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Naturwissenschaften

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Assembly and Function of Polymer  
Fullerene Solar Cells

The effect of production parameters  
on energy conversion efficiency

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# Abstract

The demand for alternative energy is steadily increasing and because of this a major field being investigated is Solar Cell technology. Until now, most of the Solar Cells in daily life have consisted of inorganic materials. They are already quite efficient as they can reach an energy conversion efficiency of about 39%. Organic Solar Cells could only achieve efficiencies of about 4-5% in 2014. Organic Solar Cells are flexible, transparent, low-weight and they are also said to be produced at low costs. As to enhance energy conversion efficiency of Organic Solar Cells, it is being investigated how to adapt their designs and materials. One possibility is varying production parameters. This paper concentrates on the influence of the production parameters "thermal annealing" and "process additives" on the energy conversion efficiency of Polymer Fullerene Solar Cells. A series of experiments was carried out at the Institute for Chemistry and Technology of Materials, an institute of Graz University of Technology (Austria), in which different polymers were used. The energy conversion efficiency of certain blends could be increased after thermal annealing, whereas other blends did not show any enhanced energy conversion efficiency. The two used process additives had an increasing effect on the energy conversion efficiency, although further experiments will have to be carried out in order to get clearer results.

# Foreword

My passion for Chemistry started quite early: When I was 13 years old, I decided to study Chemistry at university. Until now, my plan has not changed at all. There was not one specific topic I was interested in, but it was the world of molecules and atoms that fascinated me. When I was 15, I was bent on working in a laboratory in summer and in July 2013 I took an internship at the Institute for Chemistry and Technology of Materials, an institute of Graz University of Technology. My colleagues taught me how to assemble Solar Cells on my own and I was really enthusiastic. That is why I contacted Dr. Gregor Trimmel, the head of the Solar Cell team, again and asked him if it was possible to make an experiment for this paper at the institute.

Composing this piece of work was not as easy as I had expected but within this year of writing I have gained an enormous amount of know-how in Physics and Chemistry.

Since I could not have managed dealing with this topic without competent help, I want to thank my supervisor Mag. DI Dr. Barbara Enko for her patience, support and time (even in the holidays). In addition to this, I also want to thank the teachers Mag. Elisabeth Klemm and OStR. Mag. Maria Stückler for proofreading this piece of work and pointing out ideas for improvement.

A big "Thank you" goes to Assoc. Prof. Dipl.-Ing. Dr. Gregor Trimmel, who made my internship at the Institute for Chemistry and Technology of Materials possible. I also give props to DI Dr. Verena Harum. She did not only guide me through my series of experiments at the institute, but also gave important hints for the writing process of my piece of work.

Finally, I have to thank my family. They did their best not to disturb me when I was caged in my room writing this paper. Especially my younger sister Julia managed to clear my mind with jokes and "30s dance parties".

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# 1 Introduction

At present, Solar Cell technology is a big field being investigated. World population rises and therefore the demand for energy is increasing too. Nowadays a big part of energy is still produced by nuclear power stations, even though on March 11<sup>th</sup>, 2011 an accident took place in Fukushima, Japan. After an earthquake a tsunami hit the nuclear power station and stopped the cooler of three reactors from working, which led to a meltdown [1].

Now new ways of producing energy have to be found, because accidents in nuclear power stations like Fukushima encourage the support of an opting out of nuclear energy. An environmentally friendly alternative are Solar Cells. They generate electrical current with the help of sunlight. Within the last years, Solar Cells with more useful properties such as flexibility have been produced [22]. One of these new types of Solar Cells are Polymer Fullerene Solar Cells. One of the parameters describing the quality of a Solar Cell is the energy conversion efficiency. It is explained as the ratio between the incoming sunlight and the generated electrical current. In Organic Solar Cells, such as Polymer Fullerene Solar Cells, lower efficiencies can be observed than in Inorganic Solar Cells (see chapter 3.3). Different production parameters can be varied in order to increase the energy conversion efficiency of Polymer Fullerene Solar Cells. This paper discusses the results of a series of experiments investigating the influence of thermal annealing and process additives on three kinds of Polymer Fullerene Solar Cells compared to results in scientific papers. The basis of this piece of work is the paper "Organische Solarzellen" by D. Wöhrle and O.R. Hild. It discusses the function of Organic and Inorganic Solar Cells (see chapters 3.1 and 3.2) and stresses the importance of cheap Organic Solar Cells. Synooka et al., Liu et al. and Yang et al. have published information on the influence of thermal annealing on the used polymers [26] [27] [28].

## 2 Development of Solar Cell technology

The background of Solar Cell technology shall be explained with a historical introduction. It focuses on the major developments in the 19<sup>th</sup> and 20<sup>th</sup> centuries.

### 2.1 Bequerel and the photoelectric effect

It all started in 1839 when 19-year-old Alexandre Edmond Bequerel made a discovery of scientific importance: He took two pieces of Platinum and put them into dilute acid. Then he observed that sunlight can increase the voltage of his system. The photoelectric effect, which is explained in chapter 3.1, was discovered [3]. It showed that there is an interaction between light and matter. Light can change the composition of atoms and lets them emit electrons [2].

Nobody understood the background until 1905, when Albert Einstein claimed that light is not a wave but made up of small energy packages, called photons. The energy of the light depends on its wavelength and not on its intensity [3].

Before this hypothesis, people carried out a lot of experiments in order to gather more information. That is how Willoughby Smith found out (in 1873) that Selenium is a semiconductor. In the dark electrical resistance is high, in the sun it is low. Three years later, Adams and Day proved the photoelectric effect with a crystal of Selenium. In 1954, Daryl Chapin, Calvin Fuller and Gerald Pearson produced the first Silicon Solar Cells based on this know-how [3] .

## 2.2 Batteries for the Bell telephone system

In 1952, the Bell telephone system was not reliable enough using dry-cell batteries, as climate conditions had an impact on their efficiency. Chapin, Fuller and Pearson from Bell laboratories were asked to find a solution and different types of generating electrical current were considered. Further, Chapin was interested in Solar Cells and did research with Selenium Cells (which were the only type available). The setup of Solar Cells (based on organic materials) is explained in chapter 3.2.2. Nevertheless, his results were not that good as the experiment did not produce enough current [4].

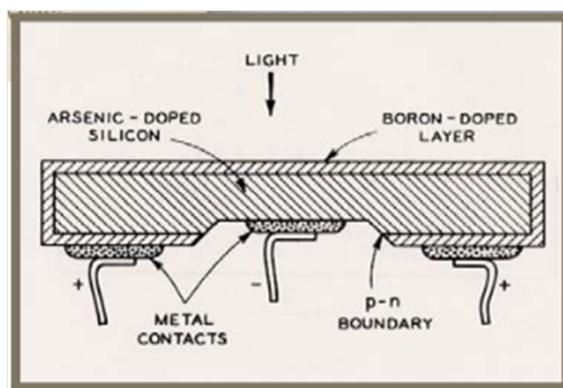


Figure 2.1: Fuller's Solar Cell assembly [5]

In the meantime Pearson and Fuller worked with Silicon. They doped a rod of Silicon with small amounts of Gallium. A positively charged layer was created, as Gallium has only three valence electrons. The rod was put into a Lithium bath, and the Lithium on the rod created a negative layer, because Lithium atoms have only one free moving valence electron. The outcome was a superior conductor with a depletion region between the doped Silicon rod and the Lithium on the rod (see chapter 3.1). The structure is comparable to figure 2.1. After lighting it up, a higher electrical current than in the dark was observed. Based on Pearson's studies, Chapin changed the material. He did tests with Silicon and came to the results that it was theoretically possible to reach an efficiency of 23% with Silicon Solar Cells, on the contrary to the Selenium Cells which were on the market in those days. (They reached only about 0.5%). One problem was that the Lithium easily penetrated into the doped Silicon rod. This switched the depletion region and decreased light absorption as the p-n junction should ideally be near the surface. Therefore the scientists switched from Lithium over to Phosphorus and it showed a bit of success. Fuller changed Chapin's Solar Cell a lot. While Chapin produced a positive layer by doping the Silicon with Gallium, Fuller's "rod" was negatively charged. He had doped it with Arsenic. He put it in a furnace and coated it with Boron in order to

## *2 Development of Solar Cell technology*

produce a thin layer, near the surface. Figure 2.1 shows the assembly of this Solar Cell. The achievements were satisfying. On April 25<sup>th</sup>, 1954 the public could see the Bell Solar Battery powering a radio transmitter. Since then it has been being tried to improve this prototype and to make it applicable for the industry and the public [4] [6].

