

Large-area, flexible electronics for space applications

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Abstract

We report the thinnest, lightest, and most-flexible organic solar cell, with fabrication processes compatible with large-area processing (1). The efficiency of these ultrathin solar cells is equal to that of their glass-based counterparts. When transferred to a pre-stretched elastomeric support our ultrathin solar cells are shown to be ultracompliant. Our solar cells have an unprecedented power output per weight of 10 W/g, which is the highest specific weight and then, enabling new applications in ambient sensor nodes, remote sensing systems, weather balloons, unmanned aircraft, and space applications.

1. Introduction

Light-weight and mechanically resilient solar power sources are of increasing interest for modern applications such as electronic textiles, synthetic skin, and robotics (2,3). Organic photovoltaic (OPV) solar cells are highly promising in this sector. The thin-film devices comprise two electrodes, a light harvesting active layer and blocking or transport layers. The total thickness of a functional OPV cell is only a few hundred nanometers. Primary benefits of OPV cells are often listed as low cost, low weight, flexibility, and compatibility with reel-to-reel processing for high volume production (4,5). Efficiency and lifetime have reached commercially acceptable levels and flexible modules with roughly 1.5% power conversion efficiency and over 1 year lifetimes are already on the market (6). Mitsubishi Chemical Holdings have attained laboratory scale cell efficiencies exceeding 10% (7). Weight and flexibility are two of the primary benefits of OPV yet these mechanical properties are entirely dominated by the device substrate, which leaves much room for the advancement of OPV.

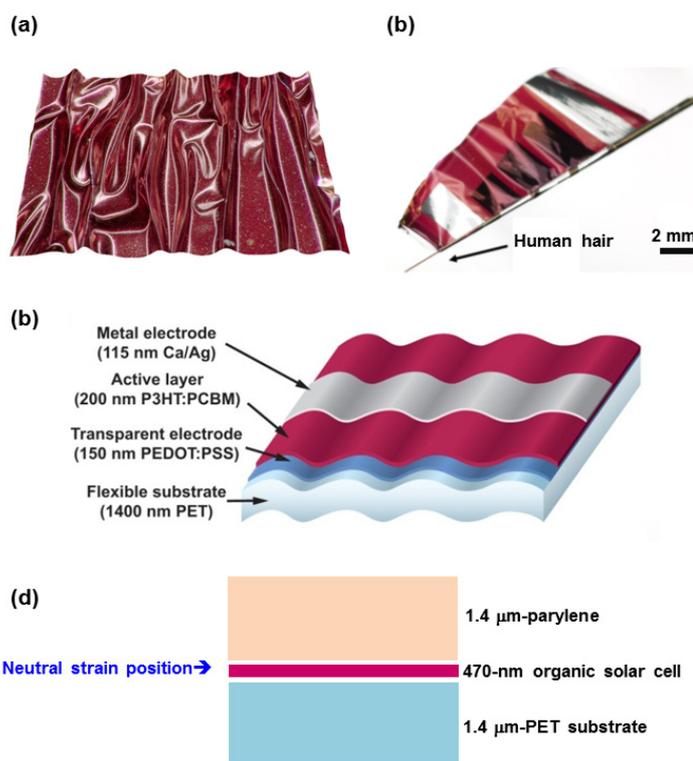


Fig. 1.(a) and (b) Pictures of a thin-film organic solar cells. (b) and (c) Schematic cross-sectional illustration.

In this work, we present ultra-thin, -light, -flexible and -compliant OPV devices constructed on only 1.4-μm-thick poly(ethylene 2,6-)naphthalate (PEN) substrates. We obtain 4.2% power conversion efficiency, an unprecedented specific weight value of 10 W/g and can reversibly attain strains of 70% on elastomeric

support. Other than the substrate, all of the materials used for our devices are well-established materials for OPV, including poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), poly(3-hexylthiophene) (P3HT), (6,6)-phenyl-C61-butyric acid methyl ester (PCBM), and Ca/Ag evaporated metal electrodes. Using these materials, OPV devices on indium tin oxide (ITO) coated glass substrates have been certified with efficiency up to 4.3% (8). Using the ITO-free architecture on PEN we obtain comparable efficiency while demonstrating the extreme mechanical properties of organic solar cells.

