

Facile Fabrication of Stretchable Touch-Responsive Perovskite Light-Emitting Diodes Using Robust Stretchable Composite Electrodes

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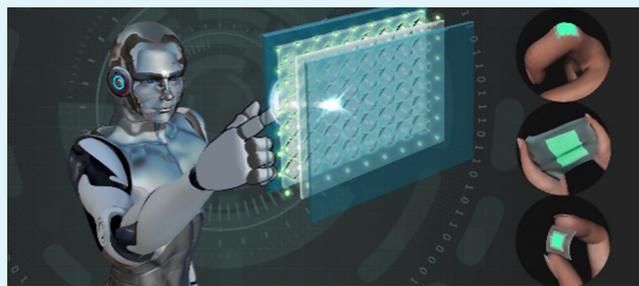
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ABSTRACT: Perovskite light-emitting diode (PeLED) has been vigorously developed in recent years. As it has demonstrated good performance on the rigid substrates, the next important direction of PeLED is its integration with stretchable components to realize stretchable, responsive device. Here, we describe a facile fabrication of stretchable perovskite light-emissive touch-responsive devices (PeLETDs) by utilizing highly transparent and conductive polyurethane/silver nanowires (PU/AgNWs) as the electrode. Meanwhile, a stretchable tricomposite perovskite emissive layer was developed by blending a small amount of poly(ethylene oxide) (PEO) and poly(vinylpyrrolidone) (PVP) with CsPbBr₃. Additionally, a thin PVP layer was introduced at the bottom of the emissive layer. On one hand, it can further improve the morphology of the emissive layer; on the other hand, it can serve as an electron-injection barrier to reduce the high nonradiative recombination at the corresponding interface. Further, to fulfill the responsive function of the fabricated PeLEDs, a poly(ethylene terephthalate) (PET) spacer with a 100 μm thickness was inserted between the top electrode and the emissive layer. A stretchable PeLETD is finally demonstrated to possess a low turn-on voltage of 2 V with a brightness of 380.5 cd m⁻² at 7.5 V and can sustain 30% uniaxial strain with a small luminance variation of 24%. More interestingly, our stretchable PeLETD exhibited high stability, which could be well touch responsivity, where the luminance is on/off switched for 300 cycles by repeatedly applying pressure.

KEYWORDS: perovskite, polymer, stretchable electrode, touch-responsive devices, light-emitting diode



1. INTRODUCTION

Stretchable optoelectronic devices have received significant attention recently owing to the advent of artificial intelligence (AI).^{1–4} In response to the growing enthusiasm toward wearable devices, such as bioinspired robotic and healthcare wearables, the development of flexible stretchable devices than can sustain high deformation and possess good adherent to irregular surfaces has become necessary.^{5–7} In addition, the stretchable devices possess much better portability because they can be folded into small volumes.

Recently, organic–inorganic perovskite has become a major research topic for its attractive optical properties, and its applications in solar cells and light-emitting diodes (LEDs) has progressed substantially in recent years.^{8–10} To date, versatile soft materials and transparent electrodes have been combined with the perovskite materials to realize stretchable LED devices. In 2009, Park et al.¹¹ first reported stretchable LED displays. Thereafter, a series of rigid organic LED and inorganic materials mounted onto a soft substrate and elastic conductors based on carbon nanotubes (CNTs) polymer or silver nanowires (AgNWs) polymer electrodes have been

reported.^{12–14} For instance, Yu et al.¹⁵ have fabricated perovskite LEDs using flexible CNTs/polymers as the bottom electrodes. The derived device could be strained to a curvature of a 5 mm radius without degrading the device performance. A similar concept has been used to prepare pressure sensor networks and solar panels in the meanwhile.^{16,17}

Researchers have also sought LEDs that can undergo stretching deformation.^{18–22} For instance, Li et al.²¹ have used solution blending to successfully fabricate a highly stretchable and conductive polymer film, which exhibits a 50% elongation and a conductivity of 172 S cm⁻¹, representing a significant breakthrough in developing polymers with high elasticity. Bade et al.¹⁵ subsequently reported an intrinsically stretchable LED based on polymer composites electrodes,

