Structural design of light-emitting fibers and fabrics for wearable and smart devices

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Abstract

Flexible light-emitting fibers and fabrics serve to bridge human–machine interactions. The desire for practical applications and the commercialization of flexible light-emitting fibers has accelerated structural progress and improvements. This review focuses on the structural design of light-emitting fibers and fabrics, starting with a summary of design principles, emission mechanisms, and structural evolution of coaxial structured light-emitting fibers. Subsequently, we explore recent advances in the helical structure design strategies that boost the mechanical sensitivity of light-emitting fibers. Following that, we analyze continuous preparation processes and the development of large-area intelligent light-emitting fabrics based on interwoven structures. Examples based on stiff and rigid inorganic-based light-emitting diodes integrated into flexible systems are also presented. Finally, we discuss the current challenges and future opportunities for light-emitting applications in the field of wearable and smart devices.

Keywords: Light-emitting fiber, Structure design, Wearable electronic, Flexible optoelectronic fiber, Large-scale fabric, Electronic textile

1. Introduction

Light-emitting (LE) fibers and fabrics are the product of the combination of rapidly developing optoelectronic technologies and tens of thousands of years of traditional fiber materials. They have emerged as a key development direction for wearable electronics [1–3]. Compared with panel and film form factors, the cylindrical shape of optical fiber-based LE devices greatly improves light output coupling efficiency [4, 5]. In addition, the natural advantages of fibers, such as flexibility, lightweight, small diameter, high aspect ratio, implantability, and long-distance signal transmission, make them a promising candidate for advanced medical, human–machine interfaces, and the Internet of Things applications [6–10]. Flexible LE fibers possess the ability to conform to the contours of the human body, seamlessly adapting to movements and facilitating the development of wearable optoelectronic systems. [11, 12]. One approach to flexibilization is to directly incorporate stiff light emitting diode (LED)-based LE devices into flexible fibers and textiles [13–15]. In these systems, tiny LE elements are embedded within the fiber or attached to the surface of the fiber/fabric to inherit the intrinsic flexibility of the fiber. Another method is to sequentially deposit the functional layers of the 2D planar LE device onto the fiber substrate, obtaining LE fibers with a coaxial structure. This method provides excellent mechanical flexibility and high luminous efficiency. Organic LE materials, with intrinsic flexibility and lightness, are ideal materials for LE fibers [16–19]. These organic material-based LE fibers can be fabricated by assembling multiple functional layers [20–22], including an emissive material, electrodes, and charge transfer channels, assembled in a coaxial structure. In some cases, additional outer layers (e.g., polydimethylsiloxane (PDMS)) are introduced to protect the inner layers and extend the life of the fiber [11, 23–26]. In addition to the organic LE materials, some inorganic phosphors (e.g., zinc sulfide (ZnS)) dispersed in a dielectric polymer can be used as flexible LE composites to deposit onto a conductive fiber substrate, forming stable and flexible LE fibers. Fig. 1 highlights the milestone structures of LE fibers in the last decade, as well as the corresponding

mechanisms of operation and preparation procedures.

The first method to fabricate LE fibers involved using planer organic light-emitting diodes (OLEDs). LE fibers with coaxial architectures are deposited layer by layer, incorporating various functional layers [24, 27, 28]. Upon applying voltage, electrons and holes are created from the cathode and anode, respectively, and move through the injection and transport layers before recombining to form excitons in the luminous active layer. The excitons return to the ground state through radiative decay and emit light in the process [29]. However, the device fabrication procedure for this type of fiber is complex and requires strict vacuum conditions. Moreover, the length and size of the obtained LE fibers are limited, making continuous fabrication difficult. In order to address the above issues, polymer light-emitting diodes (PLEDs) were developed [30, 31], which use a dip-coatable organic polymer layer as emissive layer [32]. Under an external voltage electrons and holes are injected into the polymer and transported through the polymer. When recombination occurs, the luminescent layer emits light. Compared with OLED, a breakthrough in the PLED preparation process is that the LE layer can be loaded onto the fiber substrate through simple dip coating [33–35]. However, the electrode layer of LE fibers can still only be fabricated by vacuum evaporation, and continuous preparation of LE fibers cannot be truly realized. The incorporation of polymer luminescent electrochemical cells (PLECs) into fiber architectures facilitates the continuous production of LE fibers. Here, a continuous metal wire served as the axial electrode, a twisted yarn of carbon nanotubes (CNTs) was utilized as the shell electrode, and a LE active layer containing the electrolyte was sandwiched between the two electrodes with the addition of a zinc oxide (ZnO) layer acting as an electron-transferring layer [36]. The presence of the electrolyte in the active layer is crucial for the PLEC to work. Under sufficient voltage, charges (electrons and holes) are injected into the active layer from the two electrodes, and counterions in the electrolyte are inserted near the corresponding injected charges to form doped regions (n-type doped regions and p-type doped regions) with high conductivity. Under the application of a constant

voltage the two doped regions expand forward and eventually meet to form a p-n junction, resulting in a large electron current. The electrons and holes recombine in the junction region and radiatively decay to the ground state, resulting in electroluminescence [37, 38]. Compared with PLEDs, the critical difference lies in the presence of mobile ions in the active layer, conferring electrochemical activity to the PLEC and allowing efficient light emission at low applied voltages [39]. This electrochemical property brings processing advantages when compared to PLEC fibers and do not require the use of low−work function electrodes [39, 40], so the stable metal wires and CNTs yarns could be used. A disadvantage with the PLEC fibers is that the luminescence mechanism assisted by the redistribution of mobile ions, makes their response time slow. Moreover, the electrochemical activity of the active layer makes it susceptible to degradation, especially in the presence of moisture and oxygen [41]. These disadvantages greatly limit the widespread practical use of PLEC fibers.

Alternating current electroluminescence (ACEL) systems have been developed to improve the durability. ACEL systems include an emissive layer which typically consists of ZnS-based luminophores that incorporate a dielectric material (e.g., polyethylene terephthalate (PET), PDMS). The obtained devices are insensitive to atmospheric conditions and are virtually unaffected by moisture and oxygen. In addition, the dielectric insulating layer sandwiched between the emissive layer and the electrodes effectively prevents the electrochemical reactions between them [42–45], avoiding the quenching of charge carriers in the active layer. Moreover, this insulating layer can also avoid charge accumulation in the device caused by direct current (DC) injection, which can reduce triplet−triplet or triplet−charge annihilation during device operation [46, 47]. In this device, the light emitting process consists of four main stages: the injection of electrons into the phosphor layer under a high alternating current (AC) electric field; the acceleration of the injected electrons; the excitation of luminescent centers as the high-energy electrons impact; and finally, the emission of light when the excited electron returns to the ground states [45, 48]. By utilizing organic polymers as dielectrics and binders, combined with air-stable

electrodes, all of the preparation procedures for ACEL fibers could be accomplished by simple solution-based dip-coating, which can be carried out in an ambient environment [49]. Therefore, ACEL fibers have been extensively developed in recent years, with their applications extending beyond lighting and displays. For example, based on the ACEL system, several mechanically responsive LE fibers with specific structures (here mainly helical structures) have been designed to extend the functionality of LE fibers for smart wearable electronic systems [50]. By combining the structural design of a spiral internal threaded LE layer [51], a helical-shaped electrode [52], or a helical LE fiber array [53], the mechanical responsiveness of luminescent devices has been enhanced; these can be used to improve the sensitivity of optical human motion monitoring systems. In addition, building on previous work, some large-scale LE fabrics have recently been realized using woven structures by weaving luminescent active fibers and conductive fibers together [54–56]. These fabrics have stable light emission and due to their structure, they preserve the fabric's flexibility and breathability to the greatest extent, paving the way for the continuous and mass production of LE textiles for wearable smart devices [57].

In this review, we focus on the evolution of LE fiber devices driven by structural design, with the aim of creating LE fibers suitable for continuous fabrication and large-scale applications for wearables and smart devices. First, the design principles, preparation methods, and emission mechanisms of several early coaxial-structured LE fibers are described in detail. This is followed by a discussion on progress made in device structure for mechanical sensing functions, and to facilitate continuous manufacturing processes. The discussion then shifts to the design and continuous preparation process developed for large-area LE fabrics, which is a significant step toward the scale-up of LE fibers. Inspired by the structural design of organic flexible LE fibers, we also present typical examples of flexible systems created using stiff and rigid inorganic-based LEDs. Finally, we address the challenges and opportunities for the future applications of LE fibers.

2. Simplification of coaxial structures drives the continuous fabrication of LE

fibers

LE fibers and fabrics have developed from planar LE devices. The most direct method to obtain LE fabrics involves depositing electrodes, emission materials, and functional layers directly onto a fabric. However, this approach faces obstacles due to the rough surface and heat resistance of textile substrates. To achieve optimal performance, LE devices typically require a flat and clean substrate layer and the multilayer deposition process requires high-temperature treatment. Furthermore, direct deposition results in the loss of the original flexibility and air/moisture permeability of fabrics. Instead, creating LE devices with a fiber shape and integrating them into fabrics can improve the flexibility [12], air permeability [58] and wearer comfort [59] of the resultant textile, which opens up a new frontier in wearable LE electronic devices. The first challenge is to fabricate the fiber-shaped LE device. As previously stated, the preparation of LE devices requires an extremely smooth and clean substrate. In contrast, conventional textile fibers typically have rough surfaces, and utilizing them as a substrate can result in a current buildup and localized heating (i.e., hot spots). These hot spots can cause a gradual escalation in current flow through the device, ultimately resulting in short circuits [60]. Additionally, the assembly process for each layer of the LE device must strictly control thickness, uniformity, and interfaces to ensure the effective transport of electrons and holes. Achieving these effects for fibers with high curvature surfaces can be particularly challenging. Therefore, the development of fiber-shaped LE devices requires careful consideration of the preparation process complexity, material development, structural design, and interface adjustment.

Organic LE devices typically use organic semiconductors as their electroluminescent-active materials. These devices can be fabricated by depositing an amorphous organic layer onto a substrate, avoiding lattice matching with the substrate and the high-temperature preparation process required for most inorganic semiconductor-based LE devices [61]. This makes them a favorable candidate for the fabrication of fiber-shaped LE

devices. The monolayer structure is the simplest form of OLEDs, consisting of a single layer of organic emissive material sandwiched between the anode and cathode. A disadvantage of this device structure is that it necessitates matching LUMO and HOMO energy levels between the electrodes and the emissive layer. Additionally, the organic emissive material must possess dual carrier transport properties to transport both electrons and holes. Further, it is challenging to find a material with similar electron and hole mobility. The addition of transport layers for electrons and holes can effectively enhance the carrier transport capability, resulting in a reduced operating voltage. Simultaneously, it can regulate the balance of hole and electron transport, avoiding or reducing the excess of a certain type of carrier in the device, which reduces the leakage current and improves the device's efficiency. Some LE devices may also incorporate additional layers such as hole transport, hole blocking, electron transport, electron blocking, cathode/anode interface, buffer, and other auxiliary layers, to enhance device performance. Fiber-shaped OLEDs have evolved from the multi-layer structures of archetypal planar LE devices [62, 63] (Fig. 2a, left) to a characteristic coaxial structure. The cathode, the electron transport layer, the luminescent active layer, the hole transport layer, and the anode can be successively assembled on the fiber substrate as shown in Fig. 2a (right). The charges are transmitted between the two electrodes, and luminescence occurs with the recombination of charge carriers in the luminescent active layer. The close interface between the electrodes and the active materials in a coaxial configuration is beneficial for rapid charge transfer [1], setting a key requirement for LE fiber manufacturing technology. To ensure that the thickness of each layer on the cylindrical fiber electrode is thin and uniform, a vacuum thermal evaporation process is utilized.