Among the thinnest reported substrates for OPV devices are 125 μm PET and PEN (9-11). The resulting solar cells are highly flexible with radius of curvature of 5 mm. Stretchable devices have been fabricated on pre-stretched PDMS (thickness 200-500 μm), which can then be compressed to 27% of their original size reversibly (12). A liquid metal back contact was used to improve mechanical stability. In all of these cases the actual solar cell still constitutes less than 0.25% of the total device thickness. Inorganic GaAs solar microcells (thickness 6.6 μm with polymer coatings) with high areal coverage allowed for 20% strain and wrapping around cylinders with 1.5 mm radius (13). Reducing the substrate thickness further to the 1.4 μm thin PEN films used here significantly improves the specific weight of the system but it also drastically improves the mechanical resilience under flex or compression.

2. Device manufacturing, results, and discussions

Pictures and schematic illustration of an ultra-thin solar cell is shown in Fig. 1. The total device thickness without an encapsulation layer is only 1.9 μm , where about one-quarter of the thickness is the active solar cell. The P3HT:PCBM bulk heterojunction solar cell is fabricated on a PEDOT:PSS coated 1.4 μm thick PEN substrate foil. Top electrodes are 115 nm Ca/Ag. Fig. 1b illustrates the extreme bending flexibility of the solar cell, wrapped around a human hair with a radius of 35 μm , and current density (J)-voltage (V) characteristics can be seen in Fig. 2. Fig. 3 depicts a solar cell on a 100 μm thick pre-stretched 3M VHB 4905 elastomer. As the strain of the elastomer is reduced by 30% and then 50%, it is evident that the solar cell becomes wrinkled. This enables the solar cell to be stretched back to the pre-strain defined by the elastomer.

The ultra-thin flexible substrate is completely compatible with scalable processing and, therefore, is advantageous for high throughput fabrication. During fabrication, we avoid complex transfer printing processes.

