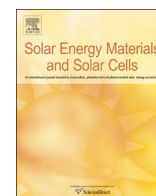




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Wearable organic solar cells with high cyclic bending stability: Materials selection criteria



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ABSTRACT

Despite the intrinsic flexibility of organic electronic materials due to their thinness, a deliberate selection of materials on the basis of their mechanical—not just charge-transport—properties is required for applications with mechanically demanding form factors, such as those that exist in the field of wearable electronics. This paper describes a skin-wearable solar cell enabled by the deliberate selection or intentional plasticization of the components to enable an extreme level of stability under cyclic bending deformations. In particular, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) films plasticized with 10% fluorosurfactant are used for both the anode and cathode, with the cathode layer further modified with polyethyleneimine (PEI) to lower the work function. Use of poly(3-heptylthiophene) (P3HpT) instead of the far more common poly(3-hexylthiophene) (P3HT) as the electron donor permitted extreme deformation because of its increased mechanical compliance (owing to its low glass transition temperature). Cells fabricated on 13- μm polyimide tape and adhered to human skin show stable performance when compressively strained by approximately 75%. These compressive strains produce convex and concave buckles with minimum radii of curvature of ca. 100 μm . Finite-element modeling predicts that the films require the ability to withstand a range of strains of 10%, when both convex and concave bends are considered. These devices, enabled by the stretchable semiconductor P3HpT, withstand up to one thousand cycles of compression with less than 20% degradation in power conversion efficiency, whereas devices based on P3HT show greater degradation after only five cycles, and fail catastrophically by fifty cycles. The usefulness of the wearable solar cells is demonstrated by their abilities to power an LED and a digital watch.

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1. Introduction

The surest strategy to promote organic solar cells (OSCs) from laboratory-scale demonstrations to use in the real world is to exploit the advantages possessed by organics that would be difficult or impossible to replicate in more-efficient competing technologies [1]. Such advantages include low cost and embodied energy [2], extreme thinness [3], tunable color [4], biodegradability [5], semitransparency [6], extreme flexibility [3], and stretchability [7]. These characteristics suggest that portable power for displays [8], mobile health monitoring devices, and mitigation of climate change triggered by burning of biomass in the developing world are—far from applications dismissible as “niche”—important problems for which organic solar cells may provide the ideal solutions [9]. While the community has produced impressive

demonstrations in ultra lightweight organic solar cells that can tolerate small bending radii [3], stability under cyclic deformation—required for real-world applications—has been poor. This fragility is a consequence of the fact that typical systems of materials are optimized overwhelmingly on the basis of efficiency on rigid substrates, as opposed to on the basis of mechanical stability. In this communication, we show that a deliberate selection of materials optimized for their mechanical properties can enable a new type of skin-mountable solar cell that can be used to power wearable electronic devices; these solar cells exhibit unprecedented stability to cyclic bending deformation. We performed these experiments to understand and anticipate routes of mechanical and photochemical degradation for all-organic solar cells under realistic operating conditions.

Despite the attractiveness of organic semiconductors for flexible and stretchable applications [1], the mechanical properties of these materials are highly variable, and there is a strong competition between compliance and charge transport [10]. The perceived compliance of optoelectronic polymers comes not because

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of their polymeric character, but because of the extreme thinness possible for the active layers of devices (ca. ≤ 100 nm) [11]. Thus unconventional active materials (e.g., liquid metals) [12], processing conditions, and device layouts [13,14] are generally required to produce devices stable enough to withstand extreme or repeated deformation, as would be encountered in ultra-thin, portable, or wearable applications [15]. The first wearable OSCs—i.e., integrated with clothing—were reported by Krebs in 2006 [16]. While these devices were flexible, the substrates and encapsulants were relatively thick (> 250 μm total thickness), and thus mechanical compliance was not maximized [16].

The first stretchable organic solar cell was reported by Lipomi et al. in 2011 [17]. In this device, an organic active layer was coated on a poly(dimethylsiloxane) (PDMS) substrate under uniaxial tension. Release of the tension produced buckles in the active layer that accommodated subsequent cycles of strain. However, because of the unencapsulated liquid top contact—eutectic gallium–indium, an expensive material necessary to conform to the topography of the buckles—the device was not wearable because it could not be inverted [17]. Later, Kaltenbrunner et al. demonstrated an OSC on an ultrathin (1.4 μm) polyester foil capable of accommodating bending radii ≥ 10 μm [3]. This device represents the state of the art in ultrathin organic cells, but it showed poor stability under repeated compression when the whole device was mounted to an elastic substrate—27% reduction in efficiency after only 22 cycles of 50% compression—and all devices were measured in a nitrogen-filled glovebox [3]. Our goal was to fabricate devices that retained at least 80% of their initial efficiency up to 1000 cycles of loading, on the skin, and in outdoor sunlight, by selecting materials optimized on the basis of mechanical stability.

