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Research Article
MEH-PPV: PCBM Solution for Screen-printing Technique in Polymer Solar Cell Fabrication

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Abstract
The fabrication and property characterization of bulk heterojunction photoactive layer made of blend of poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV) and (6,6)-phenyl C61 butyric acid methyl ester (PCBM) and fabricated by screen-printing technique was carried out. Effects of the photoactive solid concentration, temperatures at 15, 25 and 35°C and shear rate ranging from 100-10^4 sec^{-1} on the viscosity was investigated and shows that 1, 3 and 5% solid concentrated solutions are possible for using as ink in this technique in term of rheological property. Thickness of the films fabricated from different solid concentrated solutions at different print speed was measured and show a dependency of film thickness on print speed as well as exponential law dependence on solid concentration of the solution. Surface morphology and absorption coefficient of the films was also carried out and confirms that the films formed by the screen printer is well-adapted for fabricate photovoltaic devices.

Key words: Polymer solar cell, bulk heterojunction, screen printing

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

In recent time, solar cell market is ruled by silicon based technology because of its high power conversion efficiency. However, this technology suffers disadvantages such as high cost due to requirement of solar grade silicon, limits in application due to only flat shape (Carlson and Wronski, 1976; Yamamoto *et al.*, 2001). The Organic Solar Cell (OSC) is developed and paid much attention to researchers and scientists in effort to overcome the silicon based solar cell's limitation. Among the OSCs, the conjugated Polymer Solar Cells (PSC) (Yang *et al.*, 2012) have taken opportunity by the virtue of the polymer's conducting properties similar to those of the semiconductors, flexibility (Liang *et al.*, 2008), friendly-environment, easily-large-scaling fabrication (Brabec, 2004; Li *et al.*, 2012a; Krebs, 2009; Yusli *et al.*, 2009) and cost reduction in overall (Tanenbaum *et al.*, 2012). However, the shorter lifetime and the lower PCE in comparison with those of the inorganic solar cell require much effort in research and development (Jorgensen *et al.*, 2008; Chen *et al.*, 2009; Krebs and Spangaard, 2005; Shao *et al.*, 2013).

Buck heterojunction (BHJ) architecture of Donor (D) and Acceptor (A) material is a promising concept for the PSC. The PCE of 10.6% for an BHJ solar cell is reported by Li *et al.* (2012b). The conventional BHJ device structure is of an active layer consisted of A/D materials sandwiched between the transparent substrate and backside contact as anode and cathode electrode, respectively (Yu *et al.*, 1995). Operation based on principle of ultrafast photo-induced charge transfer from a conjugated polymer (D) to fullerene (A) molecule requires that an interpenetrating bi-continuous network of A and D materials formed in the BHJ active layer since the generated charges can be transported towards their respective electrodes. Indium Tin Oxide (ITO) covered with a thin layer of poly(ethylenedioxythiopene)/poly(styrene-sulfonate) (PEDOT/PSS) is commonly used as transparent electrode, whereas evaporated metal such as Ca, Al/LiF form the backside contact. Substrates selected for the BHJ solar cell affect the fabrication process and normally is glasses or Poly Ethylene Terephtalate (PET) foils in which the PET is capable to produce fully flexible solar cell due to its mechanical flexibility.

In OSC research progress, much effort has been focused on utilizing blend film of poly(3-hexylthiophene) (P3HT) and (6,6)-phenyl C61 butyric acid methyl ester (PCBM) as photoactive layer (Sharma *et al.*, 2012; Liao *et al.*, 2008; Xu *et al.*, 2012; Lira-Cantu and Krebs, 2006; Han *et al.*, 2009). The processing parameters thin film deposition of P3HT/PCBM was intensively studied by Takanezawa *et al.* (2007) and Chou *et al.* (2009). However, a few number of report has been conducted to poly[2-methoxy-5-(2'-ethylhexyloxy)-p-phenylenevinylene] (MEH-PPV) as D material blended with PCBM as A material in mean of BHJ photoactive layer. Both materials can be dissolved in organic solvent giving that it is able to apply solution based process such as screen-printing for fabrication of the active layer thin film. MEH-PPV has broad optical absorption region 400–600 nm and also higher hole mobility compared to electron mobility (Crone *et al.*, 1998).

In the present study, the effect of MEH-PPV and PCBM composition in chloroform (CF) on rheology of the blend solution is studied in order to apply for screen-printing technique. The printing parameters based on the studied solution are investigated and optimized. Performance of a fabricated PSC also discussed.

