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Stretchable Organic Semiconductor Devices

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Abstract: Stretchable electronics are essential for development of intensely packed collapsible and portable electronics, wearable electronics, epidermal and bio-implanted electronics, 3-D surface compliant devices, bionics, prosthesis and robotics. However, most of the stretchable devices are currently based on inorganic electronics, whose high cost of fabrication and limited processing area makes it difficult to produce inexpensive, large-area devices. Therefore, organic stretchable electronics are highly attractive due to many advantages over their inorganic counterparts, such as light-weight, flexibility, low cost and large-area solution-processing, reproducible semiconductor resources, and easy tuning of properties via molecular tailoring. Among them, stretchable organic semiconductor devices have become a hot and fast-growing research field, in which great advances have been made in recent years. These fantastic advances are summarized in this review, focusing on

stretchable organic field effect transistors, light-emitting devices, solar cells and memory devices.

Outline

1. Introduction

2. Design of Stretchable Electrodes and Devices

2.1 Design of Stretchable Electrodes

2.2 Design of Stretchable Devices

3. Stretchable Organic Field Effect Transistors (OFETs)

3.1 OFET Matrixes with Stretchable Interconnectors

3.2 Buckled OFETs

3.3 Intrinsically Stretchable OFETs

3.3.1 Stretchable Active Layers

3.3.2 Intrinsically Stretchable OFETs

4. Stretchable Organic Solar Cells (OSCs)

4.1 Buckled OSCs

4.2 Spring- or Mesh-shaped OSC fibers

4.3 Intrinsically Stretchable OSCs

4.3.1 Stretchable transparent electrodes

4.3.2 Stretchable Active Layers

4.3.3 Intrinsically Stretchable OSCs

5. Stretchable Organic Light-emitting Devices

5.1 Buckled OLEDs

5.2 Intrinsically Stretchable Organic Light-emitting Devices

5.2.1 Intrinsically Stretchable OLEDs

5.2.1 Intrinsically Stretchable OLECs

5.3 Stretchable OLED Displays

6. Stretchable Organic Memory Devices (OMDs)

7. Challenges and Outlook

Introduction

Stretchable electronics have received increasing attentions in recent years due to great potential in wearable electronics, epidermal and bio-implanted electronics, mechanically 3-D surface compliant in-door and out-door devices, and their integration with bionics, prosthesis and robotics.^[1-15] Stretchability offers high durability and robustness for intimate conformabilities with the surfaces of arbitrary complex-shaped static and moving objects, so that the devices can survive repeated crumpling, creasing, sharp folding or even stretching without incurring physical damage. Traditional inorganic electronic materials are normally too brittle to be used in stretchable electronics, especially in intrinsically stretchable devices. For inorganic electronics, several strategies for achieving stretchability are generally adopted, such as using stretchable interconnects between rigid individual device for realizing stretchable matrixes, using buckled-, spring-, meshed-shape in configuration design for stretchable devices, and fabricating random networks of inorganic semiconductor nanowires for intrinsically stretchable devices. However, the high cost of fabrication and limited processing area makes it difficult to produce inexpensive, large-area devices. Therefore, stretchable organic electronics have become highly attractive due to the intrinsic stretchability, low cost and large area solution processing.^[16-19]

Organic semiconductors refer to organic materials exhibiting semiconductor properties with an electrical conductivity between that of insulators and that of metals. Organic single molecules, oligomers, and organic polymers can be semiconductive. Organic semiconductor devices, defined as electronic functional devices employing organic semiconductors as active layers, have emerging as a fast developing field due to favorable advantages over traditional silicon-based inorganic semiconductor devices. These advantages include light-weight, flexibility, reproducible semiconductor resources, easy tuning of functionalities via molecular tailoring, and, most importantly, low cost, high throughput and large-area solution-processing which are compatible with screen printing, inkjet printing and roll-to-roll coating techniques.

Organic semiconductor devices mainly including organic field effect transistors (OFETs),^[20-28] organic light-emitting diodes (OLEDs),^[29-36] organic light-emitting electrochemical cells (OLECs),^[37-40] organic solar cells (OSCs)^[41-48] and organic memory devices (OMDs).^[49-61] To date, great improvements have been made for achieving high performance that is comparable to their inorganic counterparts. For example, OLEDs with external quantum efficiency (EQE) of >30% for blue^[62-65] and green^[66-70] emitting devices, and >20% for red^[71] emitting devices, OSCs^[72, 73] and organic-inorganic hybrid perovskite solar cells^[74] with power current efficiency (PCE) of >10% and 20%, respectively, OFETs with mobility of several tens $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$,^[75, 76] and OMDs with high densities, high ON/OFF ratios, fast response and long durability^[59, 77-79] have been reported.

Recently, stretchable organic semiconductor devices have become an emerging discipline, remaining very big development space and research value.^[63, 80-83] Great advances have been made in this field, which have been summarized in this review, focusing on OFETs, organic light-emitting devices (including OLEDs and OLECs), OSCs and OMDs. Some of the significant developments are depicted in **Figure 1**. Other stretchable electronic devices utilizing one or more organic elements (i.e. electrodes, electrolytes, separators, stimuli-responsive and sensing materials), such as sensors,^[84] capacitors,^[85-88] batteries,^[89] piezotronic and piezo-phototronic devices,^[5] electrochromic devices,^[90, 91] actuators,^[92-97] generators,^[98] and photodetectors,^[99, 100] are not included here, and interested readers are directed to other excellent reviews and articles on these subjects.

2. Design of Stretchable Electrodes and Devices

2.1 Design of Stretchable Electrodes

As fundamental components of organic electronic devices, thin-film electrodes firstly need to be endowed with stretchability for fabrication of stretchable devices. The stretchable electrodes should exhibit little or no change in conductivity at stretching states. Based on

flexible electrodes, various strategies are utilized to further obtain stretchability. For the metal electrodes, liquid metals such as EGaIn are demonstrated to be stretchable.^[101-104] For the conductor/polymer hybrid electrode, highly elastic polymer matrixes can be applied.^[80, 102, 103, 105-110] Some additives can be helpful to enhance the stretchability.^[111-113] These stretchable electrodes are essential for developing intrinsically stretchable devices.^[103, 109] Another method for achieving stretchability is based on configuration design. For the thin-film electrodes, buckled/waved,^[114-118] microcracked^[103] or porous^[119-122] structures can be applied. For the fiber-, tape-, wire-, **or textile-** shaped electrodes, spring-,^[118, 123, 124] serpentine-,^[11, 125-127] mesh-,^[128] kirigami,^[129, 130] origami^[131] and buckled^[118, 132, 133] structures ensure enhanced stretchability. Detailed summary of these stretchable electrodes have been reported in our previous review.^[140] **In addition, low-cost printing, patterning, and development of appropriate measurement techniques of the stretchable electrodes are necessary for the fabrication of high-density device arrays or matrixes.**^[134-139]

2.2 Design of Stretchable Devices

To realize stretchable devices, there are mainly three design strategies. The first strategy is to fabricate device matrix with stretchable interconnectors, either the rigid but winding-type interconnectors or soft elastic interconnectors, between individual rigid devices.^[80, 141-145] The second strategy is to design buckled,^[101, 114-117, 146-148] spring-^[149] or mesh-^[128] shaped device configurations. The third strategy, due to the more or less inherent stretchability of organic materials, is to develop intrinsically stretchable devices which employ as many as possible elastic elements (stretchable substrates, e.g. polydimethylsiloxane (PDMS)), stretchable electrodes, stretchable active layers, or even stretchable barriers).^[17, 102-104, 108-110, 118, 150-155] To further improve the practicability of the devices, complementary functionalities such as self-healing^[12, 156-162] and self-powering^[98] capabilities can be added.

