

## Brief Notes on Spectroscopy of Resistance for Thermal Treatment

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### Abstract

Organic photovoltaic cells (OPVs) have been studied for a long time, and due to their high photovoltaic performance of around 12%, practical realisations have also been ongoing. In this study, we created typical OPVs and looked at how annealing the MoOx layer affected the efficiency of optical-to-electrical conversion. After annealing at 160°C for 5 minutes, the photoconversion efficiency increased from 2.05% to 5.65%, and the external quantum efficiency likewise increased for all measured wavelengths between 300 and 900 nm. Among photovoltaic metrics, the short circuit current density increased in particular. During analysis of the impedance test, it was discovered that the carrier transport resistance of the photoactive layer had decreased. These findings show that the thermal annealing of the MoOx layer successfully eliminated flaws at the MoOx/organic interface. The annealed device's exceptional device performance was made possible by the efficient carrier transfer. The annealing procedure significantly decreased the relaxation and electron recombination periods, which increased photovoltaic performance.

**Keywords:** PTB7-Th; MoOx; Annealing; Impedance spectroscopy; Organic photovoltaic cells

### Introduction

The molecular structures of donor polymers and device topologies have been optimised recently to significantly increase the photovoltaic performances of organic photovoltaic cells (OPVs) [1-4]. With the help of the bulk heterojunction structure and the red-shifted absorption band of the p-type polymer, photovoltaic performance can exceed 10%. Moreover, the nanoscale shape of the donor-acceptor blend layer plays a crucial role in determining the likelihood of carrier recombination when exposed to solar radiation, which is directly related to photovoltaic efficiency [5-7]. Since most photoexcited excitons are deactivated without the donor/acceptor interface, effective exciton dissociation at the donor-acceptor interface is necessary for photocurrent generation. Although the optical absorption length is nearly identical to the organic active layer thickness of 80 to 200 nm, the conventional organic semiconductors' exciton diffusion length is shorter. Since donor-acceptor materials have a large contact area, bulk heterojunction architecture has been employed for a long time to address this issue. Several researchers have created assessment techniques for carrier dynamics in OPVs in addition to the manufacture of novel polymers, molecular stacking in the active layer, and optimised device designs [8-11]. To comprehend carrier dynamics in the OPVs, researchers have looked at a number of characterization techniques, such as the carrier mobility of organic materials [12, 1]. Through the equivalent circuit of the device, impedance spectroscopy is a crucial technique for discussing carrier dynamics, including carrier mobility and density. This method uses the phase difference between the input sinusoidal voltage and the response current to determine the impedance of the device. It is feasible to isolate and monitor components contributing to the impedance in the device that have different relaxation durations by examining the test findings over a broad frequency range (10-3 to 106 Hz). Also, because it uses a non-destructive measurement technique that can be used with a variety of electronic devices, it is one of the features that can measure a real device. In particular, one straightforward interpretation of impedance measurement holds that each layer's and interface's resistance and capacitance components can be distinguished from the equivalent circuit [8] [9]. Garcia-Belmonte et al. investigated the effect of bias voltage on the depletion layer capacitance and the minority carrier (electron), and by constructing an equivalent circuit they assessed electron mobility and lifetime [2]. Due to the creation of

a Schottky junction (band bending) at the organic/metal interface in traditional OPVs, the reverse bias capacitance typically displays Mott-Schottky-like behaviour. Also, using an equivalent circuit with charge transfer resistance, bulk layer capacitance, and the donor-acceptor interface, Leever et al. analysed the electron density and lifetime of an OPV as a function of applied voltage [1]. Recently, we showed that employing the annealed MoOx layer at 160 °C significantly increased the photovoltaic performance of the ITO/MoOx/organic active layer/LiF/Al device [2]. The oxygen vacancies in MoOx can be restored by the annealing process, which results in the efficient carrier injection at the MoOx/organic interface, according to an analysis of angle-dependent X-ray photoelectron spectroscopy. Many studies on the MoOx layer's surface have been published since it is frequently utilised as a hole transport layer in the typical OPV device architecture. However, the precise mechanism of effective carrier transport at the MoOx/organic layer contact is unknown, necessitating additional research. In order to minimise surface imperfections, the MoOx layer on the bulk heterojunction OPV in this study was annealed at 160 °C in an inert atmosphere. The equivalent circuit can be used to assess the resistance and capacitance elements in the organic layer. Also, in order to comprehend the mechanism underlying better photovoltaic performance, the relaxation time and electron lifetime of diffusion were assessed.

