

Polymer thermoelectric devices prepared by thermal lamination



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ABSTRACT

The thermoelectric properties of organic materials have been greatly improved very recently, but studies of organic thermoelectric device fabrication are still very limited. In this work, we report proof-of-concept studies of organic thermoelectric devices fabricated by thermal lamination at 100 °C using the benchmark p-type material poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate). A 1.54 cm² device shows a power output of 37 μW at a temperature difference of 50 K (a power density of 24 μW/cm²), which is one of the highest values when compared with previous studies. This approach potentially enables the fabrication of high-performance, large-area organic thermoelectric devices via roll-to-roll processing at a low temperature.

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1. Introduction

There is a huge amount of waste heat and solar thermal energy available in the environment, and the energy is usually at temperatures lower than 150 °C [1]. To harvest thermal energy for practical applications, using thermoelectric devices which can directly convert heat energy into electricity is ideal because of the simple structure and lack of moving parts in these devices [2]. Current thermoelectric materials are based on inorganic semiconductors such as bismuth-telluride (BiTe) alloys, lead-telluride (PbTe) alloys, metal silicides, and metal oxides. Most of these materials exhibit a good performance at temperatures higher than 200 °C, and the Bi–Te alloy is the best commercially available thermoelectric material for low-temperature applications (from room temperature to 200 °C) [3]. Nevertheless, it is important to note that Te is a toxic and rare metal that is not suitable for large-area applications. Furthermore, inorganic thermoelectric devices are typically fabricated by a sintering process at a high temperature, for low-temperature application, and the energy conversion efficiency is not high; therefore, the energy payback for inorganic thermoelectric devices at a low temperature could be an important issue limiting their practical applications. Very recently, high thermoelectric performance with conducting polymers [4–16] and carbon-based materials [17–23] has been reported, which shows that the performance of organic thermoelectric materials is approaching that of their inorganic counterparts. These materials

are solution processable and should be ideal for large-area fabrication to harvest low-temperature thermal energy. Although material properties have been greatly improved, studies of organic thermoelectric device fabrication are still very limited, and fabrication requires further development with an eye toward practical applications [24–31].

Typical thermoelectric devices are called π -type devices and contain both p-leg block and n-leg block (Fig. 1a). This structure is not easy to directly apply in organic thermoelectric materials for several reasons. The first is the film thickness. To achieve a reliable temperature gradient in π -type devices, the film thickness is typically on the millimeter scale, which is difficult to achieve using solution processing. The second is the anisotropy of film samples. For a film sample, the in-plane Seebeck coefficient and electrical conductivity are easy to measure and compare, but extracting the through-plane quantities is not straightforward. In π -type devices, the through-plane thermoelectric properties of the materials largely determine device performance. Our previous study shows that in conducting polymers, the through-plane thermoelectric properties are lower than the in-plane properties [32–35]. The third reason is that the choice of high-performance n-type organic thermoelectric materials is still limited [36–39]. To utilize the advantage of solution-processed organic materials, one natural consideration is printing these materials as lines on a substrate and connecting these lines in parallel or in series to make a fin-type device (Fig. 1b). The thermal gradient is then applied parallel to the substrate. The largest advantage of this structure is its ease of production. Recently, we have reported the fabrication of polymer-based thermoelectric device adopting this structure using screening printing and demonstrated their application for powering

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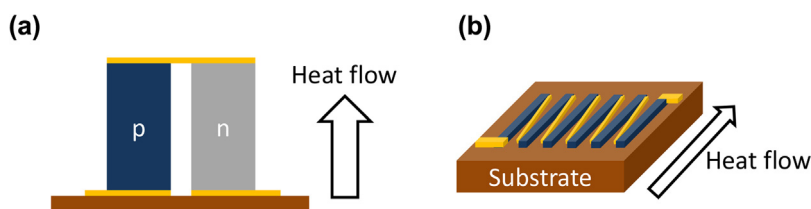


Fig. 1. Schematic representation of (a) π -type thermoelectric devices and (b) fin-type thermoelectric devices.

practical devices [27]. Sondergaard et al. and Fang et al. have also demonstrated large-scale fabrication using printing technologies [24,28]. These results suggested the possibility of using organic thermoelectric modules for harvesting heat energy at low temperatures. On the other hand, in these devices, the substrates take up volume, which means that the power density of these devices is not high. If we can minimize the volume of the substrate, we should be able to obtain compact devices with higher power densities. Compact devices with high power density are important for practical applications such as sensor networks and wearable devices.

In this paper, we report a proof-of-concept study of organic thermoelectric modules fabricated by thermal lamination at low temperature using the benchmark p-type material poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT/PSS). A 1.54 cm² device shows a power output of 37 μ W at a temperature difference of 50 K (corresponding to a power density of 24 μ W/cm²), which should be sufficient for powering practical devices such as sensors and light-emitting diodes. This approach enables the fabrication of large-area organic thermoelectric devices via roll-to-roll processing at low temperatures.

