The growing interests in polymer solar cells (PSCs)\(^1-^2\) are related to their unique advantages such as low-cost manufacturing and easy processability over large-area size via printing and roll-to-roll coating technologies,\(^3\) and compatibility with flexible substrates and materials availability, which enable them for potential applications in low-cost photovoltaic systems. Although the power conversion efficiency (PCE) of state-of-the-art PSCs has already exceeded 7% in scientific literature,\(^4,^5\) further improvements are needed for mass production and practical applications, especially simultaneous enhancement of short-circuit current density (\(J_{sc}\)),\(^6\) open-circuit voltage (\(V_{oc}\)),\(^7,^8\) and fill factor (\(FF\))\(^9\) are in urgent need. Among all of the approaches to improve the PCE, the most common and successful strategy is developing new low bandgap donor materials\(^5,^{10-12}\) to maintain a broad overlap with the solar spectrum and ensure effective harvesting of solar photons, which leads to higher \(J_{sc}\). However, since there is a trade-off between light harvesting \(J_{sc}\) and open-circuit voltage \(V_{oc}\),\(^13\) a gain in PCE can be expected only if a decrease of \(V_{oc}\) can be avoided. Apart from this, other approaches based on device physics have also been studied and proved to be effective, but these methods normally can only improve one or two key parameters of PSCs substantially at a time. For example, incorporation of an additive,\(^14\) controlling the growth rate of film,\(^15\) or optimizing film thickness in combination with proper annealing\(^16\) alter the active layer morphology and enhance \(J_{sc}\) and \(FF\); inserting an optical spacer layer solely increases the \(J_{sc}\)\(^17\) and the tandem cell approach greatly enhances either the \(J_{sc}\) or \(V_{oc}\) depending on the device structure.\(^18-^19\) Besides the methods described above, it was reported by our group that alcohol/water-soluble polymer cathode interlayers can enhance \(V_{oc}\) of PSCs prepared from polyfluorene and polycarbazole derivatives.\(^20-^22\) In parallel, other polyelectrolytes have also been reported to have similar functions\(^23,^24\) and in particular the Bazan group improved the efficiency of PSCs based on poly[N,N′-hepta-decanyl-2,7-carbazole-alt-5,5-(4′,7′-di-2-thienyl-2′,1′,3′-benzothiadiazole)] (PCDTBT) to 6.5% via incorporation of a conjugated polyelectrolyte interlayer.\(^25\) Although it is indispensable to obtain simultaneous enhancement in \(V_{oc}, J_{sc}\), and \(FF\) in order to push the PCE of PSCs towards their theoretical limit (~10%),\(^26\) there are few methods so far in the literature that are able to simultaneously improve all three parameters towards realization of the high efficiency of 10%.

In this study, we apply our method of incorporating an alcohol/water-soluble conjugated polymer, poly[(9,9-bis(3′-N,N-dimethy lamino) propyl)-2,7-fluorene-alt-2,7-(9,9-diocylfluorene)] (PFN, see Scheme S1, Supporting Information) as a novel cathode interlayer in polymer:fullerene bulk heterojunction solar cells based on [6,6]-phenyl C\(_{71}\)-butyric acid methyl ester (PC\(_{71}\)BM) (PFN, see Scheme S1, Supporting Information) as a novel cathode interlayer in polymer:fullerene bulk heterojunction solar cells based on [6,6]-phenyl C\(_{71}\)-butyric acid methyl ester (PC\(_{71}\)BM) and thieno[3,4-b]thiophene/benzodithiophene (PTB7, Scheme S1, Supporting Information).\(^28\) The synthesis of PFN has been described elsewhere\(^28\) and the applications of this material for highly efficient polymer light-emitting devices (PLEDs) and other optoelectronic devices have been reported in previous publications by our group\(^29,^30\) and other groups.\(^31\) The PSCs incorporating the PFN interlayer show significant and simultaneous enhancement in \(J_{sc}\), \(V_{oc}\), and \(FF\), which leads to a PCE of 6.79% and 8.37% for PCDTBT and PTB7 devices, respectively, and is the best reported in literature to date for PSCs. Due to drastic improvement in efficiency and easy utilization, this method opens new opportunities for PSCs from various material systems to improve towards 10% efficiency. Similar to all-printed red-green-blue (RGB) emission PLEDs demonstrated previously,\(^32\) the fabrication of these high-performance PSCs is compatible with low-cost, roll-to-roll processing,\(^33\) which is now on the eve of a critical development where round-robin or interlaboratory studies are possible.\(^33\)

