Polymer self-organization enhances photovoltaic efficiency

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By growing films of light-absorbing polymers slowly, self-organization—which improves the efficiency of polymer solar cells—is promoted.

Polymer, or plastic, solar cells have shown potential for harvesting solar energy in a cost-effective way. Significant efforts are under way to improve their efficiency to the level of practical application. By exploring a new slow-growth approach to enhance polymer self-organization, the efficiency of polymer solar cells has been significantly improved: to 4.4% under the standard AM1.5G (the standard spectrum of sunlight at the Earth’s surface) test condition.

Converting solar energy into electricity may provide a much-needed solution to the energy crisis the world is facing today. Using photovoltaic (PV) modules with 10% efficiency, the total U.S. energy demand could be met by a 10,000 mi² PV array in Nevada. But inorganic solar cells have not gained wide acceptance as an electricity source because of the high cost of manufacturing crystalline silicon. Plastic solar cells, on the other hand, can provide low cost fabrication, easy processing, and mechanical flexibility. As such, they have evolved as the most promising cost-effective alternatives to silicon-based solar cells.

Currently, low efficiency is their major limiting factor. High efficiency requires efficient light absorption and transport of charge carriers—electrons and holes. Polymers have much lower carrier mobility than inorganic semiconductors, which makes it difficult to achieve these two properties simultaneously.

Many approaches to enhancing polymer solar-cell efficiency have been tried, including morphology improvement and inserting an interfacial layer. Regioregular-Poly(3-hexylthiophen) (RR-P3HT) is a promising polymer for polymer solar cells: it has high ordering-induced mobility as well as an improved solar-spectrum absorption up to 650nm. When blended with methanofullerene (PCBM) in a bulk-heterojunction structure, however, severe absorption quenching was observed, indicating the destruction of ordering. Several groups have investigated the partial recovery of polymer ordering by thermal annealing, and device efficiency has improved to more than 3%. Inadequate absorption and incomplete ordering recovery still limit solar cell performance, however.

To push this technology forward, we have developed a new approach: reducing the film growth rate to maximize ordering by polymer self-organization. In the pure polymer phase, the highly regular chain structure of P3HT facilitates its self-organization into two-dimensional sheets by means of interchain stacking. Self-organization has been shown to improve field-effect carrier mobility in RR-P3HT by more than a factor of 100, to 0.1 cm²/Vs. We chose dichlorobenzene (DCB) as the solvent to facilitate slow film growth: it provides appropriate conditions for RR-P3HT to form a self-organized, ordered structure in the RR-P3HT:PCBM 1:1 weight-ratio blend system. The de-
The degree of self-organization can be controlled by changing the film growth rate.

Figure 1 shows the effect of film growth rate and thermal annealing on the absorbance of the blend films. The slowly grown film took ~20min to dry while the fast one took ~20s. The thickness was 210–230nm for both. The absorption in the red region of the slowly grown film was much stronger, and the vibronic absorption shoulders (peaks) much more pronounced, indicating a higher degree of ordering. After annealing at 110°C for 20min, the absorbance of fast-grown film was significantly increased and the vibronic features were clearer, indicating a partial recovery of ordering. Annealing does not change the absorption of the slowly grown film, as a very-high degree of ordering is already present in slowly grown films.

To form solar cells, we use the conducting polymer PEDOT:PSS on indium tin oxide as the anode and Ca/Al as the cathode. The P3HT:PCBM polymer blend is sandwiched in between. As shown in Figure 2, the maximum external quantum efficiency (EQE) of the slowly grown device is ~63%, more than three times that of the fast grown device (~19%). Figure 3 shows J–V characteristics (AM1.5G standard testing condition) of four devices with different solvent evaporation times (t_{evp}=20min, 3min, 40s, and 20s). The short circuit current (J_{sc}) dropped from 9.9 to 4.5mA/cm², the device series resistance (R_{s}) increased from 2.4 to 19.8 Ω•cm² and the fill factor (FF) decreased from 60.3% to 52.0% with t_{evp} reducing monotonically. The overall effect is a reduction in power conversion efficiency (PCE) from 3.5% to 1.4%.

Further improvement in solar cell efficiency was obtained by thermal annealing at 110°C (see Figure 4). Upon annealing, J_{sc} increased slightly from 9.9 to 10.6mA/cm², and FF increases from 60.3% to 67.4%, which is among the highest fill factors in organic solar cells. As a result, the PCE went up from 3.5% to 4.4%.

Figure 2. The film growth rate affects the EQE of polymer solar cells. The efficiency maximum for the slowly grown device is ~63%, which is more than three times that of the fast grown one (~19%).

Figure 3. The film growth rate affects the J–V characteristics under AM1.5G one-sun illumination of the plastic solar cells. Film growth times are: 20min (black), 3min (red), 40s (green), and 20s (blue).

Figure 4. Annealing the slowly grown film barely affects the polymer solar-cell performance. The annealing temperature is 110°C, and the annealing time varies from 0 to 30min.

Continued on next page
after 10 min annealing. These devices show excellent diode characteristics under the dark, with rectification ratios close to $10^7$ at a bias of 2 V. The thickness of the active layer eliminates pinholes and microcracks, and all devices show a very high shunt resistance of $180-640 \times 10^6 \, \Omega$, the highest reported for organic solar cells. This makes these cells ideal for photodetector applications. Annealing the slowly grown films is believed to help remove solvent residue and to improve the interface with electrode. Higher mobilities and balanced carrier transport are also detected by time-of-flight measurements in the slowly grown device: vital for high $J_{sc}$ and good FF.

In summary, efficiency enhancement is very important for the future of polymer solar cells as a cost-effective method of solar energy harvesting. The slow growth approach to enhancing polymer self-organization provides a promising tool. Our results show that this approach can achieve efficient absorption and carrier transport at the same time for high efficiency. Our next step will be to extend this approach into other materials systems as well as to fabricate large area and flexible solar cells to demonstrate the full advantage of using polymer-based devices.

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