MATERIALS AND METHODS

The MEH-PPV with molecular weight of 40,000-70,000 g mol^{-1} and the PCBM was purchased from Sigma-Aldrich and Nano-C, respectively. Blend of MEH-PPV and PCBM with relative weight ratio of 1:1 was dissolved in chloroform to obtain 1, 3 and 5% concentrated solutions.

Rheology measurement: Temperature-dependence viscosity of the photoactive solutions was investigated using Carri-Med Rheometer (CLS2 500 K) at temperature ranging from 15-35°C with constant shear rate of 10 sec^{-1}. Shear rate dependence viscosity of the 3% polymer concentrated solution was also carried out at temperature of 15, 25 and 35°C.

Photoactive film formation: Photoactive thin films deposited on glass substrate were formed at room temperature from the blend solutions using a SP002 screen printer (Essemtec) with a polyurethane squeegee setting at 45° angle at different speeds. A description of the screen printing process is as follows: The screen is placed a few millimeters above the surface of the substrate. Upon loading the polymer solution onto the screen, the squeegee is then swept with a velocity of several hundreds of millimeter per second across the surface of the screen, momentarily contacting it to the FTO substrate. At this point, solution flows from the screen to the surface of the substrate. As the squeegee then passes over a region, the screen separates from the substrate, leaving behind solution that dries to yield a continuous film. For this study, a screen with a thread diameter of 30 mm and a mesh count of 181 cm^{-1} was used.

Characterization of the photoactive film: The resulting films were quantitatively evaluated by comparing the pixel intensity in color histogram of the digital picture of the film. Thickness of the dried films was obtained by using Alpha-step IQ profiler (KLA Tencor). The UV-visible transmission characteristic of the films was investigated using UV/VIS/NIR Spectrophotometer V570-JASCO.

RESULTS AND DISCUSSION

Rheological properties: Figure 1 shows the temperature dependence viscosity of the MEH-PPV:PCBM solutions at constant shear rate of 10 sec^{-1}. It can see that the viscosity decreases with the increasing temperature for all the solutions. However, the insignificant decrease of the viscosity indicates that no gelation phenomena are appearing in this temperature for all solutions. Another hand, the temperature range maybe set for working environment conducted to screen-print stage in PSC fabrication progress. The 1% solid concentrated solution has viscosity about $2.5x10^{-2}$ and $1.7x10^{-2}$ Pa sec^{-1} at 15 and 35°C, respectively. These values are much smaller than those of the 3% (about one fourth) and 5% (about one thirteenth) solid concentrated solution. The faster increase of viscosity in comparison with increasing solid concentration may associate to MEH-PPV solubility in chloroform.

Effect of shear rate on viscosity of the 3% solid concentrated solution was also investigated at temperature of 15, 25 and 35°C and shown in Fig. 2. It can again be observed the viscosity increases when the temperature is lowered. The results also clearly shows that a dependence of the viscosity upon the shear rate in such a way the viscosity decrease with the increasing shear rate. This behavior is named as pseudo-plastic (shear thinning regime) and caused by a strong interaction among the polymer chains in the solution. The MEH-PPV component in the blend is considered significant effect on this behavior due to interaction of its long polymer chains in comparison to the interaction of small molecules of the PCBM component. Furthermore, no any Newtonian plateau occurrence at the lower as well as the high shear rate was observed.

The rheological measurement confirms that all photoactive solutions are able to use as ink for screen printing. However, in practice, it may need a change of screen printing parameters in order to obtain better printing quality due to influence of environment temperature on ink viscosity.

Photoactive film characterization: The solutions were used as ink with a fixed paste-volume of 8 cm^3 m^{-2} for screen-printer. The printed films deposited on the FTO substrate (~20 Ω/sq) were dried and characterized in terms of thickness and surface morphology.

Figure 3 shows a dependence of the film thickness to the printing speed. As indicated in this figure, the final thickness layer decreases when a higher speed is applied for the all solutions. Based on these results, attempt to obtain a film having thickness of around 100 nm is possible for the all solutions, however, a higher speed is need for the 3 and 5% solid concentrated solution comparing to speed of 250 mm sec^{-1} for the 1% solution.