3. Stretchable Organic Field Effect Transistors (OFETs)

OFETs are basic building blocks for organic logic circuits and are essential for development of printable and flexible electronic technologies, which find many applications including circuits,^[142, 146] displays,^[163, 164] memories,^[165] photodetectors,^[166-168] sensors,^[169-171] transconductance amplifiers,^[139, 172] radio frequency identification (RFID) transponders,^[173] and oscillators.^[174] Stretchable OFETs have been demonstrated with wide applications such as light-emitting display,^[145] skin-inspired electronics,^[6, 175] and implantable electronics.^[176-178]

3.1 OFET Matrixes with Stretchable Interconnectors

To fabricate stretchable OFET matrix or arrays, one general strategy is to construct the individual rigid OFET units on the soft and stretchable substrates with linkage of stretchable conductive interconnectors. Under tension, these stretchable interconnectors lengthen, so that the device performance in intersection areas is nearly unaffected. These interconnectors include metal integrated elastic mesh,^[128, 142] stretchable conductor wiring,^[80, 141] and ductile metal electrodes on soft part of stiffness-gradient substrates.^[143] Someya *et al* developed a conformable, stretchable and large-area mesh-like OFET matrix by cutting holes on stand-alone flexible planar devices, which functions electrically with a unidirectional extension of 25% and can easily be laminated with pressure and/or thermal sensors (**Figure 2a**).^[128] Seo *et al* constructed stretchable devices directly on as-prepared mesh-like elastic substrate, by designing the metal electrodes extending as winding interconnectors (**Figure 2b**).^[142] Similar mesh-like structure with winding metal interconnectors has also been demonstrated in Si-based circuits.^[179]

Another strategy is to develop soft, elastic and stretchable conductors as interconnectors. Someya group developed a stretchable and highly conductive PDMS-coated SWCNTs/ionic liquid composite film, which exhibited a conductivity of 57 S cm⁻¹ and a stretchability of 134%. Further, these elastic conductors were integrated with printed OFETs to fabricate a rubber-like active matrix with an effective area of 20×20 cm², which can be

uniaxially and biaxially stretched by 70% without mechanical or electrical damage (**Figure 3**).^[80] The third strategy is to utilize stiffness-gradient substrate with ductile metal electrode interconnectors laminated on the soft regions.^[143, 144] By constructing ductile metal source/gate interconnected electrodes on the soft regions of the substrate and active OFET units on **polyimide (PI)**-mounted stiff regions, stretchable OFET matrix tolerant to 50% strain was demonstrated.^[143] The PI stiff islands, on which the individual devices are fabricated, are defined by conventional photolithography on a stress-free elastomeric substrate.^[144] When the substrate is stretched, the deformation is mainly accommodated by the substrate. Thus, the stiff islands and above-constructed electronic devices experience relatively small strains. The combination of the elastic interconnected conductors and stiffness-gradient substrate has been demonstrated to obtain an OFET matrix sustaining up to 110% stretch without impairing functionality (**Figure 4**).^[141] The printable mesh-like elastic conductor comprised of Ag flakes, a fluorine rubber and a fluorine surfactant has a high initial conductivity of 738 S cm^{-1} and remains at 182 S cm^{-1} under 215% strain.

3.2. Buckled OFETs

Buckled devices are generally prepared on prestrained substrates which are released or relaxed after device fabrication. The full relaxation from the strain results in ripple- or wave-like shape of the device, which helps to sustain larger strain or stretch. Bettinger *et al* fabricated a wave-like OFET on the pre-strained sinusoidal topographic SiO coated PDMS substrate by conformal coating methods, which still operated under strains up to 12%.^[148] No severe decrease of device performance of the flat-configuration counterparts was observed for these buckled devices. So that the devices are less affected by the stretch, stiff islands can be utilized on the buckled and stretchable substrates.^[143] For example, the active device and the ends of source/drain electrodes were mounted on the flat rigid PI islands, while the central part of electrodes was conformably laminated on buckled surfaces. The endurable maximum

strains were estimated to be 2.28, 9.70, and 9.32% for the OFETs formed on flat, 1D-, and 2D-wavy elastomers, respectively. By constructing buckled devices onto ultrathin substrates, high flexibility and stretchability can be simultaneously obtained. The vacuum-evaporated DNTT-based OFETs were constructed on 1.2 μm -thick PEN foils coated on pre-stretched elastomer, followed by release of the device. The subsequently obtained buckled OFETs with a total device thickness of 2 μm demonstrated extreme light-weight (3 g m^{-2}), ultraflexibility (bending radius of 5 μm) and reversibly high stretchability (233%) and compressibility (70%), virtually with no performance degradation (**Figure 5**).^[115] After 200 full cycles, less than 4% change in device parameters was observed. Even after 1,000 full cycles, the device parameters retained 80% of their initial values. Using the same strategy, Fukuda *et al* further reported a full-solution-processed buckled high performance, ultraflexible (bending radius of 140 μm) and stretchable/compressive (100% strain, 50% compress) OFETs fabricated on 1- μm -thick parylene-C substrates, using printed Ag nanoparticle ink as electrodes and a soluble p-type small molecule material as active layer.^[146]

3.3 Intrinsically Stretchable OFET

3.3.1 Stretchable Active Layers

To fabricate intrinsically stretchable OFET, it is essential to employ all stretchable elements. Since ductile metals (such as Au) or liquid metals (such as EGaIn) for stretchable electrodes are commercially available, it is essential to develop intrinsically stretchable active channel materials. It has been reported that the crack onset strain of poly(3-hexylthiophene) (P3HT), a commonly used solution-processable channel polymer material, is greater than 150%.^[180] This indicates a great potential for achieving intrinsically stretchable OFETs. In addition, large anisotropic charge transport characteristics were demonstrated in the strain-induced highly-aligned regioregular (RR=99%) P3HT, because the polymer backbone is shown to align in the direction of applied strain which induces a significant face-on orientation of the originally highly edge-on oriented crystalline regions of the film (**Figure**

6a).^[181] The in-plane mobility increases in the applied strain direction (along the pi-pi intrachain direction) and decreases in the perpendicular direction (along the pi-pi interchain direction), resulting in an optical dichroic ratio of 4.8 and a charge-mobility anisotropy of 9. These results are consistent with theoretical calculations.^[182] O'Connor group also found this strain-induced face-on packing, and further found that this charge transport anisotropy will be much weakened by biaxial stretching.^[183] The mobility in the biaxially strained films is greater than that for unstrained films for channel lengths of $\leq 10\text{ }\mu\text{m}$ (**Figure 6b**), suggesting that edge-on packing is not a prerequisite for relatively high-field-effect mobility in P3HT-based OFETs.

However, they also found that dependence of anisotropic elastic modulus of oriented regioregular poly(3-hexylthiophene) films on the strain direction is opposite to that of mobility.^[184, 185] The elastic modulus of the film increases in the direction of backbone alignment and decreases in the transverse direction. Although high-RR P3HT demonstrates a relatively high mobility, it is brittle. Lowering RR improves the elastic properties, but causes a decreased mobility (**Figure 7**).^[186] Therefore, it should reach a compromise of the stretchability and mobility. Jeong group found that low-RR P3HTs generate neat long nanofibrils, whereas high-RR P3HTs form short nanofibrils, and the mixing ratio of high- and low-RR P3HTs can be optimized to enhance the device performance.^[187]

To enhance the stretchability, molecular structure tailoring^[188, 189] or integration with rubber matrix^[102] can also be employed. Copolymers of regioregular P3HT and polyethylene (PE) display outstanding mechanical toughness with elongations at break exceeding 600% and better mobility than P3HT.^[188] Relationship between stretchability and molecular structure can easily be investigated by a lamination method developed by Bao group.^[189] In addition to the mere molecular tailoring of the single-component active semiconductor materials, the addition of a secondary polymer to the high-mobility and stiff donor-acceptor polymer semiconductor can significantly increase the film ductility while maintaining charge

transport characteristics.^[190] The large increase in ductility is attributed to the vertical segregation observed in the film.

