Highly efficient carbon-nanotube and organic-polymer hybrid photovoltaics

Zhaohui Zhong and Nanditha M. Dissanayake

Discovery of extremely efficient photocurrent generation through hole transfer at individual heterojunctions could provide a breakthrough for development of high-efficiency hybrid photovoltaics.

Semiconducting organic polymers are excellent light absorbers, making them ideal for fabricating low-cost and environmentally friendly thin-film photovoltaics. However, despite tremendous developments, the power-conversion efficiency of organic photovoltaics is still well below those of other competing thin-film photovoltaic technologies such as copper indium selenide, copper monosulfide, and amorphous silicon. A major limitation is the inherently poor charge-transport behavior in organic polymers. Single-walled carbon nanotubes (SWNTs), which are quasi-1D, low-loss conductors, can be integrated with organic polymers to boost their charge transport and, consequently, their overall performance. Fabrication of such hybrid photovoltaics requires detailed understanding of the energy levels at the heterojunction between organic polymers and SWNTs.

Photoinduced charge separation at a typical polymer-SWNT junction is believed to be fundamentally governed by the alignment of the highest-occupied and the lowest-unoccupied molecular orbitals (HOMO, LUMO) of the constituents. Accordingly, heterojunctions between the widely used poly(3-hexylthiophene-2,5-diyl) (P3HT) polymer and SWNTs are presumed to function such that semiconducting (s-) SWNTs behave as efficient electron acceptors, while metallic (m-) SWNTs are thought to impede charge transfer by creating recombination centers. However, experimental investigation of charge transfer at individual SWNT/P3HT junctions has not been carried out to validate these claims. Furthermore, ensemble SWNT/P3HT photovoltaics fabricated based on these assumptions have thus far resulted in poor device performance. Accurate understanding of the nanoscale charge-transfer behavior at SWNT/P3HT junctions is needed to design novel device architectures that can yield greater power-conversion efficiencies. Therefore, we carried out a comprehensive study of photoinduced charge transfer at individual SWNT-P3HT junctions.

We grew individual SWNTs using chemical-vapor deposition on quartz substrates, which we electrically contacted using photolithography-patterned source and drain electrodes. A layer of P3HT (65nm) was spin coated onto each SWNT, forming nanoscale P3HT-SWNT junctions, while an indium tin oxide layer (200nm) was deposited onto the P3HT as a transparent top electrode: see Figure 1(a). We carried out transport measurements to separately identify each s- and m-SWNT/P3HT junction. Subsequently, we illuminated each heterojunction using a focused laser beam and measured the resulting photocurrent.

Figure 1. (a) Single-junction, single-walled carbon-nanotube (SWNT)/poly(3-hexylthiophene-2,5-diyl) (P3HT) hybrid photovoltaic. Excitons dissociate at the heterojunction, followed by hole (h) transfer into the nanotube and electron (e) collection by the indium tin oxide (ITO) electrode. Al: Aluminum. Pd: Palladium. SiO\textsubscript{2}: Silicon dioxide. (b) Short-circuit photocurrent versus gate voltage (V\textsubscript{g}) of individual semiconductor- (s-) SWNT/P3HT (black) and metallic- (m-) SWNT/P3HT (red) devices. The current is collected from the Pd source electrode while keeping ITO at zero bias and floating the Pd drain electrode. Positive (negative) photocurrent indicates hole (electron) transfer from P3HT to the SWNTs.

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Figure 2. Estimated effective external quantum efficiency (EQE) of our large-area P3HT-SWNT bilayer photovoltaic device. λ: Wavelength. (inset) Effective volume of a single P3HT-SWNT junction considering the exciton-diffusion length.

In summary, we discovered a high-efficiency photoinduced hole-transfer mechanism from P3HT to SWNTs, irrespective of the SWNT nature. This charge-transfer process is driven by an electric field formed at the nanoscale junction as a result of differences in the Fermi levels of P3HT and SWNTs. Our results suggest that the interfaces between nanotube and polymer are much more complex than previously assumed. Importantly, these findings also make selective omission of m-SWNTs redundant, promising carbon-nanomaterial-based, low-cost, high-efficiency hybrid photovoltaics. We aim at further developing this technology.

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Author Information
Zhaohui Zhong and Nanditha M. Dissanayake
Department of Electrical Engineering and Computer Science
University of Michigan
Ann Arbor, MI

Zhaohui Zhong received his PhD in chemistry in 2005 from Harvard University. He is an assistant professor of electrical engineering.

Nanditha Dissanayake received his PhD in electrical engineering in 2008 from the University of Surrey. He is a postdoctoral researcher.

References

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