. TEST METHOD

To create the solar cell, patterned Indium Tin Oxide (ITO) glass was used to laminate the photoactive layers. The surface resistivity of the ITO substrate was determined to be 15 Ω / sq. The ITO glass was prepared by complicated process. First, the ITO glass was washed with ultrasonic waves in the sonicator for 20 minutes with distilled water. Then, it was immersed in acetone and heated for 20 minutes. After that, it was immersed in isopropyl alcohol, and then heated for 20 minutes. Finally, the ITO glass was dried for 24 hours in an oven, and then exposed to UV ozone for 20 minutes. This UV treatment insures that the Poly (3,4-ethylenedioxythiophene): poly (styrenesulfonic acid) (PEDOT: PSS, Clevios PVP AI 4083), which was used as the HTL, laminated well. On the UV treated ITO glass, a 40nm thick PEDOT: PSS layer was spin coated with a spin coater at 3000rpm for 40 seconds. After that, the glass was heated at 150 ° C for 20 minutes to evaporate the water. For the single solar cells of P3HT:PC₆₁BM and PTB7:PC₆₁BM that served as controls, The P3HT:PC₆₁BM solution and the PTB7: PC₆₁BM solution was made 24 hours in advance. For the P3HT:PC₆₁BM solution, 15mg of P3HT and 10mg of PC₆₁BM was placed in 1ml of 1, 2-dichlorobenzene (ODCB) solvent, then was stirred for 24 hours at 65 °C using magnets. For the PTB7:PC₆₁BM solution, 7.5mg of PTB7 and 15mg of PC61BM was placed in 1ml of ODCB solvent, then was stirred for 24 hours at 65 °C using magnets. Then, the P3HT:PC₆₁BM was spin coated over the PEDOT: PSS hole-transport layer at 900 rpm for 40 seconds, and the PTB7:PC61BM was spin coated over the PEDOT: PSS hole-transport layer at 1000rpm for 1 minute. After the photoactive layers had been laminated, both controls were kept at room temperature for 1 hour to let the leftover ODCB solvent evaporate. After that, a LiF layer with 0.5nm thickness and an aluminum layer of 150nm that was used as a cathode was vacuum laminated under 10⁻⁷Torr of pressure.

For the P3HT:PTB7:PC₆₁BM solution for the ternary solar cell, 15mg of P3HT, 7.5mg of PTB7, and 25mg of PC₆₁BM was placed in 2ml of ODCB solvent then was stirred for 24 °C Then, hours at 65 using magnets. the P3HT:PTB7:PC₆₁BM was spin coated over the PEDOT: PSS hole-transport layer at 1000rpm for 1 minute. Then it was kept at room temperature for 1 hour to let the leftover ODCB solvent evaporate. After that, a LiF layer with 0.5nm thickness and an aluminum layer of 150nm that was used as a cathode was vacuum laminated under 10⁻⁷Torr of pressure. The structure of the two singular solar cells and the ternary solar cell is shown in Fig. 6.



Fig. 6. The structure of the solar cells used in the experiment. (a) P3HT:PC61BM, (b) PTB7:PC61BM, (c) P3HT:PTB7:PC61BM.

The photoactive layer area was $0.3 \text{ cm} \times 0.3 \text{ cm}$, and the IV curves were measured with a Keithley 2400 instrument. A solar light simulator (Newport Thermal Oriel 91192, 1000W) was used as the light source, with 100mW/cm² under AM 1.5G conditions. To investigate the effect of the annealing temperature, P3HT: PC61BM single solar cells were annealed at 60, 80, 100, 120, 140, and 160 °C for 5 minutes, and the IV curve was measured. For PTB7, which is relatively weak to heat compared to P3HT, the solar cells were annealed at 60, 70, 80, 90, and 100 °C for 5 minutes, and the IV curve was measured. For the ternary solar cell, the P3HT:PTB7:PC61BM solar cell was annealed at 80, 90, 100, 120, and 140 °C for 5 minutes, then the IV curve was measured.

IV. RESULT AND DISCUSSION

A. P3HT:PC₆₁BM Single Solar Cell

As the two donor materials of P3HT and PTB7 have complimentary absorption spectrums (as shown in Fig. 5), theoretically, the current would increase significantly in a ternary structure compared to a control solar cell. The I-V diagram of the P3HT:PTB7:PC₆₁BM ternary solar cell under different annealing temperatures from 80 to 140°C is shown in Fig. 7. The J_{SC} generally have values over 10mA/cm² for almost all temperatures, and have the maximum value of 11.949mA/cm² at the annealing temperature of 90°C. Compared to the current of singular solar cells, the current of the ternary solar cells are significantly larger: the maximum current of the P3HT:PC₆₁BM singular solar cell was 8.473mA/cm², and the maximum current of the PTB7:PC₆₁BM singular solar cell was 10.96mA/cm².