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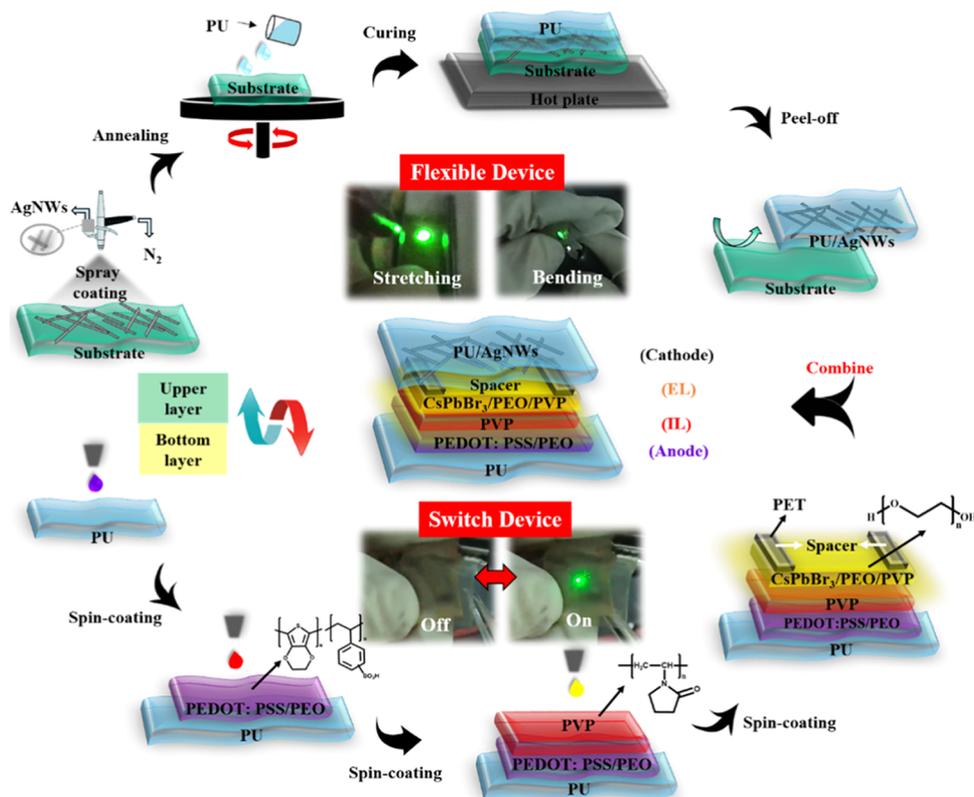


Figure 1. Schematic of the fabrication process of stretchable LETD. Photographs of stretchable LED and LETD under 5 V.

which could be stretched by 40% for 100 cycles without failure. Afterward, Yin et al.²³ fabricate an LED showing superior mechanical stability using a simple stencil-pattern transferring technology. The LED device exhibits an ultralow performance variation after 2000 stretch–release cycles between 0 and 20% tensile strain. More recently, Li et al.²⁴ employed an ultrathin and smooth AgNWs/polymide composite electrode to realize a stretchable LED showing a low turn-on voltage (3.2 V) and high luminance of 3187 cd m⁻² at 9 V. Moreover, it could accommodate stretching up to 50% tensile strain with high performance.

Thus far, several benchmarks achieved with versatile methods to present high-performance and stable stretchable LEDs.^{15,25–30} However, stretchable touch-responsive perovskite LEDs have been rarely developed and their production remains challenging because of the intrinsic instability of perovskite.^{17,31} Chou et al.³² have developed a touch-responsive perovskite LED showing an instantaneous visualization of pressure. The touch responsivity of the device is enabled by the integrated employments of a perovskite–polymer composite emissive layer and a polyurethane (PU) AgNW composite electrode. The device could be turned on because of the instantaneous formation of a Schottky contact and exhibits a low turn-on voltage of 2.5 V along with high luminance of 1030 cd m⁻² at 6 V. However, this device could not tolerate high deformation because of the rigid indium-tin-oxide (ITO)/poly(ethylene terephthalate) (PET) substrate. Another key advantage of stretchable, touch-responsive LED is its good integrability. For example, Kim et al.³³ integrated flexible quantum dot LEDs with wearable sensors and microcontrollers and reported high brightness of up to 44719 cd m⁻² at 9 V. Although the facile integrability is considerably useful, it must also be able to be driven by

complicated circuit devices.^{17,33,34} Therefore, developing a touch-responsive perovskite LED with a simplified and transparent structure is of great interest. The major challenge in realizing this that the transparent conductive electrodes and associated perovskite interfaces should have appropriate morphological features to avoid electron and hole leakages.

