Rekayasa Antarmuka Sel Surya Perovskit Berbasis Karbon Melalui Polimer Donor sebagai Agen Pasivasi dan *Hole Transport Layer* (HTL)

Program Riset Pengembangan Unggulan 3P 2026

Setiyadi Tri Utomo[1,2], Sudirman[3,4], Prof. Rachmat Hidayat, S.Si., M.Eng., Ph.D.[5], Prof. Ir. Ahmad Nuruddin, M.Sc., Ph.D.[6], Yuliar Firdaus, Ph.D.[2], Prof. Dr. Veinardi Suendo, S.Si., M.Eng.[3,7]

[1]Program Magister Nanoteknologi, ITB; [2]Pusat Riset Elektronika, BRIN; [3]KK Kimia Anorganik dan Fisik, FMIPA ITB; [4]Jurusan Kimia, Universitas Mataram
[5]KK Fisika Magnetik dan Fotonik, FMIPA ITB; [6]Advanced Functional Materials Laboratory, FTI ITB; [7]Pusat Penelitian Nanosains dan Nanoteknologi, ITB

Abstract

Carbon-electrode perovskite solar cells (PSCs) are a low-cost alternative to devices with evaporated noble-metal contacts, but their performance depends strongly on the perovskite/hole-transport layer (HTL)/carbon junction. Here, six conjugated donor polymers (P3HT, PCE10, PCE12, PM6, PM7, and PTQ10) were evaluated as dopant-free HTLs in planar FTO/SnO_2/$FAPbI_3$/HTL/carbon cells. Energy levels, surface coverage, photovoltaic response, EQE, impedance, dark current, hole-only injection, space-charge-limited current (SCLC), and light-intensity-dependent voltage were compared. The polymers with relatively shallow HOMO levels gave the most balanced contacts. PCE12 reached the highest average and champion power conversion efficiencies (PCE) of 10.83 ± 0.62% and 11.97%, with an average short-circuit current density of 21.01 ± 0.84 mA cm^{-2}. The deep-HOMO polymers gave much lower fill factors (FF), although their effective trap densities differed only slightly. PM6 produced an extraction-limited S-shaped *J–V* curve, whereas PTQ10 combined high forward injection with a lower voltage. HOMO alignment or trap density alone therefore cannot predict HTL performance. Efficient carbon-electrode devices need a balance of perovskite-side selectivity, polymer morphology and conductivity, and low-resistance hole transfer to carbon.

Keyword: Donor polymers, hole-transport layers, carbon-electrode perovskite solar cells, interfacial energetics, charge extraction.

Introduction

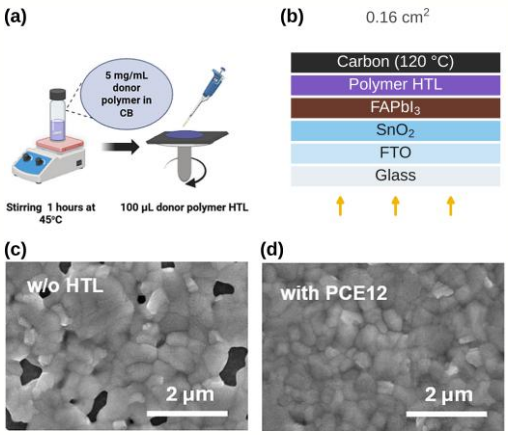
Metal-halide perovskite solar cells combine strong light absorption, long carrier diffusion lengths, and solution processability. For commercial modules, they must also be cheap, scalable, and stable. Carbon electrodes meet these needs because they are earth-abundant, chemically robust, printable at low temperature, and free from the metal diffusion and halide corrosion seen with Au or Ag. Planar carbon-electrode cells are, however, still less efficient than metal-electrode cells, mainly because carbon forms a non-ideal contact with the perovskite or the hole-selective layer.

In these devices, the HTL works at two interfaces. It has to extract holes selectively from the perovskite and suppress recombination at the absorber surface, and it then has to pass the holes to carbon with a low contact resistance. A deep HOMO can improve selectivity on the perovskite side, whereas a shallower HOMO may be needed for a near-ohmic contact with carbon (work function ≈ −5.0 eV).

Conjugated donor polymers are attractive dopant-free HTLs because their energy levels, packing, and interfacial chemistry can be tuned. These properties are strongly coupled, so the performance of a polymer cannot be predicted from its HOMO level alone. In this work, six donor polymers with HOMO levels from −5.20 to −5.54 eV were compared in carbon-electrode $FAPbI_3$ cells to separate perovskite-side selectivity from carbon-side contact formation.

