



All organic-based solar cell and thermoelectric generator hybrid device system using highly conductive PEDOT:PSS film as organic thermoelectric generator



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ABSTRACT

We report the fabrication of an all organic-based solar cell and thermoelectric generator hybrid device using an organic solar cell (OSC) and a single highly conductive PEDOT:PSS film as the solar cell and thermoelectric generator (TEG), respectively. The Seebeck coefficient of the single PEDOT:PSS film was measured as 19.8 $\mu\text{V/K}$. When the two devices were hybridized, hybridization loss (i.e., reduction in FF) caused by the internal resistance of the PEDOT:PSS film used as the organic thermoelectric generator (OTEG) decreased, because of the decreased resistance of the PEDOT:PSS film. The hybridization loss of FF was neglected when the resistance of the PEDOT:PSS film was below 1.36 Ω . The PEDOT:PSS films as OTEG were fabricated by drop-casting of the PEDOT:PSS solution on glass. The temperature difference in the PEDOT:PSS film was 5 $^{\circ}\text{C}$ in the hybrid device. As the resistance of the PEDOT:PSS film was 1.36 Ω , after hybridization, the PCE of the hybrid device was enhanced by the increased open circuit voltage (V_{oc}) generated from the PEDOT:PSS film. The result shows that the PEDOT:PSS film can be applied as an OTEG material for all organic-based solar cell and thermoelectric generator hybrid devices.

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1. Introduction

Over the past decades, significant development has been made to photovoltaic (PV) devices as next-generation renewable energy resources for solar energy conversion. The key to improve the power conversion efficiency (PCE) of a PV device is the utilization of irradiated solar light with a broad wavelength range. To this end, effort has been devoted toward the development of new active materials and device structure design (Chen et al., 2014a, b; Dou et al., 2012; Li, 2012; Scharber et al., 2006). However, most PV devices absorb only a part of the visible and infrared light in the full sunlight wavelength. Thus, long-wavelength infrared light cannot be efficiently used. In most PV devices, unabsorbed long-wavelength infrared light with low energy photons, as well as

some UV light with photons having energy higher than the band-gap energy of the active layer, are spontaneously transformed into heat. This waste heat energy corresponds to $\sim 40\%$ of the solar spectral irradiance (Nelson, 2003). Therefore, it is necessary to recycle the waste heat generated from the solar-facing PV device. A thermoelectric generator (TEG) can convert heat directly into electric energy via the Seebeck effect; hence, a PV–TE hybrid device not only allows for the utilization of a broader spectrum of the irradiated solar light, but also prevents overheating that decreases the efficiency and stability of the PV device during operation (Royne et al., 2005).

Several groups have reported PV–TE hybrid device systems based on various PV devices such as Si solar cells, dye-sensitized solar cells (DSSCs), and polymer solar cells (PSCs) (Chang and Yu, 2012; Deng et al., 2013; Park et al., 2013; Wang et al., 2011; Zhang et al., 2013). However, there is hardly any report on all organic-based PV–TE hybrid device systems using an organic-based TEG, because PV devices have mostly been hybridized with a commercialized TE module based on inorganic materials. A part of TE as well as PV device should be also needed to be organized for PV–TE hybrid device to have advantages of organic electronic device with low-cost, lightness, flexibility and mass and easy

Abbreviations: OSC, organic solar cell; TEG, thermoelectric generator; OTEG, organic thermoelectric generator; PV, photovoltaic; PCE, power conversion efficiency; DSSC, dye-sensitized solar cell; PSC, polymer solar cell; PEDOT:PSS, poly (3,4-ethylenedioxythiophene) polystyrene sulfonate; DMSO, dimethyl sulfoxide.

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fabrication. Thus, all organic-based PV–TE hybrid systems can minimize manufacturing cost and complexity in production. A PV–TE hybrid device based on organic materials, with a doped P3HT film as the organic TEG (OTEG), has been reported, but its PCE is very low, $2.1 \times 10^{-2}\%$ (Suzuki et al., 2010). In a PV–TE hybrid device system, the internal resistance of the TEG is a very important factor affecting hybridization loss because the PV and TE components are electrically connected in series. The hybridization loss due to the high internal resistance of the TEG decreases the FF of the hybrid device, which in turn results in a reduction in the PCE. Therefore, it is essential to select an organic thermoelectric material with low resistance as well as good thermoelectric characteristics as the OTEG. Among the various organic thermoelectric materials based on conductive polymers, poly(3,4-ethylenedioxy thiophene) polystyrene sulfonate (PEDOT:PSS) exhibits the highest conductivity with promising thermoelectric properties (Lee et al., 2014; Park et al., 2014; Yue and Xu, 2012). Hence, PEDOT:PSS was selected as the most suitable OTEG candidate for the all organic-based PV–TE hybrid device system in our research.

In this study, we focus on the effect of the resistance of a single PEDOT:PSS film on hybridization loss of the hybrid device, at various film thicknesses, to verify the applicability of PEDOT:PSS films as OTEG material for all organic-based PV–TE hybrid systems. The resistance of the PEDOT:PSS films decreased from 58.73 to 1.36 Ω with an increase in the film thickness from 0.19 to 12.30 μm . The single PEDOT:PSS film with a resistance of 1.36 Ω did not induce any hybridization loss that would lead to FF reduction. The results show that the PEDOT:PSS film is applicable as an OTEG material for all organic-based PV–TE hybrid devices.

2. Experimental

2.1. Materials

PEDOT:PSS solution (Clevios PH 1000) was purchased from Heraeus Ltd. Dimethyl sulfoxide (DMSO, 99%) was purchased from Samchun Pure Chemicals (S. Korea). All materials were used without further purification.