The increased current of the ternary solar cells show that the increased absorption spectrum allowed for more absorption of photons, and that much of the photons were converted into charges.



Fig. 7. The I-V curve of the P3HT:PTB7:PC61BM ternary solar cell according to temperature.

The variation of various parameters due to annealing temperatures is shown in Fig. 8 to Fig. 11. The V_{OC} showed almost no change in temperature 80 to 100°C but had a slight decrease in higher temperatures. The V_{OC} of PTB7:PC₆₁BM is around 0.71 to 0.72V, and the Voc of P3HT:PC61BM is around 0.6V, so the effect of annealing temperatures on PTB7 is more apparent in lower temperatures where P3HT is not significantly affected, and the effect on P3HT is more apparent in higher temperatures. With 120, 140°C annealing temperatures, while the current of the ternary solar cell is higher than that of P3HT, the actual efficiency is 2.39% and 2.35%, even lower than that of the P3HT singular solar cell (2.97%). This drop in efficiency can be reasonably assigned to the decrease in FF. The reason for FF decreasing even with a I_{SC} increase is thought to be because, as the annealing temperature increased, the heat-weak PTB7 actually hinders charge transportation within the photoactive layer instead of helping. The P3HT:PTB7:PC61BM ternary structure had its maximum efficiency at 90°C, where Voc was 0.7112V, JSC was 11.949mA/cm², FF was 47.28, and PCE was 4.02%. the maximum efficiency increased by 0.25% from the 3.77% efficiency of the PTB7 singular solar cell. Although the increase percentage of efficiency is not large, the viability of using two donor materials with complimentary absorption spectrums in a ternary hetero junction structure to bring about current and efficiency increase is meaningful. Also, by investigating changes resulting from annealing temperatures, the conclusion that when two donor materials with a large difference in optimal annealing temperatures are used in a ternary hetero junction structure, the donor material with the lower annealing temperature could be the limiting element for the efficiency of the total efficiency. So to optimize the efficiency of solar cells with ternary structures, it would be best to use donor materials with similar optimal annealing temperatures. Also, if factors other than annealing temperature that affect the FF could be found, there is a possibility of a much significant increase in efficiency.



Fig. 8. VOC vs Annealing temperature.



Fig. 9. JSC vs Annealing temperature.



B. PTB7:PC₆₁BM Single Solar Cell

The IV curve of the PTB7:PC₆₁BM single solar cell is shown in Fig. 12. For non-annealed solar cells, the V_{OC} was 0.7042V, J_{SC} was 8.694mA/cm², FF was 47.16%, and PCE was 2.89%. As the annealing temperature increases, the efficiency of the solar cell rises. But as PTB7 is much weaker to heat than P3HT, the PTB7 solar cell showed maximum efficiency around 80°C, which is much lower than the P3HT solar cell's 140°C, and the extent by which efficiency increased due to annealing temperature was much lower than that of P3HT as well. The maximum efficiency is reached with the solar cell annealed at 80°C, with V_{OC} at 0.6948V, J_{SC} at 10.96mA/cm², FF at 49.51%, and PCE at 3.77%. As the result of annealing, the V_{OC} decreased slightly without much change, the J_{SC} increased by 26%, and the FF by 5%. As a result, the PCE increased by about 30%. This result shows that 80°C is the optimal annealing temperature for the PTB7:PC₆₁BM single solar cell. In the P3HT solar cell, the annealing temperature had a significant effect on FF, but in the PTB7 solar cell, it affected the current more. Unlike P3HT, which held up well in high temperatures, the efficiency of PTB7, while having not much change in temperatures below 80°C, decreased dramatically in temperatures above 80°C.



Fig. 12. The I-V curve of the PTB7:PC61BM single solar cell according to temperature.

The variation of various parameters due to annealing temperatures is shown in Fig. 13 to Fig. 16. The V_{OC} had no significant change until 90°C, then decreased dramatically under temperatures higher than 90°C, and the J_{SC} had its maximum value at around 70 and 80°C. FF had no significant change until 80, 90°C, but decreased dramatically in temperatures higher than that.



Fig. 16. PCE vs Annealing temperature.

