Improvement of Solar Cell Performance by Annealing Temperature Treatment and Active Layer Ratio

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ABSTRACT: Polymer solar cells are potential candidates for providing energy as an alternative energy source. In this type of solar cell, the active layer of the semiconductor material absorbs light. The P3HT: PPV layer was used as the active layer in this study. This research aimed to determine the impact of annealing and P3HT composition on the performance of solar cells. The annealing temperatures tested in this study were 120 °C, 135 °C, 150 °C, 165 °C, and 180 °C. Based on the findings, the annealing treatment affects the morphological structure of the active layer and the self-ordering crystallinity of P3HT, which PPV hampers. The results of SEM characterization revealed the effect of annealing on the morphological structure of the active layer. An annealing temperature of 150 °C produced the most homogeneous layers. The P3HT: PPV ratio was tested at 1: 1, 2: 1, and 3: 1. The 3:1 ratio yielded the highest efficiency, which was 3.5 percent. In this condition, an electric current of 0.04 mA and a voltage of 4.098 V were also obtained.

KEYWORDS: Active layer ratio; Annealing; Polymer solar cells; P3HT: PPV.

INTRODUCTION

Organic materials and conductive conjugated polymers are popular in solar cell research [1-5]. The conjugated polymer contains an electron π that has been delocalized, allowing it to absorb sun rays, forming conveyor loads and transmitting those loads into electrical energy [6-7]. Conjugated polymer research is advancing because it offers low production costs, a simple technique, and the ability to produce higher conversion efficiencies [8-10]. In addition to conjugated polymers, organic semiconductor molecules can also be used as active materials [11].

P3HT, or poly (3-hexylthiophene-2,5-diyl), is a polythiophene subtype that has received much attention as a polymer solar cell active material [12-13]. P3HT is an electron donor referred to as a p-type semiconductor.

It has a Regular Region (RR) structure that produces good electric conductivity, melts quickly in normal organic fluid, and can form into a thin film using simple techniques such as spin-coating, dip coating, inkjet printing, and roll-to-roll printing [13-16]. P3HT has a molecular weight of 65.5 grams/mol and a work function at the HOMO level of 5.2 eV and the LUMO level of 3.2 eV [17].

Polymer PPV (poly (p-fenilen vinyl)) is a conjugated polymer with acetylene (CH₂) and benzene (C₆H₆) structures [18]. The PPV material is gaining popularity because it is simple to manufacture in thin film or fiber form [19]. PPV polymeric materials are also p-type semiconductors [20]. PPV has π electrons delocalized along the polymer's sequence, as it gives semiconductor

* To whom correspondence should be addressed. + E-mail: hardeli1@yahoo.com & hardelikimia123@gmail.com 1021-9986/2022/11/3841-3851 11/\$/6.01 characteristics [21]. The orbital energy of a PPV is -5.3 eV for HOMO and -3.2 eV for LUMO [22]. Poly [2-methoxy-5-(2-ethyl-hexyloxy)-1,4-phenylene], or MEH-PPV, is the type of PPV used in this study. MEH-PPV has good organic semiconductor material stability and barrier height enhancement properties and is widely used in electronics [23]. The absorption peaks of the pure MEH-PPV and its composites are located at 507 nm [24].

Polymer P3HT naturally donates electrons in the active layer, while polymer PPV accepts electrons. Light adsorption occurs on the active layer P3HT: PPV is a semiconductor material that produces electrons [25]. The photons' excitation energy (electrons and holes) will transfer to the donor material. The active layer, contaminated by light, will inject the electron into the semiconductor [26]. The resulting electrons and holes in the material will have varying mobilities, resulting in device efficiency [27].

The semiconductor used in this research was ITO glass. Indium Tin Oxide (ITO) thin films have high transparency in the visible area (more than 85%), have a bandgap of 3 eV, and have lower resistivity [28]. ITO consists of In_2O_3 generated by SnO₂. ITO is usually in an armored form and has low conductivity at low temperatures [29].

PEDOT: PSS (Poly (3,4-ethylene dioxythiophene) poly (styrene sulfonate) is a conductive, transparent, flexible, and stable polymer [30]. This polymer serves to load conveyors (positive or negative) to electrodes close to each other, preventing recombination. This layer can act as a path for electrons because of the switch between the active material and the electrode at the proper energy level [31].

The heterojunction is a method commonly used for polymer solar cells. This method is generally divided into the bilayer and the bulk mixture [32]. The material donor and acceptor are heaped together in the bilayer heterojunction method. Meanwhile, in the bulk method used in this study, both the material donor and the material acceptor are mixed. This method was chosen because it has several advantages over the bilayer method, including producing higher efficiencies and absorbing higher waves [33, 34]. The illustration, Fig. 2, shows the structural difference between a bilayer and a bulk heterojunction.

