Intrinsically stretchable electroluminescent materials and devices

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Abstract:

Intrinsically stretchable electroluminescent (EL) devices have emerged as pivotal components with transformative potential in various domains, including wearable technology, medical devices, human-machine interfaces, and communications. This mini-review focuses on the recent progress in the development of intrinsically stretchable EL materials, highlighting milestones and breakthroughs in the field. The article discusses the basic principles, advantages, and disadvantages associated with various EL mechanisms and materials. Specific material design strategies, particularly focusing on light-emitting layers, are thoroughly examined, detailing their implementation in EL devices and the resultant EL performance. We also provide perspectives on the active challenges and future research needs for each type of EL materials and devices for achieving stretchable designs, together with some insights into the future trajectory of stretchable EL technology.

Key words: stretchable electronics, electroluminescent materials, electroluminescent devices, light-emitting diodes, thermally activated delayed fluorescence

1. Introduction

The field of stretchable electronics has garnered significant attention for its disruptive capabilities in revolutionizing diverse domains such as wearable technology, medical devices, human-machine interfaces, and communications[1-11]. Within this realm, stretchable electroluminescent (EL) devices have emerged as one of the most pivotal devices with immense potential for a wide range of applications[12-14], such as on-skin displays[15-18], optical sensors for monitoring oxygen saturation[19], wearable imaging systems[20], and implanted optical stimulation for optogenetics[21], to name a few. The overarching development goal for stretchable EL devices is to combine high stretchability with high EL performance.

In recent years, significant progress has been made in the development of stretchable EL devices, driven by advances in materials science, nanotechnology, and device fabrication techniques. In the pursuit of stretchable EL devices, two prominent strategies have emerged: strain engineering on conventional rigid materials/devices, including the utilization of spring-like (e.g., wrinkle[22, 23], pre-stretched buckle[24-26], or serpentine configurations[27, 28]), rigid-island[11, 29-31], or fiber/textile structures[32-34]; and materials engineering for achieving intrinsic stretchability[16, 17, 35].

Strain engineering strategies involve engineering the material or device architecture to possess device-level stretchability. This general approach offers versatility and scalability to different materials and device structures, thereby enabling the fabrication of stretchable EL devices with various conventional rigid materials[11, 29]. As such, the achieved stretchable devices can best maintain the state-of-the-art EL performance typically seen in rigid devices[18, 30], positioning them as compelling contenders for the application in wearable electronics. However, the achieved stretchability often comes with sacrifice in several aspects. For instance, interconnected serpentine structures can lead to the sacrifice of EL resolutions, while wrinkled or buckled structures may result in suboptimal visualization effects. Also, the use of rigid materials with high Young's moduli renders limited mechanical compatibility with soft bio-tissues[36].

An alternative strategy to overcome these limitations involves the incorporation of intrinsic stretchability into EL materials and then devices. This design approach can both allow densely packed emissive pixels and accommodate large mechanical deformations. In addition, the moduli of such intrinsically stretchable devices are often much lower than rigid devices[35]. At the heart of this strategy is the development of novel materials that possess both excellent EL performance and mechanical stretchability. Recent advancements in intrinsically stretchable EL materials have opened up new avenues for research and development, leading to exciting breakthroughs and promising applications[16, 17, 35].

Recent articles have already provided comprehensive reviews of the development of stretchable EL devices with the strain engineering strategy[37-41]. This mini-review will focus on the recent progress in the development of intrinsically stretchable EL materials and devices. Figure 1 illustrates the milestone progresses of the field starting from 2011 when Pei and colleagues presented the pioneering intrinsically stretchable EL device based on fluorescent emitter[42]. Herein, we will first briefly discuss the basic principles and pros/cons associated with various types of EL devices and light-emitting materials. Then we will delve into specific material design strategies, with a specific focus on emitting layers (EML). Furthermore, we will explore the implementation of these EMLs in EL devices and discuss their EL performance.