As pioneers in the field, O'Connor et al. [27] used vacuum thermal evaporation technology to develop the first fiber-shaped OLEDs, featuring a cross-sectional device structure similar to that of a planer device. Fig. 2b shows the deposited layers, comprising metal cathode/anode layers, electron/hole transport layers, and a

luminescent active layer. The substrate used was a polyimide-coated silica fiber with a diameter ranging from 50 to 1×10^3 μm. The polymer coating effectively smoothed the fiber substrate, enabling the subsequent deposition of the metal layer. Active layers, 100 nm thick, were grown concentrically on the fiber substrate through vacuum thermal evaporation with the fiber substrate rotated axially during the material deposition. The deposition rates and layer thicknesses were strictly controlled throughout the preparation process. When a forward electrical bias was applied, the OLED segment around the fiber substrate emitted green light. The cylindrical geometry of the fibers eliminated the emission spectrum variations with angle caused by the microcavity effect in planar devices, resulting in an emission peak and intensity that was independent of angle. This innovation represented a significant advancement in the transition of LE devices from a planar to a fiber form-factor. The layer-by-layer evaporation technique applied to axially rotating fiber substrates has become a paradigm for producing fiber-shaped OLEDs, leading to increased research into LE fiber devices. While the external quantum efficiency of LE fibers created using vacuum evaporation methods under precisely regulated preparation conditions is comparable to that of planar LE devices, the vacuum deposition process is complex and intermittent, making large-scale manufacturing difficult. Researchers are still actively seeking ways to produce LE fibers using simple, scalable technology.

The solution dip-coating method is extensively explored due to its ability to simplify the LE fiber fabrication process and reduce preparation requirements. However, this method has limitations as solution processing causes dispersion and mixing of the well-defined multilayer structures in the LE devices, which ultimately reduces the device performance [64]. Fortunately, the use of polymer materials can overcome this problem and have good film-forming and spreading qualities. As such, they are frequently employed in the solution-based manufacturing processing of OLEDs. OLEDs that contain polymer LE materials as the active layer are commonly referred to as PLEDs. In PLEDs, a typical sandwich structure is used, with the polymer-based active

layer sandwiched between two electrodes [32]. The electroluminescent polymers of the active layers are allowed to be deposited onto the fiber substrate using a dip-coating method. While this simple sandwich structure decreases the manufacturing costs, the lack of a separate electron and hole transport layers often leads to unbalanced charge transport within the active layer and low device efficiency, resulting in poor luminescence efficiency. To improve the lighting performance of the LE devices, it is necessary to incorporate charge injection and transport layers between the emissive layer and the electrode. Several charge injection and transport materials that can be applied using dip-coating have been developed, such as ZnO nanoparticles and polyethyleneimine (PEI), which have been combined to create an effective method for reducing work-function and solution-processability. This combination of materials is often used to assemble an electron injection layer (EIL) for LE fibers. Poly(3,4-ethylenedioxythiophene) (PEDOT) is commonly used as a hole injection layer (HIL) in electronic devices due to its high work-function, strong electrical conductivity, and optical transparency. Its high work function value reduces the barrier height between the anode and polymer interface, facilitating charge injection within the device [65]. To enhance the solubility of PEDOT for aqueous dip-coating, it is common to use polystyrene sulfonate (PSS) to create complexes known as PEDOT: PSS. The PSS serves a dual purpose, it enhances the water solubility of PEDOT and acts as a negative counter ion to the positively charged PEDOT, improving the electrical conductivity of the complexes. The coating performance and electrical properties of PEDOT: PSS can be modified by adjusting the composition ratio, impurity density, dopant type, and dispersion solvent. When its conductivity reaches a sufficient level, PEDOT: PSS can be used as a replacement for rigid ITO as an anode in devices.

Utilizing these dip-cotable materials, Kwon et al. [21] demonstrated an archetypal PLED fiber as shown in Fig. 2c. The fiber employed a bottom-emission inverted structure, with the light generated from the emitting layer passing through the substrates. For the conductive polymer PEDOT: PSS was used as the cathode,

multiple layers of ZnO/PEI, Super Yellow (SY) and molybdenum oxide (MoO_3) /Al were constituted as the EIL, emitting layer, HIL and anode, respectively (Fig. 2d). All of the functional layers could be deposited by dip-coating, except for the MoO_3/Al anodes, which were prepared using evaporation. It is worth noting that this inverted structure prevented the use of unstable low−work function metals as cathodes and improved the device's air stability. Additionally, the PEDOT: PSS served as both a cathode and a planarization layer for the fiber substrate, achieving a balance in carrier dispersion within the device. The use of ZnO/PEI as the EIL decouples the chemical synthesis of the nanoparticles and the layer deposition process, avoiding the high-temperature process necessary for direct ZnO deposition. As a result, the processing temperature was lowered to 105 °C to prevent the unstable deformation of the polymer fiber substrate at high temperatures. Consequently, the developed coaxial fiber PLED showed a brightness of more than 1×10^4 cd m^{-2}, an efficiency of 11 cd A^{-1}, and an operating life of 80 h. These values are comparable to those of conventional planar PLEDs made from glass substrates. Moreover, due to the utilization of a flexible fiber substrate (PET) and organic conductive polymer layer, the resulting fiber PLEDs could withstand a tensile strain of 4.3% at a radius of curvature of 3.5 mm and could be incorporated into knitted clothes. Due to its simplicity, low cost, and excellent performance, the dip-coating method has been widely implemented for this application [30]. Fluorescent PLEDs, phosphorescent PLEDs [28], perovskite LE/light-detecting bifunctional fibers [66], and numerous other luminescent fibers have been created successively using dip-coating.

Designing fiber-based white light OLEDs (WOLEDs) for LE devices can be challenging due to their complex structure and unstable optoelectronic device performance. WOLEDs typically use a tandem structure of thermally deposited multilayer emissive layers. However, the cylindrical shape of the fiber often leads to uneven, thin layers on the edges, resulting in device instability. WOLEDs based on a single emissive layer require the simultaneous deposition of multiple materials in a guaranteed ratio, leading to process complexity and cost

issues. Hwang et al. [67] used the dip-coating method mentioned above to create a WOLED fiber. They introduced a cylindrical fiber that was dip-coated with single white emission layer (Fig. 2e). The red, green, and blue (RGB) dopants were mixed in a poly(N-vinyl carbazole) host material, which provided sufficient surface tension for the dip-coating process. The RGB solution mixing ratio was adjusted carefully, in combination with chromatographic simulations and experiments, to obtain reliable white light color indices. Specifically, the single white light emitting layer involved a competition between two main emission mechanisms: energy transfer (ET) from the blue dopant to the red dopant and charge trapping (CT) to the dopant. To ensure the devices suitability for practical applications, it was important to avoid the voltage-influenced color shift due to CT. Therefore, they chose a RGB doping ratio that was closest to the white color spectrum while ensuring that ET was dominant. The resulting fiber WOLED exhibited a reliable white emission in color coordinates, and a stable performance (maximum $\Delta x = 0.00413$, $\Delta y = 0.00497$) when the driving voltage changed from 4 to 6 V. In addition, the WOLED fiber exhibited an operation voltage of less than 6 V, a luminance of approximately 738 cd m^{-2} and a current efficiency of around 10.8 cd A^{-1}. Further, after being subjected to 1×10^3 cycles at a tensile strain of 1.5%, the WOLED fiber device remained stable. This work represented a significant advancement in the development of reliable full-color wearable displays through the use of a simple solution process to prepare WOLED fibers.

Although dip-coating simplifies the fabrication process, vacuum deposition is still necessary for constructing the anode layer and the outermost encapsulated Al_2O_3 layer [68]. However, this compromises the flexibility of PLED fibers and makes continuous production of LE fibers impossible, as the length of the LE fiber mentioned above is limited to 3−4 mm. Furthermore, vacuum deposition often leads to uneven film deposition on cylindrical fibers, resulting in decreased optoelectronic performance of the LE fiber. To develop wearable technology and textiles, highly flexible LE fiber devices are necessary which are also long enough to be knitted or

woven into textiles. To help achieve this, flexible dip-coated electrodes, such as silver nanowires (AgNWs), CNTs, hybrid composites (e.g. dielectric/metal/dielectric), have been developed as alternatives to traditional metal electrodes. However, the work function of these materials cannot meet the demands of small molecule and polymer emissive materials, rendering electron charge injection in the LE layer inefficient.

Against such a background, PLECs is an ideal structure for the continuous preparation of flexible LE fibers. PLEC is a special case of PLED, first created by Pei et al. in 1995 [38]. The essential feature of PLEC is doping of the active layer of the PLED with an electrolyte to form an *in-situ* LE p-i-n junction upon applying voltage, increasing the carrier transport capacity of the active layer [38, 69]. This characteristic makes PLECs insensitive to electrode work function, eliminating the need for air-sensitive low work function electrodes. Additionally, it enables PLECs to operate with relatively rougher surfaces than those required for OLEDs and PLEDs. This makes it possible to scale up these devices at a low cost and with high efficiency. Zhang et al. [36] developed the first fiber-shaped PLECs, capable of continuously and uniformly assembling anode materials onto fibers. This PLEC fiber had a coaxial structure consisting of a ZnO-modified metal wire cathode, a conducting aligned CNT sheet, and a polymer LE layer sandwiched between them. The fabrication process involved solution-based processes combined with continuous CNT wrapping, as shown in Fig. 3a. The electro-luminescent polymer layer was composed of a blend of an emitting polymer, an ionic conductor, and a lithium salt. When a voltage was applied, ions in the emission layer migrate to form a p-i-n junction, where electrons and holes meet to generate exciton luminescence [70]. Aligned CNT sheets were utilized to evenly coat fibers that were impregnated with ZnO and polymer luminescent layers. The obtained PLEC fibers began emitting light at 5.6 V and reach a peak luminance of 609 cd m^{-2} at 13 V. The current efficiency increased with luminance and finally reached 0.83 cd A^{-1} with an external quantum efficiency of 0.35%. Although the quantum efficiency of the PLEC was not as good as that of PLEDs with a multilayer structure, it was still superior to that of PLEDs with

the same sandwich structure [71]. Additionally, the light emitted from the PLEC fiber remained independent of the observation angle due to the uniform coating of the CNTs (Fig. 3b). In other work, a continuous CNT coating has been used as both the anode and cathode materials to prepare the PLEC fiber as shown (Fig. 3c) [72]. By utilizing CNT wrapping technology, the entire preparation process can be conducted in the air, providing a new method for the continuous production of LE fibers. A disadvantage is that the omission of the ZnO EIL from the device resulted in a reduction of the maximum brightness of this PLEC fiber to 505 cd m^{-2} (at 30 V) and the maximum current efficiency to 0.51 cd A^{-1}. In addition, it should be noted that the transient light emission response under the voltage pulse was relatively slow due to the luminescence process caused by ion migration [73]. Additionally, the limited operational lifetime of the PLEC was a legitimate concern. Compared to PLEDs, which have a half-life of over 1×10^4 h, PLECs have a half-life of only 100 h or less [34]. The rapid deterioration of the electrochemically doped active layer or phase separation of the nonpolar conjugated polymers and the polar polymer electrolyte in the active layer caused this issue. Furthermore, the p-i-n junction relaxes when the external voltage is removed due to the redistribution of ions, making it unsuitable for repeated switching in practical applications. The intensity of the emitted light was reduced when the external voltage exceeded the electrochemical stabilization window of the active material, due to the appearance of the highly doped region known as the exciton quenching sites. Therefore, further improvement of the fiber device structures and materials is necessary before practical applications can be considered.

