Allamanda Cathartica Linn and Ixora Coccinea L as Natural Dye for Bulk Heterojunction Organic Solar Cell: A Hall Effect and Electrical Conductivity Study

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This paper describes the electrical conductivity, Hall Effect study, and efficiency in bulk hetero-junction organic solar cell (OSCs) by using natural pigments Allamanda Cathartica Linn (yellow bell flower) and *Ixora coccinea L* (Ixora flower). In this work, Indium Tin oxide (ITO) glass as substrate was heated at $50 \circ C$ to $200 \circ C$. The polymer used was Poly (3-Dodecylthiophene) (P3DT) thin film. These OSCs are fabricated accordingly bulk of ITO/P3DT+Allamanda Cathartica Linn via electrochemistry method at room temperature. The electrical conductivity of sample was explored by four point probes (FPP) under dark and under light radiation (range of $10~\rm Wm^{-2}$ to $200 \rm Wm^{-2}$). FPP data revealed that electrical conductivity was increased by the increment of light intensity and substrate temperature. In order to obtain the type of sample, Hall mobility, and highest charge carrier, Hall Effect measurement was used. Both samples detected were N-type of charge carrier. The performance of OSCs was successfully improved.

Key words: Allamanda Cathartica Linn, Ixora coccinea L, Organic Solar cell, Poly (3-Dodecylthiophene), Bulk Heterojunction.

Today, world is facing the increasing energy demand, security of energy supply and reduction of emissions¹. Energy consumption which accounts for 60% global greenhouse gas emissions has mainly contributed to climate change². Renewable energy is an appropriate way to satisfy energy consumption without environmental degradation³. Solar energy is one of the most promising renewable energy sources today. In recent years, organic solar cells (OSCs) which are a promising alternative to conventional inorganic devices have received much attention.

It is due to their high performance, potential applications, including low cost materials, ease of production, and lack of harmful emissions4. Sun light is used to excite electrons of pigments, mainly metal complexes, into a higher energy level which are then transferred to the conduction band of a wide band gap semiconductor⁵. Organic solar cells could alleviate the stated draw-backs through the use of natural pigments for the conversion of solar energy into electrical energy. The pigment could be extracted from almost anything plants, minerals by simple procedure. Most natural dye colors are found in the roots, bark, leaves, flowers, skins, and shells of plants. The advantage of natural are that they are cost effective, renewable, ecofriendly⁶⁻⁷ make them good candidates as alternative photosensitizer8. The flowers of Ixora broadly used in traditional Indian systems of

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medicine for dysentery, healing of ulcers and, more recently, for an anti-tumour activity⁹. They have also been reported to have anti-inflammatory activity comparable to indomethacin¹⁰. Allamanda cathartica. L. is a perennial shrub used in traditional medicine for treating malaria and jaundice¹¹. It has been used as a purgative or emetic, febrifuge, as well as for the treatment of coughs, headaches, jaundice and enlarged spleen resulting from malaria¹².

In this paper, an organic solar cell were formed to find its electrical conductivity, Hall Effect measurement and to elucidate its efficiency in the absorption of energy. The substrates indium tin oxide (ITO) glass, Poly (3- Dodecylthiophene) (P3DT) and Allamanda Cathartica Linn (yellow bell flower) as natural dye were used. The OSCs were deposited via electrochemistry method.

EXPERIMENTAL

Sample collection and preparation

Allamanda Cathartica Linn (yellow bell flower) and Ixora coccinea L (Ixora flower) was hand-picked from three separate plants located in Universiti Malaysia Terengganu (UMT), Terengganu. P3DT was prepared through polymerization of the Dodecylthiphene.

The 2cm x 2cm ITO glass were used as a substrate. The cleaning process of substrates was done using ultrasonic vibrator (JEIOTECH model). The tank in ultrasonic vibrator was rinsed using distilled water to make sure it is cleaned¹⁴. 50ml beaker filled with distilled water and ITO substrates was putted into the ultrasonic vibrator. Then, ITO substrates were thoroughly cleaned by distilled water, the detergent. It was followed by acetone in

order to remove any contaminations that might have been formed on the substrates¹⁵ and distilled water, respectively. The time was set for 20 minutes, 30°C and mode vibration was set as medium for each cleaning. ITO substrates were dried using the dryer before kept into a Petri dish. Then, ITO substrate used was in different temperature (room temperature, 50ÚC, 100ÚC, 150ÚC, 200ÚC).