Figure 1a presents the current density versus voltage (\(J-V\)) characteristics of the best PCDTBT PSCs with and without the PFN interlayer between the active layer and the Al cathode under 1000 W m\(^{-2}\) air mass 1.5 global (AM 1.5 G) illumination. For each type, over 200 devices were made from over...
40 individual films. A high PCE of 6.55 ± 0.09% was achieved in optimized devices with interlayer (with the highest value of 6.73%), while a 3.90 ± 0.17% (with the maximum value of 4.02%) was obtained for the control devices without interlayer. Incorporation of the PFN interlayer leads to a simultaneous enhancement in $V_{OC}$ (0.90 V vs. 0.70 V), $J_{SC}$ (12.1 mA cm$^{-2}$ vs. 11.4 mA cm$^{-2}$) and fill factor (62% vs. 49%), resulting in a drastic improvement in the PCE by 68% (6.55% vs. 3.90%) over the control devices.

Conventional cathode interlayers using low work function metal such as Ca are widely applied to form ohmic contact with the active layer and improve electron extraction.\[13\] We also prepared control devices using 3 nm thick Ca film as the cathode interlayer. It shows that Ca interlayer only results in moderate improvement in $J_{SC}$ (12.4 mA cm$^{-2}$ vs. 11.4 mA cm$^{-2}$), but no improvement in $FF$ (46%) and $V_{OC}$ (0.72 V), resulting in a PCE of 4.11%. Application of the PFN interlayer between the active layer and Ca not only retains the $J_{SC}$ (12.7 mA cm$^{-2}$) but also improves $FF$ (59%) and $V_{OC}$ (0.90 V), leading to a PCE of 6.79%. Thus we believe the functions of PFN must involve a mechanism different from the Ca interlayer.

Under AM 1.5G illumination, the device with a thin (∼5 nm) PFN interlayer shows high $V_{OC}$ of 0.91 ± 0.02 V, which is significantly higher than that of the control devices (0.71 ± 0.02 V). In dark, the device with PFN interlayer exhibits a turn-on voltage of about 0.8–1.0 V while it is only 0.5–0.6 V for the control device (Figure 1b), implying that the built-in potential ($V_{bi}$) across the device, which is the upper limit of the attainable $V_{OC}$ in PSCs,\[20,26\] drastically increases upon utilization of the PFN cathode interlayer. A similar increase in $V_{bi}$ has been observed in PLEDs devices utilizing PFN interlayers. An interfacial electrical dipole due to the strong interaction and alignment of polar amino group at the side chain of PFN with the active layer and a consequent vacuum level shift at the interface was identified between PFN and active layer of PLEDs by using small angle X-ray diffraction and electroabsorption techniques.\[30,34,35\] We suspect similar interfacial dipole between PFN and PCDTBT:PC71BM active layer could be the origin of enhanced $V_{bi}$ in our devices.