# 3 Difference between Organic and Inorganic Solar Cells

Nowadays there are two different fields competing: Organic and Inorganic Solar Cells. Producing Inorganic Solar Cells is expensive and Organic ones are said to be a cheaper alternative (see chapter 3.3) [7] [8].

## 3.1 Function of Inorganic Solar Cells

Since Silicon is the most common material for Inorganic Solar Cells, the following chapter explains their function based on this element.

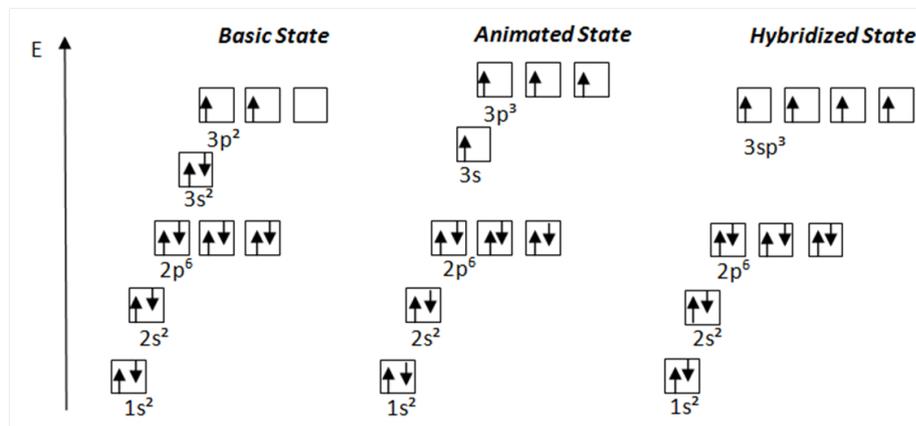


Figure 3.1: Electron configuration of Silicon in different states of hybridization

Silicon is an element with four valence electrons which are able to combine. They are located in the 3s and 3p orbitals, which is described in the basic state in figure 3.1. With energy, atoms are able to form bondings with other atoms, because one of the electrons in the 3s orbital becomes energetic richer and moves to the last free p orbital. This animated state can also be seen in figure 3.1. If a Silicon atom forms a bonding with four other atoms, theoretically, there should theoretically be two different kinds of bondings: one for the 3s, and one for the 3p orbitals. But it

### 3 Difference between Organic and Inorganic Solar Cells

has been observed that the four bondings are the same. This is possible because of hybridization. The 3s orbital and the three 3p orbitals mix and four  $sp^3$  orbitals are produced.

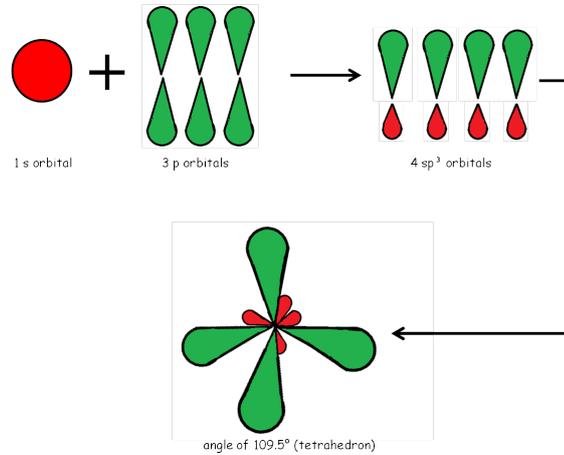


Figure 3.2: Formation of an  $sp^3$  orbital

The s orbitals can be visualised by a ball, whereas the p orbitals can be described as pairs of clubs. The  $sp^3$  orbitals look like pairs of one bigger and one smaller club. They organise in an angle which creates the biggest distance between the orbitals. Therefore the orbitals do not include an angle of  $90^\circ$ , but an angle of  $109,5^\circ$ , shown in figure 3.2. If each of the four valence electrons of hybridized Silicon combines with another Silicon atom, a crystal is formed [11] [12].

The formation of a crystal is necessary for the development of energy bands. If one single atom is observed, there are separate energy levels. But if this atom starts an interaction with another atom, the energy level becomes parted. One of these parts is energetically richer than the other. In a crystal, there are a lot of sectioned energy levels and they form bands, as shown in figure 3.4 [13].

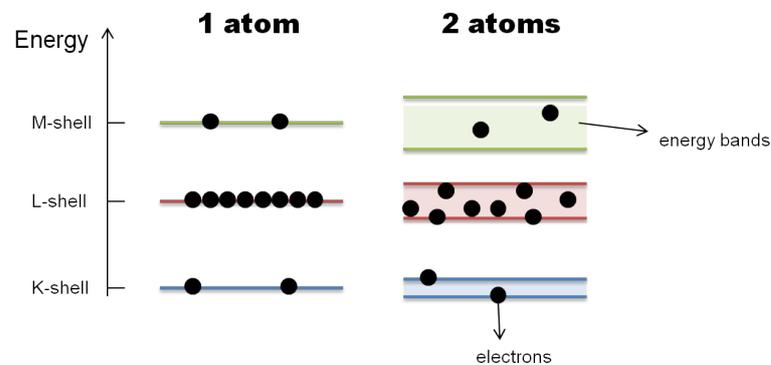


Figure 3.3: Development of bands

The model of energy bands describes the difference between conductors, semiconductors and insulators.

### 3 Difference between Organic and Inorganic Solar Cells

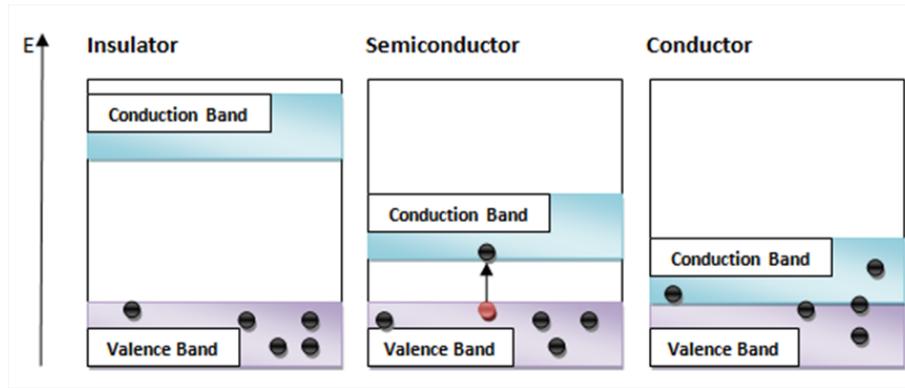


Figure 3.4: Difference between insulators, semiconductors and conductors

There are two main bands: the valence band (VB) and the conduction band (CB). Most of the electrons are in the VB, which is energetically lower than the CB. Due to a lot of electrons in a small energy field there cannot be a lot of movement. Therefore, conducting current is not possible. In conductors the VB and CB overlap each other. This means that electrons may evade into the CB. As a result, each electron has a bigger energetic range and is able to move and conduct electrical current. On the contrary to conductors, there is a gap between CB and VB in insulators. Thus, electrons are not able to change energy levels. Silicon is a semiconductor. It shows properties of conductors and insulators. Semiconductors show a gap between the CB and the VB, which is not as big as in insulators. With a special amount of energy, for example through absorption of light, electrons have the capability of overcoming the gap and of reaching the CB [13]. Figure 3.5 shows the energetic difference of CB and VB in insulators, semiconductors and conductors.

This intrinsic conductivity of Silicon is not important for the production of Solar Cells, since it changes with different production temperatures. In order to reach a conductivity comparable to metals, temperatures of more than  $100^{\circ}\text{C}$  would be needed [14]. Consequently, the material must be doped.