Extraction of dye

In the laboratory, the fresh flowers were washed under tapwater and then rinsed in distilled water three times on the same day. This is to remove the contamination from flowers and to maintain flower fresh in order to yield the best plant pigment. Fresh flower was crushed into small size and shade dried. 20g of fresh flower was put into a beaker, then 100 mL absolute ethanol was added and the mixture was immersed for 48 hours and lastly, it was vibrated into the ultrasonic vibrator for 24 hours. It is for maximizing the efficiency of natural colorant extraction¹⁶. Later, the solid residues were subjected to repeated filtration with filter paper to yield a pure natural dye solution. This extraction was protected from direct sunlight exposure and stored at about 5Ú C and used for further characterization. Figure 3 show Ixora coccinea L (Ixora flower) and Allamanda Cathartica Linn (yellow bell flower).

Fabrication of organic solar cells

The bulk heterojunction organic solar cell of P3DT+yellow bell (ITO/P3DT+YB/AU) and P3DT+ixora (ITO/P3DT+IX/AU) were prepared deposited on heated substrates by electrochemistry method using electrochemical Impedance spectroscopy (EIS) PGSTAT302. EIS located in Department of Physical sciences, school of ocean engineering, Universiti Malaysia

Fig. 1. Chemical structure of Ixora coccinea L¹³

Fig. 2. Chemical structure of Allamanda Cathartica Linn 11

Terengganu. A typical electrochemical impedance experimental set-up consists of an electrochemical cell (the system under investigation), and a potentiostat/galvanostat. Based on the concentration of the solution, the less concentration of the solution will generate more cyclic in order to ensure the desired thickness is formed¹⁷. Figure 4 show the structure of bulk heterojunction of organic solar cell.

Characterization

Electrical conductivity

The samples were measured in two conditions. They were; in the dark condition and under illumination of light. The sheet resistivity produced by films was measured using FPP (Jandel RM3 Test Unit). The two outer probes supply a voltage difference that drives a current through the film. The two inner probes pick up a voltage difference.

$$R_s = 4.532 \text{ x V/I}$$
 ... (1)

Where, R_s is the sheet resistance, 4.532 is the correction factor, V is the voltage measured and I is the current applied from the test unit. Thus, electrical conductivity (EC or \tilde{A}) can be determined



Fig. 3. (a) *Ixora coccinea L* (Ixora flower) and (b) *Allamanda Cathartica Linn* (yellow bell flower)

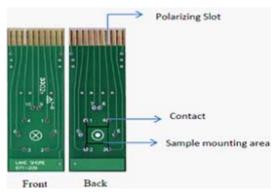


Fig. 5. The geometry of sample holder¹⁸

which it is the reciprocal of R_s , as described in Equation 2.

$$\sigma = 1/R_{c} \qquad ...(2)$$

Hall Effect Measurement (HEM)

In HEM, the samples should have well-defined geometries and good ohmic contacts in order to obtain the accurate results. The samples must have vdP geometry. The ITO substrates were placed on the sample holder as shown in Figure 5. The sample on the holder system is then connected to contacts 1, 2, 3 and 4, using the silver paint on four edges. The connection to the contact is then tested using a multimeter to ensure proper contact.

The measurements were performed using the Leois-JSF software. The software was developed and tested through the corporation of Physical Sciences Department, Universiti Malaysia Terengganu, led by Dr Salleh Harun and Nanorian Technology¹⁹. The Hardware system called Hall Effect measurement system model 7600 is supplied by Lakeshore Ltd. The important part of this HEM system is ensuring that the room temperature and set temperature was equivalent (20°C) in order to

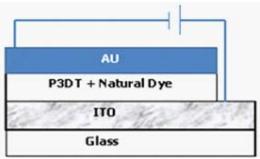


Fig. 4. Show the structure of bulk heterojunction of organic solar cell

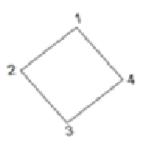


Fig. 6. The numbering for Hall calculations

prevent the power supply from breakdown. The measurement consists of two parts. The first parts are called the IV curve traces measurement and the second part is variable magnetic field measurement. The purpose of IV curve traces measurement is to make sure that all the contacts are in good connections. In this work, the magnetic field fixed was 10 kG (1 Tesla) and the current was 0.1 A²⁰. In addition, Figure 6 shows the numbering of sample for Hall calculations which used in this work.