In this work, we prepare the first stretchable, perovskite light-emissive touch-responsive devices (PeLETDs) using a polymer/perovskite hybrid emissive layer and a transparent PU/AgNWs electrode. Our proposed PeLETDs is composed of an optimized composite film CsPbBr₃/poly(ethylene oxide) (PEO)/poly(vinylpyrrolidone) (PVP) (1:0.125:0.0225) as the emissive layer, a thin poly(vinylpyrrolidone) (PVP) layer as an electron-injection barrier, and PU/AgNWs as the flexible and transparent cathode. The PVP polymer could induce uniform morphology of perovskite layers and improve the stability of stretchable PeLETDs. Consequently, a stretchable, perovskite touch-sensitive LED with green-light emission is successfully fabricated, which exhibits a low turn-on voltage of 2 V with a brightness of 380.5 cd m⁻² at 7.5 V and can sustain 30% uniaxial strain without obvious changes in brightness. Moreover, the brightness of the devices remains stable after 300 repeated pressure–release cycles under a 20% strain.

2. RESULTS AND DISCUSSION

As illustrated in Figure 1, the key to realizing a stretchable, touch-responsive LED mainly relies on the proper design of the upper and bottom electrodes. We herein used a transparent PU/AgNWs composite electrode as the upper electrode to serve as the cathode. It was prepared by the transfer method to ensure the electrode has a smooth surface and decent mechanical strength.^{28,35} The prepared composite electrode exhibited a low sheet resistance ($\sim 10 \Omega \text{ sq}^{-1}$) and high

transparency (~89%; Figure S1a in the Supporting Information). Moreover, it could be reversibly stretched up to 100% strain without failure (Figure S1b). Furthermore, the PU/AgNWs could withstand 100 times 3M scotch tape adhesion-peeling without obvious changes in sheet resistance (Figure S1c). On the other hand, the high-conductivity poly(3,4-ethylenedioxythiophene)/polystyrene sulfonate PEDOT:PSS mixed with PEO (5 wt %) was adopted as the bottom electrode to serve as the anode and deposited onto the PU substrate. The PEDOT:PSS/PEO composite electrode displayed good conductivity and served as an elastic bridge for the emissive layers (Figure S1b). PEDOT:PSS/PEO composite was found to impart significant smooth surfaces,³⁶ and this composite is explored to have good strain-insensitive electrical conductivity, which is supported by the previously published literatures.^{15,37}

To enrich the device's touch responsivity without using any excessively complicated external electrical designs, a PET spacer was introduced between the upper electrode and the emissive layer to separate them. While efficient pressure was applied onto the PU/AgNW composite electrode, they will contact with each other to form an electrical connection and induce luminescent emission, thereby fulfilling the device's touch responsivity. Finally, a stretchable, touch-responsive LED can be successfully prepared by this integrated design using PEDOT:PSS/PEO and PU/AgNWs composite electrodes. Notably, our proposed device was fabricated through a full solution process and could be operated under ambient conditions without encapsulation, and its real-time application views are displayed in the inset of Figure 1.

Besides electrode engineering, the processing of a light-emitting perovskite layer also needs to be modified to fulfill its stretchability. A thin PVP layer was deposited onto the PEDOT:PSS/PEO electrode prior to the deposition of the perovskite layer to modulate the morphology and the surface roughness of the emissive layer. The hydrophilic feature of PVP (Figure S2) enables a better wetting of the perovskite solution to improve the film quality of the prepared perovskite layer. Figure S3a,b demonstrates the scanning electron microscopy (SEM) images of the perovskite layers deposited on the PEDOT:PSS/PEO electrode and the PVP-modified electrode. As seen, inserting a PVP intermediate layer at the electrode surface large improves the film quality of the perovskite. However, the overall film quality is still not satisfactory.