This means, different atoms of other element groups are added to Silicon. In this case Boron, which has three valence electrons, is taken. Instead, Silicon has four valence electrons and as a result the p-conductor (positive conductor) lacks in electrons. In the following explanations the lack of an electron is called "hole". For the n-conductor (negative conductor) Phosphorus from the fifth element group is added and a surplus of electrons occurs. Thus, conductivity improves. Although the conductors are charged, the single atoms are still neutral.

If p- and n-conductors get into contact with each other, the electrons flow from n-conductor to p-conductor. Additionally, the holes move from p-conductor to n-

conductor. The electrons and holes combine and form a barrier, because there are not any free carriers. This barrier is called depletion region.

The depletion region on the side of the n-conductor is positively charged, whereas the area on the side of the p-conductor is negatively charged. It can be explained with the carrier transport. In the n-conductor one of the five Phosphorus valence electrons combines with a hole from the p-conductor. Therefore an electron is missing in the Phosphorus atom at the depletion region and it is positively charged. The same is true for the p-conductor. The Boron atom is neutral, but the hole combines with an electron and the Boron atom at the depletion region is negatively charged.

If light with a wave length of 1130nm at the most reaches the Solar Cell, the photons become absorbed. The photons split up neutral atoms at the depletion region and produce excitons (pairs of electrons and holes). Due to the charge of the depletion region, electrons move from p-conductor to n-conductor, in the meantime holes move from n-conductor to p-conductor. Consequently, the Solar Cell produces voltage. The process of removing electrons of a material by lighting it, is called the inner photoelectric effect [9] [17].

## 3.2 Organic Solar Cells

The basic design of Organic Solar Cells can be compared with a sandwich. The two active layers donor material and acceptor material (the terms are explained in chapter 4) are placed between two electrodes. The main difference to Inorganic Solar Cells is that the active layers are made of organic molecules [18].

### 3.2.1 Function

Organic semiconductors show a lower conductivity than inorganic semiconductors, which is wanted to be avoided. This can be explained with the  $\pi$ -bonding system. In organic semiconductors the delocated electrons depend on double bondings and aromatic molecules which compose only a part of the whole molecule. An example can be seen in chapter 4.1.1. Contrary to organic materials, Silicon for example forms a crystal (see chapter 3.1) and delocated electrons can be found all over the solid body. Therefore the energy bands in organic semiconductors are broader than in inorganic semiconductors.

### 3 Difference between Organic and Inorganic Solar Cells

In order to improve the conductivity, organic semiconductors are doped. This process is similar to doping inorganic materials (see chapter 3.1), nevertheless in organic semiconductors more external material is added (1-10 weight % in organic semiconductors and 100 parts per million up to 0.1 parts per billion in inorganic semiconductors). According to Assoc. Prof. Dipl.-Ing. Dr. Gregor Trimmel, who works at the Institute for Chemistry and Technology of Materials, an institute of Graz University of Technology, the Solar Cells do not always have to be doped as too high conductivity can have a negative effect on the Solar Cells.

In molecules the concept of energy bands (see chapter 3.1) is exchanged for HOMO (highest occupied molecule), which is similar to the VB in inorganic materials and LUMO (lowest unoccupied molecule), which is similar to the CB [9] [19].

The p-conductor is doped with the help of an electron acceptor, which has a LUMO with lower energy than the HOMO of the p-conductor. Electrons move to the acceptor and generate holes. When producing an n-conductor, an electron donor is needed with a HOMO that is energy-richer than the LUMO of the n-conductor. That is why electrons move to the p-conductor. There is a difference in charge and conductivity becomes higher.

Like in Inorganic Solar Cells, the photoelectric effect can be observed when Organic Solar Cells are illuminated (see chapter 3.1). If the energy of the photons is higher than the bandgap between HOMO and LUMO, excitons are created. Excitons are pairs of electrons and holes, which easily recombine. This recombination should be avoided in order to generate electrical current, since a voltage is needed. Thus, either electrons or holes have to reach an acceptor. Organic Solar Cells need a donor that yields electrons and an acceptor that accepts electrons. When they are doped, they are called p-conductor and n-conductor (see chapter 3.1) [9].

#### 3.2.2 Design

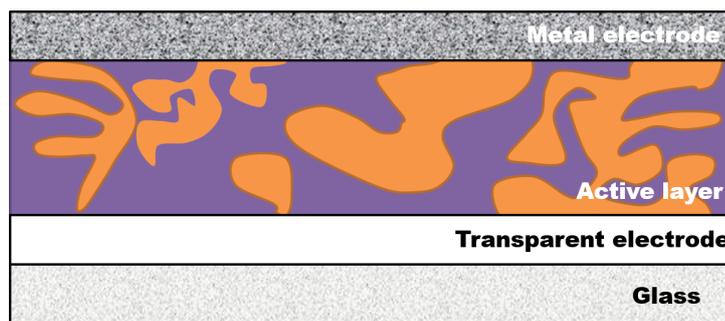


Figure 3.5: Design of an Organic Bulk Heterojunction Solar Cell

Figure 3.9 shows the design of a Bulk Heterojunction Solar Cell. Organic Solar Cells consist of several layers. The active layers of donor and acceptor are placed between two electrodes. One of the electrodes has to be transparent in order to let photons reach the surface between the two active layers. Therefore, ITO (Indium Tin Oxide) is often used as anode. It is put on a glass substrate. On the transparent electrode often another film is coated. A thin layer of Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (Pedot:PSS) helps to reduce the surface recombination at the contact to ITO. A special type of Organic Solar Cells are Bulk Heterojunction Solar Cells. As can be read in chapter 3.2.1, excitons are likely to recombine. One way of accelerating the dissociation is the enlargement of the surface between donor and acceptor. Thus, the two layers are mixed. The film of Pedot:PSS is not drawn in the picture, but is placed between the transparent electrode and the active layer [21].

## 3.3 Advantages and Disadvantages of Organic Solar Cells

Several arguments support the development of Organic Solar Cells, although the disadvantages have also to be mentioned.

Organic Solar Cells are flexible and their weight is low. Furthermore, they show a high transparency, which is important for panoramic roofs for example, and production is environmentally friendly.

Research on Organic Solar Cells was started, because they were said to be a more economical alternative to expensive Inorganic Solar Cells. It is a goal to manufacture Organic Solar Cells cheaply and use them as part of other products. Thus, new gadgets will be placed on the market (e.g. wearable Solar Cells) [22]. According to Dr. Trimmel, it is difficult to produce Organic Solar Cells at a lower cost than Inorganic ones at the moment, as the price for Silicon (see chapter 3.1) has been reduced.

The biggest disadvantage of Organic Solar Cells is their low efficiency. In 2014, 4-5% efficiency was achieved, whereas Inorganic Solar Cells have already reached 39%, although Inorganic Solar Cells on sale have achieved 15-20% [23].

It is being tried to increase the efficiency of Organic Solar Cells by enhancing the design and the materials. This paper is concentrating on different production parameters in order to find out if, for example thermal annealing or process additives

### *3 Difference between Organic and Inorganic Solar Cells*

have an impact on energy conversion efficiency of Polymer Fullerene Solar Cells and if the design can, therefore, be improved. Experiments with several polymers at different temperatures and with two process additives have been carried out. The following series of experiments has been carried out at the Institute for Chemistry and Technology of Materials (Graz, Austria).

# 4 Materials

Organic Solar Cells are composed of donor materials and acceptor materials (see chapter 3.2.1). These are the terms used before the materials are doped. When being doped, the donor yields an electron and becomes a p-conductor. The acceptor accepts an electron and becomes an n-conductor. In this series of experiments, polymers are the donors and a fullerene is the acceptor. So, the active layers used in this series of experiments were not doped. Although it can be read in literature that organic semiconductors are doped, in order to achieve a higher conductivity, it is not always common [9]. Furthermore, process additives are added.

## 4.1 Polymers

Different polymers are investigated, because the chemical structure has major influence on "[...] the electronic band structure, charge carrier mobility and absorption range." [24] (see chapter 3.2.1).