2.2. Fabrication of the hybrid device

First, the OSCs were fabricated by a previously reported procedure (Lee et al., 2015). Glass coated with ITO (150 nm thick, 10 $\Omega/\square$) was cleaned by sequential ultrasonic treatment in acetone and isopropanol. The cleaned ITO glass was dried at 100 $^{\circ}\text{C}$ under vacuum for 1 h and then treated using an ultraviolet–ozone cleaner for 15 min. PEDOT/PSS solutions (Clevios PH) mixed with PEGME in a PSS/PEGME weight ratio of 1:0.5 were spin-coated onto the cleaned ITO glass at 2000 rpm for 40 s. The PEDOT:PSS-coated ITO glass samples were then thermally annealed at 150 $^{\circ}\text{C}$ for 30 min in air and transferred to a nitrogen-filled glove box. P3HT (Rieke Metals, 4002-EE) and C61-butyric acid methyl ester (PCBM) (Nano-C, 99%) in a weight ratio of 1:0.6 were dissolved in chlorobenzene. The solution containing P3HT and PCBM was stirred for 1 d at 50 $^{\circ}\text{C}$, filtered through a 0.2 μm PTFE syringe filter, and spin-coated onto the PEDOT:PSS-coated ITO glass at 600 rpm for 40 s. The thin films were then thermally annealed on a hot plate at 120 $^{\circ}\text{C}$ for 10 min. To complete the structure of the device, top contact was formed through sequential thermal evaporation of lithium fluoride (0.6 nm) and aluminum (100 nm) through a shadow mask under vacuum (pressure: 2×10^{-6} Torr). Finally, the fabricated OSCs were encapsulated in a nitrogen-filled glove box using UV epoxy resin and covered glass.

Second, highly conductive PEDOT:PSS films were prepared to be used as the organic TEG. PEDOT:PSS solution (Clevios PH 1000)

were filtered through a 5.0 μm nylon syringe filter, spin-coated twice at 1500 rpm for 60 s onto glass, and thermally annealed on a hot plate at 150 $^{\circ}\text{C}$ for 5 min in air (thickness = 190 nm). To prepare PEDOT:PSS films with thickness of a few to several tens of micrometers, 50, 100, and 200 μL PEDOT:PSS solutions were drop-casted onto glass and annealed on a hot plate at 80 $^{\circ}\text{C}$ for 30 min in air. All the PEDOT:PSS films had dimensions of 2 cm $\times$ 1 cm.

Finally, the OSC–OTEG hybrid device was fabricated by attaching one-half of the PEDOT:PSS film, which acts as the OTEG, onto the backside of the OSC using scotch tape, so that the other half of the films protruded from the OSC.

2.3. Characterization and measurements

The resistance of the PEDOT:PSS films was measured by using a two-point probe meter with a Fluke 289 RMS multimeter after deposition of silver paste on the PEDOT:PSS films to enable Ohmic contact. The distance between two points of silver paste deposition was 6 mm. The thicknesses of the PEDOT:PSS films were measured by a surface profiler (Alpha step IQ, KLA-Tencor). Current/voltage curves were recorded by an electrochemical workstation (Keithley Model 2400) and a solar simulator (1000 W xenon lamp, Oriel, 91193), which provided a simulated AM 1.5 spectrum (100 mW/cm²). The simulated light was calibrated with a Si solar cell (Fraunhofer Institute for Solar Energy Systems, Mono-Si + KG filter, Certificate No. C-ISE269) to a sunlight intensity of 1 (100 mW/cm²). The temperature at the important points of the OSC–OTEG hybrid device was measured using a Fluke 289 RMS multimeter. The Seebeck coefficient of the PEDOT:PSS films with silver paste was measured by using a home-built setup consisting of two Peltier devices to maintain a controlled temperature gradient. The Peltier devices were controlled by using a Keithley 2400 source-measure unit and a Keithley 2200-30-5 power supply. Two thermocouples were used to measure the temperature gradient across the PEDOT:PSS films.

3. Results and discussion

3.1. OSC–OTEG hybrid device system

Fig. 1 illustrates the OSC–OTEG hybrid device system using highly conductive PEDOT:PSS films as an OTEG. The temperature gradient of the commercialized TE module based on inorganic materials used as the TEG has been formed in the upper and lower direction vertically in most previously reported studies of the PV–TE hybrid device (Chang and Yu, 2012; Deng et al., 2013; Park et al., 2013; Wang et al., 2011; Zhang et al., 2013). One of the many advantages of organic materials compared to inorganic materials is easy and large-area film coating by using a solution process. Hence, as shown in Fig. 1(a), the OSC was hybridized with the prepared PEDOT:PSS film in mainly the horizontal direction as the OTEG. This is because using the common coating method in the horizontal direction with spin, bar, and spray coating is much easier to fabricate large-area films than the coating method in the vertical direction. The range of the thickness of the prepared PEDOT:PSS film is from a few hundred nanometers to several tens of micrometers. Thus, as shown in Fig. 1(a), it is almost impossible for the temperature gradient inside the PEDOT:PSS film to be formed in the vertical direction film because the thickness of the prepared PEDOT:PSS film is not thick enough. However, as shown in Fig. 1(b), covering a part of the PEDOT:PSS films out of area of OSC by sunshade can generate a temperature gradient in the PEDOT:PSS film. Thus, the output voltage of the PEDOT:PSS film (V_{TE}) as the OTEG can be obtained using the following equation

$$V_{\text{TE}} = S(T_{\text{TEG, hot side}} - T_{\text{TEG, cold side}}) = \Delta T \quad (1)$$

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