Annealing is a material heating treatment at a specific temperature. In this study, this process helped restore the

crystallite and regularity of the polymer P3HT disrupted by the presence of other polymers (MEH-PPV). As a result, annealing from the active layer in a solar cell may form a polymer structure generality [35]. This treatment may affect interface homogeneity, interlayer mobility, and load mobility. When the active layer of a solar cell is more homogeneous, more sunray energy can be absorbed [36]. This study investigated the effect of annealing at various temperatures on the P3HT: PPV polymer mixture. In this study, annealing was performed at temperatures of 120 °C, 135 °C, 150 °C, 165 °C, and 180 °C.

Aside from the annealing temperature, the comparison amounts of two polymers used significantly impact solar cell performance. In this study, we experimented with different parts of polymer P3HT on the active material. The P3HT amount was increased two and three times over the PPV amount.

Based on the above description, an experiment was conducted to determine the effect of annealing and the ratio of active ingredients in the P3HT: PPV mixture on improving polymer solar cell devices.

EXPERIMENTAL SECTION

Materials and Tools

ITO substrate, beaker glass, stirring spoon, measure glasses, pipette, volumetric pipettes, spatula, watch glass, porcelain crucible, desiccators, absorbent balls, pincers, vial, spray bottle, graphite roller, binder clips, analytical balance, oven vacuum, spin coater, digital multimeter, ultrasonic cleaner, Oven France Etuves XU225, scotch tape, and SEM were the instruments used. Whereas PEDOT: PSS, P3HT, MEH-PPV, chlorobenzene, methanol, carbon, and aquadest were the materials used.

Polymer Solar Cell Components Preparation

• The first step was to do the ITO lithography. The substrate was cleaned before use by performing the following steps: the substrate was placed in a beaker glass half-filled with soap water, cleaned, and dried. It was put in 50 mL of methanol and sonicated for 120 minutes with an ultrasonic cleaner. After that, ITO glass was stored in an aquadest to avoid fat and dust. When the glass substrate was used, it was cleaned in a furnace at 110 °C for 60 minutes. After cleaning and sterilizing, the ITO substrate was given scotch tape on the offset area. It was to mark the part used as an experimental area.



Fig. 1: The structures of (a) P3HT and (b) MEH-PPV.



Fig. 2: The difference of structure between bilayer and bulk heterojunction.



Fig. 3: The structure of assembly polymer solar cell P3HT: PPV.

• Next was to prepare the PEDOT: PPS paste. PEDOT: PPS was scaled to 0.05 g, dissolved in 1.5 mL of aquadest, and stirred until a paste formed.

• The P3HT: PPV mixture was prepared in various ratios.

- The first step was to make a P3HT: PPV (1:1) mixture. 10 mg of P3HT was dissolved in 1 mL of chlorobenzene and thoroughly mixed. 10 mg of PPV was dissolved in 1 mL of chlorobenzene and mixed thoroughly. Both solutions were then mixed and stirred until well combined.

- The second step was to make a P3HT: PPV (2:1) mixture. 20 mg of P3HT was dissolved in 1 mL of chlorobenzene and thoroughly mixed. 10 mg of PPV was dissolved in 1 mL of chlorobenzene and mixed thoroughly. Both solutions

were then mixed and stirred until well combined.

- The last step was to make a P3HT: PPV (3:1) mixture. The procedure was the same as making a 2:1 mixture. The only difference is that the P3HT used was 30 mg.

• The following step was Counter-Electrode Carbon Preparation. Graphite from pencils was used as a carbon resource. Graphite was coated into the conductive area of the ITO substrate. The graphite was then heated at 450 $^{\circ}$ C for 10 minutes to ensure good contact with the carbon particles in the ITO substrate.

Polymer solar cell assembling

After the offset area was coated with scotch tape, the ITO substrate was coated with PEDOT: PPS paste. The layer was then heated in the oven for 60 minutes at 120 °C. Then, it was coated with the polymer active material P3HT: PPV. A spin coating at 600 rpm for 30s was used in the coating process. The active layers were then annealed for 10 minutes at 120 °C, 135 °C, 150 °C, 165 °C, and 180 °C using a vacuum oven. The activated carbon was then placed on the active polymer layer, with the carbon electrode counter facing the active layer. The structure was pinched and held in place with a clip binder.