2. Experimental section

2.1. Materials

PEDOT/PSS (PH1000, Clevios) was purchased from H.C. Starck. Ethylene glycol (>99.5%) was purchased from TCI Chemicals. Nickel foil (5 μ m, 99%) was purchased from Nilaco Co., Japan. Polyvinyl butyral spray (Hayacoat Mark 2 AY-302) was purchased from Sunhayato, Japan.

2.2. Film preparation

The free standing PEDOT/PSS films were prepared following our previous report. In brief, PEDOT/PSS solution (10 mL) containing 3 wt.% ethylene glycol was added to a polystyrene case and heated on a hot plate at 40 °C [40]. After all of the solvent had evaporated, the PEDOT/PSS film was easily detached from the case. The thickness of the film is ca. 50 μ m. The as-prepared film was then cut into a 2.2 cm \times 2.2 cm square using a precision film cutter (NOGAMIGIKEN Co., Ltd.). Nickel foil was also cut into a 2.2 cm \times 2.2 cm square using a precision film cutter. Both sides of the Nickel foil were coated with a 20 nm-thick Au layer with an area of 0.1 cm \times 2.2 cm. The rest of the Ni foil was coated with polyvinyl butyral spray and the thickness was around 10 μ m. The as-prepared Ni foils and free standing PEDOT/PSS films were stacked inside a mold for alignment and laminated at 100 °C using a hydrostatic pressurizer with hot plates (AH-1TC, AS ONE, Japan).

2.3. Characterization

The film thickness was measured using a high-resolution digimatic measuring unit (VL-50-B, Mitsutoyo) and a surface profilometer (Surfcoder ET 200, Kosaka Laboratory Ltd.). The

conductivity was measured with a four-probe conductivity test meter (MCP-T600, Mitsubishi Chemical Corp.). All the electrical characteristics of the devices were measured using a semiconductor parameter analyzer (4200-SCS, Keithley). The temperature on different sides of the device was controlled by two Peltier units (Ampere, UT40U100F). The elastic properties were measured with force gauges (IMADA Co., Ltd.). The glass transition temperature of the polyvinyl butyral films was measured with a differential scanning calorimeter (Netzsch Instruments, Inc.).

3. Results and discussion

Fig. 2 shows a schematic representation of the fabrication of the thermoelectric devices using thermal lamination. This process started from a 5 μ m-thick Ni foil because of the negative Seebeck coefficient (-20μ V/K) of Ni at room temperature. The edge of both sides was first coated with 20 nm of Au to reduce the contact resistance with PEDOT/PSS (See discussion later). The rest was coated with ca. 10 μ m-thick polyvinyl butyral (PVB) as an insulating layer. After drying at 70 °C for 30 min, the foil was cut into a 2.2 cm \times 2.2 cm square using a precision film cutter. The free standing PEDOT/PSS films were prepared in a polystyrene case. 10 mL of PEDOT/PSS solution containing 3 wt.% ethylene glycol was added to a polystyrene case, and the case was then heated on a hot plate at 40 °C for 12 h. After all of the solvent had evaporated, the film was detached from the polystyrene case. The films, dried at 40 °C, were flat and uniform. The film was also cut into a 2.2 cm \times 2.2 cm square using a precision film cutter. The film thickness was ca. 50 μ m, and the electrical conductivity was larger than 800 S/cm. The as-prepared Ni foils and free-standing PEDOT/PSS films were stacked inside a mold for alignment and laminated at 100 °C. The number of layers typically varied from 10 to 100 depending on the required device area.

Two key points are critical for making a good device using this lamination approach. The first is the choice of insulating layers. From the device design, all layers should be connected in series. This layer should be thin but uniform and have good insulating characteristics to avoid direct contact between the PEDOT/PSS and the Ni foil in the center of the films. Once one of the layers connects in parallel, the open-circuit voltage could be significantly reduced. The insulating layer should also show good adhesion ability on Ni as well as PEDOT/PSS. This is because relatively thick PEDOT/PSS films cannot attach to the substrate well; although, it is used as a conductive glue in some cases. Polyvinyl butyral was chosen here because it meets these two requirements. It is well known that PVB is used as an insulating coating layer for integrated electronics to avoid corrosion [41]. The resistivity of the PVB layer is larger than $10^{13} \Omega$ cm and the breakdown voltage is larger than 1 kV for a 10 μ m-thick film. This is good enough for our purposes because both PEDOT/PSS films and Ni foil show much lower resistivity and the voltage drop between different layers is only several millivolts. PVB is also used as a glue showing good adhesion to many surfaces and ductility under pressure. In fact, PVB is used in lamination processes such as the preparation of safety glass.

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