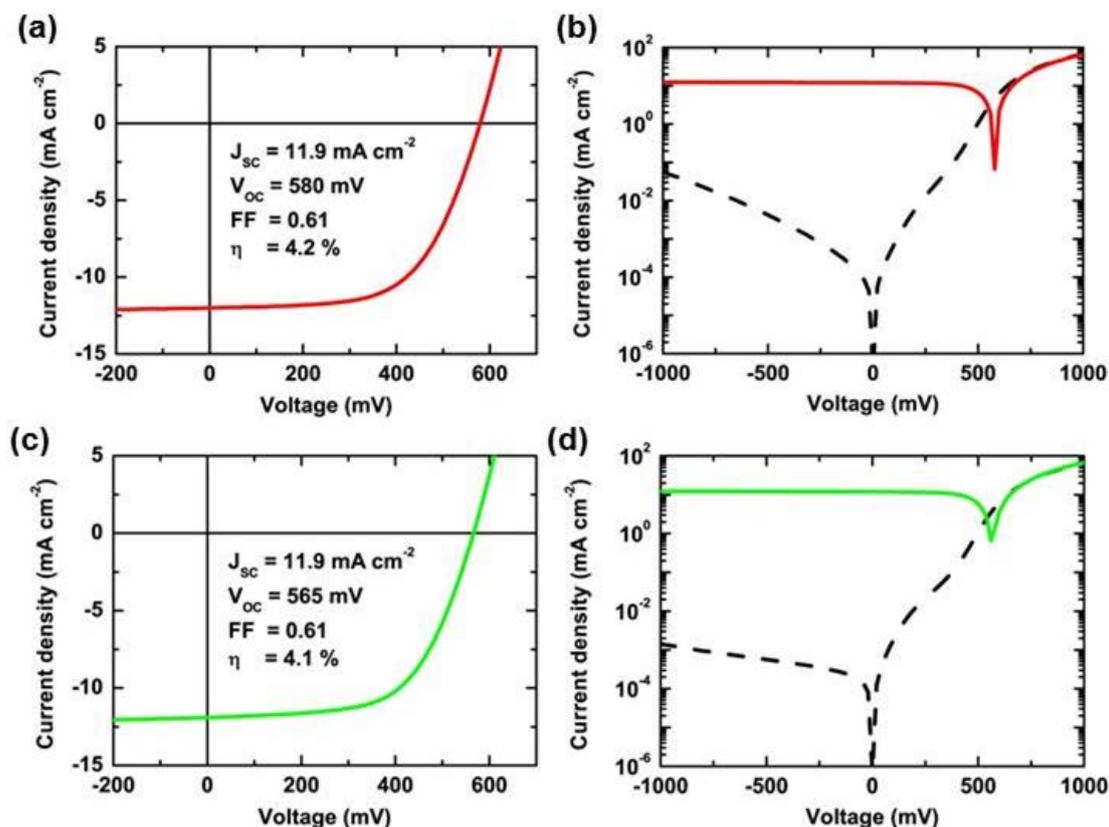


Fig. 2. Current density (J)-voltage (V) characteristics obtained from organic solar cells on 1.4- μm -thick PET substrate (a) and (b), and on glass (c) and (d). Dashed lines represent dark current density as a function of voltage.

For laboratory-scale device construction, the substrate is placed onto a supporting glass plate for processing ease. The PEN adheres to the PDMS coated glass via weak van der Waals bonding, which allows standard laboratory processes such as spin coating, thermal annealing, and vacuum evaporation. The primary functions of the glass support are to maintain planarity during processing and to ease the handling of the device, and the PEN can be easily removed by peeling it away. Exact processing details are found in the experimental details section.

Prior to removing the PEN from the supporting glass, the device J-V characteristics are measured under the solar simulator (approximately the A.M 1.5 Global spectrum with 100 mW/cm^2 intensity and spectral mismatch correction), and in the dark. After removal from the supporting glass, it is difficult to maintain planarity in the device, and therefore to know the incident area for calculation of current density (J) and power conversion efficiency (η). All measurements while the device is still attached to the glass support are scaled per unit area, using a conservative area estimate as the PEDOT:PSS transparent electrode is difficult to pattern exactly. The data reported in subsequent sections, for mechanical stretching and cycling, is not scaled to area and only total current and power are reported for comparison during mechanical manipulation. The J-V characteristics of an optimized device are shown in Fig. 2. This particular device performs with $V_{OC} = 580 \text{ mV}$, $J_{SC} = 11.9 \text{ mA/cm}^2$, F.F. = 61%, and $\eta = 4.2\%$ under 1 sun illumination. This is a representative sample from over 50 devices constructed with various parameters. Of primary concern here are the transparency and conductivity of the PEDOT:PSS transparent electrode. By varying the thickness of the layer, there is a trade-off between F.F. and J_{SC} , as a thicker electrode will be more conductive but less transparent.

The device performance shown in Fig. 2a shows one of the best-performing devices. However, through mass construction of the devices with slight variations, we have obtained a wide range of F.F. (up to 71%) and J_{SC} (up to 12 mA/cm^2) though not in the same device. The average η of the fabricated samples was 3.8%, and $\eta > 3.5\%$ was easily reproducible. As seen in Fig. 2, the device shows reasonable dark diode rectification.

These results are very comparable to leading BJJ OPV devices using P3HT:PCBM on ITO-coated glass substrates (8). The scarcity and cost of indium, and the requirement of smooth substrates for thin films are considered to be significant hurdles for mass production of OPV devices (14,15). The surface roughness of the PEN substrate, and the high-conductivity PEDOT:PSS in place of ITO do not significantly reduce the device efficiency.

The primary goal in constructing such thin devices is to demonstrate continuous opto-electronic operation under extreme mechanical duress. In order to do so, we transfer the device from the glass support to a pre-stretched elastomer. (3M VHB 4905) The device on PEN sticks to the elastomer very well, and the glass support can be removed. Two rigid plastic spacers are placed on the elastomer, leaving a gap of 1 cm for the compression and stretching of the device. This assures that the strain will be nearly one dimensional and concentrated in the region of the active device, which occupies roughly half of the 1 cm gap. The rigid spacers are brought closer together in steps of 1 mm, corresponding to 10% compression. At each step the I-V characteristics are measured under the solar simulator, and the results are shown in Fig. 3. It is clear that the device remains a functional solar cell down to 80% compression. Upon re-stretching the device, it returns to its pre-compression operation state. The V_{OC} , I_{SC} , F.F., and output power are plotted as a function of compression in Fig. 3e-f, all normalized to their pre-compression value. As the incident area should track linearly 1 to 1 with the compression, we would expect the I_{SC} and power to scale exactly with the compression, following the black line in Fig. 3f. We can see that both values are consistently higher than the device area scaling.