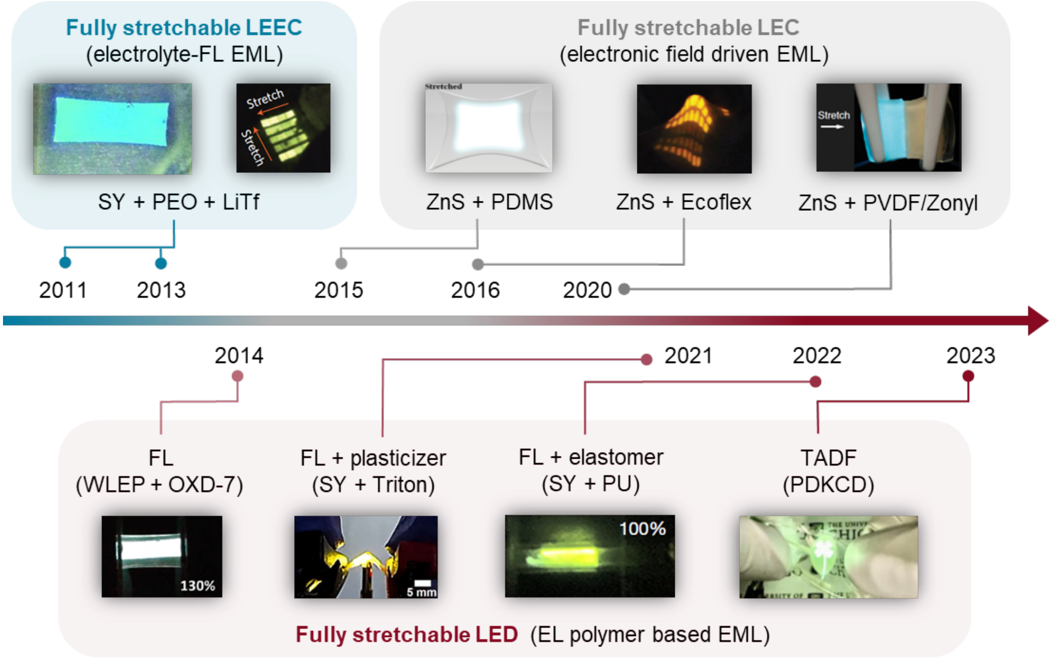

Figure 1| Timeline and key milestones of the development of intrinsically stretchable EL devices based on different EML designs. LEEC: light-emitting electrochemical cells; LEC: light-emitting capacitors; LED: light-emitting diodes. Reproduced with permission from ref [42]. Copyright 2011 Wiley-VCH. Reproduced with permission from ref [43]. Copyright 2013 Springer Nature Limited. Reproduced with permission from ref [44]. Copyright 2014 American Chemical Society. Reproduced with permission from ref [35]. Copyright 2021 AAAS. Reproduced with permission from ref [16]. Copyright 2022 Springer Nature Limited. Reproduced with permission from ref [17]. Copyright 2023 Springer Nature Limited. Reproduced with permission from ref [45]. Copyright 2015 Wiley-VCH. Reproduced with permission from ref [46]. Copyright 2016 AAAS. Reproduced with permission from ref [47]. Copyright 2020 Springer Nature Limited.

2. Working mechanisms of EL devices and emitters

To date, intrinsically stretchable EL devices have been investigated based on different working mechanisms on the device and emitter levels. Before reviewing the strategies for incorporating stretchability into EL materials and devices, it can offer values to first discuss their working principles. In this part, we will briefly introduce those distinct principles and comment on their advantages and disadvantages.

According to the different device working mechanisms, there have been mainly three kinds of intrinsically stretchable EL devices: light-emitting electrochemical cells (LEEC) (Figure 2a), light-emitting capacitors (LEC) (Figure 2b), and light-emitting diodes (LED) (Figure 2c).

LEEC was initially designed to simplify the device architecture found in conventional LED by achieving efficient charge injection with fewer device layers (Figure 2a)[48, 49]. The phototype LEEC consists of an active ion-electron mixed conducting EML sandwiched between two electrodes. Usually, the EML is a composite of several components that include EL-active materials, ionic conducting materials (i.e., electrolytes), and salts, as well as other binder/host additives that are necessary for efficient light emission. Under bias voltages exceeding the bandgap (i.e., E_g/e) of the electronic conducting materials, the electrochemical doping process occurs within the EML, leading to n-/p-doping of the semiconductor at the anode/cathode interfaces, respectively, thereby promoting the injection of electrons/holes[50]. Apart from the electrochemical doping process, LEEC works similarly to OLED in terms of charge transport, recombination, and emission. Nevertheless, the doping process is relatively slow, thereby limiting the operational speed of LEEC. Typically, after a bias is applied, LEEC exhibits a delay of several minutes before turning on. Upon the removal of the bias, the resulting p-i-n structure that represents a metastable equilibrium state will gradually vanish. Regarding the state-of-the-art EL performance, LEEC's light-emitting brightness and efficiency are still much lower than LED, even when using the same light-emitting materials.

LEC typically features a capacitor-type device structure (Figure 2a), with an active layer of luminescent phosphor micro/nanoparticle embedded in a dielectric matrix sandwiched between two electrodes. LEC operates under a hot-electron injection and impact-excitation mechanism. Specifically, an alternating current (AC) voltage is applied on the two electrodes to generate a high electric field across the dielectric layer, which polarizes the medium surrounding the luminescent phosphor and stimulates the phosphors to the excited state for the photon emission. Generally, the dielectric layer must be thick enough ($> 50\ \mu m$) to prevent the leakage current. Meanwhile, high AC electric fields ($> 1\ V\ \mu m^{-1}$) are required to excite luminescent centers to achieve decent brightness ($> 100\ cd\ m^{-2}$), which leads to a high working voltage with a high operation frequency (> 1 kHz). It is reported that dielectric materials with high dielectric constants can effectively lower the working voltages[47, 51, 52]. For such voltage-driven (rather than current-driven) EL operations, the requirements for the conductivity of electrodes are more relaxed, thereby giving more options for stretchable designs. Benefiting from the relatively simple device architecture and abundant materials selections, LEC has been imported with superhigh stretchability (> 500%). In addition, the switching speed of LEC can be incredibly fast, typically reaching sub-millisecond levels. However, along with the high working voltage and high operation frequency, low efficiency is another obstacle that limits LECs in real applications.

