Impedance analysis of the multilayered organic solar cells with and without hole buffer layer

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Abstract

We have investigated the capacitance and conductance of the multilayered organic photovoltaic devices consisting of ITO/(hole buffer layer)/p-type tetraphenyldibenzoperiflanthene(DBP)/ fullerene C60/ bathocuproine (BCP)/Al devices in dark and under AM1.5 illumination. DBP without hole buffer layer act as a dielectric layer and most of the external voltage applied to DBP. The thin molybdenum oxide layer reduces the interfacial resistance and contributes to the increment of V_{oC} and the conductance across the film resulting to the improved photovoltaic properties.

1. Introduction

Organic semiconductors are considered as a potential candidate for fabricating low-cost, flexible, and efficient organic photovoltaic devices (OPVs). The power conversion efficiency (η_P) of solar cells depends on the open-circuit voltage (V_{OC}), the short-circuit current density (J_{SC}), and the fill factor (FF), and all of which still need improving for practical applications.[1-4] Since the upper limit of the V_{OC} is considered as the energy difference between the lowest unoccupied molecular orbital (LUMO) level of the acceptor (n-type) and the highest occupied molecular orbital (HOMO) level of the donor (p-type) ΔE at the heterojunction (HJs) interface, the relatively low gap donor materials with large ionization potential (IP) are preferred for the application of organic solar cells. However, a larger IP value basically increases the hole injection barrier height at the electrode/donor interface. One of the strategies to improve the efficiency of organic solar cells with a given set of organic semiconductor materials is to insert an interfacial buffer layer, such as PEDOT-PSS and transition metal oxide, between active layers and electrodes to improve the charge transport and extraction.[6-12] On the other hand, the organic film without carrier doping acts as a dielectric material, while it acts like the inorganic semiconductors when the carriers are generated in the films. Therefore, the capacitance behaviors under the external voltage and photoillumination should give important information for understanding the electrical, charge behavior in the organic photovoltaic devices.

In this study, we have investigated impedance behaviors of the multilayered organic photovoltaic devices consisting of ITO/hole buffer layer/ donor/ fullerene C60 (50 nm)/ bathocuproine (BCP)/ Al structures. We mainly focused on tetraphenyldibenzoperiflanthene (DBP) as a donor material and compared the devices with and without the hole buffer layer. We then investigated the impedance components, such as the capacitance and conductance, and their changes as the functions of the frequency, voltage, and the thickness of DBP layer.

2. Experimental

All devices were fabricated on 150-nm-thick ITO-coated glass substrates. Figure 1 shows the chemical structures and energy diagrams of the materials used in this study. After patterning the ITO (2mm in width), we prepared a known multilayered structure based on ITO/hole buffer layer/DBP (20, 40, 60 nm) /C60 (50 nm)/BCP (10 nm)/Al(100 nm). We adopted molybdenum oxide and PEDOT-PSS as the hole buffer layers. They were deposited by evaporation technique in a vacuum of $\sim 3 \times 10^{-4}$ Pa except for PEDOT-PSS. The deposition rates of MoO_3 , DBP, C_{60} , BCP, and Al were 0.05, 0.1, 0.1, 0.1, and 1 nm/s, respectively. Finally, 2-mm-wide, ca. 100-nm-thick Al electrodes were evaporated on the organic layers in a different vacuum chamber after brief exposure to air. The effective area is 4 mm². Electrical characterizations were performed in a grove box filled with argon with an ammeter Keithley 6487, chemical impedance analyzer (Hioki IM3590), and a global 1 sun AM1.5 solar simulator (SAN-EI Electric model XES-40S1). All devices were illuminated through ITO electrodes.