### 4.1.1 Structure and properties of P3HT

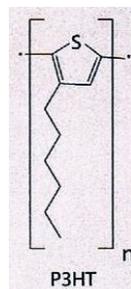


Figure 4.1: Structure of P3HT [9]

Poly(3-hexylthiophene) (P3HT) is the most common polymer when it comes to Organic Solar Cells. Its structure is drawn in figure 4.1. It is sought to work

with pure P3HT, but purification is a difficult process. Residues, for instance from catalysts, can have an impact on charge-carrier transport and recombination rates.

If P3HT is in solid state, it has a high molecular weight. The polymer shows a lamellar structure and there are interlamellar zones in between. This property might improve hole mobility, because it is verified that "[...] the higher the molecular weight, the higher the hole mobility in pure P3HT." [25].

#### 4.1.2 Structure and properties of PCDTBT

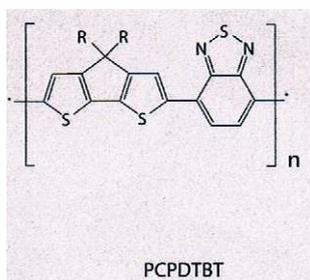


Figure 4.2: Structure of PCDTBT [9]

Another polymer used is poly[N-9'-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)], also called PCDTBT (see figure 4.2). Leclerc et al. incorporated it in 2007. Properties of PCDTBT are "high solubility, nearly perfect internal quantum efficiency in samples annealed below 80°C, and excellent thermal stability, with operating lifetimes estimated to be 7 years". The reason for the high energy conversion efficiency of PCDTBT Solar Cells is the fast charge carrier generation and recombination dynamics [26].

#### 4.1.3 Structure and properties of PTB7

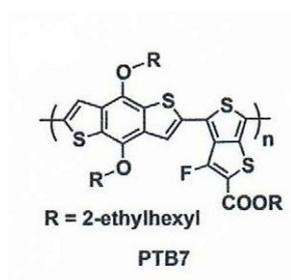


Figure 4.3: Structure of PTB7 [27]

The polymer (poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]) (PTB7) is also a good

donor for Organic Solar Cells. Liu et al. have published that "Solution-cast thin films of PTB7 show a 'face-on' crystal orientation, although the crystallinity was shown to be low". PTB7 shows a low HOMO, which increases the open circuit voltage ( $V_{OC}$ ) [27].  $V_{OC}$  is the charge difference between cathode and anode when there is no electrical current. It is explained in detail in chapter 6. There is a linear relationship between the effective bandgap and  $V_{OC}$ . The lower the HOMO, the higher the  $V_{OC}$  [28]. The structure of PTB7 can be seen in figure 4.3.

## 4.2 Fullerene

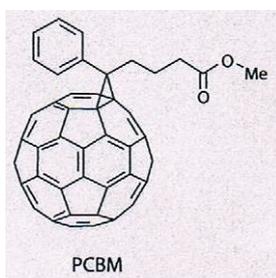


Figure 4.4: Structure of PCBM [9]

[6,6]-phenylC<sub>71</sub>butyric acid methyl ester, abbreviated to PCBM, can be seen in figure 4.4. It is the acceptor in our devices [26].

Fullerene was discovered in 1985 by Harold W. Kroto, Robert F. Curl and Richard E. Smalley. It has been being synthesized since 1990: Carbon out of graphite-electrodes is evaporated by an arc in an inert atmosphere (helium). The vapor condenses. This condensate consists of a lot of C<sub>60</sub>-molecules, which is the most common fullerene [29] [30].

Fullerene is similar to graphite. Graphite is a solid body made up of hexagons, which form parallel layers. Every atom has three electrons in bonding. The fourth electron is delocated because of double bondings. The delocated electrons are responsible for the metallic properties of graphite.

In fullerenes, there are a number of pentagons instead of hexagons. The pentagons are the reason for the bending of the molecules, which look like a football. In the pentagons there are not any double bondings. That is why there are not as many delocated electrons in fullerenes as in graphite and fullerenes do not conduct electrical current as well as graphite does [30].

The fullerene can be doped (for example with alkali-metals), as to get the properties

of a metal or even a superconductor. In general, intramolecular attraction is higher than intermolecular attraction, but with pressure, doping and light irradiation the fullerenes can be polymerized. Covalent (strong) bondings between the molecules are formed [31] [32].

### 4.3 Process Additives

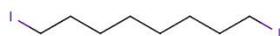


Figure 4.5: Structure of 1,8-Diiodooctane [33]

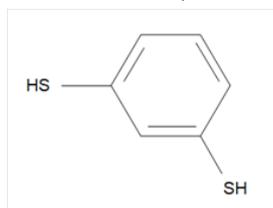


Figure 4.6: Structure of Benzene-1,3-dithiol

Energy conversion efficiency of Polymer Fullerene Solar Cells can be improved by adding process additives. In this series of experiments, 1,8-Diiodooctane (DIO) (see figure 4.5) and Benzene-1,3-dithiol (BED) (see figure 4.6) are used. When seeing the different structures of the two process additives used, the question arises, what characteristics are important for process additives. According to Dr. Trimmel, the structure of the process additives is not crucial for the decision which materials to use. What is decisive, is the temperature of ebullition.

# 5 Methods

## 5.1 Assembly

Bulk Heterojunction Solar Cells, as explained in chapter 3.2.2, are produced and the active layer consists of a mixture of a polymer and a fullerene.

### 5.1.1 General

1. **CLEANING:** ITO (Indium Tin Oxide) substrates are cleaned with acetone. They are put in an ultrasonic bath and plasmaetched, as shown in figure 5.1.

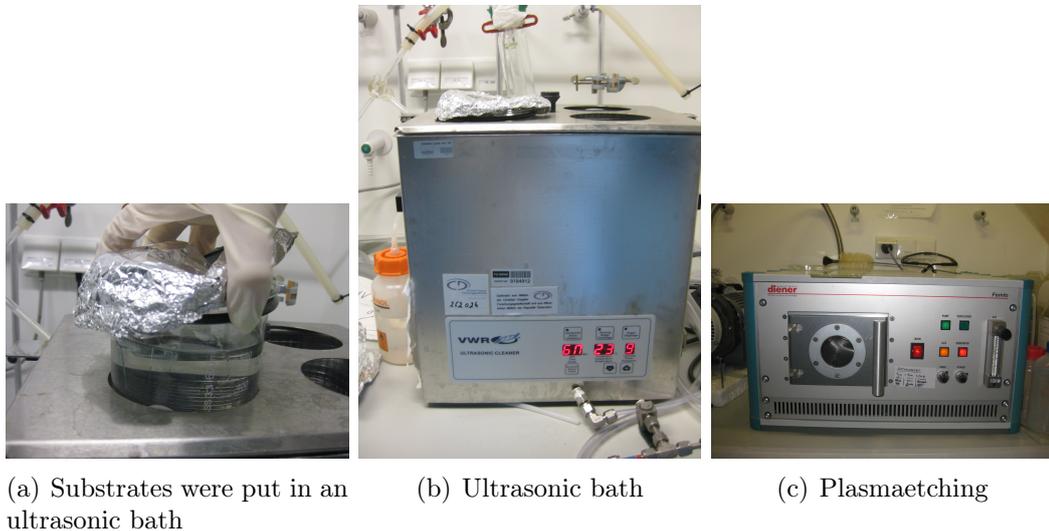


Figure 5.1: Cleaning the substrates

2. **SPINCOATING:** The process of Spincoating is used in order to produce a thin layer of Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (Pedot:PSS). This helps to reduce the surface recombination at the contact to ITO (see chapter 3.2.2) [21]. In a protective atmosphere the substrates are tempered at a temperature of 150°C for 10 minutes. Figure 5.2 is a photograph of a spincoater and of the substrates being tempered.