LED discussed in this review mainly includes organic LED (OLED)[53-55], quantum dot LED (QLED)[56, 57], and perovskite LED (PeLED)[58-60], which are categorized based on EMLs. They have similar device working mechanisms and device architectures (Figure 2a), typically with a multi-layer structure for hole/electron injection, transportation, and recombination. Generally, the holes/electrons are injected from anode/cathode electrodes, transported in hole/electron transporting layers (HTL/ETL), and recombined into the excitons in the EML followed by the light emission from the decay of the excitons. The energy barriers at the interfaces between different layers need to be precisely controlled for efficient charge injection and exciton confinement in the EML, which usually involves additional hole/electron injection (HIL/EIL) and blocking layers (HBL/EBL) in the device structure to reach high efficiency. Because of such complex device structures with various functional layers, achieving high stretchability in LED is more challenging than LEEC and LEC. However, OLED/QLED intrinsically exhibits low operation voltage (< 10 V), high efficiency (100% internal quantum efficiency (IQE)[55, 61], over 30% external quantum efficiency (EQE))[62-65], and high working frequency (hundreds of MHz)[66], which give an attractive future for various stretchable EL applications. Owing to these advantages, so far, LED has been widely used as the most advanced EL technology for lighting, displays, and imaging.

The key luminescent materials unutilized in the EL devices can be classified into two groups, inorganic and organic emitters. Inorganic emitters, such as traditional phosphors (e.g., ZnS), transition metal dichalcogenides (e.g., WSe_2, MoS_2), perovskite, quantum dots (e.g., CdS), etc., are not inherently stretchable and need to blend with elastomers to realize stretchability, which is often accomplished with sacrificed EL performance. According to the ability and mechanisms for harvesting excitons, organic (including polymeric) emitters are generally classified into three generations, as shown in Figure 2d. In EL processes, excitons formed by injected electrons and holes are composed of singlet excitons with zero spin, and triplet excitons with non-zero spin, which make up 25% and 75% of all the excitons, respectively[67-69]. The first-generation emitters, so-called fluorescent (FL) emitters, can only use radiative singlet excitons for light emission, resulting in a maximum IQE of 25%.[70] The second-generation emitters are phosphorescent (PH) emitters which incorporate heavy metal elements in emitters' chemical structures to induce strong spin-orbit coupling, enabling direct triplet emissions and thereby achieving a 100% IQE[61]. Besides, the third-generation emitters based on the mechanism of thermally activated delayed fluorescence (TADF) can also harvest triplet excitons for radiative decay, thereby achieving 100% IQE[55]. For TADF, non-radiative triplet excitons can convert into singlet excitons by an efficient reverse intersystem crossing (RISC) process enabled by significantly reduced energy level splitting (ΔE_{ST}) between singlet (S_1) and triplet (T_1) excited states. Such TADF emitters can avoid the need to incorporate heavy metal ions, which helps to ensure minimal biological/environmental toxicity and lower prices. These attributes are highly desirable for human-integrated applications.