Referring to Figure 6, the Hall voltage $V_{_{+31,42\,(+B)}}$ is measured between contact point 4 and 2 when a current $I_{_{+31}}$ is passed from points 3 and 1. With field and current reversal, there will be eight voltages can be measured. According to our previous paper²⁰, HEM can be calculated as follow, Hall voltage average, V_{Have} .

$$= \left[V_{+31,42(+B)} - V_{+31,42(-B)} + V_{-31,42(-B)} - V_{-31,42(+B)} + V_{+42,13(+B)} - V_{+42,13(-B)} \right]...(3)$$

$$+ V_{-42,13(-B)} - V_{-42,13(+B)} / 2$$

The R_{H} in vertical (R_{Hc}) and horizontal (R_{HD}) directions respectively are given as;

$$R_{HC} = 10^8 \frac{t[cm]}{B[G]} \cdot \frac{V_{+31,42(+B)} - V_{-31,42(+B)}}{I_{+31(+B)} - I_{-31(+B)}} \dots \frac{+V_{-31,42(-B)} - V_{+31,42(-B)}}{+I_{-31(-B)} - I_{+31(-B)}}$$

And;

$$R_{HD} = 10^{8} \frac{t[cm]}{B[G]} \cdot \frac{V_{+42,13(+B)} - V_{-42,13(+B)}}{I_{+42(+B)} - I_{-42(+B)}} \dots (4)$$

$$\frac{+V_{-42,13(-B)} - V_{+42,13(-B)}}{+I_{-4(-B)} - I_{+42(-B)}}$$

The Hall coefficient average, $R_{{\scriptscriptstyle H}\;{\scriptsize avg}}$ calculated by;

$$R_{H \ avg} = \frac{R_{HC} + R_{HD}}{2} [cm^3.C^{-1}] \dots (5)$$

If the thickness is unknown, the layer or sheet carrier concentration, $n_s = n$, is used instead of the bulk density,

$$n_s = \frac{8 \times 10^{-8} \times IB}{qv_{H (total)}} [cm^{-2}]$$
 ...(6)

The unit of R_H will be m³C⁻¹, if t is in meter,

B in Tesla, voltages in volt and current, I in ampere. If t is excluded, then R_H becomes the sheet Hall coefficient. Where, B is in Gauss unit, I in ampere, and V_H in volt. Then the Hall mobility (${}^{1}\!\!/_{\!\!4_H}$) is given by

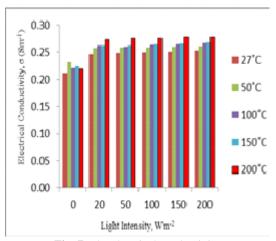


Fig. 7. The electrical conductivity for ITO/P3DT+YB organic solar cell

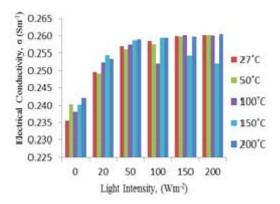


Fig. 8. The electrical conductivity for ITO/P3DT+IX organic solar cell

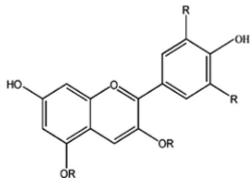


Fig. 9. Anthocyanin structure and the substitute groups (R)²³

$$\mu_H = \frac{|V_{H \, avg}|}{\rho_{\, avg}} \, [cm^2. V^{-1}. s^{-1}] \quad ...(7)$$

Lastly, types of charge carrier. It is determined by the polarity sign of V_{Havg} from (1) and polarity sign of R_{Havg} in (5). If the polarity sign is positive, the type of charge carrier is holes and called P-type. In contrast, if negative sign, it is electrons and called N-type.