ACEL devices avoid charge buildup by frequently flipping the external electric field, which extends device operating life and improves power efficiency. Typically, the luminescent active layer consists of a blend of polymer elastomer-encapsulated phosphors [49, 74], which are robust and inert to moisture or oxygen. The charge carriers in the emissive layer are accelerated to high energy under an AC electric field, resulting in the generation of electron-hole pairs in the phosphor particles. Due to the restriction of the surrounding elastomer

network, the electron-hole pairs eventually recombine and emit light [45, 75]. Active phosphor typically consist of ZnS or metal-doped ZnS particles [49, 76–78]. These ZnS-based particles are cost-effective, versatile in color, and do not require strict uniformity in the active layer, which provides a promising solution for the large-scale production of LE fibers. The luminescent active layer should be sandwiched between the electrodes, giving rise to three types of ACEL fiber structures: coaxial electrodes, parallel electrodes, and single-electrode ACEL fiber structures. The coaxial structures, like in OLED, PLED, and PLEC fibers, are the most common ACEL fiber structures.

The coaxial structure of ACEL fibers is usually stacked layer by layer over a core electrode, with one or more layers of insulating media, luminescent active particles, a shell electrode, and an exterior protective layer. Since the preparation conditions are not strict, coaxial ACEL fibers could be fabricated entirely using the dip-coating method. For example, Liang et al. [42] developed a simple all-solution dip-coating process to prepare ACEL fibers with coaxial electrodes (Fig. 4a). Flexible and mechanically stable AgNWs with high-aspect ratios were used as the internal and external electrodes, sandwiching the ZnS luminescent active layer between them. The as-prepared ACEL fiber started emitting light at around 30 V, and the intensity then rapidly increased, reaching 202 cd m^{-2} at 195 V. To enable the continuous production of long ACEL fibers, a silicone binder was used during the dip-coating process of each layer. Fig. 4b illustrates that ACEL fibers, approximately 12 cm in length, can be produced using various LE particles that emit red, green, and blue light. It is worth noting that, in addition to binders, silicones are also utilized as insulators, flexibility mediums, and encapsulants in ACEL fibers. According to the principle of operation, ACEL requires high electric fields for electron tunneling, however, applying high electric fields (>100 V) can cause the electric field in the phosphor layer to exceed 107 V m^{-1} [43]. This may result in an electrical breakdown if there are some defects in the active layer, and lead to device failure. Therefore, silicone was used as the insulating materials between the electrodes and phosphor to

control carrier injection. Additionally, the silicone, combined with the excellent mechanical properties inherent in AgNWs, provides ACEL fiber with exceptional flexibility. Fig. 4c shows that after 500 bending cycles at a fixed position with a bend radius of 2 mm and a frequency of 1 Hz, the brightness of the ACEL fiber only decreased by 9.1%. The outermost silicone encapsulation layer also improved the operational durability and environmental stability of the ACEL fiber, making it suitable for wearable applications.

To emit light, the ACEL only required spatial contact between the electrode and luminescent layer, allowing it to function when the active layer deformed. Therefore, ACEL is suitable for designing highly elastic LE fibers [79–81] and as elastic fibers can adapt to the frequent and large-scale movements of the human bodies, they can potentially be used in wearable applications. Although elastic PET was used as the fiber substrate and silicone elastomers were extensively used as the binders for the ACEL fibers as described above, it was not possible to obtain stretchable ACEL fibers. This was due to the fact that the AgNWs electrodes could not withstand high stretching strains. The interconnected AgNWs could break due to stretching, resulting in a rapid increase in their resistance under strain [43]. Hydrogel is a highly elastic and transparent material with a resistivity that remains independent of deformation. Its resistivity increases with strain at a much slower rate than that of AgNW or other commercially available stretchable conductors, such as conductive polymers and carbon nanomaterials [81]. This makes hydrogel an ideal electrode material for highly elastic LE fibers. For example, Yang et al. [79] used a polyacrylamide (PAAM) hydrogel as the ionic electrodes when preparing a coaxial structure ACEL fiber. The fiber exhibited good elasticity, and was capable of being stretched to about 2.5 times its original length. In this coaxial structure, the luminescent layer composed of copper-doped ZnS (ZnS:Cu) particle-dopped PDMS which was sandwiched between two layers of PAAM hydrogel electrodes, with an outermost PDMS layer encapsulated around the hydrogel LE fiber to restrict the loss of water (Fig. 4d and e). To fabricate this hydrogel-based LE fiber, a multi-step dip-coating technique was utilized. It should be noted

that wetting was a prerequisite for the smooth implementation of this muti-step dip-coating process (i.e., good spreading of the dipping solution over the surface of the fibers to be coated in each layer). Through wettability experiments, it was found that the PDMS solution was able to easily wet the hydrogel fiber substrate, which was directly related to the surface energies of the dip-coating solution and the substrate, as well as their interfacial energies ($\gamma_{\text{hydrogel precursor}} > \gamma_{\text{PDMS substrate}} + \gamma_{\text{PDMS substrate–hydrogel precursor}}$, γ: surface energy). For hydrophobic PDMS solid surfaces, treatment with a hydrophilic precursor is required to effectively dip-coat PAAM precursor solutions. It was essential to ensure covalent anchoring between the hydrogel and the PDMS elastomer as well as enhance the elastic recovery of the luminescent hydrogel fiber. In this case, using silane as an anchor, the PDMS was modified with silane and immersed into a primer of silane-modified PAAM polymer. Afterwards, the silane on the surface of the PDMS hydrolyzed in the presence of water, and condensed with the silane on the PAAM polymer. As a result, a thin layer of covalently anchored PAAM chains was formed on the PDMS, making it hydrophilic. After dip-coating another, thicker, hydrogel layer and an exterior PDMS layer, the fiber was held at 65 °C to allow the silanes to condense into crosslinked PAAM chains and interconnect the PAAM with PDMS networks (Fig. 4e, right). Eventually, a strong, stretchable, and transparent adhesion between the hydrogel and the hydrophobic elastomer was formed. The luminescent fiber could still maintain its performance after 1×10^4 cycles of stretching to 1.5 times its initial length, showing great potential for use in wearable optoelectronic devices (Fig. 4f). However, the optimization of the brightness of the ionotronic fibers is necessary. At a fixed amplitude of the nominal electric field of 6 V μm^{-1}, brightness' of was about 16 cd m^{-2} could be achieved. The electrical conductivity of the hydrogel electrodes may have been affected by the degree of cross-linking and the infiltration of the silane coupling agent. Furthermore, the preparation process has a very long cycle time of over 24 h. This was primarily due to the extended period required for the cross-linking of the hydrogel, the curing of the elastomer, and the condensation of the silane coupling agent.

Peng and colleagues [81] shortened the process by using a one-step continuous preparation method, preparing ACEL fibers with parallel hydrogel electrodes (Fig. 4g). The two parallel hydrogel electrodes and the external luminescent layer were extruded concurrently. As shown in Fig. 4h, the hydrogel electrodes were designed in an elliptical shape to increase the face-to-face area between the electrodes and generate more light. The hydrogel system composed of poly(vinyl alcohol) (PVA) and poly(ethylene oxide), which were chosen to control the crosslinking of the composite hydrogel after extrusion. This resulted in a fiber electrode with high ionic conductivity and good tensile properties. The ionic conductivity reached approximately 0.29 S cm^{-1}, which surpassed that of many other reported hydrogel ionic conductors. Under an AC electric field, the maximum luminance could reach 242.6 cd m^{-2}. Moreover, the LE fibers exhibited exceptionally high elasticity due to the good cross-linking of the hydrogel, which could be stretch up to 800%, and maintain their luminescent properties through 100 cycles at 300% strain (Fig. 4i and j). The LE fibers, which are prepared by the one-step method, are flexible, elastic, and three-dimensionally twistable, allowing for the fabrication of display textiles using a loom. These display textiles can communicate with computers and mouse brains for smart display and camouflage applications. However, parallel electrode-structured fibers have some limitations. For example, the luminance was angle-dependent because the electric field was generated between two parallel hydrogel electrodes. By contrast, LE fibers with coaxial structure have a LE layer that is completely wrapped around the cylindrical surface, avoiding the angle dependence of light emission. Consequently, researchers are still exploring simple fabrication methods for coaxially structured LE fibers.

The preparation method for coaxial single-electrode fibers is much simpler than that of dual-electrode fibers, which is more promising in terms of continuous manufacturing. Li et al. [82] recently reported a unique single-electrode electroluminescent fiber that was prepared continuously by one-step coaxial wet spinning. The fiber was obtained by using a coaxial spinning nozzle to wrap the ZnS/ polyurethane (PU) EL solution around a

silver-plated nylon wire core and uniformly extrude it into a solidification bath. A conductive liquid was used as a bridging medium to connect to the prepared fiber (Figs. 4k–m). When an external power supply is applied, an AC electric field was generated between the conductive liquid and the inner electrode fiber, lighting up the luminescent layer. The luminance was related to the porous structure of the shell layer. The abundant pore network facilitated the combination of electrons and holes, leading to higher luminescence (149.08 cd m^{-2} at 4 V μm^{-1} and 6 kHz), which was 38.41% higher than that of a dense-LE-fiber. As a proof of concept, single-electrode electroluminescent fibers were stitched into various designs. As shown in Fig. 4n, the cat design becomes brighter when immersed in a conductive liquid (e.g., water). Traditional weaving processes can be used to combine single-electrode fibers into fabrics with a plain weave texture structure. When stimulated by an electric field, the fabric coated with small water droplets generates a bright blue light, revealing the shape and size of the droplets (Fig. 4o). Therefore, these conductive liquid-bridged LE fabrics show potential for applications including water sensing, visual interaction, and for environmental warning systems. In addition, since various liquids have different ionic conductivities, this conductive liquid-bridged single-electrode fiber fabric exhibited optical responses to different liquid electrodes (such as water, alcohol, glycerol and NaCl solutions), and was expected to be used for liquid identification (Fig. 4p).