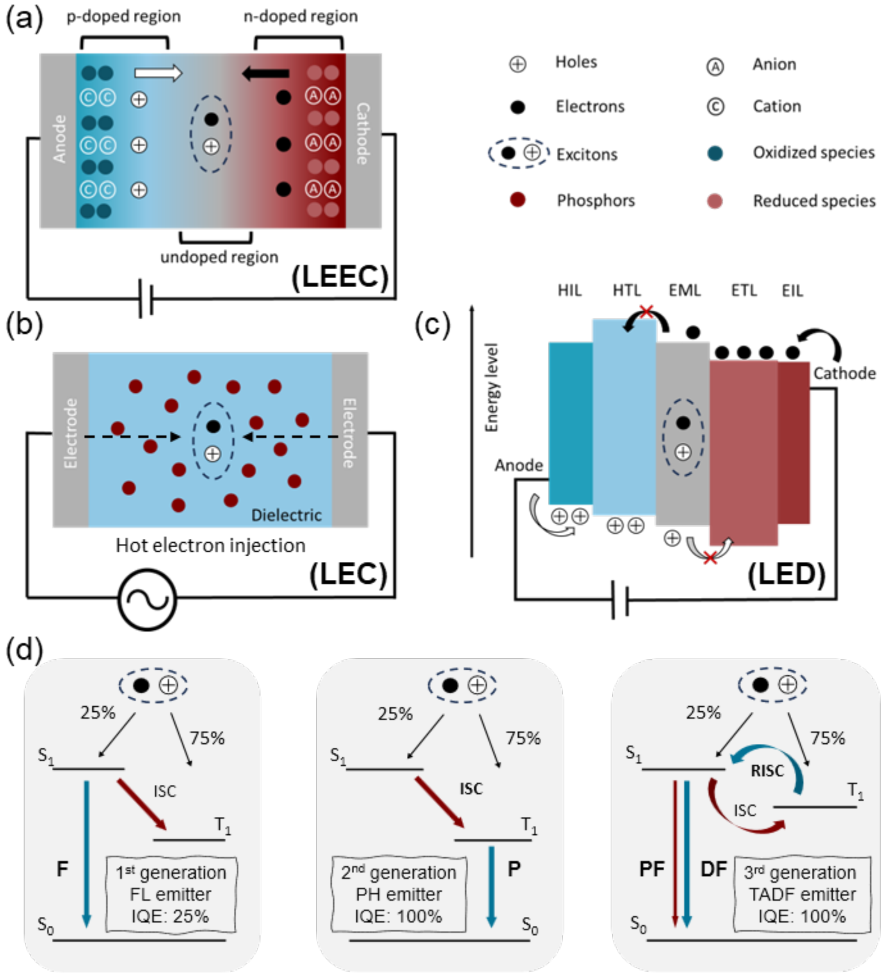

Figure 2 | Working mechanisms of different EL devices. (a) LEEC. (b) LEC. (c) LED. (d) Working mechanisms of three generations of organic emitters: FL (fluorescent), PH (phosphorescent), and TADF (thermally activated delayed fluorescence). S_1, T_1, and S_0 stand for the first-excited singlet state, first-excited triplet state, and ground-state energy levels, respectively. ISC, RISC, F, P, PF, and DF stand for intersystem crossing, reverse intersystem crossing, fluorescence, phosphorescence, prompt fluorescence, and delayed fluorescence, respectively.

To date, the commercialization and development of OLED technology have been dominated by small molecules. However, for achieving stretchability, polymer designs provide the most distinct promise due to their inherent flexibility, unique intermolecular entanglements, versatile chemical and physical tunability, as well as solution processability and cost-effectiveness. So far, research efforts have been made to create polymeric emitters based on different mechanisms, including FL, PH, and TADF. However, the reported EL polymers usually contain a conjugated backbone, leading to relatively rigid polymer chains and low polymer chain dynamics. So, the EL polymers are naturally not very stretchable. For example, the film made of mostly well-known EL polymer, poly(1,4-phenylenevinylene) (Super Yellow, SY), will generate cracks at strain around 40%[43], which is less than skin deformations up to 50% strain. Therefore, innovations in material designs are needed to significantly increase the stretchability of EL polymers, while keeping high light-emitting efficiencies.

3. Strategies for realizing stretchable EL materials and devices.

3.1. Stretchable light-emitting electrochemical cells (LEEC)

Because of the simplicity of device structures, historically, the efforts in intrinsically stretchable EL devices started from LEEC devices.

3.1.1. Intrinsically stretchable EMLs for LEEC

For use in LEEC, EMLs need to have mixed ion-electron conducting properties, together with light-emitting properties. So far, there have been mainly two strategies for realizing stretchable EMLs for LEEC: 1) blending EL polymers with electrolyte, which serves as the plasticizing agent, and 2) blending ionic EL small molecules into an elastomer such as polydimethylsiloxane (PDMS).

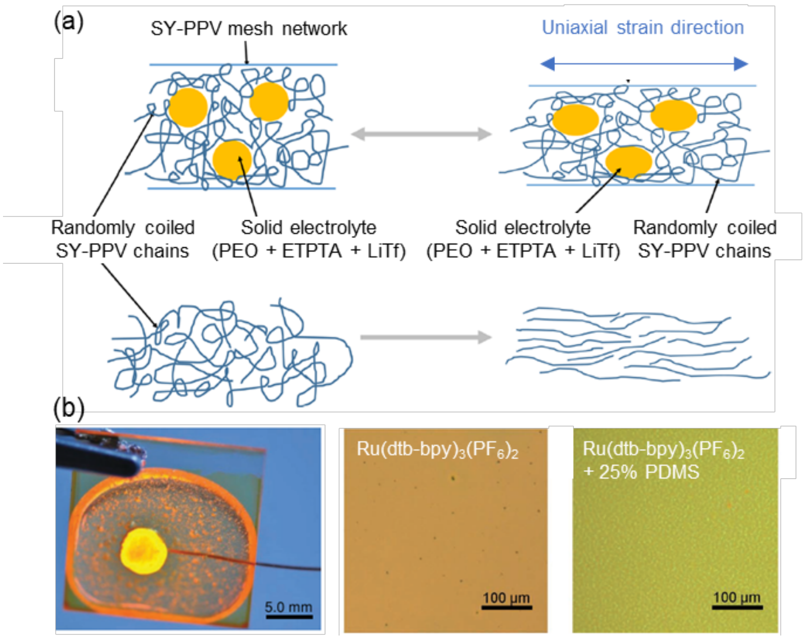

Figure 3 | The designs of intrinsically stretchable EMLs for LEEC. (a) Schematic view of the phase morphology of an IPN film in response to uniaxial stretching and nest SY in response to uniaxial stretching. Reprinted with permission from ref [71]. Copyright 2016 American Chemical Society. (b) Photograph of the LEEC device (left). Optical microscopy image of a Ru(dtb-bpy)$_3$(PF6)$_2$ film on ITO-coated glass with no polymer matrix (middle). Optical microscopy image of a Ru(dtb-bpy)$_3$(PF6)$_2$ film containing ~25% PDMS (right). Reprinted with permission from ref [72]. Copyright 2016 Wiley-VCH.

