Methylammonium lead iodide perovskite/fullerene-based hybrid solar cells

Jun-Yuan Jeng, Yi-Fang Chiang, Mu-Huan Lee, Shin-Rung Peng, Tzung-Fang Guo, Peter Chen, and Ten-Chin Wen

A novel design simplifies fabrication of all-solid-state, ultrathin organic/inorganic photovoltaics with respectable efficiency.

Turning solar energy into a clean, practical alternative power source will require new types of devices that generate electrical power directly from solar irradiation with high efficiency and at low cost. Recent studies report solar cells based on materials known as organometal halide perovskites that have promising photovoltaic efficiencies. These devices employ a submicron, crystalline, and well-oriented thin film of organolead iodide perovskite as the light absorber (or as both light absorber and carrier conductor) deposited on the surface of meso (i.e., 1–1000nm)-structured titanium dioxide (TiO₂) or aluminum oxide layers through a simple spin-coating process.

The function of methylammonium lead iodide (CH₃NH₃PbI₃) perovskite is akin to that of the ‘electron donor’ material in donor-acceptor polymer/organic planar- and bulk-heterojunction solar cells (i.e., PHJs and BHJs, two typical configurations of these types of cells). Accordingly, we propose depositing or growing a thin layer of acceptor material on CH₃NH₃PbI₃ perovskite film to create a donor-acceptor contact interface for charge separation, in which the hybrid CH₃NH₃PbI₃ perovskite/acceptor PHJ yields the photovoltaic effect under irradiation.

Several studies have reported that a thin layer of crystalline CH₃NH₃PbI₃ perovskite is easily prepared by spin-casting a solution of equimolar CH₃NH₃I and PbI₂ (precursor solution) on the substrate. However, the process of crystal growth can coarsen the film and markedly degrade device performance or result in device failure. To ensure rapid solvent evaporation and, therefore, inhibit crystal coarsening, we preheated the substrate at an elevated temperature of 60°C for 5min and cast the precursor solution at a high spinning speed of 6000rpm.

Figure 1 shows the CH₃NH₃PbI₃ perovskite solution and spin-coating process we use to prepare the film on an indium tin oxide (ITO)/glass substrate. The film is post-annealed at 100°C for 15min to a thickness of approximately 20–30nm. Figure 2 shows the UV-visible spectrum and a photograph of the film. Its absorption covers a wide range of light from the visible to

Continued on next page
Figure 3. Diagram showing (left) the device configuration and (right) energy levels of each layer in the device. Al: Aluminum. BCP: Bathocuproine. C₆₀: Fullerene. PEDOT:PSS: Conducting polymer.

Figure 4. The current density-voltage (J-V) curves for devices of (a) γ-butyrolactone film and (b) DMF film. PCBM: [6,6]-Phenyl C₆₁-butyric acid methyl ester. ICBA: Indene-C₆₀ bisadduct.

the near-IR region, indicating the formation of CH₃NH₃PbI₃ perovskite on the substrate.

Depositing a thin acceptor layer, such as fullerene (C₆₀) or a C₆₀ derivative, on top of the film creates a donor-acceptor interface for charge separation. A thin bathocuproine (BCP) film is an exciton (coupled electron-hole pair)- or hole-blocking layer that, combined with an aluminum (Al) layer, acts as the negative electrode. Figure 3 shows the device configuration (glass/ITO/PEDOT:PSS (a conducting polymer)/CH₃NH₃PbI₃ perovskite/C₆₀ [30nm]/BCP [10nm]/Al) and the energy levels of each layer in the hybrid CH₃NH₃PbI₃ perovskite/C₆₀ PHJ solar cell.

The device exhibits open-circuit voltage (V_OC) = 0.55V, short-circuit current (J_SC) = 5.21mA/cm², and fill factor (FF) = 0.57, corresponding to a power conversion efficiency (PCE) of 1.6% under a standard one sun of simulated solar irradiation (AM1.5). Applying higher LUMO (lowest unoccupied molecular orbital) energy level [6,6]-phenyl C₆₁-butyric acid methyl ester (PCBM) or indene-C₆₀ bisadduct (ICBA) instead of C₆₀ as the acceptor elevates the magnitude of V_OC and PCE to 0.65 and 0.75V, 2.4 and 2.1%, respectively. Figure 4 presents the current density-voltage (J-V) curves of the devices. These results verify the formation of a donor-acceptor interface at the CH₃NH₃PbI₃ perovskite/C₆₀ PHJ due to the modulation of photovoltaic performance by acceptors of varied LUMO levels. The glass/ITO/PEDOT:PSS/CH₃NH₃PbI₃ perovskite (from DMF solution)/C₆₀ (30nm)/BCP (10nm)/Al device configuration delivers V_OC = 0.55V, J_SC = 9.02mA/cm², and FF = 0.61, corresponding to a PCE of 3.0%. As shown in Figure 4(b), the CH₃NH₃PbI₃ perovskite (DMF)/ICBA PHJ solar cell has a V_OC = 0.58V, J_SC = 10.03mA/cm², and FF = 0.58, corresponding to a PCE of 3.4%. The CH₃NH₃PbI₃ perovskite (DMF)/PCBM (25nm) PHJ device exhibits a V_OC = 0.60V, J_SC = 10.30mA/cm², and FF = 0.63, corresponding to an optimized PCE of 3.9%.