## 5 Methods

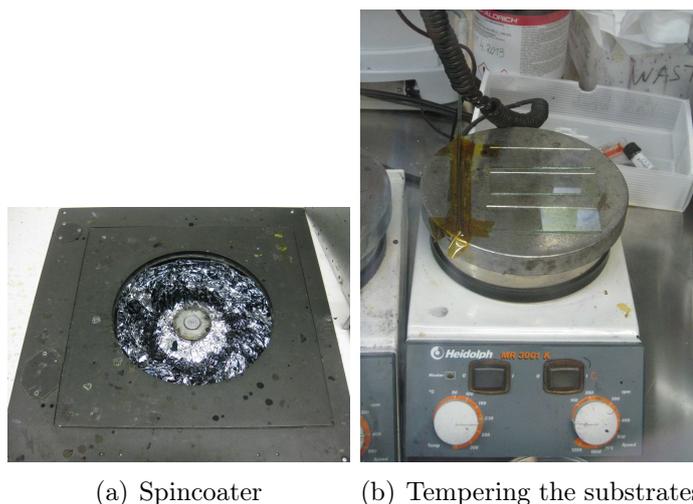


Figure 5.2: Spincoating and tempering the substrates

3. COATING THE ACTIVE LAYER: After a solution of the polymer P3HT in chlorobenzene with a concentration of 10mg/ml has been produced, 4mg of PCBM are added. The mixture is coated onto the substrates with a doctor blade and the layer is coated with four different squeegee speeds: 15, 20, 25 and 30mm/s. The height of the doctor blade is 100 $\mu$ m and the amount of Polymer-Fullerene used for each substratum is 30 $\mu$ L. This process has to be carried out in a protective atmosphere, therefore the cells were produced in a so-called Glovebox (see figure 5.3).

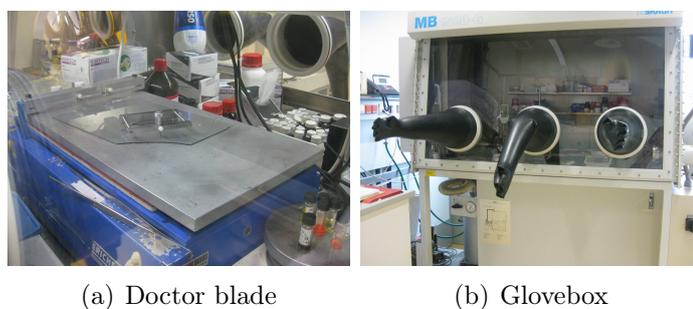


Figure 5.3: Coating the active layer

4. EVAPORATION: Aluminum contacts are vapor positioned.
5. THERMAL ANNEALING: The cells are tempered at a temperature of 100°C for 30 minutes as to investigate the impact of thermal annealing on energy conversion efficiency. Chapter 6.1 describes the background on it.

### 5.1.2 Variations

1. POLYMER: The experiment is carried out with the polymers P3HT, PCDTBT (in this case 5mg PCBM) and PTB7 (7,5mg PCBM).
2. THERMAL ANNEALING: The substrates are tempered at a temperature of 100°C, 140°C, 180°C and some are not tempered at all.
3. PROCESS ADDITIVES: The cells with the highest efficiencies of each polymer (temperature, squeegee speed) are produced again. In order to have a deeper look into the influence of process additives on energy conversion efficiency, DIO is added to the Polymer-Fullerene solution at amounts of 5, 10, 15, and 20  $\mu\text{L}$ . Cells with the same amounts of the process additive BED are manufactured.

## 5.2 Measurements

The energy conversion efficiency and other parameters of the cells are measured before and after thermal annealing. The substrates are put in a measuring box, which is pictured in figure 5.4, and illuminated.

Finally, the layer thickness and roughness are measured with a profilometer. This device can also be seen in figure 5.4.

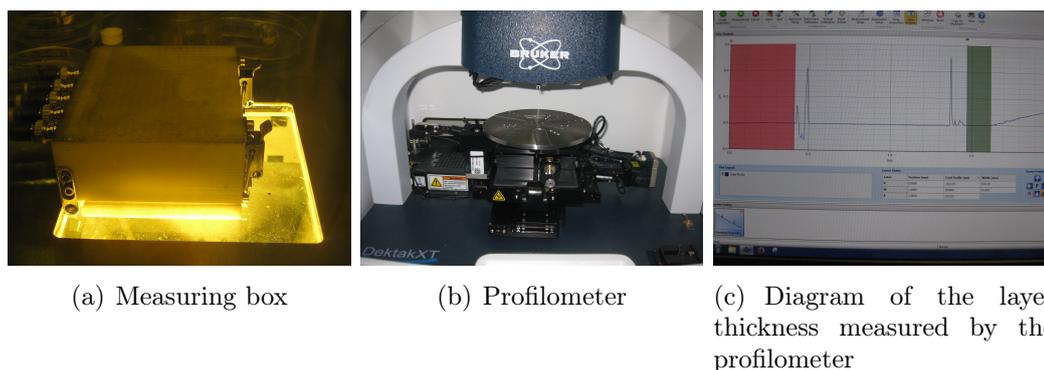


Figure 5.4: Measurements

## 5.3 Data evaluation

The data from the measuring box is exported with a purpose-built computer programme. There are twenty Solar Cells on each substrate. Theoretically, they should

## 5 *Methods*

show the same efficiencies, but this is not the case because of environmental and other circumstances. Per substrate, average and standard deviation of the five highest efficiencies are calculated.

## 6 Results and Discussion

One of the most important parameters describing the quality of a Solar Cell is the energy conversion efficiency. This is the ratio between the incoming light energy and the generated energy [9]. The goal of the series of experiments is to find out how special production parameters influence the energy conversion efficiency of Polymer Fullerene Solar Cells. Devices with three different polymers P3HT, PCDTBT and PTB7, are produced and the temperature of thermal annealing varies (no thermal annealing, 100°C, 140°C, 180°C). Furthermore, the impact of different amounts of the process additives DIO and BED is investigated.

The energy conversion efficiency ( $\eta$ ) is defined as:

$$\eta = \frac{I_{SC} \times V_{OC} \times FF}{P_{in}}$$

$I_{SC}$  (short circuit current density) describes the current density when there is no voltage. It shows if the Solar Cell absorbs sunlight well and if it is good at transporting charges.  $V_{OC}$  (open circuit voltage) describes the voltage at zero current. Theoretically, it can reach values up to the effective bandgap, which results from the difference between the HOMO of the donor and the LUMO of the acceptor. FF (fill-factor) describes the quality of the device. At the moment, Inorganic Photovoltaics can reach an FF of about 0.9, whereas Organic Photovoltaics can reach values up to 0.6.  $P_{in}$  is the incident input power. Therefore,  $\eta$  depends on  $V_{OC}$ ,  $I_{SC}$  and FF. If thermal annealing affects  $\eta$ , it must also affect at least one of these parameters. All of these parameters are measured, but this paper focuses only on the change of  $\eta$  [28].

The efficiencies are compared at different squeegee speeds. The squeegee speed is the speed at which the doctor blade coats the active layer. It causes different layer thicknesses and influences the energy conversion efficiency. Chapter 6.1.1 explains why.

## 6.1 Influence of thermal annealing on energy conversion efficiency

Thermal annealing is said to influence Bulk Heterojunction Solar Cells in two ways:

- "Dispersion of donor and acceptor phase in the photoactive layer to allow for large interfacial area which leads to efficient excitonic dissociation." [28]
- "Establishing of continuous conducting pathways for good charge transport." [28]

The data in the following chapters are structured by polymers. For each polymer the diagrams show efficiencies at a certain temperature at different squeegee speeds. The values are compared with Solar Cells which are not tempered. What is interesting in reading the diagram is that there are differences between energy conversion efficiency of annealed and unannealed devices. Energy conversion efficiency can be increased, but also decreased. It depends on the polymer. Also other parameters at which the highest efficiencies are achieved (such as squeegee speed and annealing temperature) vary from polymer to polymer. Another diagram matches the values at the most efficient squeegee speed with different annealing temperatures.

### 6.1.1 P3HT

The first series of experiments is carried out with the polymer P3HT.

As figure 6.1 shows, the squeegee speed has an impact on the efficiency. Measuring the layer thickness shows that the squeegee speed influences the layer thickness. Figure 6.2 describes the relationship between squeegee speed and layer thickness at an annealing temperature of 180°C. When increasing the squeegee speed, an increase of layer thickness can be observed.