3.3.2 *Intrinsically Stretchable OFETs*

Fukuda and coworkers obtained fully solution-processed, high performance stretchable OFETs with mobility of over $1.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for single device and $0.80 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for OFET arrays.^[150] However, the low stretchability (<2% strain) is not desirable for practical applications. Bao group developed a highly stretchable transistors which exhibited transistor characteristics under 265% strain, by utilizing a microcracked P3HT layer inserted between two pre-strained elastic layers (**Figure 8**).^[103] One elastic layer is carbon nanotube (CNT)-based source/drain electrodes embedded in polyurethane (PU) elastomer, and the other elastic layer is PU dielectric layer. A stretchable liquid metal EGaIn serves as the top gate electrode. Using the rubber poly(styrene-*b*-butadiene-*b*-styrene) (SBS) fiber mat instead of elastic PDMS substrate ($\varepsilon = 0.4$) can much enhance the stretchability of the P3HT nanofibers (from $\varepsilon = 0.6$ to $\varepsilon > 0.9$).^[151] By using another two stretchable components—Au nanosheet electrodes and the polyelectrolyte gel gate dielectric, they obtained a transistor sustainable to 70% strain. The device exhibited a high hole mobility ($\mu = 18 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) at $\varepsilon = 0.7$ and showed excellent electrical stability over 1,500 stretching cycles. Recently, they reported a strategy to achieve ultra-stretchable materials with self-healing ability by embedding dual-strength dynamic metal-ligand coordination bonds within the soft PDMS polymer backbone.^[191] Based on this, a series of fully stretchable, hysteresis-free and high-performance n-type and p-type OFETs, using such self-healing cross-linkable elastomers, have been demonstrated.^[192] These devices exhibit good mechanical robustness and stable electrical performance with low gate leakage current, even after 1000 cycles at 100% strain. The hysteresis-free transfer characteristics are due to the low ion conductivity in PDMS and the strong columbic interaction between metal cations and the small Cl^- anions which can prevent mobile anions drifting under gate bias. Both the dielectric properties of various gate

dielectric elastomers and the mobility of thin film transistors can easily be quantified by a charging measurement technique using a resistor–capacitor circuit.^[193] Further investigation found that the in-situ phase separation of the self-assembled P3HT nanofibril bundles with indentation into the surface of the polystyrene block-poly(ethylene-co-butylene)-block-polystyrene (SEBS) rubber matrix endows much higher stretchability (with crack onset strain of $\varepsilon = 0.7$) than unbundled nanofibrils on SEBS surface (with crack onset strain of $\varepsilon = 0.3$).^[102] A top-gated fully-stretchable transistor ($\varepsilon = 0.5$) was fabricated by employing all stretchable elements—stretchable liquid metal EGaIn gate electrode, SEBS rubber dielectric layer, Au nanosheet S-D electrodes, and P3HT/rubber (SEBS) composite channel semiconductor. The device performance and stretchability of some typical stretchable OFETs are summarized in **Table 1**.

In general, the OFET matrixes with stretchable interconnectors and buckled OFETs have advantages of using conventional or previously reported high-mobility channel materials and directly applying high-performance unstretchable device structures, but have intrinsic disadvantages of delamination after many stretching cycles due to difference in elastic modulus between different parts or layers. In addition, the high fabrication cost remains as another challenge. Adopting intrinsically stretchable OFETs seems to be an effective way to overcome these problems. However, development of efficient channel and dielectric materials with both high performance and high elasticity still needs vast investigations in material design communities.

4. Stretchable Organic Solar Cells (OSCs)

An OSC is a device that converts solar energy into direct current electricity using organic photovoltaic semiconducting materials for light absorption and charge transport to produce electricity from sunlight. When these materials absorb a photon, an excited state which can be regarded as an exciton or an electron-hole pair, is created and confined to a

molecule or a region of a polymer chain. Then, by effective fields set up by creating a heterojunction between two dissimilar materials of donor and acceptor, the excitons are dissociated into free electrons and holes with directional migration, thus causing the current electricity. OSCs indicate great potentials in low-cost, large area and flexible photovoltaic applications.

4.1 Buckled OSCs

Using the similar buckling strategy applied in OFETs, Bao group developed a stretchable OSC which can reversibly accommodate strain up to 27%.^[101] Using the liquid metal EGaIn as electrodes, they fabricated the device on a pre-strained PDMS substrate. After releasing from the stretch, the buckled device is stretchable almost without decline in performance under strain less than the pre-strained stretch. The buckling of P3HT:PCBM/PEDOT:PSS films can obviously be observed in their optical images. Kaltenbrunner *et al* constructed a P3HT:PCBM-based OSC on the pre-strained ultrathin (1.4 μm -thick) PET foil. The device exhibits a total thickness of only 1.9 μm and unprecedented lightweight (4 g m⁻²), with nearly identical power conversion efficiency (PCE, 4.2%) to their glass-based counterparts. After releasing, the resulting buckled device can reversibly withstand extreme mechanical deformation ($R_b = 35 \mu\text{m}$), and remains operational under reversible alternation between 80% compression and 400% tensile strain (**Figure 9**).^[114] The cycling test showed a gradual decrease in the short-circuit current (I_{SC}), the fill factor (FF) and the output power, resulting in a 27% decrease in power after 22 cycles. Further by employing the organolead halide perovskites as the photoactive layer, they improved the PCE as high as 12%. This buckled perovskite solar cells can withstand both uniaxial and radial large stretch/compression.^[147]

4.2 Spring- or Mesh-Shaped OSC fibers

For the fiber-like shaped OSCs, the configuration design into spring or mesh shapes can effectively make the devices durable to stretching. Peng *et al* reported an OSC fiber by fabricating a multilayer spring, consisting of Ti wire core as cathode, TiO₂ as electron transporting layer, P3HT:PCBM as photoactive layer and PEDOT:PSS as hole transporting layer, around an elastic rubber fiber and finally covered by a SWCNT sheet (**Figure 10a**).^[149] The fiber-shaped OSCs exhibit only a small decrease of PCE (~ 3%) after undergoing 100 cycles of 30% stretch, and the flexible and bendable textile fabricated by these OSC fibers maintains 90% of PCE after 50 cycles of 30% stretch (**Figure 10b**). These OSC fibers indicate great potentials for wearable electronics. The stretchable OSC constructed on a mesh-like ITO-coated PUA substrate, is reported to exhibit little change in performance after transferring onto a hemispherical surface.^[142]

