

Kirigami-Inspired Stretchable Conjugated Electronics

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 π -Conjugated polymers have drawn broad interest in flexible electronics due to their solution processability, light weight, and a combination of conducting and light-emitting properties. However, achieving mechanical endurance and stretchability in freestanding conjugated polymers is still difficult. Surface-assembly-induced light-emitting polymer nanosheets with prodigious mechanical strength and charge transport are reported. Transferring freestanding polymer films onto various templates with conformal contact results in electrical and optical strain sensors with a gauge factor of \approx 29. Subsequent geometric engineering into kirigami structures of the polymer sheets further extends the strain accommodations 20-fold without compromising electric conductivity or fluorescence properties. These as-prepared semiconducting polymers represent a possible new material for emerging stretchable electronics.

1. Introduction

Flexible and stretchable electronics with high mechanical strength are desirable for next-generation strain sensors,^[1,2] stretchable energy harvesters,^[3,4] artificial skins,^[5,6] humanactivity monitoring, and personal healthcare systems,^[7–10] etc. Among the flexible and stretchable materials, such as conductors,^[11,12] semiconductors,^[13,14] and insulators,^[15,16] π -conjugated semiconducting polymers have raised significant

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interest due to their low cost, abundance, lightweight, and solution processability.^[17,18] Stretchable polymer conductors have been applied to the interconnects. electrodes, and piezoresistive sensors.^[19] The conducting nature of π -conjugated polymers enables the strain-dependent electronic and capacitance sensor. Its light-emitting nature facilitates its optical strain sensing due to the emission change under strain, which is unaccessible in the conductor and insulator materials. The semiconducting materials used to form stretchable components are typically achieved by transferring the films onto the soft stretchable substrate. Random polymer chain distribution with defects would exist, and a significant amount of cracks could occur during the transferring

process. The geometric engineering has been applied to simultaneously achieve high stretchability and conductivity of conjugated polymers.^[20-22] In this context, the kirigami, which refers to cutting materials into certain patterns to acquire desirable structures, has inspired the programmable stretchable electronics.^[23,24] The kirigami structures reveal excellent potential to achieve the desired stretchable electronic properties. Here, we report a liquid/air assembly to grow large-area freestanding poly2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene (MEH-PPV) nanosheets, while their unique electronic and light-emitting properties arise from the electron coupling strength between π -stacking units and further high degree of molecular packing arrangements. The freestanding MEH-PPV nanosheets at the water-air interface ensure the conformal contact onto various surfaces. These contribute to the development of conformal sensors based on the sensitivity of band gap to π - π stacking. Furthermore, the geometric engineering with kirigami structures on the freestanding MEH-PPV films not only extends its stretchability but also sustains its conductive and light-emitting properties, which significantly enhance its stretchable applications.

2. Results and Discussions

The MEH-PPV is a light-emitting conducting polymer, which makes it a good candidate in optoelectronics.^[25,26] The free-standing and mechanically robust MEH-PPV nanosheets are assembled on a water–air interface in the ambient environment condition (**Figure 1**a–c). The MEH-PPV solution spreads throughout the water surface spontaneously, if the spreading







Figure 1. The self-assembly, transferring, and structure characteristics of freestanding MEH-PPV nanosheets. a–c) Schematic illustration of selfassembly of MEH-PPV nanosheets at the water–air interface. d) The substrate transferring processing method for MEH-PPV nanosheets. e,f) The transferred nanosheets on the flexible substrates, such as polyethylene terephthalate (PET) and PDMS. g) Freestanding MEH-PPV film with high photofluorescence under 365 nm UV light excitation in the inset. Typical h) optical, i) TEM images of MEH-PPV nanosheets show the flat, compact, and continuous morphology. j) X-ray diffraction patterns of self-assembled MEH-PPV nanosheets and spun-cast control films.