By carefully selecting layer materials and optimizing the fibers structural design, LE fibers have progressively advanced toward using simplified preparation processes. This progress has led to the development of numerous LE fiber devices exhibiting excellent flexibility, elasticity, and smart sensing capabilities. The novel textiles created by incorporating these LE fibers into fabrics exhibit excellent flexibility, stretchability, breathability and seamless integration to the contours of human bodies, laying a foundation for the commercial preparation and practical application of LE fiber in the field of wearable display electronics.

3. Helical structure design enhances the mechanical responsiveness of LE fibers

Endowing LE fibers with multiple functions is crucial for the future of smart fabrics. Mechanical sensing is one of these highly valuable functions, which enable wearable devices to sense the daily movements of the human body and express them through optical and/or electrical signals [26, 50, 83, 84]. ZnS:Cu inorganic hybrid semiconductors are shown to be ideal candidates for demonstrating mechanoluminescent phenomenon, which is derived from electron de-trapping electroluminescence triggered by the piezoelectric effect. The energy transfer from impact force to the ZnS:Cu inorganic phosphors is important for luminescence generation. When the device is subjected to an external force, the stress is redistributed and concentrated at the local sites, the excited emission is generated mainly at the stress concentration area. Compared to conventionally employed cylindrical fibers, the design of helical fiber structures has attracted great interest for mechanical sensing.

Zhou et al. [51] developed a pressure-responsive LE fiber sensor that could both detect and visualize external force stimuli in a single device. They used a helical steel wire as a template to prepare the LE fiber sensor. Following chemical etching, the uniaxial folded microstructures distributed on the inner surface of the sheath were generated (Figs. 5a–d). First, a steel wire was taken as the core fiber and another steel fiber was coiled on around it to produce a spring-shaped fiber template with a thread structure. Subsequently, a blend of ZnS:Cu/PDMS both as a dielectric layer and electroluminescence layer was coated on the outer surface of the above helical fiber template. When the template was removed by etching, ZnS:Cu/PDMS hollow fibers were generated with internal voids. Finally, the LE fiber was obtained by inserting steel fibers into the fiber hollow as inner electrodes (Fig. 5a and b). Compared to a control fiber without voids (Fig. 5c), the resulting fiber sensor with the helical micro-pattern showed a high-pressure sensitivity coefficient ($\Delta C/C_0$ per N) of 16.81 N^{-1} (< 0.05 N, low region) and 0.91 N^{-1} (> 0.05 N, high region) in the low and high regions, respectively. For fiber sensors without a helical micro-pattern, the sensitivity coefficient was much lower (only ~2.20 N^{-1} and 0.16 N^{-1} in low and high regions, respectively). The analysis of the factors that influence the change in capacitance of the two

structures was based on the equation $C = \varepsilon_{eff}(A/d)$, where C is capacitance, ε_{eff} is the dielectric material's efficient dielectric constant, A is the contact area, and d is the distance between the fiber electrode and the epidermal electrode (Fig. 5d). The significant improvement in the sensitivity of the sensor with the helical micro-pattern may be ascribed to the dielectric layer's good deformability and an increased ε_{eff}: The fiber with helical micro-patterns was easy to deformed, as the local force was mostly centered at the ridge points and ε_{eff} decreased significantly as the uniaxial folded microstructures dramatically deformed under an external force (Fig. 5d). In such a well-designed microstructure, the dielectric layer's force-induced thickness and the dielectric constant could be modulated effectively, leading to a substantial increase in the sensor's sensitivity. In addition to sensing force by changes in capacitance, the application of force could also be visualized by changes in brightness. As shown in Fig. 5e, the device exhibited a virtually noise-free electroluminescent response under alternating 0.5 N/1 N loading/unloading forces. The luminescence sensing was proven to be reliable, as the luminescence intensity retained its original level after 1,500 loading and unloading cycles.

Designing fibers into helical shapes is a common strategy to transform non-stretchable materials or devices into highly stretchable elastic materials. A typical example is a highly elastic spring obtained from rigid metals tailored by helical shapes. Li et al. [85] recently explained this phenomenon as a macroscopic helical structural conformation leading to the macroscopic elastic behavior of materials, inspired by the classical theories that polymer chain conformation affects the macroscopic properties of materials [86]. By designing helical conformations, it is possible to escape from the dependence on elastic materials in conventional stretching devices. Therefore, it is of great interest to achieve simple and stable luminescence performance of mechano-sensing LE fibers by imparting or enhancing their tensile properties through helical structure without changing the LE fiber material. Using the helical shape, He et al. [52] designed a tensile-responsive luminescent fiber where the coiled springs acted as electrodes rather than templates (Fig. 5f and g). Specifically, a spring was used as the

inner electrode and support layer, and a ZnS:Cu/PDMS layer was coated around the helical spring. The spring provided good stretchability and a large contact area between the electrode and luminescence active layer, endowing the fiber sensor with good sensitivity. The resulting fiber demonstrated the ability to sense human movement, showing both electrical and luminescence signals. As shown in Fig. 5h, various physiological signals, including movements of the finger, wrist, neck, elbow, shoulder, knee, ankle, sole, and other joints were distinguished and monitored in real-time, visually, and in a non-invasive way.

In addition to the introduction of helical structures as the electrode of the fiber device, the illuminating units have also been processed into a spring-like structure, serving as illuminating pixel points, arranged vertically or horizontally between two transparent PDMS films [53]. The resulting flexible luminescent devices remained stable after bending or twisting. When subjected to external pressure, the associated PDMS pushed the surrounding LE units to deform together, resulting in the spring spots (Fig. 5i). It was found that horizontally aligned mechanoluminescent devices exhibit a brighter emission than vertically aligned mechanoluminescent devices due to their effective deformation and larger LE area. Compared with vertically arranged helical fibers, horizontally arranged luminescent units produce more effective material deformations when subjected to the same external force, resulting in the emission of brighter luminescence at low pressures (Fig. 5j). Consequently, the device's sensitivity to pressure can be properly adjusted by designing different luminescent unit structures and assembly forms. Moreover, the helical fiber device showed excellent flexibility, which could recover to its original shape after bending and twisting, thanks to its inherent geometry (Fig. 5i). This research provides a novel method for regulating piezoelectric luminescence.

4. Interwoven structures encourage continuous LE fabric manufacturing and large-area LE fabric development

Given that planar OLED panels have already been commercialized using vacuum evaporation technology,

developing OLED devices on planar fabrics is considered a more straightforward approach for large-area fabrication than developing OLED devices on cylindrical fibers. Kim et al. [87] reported the fabrication of PLEDs on fabrics using spin-coating. The process involved planarizing the fabric surface with Al_2O_3/PVA and then assembling PLEDs on it by solution spin-coating. The fabricated devices exhibited high luminous efficiency (9.72 cd A^{-1}) and power efficiency (7.17 lm W^{-1}). In addition, the PLEDs fabrics showed stable operation under bending, with a bending radius of 2.5 mm. Kim et al. [88] planarized the fabrics with PU and encapsulated them with multilayer Al_2O_3/PVA barrier films, which resulted in the operating life of the fabric-based OLEDs of over 1×10^3 h. However, braided and opaque textile substrates cause light loss, and the deposition of light-emitting devices affects the air and moisture permeability of the fabric. Therefore, the majority of these LE fabrics can only exhibit illumination over small sections and in pre-designed patterns. Creating large areas of luminous display with textiles remains a challenge.

In order to address the above problem, researchers have recently developed several technologies that hold promise for industrial-scale manufacturing of large-scale luminous textile displays. For example, Chang et al. [89] created an adhere-coating method for the continuous spinning of LE fibers (Fig. 6a). Specifically, commercial PU fibers were first dip-coated with dilute PDMS and then immersed in a bath of ZnS:Cu luminous particles. After rapid curing with a heat tube, the phosphors adhered firmly to the fiber. Finally, the interior was encapsulated with PDMS again and cured to form a continuous luminous fiber. This produces meter-scale LE fibers with good brightness and flexibility, which were then processed into luminescent weft-knitted fabrics and used for mechanical sensing (Fig. 6b). This continuous spinning process was limited to the manufacture of mechanoluminescent or photoluminescent fibers. The development of smart optical clothing often requires integrated optoelectronic devices and their incorporation displays into knitted or woven textiles has the potential to revolutionize our interaction with electronic devices. Recently, Lee et al. [90] effectively realized the

automatic integration of optoelectronic fiber devices and textiles by automatically weaving fiber quantum dot light-emitting diodes (F-QLEDs). In order to avoid mechanical damage to the fiber devices during the automated weaving process, the as-prepared F-QLEDs were first encapsulated using PET. Then these fiber devices were inserted into the textiles using an automated weaving. The weaving parameters were optimized according to the mechanical stress applied to the fiber device on the loom. Laser welding was then used to form conductive paths (Fig. 6c and d) and the curing time was controlled within 1 s to avoid thermal damage to the devices. These optimized device architectures and processes ensured the reliable integration and performance of the devices. As a paradigm, this automated integration approach has also been used to encapsulate other fiber devices, such as fiber optic photodetectors (F-PD), fiber optic supercapacitors (F-SC), and fiber optic field effect transistors (F-FETs) (Fig. 6c). Therefore, through the integration of various fiber optic devices in single and multiple units, specific and scalable functions such as optical sensing, electrical actuation, and energy storage can be simultaneously achieved (Fig. 6d). The work is a significant step towards the integration of current electronics technology textile engineering.

Another approach to the large-scale manufacture of LE textiles is to assemble small LE units over large areas. Shi et al. [54] designed a large-area display fabric that was 6 m long and 25 cm wide, containing 5×10^5 electroluminescent units spaced approximately 800 μm apart from each other (Fig. 7a). The display fabric showed a unique interwoven structure, woven from luminous warp yarns and conductive weft yarns. At the contact points of weft and warp yarns, micron-scale electroluminescent units were formed (Fig. 7b). The conductive weft yarn was fabricated by melt-spinning ionic-liquid-doped PU, while the luminescent warp yarn was obtained by coating ZnS phosphor slurry onto the surface of a silver-plated yarn (Fig. 7c). Triggered by an AC, the ionic liquid in the core layer of the weft yarn and the conductive warp yarn formed an electric field, resulting in light emission from the ZnS phosphor at the weft–warp contact area. Each interlaced point between the weft

and warp formed an EL unit, which produced a luminescent pixel (Fig. 7d). Based on this principle, the fabric can be worn like a large screen (Figs. 7e–g). The brightness of the LE units varies by less than 8%, and the fabric was not only stretchable, breathable, and durable, but could also withstand repeated machine washes, demonstrating that it would be practical to use for navigation or healthcare displays.

5. LED-incorporated flexible LE fiber

LEDs are solid-state semiconductor devices that have the advantages of high efficiency, long lifetime and a large color palette, which make them popular in display and fashion applications [91]. LED chip production involves epilayer growth on a bare wafer and electric contact pad attachment, making them rigid, thick, and difficult to apply to wearable devices. Utilizing flexible fibers with a high aspect ratio to anchor LED chips is feasible to produce fiber-shaped flexible devices. As a result, the inherent drape, consistency, and stretchability of the fibers counteract the LED's rigidity, producing flexible LE textiles [13]. Several discrete approaches have been employed to incorporate LEDs into fibers and yarns. One of the most common structures is an LE fiber formed by mounting multiple LED devices on the surface of a fiber or yarn.