4.3 Intrinsically Stretchable OSCs

4.3.1 Stretchable transparent electrodes

To fabricate intrinsically stretched OSCs, it is required that both the transparent, conductive electrodes and the photoactive layer can withstand high stretch. Liquid metals or other ductile metals can be used for the non-transparent cathodes. For stretchable transparent, flexible and conductive (TFC) anodes, the commonly used electrodes in OSCs are PEDOT:PSS-based electrodes. However, the PEDOT:PSS electrodes present a number of problems such as relatively low conductivity, low adhesion to hydrophobic substrates, and low stretchability causing decreased conductivity at large strains. These properties can be improved by using some additives — i.e., dimethylsulfoxide (DMSO), Zonyl fluorosurfactant (Zonyl), poly(ethyleneimine) (PEI), and Triton X-100.^[194, 195] After adding a small amount (~1%) of Zonyl, the R_s of film increases by a factor of only two at 50% strain and retains significant conductivity up to 188% strain.^[111, 112] The buckled films deposited on a pre-strained PDMS substrate are reversibly stretchable with almost no change in R_s during the course of over 5000 cycles of 0 to 10% strain. Significant cracking occurs at 30% uniaxial

strain, beyond which the films are not reversibly stretchable. The decrease of conductivity caused by Zonyl additive can be offset by depositing multilayers, to reach comparable device performance to the ITO-electrode-based counterparts.^[111] Moreover, the Zonyl additive renders spin-coating of PEDOT:PSS amenable for deposition on hydrophobic surfaces, including both pre-deposited, annealed films of PEDOT:PSS (enabling the deposition of thick, highly conductive, multilayer films) and stretchable PDMS substrates (enabling stretchable electronics). Jeong *et al* produced a stretchable and highly conductive PEDOT:PSS thin-film coating on hydrophobic substrates, simply by adding a nonionic surfactant (Triton X-100) to an aqueous PEDOT:PSS solution.^[195] This surfactant significantly reduces the surface energy of the PEDOT:PSS solution, which permits conformal coatings of PEDOT:PSS thin film on a diverse range of hydrophobic substrates, and induces the formation of PEDOT nanofibrils during coating, which leads to the high conductivity and mechanical stability at larger strains ($\epsilon = 10.3\%$). Buriak group reported a spray coated high-conductivity ethylene glycol (EG)-modified PEDOT:PSS transparent electrode whose R_s increases 30% at 20% strain and 100% at 100% strain.^[113] The brush-painted CNT electrodes with R_s increase of $\sim 3\%$ at 5.5% strain,^[196] and stretchable TFC electrodes based on a ITO-coated on PUA mesh with R_s increase of $<2\%$ at 70% strain have also been demonstrated.^[142]

4.3.2 Stretchable Active Layers

The photoactive layer in OSCs is generally a heterojunction composite blend of electron donor and electron acceptor materials, which induces photo charge separation for generating current. The elastic properties are determined by the component molecular structures, doping ratios and even post treatment methods. P3HT:PCBM blends are usually applied as photoactive materials. However, Lipomi group found that the tensile modulus of P3HT, a polymer of alkyl substituted thiophene monomer, is much enhanced after blending with PCBM, consistent with the enlarged buckling intervals in their films.^[153] By increasing the length of the alkyl substitute (C_nH_{2n+1}) on thiophene monomers, this detriment to the

elasticity in blends could be much reduced (**Figure 11a**). Moreover, increase of the length of these alkyl side chains also benefits for reducing the fragility of pristine poly(3-alkylthiophenes) (P3AT) films. The on-crack strain is 6% for P3BT ($n=4$) and 60% for P3OT ($n=8$). However, there is an offset between the elasticity of active layer and device efficiency. The inherent competition between deformability and charge transport arises because the properties needed for high carrier mobilities — e.g., rigid chains in pi-conjugated polymers and high degrees of crystallinity in the solid state — are antithetical to deformability.^[197] For $n \leq 6$, the OSC exhibits higher PCE but poor elasticity and ductility. For $n \geq 8$, high elasticity and ductility are obtained, but with inferior PCE. The co-optimization of both PCE and elasticity was achieved based on P3HpT ($n=7$) polymers (**Figure 11b**).^[198, 199] Stretchability can be further enhanced by encapsulating the whole device in thermoplastic polyurethane which delocalizes strain around defects in the films.^[200] Although the tensile modulus can also be tuned by blending, copolymerization or block-polymerization of P3HT ($n=6$) with P3OT ($n=8$), these blends or polymers generally cannot achieve a higher PCE than P3HT-based OSCs. They further developed an approach to increase the elasticity of the blends with polymers by statistical incorporation of unlike monomers.^[104, 201] The donor–acceptor polymer DPPT-TT, whose repeat unit comprises of adiketopyrrolo-pyrrole moiety, thiophene and thienothiophene, has been synthesized. It is found that the elastic properties of DPPT-TT:PCBM film is significantly improved with comparison to P3HT:PCBM film.^[104] They further demonstrated a material design method to increase the mechanical resilience of semiconducting polymers for OSCs without having a deleterious effect on the photovoltaic properties. The alternating copolymer, PDPP2FT, comprising an N-alkylated diketopyrrolopyrrole (DPP) unit flanked by two furan rings (2F) alternating with thiophene (T), and the "segmented" polymer, PDPP2FT-seg-2T, in which the DPP is exchanged for a tail-to-tail coupled unit of two 3-hexylthiophene rings (bithiophene, 2T) in an average of one of approximately five repeat units, have been synthesized, respectively (**Figure 11c**).^[201] The

tensile modulus of the segmented polymer PDPP2FT-seg-2T (0.93 ± 0.16 GPa) is lower than that of the homopolymer PDPP2FT (2.17 ± 0.35 GPa). Meanwhile, when blended with PC₆₁BM, the segmented material produces devices with PCE of $2.82 \pm 0.28\%$, which is similar to that of PDPP2FT-based device ($2.52 \pm 0.34\%$). In addition, the stiffening after PCBM blending is much reduced, with improved PCE than the P3HT-based OSCs. In addition, blending ratio, film thickness and morphology and post-treatment also affect the elastic properties.^[202, 203] In blends of P3HT with either [60]PCBM or [70]PCBM, the stiffness decreases with the reducing PCBM doping ratios. And the thermally annealed blended films exhibit a higher tensile modulus than that of the as-cast films (**Figure 11d**).^[202] The mechanical properties of these organic semiconductors and their blends with C₆₀ can be predicted using coarse-grained molecular dynamics simulations.^[204] Other series of donor-acceptor low-band-gap polymers and their heterojunction blends have also been investigated to understand the relationship between molecular structures and mechanical properties.^[205, 206] Besides the modification of organic donors, using of organic acceptors instead of the traditional but brittle PCBM has been demonstrated as an effective way to enhance the mechanical robustness.^[207] The tensile modulus of the all-polymer PBDTTTPD:P(NDI2HD-T) heterojunction blend film was only 0.43 GPa, and its elongation at break was 7.16%, which was a 60-fold enhancement over that of PBDTTTPD:PCBM (1.76 GPa and 0.12%) (**Figure 11e, f**). In addition to the molecular structure, the film morphology and intermolecular packing modes also affect the device performance. For example, for the P3HT-based OSCs, the face-on stacking of P3HT has a positive correlation to the OSC open-circuit voltage (V_{oc}), attributed to a shift in the highest occupied molecular orbital (HOMO) energy level.^[208]

4.3.3 Intrinsically Stretchable OSCs

Using stretchable Zonyl-modified PEDOT:PSS as highly conductive anode, liquid metal EGaIn as top electrode/cathode, and various stretchable active layers, intrinsically stretchable OSCs can be fabricated. Lipomi *et al* applied DPPT-TT: PCBM as the active layer

with crack-onset strain of 6~12%, and fabricated stretchable OSCs which sustain 20% strain.^[104] The DPPT-TT:PCBM film exhibits a lower tensile modulus than that of the P3HT:PCBM film, which is demonstrated by the much fewer cracks in the film under the same strain. These devices show a narrower spread in properties due to cyclic stretching with strain from 0 to 10%. Based on the stretchable P3OT:PCBM active layer using elastic and ductile poly(3-octylthiophene) (P3OT), they fabricated stretchable OSCs which exhibit typical photovoltaic properties when bonded to hemispherical glass substrates with radius of 8 mm, with equivalent biaxial tensile strains of 24%.^[152] In contrast, the OSCs based on the relatively brittle poly(3-hexylthiophene) (P3HT) exhibit extensive cracking, which degrades the photovoltaic effect significantly. Using the longer side alkyl substituted P3DDT, the OSCs exhibit even higher PCE (0.381%) under 10% strain than the un-stretched ones (PCE=0.291%), while the P3HT:PCBM-based OSCs shows a significantly damaged device performance (PCE=0.008% under 10% strain, while PCE=0.594% under 0% strain).^[153] Consistently, compared with obvious cracks on the surface of P3HT:PCBM-based devices, almost no cracks are observed in the P3DDT:PCBM-based OSCs (**Figure 12**). **Kim group** developed highly efficient and mechanically robust all-polymer solar cells that are based on the PBDTTTPD polymer donor and the P(NDI2HD-T) polymer acceptor, which exhibit comparable high PCE of 6.64% to the control polymer-fullerene devices (6.12%) and afford greater tolerance to severe deformations with 60- and 470-fold improvements in elongation at break and toughness, respectively. The device performance of these stretchable OSCs has been summarized in **Table 2**.