coefficient is positive, $S = \gamma_{bv} - \gamma_{tv} - \gamma_{tb}$, where the surface tension of the water substrate γ_{bv} is larger than the sum of the surface tension between water and the surrounding vapor, γ_{tv} and the interfacial tension between the spreading solution and water $\gamma_{\rm tb}$ ^[27,28] When the solvent is gradually evaporated, the MEH-PPV chains aggregate together in the lateral direction due to the aromatic interactions and hydrophobic interactions. The further evaporation of the solvent would consume the remaining precursor, leading to the nanosheet growth. The freestanding nanosheet film spreading on water surface can be readily transferred onto different types of flexible substrates (polyethylene terephthalate [PET], paper, aluminum foil, polydimethylsiloxane [PDMS], etc.; Figure 1d-f and Figure S1, Supporting Information). In addition, the freestanding MEH-PPV nanosheets can be picked up directly without the substrate, stemming from its high mechanical strength, which facilities its applications in the substrate-free devices (Figure 1g and Figure S2, Supporting Information). High fluorescence is observed in the freestanding MEH-PPV nanosheet (inset of Figure 1g).

The morphology of the as-prepared MEH-PPV nanosheets is examined by the optical and transmission electron microscopy (TEM), respectively. A large-area, smooth, and continuous thin film morphology is shown (Figure 1h,i), while the wrinkles on the film reveal its flexibility characteristic. The similar aberration chromatism could further confirm the uniformity of the MEH-PPV across the film in the optical image, as well as the chemical composition mapping image with a uniform elemental distribution (Figure S3, Supporting Information). The corresponding atomic force microscopy (AFM) image confirms the compact and smooth thin film morphology with a small surface roughness of ≈0.5 nm (Figure S5, Supporting Information) with an average thickness of 40 nm. Moreover, we performed X-ray diffraction (XRD) to get insight into the structure packing prepared by the interfacial assembly and the spuncast control films (Figure 1j). The spun-cast MEH-PPV films show the amorphous structure without distinct diffraction peaks, resulting from the random and staggered distribution of MEH-PPV chains. In comparison, two main diffraction peaks assigned to (100) and (200) planes could be found in the freestanding self-assembled nanosheets, which are attributed to the ordered MEH-PPV packing arrangement with strong aromatic interactions.

The thickness of MEH-PPV nanosheets from several to hundreds of nanometers could be controlled by the solution concentration (**Figure 2**a). The inset shows the MEH-PPV nanosheets with various thickness transferred onto the substrates. With increasing the average thickness from 8.0 to







Figure 2. Thickness control of MEH-PPV nanosheets and corresponding optical and mechanical properties. a) Concentration-dependent thickness film. The inset shows the transferred MEH-PPV nanosheets with different thickness. b) Absorption of MEH-PPV nanosheets with different thickness. c) Force–displacement curve of a \approx 400 nm thick MEH-PPV nanosheets. The inset is the ultra-fast extreme property mapping (XPM) applied on the surface of the nanosheets spanning an area of 5 × 5 μ m² with a 5 × 5 grid. d) Thickness-dependent Young's modulus of MEH-PPV nanosheets.

420 nm, the continuous MEH-PPV films changes from near transparency to crimson (the inset of Figure 2a and Figures S4 and S5, Supporting Information), sustaining the smooth surface and compact morphology shown in the optical images (the inset of Figure 2b). The photoabsorption also increases gradually with increasing the thickness of MEH-PPV films (Figure 2b). The nanoindentation is further employed to get the mechanical strength of MEH-PPV nanosheets by applying an external load through a nanoindenter tip at the sheet surface. Figure 2c describes the displacement-dependent loading force on a 400 nm thick MEH-PPV nanosheet. The measurements are conducted in the displacement-control mode in which the displacement is kept lower than 10% of the film thickness. The Young's modulus can be obtained from the force–displacement behavior as follows.^[29]

$$\frac{1}{E_{\rm r}} = \frac{(1-\nu^2)}{E} + \frac{(1-\nu_{\rm i}^2)}{E_{\rm i}}, E_{\rm r} = \frac{\sqrt{\pi}}{2\beta} \frac{S}{\sqrt{A}}$$
(1)

where *A* is the contact surface area at peak load, *S* is the initial unloading contact stiffness calculated using $S = \frac{dP}{dh}$, β_{is} an indenter geometry-dependent dimensionless parameter, E_r is the reduced modulus, E_i is Young's modulus of the indenter, *v*, v_i are the Poisson's ratio of the specimen and the indenter, respectively. An average Young's modulus of 4.2 GPa is obtained in the MEH-PPV nanosheets. The uniform force–displacement curves of MEH-PPV nanosheets suggest homogeneously distributed stress relaxation throughout the material. We further perform ultra-fast extreme property mapping (XPM)