To visualize the interfacial dipole upon the incorporation of the PFN interlayer, we directly probe the surface potential change using scanning Kelvin probe microscopy (SKPM) with the experimental setup depicted in Figure 2c. The surface potential is uniform over both the active layer and the interlayer areas, but the surface potential of interlayer area is about 300 mV more positive than that of the active layer without interlayer (Figure 2b,d). At the edge of the interlayer area, the topography shows an abrupt bump because of accumulated material, but the surface potential remains the same as the rest of the interlayer; therefore, the difference in surface potential is due to chemical and/or physical nature of the interlayer rather than the height contrast. This indicates a microscopic electric dipole moment with the positive charge end pointing toward the Al electrode and the negative charge end pointing toward the active layer. The direction of this dipole moment is aligned with the built-in potential originated from the asymmetric contact at the electrodes; therefore, the actual built-in potential across the device is reinforced as a result of the superposition (Figure 2f). For an order-of-magnitude rough estimate, the electrical field...
due to the interfacial dipole layer is about $6 \times 10^5 \text{ V cm}^{-1}$ (0.3 V potential difference over a thickness of 5 nm) (Figure 2f), while the built-in field in devices without the PFN interlayer under short-circuit condition (with zero external applied bias) is about $7 \times 10^4 \text{ V cm}^{-1}$ (0.7 V potential difference over a thickness of 100 nm) (Figure 2e). Therefore, the incorporation of the PFN interlayer not only enhances $V_{bi}$, but also exerts a strong electrical field at the active layer/cathode interface that may strongly influence charge transport and extraction. This new approach of increasing $V_{bi}$ and $V_{OC}$ of the device by exploiting an interfacial dipole layer does not directly change the energy levels of active materials and their bandgap. It retains the solar spectral range utilized by the active material and does not sacrifice light absorption. Therefore, the trade-off between $V_{OC}$ and $J_{SC}$ can be avoided and we even observe simultaneous improvement as discussed in following sections.

In addition to the enhancement of $V_{bi}$, the PFN interlayer also avoids Fermi level pinning between the metal cathode and the acceptor material PCBM in the active layer. Therefore, the PFN/Al cathode can be expected to be an electron-selective electrode in nature. Indeed, the electrical leakage for the devices with an interlayer is considerably suppressed (Figure 1b). The superior diode quality to that of the control devices (characterized with much higher rectification ratio at -2 V, i.e., 283 000 vs. 5000) indicates that the interlayer is efficient in blocking holes and collecting electrons. As a consequence of reduced leakage current, a slightly higher $V_{OC}$ can be expected according to the Shockley equation. In addition, a reduced series resistance in the devices with an interlayer can further ensure a reduced $V_{OC}$ loss due to potential drop at the bulk of the active layer and the interface with the electrodes.

Besides the significant improvement in $V_{OC}$, an increase of $J_{SC}$ from 11.4 mA cm$^{-2}$ to 12.1 mA cm$^{-2}$ (by an extra increase of 5%) was observed upon the incorporation of the interlayer. The external quantum efficiency (EQE) spectra of the devices illuminated by monochromatic light are shown in Figure 1c. An average EQE of 70% in range of 350–650 nm are observed for both the devices with and without the interlayer. Note that theoretical $J_{SC}$ obtained by integrating the product of the EQE data in Figure 1c and the AM 1.5G solar spectrum (12.0 mA cm$^{-2}$ and 11.4 mA cm$^{-2}$ for the devices with the interlayer and without the interlayer, respectively) perfectly matches the values measured from $J$–$V$ characteristics (Figure 1a; 12.1 and 11.4 mA cm$^{-2}$, respectively).

Simultaneously with the enhancement in $V_{OC}$ and $J_{SC}$, the FF of the optimized devices is found to be 62%, much higher than that of the control devices (49%), suggesting that the charge transport properties are substantially improved. In order to make a realistic evaluation on the apparent charge carrier mobility in the active layer, we measure $J$–$V$ characteristics of single charge carrier devices and then fit the results using the space-charge-limited current (SCLC) model (Figure 3) and the Mott–Gurney law that includes field-dependent mobility, given by $J = \frac{2e\varepsilon_0\mu_r(V-V_{bi})^2}{L^2} \exp\left(\frac{\beta(V-V_{bi})}{L}\right)$, where $\varepsilon_0$, $\mu_r$ is the dielectric permittivity of the active layer, $L$ is the thickness of the active layer, $\mu_0$ the zero-field mobility, and $\beta$ the field activation factor.
is used (Figure S2 and Table S1, Supporting Information). At a typical electric field of $10^5 \text{ V cm}^{-1}$ (corresponding to an applied voltage of 1 V across the bulk of a 100 nm device), apparent hole mobilities of $1.7 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $3.2 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and apparent electron mobilities of $5.8 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $3.3 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ have been determined for the devices with and without the PFN interlayer, respectively. In other words, upon the incorporation of the PFN interlayer, both the hole and electron mobilities increase and a more balanced charge transport in the devices can be achieved. While the microscopic origin for the enhanced mobility remains uncertain at this point, we speculate that the increased $V_{bi}$ across the device and the strong local field at the active layer/cathode interface due to the PFN interlayer may facilitate charge carriers to escape shallow traps and thus improve their mobility (Figure 2f).