The optimistic explanation for this would be an enhancement in light harvesting due to micro-texturing upon folding of the device. Light not absorbed on the first pass through the active layer would be reflected back into the device on the other side of the crease, allowing for improved overall absorption. Additionally, the compression does not decrease the device area uniformly. As the device area is defined by the metal back electrode, it is likely that the overall substrate compresses more on the edge. Thus, when the total compression is 50%, the device area is not reduced by exactly the same value. At compression greater than 60%, there is a sharp drop off in the I_{SC} and the power output. At such values, the elastomer no longer sticks to the rigid plastic delimiters, so the device begins to fold slightly behind and is shaded from the illumination.

Biaxial compression and stretch testing was also performed. The OPV devices on PEN substrates were attached to the same elastomer, now pre-stretched in two directions. In order to do so, the rigid plastic pieces that constricted the compression to one dimension were left off, which makes the actual compression of the device much less certain. We estimate 50% compression in one direction with an additional 20% compression in the other. The overall biaxial stretch behavior is similar to the uniaxial stretch described above. This approach for stretchable solar cells has several distinct advantages. The planar-only fabrication processes are fully reel-to-reel compatible. The complete solar cells show large, reversible quasi-linear stretching ratios over 70% when

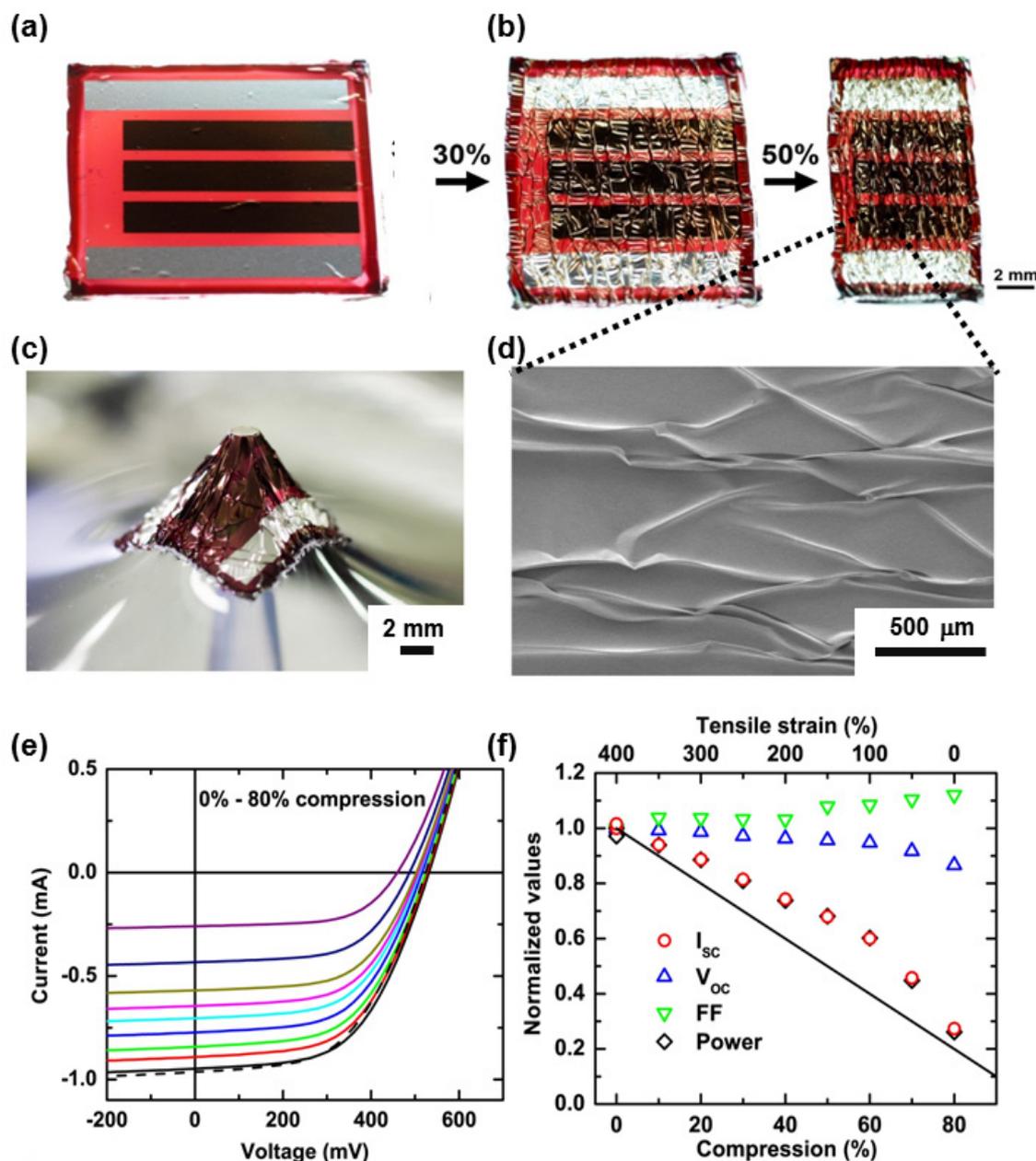


Fig. 3.1.4- μm -thick organic solar cell on rubber substrate with (a) 100%-pre-stretching (b) 50%- and 30%- compression., (c) An organic solar cell with three-dimensional deformation. (d) Scanning electron microscope image of the surface of organic solar cell. (e) J-V characteristics with changing compressive strain from 0 to 80%. (f) I_{sc} , V_{oc} , FF, and Power as a function of compression.

transferred onto an elastomeric support.

Though compression and stretching of the device induces very little change in the device performance, there may be small changes that accumulate over many cycles. To investigate the durability of the devices under repeated stretching and compression, we cycled the device from 0% (original size) to 50% compression and back more than twenty times. At each cycle the I-V characteristics were measured under the solar simulator, both in the compressed and extended state. The cycling test showed gradual decrease in the I_{sc} , the FF, and the output power, resulting in a 27% decrease in power after 24 cycles.