### Experimental

#### Materials

Poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b;4,5-b']],

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dithiophene-2,6-di-yl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene)-2-carboxylate-2-6-diyl)] (PTB7-Th) and [6,6]-phenyl-C71-butyric acid methyl ester (PC71BM, purity: >99.0%) were bought from Solene BV and 1-Material Inc., respectively. Kojundo Chemical Laboratory sold MoOx powder, which was acquired. These substances weren't further purified before being employed.

### Manufacturing of OPV

We created the OPV by following the steps. Using a chemical etching procedure, an indium tin oxide (ITO) anode with a thickness of 150 nm was first etched on a glass substrate. Acetone, isopropyl alcohol, and pure water were then applied in turn using an ultrasonic cleaner. The UV ozone cleaner was then used to clean the substrate for 20 minutes. The ITO layer was then thermally evaporated to create a MoOx layer that served as a hole transport layer with a controlled thickness of 40 nm. In order to prevent the unanticipated oxidation by air, the sample was then annealed at 160 °C for 5 min under a nitrogen environment (called as device A).

### The Findings and Discussion

The current density-voltage characteristics and EQE spectra of devices A (with MoOx annealing) and B (without MoOx annealing), respectively, are shown in Figures 1(a) and 1(b). Our results clearly demonstrate that when the MoOx layer was annealed at 160 °C for 5 min, enhanced photocurrent and increased EQEs across the entire wavelength band from 300 to 900 nm were seen. These outcomes line up with the 2.44-times-increased short circuit current density ( $J_{sc}$ ) in Table 1. Devices A and B had calculated photo conversion efficiencies (PCEs) of 5.65% and 2.05%, respectively. Also, Table 1 summarises the PCE,  $J_{sc}$ , open circuit voltage ( $V_{oc}$ ), and fill factor of devices A and B. The thermal annealing for the MoOx improved  $J_{sc}$  in particular, leading to higher PCE. This pattern supports the findings of our earlier research that annealing MoOx can enhance OPVs' photovoltaic performance. In the previous work, we came to the conclusion that the annealing process fills oxygen flaws that may operate as electron traps on the surface of MoOx, which is the reason why the OPV properties have improved. Additionally, this impact was equally validated at temperatures between 160 and 200 °C and for durations between 5 and 30 min (data not shown). A temperature of 160 °C for five minutes is a sensible option for high compatibility with other procedures, including the annealing of P3HT. For the measurement, 0 and 100 mV were chosen as the bias and AC voltages, respectively. Also, measurements were performed in the dark. Impedance spectroscopy is often carried out in complete darkness, however when discussing the formation of VOC, it is possible that measurement under light irradiation is required since the movement of photo carriers affected the electronic state at the interface. The cole-cole plots for both devices exhibited a nearly identical single semicircle shape, with the exception of the radius, which was decreased during the annealing process for the MoOx layer. Fitting curves matched the impedance spectra satisfactorily in the entire measurement frequency range from 0.1 Hz to 1 MHz by assuming a single semicircle. The resistance component of the PTB7-Th: PC71BM layer is corresponding to the semi-radius circle's. PTB7-Th: PC71BM layers in devices A and B had calculated resistance components of 6.5 and 17 M $\Omega$ , respectively. This outcome suggests that following annealing for the MoOx layer, the resistance component of the PTB7-Th: PC71BM layer was decreased. In our work, the PTB7-Th: PC71BM layer spin-coating conditions were exactly the same for both devices, and the PTB7-Th molecule orientation was also the same for both devices because annealing was done before spin-coating. As a result, the