The PFN interlayer is also found to prevent the build-up of space charge under device working conditions. Figure 4a,b show the experimental power law dependence of photocurrent on incident light intensity. At a high effective voltage, $V_{eff}$ of 2.1 V, where $V_{eff} = V_0 - V$, and $V_0$ is the compensation voltage at which net photocurrent $J_{ph} = 0$ and $V$ is the applied voltage; $V_{eff} = 2.1 \text{ V}$ corresponds to negative biases of $-1.4 \text{ V}$ and $-1.2 \text{ V}$ for devices with and without the PFN interlayer, respectively. The power exponents are both 0.90, much higher than the 0.75 exponent for the space-charge-limited scenario,$^{37,38}$ indicating no build-up of net space charges in either devices even at the highest illumination intensity; this is due to the high electric field as well as high and balanced charge carrier mobilities, as confirmed by the SCLC measurements. However, at a low effective voltage of 0.1 V, which corresponds to an external bias of 0.6 V for devices without the PFN interlayer, the power exponent decreases to 0.84 (Figure 4b) for the device without the PFN interlayer, implying a buildup of space charge and the subsequent bimolecular recombination.$^{38}$ In contrast, the power exponent for devices with the PN interlayer remains 0.90 at $V_{eff} = 0.1 \text{ V}$, corresponding to an external bias of 0.8 V for devices with the PFN interlayer (Figure 4a) and indicating no space charge build-up. The results clearly identify that the PFN interlayer effectively prevents space charge build-up even under low effective voltages, at which maximal power output condition of PSCs usually appears.

The incorporation of the PFN interlayer also improves the charge collection efficiency under working conditions. Figure 4c shows the $J-V$ characteristics in a wide reverse bias range under AM 1.5G illumination. The results are plotted as the net photocurrent ($J_{ph} = J_L - J_D$) dependence on the effective applied voltage $V_{eff}$, where $J_L$ and $J_D$ are the current density under illumination and in the dark, respectively, $V$ is the applied voltage, and $V_0$ is the compensation voltage at which $J_{ph} = 0$. At a large reverse voltage (i.e., $V_{eff} = 2 \text{ V}$), $J_{ph}$ reaches saturation for both devices, suggesting all photogenerated excitons are dissociated into free carriers and all carriers are collected at the electrodes without any bimolecular recombination.$^{11,14,19}$ In this case saturation current density $J_{sat}$ is only limited by the amount of absorbed incident photon flux ($\phi_{\text{photon}}$), thus the maximal obtainable exciton generation rates ($\phi_{\text{max}}$) given by $J_{sat} = \epsilon \phi_{\text{max}} L$ where $L$ is the thickness of the active layer) are essentially the same ($9.4 \times 10^{-7} \text{ m}^{-2} \text{ s}^{-1}$) in both devices under AM 1.5G illumination. At the slightly lower effective voltage range ($0.5 \text{ V} < V_{eff} < 2 \text{ V}$), $J_{ph} - V_{eff}$ characteristics for both devices approach saturation and overlap with each other. For example, at the short current density conditions, the photocurrent densities $J_{ph}$ are 94% and 90% of the saturation current density $J_{sat}$ ($J_{ph}$ at $V_{eff} = 2 \text{ V}$) for devices with and without the interlayer, respectively. Interestingly, in the low effective voltage range ($V_{eff} < 0.5 \text{ V}$), $J_{ph} - V_{eff}$ characteristics for the two devices show large differences. For example, at the maximal power output condition (corresponding to $V_{eff} = 0.2 \text{ V}$ for both devices), $J_{ph}/J_{sat}$ is 74% for the device with the PFN interlayer while it
is only 61% for the control device without the PFN interlayer. Since the ratio $J_{ph}/J_{sat}$ is essentially the product of exciton dissociation efficiency and charge collection efficiency,[40] a decreased $J_{ph}/J_{sat}$ suggests either a reduced exciton dissociation efficiency or charge collection efficiency, with the latter suggesting that bimolecular recombination begins to dominate and usually lead to a lower $FF$.[39] The recombination in devices is manifested in the deviation of the photocurrent from the square-root dependence on effective voltage, which is one of the signatures of recombination-limited photocurrent in PSCs.[37] The superior $J_{ph} - V_{eff}$ characteristics from devices with interlayer clearly identifies the effect of interlayer on reducing bimolecular recombination at the low effective voltage, at which maximal power output condition of PSCs usually appears.