Hardy et al. [13] have developed yarns with a diameter of around 1 mm where a small LED (0402 SMD packaging: 1.0 mm × 0.5 mm × 0.5 mm) has been soldered onto copper wires, encapsulated within a small, rigid polymer pod along with supporting yarns, and covered in a fibrous sheath consolidating the structure. These LED embedded electronic yarns (E-yarns) have been integrated into textiles by using both couching [13] and weaving [92] with the E-yarn acting as the weft. LED E-yarns have been demonstrated to be able to survive at least five machine washing and drying cycles [93], however revisions to the E-yarn design including the use of a robust Litz wire for interconnections and a braided outer structure, as demonstrated for inertial motion unit embedded electronics yarns [94], are believed to have significantly enhanced their durability. While the E-yarns are thicker than some of the other innovations in this section, the use of smaller SMD LEDs would facilitate

the creation of much thinner yarns. In addition, the research group has made progress towards the automated production of the electronic yarns [95] to realize the large-scale production of the E-yarns.

Zheng et al. [96] utilized a surface mounting method to develop an LE fiber device that decorates a helical fiber with multiple mini-LEDs, which can be implanted onto specific nerves and induce muscle responses by stimulating the nerves with the mini-LEDs. PU was chosen as the fiber matrix due to its thermally-induced shape-memory properties and biocompatibility. As shown in Fig. 8a, the PU was first ejected from a spinneret and formed the original straight fiber. Then the mini-LEDs, which are insulated with PDMS, were fixed on the PU fiber using cyanoacrylate and epoxy resin adhesives. The resulting PU fiber/mini-LEDs were then rolled onto a metal rod and heated above the deformation temperature to form a spiral shape. The diameter of the helix could be custom-tuned, and the angle and distance between the mini-LEDs could be modified according to the size of the targeted nerve. As a result, this optoelectronic device successfully stimulated the movement of selective peripheral nerves (Fig. 8b). Despite the complex wiring of the conductive fibers, this work may inspire further applications of LE fibers in neurosurgery and neuroscience research.

For LED-based LE fabrics, it is crucial to maintain adequate conductive pathways for each component while avoiding excessive wire laying. This is essential for the fabric to be lightweight and wearable. A common solution is to design a set of bus lines in the fiber or fabric, where multiple LED primitives are embedded in the fiber or fabric bus using a bonding material. This strategy makes it relatively easy to integrate LEDs into fabrics. A concept presented by Komolafe et al. [97] has seen filament circuits created by etching a copper clad Kapton board creating very fine conductive interconnection onto which components could be soldered. This concept has been used to create complex circuits that include LEDs. These thin boards could then be incorporated into a channel within a woven textile. Glob-top encapsulation could be used to mechanically protect components, and this was seen to enhance its durability to repeated bending. However, applying this strategy to the individual

fibers of an LED inlay can be challenging.

Rein et al. [14] demonstrate a method to obtained flexible single LE fibers by pre-laying the conductive path in a prefabricated part and then performing a thermal drawing process. High-performance LED semiconductor devices were then incorporated into the polymer–clad preforms (Fig. 8c). During the heat drawing process, the viscous polymer cladding simultaneously facilitated *in-situ* electrical connection and device packaging. Specifically, micro-scale semiconductor devices were placed in a fiber-performing body, followed by inserting copper or tungsten wires into the hollow channel. When heated, the preform was stretched into a fiber, and the micro-scale semiconductor device within the preforms was then separated. Simultaneously, the conductive wire and micro-semiconductor device were pressed against each other until an electrical contact between them was made. Rein et al. [14] have managed to create single fibers with hundreds of micro semiconductor devices connected in parallel within them (Fig. 8c). This is the first time a long fiber containing LEDs which emits light along the entire length of the fiber when powered by an electrical field has been produced in a single thermal drawing process. This thermally-drawn method could also be applicable to other electronic devices. Moreover, two micro-devices, such as LEDs and light-detecting p-i-n diodes, could be woven into the fabric, creating a bidirectional optical communication link between the two fabrics (Fig. 8d). Finally, the potential for implementing physiological signal monitoring in all-textile systems was also demonstrated (Fig. 8e). This design and application of the above-mentioned LED-based flexible optical fiber devices shows significant value in future wearable electronics and smart optical fibers. Further, they inspire the future development direction of OLED, PLED, PLEC, and ACLE flexible LE fibers. In addition to applications in display fabrics and wearable electronics, applications of these fibers in the communications and medical industries may make them even more valuable for future wearable devices.

Textiles with normal properties have been created using LED-incorporated flexible LE fibers, principally

using weaving (as shown by [97] and [14]). LED-incorporated flexible LE fibers have also been demonstrated to be extremely durable, with Rein et al. [14] reporting that their devices could remain undamaged when subjected to tensile stress of up to 70 MPa. Most devices have been shown to be wash-durable. Rein et al. [14] report their diode fibers to continue to function after ten machine wash cycles, however full details of this test are absent in their 2018 article. LED E-yarns and LED's mounted on filament circuits created by etching a copper clad Kapton were both tested for wash durability as part of the same study, seeing both types of device survive up to five washing cycles followed by being line dried [93]. Here E-yarns performed slightly better when tumble drying was employed, and all of the E-yarn samples survived five machine washing and tumble drying cycles. Since this study, further work has been conducted on the packaging of similar E-textile devices created using filament circuits, with wash trials showing the E-textiles surviving 10–15 wash cycles [98]. This implies that newer E-textile filament circuits with LEDs would survive a greater number of wash cycles than previously reported. The use of thermal drawing to integrate optical devices into fibers currently appears to be the most scalable, allowing for the population of hundreds of devices onto a single fiber. Common to all of the integration methods discussed in this section, the use of wires and non-stretchable materials at the core of these flexible LE fibers will restrict the extensibility of the fibers, which may be undesirable for some applications. Different techniques for incorporating the fibers into textiles may help overcome this limitation; for example, most knitted structures are inherently stretchable, so knitting fibers could provide a final textile that can be stretched.

6. Conclusions and Perspectives

The structural design of LE fibers and fabrics for wearable electronic garments is constantly evolving. In recent years, fiber-shaped OLEDs, PLEDs, PLECs, and ACELs have been developed to be woven or knitted

into fabrics to obtain wearable LE clothing. The simplification of the structure of these LE fibers has improved production continuity. In the past, multilayer OLEDs required a rigorous vacuum vapor deposition process for preparation that could only produce LE fibers in millimeter lengths. The development of polymer-based PLED fibers has simplified the preparation process by enabling the coating of the luminescent layer through a solution method. The development of PLEC has further enabled the use of continuously wrapped CNTs as electrodes and simplified the structure to a true sandwich structure. The ACEL device utilizes phosphors and elastomers to create a stabilized luminescent layer, reducing the need for electrodes and fiber substrates. This enables the production of ACEL long fibers through either a full solution process or a one-step extrusion method. The LE devices in fiber form are approaching a continuous scale. In addition, the high elasticity and simple preparation conditions of ACEL fibers have led to the development of fibers with a mechano-luminescence response through the design of helical microstructures or macroscopic helical shapes, which have been used for human motion monitoring. LE fabrics based on textile platforms have also shown promising progress. A large-area LE textile was developed by interweaving materials. One yarn in the structure acted as an electrode, while the other contained the rest of the LE diode material. The preparation of LE fibers was continuous and stable, which has promoted research on the application of large fully flexible display fabrics. In addition, the technology for manufacturing LED-based flexible LE fibers and fabrics is constantly improving, including upgrades to materials, structures, circuits, and automated production processes. Although significant progress has been made in LE fibers and fabrics, there are still many challenges to achieving their large-scale application. Three areas for future development have been identified.

1. Striking a balance between structural simplicity and fiber performance. Material and manufacturing costs must be considered during the material commercialization process. However, the challenge persists in creating LE fiber devices that strike a balance between a simple structure, high LE performance, low energy

consumption, and excellent flexibility and elasticity. Although LEDs offer high energy efficiency and stability, the device stiffness limits their use in fabrics. OLEDs greatly improve device flexibility while maintaining high energy efficiency. However, the high cost of evaporation technology prevents their large-scale application. While dip-coating has advanced the solution process for PLEDs, vacuum deposition and tight encapsulation remain necessary. PLEC fibers offer the advantages of not requiring low-work-function electrodes, being easy to fabricate continuously, and having high flexibility and low operating voltage. However, their operating life and stability present challenges that must be addressed for practical applications. ACEL fibers offer several advantages, such as stable luminescence and good mechanical properties. For instance, ACEL employs a water/air resistant ZnS-based phosphor/elastic dielectric material as the luminescent active layer, making the processing requirements low. Direct assembling of luminescent layers in fibers generates leakage currents and non-radiative transitions due to their surface texture. However, in ACEL, elastomeric dielectrics can be utilized to accommodate these rough surfaces, resulting in high LE efficiency. Additionally, ACEL fibers possess excellent mechanical properties as elastomeric media are extensively used in these devices. In terms of electrodes, ACEL devices do not necessitate charge injection/transport layers or air-sensitive work function electrodes. The tensile stability of the electrode materials can be enhanced by utilizing flexible silver nanowires, ionic liquid electrodes, and hydrogel electrodes. ACEL-based LE fibers are a viable solution for achieving a balance between structure and performance. The luminescence principle of ACEL is also applicable to fabrics, as it only requires spatial contact between the electrode material and the luminescent layer to produce luminescence. The production of large-area display fabrics has been accomplished by creating micro LE devices at the intersections of the warp and weft fibers to form LE pixels, using industrial weaving equipment. As a result, further LE fibers and fabrics based on the structure of ACEL devices are expected to be produced in the future. Despite promise, limitations

include the high operating voltage needed for large-scale application as a portable electronic device. Furthermore, it is crucial to take into account that the LE structures may suffer damage from collisions, friction, bending, stretching, and repeated washing of the fabrics during abrasion, which can ultimately result in device failure. In their study, Shi et al. [54] aimed to achieve stable luminescence from the fabric during bending and stretching by adjusting the modulus of the highly elastic polymer conductive fibers. Interweaving LE warp threads with the fabric results in adaptive elastic deformation, creating stable contact. However, this process can cause the fabric to become stiff, which may affect comfort when worn. Achieving the perfect combination of LE devices and textiles remains a challenging task for the future.

2. Integrating LE fibers with other functional fiber devices into fabrics to develop human-oriented smart electronic devices. Interconnections between fiber-based electronic devices can replace rigid components in integrated devices and enhance the wearability of fabric devices. For instance, integrating electrically driven LE devices into clothing requires a crucial power supply module. Typically, DC or AC power sources are utilized in the laboratory. However, in practice, adding an extra power supply would increase the weight of lightweight fabrics, which compromises their wearability and comfort. Integrating flexible fiber energy storage devices, such as fiber batteries and supercapacitors, and fiber energy harvesting devices, such as biofuel cells and nanogenerators, can provide energy to an integrated system. Fiber electronic devices have shown promise as wearable LE fabrics that provide multiple electronic functions [1, 2, 99, 100]. Shi et al. [54] have developed a functional integrated system of photovoltaic fabrics, energy storage fabrics, touch sensing fabrics, and display fabrics using a weaving method. This allows for a fabric system that can perform multiple functions, including energy conversion and storage, sensing, and display. The system has promising applications for the Internet of Things and human−computer interaction, such as real-time positioning, intelligent communication, and medical assistance. However, the tolerance or power limitations of

these fiber electronic devices and the rigid electrical connections between the integrated devices hinder their further development. Addressing this issue necessitates collaboration among researchers from diverse fields, including experts in electronic circuits, optics, information, medicine, and people from industry [6].