3.1.1.1. EL polymers with electrolyte

To realize stretchable emitter for LEEC, one reported strategy is to blend solid electrolytes, such as poly(ethylene oxide) (PEO) and lithium trifluoromethanesulfonate (LiTf), into an EL polymer matrix to form an interpenetrating polymer network (IPN)[71], as shown in Figure 3a. In this morphology, EL polymers form a porous network with pores filled by the PEO phase. The two domains are separate and evenly distributed over the film due to inter-chain excluded volume, which facilitates the independent charge (electron/hole) and ion transport in their own phases with continuous pathways. Under strain, the electrolyte acts as a plasticizer for the EL polymers, allowing the IPN film to withstand greater strain compared to the pristine EL polymer.

The first example of this design strategy dates to 2011[42]. Yu et al. reported a fully stretchable LEEC based on a stretchable EML that combines polyfluorene copolymer (PF-B) as the FL polymer, ion conductor (poly(ethylene oxide) dimethacrylate (PEO-DMA)) as the electrolyte, and lithium trifluoromethanesulfonate (LiTf) as the salt. After that, similar design strategies based on the SY polymer and PEO were reported in multiple works[43, 71, 73]. It was reported that the stretchability of the IPN film based on SY, PEO, exoxylated trimethylolpropanetriacrylate (ETPTA), and LiTf show excellent stretchability without crack formation after 1,000 strain cycles between 0 and 50%.[43, 71] To increase the stretchability of the IPN film limited by PEO, Liu et al. employed a supramolecular polymer that consists of polytetramethylene glycol, tetraethylene glycol soft domains, and reversible quadruple H-bonding cross-linkers as the ion conductor, which blended with SY, ETPTA, and LiTf[74]. It was found that the stretchability of the blended film increased from less than 10% to nearly 50% by increasing the ratio between ionic polymer/SY from 0 to 2.

3.1.1.2. Ionic EL molecules with elastomers

A slightly different strategy than the above works is to use an ionic emitter to blend with an elastomer, which, to the best of our knowledge, has been demonstrated by just one work so far. Filiatrault et al. incorporated ionic PH emitter $Ru(dtb\text{-}bpy)_3(PF_6)_2$ (dtb-bpy = 4,4′-di-tert-butyl-2,2′-dipyridyl) into a PDMS matrix to construct stretchable EL layers for LEEC[72] (Figure 3b). It was found that the film quality of the composite improved compared with the pure emitter due to the polymer matrix, which potentially can prevent the problematic current leakage in the devices because of the pinhole in the film. However, this composite EML shows a low maximum EQE of 1% in the rigid devices. Even though this strategy is straightforward for realizing stretchable EML for LEEC, the charge transport of such small-molecule-based emitters is more vulnerable to the blended elastomer, which results in a greater trade-off between EL performance and stretchability.

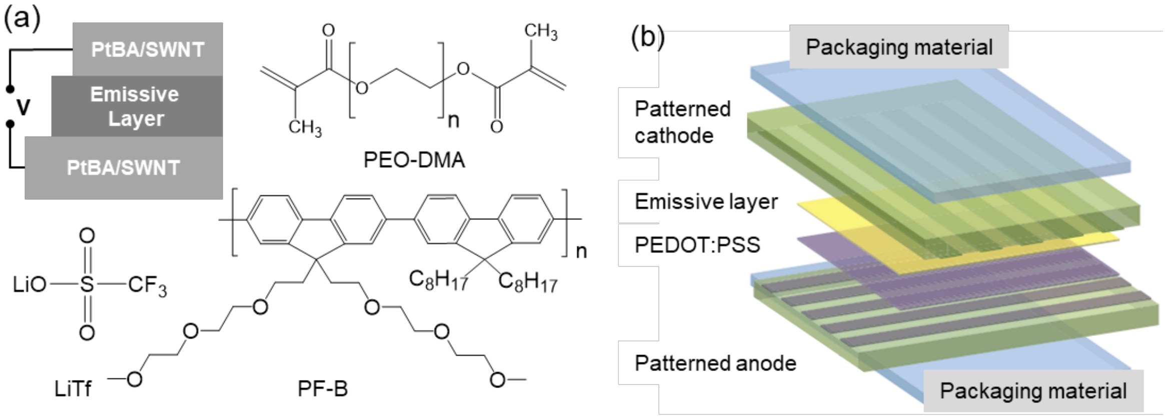

Figure 4 | Fully stretchable LEEC devices. (a) Fabrication process of a LEEC using SWNT-polymer composites as both electrodes, and the chemical structures of materials in the emissive layer:

polyfluorene copolymer, PF-B, PEO-DMA, and LiTf (left figure). Current density-voltage-luminance responses of a PLEC device before and after flexing to a radius < 1 mm (right figure). Reproduced with permission from ref [42]. Copyright 2011 Wiley-VCH. (b) Schematic illustrations of an encapsulated fully stretchable LEEC display (left figure). Current density–luminance–driving voltage characteristics of an elastomeric LEEC device (right figure). Reproduced with permission from ref [43]. Copyright 2013 Springer Nature Limited.