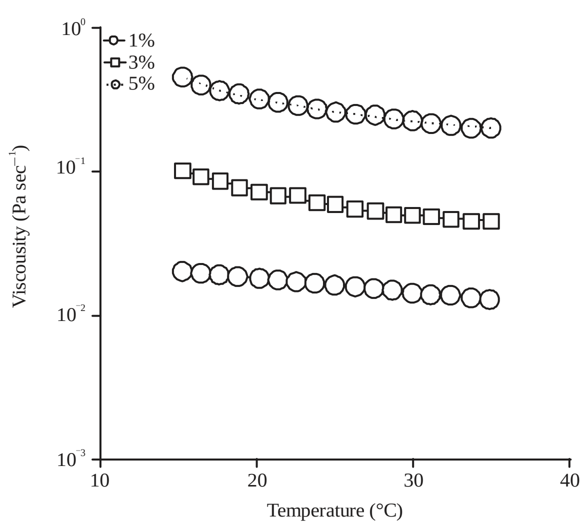

Fig. 1: Temperature-dependence viscosity at shear rate of 10 sec^{-1} of the MEH-PPV:PCBM solution

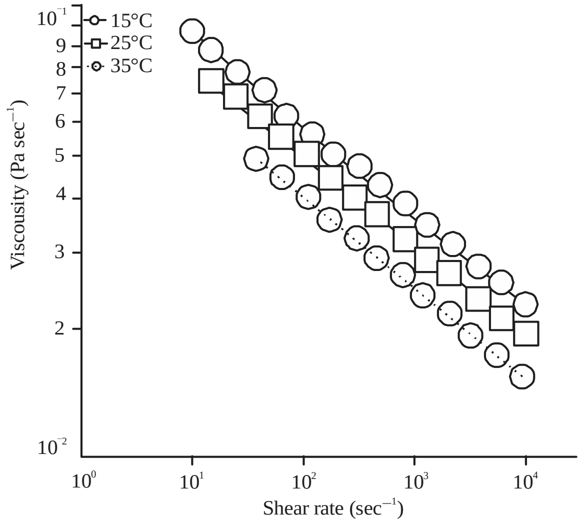

Fig. 2: Shear rate-dependence viscosity at different temperatures of the MEH-PPV: PCBM solution

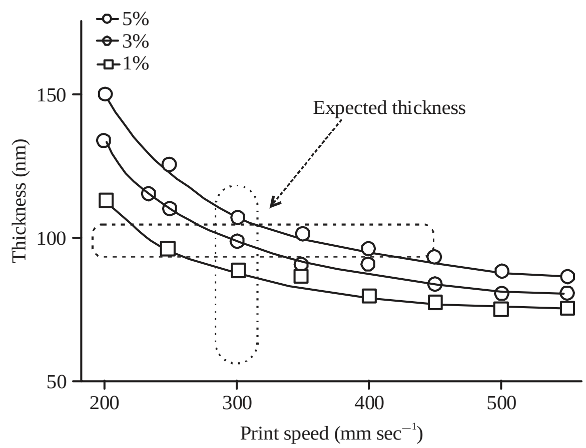

Fig. 3: Print speed dependence-film thickness of the MEH-PPV:PCBM solutions

The solid-concentration-dependent-film thickness is shown in Fig. 4. The thin films is performed at print speed of 300 nm sec^{-1}. A fit curve was carried out and shows that the thickness is correlated to the solid content by exponential law. A number of films produced confirm that this relationship was well-adapted.

Surface morphology of photoactive layer deposited on glass from different solid concentration solutions at print speed of 300 mm sec^{-1} was investigated and shown in Fig. 5.

The analyzed results show that the root mean square (rms) roughness increased significantly from 3.84-12.48 nm when the solid concentration decreased from 5-1%, respectively. This result means that the rougher surface may provide larger contact area between the photoactive layer and metal contact, leading to better collection of charge carrier (Lee and Park, 2000; Shen *et al.*, 2012). In addition, the thinner film obtained from the lower solid concentration solution may also result in better performance of photovoltaic cell.