3. Promoting the industrialization and large-scale application of LE fibers and fabrics. The ideas and key technologies for preparing LE fibers are based on planar LE devices. This review explores the preparation methods of LE devices applicable to fibers, taking into account the high curvature and rough surface characteristics of fibers. However, the preparation of LE fibers still lacks equipment standards, and testing methods and standards for characterizing these fibers vary. This makes it difficult to quantitatively compare their performance and hinders their application [42]. Therefore, the urgent development of industry standards for fibrous electronic devices is necessary. Although it may require a significant amount of time, the effort is worthwhile to provide people with clear guidelines and a path forward towards future industrialization and application. Furthermore, promoting the large-scale application of LE fibers and fabrics necessitates multidisciplinary cross-development. The development of LE fibers involves the synthesis of luminescent molecules, the design and assembly of LE devices, the construction of device circuits, the fabrication of LE textiles, and the development and improvement of equipment for the preparation process [3, 100–103]. This requires collaboration between various fields, including chemistry, materials science, physics, mechanics, electronics, and textiles. Effective communication and a high level of trust between different disciplines are crucial for successful collaboration and scale-up. Additionally, cooperation with industry partners is necessary to move LE fibers from the laboratory to the factory.

Conflict of Interest Statements

The authors declare that they have no conflict of interest.

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Author Contributions

Xiaoxiao Yu and Yanhua Cheng contributed to the conceptualization and writing of the manuscript. Linfeng Chen assisted with the literature search and picture drawing. Junyan Zhang, Wei Yan, Theo Hughes-Riley, Yanhua Cheng and Meifang Zhu reviewed and revised the original draft. All of the authors discussed and contributed to the final manuscript.

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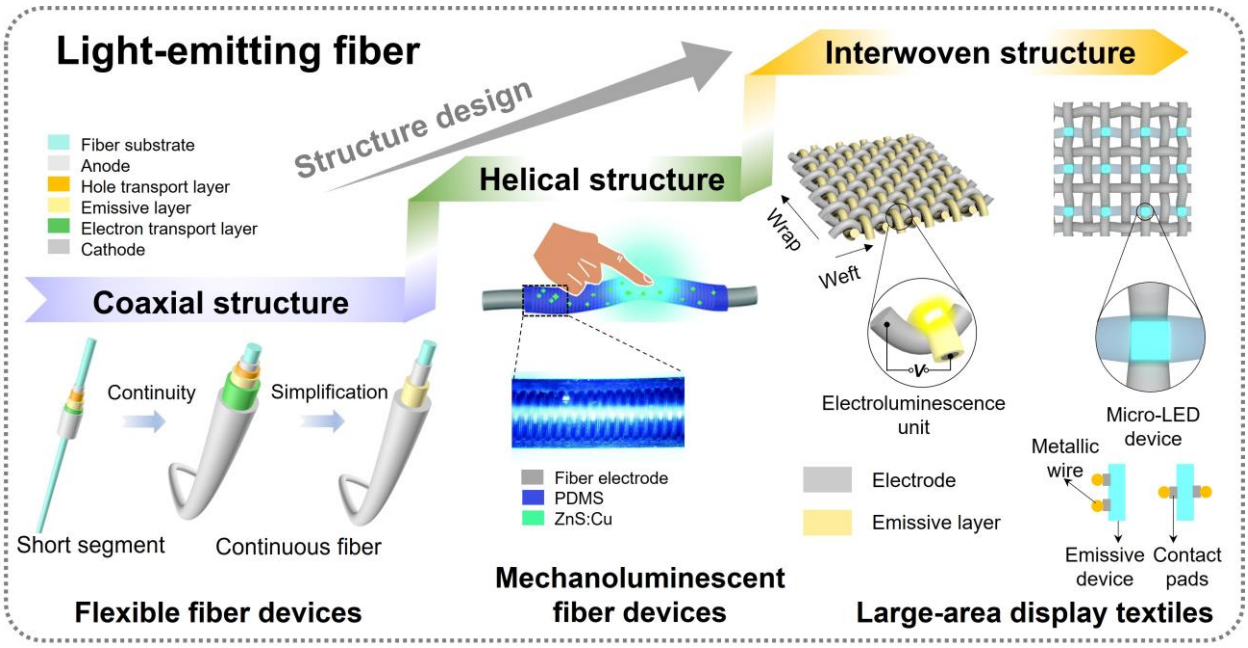
Figure



Graphical abstract

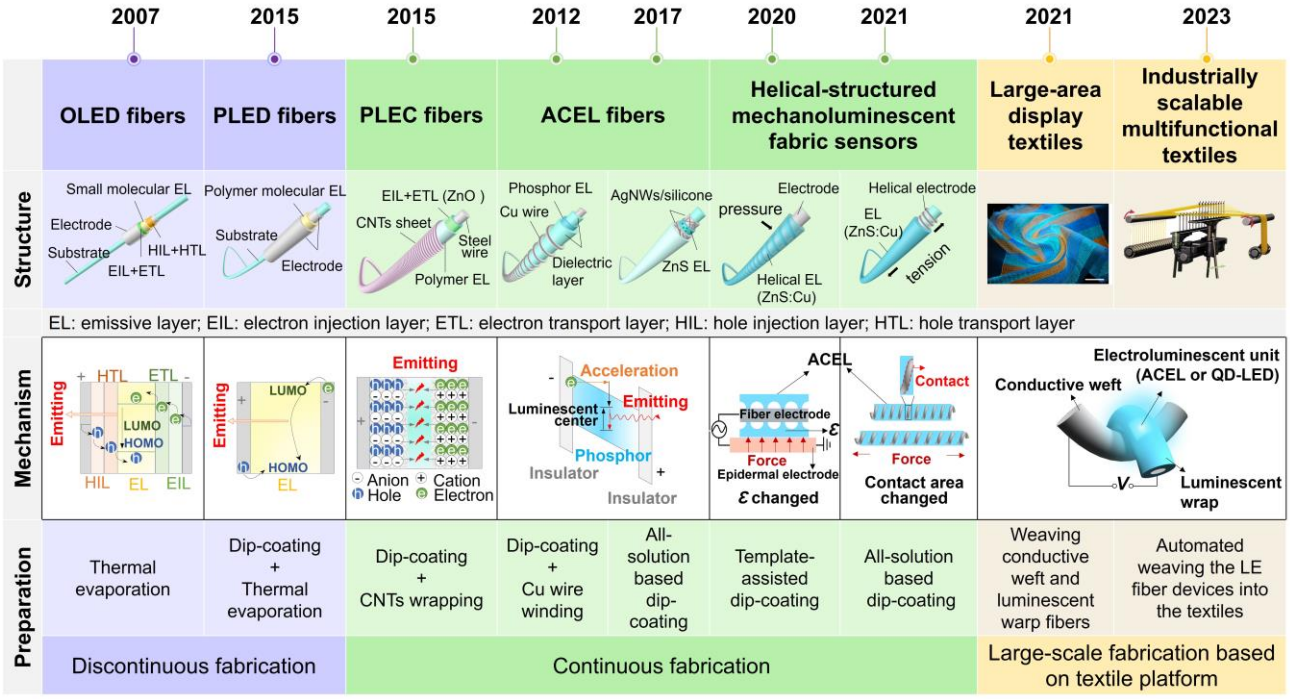


Fig. 1 (Color online) Brief chronology of the advances in light-emitting (LE) fibers for wearable and smart devices. The key milestones include the first demonstrations of the OLED fiber, PLED fiber, PLEC fiber and ACEL

fiber, as well as the helical-structured mechanoluminescent fabric sensor, the large-area display textile and the industrially scalable multifunctional textile. LE fiber fabrication has evolved from discontinuous to continuous, and then to large-area textile manufacturing.

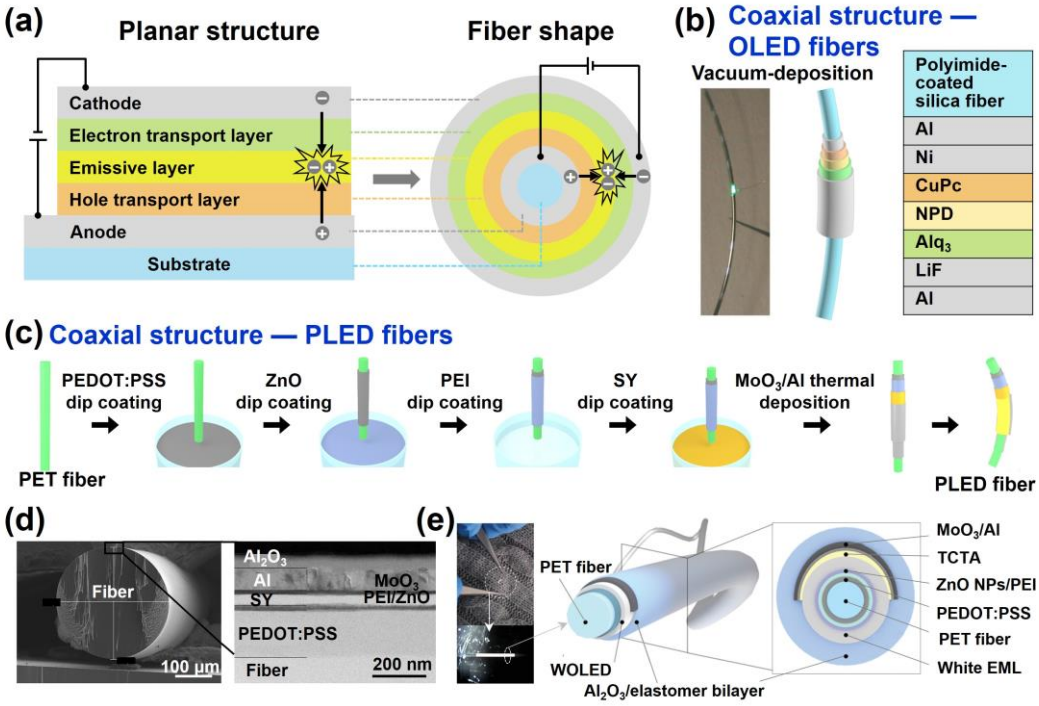


Fig. 2 (Color online) OLED and PLED fibers. (a) The multilayer structure of planar and fiber-shaped LE devices. (b) Photograph (left) and schematic diagram of device structure (right) of OLED fiber. Reprinted with permission from reference [27]. Copyright 2007, Wiley-VCH. (c) Schematic fabrication of a PLED fiber mainly using the dip-coating method. (d) Cross-sectional scanning electron microscope (SEM) images of the PLED fiber fabricated in (c) with multilayer. (c–d) Reprinted with permission from reference [21]. Copyright 2017, American Chemical Society. (e) Photograph (left panel) of a WOLED fiber and the schematic illustration (right panel) of the cross-section structure. Reprinted with permission from reference [67]. Copyright 2022, Wiley-VCH GmbH.