Usually organic materials have a small exciton diffusion length. Exciton diffusion length describes the smallest thickness of a layer at which excitons are able to reach the donor/acceptor interface where charge separation takes place [34]. If the layer is too thick, energy conversion efficiency can be decreased because of the small exciton diffusion length (5-10 nm [35]). On the other hand, a special thickness (100 - 200nm in 2011) is needed in order to absorb all of the photons. Thus, the ideal thickness

## 6 Results and Discussion

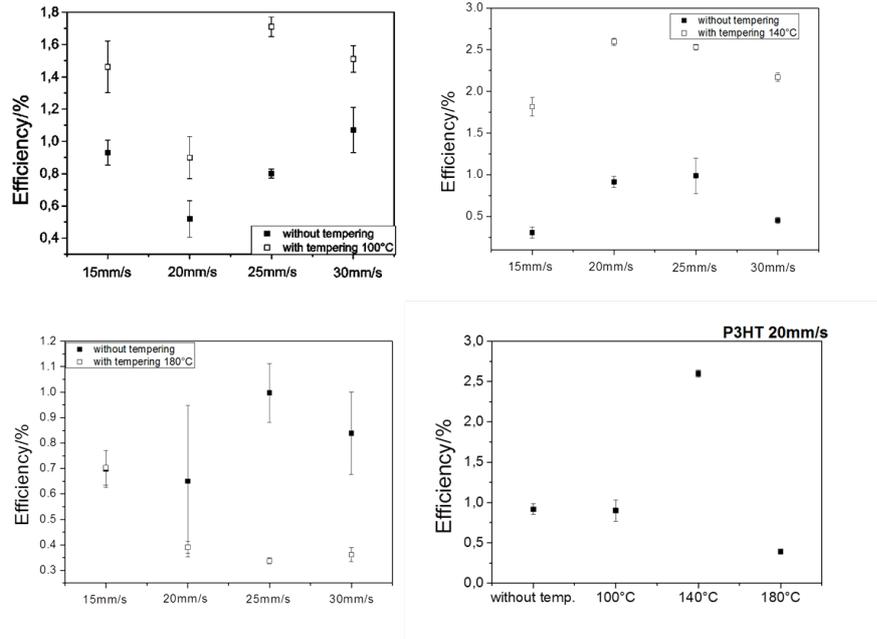


Figure 6.1: Influence of thermal annealing on P3HT:PCBM blends

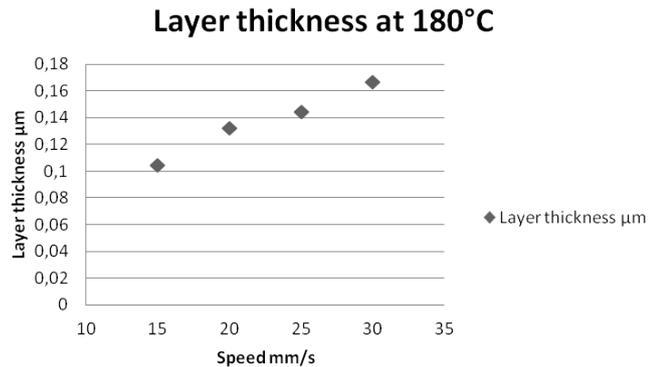


Figure 6.2: Layer thickness of P3HT:PCBM blends tempered at 180°C

depends on the polymer and its morphology [18]. A photon reaches the p-n junction and an exciton is created.  $L_D$  is the exciton diffusion length and  $d$  is the domain size.

Within the last years layer thickness has been reduced for example by enhancing the design with a Bulk Heterojunction layer (see chapter 3.2.2). It allows a donor/acceptor interface throughout the film [35].

As can be seen in figure 6.1, the highest total value of energy conversion efficiency is achieved at a squeegee speed of 20mm/s at a temperature of 140°C. Thus, the lower diagram on the right side of figure 6.1 compares the temperatures at the speed of 20mm/s. The highest values are reached after thermal annealing at 140°C.

In an unannealed state, P3HT is a poor charge carrier, due to its vague structure and morphology. Therefore,  $I_{SC}$  and FF are low. Thermal annealing has an organizing effect and brings crystallinity into the molecules. The crystallinity improves charge transport to the electrodes and increased order allows polymer chains to "[...] move freely at higher temperatures" [28]. This leads to higher  $I_{SC}$ , which means that the absorption of photons increases [28]. Nevertheless, if the Solar Cell is tempered at too high temperatures, PCBM phases can spread and aggregates turn up. This can make exciton dissociation more difficult [28] [37].

### 6.1.2 PCDTBT

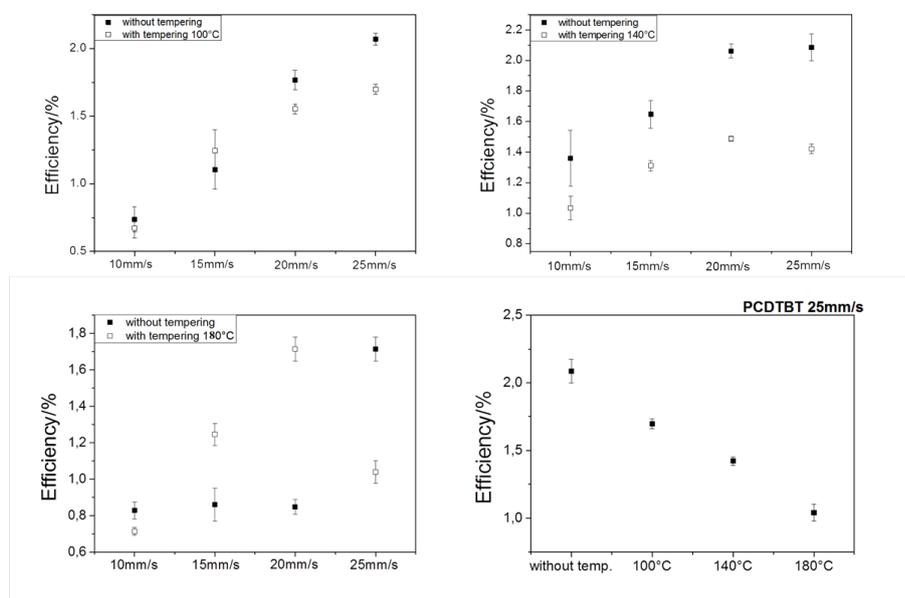


Figure 6.3: Influence of thermal annealing on PCDTBT:PCBM blends

Contrary to P3HT (see chapter 6.1.1), tempering PCDTBT improves the efficiency in most of the Solar Cells. Nevertheless, the overall highest energy conversion efficiency is achieved at a squeegee speed of 25mm/s (see figure 6.4). The lower picture on the right side of figure 6.4 shows the comparison of the temperatures at a squeegee speed of 25mm/s.

Further experiments could illustrate, if the efficiency would still increase at a squeegee speed of 30mm/s.

Synooka et al. claim that thermal annealing reduces the energy conversion efficiency of PCDTBT:PCBM blends due to the structural properties of PCDTBT [26] and Chen et al. have published that "[...] post-treatments are incapable of improving the device characteristics" [35].