Such reduced bimolecular recombination in devices with an interlayer can be attributed to the observed higher charge mobility therein. Since charge transport is described by drift-diffusion equations and diffusion-controlled photocurrent is dominant in the low effective voltage regime,[40] the diffusion coefficients of the hole and electron, which are associated with mobility and given by the Einstein relation $D = k_B T e$ where $D$ is the diffusion coefficient, $k_B$ is the Boltzmann's constant, $T$ is the temperature, $e$ is the electron charge, and $\mu$ is the mobility) will increase in the device with interlayer. Therefore, the enhancement in charge diffusion and charge transport is responsible for the distinctly different $J_{ph}/J_{sat}$ when compared with the control device.

To further investigate the effect of the interlayer on photocurrent generation and collection and fully quantify the device enhancement, we applied two independent numerical models to adequately describe the characteristics of the devices. The first model is based on the geminate recombination theory of Onsager,[40, 41] including drift and diffusion of charge carriers and a field- and temperature-dependent photocurrent generation rate. The second model involves a field-independent charge pair generation rate and a charge-density-dependent recombination coefficient that can explain the dependence of the photocurrent on the bias in the operating regime, as proposed by Shuttle et al.[39] (see Supporting Information for details of the modeling). Using input parameters from Table S2 (Supporting Information) for charge mobility and most of the other remaining the same (Table S3, Supporting Information) for the devices with and without the PFN interlayer, the experimental $J - V$ characteristics for the actual solar cells under AM 1.5 G illumination and in dark are consistently described by the both models, as shown in Figure S3 (Supporting Information).

The lines in Figure 4c denote the simulation results made with the same set of input parameters, which are in good agreement with the experimental data. It is clear that higher mobilities in the devices with the interlayer are responsible for the enhanced $J_{ph}/J_{sat}$ in the low field regime. In a high field regime, both simulation and experimental data indicate that $J_{ph}/J_{sat}$ becomes field-independent and thus weakly dependent on charge carrier mobility, which can be employed to explain the nearly identical $J_{ph}/J_{sat}$ in the high field regime for both devices.

It is worth mentioning that this approach was also successfully applied in other material systems. Simultaneous enhancement in the $J_{SC}$, $V_{OC}$, and $FF$ was also demonstrated in PTB7:PC$_{71}$BM PSCs when the PFN interlayer was incorporated.

Figure 4. The effect of the PFN interlayer on PCDTBT:PC$_{71}$BM solar cell incident light intensity dependence. Double logarithmic plot of photocurrent density as a function of the incident light intensity for PCDTBT:PC$_{71}$BM solar cells with (a) and without (b) the PFN interlayer measured under effective voltage of 2.1 V (solid black squares) and 0.1 V (open circles). The lines represent the best power fitting. c) Photocurrent density versus effective voltage ($J_{ph} - V_{eff}$) characteristics of devices with (solid circles) and without the interlayer (open circles) under constant incident light intensity (AM 1.5G, 1000 W m$^{-2}$). Solid lines represent the simulation results.
We achieved a high PCE of 8.22% for devices by incorporating the PFN cathode interlayer.

After encapsulation, the devices were sent to the National Center of Supervision & Inspection on Solar Photovoltaic Products Quality of China (CPVT) for certification, and a certified PCE of 8.37% was obtained (Figure 5b and Figure S4, Supporting Information). Independent certification by Solarenergy Inc. yielded a slightly lower PCE of 7.65%, which can be attributed to lag time (18 d) between two the laboratories.