It is known the surface roughness of the emissive layer is a major influential factor governing the resulting performance. To further optimize the film quality, we further blend the perovskite with polymers. Li et al.³⁸ have proven that the surface morphology of the perovskite layer can be effectively modulated by the incorporation of PVP. PVP with high polarity could coordinate with Pb²⁺ through the lone pair of electrons of nitrogen atoms to form complexes, resulting in the uniform distribution of perovskite inside the polymer matrix. Besides PVP, the emissive layer was also blended with PEO to facilitate the smooth morphology and enhance the stretchability of the emissive layer. The morphological optimization was simply based on a CsPbBr₃/PEO ratio of 1:0.125, with further tuning with the PVP addition to harvest better surface coverage and smooth film surface. Furthermore, the entanglement of PEO chains may impart elasticity as well as mechanical toughness to enrich the stretchable characteristics of the derived emissive layer. To examine the effects of blending PVP

and PEO, the surface morphology of the prepared films was examined by SEM and atomic force microscopy (AFM), as presented in Figures S4 and S5. As seen, the coverage and morphology of the perovskite layer were considerably improved after blending with polymers, which is very important for reducing the current leakage of the derived devices. Figures S4e and S5e present the best film condition, for which the optimized weight ratio of CsPbBr₃, PEO, and PVP is 1:0.125:0.0225. The enhanced surface coverage of perovskite grains and defect passivation contributed to the enhanced photoluminescence quantum yield (PLQY) of 58% of the CsPbBr₃/PEO/PVP film, surpassing the value (5%) of the pure perovskite film, being similar to the case reported in the literature.³⁹ Besides, the electroluminescence (EL) spectra of the studied films are presented in Figure S6a and the International Commission on Illumination (CIE) x - y chromaticity diagram of the optimized emissive layer (CsPbBr₃/PEO/PVP = 1:0.125:0.0225) is shown in Figure S6b, demonstrating an ideal color-saturated green emission peak maximum at 524 nm with CIE color coordinates of (0.146, 0.778). These results manifest that PEO and PVP incorporation enhances the EL emission of the perovskite film, which is beneficial for achieving high-performance LED.

Figure S7 presents the time-resolved photoluminescence (TRPL) decay spectra of the prepared perovskite films. The TRPL curves were fitted as follows

$$A(t) = A_1 \exp\left(\frac{-t}{\tau_1}\right) + A_2 \exp\left(\frac{-t}{\tau_2}\right) \quad (1)$$

where τ_1 is associated with the trap-assisted recombination at grain boundaries or surfaces, τ_2 is ascribed to the radiative recombination inside the bulk perovskite phase, and A_1 and A_2 are the fractions of these two contributions. Detailed fitting parameters are listed in Table 1. As can be seen, the PL lifetime

Table 1. Detailed Parameters of Time-Resolved PL Measurements

different ratio	τ_1	A_1	τ_2	A_2	τ_{avg}
CsPbBr ₃	0.20	7116	5.48	3489	5.11
CsPbBr ₃ /PEO (1:0.125)	3.75	4304	15.06	4093	12.71
CsPbBr ₃ /PEO/PVP (1:0.125:0.012)	3.57	3711	13.10	4918	11.47
CsPbBr ₃ /PEO/PVP (1:0.125:0.0225)	3.01	4102	35.84	2186	31.37
CsPbBr ₃ /PEO/PVP (1:0.125:0.045)	3.29	6759	20.66	2384	15.26

of the perovskite film was considerably increased after blending with PVP/PEO, and it was further increased after optimizing the composition of the blends. The results indicated that the blending of polymers can effectively improve the crystallinity and quality of the perovskite film. Furthermore, the PVP layer inserted between the emissive layer and the anode could also form an electron-injection barrier to improve the charge-injection balance.