## 6.1.3 PTB7

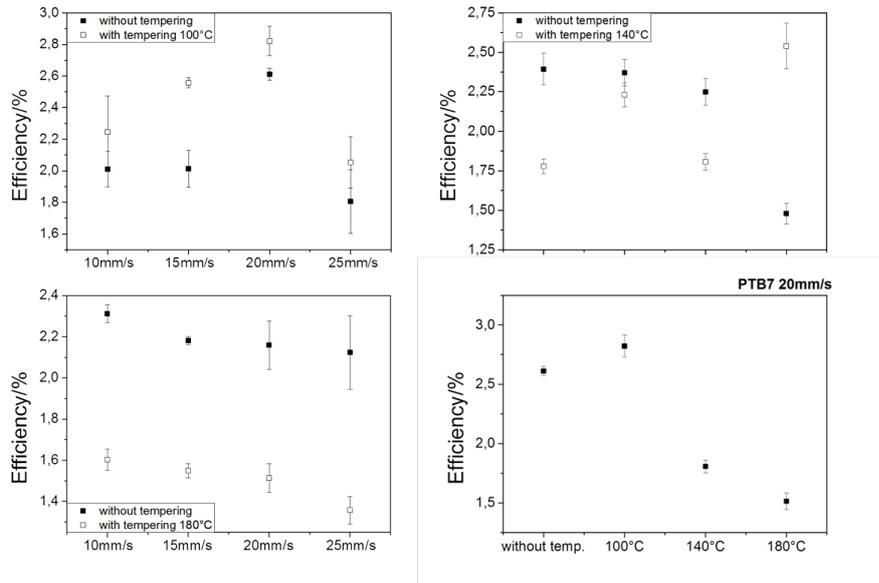


Figure 6.4: Influence of thermal annealing on PTB7:PCBM blends

Figure 6.5 shows that the highest efficiencies are achieved at a squeegee speed of 20mm/s. Consequently, further experiments are carried out at this speed. In this series of experiments, the highest efficiencies can be observed after thermal annealing at 100°C. This might be an outcome which is not that trustworthy. Literature states that "Thermal annealing the as-spun film at 150°C for 10 min did not change the crystal structure or orientation, but increased the persistence of the  $\pi$ - $\pi$  stacking slightly to 6-7 planes. In the thin-film blends [...], the face-on orientation was reduced [...]" [27]. Since different structure and orientation can have an impact on the efficiencies (see chapter 6.1), a repeated series of experiments might show decreased energy conversion efficiencies after tempering. The reduction of energy conversion efficiency is indicated by decreased orientation. Theoretically, the best results should be achieved at room temperature. Due to limited time resources it is trusted in literature as it says thermal annealing decreases the energy conversion efficiency and the following PTB7 experiments are carried out at room temperature.

So P3HT shows its best properties after coating the active layer at a squeegee speed of 20mm/s and tempering it at 140°C, contrasted to PCDTBT, which is more efficient at room temperature and a squeegee speed of 25mm/s. PTB7 should have its most efficient cells at room temperature too, but shows its best results in this experiment after having reached a temperature of 100°C and a squeegee speed of 20mm/s.

## 6.2 Influence of process additives on energy conversion efficiency

The Solar Cells of each polymer which show the highest efficiencies are produced again and the impact of process additives is investigated. The process additives 1,3-Diiodooctane (DIO) and Benzene-1,3-dithiol (BED) are compared. According to data from 2010, process additives can increase the power conversion efficiency from 5% up to 10%. The non-reacting substances are said to cause enhanced efficiencies by increasing hole mobility and forming microcrystals. A. Pivrikas et al. have published that DIO adapts the film morphology of BHJ (Bulk Heterojunction) Solar Cells [37]. To the best of the author's knowledge, there are not a lot of research and published papers describing the influence of BED on energy conversion efficiency of BHJ. Thus, the data in the following chapters cannot be compared and explained by literature.

The following diagrams show the relationship between the volume of added process additive and energy conversion efficiency. It is investigated, if process additives influence the energy conversion efficiency, and which process additive increases the energy conversion best.

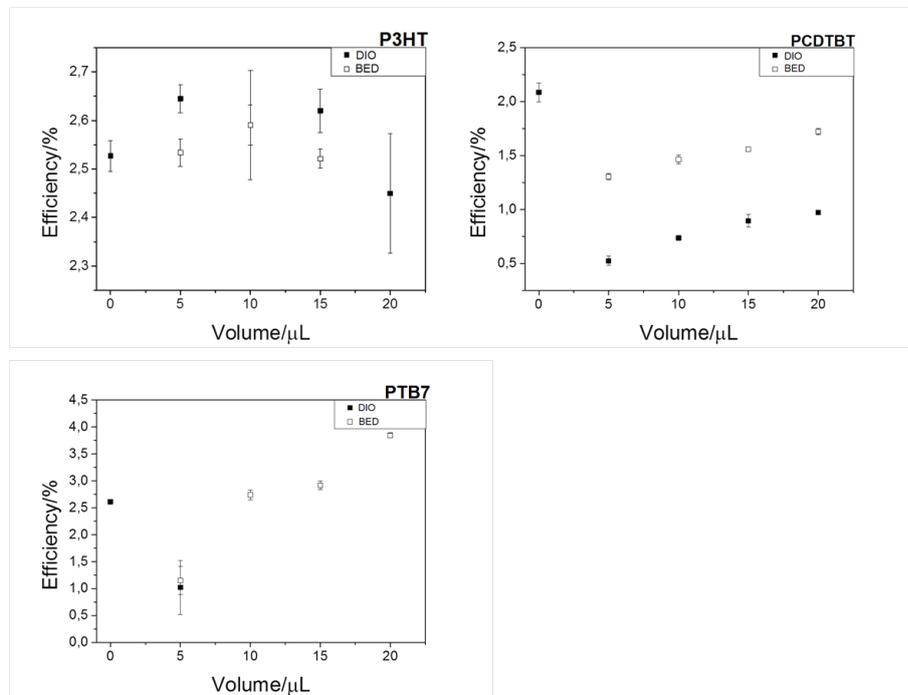


Figure 6.5: Influence of processing additives on energy conversion efficiency

### 6.2.1 P3HT

The Solar Cells are fabricated at a squeegee speed of 20mm/s and tempered at 140°C. One of the process additives 1,3-Diiodooctane (DIO) and Benzene-1,3-dithiol (BED) is added to the P3HT:PCBM solution. According to figure 6.6, each of the process additives has an impact on energy conversion efficiency. DIO reaches higher efficiencies than BED. The highest efficiencies for DIO are achieved with 5 $\mu$ L. The efficiencies start to decrease with an amount of 20 $\mu$ , and the value is lower than without any process additive, although a huge error bar can be observed.

Opposed to DIO, BED has its best efficiency with an amount of 10 $\mu$ L, but after adding more of the process additive it shows a similar behavior to DIO.

The enhancement of energy conversion efficiency by using a special amount of process additive (either DIO or BED) matches with the explanation in chapter 6.2.

### 6.2.2 PCDTBT

Solar Cells are fabricated at a squeegee speed of 25mm/s and not tempered. One of the process additives DIO and BED is added to the PCDTBT:PCBM solution. The efficiency decreases when the process additives are added. However, by increasing the amount of the additive, also the efficiency rises. Further experiments can show, if Solar Cells enriched with more than 20 $\mu$ L of process additive reach higher efficiencies than Solar Cells without process additives. Such an enhancement would match with the explanation in chapter 6.2, but comparable data are not found in literature.

Contrasted to P3HT Solar Cells, PCDTBT:PCBM Solar Cells achieve higher efficiencies processed with BED than with DIO.

### 6.2.3 PTB7

It is remarkable, that the values of DIO and BED are exactly the same at an amount of 10, 15 and 20 $\mu$  additive. There might have been mistakes when carrying out the experiment, such as mixing up the Solar Cells or measuring the same cells twice. Probably the value in the diagrams belongs to DIO. The reason for this assumption is that the Solar Cells with DIO were built and measured at first. The cells with the process additive BED were fabricated and put on a place next to the DIO cells. Then the BED Solar Cells were measured.

Nevertheless, the experiment shows that there is a process additive that improves energy conversion efficiency. After adding the additive, the value decreases, but as soon as  $10\mu\text{L}$  are added, the Solar Cell improves and the more additive, the higher the conversion efficiency. Like in the experiment with PCDTBT (see chapter 6.2.2), another series of experiments can show if further improvements are observed after going on adding process additives.

Since Liu et. al. claim that DIO improves the energy conversion efficiency of PTB7 Solar Cells, this would be a plausible explanation. DIO is said to enhance  $I_{\text{SC}}$  of PTB7:PCBM blends and results an enlarged area between donor and acceptor and in refined morphology. Also FF increases [27].