When considering solar power for certain applications, the power output per weight (specific weight) may be the most critical metric. This is particularly true for situations that require portable power; where payload is a premium such as aircraft, spacecraft, or personal pack-load. The specific weight of various solar technologies (panels and cells) for terrestrial and space applications is compared in Fig. 4. Commercially available space rated high efficiency Si and triple junction cells have 0.82 W/g and 0.39 W/g respectively (17). A recent report showed

CIGS cells constructed on thin (25 μm) polyimide films with 18.7% efficiency (17). These devices have extremely high specific weight of roughly 3 W/g. The OPV devices constructed on PEN presented here have a per-area mass of 4 g/m^2 , and 4% efficiency, giving 10 W/g.

This extreme specific weight may be advantageous for certain applications. Total power and device lifetime are obviously primary factors for high-cost situations, like most space-based uses. But weather balloons, unmanned aircraft, or any other remote sensing systems may prioritize specific weight and have known project duration. The same is true for pack-weight for remote wilderness use, where there is often a need for light-weight power sources. Ultra-thin OPV may be ideal for such applications. Polymer based solar cells have been constructed on 1.4 μm thick PEN substrate material. The devices use standard OPV materials and obtain nearly identical efficiency to those constructed on ITO coated glass substrates with over 4% power conversion efficiency. Here, the high conductivity PEDOT:PSS electrode replaces the ITO which is considered a cost and volume limiting material (14). The polymer electrode is highly flexible compared to the ternary oxide (19). The device layers consist of roughly 450 nm total thickness, so the total device including substrate is less than 2 μm .

These ultra-thin, flexible solar cells show unprecedented mechanical resilience. By adhering the flexible device to a pre-stretched elastomer we have shown that the devices survive quasi-linear compression to below 70% of their original area. Furthermore, we have shown cyclic compression and stretching to 50%, over more than 20 full cycles with marginal loss in device performance and no visible defect formation beyond the external contact points. Though the electrically active components of this device are in principle no different than other standard OPV devices, the solar cell now constitutes roughly 25% of the total thickness and 45% of the total weight. OPV is emerging as a viable technology where temporary, high-volume, and low-weight power sources are required. The extreme flexibility and specific weight demonstrated here realize the unique potential for OPV as an ultra-thin-film power source.