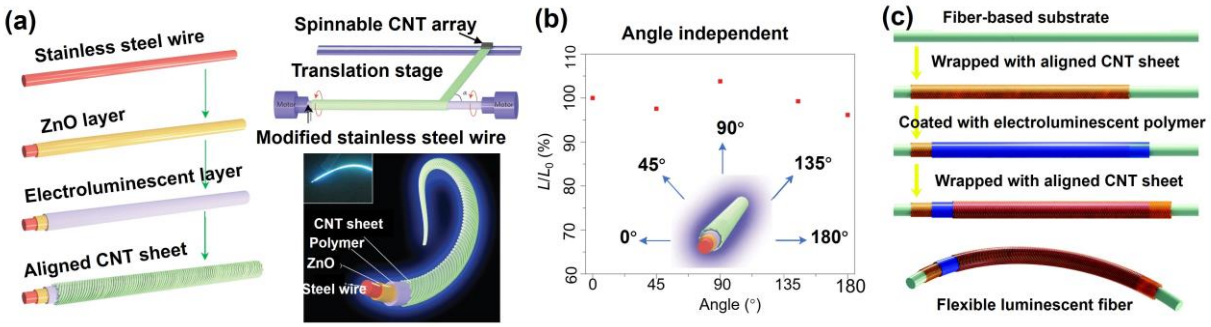


Fig. 3 (Color online) PLEC fibers. (a) Schematic fabrication of a PLEC fiber. First, a layer of ZnO is dip-coated on the stainless-steel wire, and then a layer of electroluminescent polymer is applied. Finally, the arranged CNT sheets are continuously wound on the modified fiber to finally obtain the PLEC fiber. (b) Angle-independent brightness of the PLEC fiber, where L_0 and L are the luminance measured at 0° and a specific angle, respectively. (a–b) Reprinted with permission from reference [36]. Copyright 2015, Springer Nature. (c) Schematic fabrication of a PLEC fiber using the aligned CNT sheet as the cathode and anode. Reprinted with permission from reference [72]. Copyright 2015, The Royal Society of Chemistry.

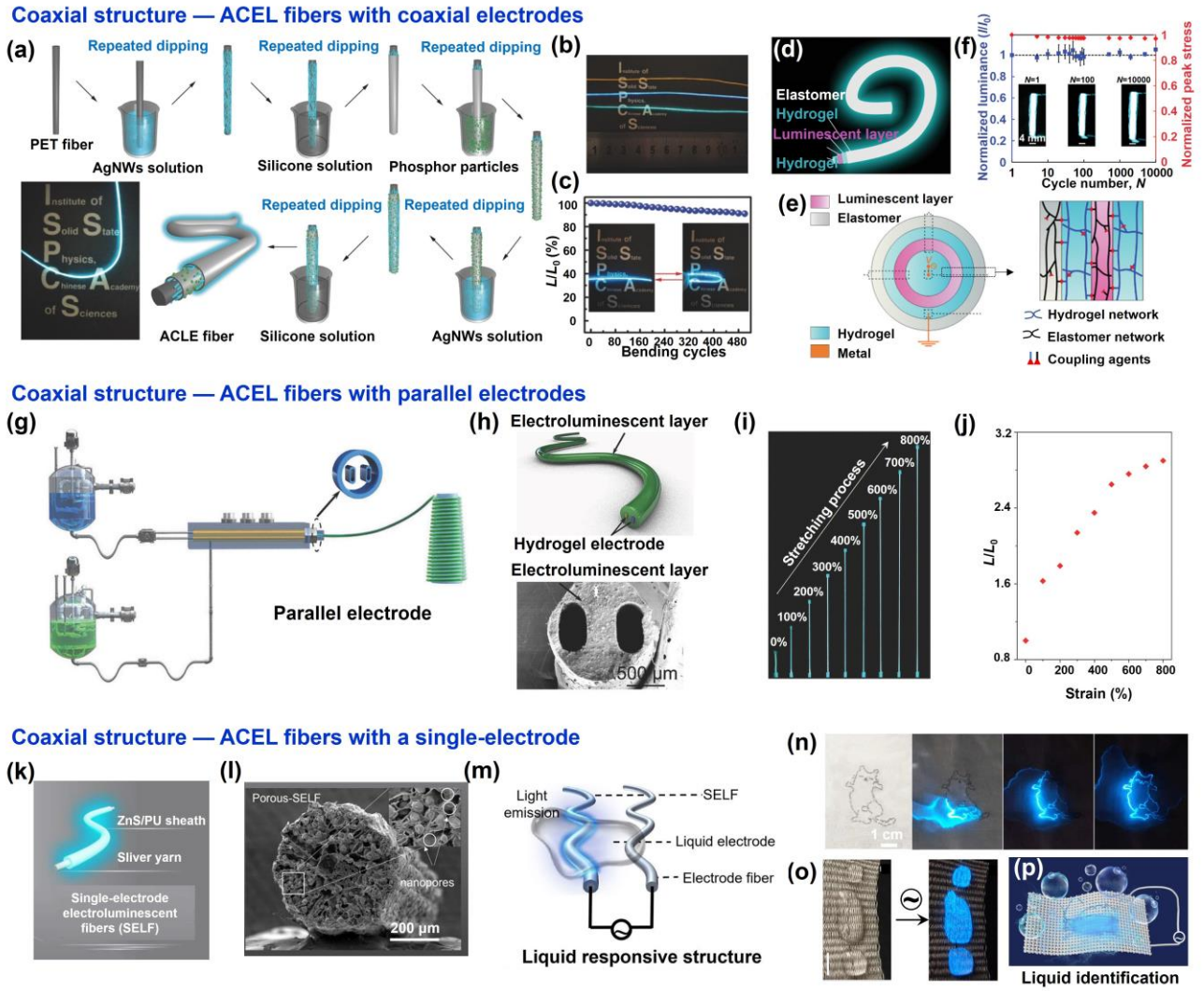


Fig. 4 (Color online) ACEL fibers with coaxial electrodes (a–f), parallel electrodes (g–j) and a single electrode (k–p). (a) Schematic of fabrication of ACEL fibers with coaxial electrodes through all-solution-based dip coating. (b) Photograph of ~12 cm long ACEL fibers with red, green, and blue light emission. (c) Recovery of flexible ACEL fiber after 500 bending cycles, where L_0 and L are the luminance in the original state and after a given cycle of bending, respectively. The insets are the fiber's luminance in the original (left) and bent (right) states. (a–c) Reprinted with permission from reference [42]. Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Schematic structure of an elastic ACEL fiber with coaxial hydrogel electrodes. (e) Left: schematic structure of the ACEL fiber, with a luminescent layer sandwiched between coaxial hydrogel electrode coated with an outer elastomer layer; right: the crosslinked network in the hydrogel fiber and the interconnected polymers between different layers.

(f) Cyclic tensile performance test of elastic ACEL fiber (stretch ratio = 1.5). The insets are photos of the fibers stretched 1.1×10^3, and 1×10^4 times. (d–f) Reprinted with permission from reference [79]. Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (g) The fabrication of an ACEL fiber with parallel hydrogel electrodes in a sequential one-step extrusion process. (h) The schematic structural diagram (up) and SEM image (bottom) of an ACEL fiber in which inner parallel hydrogel electrodes are surrounded by an outer electroluminescent layer. Photographs (i) and corresponding luminance ratio change (j) of the ACEL fiber being stretched from 0% to 800%. (g–j) Reprinted with permission from reference [81]. Copyright 2020, Wiley-VCH GmbH. (k) Schematic structural diagram and (l) SEM image of a single-electrode ACEL fiber with the sliver yarn core as the electrode and a ZnS/PU sheath as the luminescent active layer. (m) Schematic illustration of an LE device using a conductive liquid as the bridging medium. (n) The lighting process occurs when water seeps into the embroidered “Cat”. (o) The images of water droplets on the LE fabric before (top) and after (bottom) application of an external electric field. Scale bars, 4 mm. (p) Schematic illustration of the liquid recognition properties of a conductive-liquid-bridged LE fabric. (k–p) Reprinted with permission from reference [82]. Copyright 2023, American Chemical Society.