C. P3HT:PTB7:PC₆₁BM Ternary Solar Cells

From the results of the two control experiments, it is reasonable to assume that the P3HT:PTB7:PC₆₁BM ternary solar cell will have an optimal annealing temperature between 80 and 140°C. As the two donor materials of P3HT and PTB7 have complimentary absorption spectrums (as shown in Fig. 5), the hypothesis was that the current would significantly increase with the hetero junction structure. The I-V diagram of the P3HT:PTB7:PC61BM ternary solar cell under different annealing temperatures from 80 to 140°C is shown in Fig. 17. The J_{SC} generally have values over 10mA/cm² for almost all temperatures, and have the maximum value of 11.949mA/cm² at the annealing temperature of 90°C. compared to the current of singular solar cells, the current of the ternary solar cells are significantly larger: the maximum current of the P3HT:PC₆₁BM singular solar cell was 8.473mA/cm², and the maximum current of the PTB7:PC61BM singular solar cell was 10.96mA/cm². The increased current of the ternary solar cells show that the increased absorption spectrum allowed for more absorption of photons, and that much of the photons were converted into charges.



Fig. 17. The I-V curve of the P3HT:PTB7:PC61BM ternary solar cell according to temperature.

The variation of various parameters due to annealing temperatures is shown in Fig. 18 to Fig. 21. The V_{OC} showed almost no change in temperature 80 to 100°C but had a slight decrease in higher temperatures. The V_{OC} of PTB7:PC₆₁BM is around 0.71 to 0.72V, and the V_{OC} of P3HT:PC₆₁BM is around 0.6V, so the effect of annealing temperatures on PTB7 is more apparent in lower temperatures where P3HT is not significantly affected, and the effect on P3HT is more apparent in higher temperatures. With 120, 140°C annealing temperatures, while the current of the ternary solar cell is higher than that of P3HT, the actual efficiency is 2.39% and 2.35%, even lower than that of the P3HT singular solar cell (2.97%). This drop in efficiency can be reasonably assigned to the decrease in FF. The reason for FF decreasing even with a I_{SC} increase is thought to be because, as the annealing temperature increased, the heat-weak PTB7 actually hinders charge transportation within the photoactive layer instead of helping. The P3HT:PTB7:PC₆₁BM ternary structure had its maximum efficiency at 90°C, where V_{OC} was 0.7112V, J_{SC} was 11.949mA/cm², FF was 47.28, and PCE was 4.02%. the maximum efficiency increased by 0.25% from the 3.77% efficiency of the PTB7 singular solar cell. Although the increase percentage of efficiency is not large, the viability of using two donor materials with complimentary absorption spectrums in a ternary hetero junction structure to bring about current and efficiency increase is meaningful. Also, by investigating changes resulting from annealing temperatures, the conclusion that when two donor materials with a large difference in optimal annealing temperatures are used in a ternary hetero junction structure, the donor material with the lower annealing temperature could be the limiting element for the efficiency of the total efficiency. So to optimize the efficiency of solar cells with ternary structures, it would be best to use donor materials with similar optimal annealing temperatures. Also, if factors other than annealing temperature that affect the FF could be found, there is a possibility of a much significant increase in efficiency.





V. CONCLUSION

To increase the efficiency of organic solar cells, a ternary structure using two donor materials and one acceptor was developed and tested. Two high polymer donor materials with complimentary absorption spectrums were used with a high polymer acceptor material to create a ternary structure. The donor materials P3HT with an absorption spectrum of 550nm and PTB7 with an absorption spectrum of 650nm was used with an acceptor material PC₆₁BM. Because the energy gap between P3HT and PC₆₁BM is much larger than the gap between P3HT and PTB7 or the gap between PTB7 and PC₆₁BM, charges in P3HT are expected to jump to PC₆₁BM directly rather than jump to PTB7, then to PC₆₁BM, which implies a result similar to a parallel solar cell, or an increase in current.

To optimize the efficiency of ternary organic solar cells using two donor materials with complimentary absorption spectrums, the effect of annealing temperature on the efficiency of the solar cell was investigated. Since the solar cell used a ternary structure with two donor materials of P3HT and PTB7, it is reasonable to assume that the optimal annealing temperature will be between the optimal annealing temperature of P3HT and the optimal annealing temperature of P3HT and the optimal annealing temperature of PTB7, which is respectively 140°C and 80°C. The results from using different annealing temperatures from 80°C to 140°C showed that the optimal temperature was approximately 90°C, with the V_{OC} of 0.7112V, the J_{SC} of 11.949mA/cm², the FF of 47.28, and the PCE of 4.02%.

The ternary structure significantly increased the current of the solar cell as expected, and also increased the PCE from 3.77% to 4.02%. Although this is not a huge increase, the results are significant because it showed that it was possible to increase the absorption spectrum, current, and total efficiency of an organic solar cell by using two donor materials with complimentary absorption spectrums in a ternary structure.

The effect of annealing temperature on the effectiveness of the solar cell showed that the annealing temperature of the ternary solar cell is more dependent on the donor material with the lower annealing temperature, which could be considered to be the limiting factor.

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