To date, although the intrinsically stretchable OSCs have easier fabrication and overcome the inherent delamination problem of the buckled OSCs, they exhibit low stretchability which remains a big obstacle for practical application and potential commercialization. Therefore, design of highly elastic and high-performance heterojunction intrinsically stretchable materials remains a big challenge in this field.

5. Stretchable Organic Light-emitting Devices

Organic light-emitting devices mainly include OLEDs and OLECs, which utilized organic small-molecule or polymer as light emitting semiconductor materials. OLED is a light-emitting diode in which the electroluminescent emitting layer is a film of organic compound that emits light in response to an electric current. This organic semiconductor layer is sandwiched between two electrodes; generally, one electrode is transparent to escape the produced light. The hole- and electro- transporting layer are usually inserted between the active light-emitting layer and electrodes to facilitate the carrier transporting and enhance the device efficiency. OLEDs have been widely used in digital displays such as televisions, computer monitors and portable systems, and are highly prospective in next-generation of solid-state lighting technology.

OLEC is a solid-state device that generates light from an electric current, namely electroluminescence. OLECs are usually composed of two metal electrodes connected by (e.g. sandwiching) an organic semiconductor containing mobile ions. Unlike OLEDs, OLECs do not require low-work function cathodes, hole/electron transporting layers, or small roughness between the interfaces, and usually possess a simple device structure composed of two metal electrodes connected to an organic semiconductor.^[40, 209]

5.1 Buckled OLEDs

By adhering an ultrathin **polymer** based OLED, i.e., polymer light-emitting diodes (PLEDs) with total thickness of 2 μm onto a pre-strained elastomeric PDMS substrate followed by relaxing, the PLED forms a random network of folds to accommodate reversible strain and compression, as well as ultraflexibility with bending radius of only 10 μm (**Figure 13a, b**).^[116] Their extreme flexibility renders the devices fully stretch-compatible, with demonstrated cyclic tensile strains up to 100%. This value is limited by the prestrain of the supporting elastomer and not by the PLED itself. The current–voltage traces of an AnE-

PVstat-based PLED in the flat and 30% compressed state are virtually indistinguishable. Compression and stretching of this scale can be repeated many times without damage to the device. Moreover, it is demonstrated that the buckling structures with broad distribution and directional randomness can enhance the light extraction efficiency without introducing spectral changes and directionality, thus exhibiting improved current and power efficiencies.^[210] These buckled OLEDs are practical and attractive for use in fabricating highly flexible and efficient full color and white OLEDs. Recently, Sun group have developed a laser-programmable buckling process to realize a highly stretchable OLED with high device efficiency and mechanical robustness, which displays a current efficiency (CE) of 70 cdA⁻¹ under 70% strain.^[211] The highly regular bucking structure endows the device with excellent mechanical robustness which can accommodate 100% strain while exhibiting only small fluctuations in performance over 15,000 stretch-release cycles (**Figure 13c-f**).

5.2 Intrinsically Stretchable Organic Light-emitting Devices

5.2.1 Intrinsically Stretchable OLEDs

Pei group developed transparent SWNT/PtBA (a shape memory polymer with glass transition temperature of 56°C) composite electrodes, and obtained an intrinsically stretchable sky-blue PLED, that can be stretched up to 45% strain.^[110] The devices could be fully folded upon heating at 70°C for several cycles. After one folding–unfolding cycle with $R_b < 1$ mm, the CE dropped slightly to 1.16 cd A⁻¹ from original 1.24 cd A⁻¹ at 200 cd m⁻² while maintaining operating voltage almost unchanged. They further developed fully stretchable white PLEDs fabricated with graphene oxide (GO)-AgNW/PUA composites electrodes.^[154] These devices can be stretched up to 130% linear strain at room temperature and survive after 100 stretching cycles between 0 and 40% strain. Here, PUA is a copolymer of a siliconized urethane acrylate oligomer (UA) and an ethoxylated bisphenol A dimethacrylate (EBA). UA was chosen for the high transparency and excellent stretchability, and EBA for the good bonding force with AgNWs. And, GO was applied to solder the junctions between the

AgNWs in the composites, to further enhance the stretchability of the electrodes. During 100 cycles of stretching-relaxing between 0 and 20% strains, the GO-AgNW/PUA electrodes exhibit a slower resistance increase 2.3~2.7 times its initial value (2.9 ~3.3 times for the unsoldered but heat annealed counterparts).

5.2.2 Intrinsically Stretchable OLECs

With no requirement of thermal treatment, Pei designed an improved polymer-based OLEC, i.e., polymer light-emitting electrochemical cells (PLEC), sustainable up to 120% strain, by sandwiching an elastomeric light-emitting layer into the highly conductive and transparent AgNW-PUA composite electrodes (**Figure 14**).^[109] The fabrication process is facile and scalable. Strikingly, the device shows significantly improved efficiency in the stretched state. The CE shows a 200% increase from 1.0 cd A⁻¹ before stretching to 3.0 cd A⁻¹ at 40% strain. It levels off at up to 80% strain and then begins to decrease, but still retains a fairly high value of 2.1 cd A⁻¹ at 120% strain, which is still more than 100% higher than its original value. Another intrinsically stretchable OLEC, using all stretchable components—an elastic electroluminescent of Ru complex/PDMS blend, the Au/PDMS and EGaIn electrodes, has also been reported.^[108] The charge injection enhancement enables simple device architecture with no need of additional electron/hole- injection, transport, and blocking layers.

5.3 Stretchable OLED Displays

Using the printable, highly conductive and stretchable elastic conductors and pastes, Someya group fabricated a rubber-like stretchable active-matrix OLED display, comprising 16×16 driving cells with two organic transistors and one capacitor (2T1C), and OLEDs, which can be stretched by 30–50% and spread over a hemisphere without any mechanical or electrical damage (**Figure 15**).^[145] These printable elastic conductors, manufactured by uniformly dispersing single-walled carbon nanotubes (SWNTs) bundle networks into a fluorinated rubber by use of an ionic liquid and jet-milling, exhibit high conductivity of more than 100 S cm⁻¹ and stretchability of more than 100%, and did not show changes in

conductivity after 1,000 cycles of stretching by 70%. Bending or crumpling caused no mechanical or electrical damage of the displays because of the excellent conductivity and mechanical durability of the printed elastic conductors, organic transistors and OLEDs manufactured on the PDMS rubber. Moreover, the printing ability of the elastic conductors enables narrow linewidths of 10~100 μm for realizing higher resolution and miniaturization of display cells. Pei utilized the above mentioned highly elastic AgNW-PUA composite electrodes in cross-bar patterning, stretchable emitting and packaging layers and fabricated a simple foldable and stretchable passive matrix monochrome display featuring a 5×5 pixel array, which retains the rubbery elasticity of the individual intrinsically stretchable PLEC pixels.^[109] The device performance and stretchability of these stretchable light-emitting devices have been summarized in **Table 3**.