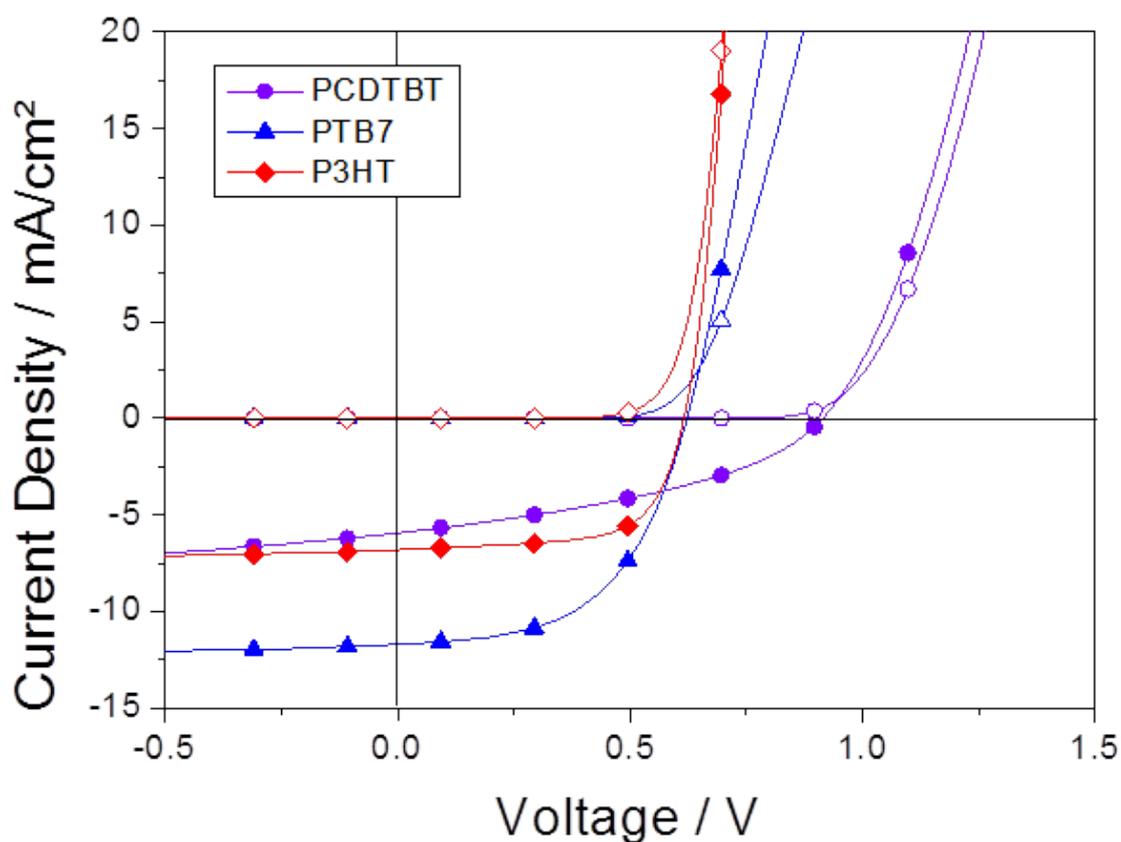


Figure 6.6: Characteristic curve of the three different Solar Cells

Figure 6.7 shows the comparison of the best cells of each polymer. Current density is plotted against voltage. The colored signs describe the graphs of the illuminated cells, whereas the white signs stand for the unlighted cells. The point of intersection of graph and x-axis is  $V_{\text{OC}}$ .  $I_{\text{SC}}$  is the absolute value of the intersection of graph and y-axis. Unlighted Solar Cells do not generate electrical current, that is why they have got  $I_{\text{SC}}=0$ . Since energy conversion efficiency is defined as:

## 6 Results and Discussion

$$\eta = \frac{I_{SC} \times V_{OC} \times FF}{P_{in}} \text{ see chapter 6}$$

The information that can be gathered from figure 6.7 matches with the results concerning the energy conversion efficiency in many points. The highest energy conversion efficiency is achieved with the polymer PTB7, which is processed with 20 $\mu$ L of DIO. In figure 6.7, the high energy conversion efficiency is indicated by the high  $I_{SC}$ . The most efficient PCDTBT blend, which is not tempered and not processed with an additive, shows lower efficiencies than the processed PTB7 blend. It is indicated in figure 6.7 by the shorter  $I_{SC}$ . According to figure 6.7, the most efficient P3HT blend should show the lowest energy conversion efficiency, because  $I_{SC}$  is shorter than in PTB7 and  $V_{OC}$  is shorter than in PCDTBT. Nevertheless, the values of the energy conversion efficiency of processed and tempered P3HT blends are higher than the values of the energy conversion efficiency of the PCDTBT blends.

## 7 Conclusion

In conclusion, there are different production parameters that influence the energy conversion efficiency of Polymer Fullerene Solar Cells. Dependent on the used materials, these production parameters can either increase or decrease the efficiencies.

In this piece of work, the effect of the production parameters "thermal annealing" and process additives on Polymer Fullerene Solar cells is discussed. A series of experiments is carried out and Solar Cells produced. The three different polymers P3HT, PCDTBT and PTB7 are used and the blends are tempered at four temperatures (room temperature, 100°C, 140°C and 180°C). Two process additives (BED, DIO) are added and the energy conversion efficiency of the blends is measured.

The squeegee speed is responsible for the layer thickness of the blends. The higher the speed, the thicker the layer. On the one hand, thick layers are desirable due to improved light absorption, but on the other hand, organic materials show short exciton diffusion lengths. Therefore the layers should also be thin, in order to maximize exciton separation. An agreement between high and low squeegee speed has to be found, which differs from polymer to polymer.

Thermal annealing is said to enhance energy conversion efficiency through increased order and crystallinity in the morphology of the blends. P3HT:PCBM blends show increased efficiencies after thermal annealing at 140°C. Higher annealing temperatures are the reason for forming aggregates and therefore hindered exciton dissociation. Due to the chemical structure, PCDTBT:PCBM and PTB7:PCBM Solar Cells do not show increased efficiencies after thermal annealing. The diagram of the influence of thermal annealing on PTB7:PCBM Solar Cells shows increased efficiencies after thermal annealing at 100°C, but according to literature that thermal annealing does not enhance energy conversion efficiency. Due to a lack of time, the experiments could not be repeated and an explanation has not been found.

Process additives lead to higher energy conversion efficiencies because they tailor the morphology. Hole mobility is being increased and microcrystals are formed. Due to the lack of information about the influence of BED as process additive on

## 7 Conclusion

Polymer Fullerene Solar Cells, the data of the discussed series of experiments cannot be explained and compared. It is clearly found out that both process additives increase the energy conversion efficiency of P3HT:PCBM Solar Cells, although the enhancement is higher when DIO is used instead of BED. PCDTBT:PCBM blends show a decreased efficiency after adding a process additive. Nevertheless, the value increases when the amount of additive is increased too, although efficiencies are still lower than without any process additive. Also PTB7:PCBM blends show a similar behavior with lower efficiencies that increase with the volume of additive added, but energy conversion efficiency at a special amount of additive exceeds the efficiency without additive. The question arises, if this can also be observed when continuing the PCDTBT experiment and adding a larger amount of additive. This would match the explanation which states that process additives increase energy conversion efficiency. The volume of process additives that leads to higher efficiencies also differs from polymer to polymer.

When discussing the influence of production parameters on energy conversion efficiency of PTB7 blends, it has to be noticed, that the diagram shows the same data for BED and DIO at three different amounts of process additive. Having a deeper look into the measured data, exactly the same values for both process additives are observed. There must have been a mistake, since achieving exactly the same values is nearly impossible. The surrounding conditions prohibit it. Probably the BED processed Solar Cells and the DIO processed Solar Cells were mixed up or measured twice. The values visible in the diagram might belong to DIO, as the blends were measured first and placed next to the BED cells.

These examples of production parameters are part of Solar Cell technology. Improving the production parameters and consequently design and materials of Organic Solar Cells increases the energy conversion efficiency and lays the foundation of a widely ranging application of Organic Solar Cells in daily life.

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**Selbstständigkeitserklärung**

Ich erkläre, dass ich diese vorwissenschaftliche Arbeit eigenständig angefertigt und nur die im Literaturverzeichnis angeführten Quellen und Hilfsmittel benutzt habe.

**Mellach, 25.02.2015**

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