In conclusion, we have successfully demonstrated that simultaneous enhancement in the open-circuit voltage, short-circuit current density, and fill factor can be achieved in highly efficient polymer solar cells by simply incorporating a thin layer of alcohol/water-soluble polymer as the cathode interlayer, resulting in a power conversion efficiency up to 6.79% and a certified 8.37% for PCDTBT and PTB7 devices, respectively. The effects of the interlayer on the improvement of device performance is shown to be threefold: an enhanced built-in potential across the device due to the existence of interface dipole, improved charge-transport properties, elimination of the buildup of space charge, and reduced recombination loss due to the increase in built-in field and charge carrier mobility. Further optimization is conceivable because the interfacial dipole of thicker interlayer can result in as large as 0.5 V enhancement in built-in potential. The approach reported here provides a simple and versatile method to optimize polymer solar cells and may set the efficiency of devices towards the goal of 10% from various material systems.

**Experimental Section**

**Fabrication of Polymer Solar Cells:** PCDTBT and PTB7 were purchased from 1-material Chemscitech Inc. (St-Laurent, Quebec, Canada) and used as received. The device structure was ITO/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS)/polymer:PC 71 BM blend/Al. The PCDTBT:PCBM (1:4 by weight) active blend layer with a thickness of 90 nm was prepared by spin-coating the chlorobenzene solution at 1000 rpm for 2 min. The blend ratio of PTB7:PC 71 BM was 1:1.5 by weight and the active layer was spin-cast from mixed solvent of chlorobenzene/1,8-diiodoctane (97:3 vol%). The interlayer material was dissolved in highly polar solvents such as methanol under the presence of small amount of acetic acid and its solution was spin-coated on the top of the obtained active layer to form a thin interlayer of 5 nm. Determination of the thickness of the interlayer followed a previously published methods and thickness control was adjusted by the concentration of the solution and the spin speed between 600 rpm to 2500 rpm. The thickness of the interlayer was determined by a surface profiler (Alfa Step-500, Tencor), in combination with extrapolation from an absorbance–thickness curve that assumed a linear dependence of absorbance at 380 nm on film thickness. Al electrode (100 nm) was evaporated through a shadow mask to define the active area of the devices (2 × 8 mm²).

**Characterization and Measurement:** SKPM measurements were carried out on an Agilent 5300 atomic force microscope (Agilent technologies, USA) equipped with a homemade vacuum glove box. The samples were transferred into a glove box and the active layer/interlayer boundary was identified using an optical field monitor on the atomic force microscope and then imaged under nitrogen gas environment using the standard SKPM mode. Conducting atomic force microscopy (AFM) tips (NSC19/Ti-Pt, Mickomasch, San Jose, CA) used for this study had a typical spring constant of 0.68 N m⁻¹ and a resonance frequency of 80 kHz. Typical scan line frequency was 0.3 Hz and each image contained 512 × 512 pixels. The AC modulation for the Kelvin probe scan was 3 V in amplitude and 10 kHz in frequency. The resulted topographical images were plane fit with Picoimage software but the surface potential images were unprocessed original data.

The PCE was determined from J–V curve measurement (using a Keithley 2400 sourcemeter) under 1 sun, AM 1.5G (air mass 1.5 global)
spectrum from a solar simulator (Oriel model 91192) (100 mW cm\(^{-2}\)). Masks made from laser beam cutting technology with well-defined area size of 2.00, 3.14, or 10.0 mm\(^2\) were attached to define the effective area for accurate measurement. All of the masked and unmasked tests gave consistent results with relative errors within 3%. Solar simulator illumination intensity was determined by a monochromatic silicon reference cell (Hamamatsu S1333, with KG-5 visible color filter) calibrated by the National Renewable Energy Laboratory (NREL). The theoretical \(j_{sc}\) obtained by integrating the product of the EQE with the AM 1.5G solar spectrum perfectly agreed with the measured value to within 1%. The spectral mismatch factors (\(M\)) were calculated according to standard procedure\([12]\) and \(M\) values of 0.977 and 1.011 were used to obtain correct photocurrent and efficiency for PCDTBT and PTB7 devices, respectively.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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