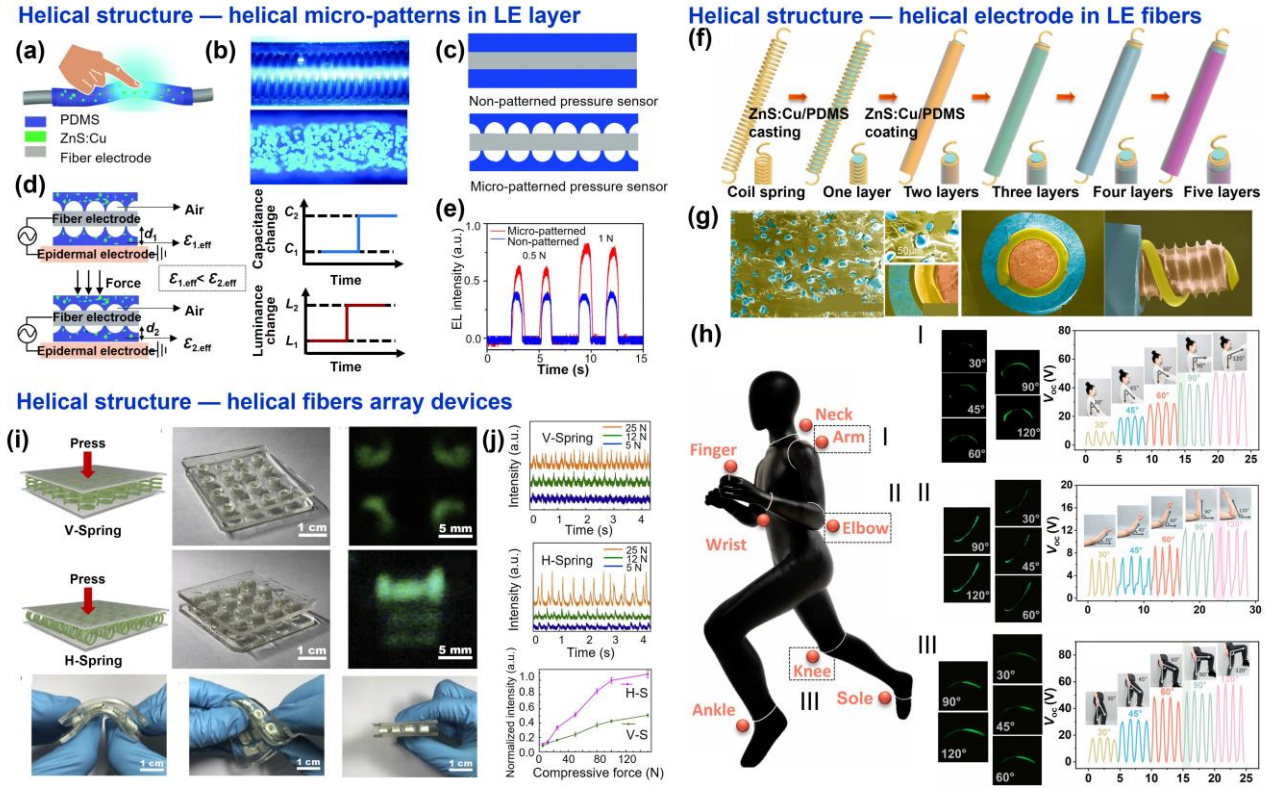


Fig. 5 (Color online) Mechanical responsive LE fiber with helical structures. (a) Schematic structural diagram of a pressure-responsive LE fiber. (b) Optical micrographs of a bare PDMS hollow fiber with helical micro-pattern (top) and PDMS hollow fiber containing ZnS:Cu particles under UV light irradiation (bottom). (c) Schematic illustration of the cross-sectional structure of pressure-sensitive fibers with (bottom) and without (top) helical micro-pattern. (d) Schematic working principle of the pressure-sensitive fibers with helical micro-pattern and changes in capacitance and luminance under external force. (e) Luminance changes of pressure-responsive fibers with and without helical micro-patterns under different forces of 0.5 N and 1 N. (a–e) Reprinted with permission from reference [51]. Copyright 2020, The Royal Society of Chemistry. (f) Schematic structure and fabrication process of a tensile-responsive LE fiber. (g) SEM images of surface morphology of mechanosensitive LE fibers doped with ZnS:Cu particles (blue) in PDMS (yellow). (h) Dual electrical and luminescence signal response to monitor physiological signals throughout the body, including fingers, wrists, neck, arms, elbows, knees, ankles, soles, and other joint movements. Motion detection of the arm (I), elbow (II), and knee (III). (f–h) Reprinted with permission from reference

[52]. Copyright 2021, Elsevier Ltd. (i) Schematics and photographs of a mechanosensitive luminescent devices with vertical (V–Spring) or horizontal (H–Spring) arrangement of helical LE fibers. It glows under pressure and the device remains stable under bending and twisting. (j) Comparison of the luminescence response of the device with V– and H–Spring array under different forces. (i–j) Reprinted with permission from reference [53]. Copyright 2019, Science China Press.

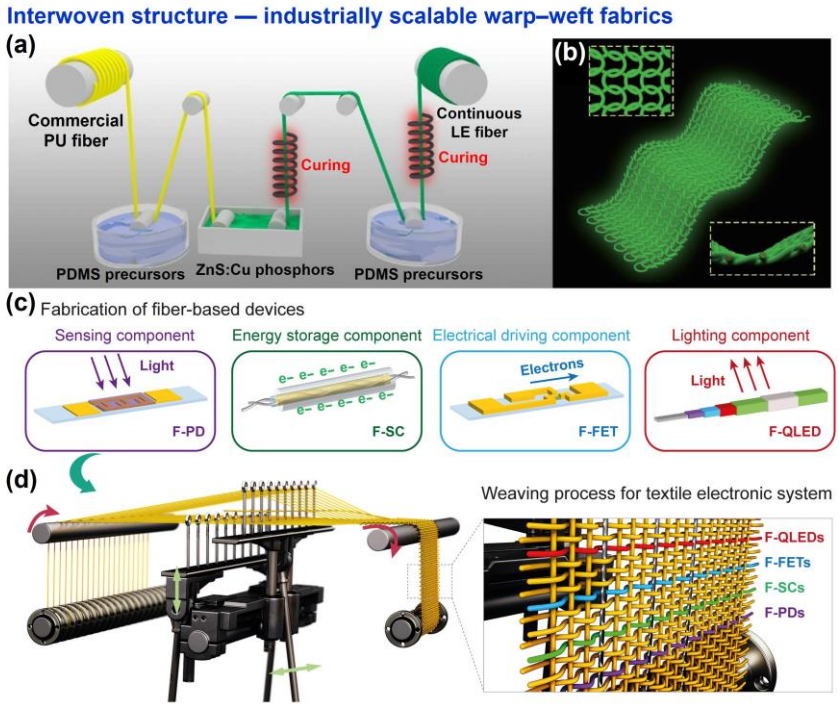


Fig. 6 (Color online) Continuous manufacturing and automated integration of LE fabrics. Schematic illustration of the continuous spinning process to produce (a) the meter-scale LE fiber and (b) the LE fabric with a weft–knit structure. Reprinted with permission from reference [89]. Copyright 2023, Tsinghua University Press. (c) Fiber components for integration into the electronics textile, include the sensing component F-PD, the energy storage component F-SC, the electric actuation component F-FET, and the lighting component F-QLED. (d) Schematic illustration of an automated weaving process for integrating the above fiber mentioned components into textiles. Fiber components and conductive threads are inserted along the weft and warp directions respectively. (c–d) Reprinted

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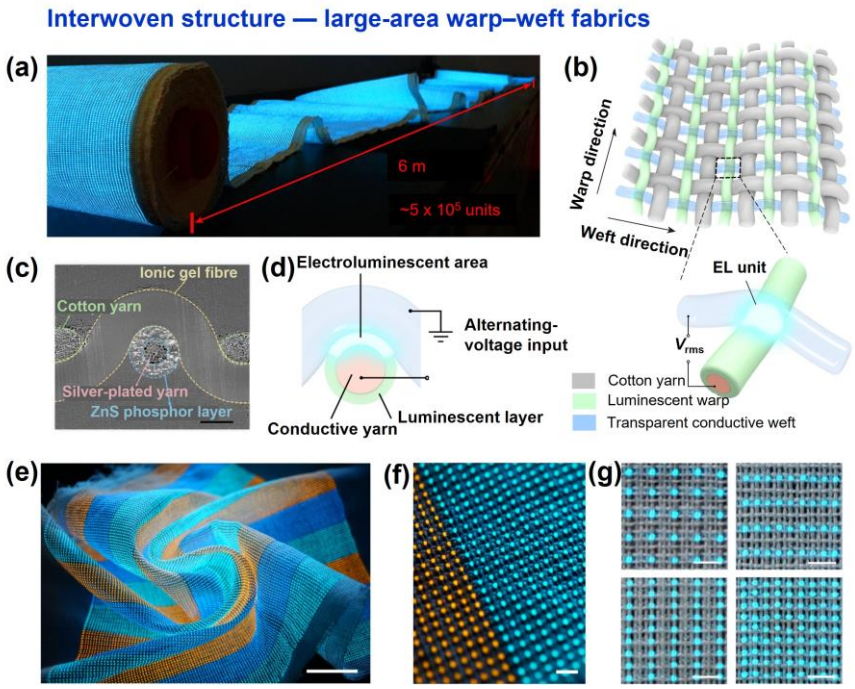

Fig. 7 (Color online) Large-area LE fabrics. (a) Photograph of a 6 m long LE fabric with ~5×10^5 electroluminescence units. (b) Schematic structure of LE fabric and electroluminescence mechanism of cross pixels. (c) Cross-sectional SEM image of fabric with interwoven structure. Scale bars, 200 μm. (d) Electroluminescence units (EL) at the contact points of the luminescent warp yarns and the conductive weft yarns emit light under AC voltage. (e) Photograph and (f) the enlargement of the three-color LE fabric. Scale bars, 2 cm. (g) Photographs of LE fabrics with electroluminescence units at different distances, made with different weaving parameters. Scale bars, 2 mm. Reprinted with permission from reference [54]. Copyright 2021 Springer Nature.

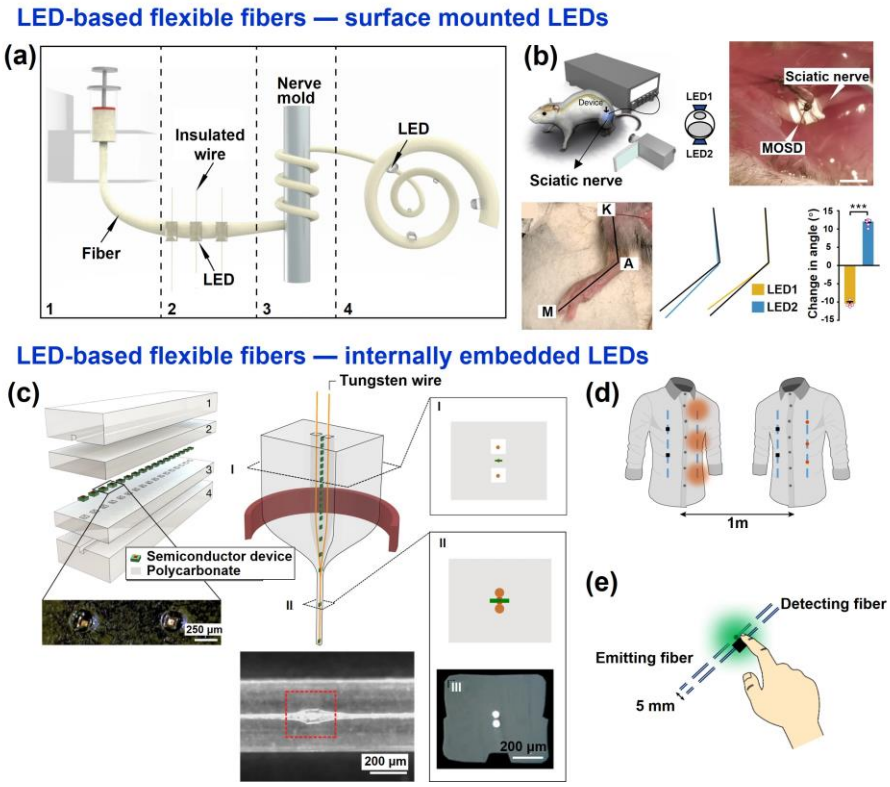

Fig. 8 (Color online) LED-Based Flexible prepared by surface mounting and internal embedding methods.

(a) Schematic design and structure of surface-mounted LED device. 1: a polyurethane fiber was first fabricated; 2: multiple mini-LEDs were attached to the fiber; 3: the resulting fiber was shaped into a spiral shape through a nerve mold; 4: the ultimate LED-based flexible device. (b) The device was implanted into an anesthetized mouse and wrapped around the sciatic nerve, and the motion of the ankle joint in response to mini-LED was induced. (a–b) Reprinted with permission from reference [96]. Copyright 2019 Springer Nature. (c) Schematic fabrication and structure of long LED-based fiber. The cross-section schematic images and optical micrograph images show that the devices and wires are well embedded in the fiber cladding, and the LEDs are well in contact with the wires. (d) The LE fibers and light-detecting fibers enable bi-directional communication between the two garments. Light from the LE fiber (light blue dashed line with red circles) is detected by the light detection fiber (light blue dashed line with black squares). (e) Application of luminescence and photodetection fabrics to pulse measurements. Placing a finger on LE fibers and photo-detecting fibers to record the reflected light. The light reflected by human skin is sensitive to blood circulation, which is associated with a pulse. (c–e) Reprinted with permission from reference [14].

Author biography

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