RESULTS AND DISCUSSION

Electrical conductivity

Figure 7 and 8 represent the electrical conductivity measured in different light

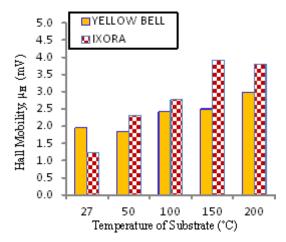


Fig. 10. The Hall mobility for bulk heterojunction organic solar cell

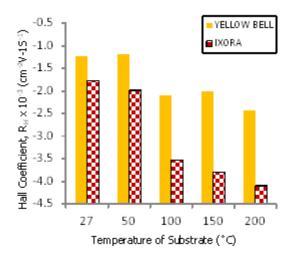


Fig. 12. The Hall coefficient for bulk heterojunction organic solar cell

illumination. The electrical conductivity for ITO/P3DT+YB and ITO/P3DT+IX sample is rises with the increment of light intensity and substrate temperature. This happen as the samples were absorbed the light energy and converted to the electrical energy²¹.

In Figure 7, ITO/P3DT+YB blended samples shows that the sample was not stable under dark condition (0 Wm⁻²). At 27ÚC to 50ÚC of substrates temperature, electrical conductivity growth then it was decrease. In the Figure 8, ITO/P3DT+IX samples show an upward trend after 100°C of substrate temperature. The ranging of electrical conductivity of both sample detected was 0.212 Sm⁻² to 0.242 Sm⁻² under dark condition.

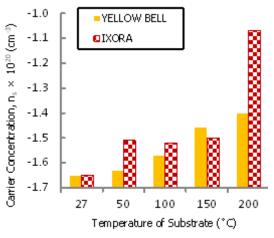


Fig. 11. Carrier concentration for bulk heterojunction organic solar cell

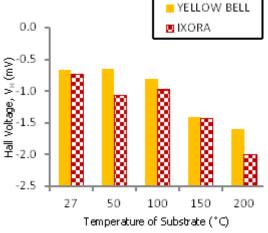


Fig. 13. The Hall voltage for bulk heterojunction organic solar cell

In illuminated condition, the plot shows that the electrical conductivity in different substrate temperature progressively rises as light intensity increased. The bottommost mark detected was using 27ÚC substrate temperature (unheated substrate) while the highest point was using 200ÚC substrate temperature for both sample when exposed to light illumination. This is due to natural phenolic compound namely anthocyanin. Anthocyanin (Figure 9) is involved in photo system assembly and contributes to light harvesting by absorbing light energy in a region of the visible spectrum [14]. Thus, the range of electrical conductivity of ITO/P3DT+YB detected was between 0.212 Sm⁻¹ to 0.277 Sm⁻¹ and for ITO/ P3DT+IX detected was between 0.235 Sm⁻¹ to 0.2604 Sm⁻¹.

The electrical conductivity was higher as substrate temperature increase. This is due after heat treatment, the particle size becomes larger, and the particle shape becomes more regular. Then, the film atoms more energetic at higher temperature to migrate and more un-perfect crystal nucleuses sufficiently grow, leading to the reconfiguration of the grains and the formation of particles with perfect crystal structure²². As a result, higher substrate temperature was led to the formation of lower resistance films, then causes a rises on electrical conductivity. This is basically due to the increase of the mobility and/or carrier concentration at higher substrate temperature. The increase in substrate temperature may have led to oxygen deficient films, resulting in an increase in carrier concentration as illustrated in Figure 10 and 11.

Hall Effect study

The Hall effects is a useful technique for making some electrical property measurements related to transport, such as the carrier concentration, Hall mobility, resistivity, and conduction type The Hall measurements were performed under room temperature for ITO/P3DT+YB and ITO/P3DT+IX blend thin film samples at different substrate temperature.

Hall Effect measurements revealed that all the films exhibited N-type carrier. On increasing the substrate temperature from 0 to 200ÚC, the carrier concentration increased from -1.65 \times 10^{20} cm $^{-3}$ to -1.15 \times 10^{20} cm $^{-3}$, while Hall voltage and Hall Coefficient decrease from -1.17 \times 10^{-3} cm $^{-2}V^{-1}S^{-1}$ to -2.79 \times 10^{-3} cm $^{-2}V^{-1}S^{-1}$. The Hall mobility also

underwent a change from 1.23 to 3.9 mV

Figure 10 clearly shows an increase as substrate temperature increases. Hall mobility is lower for unheated substrate (27ÚC) than heated substrate (50ÚC to 200ÚC) for all natural dyes blended P3DT samples. For unheated substrate, ITO/P3DT+IX exhibited the lowest Hall mobility with 1.23 mV and an upward tendency shows as substrate temperature increase (heated substrate). ITO/P3DT+YB were slowly increased as substrate temperature increase.