P3HT: PPV Active Layer Thin Film Characterization

SEM was used to identify the active layer morphology. Material characterization was carried out at a voltage of 20 kV and a compression of 10 pa. The characterization was conducted in the Laboratory of Material Characterization division, Material and Metallurgy major, Industrial Technology faculty, Institut Teknologi Sepuluh Nopember.

Solar Cell Current Electrical Examining

A direct voltage check was performed on the assembled solar cells. A digital multimeter was used to measure the scale resistivity of the solar cells. A UV lamp of 30 W was used as the light source. The efficiency of a solar cell was calculated using the voltage and current results. The mentioned equation was used to calculate the conversion efficiency value of polymer solar cells:

$$\eta = \frac{P_{\text{max}}}{P_{\text{in}}} \times 100\%$$
$$= \frac{V_{\text{max}} X I_{\text{max}}}{I_{\text{cahaya}} X L} \times 100\%$$
[37]

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RESULT AND DISCUSSION

Analyzing the impact of annealing on solar cell electrical properties

The active layer of P3HT and PPV, known as photon energy absorbers and semiconductor materials, are polymers with p-electron conjugation in their structure. The absorption of photons from the active layer of P3HT: PPV influences the measurement results of the electrical properties of solar cells. Electrical features such as voltage and current were examined to determine the amount of efficiency produced by a solar cell. Power testing was performed in the room with a 30 W lamp as a UV source. Using a UV source from the lamp was intended to obtain a consistent and accurate UV source because it was feared that using direct sunlight would result in different light intensities.

The annealing treatment tested on the active layer P3HT: PPV was at temperatures of 120 °C, 135 °C, 150 °C, 165 °C, and 180 °C, where the lowest temperature was chosen to be higher than the glass transition temperature, where the glass transition temperature was around 110 °C. The highest temperature was limited to 180 °C to avoid discoloration of the active layer due to the polymer reaching its melting point [38].

The active polymer molecules in the film will move freely during the annealing process. As a result, P3HT will experience relaxation and the process of reassembling the structure [39]. Figure 4 shows the voltage value of solar cell production for annealing processing.

Figure 4 shows that the voltage generated by solar cells at different annealing temperatures has different values. It was proven that annealing affects the voltage produced by the solar cell. The resulting voltage was 1.014 V at a temperature of 120 °C. A higher voltage, 1.127 V, was obtained at a higher annealing temperature, 135 °C. At an annealing temperature of 150 °C, a significant increase was observed. The voltage obtained was 4.018 V. This was the highest voltage obtained in the experiment because the value decreased as the temperature increased. The voltage obtained at 165 °C annealing temperature was 4.002 V, which is less than the voltage received at 150 °C annealing temperature. The voltage measured at the maximum temperature tested (180 °C) was 1.146 V.

Figure 4 shows that the highest voltage value generated by polymer solar cells using the active layer P3HT: PPV at an annealing temperature of 150 °C is 4,018 V. At lower



Fig. 4: The result of voltage on P3HT: PPV (1:1) at various annealing temperatures

annealing temperatures, a low voltage was obtained, the annealing effect was not maximal. The voltage decreases at temperatures above 150 °C due to the influence of photon energy absorbents on the active layer. According to *Yunzhang Lu*, the more the polymer reaches 200 °C, the more the active layer's color changes as it approaches its melting point [38].

The effect of annealing temperature on current power production was similar to annealing temperature on voltage. It was due to the fact that the current power generated by solar cells was equivalent to the number of absorbent photons and was dependent on the performance of the active layer used [37]. Figure 5 shows the effect of the annealing temperature on the current power generated by the solar cell.

According to the experiment, the current generated at an annealing temperature of 120 °C was 0.01 mA. The increase was observed at an annealing temperature of 135 °C, with a resultant current of 0.02 mA. The highest current produced in the experiment was 0.03 mA at an annealing temperature of 150 °C. Following 150 °C annealings, the current dropped to 0.02 mA for 165 °C and 180 °C annealings.

Figure 5 shows that the maximum current power was discovered when the solar cell was annealed at 150 °C with an existing capacity of 0.03 mA. Thus, the production of current power was relatively small. Current power was influenced by photon light sources [40]. Small photon sources will produce a small current for minimal photon energy absorbed by the active layer. This insignificant current power also causes the hole and heap on the active layer P3HT: PPV to be photon absorbent.



Fig. 5: The result of current power on P3HT: PPV (1:1) at various annealing temperatures.