on the MEH-PPV film to obtain the spatial distribution maps of the elastic modulus (the inset of Figure 2c). The measurement is applied on the surface spanning an area of $5 \times 5 \ \mu m^2$ with a 5×5 grid. The MEH-PPV nanosheets exhibit an average Young's modulus of 4.1 GPa with a small variation, which is larger than that of the spun-cast films (2.7 GPa; Figure S6, Supporting Information). The consistent nanoscale and bulk mechanical testing confirm the high mechanical strength and uniformity of self-assembled MEH-PPV nanosheets. The Young's modulus slightly increases with increasing thickness of the nanosheets (Figure 2d), mainly resulting from a higher crystallinity of packing regularity during the self-assembly process. The large-area flexible MEH-PPV nanosheets with enhanced mechanical strength facilitate its application in conformal strain electronics.

The current–voltage (*I–V*) curves of freestanding MEH-PPV nanosheets shown in **Figure 3**a exhibit a gradually increasing tendency of resistance with the increasing strain from 0% to 6%. The corresponding relative resistance change reaches 1.47 with the applied strain of 6% (Figure 3b). The decrease of its conductivity mainly results from the reduced electron coupling due to the strain-induced increase of the π – π stacking distance. A fundamental parameter to evaluate the strain sensor is its sensitivity to strain, defined as the gauge factor, which is the ratio of the fractional change in relative resistance change to the strain, presented as $\pi^{\sigma} = \frac{\Delta R}{R \times X}$, the ΔR and R are the resistance change and resistance without strain, and X is the strain.^[30] The freestanding MEH-PPV nanosheets exhibit a high gauge factor of \approx 26 with the strain of 6%, which is







Figure 3. Mechanoelectronic properties of MEH-PPV nanosheets and its conformal strain/pressure sensing. a) The current–voltage (I–V) curves of MEH-PPV nanosheets with various strains from 0% to 6%. b) Strain-dependent relative resistance change where the inset shows the corresponding conductivity change with various strain applied on the nanosheets. c) Conductivity stability testing when stretching and releasing on MEH-PPV nanosheets for 10⁴ times. d) Pressure-dependent relative current change where MEH-PPV nanosheets act like a conformal strain/pressure sensor to sense the pressure fluctuation when wrapping on an artificial blood vessel.

larger than most of the strain sensors based on the metal composite,^[31,32] graphene,^[33,34] carbon nanotube,^[35] etc. Stability is another parameter to evaluate the quality of a strain sensor. As shown in Figure 3c, the strain sensor based on MEH-PPV nanosheets exhibit a repeatable and reversible current on and off behavior for more than 10⁴ times by applying a cyclable strain from 0% to 6%. The as-prepared MEH-PPV nanosheets own enhanced flexibility and mechanical strength, which facilitate its conformal contact on various anomalistic-shaped targets, due to its excellent morphological adjustability. The freestanding MEH-PPV nanosheet is then wrapped in an artificial blood-vessel-like rubber tubing to monitor the pressure variation within the tube. Figure 3d shows the pressure-dependent current change ranging from 0 to 2000 Pa, where the inset is the repeatable ON/OFF current change with fast strain/ pressure response (<0.1 ms) by expanding or releasing the pressure in the tube. The high sensitivity, fast response time, and conformal adjustability characteristics of flexible MEH-PPV nanosheets are promising for the strain/pressure sensing application.^[36]

The light-emitting nature of MEH-PPV facilitates its wireless non-contact strain sensing due to the emission change under strain. The changes in optical properties under strain, such as fluorescence, are particularly advantageous because of the high sensitivity of fluorescence detection. Fluorescence emission, capable of providing in situ analysis through simultaneous optical excitation and detection, has been recognized as a promising mechanism to real-time monitor mechanical strain in materials.^[37,38] The optical property of MEH-PPV under various external strain is investigated under the 400 nm wavelength laser photoexcitation. As the strain increases, the corresponding fluorescence intensity decreases and reaches 20% at 6.0% of strain (**Figure 4**a). The fluorescence images of MEH-PPV under strain are presented in the inset of Figure 4a. We attribute such high sensitivity to the increase of structure disordering and π - π stacking distance under strain. In addition, the fluorescence could be reversed back to the pristine state after the strain release (red curve in Figure 4a and Figure S7, Supporting Information).