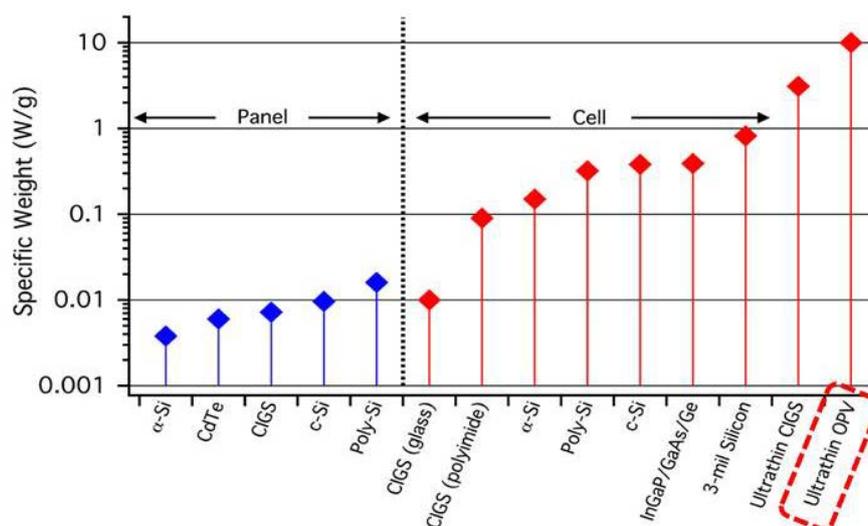


Fig. 4. Specific weight of various photovoltaic technologies for complete modules (left) and individual cells (right). Module data is taken from commercially available panels and should be representative: Kaneka 12V 55W PV Module, First Solar FS272, Sunforce 70W Pro Series, Bosch Solar Module c-Si M60, and Kyocera KD245Gx-LPB. Cell data was taken from published academic results. In many cases, the cell weight was not published and so was estimated from published layer thicknesses and known material densities, assuming 2% areal coverage for metal grids contacts (17,18,22-26).

Methods Summary

Rigid glass slides, coated with a ≈ 500 nm thick polydimethylsiloxane (PDMS) layer served as supporting substrates for the 1.4 μm thick PEN foils (obtained from Pütz GmbH + Co. Folien KG). Adhering the PEN film to the supports readily enabled all subsequent planar processing steps including spin-coating, thermal annealing and vacuum deposition. The devices could be peeled of the supporting stack without causing damage to the solar cells. We spun PEDOT:PSS (Clevios PH1000, with 5 vol% dimethylsulfoxide, and mixed with 0.5 vol% Zonyl FS-300 fluorosurfactant from Fluka to promote wetting on the hydrophobic PEN) at 1000 rpm for 60 s and then 2000 rpm for 60 s onto the PEN films. The samples were dried on a hotplate at 120 $^{\circ}\text{C}$ for 30 min, yielding 150 nm thick PEDOT:PSS layers with ~ 100 Ω/sq . A solution of 30 mg/ml P3HT (obtained from Plextronics and purified by repeated reprecipitation) and 30mg/ml PCBM (from Solenne BV) in dichlorobenzene at 1500 rpm for 120 s. The P3HT:PCBM was partly wiped away with a toluene-soaked cotton swab, exposing some of the

PEDOT:PSS to allow for contacting. 200 nm thick P3HT:PCBM films were obtained. The samples were transferred to a nitrogen-filled glovebox and annealed on a hotplate at 150° C for 15 min. Finally, 15 nm Ca and 100 nm Ag were thermally evaporated at 1 Å/s and pressure of 10⁻⁶ mbar. Typical devices had an active area of 0.1 cm². The devices were first characterized in the dark and under illumination of a solar simulator (A.M 1.5 Global spectrum with 100 mW/cm² intensity with spectral mismatch correction). I-V characteristics were recorded using a Keithley 236 source meter. Stretching experiments were performed with a purpose-built stage. All measurements were carried out in a nitrogen-filled glovebox. Layer thicknesses were measured with a Digital Instruments Dimension 3100 AFM.