Based on the scaling of current value and voltage, the highest efficiency at the annealing temperature was as in Fig. 6. According to the findings, the efficiency of fabricated solar cells at a temperature of 120 °C was 0.22 percent. The next tested temperature, 135 °C, resulted in a 0.48 percent increase in efficiency. According to the previous voltage and current values, the highest efficiency of 2.57 percent was obtained at 150 °C. After 150 °C, the efficiency gradually decreases at 165 °C and 180 °C. The efficiency was 1.71 percent and 0.49 percent at these two temperatures, respectively.

The same results were found in a previous study, which discovered that the best annealing temperature for the P3HT: PCBM and PPV: PCBM was 150 °C [26, 41-42]. As a result, thermal annealing is thought to improve cell performance by optimizing donor/acceptor morphology in the BHJ active layer and interfacial contact between the metal electrode and the active layer [42].

Morphology characterization

Annealing is a material heating treatment at a specific temperature that is commonly used to restore the crystallite and regularity of the polymer that has been disrupted by the presence of other polymers [32]. It has also been reported that annealing was helpful for phase separation, removing residual solvents, and influencing conformations and crystallization [43]. As a result, annealing from the active layer may form a generality on the polymer structure in a solar cell. This treatment may affect interface homogeneity, interlayer mobility, and load mobility. Based on the previous results, the optimum temperature for



Fig. 6: polymer solar cell parameter on P3HT: PPV (1:1) at various annealing temperatures.

annealing the active layer of the P3HT: PPV is 150 °C. The appearance of each layer in each treatment variation will be discussed in this section.

The resulting polymer solar cells were made by layering an active layer of P3HT: PPV on a lithography layer of PEDOT: PPS on an ITO glass substrate. The prepared PEDOT: PPS paste was a dark blue paste, whereas the resulting P3HT: PPV active layer was brownish-red. The purpose of analyzing a solar cell using SEM was to investigate the active layer mono structure P3HT: PPV that attaches to the PEDOT: PPS layer. P3HT: PPV, coated in conduction, was heated to reconstruct the polymer structure. The annealing process on the active layer was to control the active layer morphology because it influences load (electron) mobility production. The active layer relaxed when annealing was applied, and the structural assembly process improved because the polymer molecules could move freely on the thin film layer.

In this study, SEM characterization was performed at annealing temperatures of 135 °C, 150 °C, and 165 °C because, in previous experiments, the highest efficiency was found at these three temperatures. Figure 8 shows the SEM results with the surface morphology sample magnified 40,000 times. According to the research findings, samples annealed at 150 °C have a more homogeneous and flatter surface than samples annealed at 135 °C or 165 °C.

The annealing result at 150 °C was a layer with only minor holes. The formation of these tiny holes was caused by areas not entirely covered by the polymer layer deposited during the spin-coating process. According to



Fig.7: (a) PEDOT: PPS paste and (b) P3HT: PPV mixture preparation.



Fig. 8: The result of SEM characterization; surface display on aggrandizement 40.000 times (a) 135 °C, (b) 150 °C and (c) 165 °C.

research, a sample at 150 °C has a more homogeneous surface because P3HT rearrangement is complete at this temperature [39]. At this temperature, it is known that the molecules involved move to make the structure homogeneous and even. This homogeneity and evenness of the active layer facilitate the movement of charges, resulting in better electrical properties of the coating. As a result of all that, the resulting device efficiency was also good. It was in accordance with previous studies where, using the active ingredient mixture of P3HT: PCBM and PPV: PCBM, the maximum annealing temperature was also obtained at 150 degrees Celsius [26,41].

The annealed active layer at 165 °C appears uneven and has more accumulated parts, whereas valleys and hills are more visible at 135 °C. At an annealing temperature of 135 °C, it seems that there are many large depressions. It was because the coating process using spin coating couldn't make the coating flat entirely. Additional processing was required through annealing. But at this temperature, the annealing process was not maximized, so the homogenization and flattening of the samples were still not adequate.

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Meanwhile, during the annealing process at 165 °C, a layer with lumpy parts was formed, while the other elements that did not have lumps appeared flat and homogeneous. Previous research has also found that heating the polymer for an extended time at a high temperature causes it to cluster [44]. The piled-up part could be caused by exceeding the ideal annealing temperature, causing the active layer to be damaged and uneven again. It was familiar when dealing with organic compounds, as excessively high temperatures easily damaged organic compounds.

The results of the SEM characterization showed that the polymer coating was mainly covered on the surface, but there was still a hole and heap on the active layer. Other factors that affect the presence of solution particles in steam influence the active layer morphology. When annealed, the solution and steam left on the active layer evaporate, resulting in a hole because the active layer does not flatten [33]. Furthermore, the spin coating process contributed because the active material coating speed was still set manually.



Fig. 9: The voltage produced at various active layer ratios.