The photoexcitation induces the exciton formation in MEH-PPV nanosheets. Because the electron and hole carriers carry spin-1/2, these bound states can be spin singlets or triplets. Most of the charge carriers would recombine to induce the singlet fluorescence. While under the magnetic field, the spinconserving and spin-mixing competition modify the density of spin-parallel and spin-antiparallel states, which subsequently changes the photoluminescence intensity.[39-41] Figure 4b shows the fluorescence intensity change on the self-assembled MEH-PPV nanosheets when applied with external magnetic field steadily evolving from 0 to 4000 Oe. The increase of the light-emitting fluorescence intensity confirms the triplet to singlet conversion with fast response time (inset of Figure 4b). To get insight into the magnetic field effects, we further perform the magnetoconductance (MC) measurements to gather the relative current change before and after applying external magnetic field. Figure 4c demonstrates a negative MC under magnetic field, which stems from the scattering interaction between triplet state and polarons.[42-44]







Figure 4. Strain and magnetic modulation on the fluorescence of MEH-PPV nanosheets. a) Strain-dependent fluorescence change on the MEH-PPV nanosheets (black) and its recyclability testing (red) when gradually releasing the film. The inset shows the fluorescence image during the strain. b) Time-dependent fluorescence intensity with various magnetic fields of the MEH-PPV nanosheets. The inset shows the magnetic-field-dependent fluorescence intensity change from 0 to 4000 Oe. c) The magneto-conductance (MC) effect under different magnetic fields and voltage.

To achieve excellent performance for high load stretching and twisting applications, geometric engineering through the kirigami structure on MEH-PPV is applied to simultaneously achieve its stretchability, conductivity, and light-emitting stability. The kirigami-inspired MEH-PPV device is depicted in Figure 5a. The MEH-PPV is cut into different kirigami geometric parameters (Figure 5b), which refer to different maximum tensile strains. The maximum tensile strains are increased from 1000%, 1500% to 2000% when increasing the length-width ratio from 10:1 to 20:1 (Figure 5b and Figures S8-S10, Supporting Information). The kirigami films could be highly stretched, twisted, and bent (Figure 5c), indicating its excellent mechanical property. The kirigami structure with the maximum tensile strain of 2000% is selected for the following stretching studies. Figure 5d shows the stretching process of kirigami MEH-PPV films from 0% to 2000% strain under 365 nm UV excitation. No obvious deterioration of fluorescence intensity and the current density is observed (Figure 5e). Finiteelement modeling (Figure 5f) is applied to illustrate the sustained optical and electric signal throughout the kirigami structure, which shows the uniform strain distribution with nearly zero strain (less than 0.01%) in most area except the stress-concentrated region at cross-linking dissected area (the cracking failure susceptible area with the applied strain). While even at the cross-linking dissected area, no more than 3% strain is found. The detailed corresponding electric and optical signals are recorded in Figure 5g, showing minimal fluctuation, which is fit well with the finite-element modeling. Furthermore, kirigami electronics also shows high stability after stretching for

more than 10^4 times cycling (Figure 5h), indicating its feasibility for stretchable sensor applications.

3. Conclusion

In conclusion, large-area freestanding MEH-PPV nanosheets with advantageous electronic, mechanical, and light-emitting properties are presented through interfacial self-assembly method. Many of these unique features arise from the electron coupling strength between π -stacking units and a further high degree of molecular packing arrangement. The conformal electric/optical strain sensors exhibit high gauge factor of 29 using freestanding MEH-PPV nanosheets. The kirigami-inspired MEH-PPV structures enable stretchable electronics, on which the strain can reach 2000% without compromising the conductivity and light-emitting properties. The work presented here provides an advancement in the development of flexible strain sensors and wearable electronics.