For stretchable OLECs, although exhibiting relatively slow turn-on and low efficiency, their simple device structures and easy processing make them potential candidates for cheap and disposable applications. However, to obtain high device performance, it is favorable to utilize OLED structures with more efficient and stable organic luminescent materials such as phosphorescent materials and thermally activated delay fluorescent materials. For further low-cost and easy fabrication of intrinsically stretchable devices, the material design of stretchable host materials and emitters are required.

6. Stretchable Organic Memory Devices (OMDs)

OMDs provide an ideal solution for inexpensive but high density and high performance applications. OMDs include organic resistors,^[212] OFET memories,^[77] and other complex-structured memories.^[213] To date, very few publications are reported about stretchable inorganic memory devices which are based on buckled structures^[214] and crosspoint arrays^[196]. Even fewer publications about stretchable OMDs have been reported.^[117, 215] Chen fabricated a stretchable organic resistive memory with a buckled

structure using a mechanically flexible and elastic graphene bottom electrode and stretchable P3BT:PMMA polymer semiconductor.^[117] The wrinkled memory device demonstrated electrical bistability with typical write-once-read-many times (WORM) memory features and a high ON/OFF current ratio ($\sim 10^5$) when stretched to 50% strain (**Figure 16**). Even after 500 stretching cycles from 0 to 30% strain, the stretchable OMD exhibited excellent electrical switching functions and memory effects. Another publication demonstrated non-volatile organic ferroelectric memory transistors fabricated using rigid PI islands on an elastomer substrate, which exhibit no noticeable change in electrical performance up to 50% strain.^[215]

However, the relatively low yield of OMDs, especially the organic resistive memristors, remains a big obstacle in practical high density memory applications. In addition, due to their high sensitivity to strain, it is difficult to realize intrinsically stretchable electrical memory devices, although they may have simpler fabrication process. Therefore, to develop high-performance stretchable OMDs, fatigue- and strain-resistant electrodes and memorable materials (the conductivity is little changed with strain) are highly desired.

7. Challenges and Outlook

Generally speaking, the organic semiconductor devices have advantage of intrinsic stretchability over their inorganic counterparts, as well as low cost of organic materials, the relaxed requirements of material purity, the minute thicknesses of material required (≤ 100 nm), and the relative ease concerning the solution processability. However, to date, for achieving desirable stretchable organic semiconductor devices, there still remains several challenges as follows: 1) For the fabrication process, a feasible fabrication method to reduce total manufacturing cost, a low-temperature process (e.g. solution process) to prevent deformation of plastic substrates, and a desirable stretchable platform or architecture that allows bending, folding or stretching are required. Employing the solution processable techniques, such as roll-to-roll coating, inkjet printing and screen printing, to fabricate low-cost

and large-area stretchable devices will facilitate the development of this field. 2) For the practical applications, high device efficiency to lower the power consumption, high mechanical and electrical stability to endure repetitive stretching, long-term reliability in ambient atmosphere (especially resistance to moisture and oxygen) to maintain fair device performance.

Current efforts are primarily aimed at improving the device efficiency and environmental stability to approach or even surpass their inorganic counterparts, in order to make these devices be widely adopted for practical and robust applications. Although the buckled devices can directly use the previously developed high-efficiency unstretchable thin-film devices, the complex fabrication causes high cost and makes it difficult for large-area manufacture because the prestrain procedure is not compatible with current printing techniques. In addition, the wave-like configuration has the intrinsic delamination problem during the cyclic stretching due to the large modulus difference between the soft substrate and the stiffer device structure layers, easily producing defects at the wave-peak and wave-valley areas, and is liable to cause failure of oxygen/water-resistant encapsulation layer. The stretchable matrix with rigid serpentine or soft elastic interconnectors also easily causes the crack formation both in the connection areas, due to the modulus difference between connected parts, and in the active device areas subjected to surrounding, although relatively small, strain forces. Therefore, intrinsically stretchable devices are highly attractive due to its easy fabrication and intimate effective encapsulation. In this field, development of high-efficiency and high-elasticity active organic semiconductor materials still needs vast investigations in the near future. The structure-property relationship, i.e. how the molecular structure, the intermolecular stacking mode and film morphology affect the functionalities such as mobility, carrier transporting/donating/accepting capability, frontier energy level, device performance and so on, should be studied systematically to provide rational guides for material design. Besides, stretchable encapsulating materials and techniques are worthy of

great attention in the scientific communities. Moreover, for expanding the application scope, multifunctional organic semiconductor devices with some other complementary properties such as integration with other functional devices, and high durability for application under extreme conditions such as high thermal and pressure stability, are expected.

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Figure 1. The development of the stretchable organic semiconductor devices (OLED: organic light-emitting diode, OLEC: organic light-emitting electrochemical cell; OFET: organic field effect transistor; OSC: organic solar cell; OMD: organic memory device). Stretchable OFET matrix (2008)^[80] (2015)^[141] Reproduced with permission from [ref. 80] and [ref. 141]. Copyright 2008 AAAS, and 2015 Nature Publishing Group, respectively; Stretchable AMOLED display (2009)^[145] Reproduced with permission from [ref. 145]. Copyright 2009 Nature Publishing Group; Intrinsically stretchable OSC (2011)^[101] Reproduced with permission from [ref. 101]. Copyright 2011 WILEY-VCH; Intrinsically stretchable OLED (2011)^[110] Reproduced with permission from [ref. 110]. Copyright 2011 WILEY-VCH; Intrinsically stretchable OLEC (2012)^[108] Reproduced with permission from [ref. 108]. Copyright 2011 WILEY-VCH; Stretchable & compressive OSC (2012)^[114] and perovskite SC (2015)^[147], Reproduced with permission from [ref. 114, 147]. Copyright 2012, 2015 Nature Publishing Group; Intrinsically stretchable OLEC (2013)^[109] Reproduced with permission from [ref. 109]. Copyright 2013 Nature Publishing Group; Stretchable & compressive OFET (2013)^[115] (2014)^[146] Reproduced with permission from [ref. 115, 146]. Copyright 2013, 2014 Nature Publishing Group; Stretchable & compressive OLED (2013)^[116] Reproduced with permission from [ref. 116]. Copyright 2013 Nature Publishing Group; Intrinsically stretchable OFET (2014)^[151] (2015)^[102] Reproduced with permission from [ref. 151, 102]. Copyright 2014, 2015 WILEY-VCH; Intrinsically stretchable OMD (2014)^[117] Reproduced with permission from [ref. 117]. Copyright 2014 Nature Publishing Group.

Figure 2. (a) Schematic illustration of a plastic mesh-like film with organic transistors and pressure-sensitive rubber, mechanically extendable by 25%. A magnified view of extended net-structures is also shown.^[128] Reproduced with permission from [ref. 128]. Copyright 2005 National Academy of Sciences. (b) Bottom gate OFETs fabricated on the PUA substrate. Inset is a photo image of the PUA substrate with the OFETs, after transference on to a plastic hemispherical surface.^[142] Reproduced with permission from [ref. 142]. Copyright 2015 WILEY-VCH.

Figure 3. (a) Left: Image of the curved surface of large-area stretchable active matrix, comprising 19×37 printed OFETs and wiring using the SWNT elastic conductor, covered with the stretchable active matrix. The printed OFETs function as active components, and the SWNT elastic conductor functions as word lines and bit lines for interconnection among the transistors; Right: Magnified image of one cell. A SWNT elastic conductor having a net-shaped structure was formed with a mechanical punching system and then coated with silicone rubbers. (b) Schematic illustrations of the stretchable active matrix. (c) Images captured in the initial state before stretching and under uniaxial and biaxial stretching. Under uniaxial (d) and biaxial (e) stretching, the transistor transfer characteristics (left) and channel currents (right) measured. The channel currents (I_{DS}) were normalized, with I_{DS} being measured before the experiments as the initial state.^[80] Reproduced with permission from [ref. 80]. Copyright 2008 AAAS.