3.1.2. Device designs for fully stretchable LEEC

As LEEC has relatively simple device designs, fully stretchable LEEC devices can be made by combining stretchable EMLs with stretchable conductors for the two electrodes. In the work by Yu et al., the EML was sandwiched between two electrodes to construct fully stretchable LEEC devices (Figure 4a)[42]. Both anode and cathode are made of the composite of single-walled carbon nanotube (SWNT) and elastomer poly(tert-butyl acrylate) (PtBA). This device achieved a turn-on voltage of 4.8 V and a maximum efficiency of around 1.24 cd A^{-1} at 200 cd m^{-2}, and could be stretched to 45% strain without efficiency loss and moderate luminance decrease. To pursue better device performance, electrodes can be replaced with more conductive options, such as a thin silver nanowire (AgNW) network inlaid in the surface layer of a rubbery poly(urethane acrylate) (PUA) matrix[43]. A thin layer of PEDOT:PSS was coated on the anode side to protect the PUA matrix from being dissolved by following EML coating. The device exhibited a turn-on voltage of 6.8 V, a maximum brightness of 2200 cd m^{-2} at 21 V, the highest current efficiency of 5.7 cd A^{-1}, and a very high stretchability of 120% (Figure 4b). Similar device structures can also be found in two other works[71, 74]. Besides the optimization of electrodes, it was reported that solvent-vapor-assisted lamination (SVAL) can significantly reinforce weakly laminated interface and improve the device performance. The resulting device achieved a record low turn-on voltage of 3.7 V with a maximum luminance of 1754 cd m^{-2} at 11 V, which can endure 30% strain without much significant luminance change[73].

So far, most of the studies on fully stretchable LEEC have primarily focused on optimizing device performance, including efficiency, brightness, working voltage, and stretchability. However, there has been little attempt to address the stability issue of these devices. Fully stretchable LEEC devices are susceptible to moisture and oxygen, necessitating hermetic encapsulation. Without appropriate encapsulation technology, the devices would exhibit poor air stability with a T_{50} (the lifetime of luminance degraded to 50% of the initial value under constant driving) of less than 2 minites[73]. In contrast, a similar LEEC tested in the dry nitrogen-filled glovebox could have a T_{50} as long as 10 hours. With proper sealing using a thermally crosslinked polyurethane (TCPU), LEEC devices could work outside the glovebox. However, the storage lifetime of such TCPU-sealed devices in air was only about one week, and no corresponding device lifetime was studied[43]. Besides, the rough surface of the electrodes also significantly decreases the device stability[43].

In summary, though studies on stretchable LEEC materials and devices started relatively earlier than other types of EL devices, the strategies to achieve intrinsically stretchable EMLs are mostly just based on blending EL polymers with electrolytes. Therefore, it is necessary to explore diverse designs of EL materials that can enhance the efficiency and brightness of the LEEC devices. Besides, the relatively low response speeds (usually several minutes) of LEEC devices, which are caused by slow electrochemical doping processes, need to be addressed. More importantly, to improve the air stability of the devices, there is an urgent need to develop more stable EMLs, electrodes with smooth surfaces, and most importantly, advanced encapsulation techniques.

3.2. Stretchable light-emitting capacitors (LEC)

As we discussed in section 2, LEC operates through the excitation of phosphors under a high electric field. A typical design of LEC-EMLs is to blend EL nano/microparticles, e.g., ZnS, into insulating matrices. To achieve stretchable properties, a straightforward strategy is to use dielectric elastomers as such insulating matrices. Silicone elastomers are found to be good choices owing to their low modulus and facile miscibility with ZnS phosphors. In one of the early works, Wang et al. reported a strategy of blending ZnS nanoparticles into a PDMS matrix, which showed excellent stretchability with a sustained EL performance under strain up to 100% (Figure 5a)[45]. Later, Ecoflex is also used as the dielectric matrix to realize even softer and more stretchable properties (Figure 5b)[46]. However, the dielectric constants (*k*) of silicone elastomer are relatively low (i.e., 2.3-2.8), which is unfavorable for creating a high electrical field on the phosphors, thereby leading to low luminance of less than 250 cd m^{-2} under a high operation voltage of 300 V.