We next explore the efficiency of our proposed stretchable, touch-responsive LED, and Figure 2a shows the cross-sectional SEM of the device fabricated on ITO/glass without the top electrode, where the optimized thicknesses of the PEDOT:PSS/PEO, PVP, and CsPbBr₃/PEO/PVP layers were ~45, 3, and 300 nm, respectively. Figure 2b displays the images of the energy-dispersive X-ray spectroscopy (EDS) of the

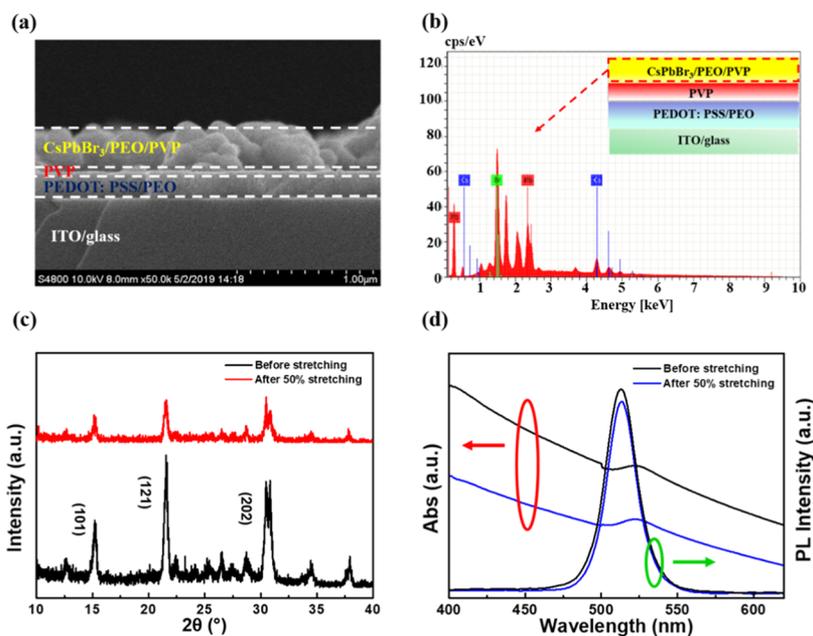


Figure 2. (a) Device structure of ITO-glass/PEDOT:PSS-PEO/PVP/CsPbBr₃-PEO-PVP. PEDOT:PSS was used as the anode. PVP was used as an insulating layer to improve the smooth perovskite morphology and interface defects. (b) Cross-sectional EDX image of the CsPbBr₃ perovskite. (c) XRD patterns of the CsPbBr₃/PEO/PVP (1:0.125:0.0225) composite thin films. (d) Photoluminescence (green) and absorbance spectra (blue) of the film with a CsPbBr₃/PEO/PVP (1:0.125:0.0225) sample.

hybrid emissive layer, wherein the presence of Cs, Pb, and Br elements confirms the existence of CsPbBr₃. The crystallinity of the composite films was analyzed through X-ray diffraction (XRD), as shown in Figure 2c. The result clearly indicates that the blending polymers did not affect the crystal structure of the perovskite, even under 50% strain. The UV-vis absorbance and PL spectroscopy of the film are shown in Figure 2d. It shows the PL maxima at 524 nm (grain size ~250 nm from Figure S4e), similar to the results reported in the literature.⁴⁰ The peak in the PL spectrum and the sharp transition in the absorption spectrum clearly appear at the wavelength of 524 nm for both pristine and 50% strain films. The full width at half-maximum (FWHM) of the PL peaks was 20 nm for both pristine and 50% strain films, suggesting the good stretchability of the emissive layer. Notably, the decreased intensity of the associated characteristics in XRD, UV-vis, and PL spectra under 50% strain might be attributed to the reduced film thickness in the stretched state.

As mentioned earlier, to fulfill the stretchability of our proposed LED, the bottom PEDOT:PSS/PEO composite electrode is prepared on a PU substrate and In-Ga is primarily used as the cathode, as illustrated in Figure 3a,b, which presents the luminance-strain characteristics of the fabricated LED. As shown, the device exhibited luminance of 1653 and 1258 cd m⁻² at 0 and 30% strain, respectively; that said, only a small variation of 24% was observed at 30% strain. However, the luminance was sharply decreased as the strain was increased to 50–70%.