3. Results and Discussion

Figure 1 shows the frequency dependence of the capacitance and the loss (conductance G/ω) under various forward biasing in the device consisting of ITO/DBP (40nm)/C60/BCP/Al device in dark condition. The device without hole buffer layer act as a dielectric film in dark, and the capacitance converges to the series capacitance of DBP (hole transporting p-type layer) and C60/BCP (electron transporting n-type layer) at the high frequency, while it converges to the capacitance of DBP single layer at low frequency. The loss becomes inversely proportional to the frequency below 1 kHz under the external voltage above 0.6V due to the increase of conductance across the film. The peaks or shoulders appear in the loss-frequency curves due to the charge accumulation at the DBP/C60 double layered dielectric interface based on the Maxwell Wagner effect. It is the evidence of the dielectric behavior in p-type



Fig. 1 C-f and Loss (G/ ω)-f curves of ITO/DBP(40nm) /C60/BCP/A1 in dark condition

DBP layer due to the extremely low charge carrier density in dark condition.

Figure 2 shows (a) the current density-voltage (J-V) curves under white light illumination, (b) C-V, and (c) conductance G-V curves in dark and under illumination measured at 100 Hz, respectively. Similar to the discussion in Fig. 1, the DBP layer acts as a dielectric layer in the device without buffer layer and the capacitance converges to that of DBP single layer at the voltage above V_{OC} of ~0.6 V. On the other hand, the capacitance of the device with MoOx layer decreases above V_{OC} of ~0.9 V due to the marked reduction in the impedance of multilayered device. The increase of V_{OC} with MoO_x is ascribed to the increment of built-in potential across the film and the reduction of hole injection barrier causes the steep increment of congdactance G. It is interestingly noted here that the peaks are observed in both capacitance and conductance curves and it is probably ascribed to the accumulation of photo-generated carriers in the HJs, and it decreases with the reduction of external voltage owing to the reduction of carrier density by the increment of internal electric field (swept out of photo-carriers by high electric field). On the other hand, C and G decrease and show minimum value at V_{OC} probably owing to the recombination of carriers and the reduction of internal electric field. The steep reduction of G in the device without hole buffer layer corresponds well to the S-shape curves in the J-V curves. The thickness dependence will be also discussed in the conference.

3. Conclusions

We have investigated the capacitance and conductance of the multilayered photovoltaic devices with and without buffer layers. The DBP acts as a dielectrics and causes the



Fig. 2 (a) J-V curves under AM1.5 illumination, (b) C-V, and (c) G-V curves of ITO/(MoOx)/DBP(40nm)/C60/BCP/Al devices in dark and under illumination (AM1.5).

S-shape curves. It is improved by the insertion of buffer layer, and related with the photo-generated space charges.

References

[1] P. Peumans, A. Yakimov, and S. R. Forrest: J. Appl. Phys. 93 (2003) 3693.

[2] P. W. M. Blom, V. D. Mihailetchi, L. J. A. Koster, and D. E. Makov: Adv. Mater. 19 (2007) 1551.

[3] B. P. Rand, D. P. Burk, and S. R. Forrest: Phys. Rev. B 75 (2007) 115327.

[4] T. Taima, J. Sakai, T. Yamarani, and K. Saito: Jpn. J. Appl. Phys. 45 (2006) L995.

[5] J. Y. Kim, S. H. Kim, H. Lee, K. Lee, W. Ma, X. Gong, and A. J. Heeger: Adv. Mater. 18 (2006) 572.

[6] J. Y. Kim, K. Lee, N. E. Coates, D. Moses, T. Nguyen, M. Dante, and A. J. Heeger: Science 317 (2007) 222.

[7] V. Shrotriya, G. Li, Y. Yao, C. W. Chu, and Y. Yang: Appl. Phys. Lett. 88 (2006) 073508.

[8] Y. Kinoshita, R. Takenaka, and H. Murata: Appl. Phys. Lett. 92 (2008) 243309.

[9] M. D. Irwin, D. B. Buchholz, A. W. Hains, R. P. H. Chang, and T. J. Marks: PNAS 105 (2008) 2783.

[10] E. Itoh, Y. Highchi, D. Furuhata, and T. Shirotori: Jpn. J. Appl. Phys. 50 (2011) 01BC14.

[11] E. Itoh, Y. Higuchi, D. Furuhata, and T. Shirotori : IEICE Trans. Electron. E94-C, (2011) 181.

[12] E. Itoh and T. Shirotori: Jpn. J. App. Phys. 51(2012) 02BK14.