To address this issue, the overall strategy is to increase the dielectric constant of the matrix, by either blending high-*k* additives or using high-*k* elastomers. As an example of the former strategy, barium titanate ($BaTiO_3$) microparticles were added to the phosphor-PDMS blends, which increases the overall dielectric constant by more than 2 folds[51]. Benefiting from this, the luminescent brightness has a remarkable increase from 12 cd m^{-2} to 121 cd m^{-2} at a working voltage of 250 V. And the fully stretchable LEC device keeps stable emission even after 500 stretching cycles to 50% strain. Another demonstrated method for increasing the dielectric constant is to replace silicone elastomers with high dielectric constant elastomers such as poly((vinylidene fluoride)-hexafluoropropylene) (PVDF-HFP)[47, 52, 75]. In the work by Tan et al, the reported composite of (PVDF-HFP) and fluorosurfactant (FS-300) achieved very high dielectric constants of 10 to 27, which is 2.5- to 5.8-fold higher than that of silicone elastomers (Figure 5b). The resulting fully stretchable LEC achieved a low turn-on voltage of 23 V and a maximum brightness of 1460 cd m^{-2} at 2.5 V um^{-1} with stable emission up to a maximum of 800% strain[47].

Due to the highly simple device structure and broader choices for electrode materials, fully stretchable LECs have been realized with very high stretchability and mechanical robustness[76, 77]. These enabled some proof-of-concept applications, such as EL skin display[15, 75] and soft robots with the function of signaling and tactile sensing[46]. Besides, fully stretchable LEC devices have been embodied into fiber[34, 78-80] or textile[81] structures for clothing-compatible wearable displays. In addition, such fiber and textile devices also achieved good stretchability, breathability, and washability for wearable displays (Figure 5c). That being said, it still remains challenging to increase brightness, improve efficiency, and reduce the working voltage for LEC.

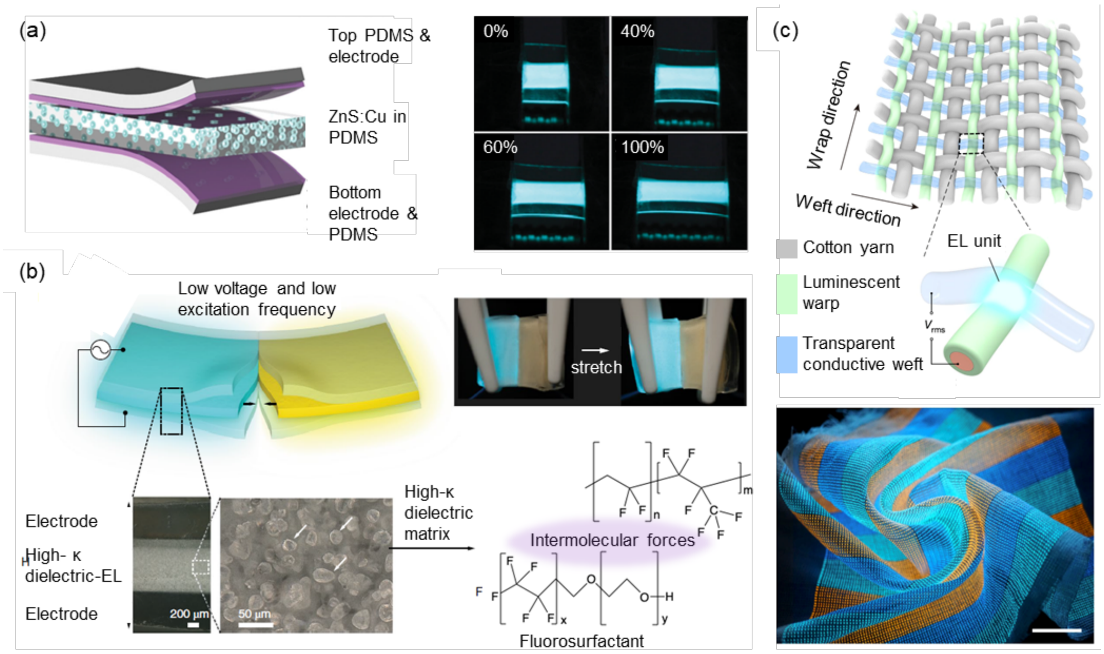

Figure 5 |EMLs and devices designs of fully stretchable LEC. (a) Schematic images showing the device geometry of the fully stretchable LEC (left). Photographs of the EL device stretched to strains of 0%, 40%, 60%, and 100% (right). Reproduced with permission from ref[45]. Copyright 2015 Wiley-VCH. (b) Schematic illustrating the fully stretchable and self-healable LEC device architecture. The optical micrographs (bottom left) show the three-layered fully stretchable LEC structure. The white arrows indicate phosphor particles. The high-κ dielectric matrix consists of a fluoroelastomer with non-ionic fluorinated surfactants. Reproduced with permission from ref[47]. Copyright 2020 Springer Nature Limited. (c) Schematic showing the weave diagram of the display textile with each contacting

luminescent warp and transparent conductive weft forms an EL unit (top). Photograph of a functional multicolour display textile under complex deformations, including bending and twisting (bottom). Reproduced with permission from ref [34]. Copyright 2021 Springer Nature Limited.