To understand the changing dynamics of stretching device performance, we investigated the morphology of the perovskite film under various strains through field-emission SEM (FE-SEM). Figure 3c–h demonstrates that the luminance of the device is gradually decreased as the strain increases. Notably, cracks appeared at 60% strain, which resulted in a decreased device performance. Such morphological features impacted heavily in terms of a sharp decrease in luminance over 50–70%

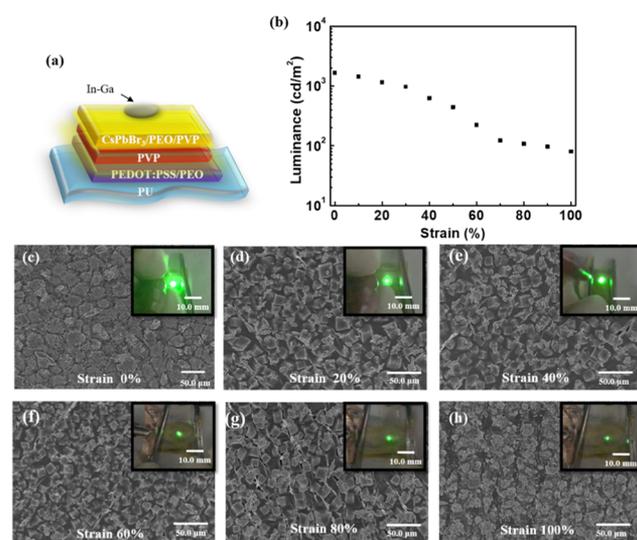


Figure 3. (a) Photographs of a complete stretchable perovskite LED with an In-Ga top electrode at 7.5 V and (b) luminance under a uniaxial strain from 0 to 100%. Uniaxial strains of (c) 0%, (d) 20%, (e) 40%, (f) 60%, (g) 80%, and (h) 100%.

strain. FE-SEM images (Figures S8 and 3c–h) of CsPbBr₃, CsPbBr₃/PEO, and CsPbBr₃/PEO/PVP films were prepared under varied strains. A tricomposite CsPbBr₃/PEO/PVP film presents nearly identical images (40%), indicating the improved stretchability by adding PEO and PVP (Figure S9). Such strain-independent morphological characters will ultimately promote the realization of stretchable LEDs.

As previously mentioned, we have employed a PU/AgNW composite electrode (Figure S1) with PET spacer to enrich device's touch responsivity, as illustrated in Figure 4a.⁴¹ When adequate pressure was applied on the PU/AgNW surface, a deformation enables the PU/AgNW electrode to contact the