Research Method

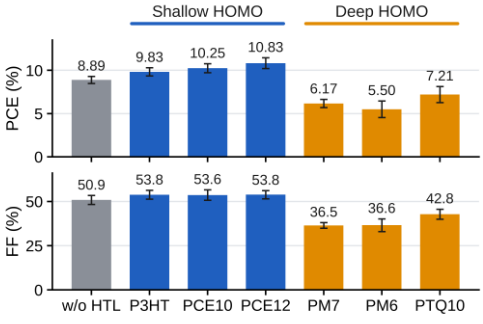
Materials. Six donor polymers were compared as dopant-free HTLs. They were grouped by HOMO level relative to the $FAPbI_3$ valence band (−5.40 eV) into a shallow group (P3HT −5.20, PCE10 −5.24, and PCE12 −5.33 eV) and a deep group (PM6 −5.45, PM7 −5.52, and PTQ10 −5.54 eV).



Picture 1. (a) HTL deposition, (b) device structure, and SEM images (c) without HTL and (d) with PCE12.

Device fabrication. A SnO_2 layer was spin-coated from $SnCl_4 \cdot 2H_2O$ on cleaned FTO and annealed at 180 °C. $FAPbI_3$ containing MACl was deposited by a one-step antisolvent method and annealed at 100 and 140 °C. Each polymer (5 mg mL^{-1} in chlorobenzene, stirred at 45 °C) was spin-coated at 3000 rpm and annealed at 100 °C (Picture 1a). Carbon paste was slip-cast and cured at 120 °C to give FTO/SnO_2/$FAPbI_3$/HTL/carbon cells with an active area of 0.16 cm^2 (Picture 1b). Cells without an HTL were used as the control.

Characterization. SEM, *J–V* under AM 1.5G (100 mW cm^{-2}), EQE, EIS (10 Hz–200 kHz), dark *J–V*, hole-only devices (FTO/PEDOT:PSS/$FAPbI_3$/HTL/carbon) with SCLC analysis, light-intensity-dependent V_{OC}, and stability during N_2 storage for 2500 h.



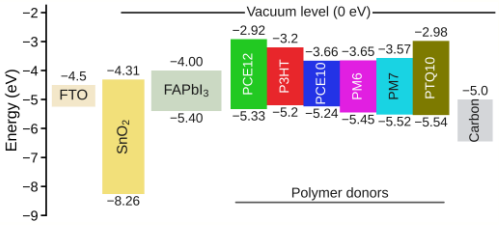
Picture 2. Average PCE and FF (± SD) for each HTL.

Discussion & Result

SEM shows dark pinholes on the bare $FAPbI_3$ film (Picture 1c), through which the carbon paste could touch the perovskite directly. The polymers cover most of these openings, and PCE12 gives the most compact surface (Picture 1d).

Picture 2 shows how the HTLs change the device performance. The shallow-HOMO polymers raise the average PCE from 8.89% for the control to 9.83%, 10.25%, and 10.83% for P3HT, PCE10, and PCE12. The gain comes mainly from a higher J_{SC} (21.01 mA cm^{-2} for PCE12), while V_{OC} stays at 0.96–0.97 V. In contrast, the deep-HOMO polymers lower the FF to 36–43% and the average PCE to 5.50–7.21%, below that of the control.

The energy diagram in Picture 3 explains this split. The HOMOs of P3HT, PCE10, and PCE12 lie between the $FAPbI_3$ valence band (−5.40 eV) and the carbon work function (−5.0 eV), so holes reach carbon in small steps. PM6, PM7, and PTQ10 lie at or below the valence band. This may help selectivity on the perovskite side, but it leaves an offset of about 0.5 eV to carbon, and it did not raise V_{OC} (0.89 V for PM6 and 0.92 V for PTQ10).



Picture 3. Energy-level diagram.

EQE and EIS support this interpretation. PCE12 and PCE10 give the highest EQE plateau and the largest recombination resistance, whereas the deep-HOMO devices show neither, so their losses occur during extraction rather than generation.

Hole-only devices probe the carbon side directly. PM6 shows weak hole injection from carbon, in line with its S-shaped *J–V* curve. PTQ10 injects holes well but still has a low V_{OC}, suggesting poor electron blocking. Both give almost the same SCLC trap density (≈7.6 × 10^{15} cm^{-3}), so trap density alone does not set the FF. With PCE12, the slope of V_{OC} versus light intensity falls from 2.63 to 1.51 kT/q, consistent with less trap-assisted recombination when both interfaces work well.

Conclusion

The HTL in carbon-electrode $FAPbI_3$ cells has to work at two interfaces. Shallow-HOMO polymers, especially PCE12 (10.83% average and 11.97% champion PCE), improve hole collection at carbon, whereas deep-HOMO polymers lose FF through contact resistance and mismatch with carbon. Future HTLs should combine perovskite-side passivation with quasi-ohmic hole extraction to carbon, for example in a bilayer.