annealing of MoOx had no effect on the carrier mobility of PTB7-Th: PC71BM. These findings suggest that the MoOx/PTB7-Th: PC71BM interface's carrier injection is what caused the lowered resistance. Our earlier research showed that annealing might minimise the surface flaws of MoOx, which are brought on by the material's oxygen vacancy [1]. For device A, it is possible to realise an effective carrier injection at the MoOx/PTB7-Th: PC71BM layer contact. DC bias voltage obtained from an impedance test. The relationship between the resistance and the bias voltage, which ranges from 0 to 5 V for devices A and B, is depicted in Figure 2(b). Such an impedance pattern is an example of an ordinary response, in which diffusion-recombination between nano-absorbing contacts controls carrier transport [4]. Moreover, the PTB7-Th: PC71BM active layer can diffuse injected minority carriers (i.e., electrons) from the Al electrode, and the impedance model consists of an equivalent circuit as shown in Figure 3 [2]. It includes distributed resistors  $r_t$ , distributed capacitance  $C_n$ , and recombination resistance  $r_{rec}$ , which account for the processes of electron recombination. Due to the carrier injection caused by providing DC bias voltage in both cases of devices A and B, the  $r_{rec}$  constantly dropped with rising bias voltage. At all of the DC voltage values, thermal annealing lowered electron recombination resistances. It suggests that the thermal annealing of the MoOx layer increased the carrier injection efficiency at the MoOx/PTB7-Th: PC71BM. Following the thermal annealing of MoOx, device A's photovoltaic performance is enhanced [12]. We then assessed the modulus cole-cole plot to look into the capacitance component of the PTB7-Th: PC71BM layer for both devices because distributed capacitance and electron recombination resistance are both known to be significant parameters to affect the carrier injection/transport at the interface and inside the organic active layer. The bias voltage was set to 0 V for OPVs. Only a portion of the semicircle was seen because the measuring frequency range was between 0.1 Hz and 1 MHz, as is seen in Figure 4. (a). However, applying the Debye relaxation model in all measurement frequencies allowed the measured modulus cole-cole plot to be fitted as a semicircle [1]. After annealing the MoOx layer, the distributed capacitance component ( $C_n$ ) of the PTB7-Th: PC71BM layer, as represented by the radius of modules cole-cole plot in Figure 3, rose from 0.040 nF to 0.068 nF. As a result, only the capacitance component of the PTB7-Th: PC71BM layer rose after the thermal annealing of the MoOx layer at 160 °C, which was the opposite trend of the electron recombination resistance component in Figure 2. The lowered electron recombination resistance influences the carrier transport from the PTB7-Th: PC71BM layer to the MoOx layer, however the effect of annealing on electron recombination resistance was greater than that of the capacitance component.

### Conclusion

We created a standard OPV using a PTB7-Th: PC71BM photoactive layer and a MoOx hole transport layer, and we used impedance spectroscopy to analyse the mechanism behind the increased photo conversion efficiency. The electron recombination time was determined from the electron recombination resistance and the dispersed capacitance by analysing the electrical and modulus cole-cole plots. The MoOx layer's annealing procedure reduced the electron recombination time, which enabled efficient carrier dissociation in the PTB7-Th: PC71BM layer. Also, as a function of measurement wavelength, the relaxation time was inferred from the imaginary portion of impedance. The annealing process also reduced the relaxation time. These findings therefore show that the PTB7-Th: PC71BM layer's photo-induced carriers were effectively removed, leading to the increased PCE. By increasing measurement precision and speed in the future, this nondestructive evaluation approach can be used for in-line assessment,

enhancing OPV productivity and dependability.

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### Competing Interest

The authors say they have no competing interests.

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