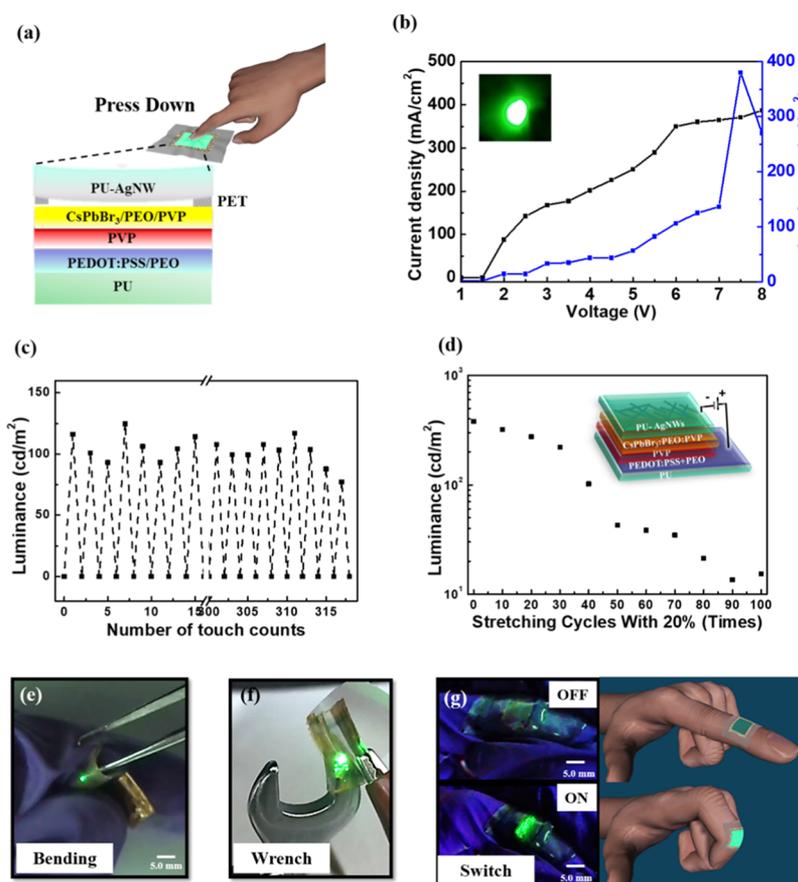


Figure 4. (a) Photograph of stretchable LETDs, (b) current–density–voltage and luminance–voltage characteristics of the stretchable perovskite LETDs, (c) durability test results of the devices under 3 V. (Before and after pressure was applied onto the surface of the AgNW/PU electrode.) (d) Luminance–stretching cycle characteristics of the devices after repetitive stretching cycles at strains of 20%. (e) Bendable perovskite LED (f) touched by the wrench (under 7 V) and (g) attached on the finger (under 7 V).

emissive layer, allowing the injection of electrons and holes when a voltage was applied (Figure S10). As the pressure was removed, the PU/AgNW electrode was detached from the emissive layer and returned to its original position, breaking the circuit. The luminance–voltage–current density characteristics of our fabricated stretchable and touch-sensitive LED are presented in Figure 4b. The device possesses a low turn-on voltage of 2 V, with a luminance of 380.5 cd m^{-2} at 7.5 V. Figure 4c reveals the good touch responsivity of the device, where the luminance is switched on/off for 315 cycles by applying repeated pressure. The luminance–strain cycle of the device with a 20% strain is shown in Figure 4d. As shown, our fabricated device can retain comparable performance before 40 stretching cycles, reflecting a certain stretchability of our touch-sensitive LED. More quantitative data are shown in Figure S11 for PeLETDs with a strain ranging from 0 to 100% and after repetitive stretching cycles at a strain of 20%. It is worth noting that the PeLETD based on the optimized perovskite active layer delivers much-enhanced performance and stability compared to other emissive systems. The device also displayed good flexibility. As presented in Figure 4e, instantaneous light emission in the regions under pressure with tweezers when bent at a 5 mm radius is observed. Because of the high flexibility and stability, the touch-sensitive LED could still show luminance while being attached to the wrench (irregular complex surface), as displayed in Figure 4f. It could be also attached to a finger to demonstrate its instantaneous emission

by bending the finger, as presented in Figure 4g. All these results unveil that our device can be easily integrated on any irregular complex surface, possessing the promising potential for practical applications. It is also important to mention that the decent performance of our device under strain (20–30%) is among the best results reported for the stretchable, touch-responsive perovskite LEDs.

3. CONCLUSIONS

In summary, we fabricated intrinsically stretchable perovskite light-emissive touch-responsive devices (PeLETDs) with good stability and low operation voltage by integrated device engineering that includes PVP modification and transparent PU/AgNWs electrode. The PEO and PVP polymer matrixes provided an elastic medium and induced uniform morphology for the perovskite crystals, thereby enabling the formation of stretchable perovskite emissive layer. Our PeLETDs performed well even under the strain of 30%, with a small luminance variation of 24% and the reproducible brilliant on–off response due to finger bend relax cycles demonstrating its good reproducibility and reliability. These interesting results unveil that our proposed PeLETDs can be easily integrated on any irregular complex surface, possessing the promising potential for practical applications. The decent performance of our device under strain (20–30%) is among the best results reported so far for stretchable touch-responsive perovskite LEDs.

4. EXPERIMENTAL SECTION

4.1. Materials. Cesium bromide (CsBr, 99%), lead bromide (PbBr₂, 99%), dimethyl sulfoxide (99.9%), *N,N*-dimethylformamide (anhydrous, 99.8%), chlorobenzene (anhydrous, 99.9%), poly(ethylene oxide) (PEO; $M_w \approx 5\,000\,000$), PVP ($M_w = 130\,000$), lithium fluoride (LiF, 99.99%), and indium–gallium eutectic (99.99%) were purchased from Sigma-Aldrich. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS; high conductivity grade 1.1 wt % in H₂O) was purchased from Ossila. Poly(dimethylsiloxane) (PDMS) was purchased from Dow Corning (Sylgard 184). Norland Optical Adhesive 63 (NOA63) was purchased from Norland Products. Polyurethane (PU) was purchased from RainEmpire. Silver nanowires (average diameter = 55–75 nm, average length = 20–40 μm) dispersed in isopropyl alcohol (IPA) (0.66%) were purchased from Zhejiang Kechuang. The ITO (Patterned Glass 15 Ω square, size = 30 × 30 × 0.7 mm³) was purchased from Ruilong.