Our laboratory and others [18–20] have elucidated many of the molecular and microstructural parameters that permit the coexistence of good electronic performance with extreme mechanical compliance [7,15]. The process and materials we used are illustrated in Fig. 1 (see Supporting information for experimental details). We designed the device to have the “inverted” geometry (cathode on the bottom) and used a thin (13 μm) polyimide (PI) tape as the substrate. While substrates as thin as 1.4 μm have been used before [3], these substrates are not available with an adhesive, and are mechanically more fragile than thicker ones. Crucially, we used the elastomeric conjugated polymer poly(3-heptylthiophene) (P3HpT) [21]—in place of the typical poly(3-hexylthiophene) (P3HT)—because P3HpT is the first poly(3-alkylthiophene) with increasing length of the alkyl side chain whose glass transition is substantially below room temperature [22]. This property renders P3HpT about an order of magnitude less stiff than P3HT, and substantially more ductile [21]. The relative compliance of P3HpT compared to P3HT is preserved even when mixed with the typical fullerene acceptor, [6,6]-phenyl C_{61} butyric acid methyl ester (PCBM), which is an antiplasticizer for poly(3-alkylthiophenes) (P3ATs) [21]. (In fact, the stiffening qualities of [60] PCBM are reduced when contaminated by up to 10% [70] PCBM—termed “technical grade” by manufacturers; thus, incompletely separated blends of methanofullerenes are substantially more compliant than pure samples, though we used 99% grade in this study for the sake of reproducibility) [23]. We used the typical transparent conductive polymer poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) as both the anode and cathode [24]. We added 10% w/w Zonyl fluorosurfactant to the PEDOT:PSS solution to permit wetting on the PI substrate and also to plasticize the films by a factor of 100 relative to the unmodified

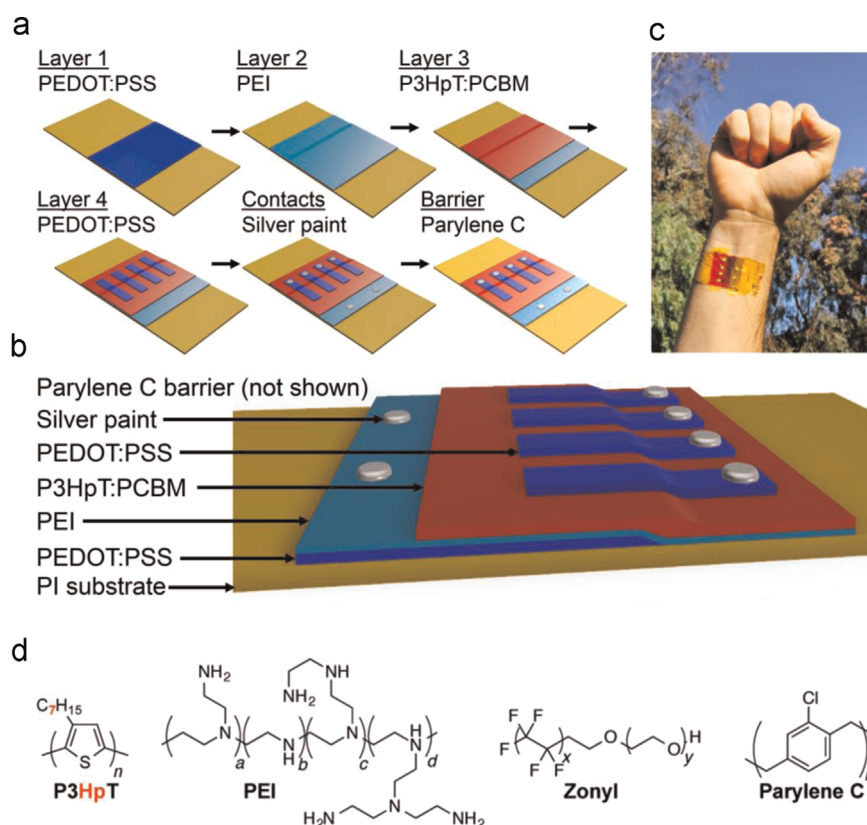


Fig. 1. Fabrication of ultra-flexible, wearable OSCs. (a) Schematic summary of the process used to fabricate wearable OSCs. Thin (13 μm) PI adhesive substrates were spin-coated with poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), polyethyleneimine (PEI), and P3HpT:PCBM. PEDOT:PSS top-contacts are then transferred on top of the device using thermal release tape, and a strip of the active layer was wiped away to expose the bottom electrode. Silver paint was then applied to make electrical contact. (b) Schematic diagram of the cross section of a wearable OSC. (c) A photograph of the wearable solar cell on skin. (d) Chemical structures of the critical materials used in this study.

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