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References

- [1] Kaltenbrunner, M. et al., "Ultrathin and lightweight organic solar cells with high flexibility". *Nature Communications*, vol. 3, 770 (2012).
- [2] Lipomi, D. J. & Bao, Z. "Stretchable, elastic materials and devices for solar energy conversion". *Energy Environ. Sci.* vol. 4, pp. 3314-3328 (2011).
- [3] Baca, A. J. et al. "Compact monocrystalline silicon solar modules with high voltage outputs and mechanically flexible designs". *Energy Environ. Sci.* vol.3, pp.208-211 (2010).
- [4] Brabec, C. J., Sariciftci, N. S. & Hummelen, J. C. "Plastic solar cells". *Adv. Funct. Mater.* 11, 15-26 (2001)
- [5] Coakley, K. M. et al. Conjugated polymer photovoltaic cells. *Chem. Mater.* vol.16, pp.4533-4542 (2004)
- [6] Hauch, J. A. et al. "Flexible organic P3HT:PCBM bulk-heterojunction modules with more than 1 year outdoor lifetime". *Solar Energy Materials and Solar Cells* vol.92, pp.727-731 (2008).
- [7] Aramaki, S., et al. Private communication (2011).
- [8] Li, G. et al. "High-efficiency solution processable polymer photovoltaic cells by self-organization of polymer blends". *Nat. Mater.* vol.4, pp.864-868 (2005).
- [9] Rowell, M. W. et al. "Organic solar cells with carbon nanotube network electrodes". *Appl. Phys. Lett.* vol.88, 233506 (2006).
- [10] Galagan, Y. et al. "ITO-free flexible organic solar cells with printed current collecting grids". *Solar Energy Materials and Solar Cells* vol.95, pp.1339-1343 (2011).
- [11] Yambem, S. D., Liao, K. S. & Curran, S. A. "Flexible Ag electrode for use in organic photovoltaics". *Solar Energy Materials and Solar Cells* vol.95, pp.3060-3064 (2011).
- [12] Lipomi, D. J., et al. "Stretchable organic solar cells. *Adv. Mater.* vol.23, pp.1771-1775 (2011).
- [13] Lee, J. et al. "Stretchable GaAs photovoltaics with designs that enable high areal coverage". *Adv. Mater.* vol.23, pp.986-991 (2011).
- [14] Fortunato, E., et al. "Transparent conducting oxides for photovoltaics". *MRS Bull.* vol.32, pp.242-247 (2007).
- [15] Tenent, R. C. et al. "Ultrasoft, large-area, high-uniformity, conductive transparent single-walled-carbon-nanotube films for photovoltaics produced by ultrasonic spraying". *Adv. Mater.* vol.21, pp.3210-3216 (2009).
- [16] Graz, I. M., et al. "Extended cyclic uniaxial loading of stretchable gold thin-films on elastomeric substrates". *Appl. Phys. Lett.* vol.94, 071902 (2009).
- [17] Fatemi, N. S., Pollard, H. E., Hou, H. Q. & Sharps, P. R. "Solar array trades between very high-efficiency multi-junction and Si space solar cells". *Conference Record of the 28th IEEE Photovoltaic Specialists Conference*, Anchorage, AK, USA (2000).
- [18] Chirila, A. et al. "Highly efficient Cu(In,Ga)Se₂ solar cells grown on flexible polymer films". *Nat. Mat.* advance online pub. 18 Sept. (2011).
- [19] Sierros, K. A., et al., "Stress-corrosion cracking of indium tin oxide coated polyethylene terephthalate for flexible optoelectronic devices". *Thin Solid Films* vol.517, pp.2590-2595 (2009).
- [20] Netter, T., "CIGS on plastic: efficiency goes high". http://www.interpv.net/magazine/mag_view.asp?idx=248&part_code=01 (2010).
- [21] Soderstrom, T. Haug, F. J., Terrazoni-Daudrix, V. & Ballif, C. "Optimization of amorphous silicon thin film solar cells for flexible photovoltaics". *J. App. Phys.* vol.103, 114509 (2008).
- [22] Rath, J. K., et al. "Fabrication of thin film silicon solar cells on plastic substrate by very high frequency PECVD". *Solar Energy Materials and Solar Cells* vol.94, pp.1534-1541 (2010).
- [23] Zhao, J., Wang, A., Green, M. A. & Ferrazza, F. "19.8% efficient „honeycomb“ textured multicrystalline and 24.4% monocrystalline silicon solar cells". *Appl. Phys. Lett.* vol.73, pp.1991-1993 (1998).
- [24] Zhao, J., Wang, A., Altermatt, P. & Green, M. A. "Twenty-four percent efficient silicon solar cells with double layer antireflection coatings and reduced resistance loss". *Appl. Phys. Lett.* vol.66, pp.3636-3638 (1995).