Absorption coefficient of the composite film formed on glass substrate at print speed of 300 mm sec^{-1} from 1% solid concentration solution was determined using a UV/VIS/NIR Spectrophotometer V570-JASCO. The result is given in Fig. 6 and shows that absorption coefficient is function of wavelength. The absorption peak at 500 nm is attributed to MEH-PPV consistence in the composite whereas the peak of 340 nm at UV region and of 635 nm is well-associated to the acceptor component in the composite film. The optical band gap (E_g^{opt}) was calculated from onset wavelength (λ_{onset}) according to the following equation:

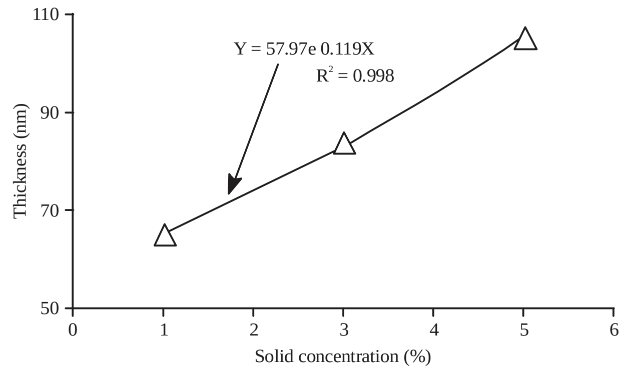

Fig. 4: Solid concentration dependence-film thickness of the MEH-PPV: PCBM solutions at the speed of 300 nm sec^{-1}

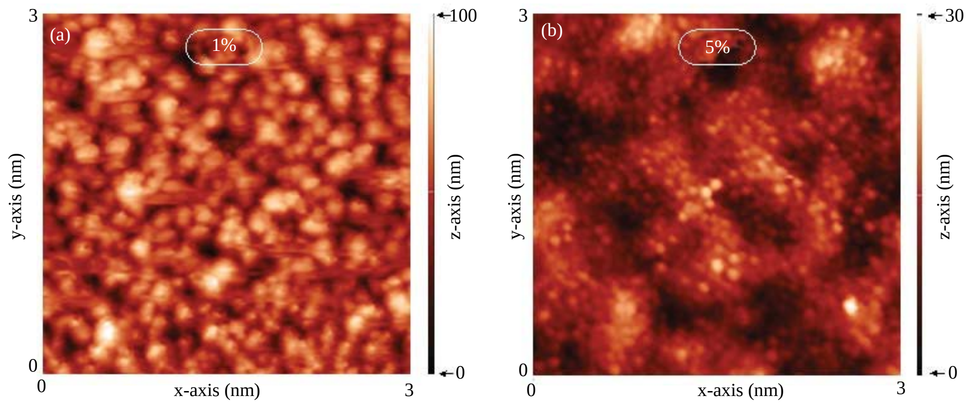

Fig. 5(a-b): AFM image of film surface formed on glass substrate at the speed of 300 nm sec^{-1} from 1 and 3% MEH-PPV: PCBM solution

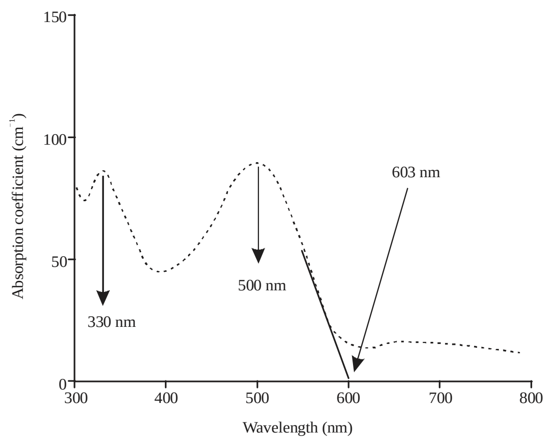

Fig. 6: Absorption spectra for MEH-PPV:PCBM film formed on glass substrate at the print speed of 300 nm sec^{-1} from the 1% solid concentration solution

$$E_g^{opt} = \frac{hc}{\lambda_{onset}}$$

where h is Plank constant and c is light speed in vacuum. The λ_{onset} and the corresponding E_g^{opt} of the film is 603 nm and 2.054 eV.

CONCLUSIONS

The MEH-PPV:PCBM (1:1 weight ratio) bulk heterojunction photoactive film was fabricated by screen-printing technique. Effects of the photoactive solid concentration, temperatures at 15, 25 and 35°C and shear rate ranging from 100-10^4 sec^{-1} on the viscosity was investigated and shows that 1, 3 and 5% solid concentrated solutions are possible for using as ink in this technique in term of rheological property. Thickness of the films fabricated from different solid concentrated solutions at different print speed was measured and show a dependency of film thickness on print speed as well as exponential law dependence on solid concentration of the solution. Surface morphology and absorption coefficient of the films was also carried out and confirms that the films formed by the screen printer is well-adapted for fabricate photovoltaic devices.

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