It is apparent that from Figure 11, the carrier concentration increases with increasing temperature of substrate for both samples. This can be explained by the lack of Oxygen to compensate possible divacancies and so, to an enhancement on the bulk defects²⁴. Furthermore, Oxygen vacancies induce free electrons as conduction carriers²⁵. The Hall mobility and carrier concentration increase with increasing temperature [26]. Free charge carriers are easily activated at relatively high temperature. This relationship is found to be in good agreement reported by²⁷ and²⁸. ITO/P3DT+IX blended samples reached a peak were -1.07×10^{20} cm⁻³ (at 200ÚC). The carrier concentration for heated ITO substrate was higher for all samples compared to samples deposited on unheated ITO substrate.

Referring Figure 12, for unheated substrate, the maximum Hall coefficient value observed was -1.77 \times 10⁻³ cm⁻²V⁻¹ s⁻¹ and -1.23 \times 10⁻³ cm⁻²V⁻¹ s⁻¹ for ITO/P3DT+IX and ITO/P3DT+YB respectively. In contrast, for heated substrate the greatest Hall coefficient value observed was the same sample at 50ÚC of temperature. After that, the Hall coefficient was decline as substrate temperature incline for both blended samples. This is revealed that, the heat treatment of substrate was affecting the Hall coefficient of blended samples.

In this case, light was used more effectively because the charge separation site increased by blending the component materials.

Figure 13, clearly shows that the Hall voltage is decreasing with the temperature of substrate. The unheated substrate (27 \circ C) deposited exhibited the maximum point. The lowest Hall voltage detected for ITO/P3DT+IX was -2.0 mV (at 200 \circ C). The range of Hall voltage obtained for these samples was between -0.67 mV to -2.0 mV.

This graph clearly shows that the Hall mobility of heated substrate was lower than unheated substrate for both blended samples. This phenomenon could be attributed to the increase of charge carrier concentration, n_s^{20} .

The charge carrier type can be determined by the polarity of the Hall voltage and Hall coefficient. If negative, the charge carriers are electrons and the material is of the N-type. In this work, it was found that, the Hall coefficient is negative for all samples. This finding indicates that the majority carriers are electrons, according to the concept of semiconductors. From the above discussions, we conclude that our samples deposited on ITO substrate are successfully investigated by Hall Effect measurement and improve the electrical properties of the commercial ITO substrate.

CONCLUSION

The ITO/P3DT+YB, ITO/P3DT+IX dye as Bulk Heterojunction Organic Solar Cell deposited on the heated ITO substrate were successfully prepared by electrochemistry method. The effect of substrate temperature on electrical conductivity, and Hall Effect of blended thin film was studied. The highest electrical conductivity at 200ÚC of temperature substrate was $2.27 \times 10^{-1} \, \text{Sm}^{-1}$ for ITO/ P3DT+YB For Hall Effect study it was found that, the polarity sign of Hall coefficient and polarity of Hall Voltage obtained were negative for all samples. This finding indicates that the majority carriers are electron. The ITO/P3DT+YB thin film shows higher electrical properties compared to ITO/P3DT+IX thin film samples. The presence of carbonyl and hydroxyl groups in the anthocyanin molecules bound to the surface of ITO, is in favor to facilitate the photoelectric conversion effect. From results obtained, we conclude that our samples deposited on ITO substrate are successfully investigated and improve the electrical properties of the commercial ITO substrate. From the results we may conclude that the substrate temperature play major role in controlling the electrical properties of the blended natural dye thin film. Futhermore, Allamanda Cathartica Linn (yellow bell flower) and Ixora coccinea L (Ixora flower) are cost effective, renewable, eco-friendly. Hence, the study of these dyes is promising and can promote additional

studies oriented to educate people on renewable energy sources, and disseminate knowledge for the optimization of solar cell components compatible with such dyes.

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