4.2. Composite Electrode Fabrication (PDMS, NOA63, and PU). AgNWs were sprayed onto a substrate and annealed at 100 °C for 10 min, and then PU solution was poured on top of the poly(tetrafluoroethylene) (PTFE) substrate and consequently cured at 40 °C for 1 day. Through this process, the AgNWs adhered to the drop-cast PU film, which was then peeled off the PTFE substrate as elastomer transparent composite electrodes. The PDMS was mixed with a curing agent at a 10:1 ratio and spin-coated onto the glass. The film was cured at 100 °C for 5 min. The AgNW solution was sprayed onto the PDMS film and then annealed for 10 min at 120 °C. After annealing, the AgNWs became attached to the PDMS, which was then peeled off from the glass. The NOA63 solution was spin-coated onto the glass and subjected to ultraviolet light at 365 nm for 5 min. When the NOA63 was cured solid, AgNWs were sprayed onto the NOA63 surface. Subsequently, the AgNW/NOA63 film was peeled off from the glass.

4.3. Stretchable LED Fabrication on Polyurethane Substrate (PU/PEDOT:PSS-PEO/PVP/CsPbBr₃-PEO-PVP/PU-AgNWs). PU substrates were prepared by being spin-coated onto the PTFE substrate and being cured at 40 °C for 1 day. The PU substrates were treated with oxygen plasma for 3 min and the PEDOT:PSS/PEO solution was spin-coated onto the treated PU substrate at 1000 rpm for 30 s. Subsequently, the film was annealed at 80 °C for 15 min to remove residual solvents. After being cooled down, the substrate was transferred into an N₂-filled glovebox and spin-coated with the PVP, CsPbBr₃/PEO/PVP layers. For the PVP layer, it was spin-coated at 4000 rpm for 60 s and annealed at 80 °C for 10 min. The perovskite layer was prepared by being spin-coated at 3000 rpm for 60 s and annealed at 70 °C for 10 min, and then the spacer (PET) was introduced onto the perovskite layer. The prepared PU/AgNW electrode was then faced down and stacked onto the emissive layer that plays the role of a cathode (upper electrode).

4.4. LED Measurement. Current density–voltage and luminance–voltage characteristics were measured with a Keithley 2400 source meter and a silicon photodiode. The silicon photodiode was calibrated with a photo research PR-670 spectroradiometer, which was used to analyze the picture and estimate the active areas of the LEDs. The voltage was applied from 0 to 11 V, and the direction of current flows was from the upper to the bottom electrode for each measurement.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsami.9b23291>.

Transmittance spectra for composite electrodes; the sheet resistance for composite electrodes with increasing strain; 3M Scotch tape adhesion/peeling tests with PU/AgNWs electrodes; FE-SEM images of PU-AgNWs thin films under different strains; contact angle measurements for H₂O deposited onto PEDOT:PSS and PEDOT:PSS/PVP surfaces; FE-SEM image of the

samples with CsPbBr₃ deposited on PEDOT:PSS, PEDOT:PSS/PEO, and CsPbBr₃/PEO/PVP on PVP; FE-SEM image of the samples; AFM image of the samples; EL spectra of the CsPbBr₃/PEO/PVP film with different ratios; CIE 1931 *x*–*y* chromaticity diagram of the green perovskite; time-resolved PL spectra of different polymer ratio samples and different structures; the SEM image of the CsPbBr₃ and CsPbBr₃/PEO (1:0.125) composite thin film in different strains; photographs of the CsPbBr₃/PEO/PVP (1:0.125:0.0225) composite film under 365 nm UV irradiation in different strains; the transient current response; and external quantum efficiency (EQE) characteristic of PeLEDs under strain from 0 to 100% and after repetitive stretching cycles for strain 0–20% (PDF)

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Notes

The authors declare no competing financial interest.

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