Figure 4. (a) Illustrations of soft and stretchable OFET active matrix. Upper left: whole structure; Upper right, structure of an OFET cell on a rigid island with elastic conductor wiring; Lower: schematic structure of an OFET cell on the rigid island, the light green box corresponds to the entire OFET device. (b) Active matrix is relaxed (left) and stretched to about 60% (right). (c) Single stretchable transistor mobility (left, the red circles represent normalized mobility) and transfer characteristics (right) dependence on tensile strain. The inset shows the transistor relaxed (upper) and stretched to 100% (lower). Scale bars, 3 cm. (d) Mobility (left) and transfer characteristics (right) dependence on tensile strain of four transistors in a 2×2 stretchable active matrix. Scale bars, 1 cm. The inset shows the device in relaxed (upper) and stretched (110%, lower) configurations.^[141] Reproduced with permission from [ref. 141]. Copyright 2015 Nature Publishing Group.

Figure 5. Device performance of a buckled OFET on ultrathin PEN foil: (a) Normalized on-current (red circles), mobility (green diamonds) and threshold voltage (blue triangles) as functions of uniaxial compressive and tensile strains. No irreversible degradation was observed during the experiment. Inset: a transistor in relaxed (left) and compressed (right) states. (b) Transfer curves corresponding to (a), demonstrating the mechanical stability of the hybrid gate dielectric: no increase in gate leakage current was observed. (c) Normalized on-current (red circles), mobility (green diamonds) and threshold voltage (blue triangles) as functions of biaxial compressive and tensile strains. The inset shows the transistor in relaxed (left) and compressed (right) states. (d) Mechanical durability during repeated uniaxial 50% compression and re-stretching to 100% tensile strain. Plot of on-current (red circles), mobility (green diamonds) and threshold voltage (blue triangles) against stretch cycles.^[115] Reproduced with permission from [ref. 115]. Copyright 2013 Nature Publishing Group.

Figure 6. The strain-dependent characteristics of P3HT: (a) Illustration of the P3HT backbone stacking orientation under uniaxially strained films. I) Top view of a small portion of the polymer illustrating the semicrystalline nature of the polymer of randomly oriented crystals (dark red) within an amorphous matrix (light red). The black lines represent the polymer backbones; II) Oblique view of the polymer stacking direction illustrating an edge-on morphology, which is observed for an unstrained film. III) Under strain, the polymer crystals are reorienting with the polymer backbones aligning in the direction of applied strain, while the amorphous material remaining highly disordered. IV) Oblique view showing the increasing level of plane-on P3HT polymer in the alignment direction, while the crystals with the backbone initially in the direction of applied strain remaining edge-on; and (b) Saturated mobility for strain-aligned films with the transistor channel parallel and perpendicular to strain for 5 and 10 μm channel lengths (L).^[181] Reproduced with permission from [ref. 181]. Copyright 2011 WILEY-VCH. (c) Illustration of the P3HT backbone stacking orientation under biaxially strained films and the two-dimensional X-ray diffraction image of P3HT film under 100% $\times$ 100% biaxially strain; and (d) Saturated field-effect mobility as a function of channel length for the 100% $\times$ 100% biaxially strained films and unstrained films. Inset: Saturated field-effect mobility of films strained by 100% in the first direction and strained to varying extents in the second strain direction for 5 μm channel length devices.^[183] Reproduced with permission from [ref. 183]. Copyright 2013 American Chemical Society.

Figure 7. (a) Charge carrier mobility of P3HT polymers as a function of RR determined from transfer curves at $V_{DS} = -100$ V (bottom-gate, top-contact configuration with OTS-treated 300 nm SiO₂/Si wafer). (b) Tensile modulus and elongation at break of the P3HT thin films as a function of RR. ^[186] Reproduced with permission from [ref. 186]. Copyright 2015 American Chemical Society.

Figure 8. (a) Schematic device structure of stretchable OFET based on microcracked organic P3HT semiconductors; (b) Microscope image of a device with zero and 150% applied strain, respectively. (c) Morphology of PU/P3HT-t, PU/P3HT-s, and PU/P3HT/PU films under 200% strain values. PU/P3HT-t: P3HT transferred onto a PU substrate; PU/P3HT-s: PU spin-coated onto P3HT and then transferred onto a PU substrate; PU/P3HT/PU: PU/P3HT-t with a PU dielectric spin-coated on top. (d) Transfer curves for a device stretched to 160% and released to 20%. The source drain bias was -20 V and the W/L is 40. (e-f) Electrical characteristics of devices with stretching: (e) On and off currents for devices stretched parallel and perpendicular to the charge transport direction; (f) Mobility and leakage currents with strain. ^[103] Reproduced with permission from [ref. 103]. Copyright 2014 WILEY-VCH.

Figure 9. Stretchable and compressible 2- μm -thick OSC on ultrathin PET substrate: (a) Scheme of the ultra-light and flexible organic solar cell. (b) Extreme bending flexibility demonstrated by wrapping a solar cell around a 35- μm -radius human hair. Scale bar (also in c) 2 mm. (c) Stretchable solar cells made by attaching the ultrathin solar cell to a pre-stretched PDMS elastomer, shown at 50% quasi-linear compression. (d) I-V curves during compression on pre-stretched elastomer. The solid black line represents 0% and the purple line represents 80% compression with the intermediate steps of 10% distributed between accordingly. The dashed black line represents the device after re-stretching from 80% compression to its initial flat state. (e) Normalized device parameters under 0–80% compression, which correspond to 400–0% tensile strain or five times change in size.^[114] Reproduced with permission from [ref. 114]. Copyright 2012 Nature Publishing Group.

Figure 10. Stretchable OSC fibers and textiles: (a) Schematic illustration of the fabrication of a stretchable PSC fiber with a spring-like inner structure; (b) dependence of energy-conversion efficiency of a stretchable PSC textile integrated into clothes on the stretching cycles with 30% strain (the inserted image schematically shows the stretching process).^[149] Reproduced with permission from [ref. 149]. Copyright 2015 WILEY-VCH.

Figure 11. (a) The tensile modulus of the P3ATs and P3AT:PCBM blends as a function of alkyl side-chain length,^[153] Reproduced with permission from [ref. 153]. Copyright 2014 WILEY-VCH. (b, c) Plot of PCE of OSCs based on active layer of polymer:PC₆₁BM blends vs tensile moduli of (b) the corresponding P3AT-based pure polymers^[198], Reproduced with permission from [ref. 198]. Copyright 2014 American Chemical Society, and (c) the blends of PC₆₁BM:synthesized polymers,^[201] Reproduced with permission from [ref. 201]. Copyright 2014 Royal Society of Chemistry. (d) quantitative summary of the effect of mixed grades of methanofullerenes (AC: as-cast; AN: annealed) on the mechanical properties of P3HT:methanofullerene blends,^[202] Reproduced with permission from [ref. 202]. Copyright 2015 American Chemical Society. (e, f) tensile test of PBDTTTPD:PCBM and PBDTTTPD:P(NDI2HD-T) blend films. (e) strain–stress curves and (f) toughness of PBDTTTPD:PCBM and PBDTTTPD:P(NDI2HD-T) blend films. (The inset in e shows photographs of the bulk-heterojunction blend film floating on water. The specimens were gripped by the PDMS-coated Al grips and the films were prepared under the optimized device condition).^[207] Reproduced with permission from [ref. 207]. Copyright 2015 Nature Publishing Group.