Continued on next page
Figure 5. High-resolution scanning electron micrographs of CH$_3$NH$_3$PbI$_3$ perovskite film from (a) γ-butyrolactone solution and (b) DMF solution.

Figure 5 shows high-resolution scanning electron micrographs of CH$_3$NH$_3$PbI$_3$ perovskite film spin-cast on a glass/ITO/PEDOT:PSS substrate. The crystal sizes in the cluster-domain regions of the perovskite are around 100–150nm in diameter—see Figure 5(a)—and around 150–200nm in Figure 5(b). Note that the films shown in Figure 5 are not morphologically ideal, yet the film’s topography as seen by the naked eye is still smooth and reflective: see Figure 2(b). Film prepared using DMF solution has a relatively higher contact area in PHJ and channels formed by interconnected crystalline strips that are suitable for transporting charge carriers.

In conclusion, we have reported a device made of CH$_3$NH$_3$PbI$_3$ perovskite film spin-cast from DMF solution with a respectable PCE of 3.9%. Applying acceptors of varied LUMO energy levels modulates the magnitude of $V_{OC}$ and PCE. We believe this progress represents a new concept for the design of ultrathin excitonic photovoltaic devices. Our approach greatly simplifies fabrication of all-solid-state, hybrid CH$_3$NH$_3$PbI$_3$ perovskite-based solar cells that perform acceptably without having to resort to TiO$_2$ nanosheets or meso-mesoporous metal oxide layers and additional hole-transport layers. This alternative might prevent the pore-filling or reproducibility issues of mesoscopic junction devices.

Our current efforts are focused on fabricating CH$_3$NH$_3$PbI$_3$ perovskite/C$_60$ or C$_{60}$-derivative PHJ solar cells at relatively low temperatures on ITO-patterned plastic substrates, for potential use in flexible, lightweight, portable devices. Adjusting the solvent, solution concentration, preheating temperature, or synthesizing new perovskite-based materials will make it possible to optimize the thickness, morphology, and topography of an ideal CH$_3$NH$_3$PbI$_3$ perovskite film to further enhance photovoltaic performance for real-world applications.

The authors would like to thank the National Science Council of Taiwan (NSC99-2113-M-006-008-MY3) for financially supporting this research.

Author Information

Jun-Yuan Jeng, Yi-Fang Chiang, Mu-Huan Lee, Shin-Rung Peng, Tzung-Fang Guo, and Peter Chen
Department of Photonics
Advanced Optoelectronic Technology Center
National Cheng Kung University
Tainan, Taiwan

Jun-Yuan Jeng received a BS in applied physics (2006) from the National University of Kaohsiung and an MS in photonics (2008) from the Department of Photonics at the National Cheng Kung University, where he is currently a PhD candidate in the Department of Photonics. His research interests include organic photovoltaic and organic electronic devices.

Yi-Fang Chiang received a BS from the Department of Earth Sciences (2005) at the National Cheng Kung University and an MS from the Department of Materials Science (2007) at the National University of Tainan. She is currently a PhD candidate at the National Cheng Kung University. Her research interests include organic photovoltaic and dye-sensitized solar cells.

Mu-Huan Lee received a double BS from the Departments of Photonics and Earth Sciences (2011) at the National Cheng Kung University, where he is currently working toward his MS. His research interests include organic photovoltaics.

Continued on next page
Shin-Rung Peng received his BS and MS in mechanical engineering (2001 and 2002, respectively) and his PhD degree in mechanical engineering (2009) all from the National Chung Cheng University. His research interests include contact mechanics, nanomechanics, molecular simulations, and organic photovoltaics.

Tzung-Fang Guo received a BS in chemistry (1993) from Soochow University, Taiwan, and an MS in chemistry (1995) from the National Chung Cheng University, Taiwan, and a PhD in materials science and engineering (2002) from the University of California, Los Angeles. In 2003, he joined the Department of Photonics, the National Cheng Kung University. His research focuses on high-performance organic and polymer LEDs, n-type pentacene organic thin-film transistors, polymer/organic solar cells, and the magneto response of organic electronics.

Peter Chen received his BS and MS (1998 and 2000, respectively) from the National Cheng Kung University. In 2004, he entered the Photonics Program at the Swiss Federal Institute of Technology, Lausanne, where he received his PhD (2009). In 2010, he joined the Department of Photonics at the National Cheng Kung University. His research interests include photovoltaic devices (dye-sensitized and other nanostructured solar cells), novel materials for solar cells, and characterization methods.

Ten-Chin Wen
Department of Chemical Engineering
National Cheng Kung University
Tainan, Taiwan

Ten-Chin Wen received his BSc in chemical engineering from the National Cheng Kung University (1977), his MSc in chemical engineering from the National Taiwan University (1979), and a PhD in chemical engineering from Lamar University, Texas (1986). His research focuses on lithium batteries, polymer electrolytes, conducting polymers, photoelectric materials, and novel electrode materials.

References