4. Experimental Section

Self-Assembly of MEHPPV Nanosheets at Water-Gas Interface: The MEH-PPV nanosheets were grown on a toluene-water interface. The Petri dish containers were ultrasonic treated with toluene/acetone, isopropanol, and deionized water for 20 min, respectively, and then plasma treated for 20 min before using. After that, deionized water was filled in the Petri dish to half-height. When the solvent was stationary, 1 mL toluene with various solute concentrations of MEHPPV from 0.1

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Figure 5. Geometric engineering (kirigami structure) on MEH-PPV nanosheets and corresponding mechanoelectronic and fluorescence property. a) Scheme illustration of the kirigami cutting on the MEH-PPV nanosheets. b) Kirigami cutting on MEH-PPV nanosheets with different parameters, with the corresponding stretched state. c) The photos of kirigami-inspired MEH-PPV nanosheets in stretching, twisting, and bending states. d) The photos of kirigami MEH-PPV nanosheets with different stains under 365 nm UV excitation. e) The *I*–V curves of kirigami MEH-PPV nanosheets in the pristine, 2000% stretched, and 2000% stretched after 104 times states. f) Strain distribution simulation on kirigami MEH-PPV nanosheets. g) Strain-dependent conductivity and fluorescence change on kirigami MEH-PPV nanosheets. h) Strain stability testing on the kirigami MEH-PPV nanosheets.

to 15 g L⁻¹ was dropped on the water surface. The organic solvent was then spread throughout the water surface. Para-film was used to cover at the top of the Petri dish with several holes to control the evaporation rate and stability. Along with the slow evaporation of the solvent, the nanosheets spread gradually on the top of the water surface. The film was then transferred by picking up by various templates (PET, paper, aluminum foil, PDMS) for further use.

Morphology and Structure Characterization: The optical images were recorded by an Olympus BX51 microscope. SEM images were taken from FEI Quanta450FEG. The TEM images were taken with a JEOL JEM-1400. AFM height images were recorded on a Bruker dimension icon atomic force microscope equipped with a Cr/Pt-coated Si probe (radius < 25 nm, resonant frequency, 13 kHz). XRD analysis was carried out using a Rigaku CCD diffractometer with Cu-K α radiation (λ = 1.54184 Å). UV-vis spectra were recorded on an Agilent Model HP8453 UV-vis spectrophotometer.

Mechanoelectric and Optical Measurement: Different tensile strain to the samples was performed by Instron 5944 Single Column Tabletop Low-Force Universal Testing System. The current information was recorded with a CHI 422 Series Electrochemical Workstation linked to the device. The displacement–force curves and XPM images are recorded by Hysitron TI 980 TriboIndenter. Fluorescence was excited with 488 nm laser (or various magnetic fields) on a MEHPPV-coated PDMS substrate and collected with a Princeton Instruments ACTON SpectraPro 2500i/ Spec-10 spectrometer using a 20× objective.

Pressure Sensor Fabrication on an Artificial Blood Vessel: Due to the morphology adjustability, the freestanding electrode deposited MEH-PPV film without substrate can be directly wrapped (directly picked from water or wrapped from a freestanding film with the help of vaseline) on an elastic-rubber-tube-based artificial blood vessel. The pressure was precisely controlled by external gas flow from syringe. The stable pressure in the vessel was recorded by a linked barometer. The strain-induced current change was also recorded by CHI 422 Series Electrochemical Workstation.

Temperature and Pressure Sensor Fabrication on an Artificial Blood Vessel: Due to the morphology adjustability, the freestanding electrode deposited MEHPPV film without substrate can be directly wrapped on an elastic-rubber-tube-based artificial blood vessel with inner and outer diameter of 4 and 4.2 mm. The water flow was controlled by LKB BROMMA Pump LKB BROMMA 2120 Varioperpex. The temperature of the water was controlled by a conventional thermoelectric-based heating module. The current change of the sensor was recorded by CHI 422 Series Electrochemical Workstation when the heated and cooled water flew through the sensor (note that capacitance, dielectric constant, and www.advancedsciencenews.com

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polarization can all be used as the sensor parameters with different instruments). For the pressure measurement, the film was also wrapped on the artificial blood vessel while the pressure was precisely controlled by external gas flow from syringe. The stable pressure in the vessel was recorded by a linked barometer. The strain-induced current change was also recorded by CHI 422 Series Electrochemical Workstation.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

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