These findings imply that molecule crystallization may not occur spontaneously at room temperature and that additional processing is required to achieve efficient charge transport in these materials [45-46]. Furthermore, a suitable temperature was needed to maximize the annealing process because a too-low temperature would cause the process to be inefficient. In contrast, a temperature that is too high will harm the polymer.

Polymer solar cell current electricity examining with annealing effect active layer ratio effect

The increasing amount of particle P3HT on the active layer was also investigated in this study, with comparisons of 2:1 and 3:1 to PPV. Annealing was performed on samples from various comparisons at a temperature of 150 °C because previous experiments indicated that this temperature was the best annealing temperature for this polymer mixture.

Figure 9 presents the voltage measurement results for the three comparisons used. According to the data, the voltage generated by a solar cell with an active layer ratio of P3HT: PPV 1: 1 was 4.018 V. In comparison, the voltage generated by a solar cell with an active layer ratio of 2: 1 was 4.034 V. The ratio of 2:1 voltage was higher than the active layer's ratio of 1: 1. However, the highest voltage data generated was from solar cells with an active layer ratio of 3: 1, generating 4.098 volts. According to the research findings, increasing the amount of P3HT can increase the voltage on the solar cell. The higher the P3HT ratio in the active layer, the higher the voltage produced by the solar cell preparation.



Fig. 10: Comparison of the active layer ratio on the current power (mA).

The current power generated in the solar cell, arranged in different comparisons, produces results similar to voltage measurements. The results are depicted in Figure 10 below. Based on the test results, it was known that the power generated by the solar cell with the active ingredient P3HT: PPV 1: 1 was 0.03 mA, while the ratios of active ingredients 2: 1 and 3: 1 produced the same result, 0.04 mA. Based on the results, it was clear that a higher ratio of P3HT to PPV made more power.

Based on the resulting voltage and current data, the efficiency of each ratio was compared. The graph below shows the efficiency produced by the three solar cells at various active layer ratios. According to the results, the efficiency of the active layer with a 1: 1 ratio was 2.57 percent, the efficiency of the active layer with a 2: 1 ratio was 3.44 percent, and the efficiency of the active layer with a 3: 1 ratio was 3.50 percent. Based on the results, it was known that the active layer has the highest efficiency at a 3: 1 ratio.

It clearly shows that increasing the amount of polymer P3HT on the active layer affected solar cell performance. It was consistent with previous research investigating the effects of the P3HT: PCBM and PPV: PCBM mixture ratio [26, 47]. When exposed to UV light, P3HT on the active layer acts as an electron donor. Increasing the amount of P3HT on the active layer may improve light-to-electrical energy conversion because more photon energy absorption results in more electronhole formation. The amount of photon energy absorbed by the active layer increases as the number of donor particles on the active layer increases. The efficiency



Fig. 11: Comparison of active layer ratio to the solar cell efficiency.

was higher at 150 °C annealing temperature, at 3.5 percent. It was due to more photons being absorbed on the active layer surface. An increase in the number of electron donors on the active layer would increase photon energy absorbency.

Increasing efficiency by 3:1 is considered very small. According to the reference [48], photon absorption was comparable to rapid electron-hole formation. The semiconductor conduction band would be reached by electron excitation. The electron in the band would move freely, but it would disrupt the electron's rapidity if the electron were too large. It can cause an electron that was passing outside of the series to the counter electrode to descend.

This study's findings were inconclusive when compared to current solar cell efficiency. However, it outperforms previous research by reference [43] with a result of 0.51×10^{-3} percent and by reference [40] with a result of 0.81 percent by varying the intensity of the xenon lamplight. Several factors contribute to low solar cell efficiency. Some of them include solution and steam on the active layer after the spin coating process. When it was annealed, the solution and steam evaporated, forming a hole and causing the active layer morphology not to be flat. Oxygen affects solar cell performance because it can bind the carbon at the top of the polymer double chain. Furthermore, binding would result in a reaction capable of disrupting P3HT conjugated [39]. Because photon energy resources are directly compared to solar cell performance, using fewer photon energy resources affects solar cell performance [40].

Based on the research and discussion, it can be concluded that:

1- At an annealing temperature of 150 °C, the active layer morphology was more flat and homogeneous.

2- As the amount of P3HT on the active layer of a polymer solar cell increases so does its efficiency. The highest efficiency was obtained with a 3:1 ratio.

3- At 150 °C and 3:1 ratio, the highest current power and voltage produced by a solar cell using P3HT: PPV as an active material are 0,03 mA and 4,098 V in sequence, representing a 3.5% efficiency.

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