Figure 12. Photographs of (a) P3HT:PCBM and (b) P3DDT:PCBM devices under 10% strain; characteristic photovoltaic properties of (c) P3HT:PCBM and (d) P3DDT:PCBM devices; the insets are optical micrographs of the surfaces of the devices, the scale bar is 0.5 cm.^[153] Reproduced with permission from [ref. 153]. Copyright 2014 WILEY-VCH.

Figure 13. (a,b) Demonstrations of extreme deformation attainable with ultrathin PLEDs. (a) Images of a free-standing ultrathin PLED operating during crumpling. The device is suspended between two pieces of glass that are moved closer together and simultaneously twisted by 90°. (b) Images of an ultrathin PLED adhered to a prestrained elastomer tape. The images from left to right represent the wrinkled PLED (unstrained elastomer) state being stretched to the flat PLED (stretched elastomer) state. Each pixel is $\sim 3 \text{ mm} \times 6 \text{ mm}$.^[116] Reproduced with permission from [ref. 116]. Copyright 2013 Nature Publishing Group. (c-f) High-efficiency laser-programmable buckled OLEDs. (c) Fabrication process: attaching the OLED/polymer film onto the prestretched elastomeric substrate with long-period gratings by laser ablation, and releasing the prestrains and obtaining OLEDs with ordered buckles. (d) Photographs of the stretchable OLED mounted on a bent finger joint. (e) Current efficiency-driving voltage characteristics of stretchable and planar devices. (f) Normalized luminance and current efficiency vs the number of stretch-release cycles for the stretchable OLEDs at 5V between 0% and 40% (b) strain.^[211] Reproduced with permission from [ref. 211]. Copyright 2016 Nature Publishing Group.

Figure 14. Device characterization of an intrinsically stretchable polymer-based OLEC (original emission area of $3.0 \times 7.0 \text{ mm}^2$): (a) Current density–luminance–driving voltage characteristics of an elastomeric PLEC device. Inset is an image of a PLEC (biased at 12 V) wrapped around the edge of 400-mm-thick cardboard. (b) Current efficiency–luminance characteristics of the device. Insets: photographs of the PLEC unbiased, biased at 12 V, and deformed to show light emission from both surfaces (from left to right). (c) Current density and luminance characteristics of a PLEC device at 12 V with increasing strain. (d) Current efficiency characteristics of the device with strain. All measurements were carried out at room temperature.^[109] Reproduced with permission from [ref. 109]. Copyright 2013 Nature Publishing Group.

Figure 15. Luminance of a stretchable display. (a) Luminance of a display cell as a function of V_{Scan} . Luminance was investigated for three configurations of display cell: 1) an OLED integrated with a 2T1C driving cell and electrical wirings made of 1-μm-thick Cu; 2) an OLED integrated with a 2T1C driving cell and electrical wirings made of printed elastic conductors (conductivity, 50 S cm⁻¹; length, 100 mm; linewidth, 500 μm; thickness, 100 μm); 3) an OLED integrated with a 2T1C driving cell and electrical wirings made of conventional conducting rubber (conductivity, 0.1 S cm⁻¹; length, 100 mm; linewidth, 1 mm; thickness, 100 μm). V_{Bias} and V_{Data} were -30V and -40 V, respectively. I_{OLED} and luminance are also shown. The effective size of the light-emitting area was 3×4 mm². (b) A demonstration of a stretchable display that can be spread over arbitrary curved surfaces. The stretchable display is functional even when folded in two or crumpled, indicating excellent durability.^[145] Reproduced with permission from [ref. 145]. Copyright 2009 Nature Publishing Group.

Figure 16. (a, b) Stretchable buckled organic resistive memory device. (a) Images of the device under 50% strain, and (b) I-V curves of the device under different post-strain.^[117] Reproduced with permission from [ref. 117]. Copyright 2014 Nature Publishing Group. (c, d) Stretchable cross-point organic ferroelectric memory transistors. (c) Images of the device fabricated on a rigid PI island/PDMS elastomer structure under 50% strain, and (d) transfer curves of the device under substrate stretching (0 to 50%).^[215] Reproduced with permission from [ref. 215]. Copyright 2016 Royal Society of Chemistry.

Table 1. The device performance of stretchable OFETs

Fabrication strategy	Active channel material	Field-effect mobility [cm ² V ⁻¹ S ⁻¹]	ON/OFF ratio	Operating voltage [V]	Tolerable maximum strain[%]	Ref
OFET Matrixes composed of rigid devices with stretchable interconnects	Pentacene	0.11	8.6×10 ³	-5	13	[144]
	Pentacene	0.5	10 ⁴ ~10 ⁶	-10 ~-5	70	[80]
	DNTT	1.8	—[a]	—[a]	110	[141]
Buckled OFETs	DNTT	1.6~3	>107	3	233	[115]
	Merck Iisicon S1200	1	106	10	100	[146][b]
Intrinsically stretchable OFETs	P3HT	(3.4±1.6)×10 ⁻²	591 ~461	—[a]	265	[103]
	P3HT nanofibers	18~22	105	-3.0 ~ -2.7	70	[151]
	tetrathienoacene-diketopyrrolopyrrole based polymers	0.19 ± 0.03	-15±2	5.9×10 ³	100	[192]

[a] not available; [b] fully printed devices.

Table 2. The device performance of stretchable OSCs (PCE: powder conversion efficiency).

Fabrication strategy	Active heterojunction material	PCE [%]	Tolerable maximum strain[%]	Ref
Buckled OSCs	P3HT:PCBM	2	27	[101]
	P3HT:PCBM	4.2	300	[114]
	Perovskite:PCBM[a]	12	100	[147]
Fiber-shaped OSCs	P3HT:PCBM	1.2	40	[149]
Intrinsically stretchable OSCs	P3DDT:PCBM	0.3-0.4	10	[153]
	PBDTTPD:P(NDI2HD-T)	6.6	7	[207]

[a] Perovskite, methyl ammonium lead trihalide(CH₃NH₃PbI_{3-x}Cl_x), is actually a hybrid organic-inorganic lead halide-based material.

Table 3. The device performance of stretchable OLEDs and OLECs (CE: current efficiency; PE: powder efficiency).

Fabrication strategy	Active luminescent layer	CE [cd/A]	Tolerable maximum strain[%]	Ref
Buckled OLEDs	AnE-PVstat	0.17±0.06	100	[116]
	Ir(ppy) ₃	70	100	[211]
Intrinsically stretchable OLEDs	PF-B/PEO-DMA/LiFf	1.24	45	[110]
	a white-light-emitting polymer [a]: OXD-7	2.0	130	[154]
Intrinsically stretchable OLECs	Ru(dtb-bpy) ₃ (PF6) ₂ /PDMS	—[b]	27	[108]
	Ru(dtb-bpy) ₃ (PF6) ₂ /PMMA	—[c]	17	
	SuperYellow/ETPTA/PEO/LiFf	11.4	120	[109]

[a] obtained from the authors' collaborators; [b] EQE = 0.16%; [c] EQE = 0.4%.

((Author bio(s), max. 75 words))

Author Photograph(s) ((40 mm broad, 50 mm high))

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The Table of Contents

Stretchable organic semiconductor devices are essential for development of low-cost, large area collapsible and portable electronics, wearable electronics, epidermal and bio-implanted electronics, 3-D surface compliant devices, bionics, prosthesis and robotics. Great advances have been made in this field recent years, which are summarized in this review, focusing on stretchable organic field effect transistors, light-emitting devices, solar cells and memories.

Keywords: Stretchable electronics, organic field effect transistors, organic light-emitting devices, organic solar cells, organic memory devices

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Stretchable Organic Semiconductor Devices

TOC figure