3.3. Stretchable light-emitting diodes (LED)

With LED being developed and utilized as the main technology for the current display and lightning industry, in recent years, the efforts in developing stretchable light-emitting devices have gradually shifted to LED, with the initial work published in 2014. For stretchable OLED, EML designs so far have been realized on four types of emitters: inorganic perovskite nano/microparticles, organic/polymeric FL, PH, and TADF emitters. To introduce stretchability into the EML of LED, a straightforward and efficient approach is to physically blend these emitters as either nano/microparticles, small molecules or polymers, with soft elastomer matrices. Besides, chemical engineering on the EL polymer's structure, either on the main chain or side chains, has been explored as another effective avenue to achieve stretchable EMLs. In this section, we will review the fundamental mechanisms underlying each strategy and discuss the main research findings.

3.3.1. Intrinsically stretchable EMLs by blending perovskite nano/microparticles with elastomers

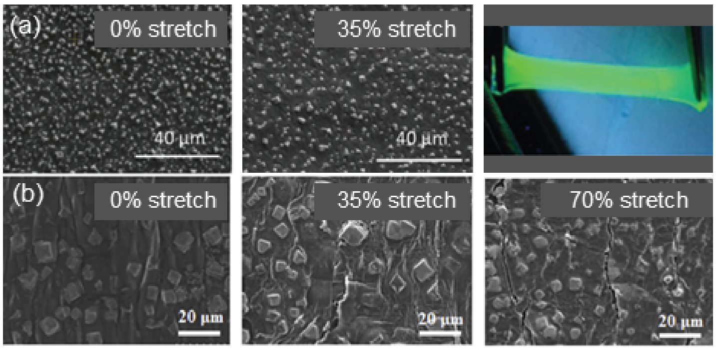

Figure 6 | Stretchable EMLs based on perovskite microparticles. (a) SEM images of the $MAPbBr_3$/PEO composite thin film before stretching and after stretching to 30% strain (left), and photos of the $MAPbBr_3$/PEO composite film under 365 nm UV irradiation before and after stretching to 100% strain (right). Reproduced with permission from ref [82]. Copyright 2017 Wiley-VCH. (b) SEM images of the $MAPbBr_3$/PEO composite film under tensile strain of 0 (left), 35 (middle), and 70% (right). Reproduced with permission from ref [83]. Copyright 2020 Elsevier.

While inorganic EL materials, such as perovskite are generally brittle, they often exhibit excellent EL properties, such as high PLQY, a wide range of tunable emission colors, pure emission with a narrow emission width, and high charge-carrier mobility[58]. In order to confer stretchability, emissive perovskite can be synthesized as nano/microparticles and then incorporated into an elastomer matrix. In 2017, Bade et al. reported a composite film consisting of methylammonium lead tribromide ($MAPbBr_3$) microparticles and PEO as the stretchable EML (Figure 6a)[82]. The composite shows a pin-hole-free film morphology with a dense distribution of the micrometer-size $MAPbBr_3$ crystals partially embedded into the PEO polymer matrix. Upon stretching, perovskite crystals exhibit continuums density decreases. Tears in the PEO polymer matrix start to appear at around 50% strain. The composite EML is able to withstand at least 100 cycles of repetitive stretching up to 40% strain without EL failure. Later, similar designs were reported in other works[83, 84]. These works showed that $MAPbBr_3$ crystals can maintain the luminescent properties during the stretching because it was less affected by the tensile strain inside the PEO matrix (Figure 6b).

For this type of EML design, although it offers a possibility to benefit from the excellent properties of inorganic EL materials, the achieved stretchability is still relatively limited. Also, there is still a lack of clear understanding of the design principles in terms of the choices of inorganic EL materials and elastomers to realize both high-performance EL properties and high stretchability. In addition, future developments can also benefit from the extended uses of more advanced forms of inorganic emitters, such as perovskite nanoparticles[85] and quantum dots[86], which could help to achieve even higher EL performance.

3.3.2. Intrinsically stretchable FL-based EMLs

Along the line of OLED technologies based on organic emitters, the development of stretchable emitters initially started from and so far has been mostly based on the first generation, i.e., FL emitters. The main reasons behind this are the structural simplicity and commercial availability of certain FL emitters (such as Super Yellow). The explored strategies for imparting stretchability can be divided into several categories, as summarized below.

3.3.2.1. Blending FL polymers with small-molecule plasticizers

Blending plasticizers (or surfactants) into conjugated EL polymers is a simple-to-implement strategy to increase stretchability by decreasing interchain interactions and increasing the free volume. A representative example is blending SY with Triton-X[35, 87]. It was found that Triton-X occupies the spaces between adjacent SY chains. As a result, the conformation of polymer chains was changed from coiled to linear. This increases the polymer chain dynamics during stretching (Figure 7a). As a result, decreased Young's modulus, decreased T_g, and higher stretchability were obtained from the composite film, as compared to the pristine SY film. Interestingly, despite the insulation nature of Triton-X, higher charge (both hole and electron) mobility than that of SY was obtained under optimized the blending ratios of Triton-X.

While this blending approach can effectively enhance the stretchability of EL polymers, further developments still need to address the challenges of compromising the EL properties and the possibility of gradual phase separation and leaching out in terms of long-term stability.

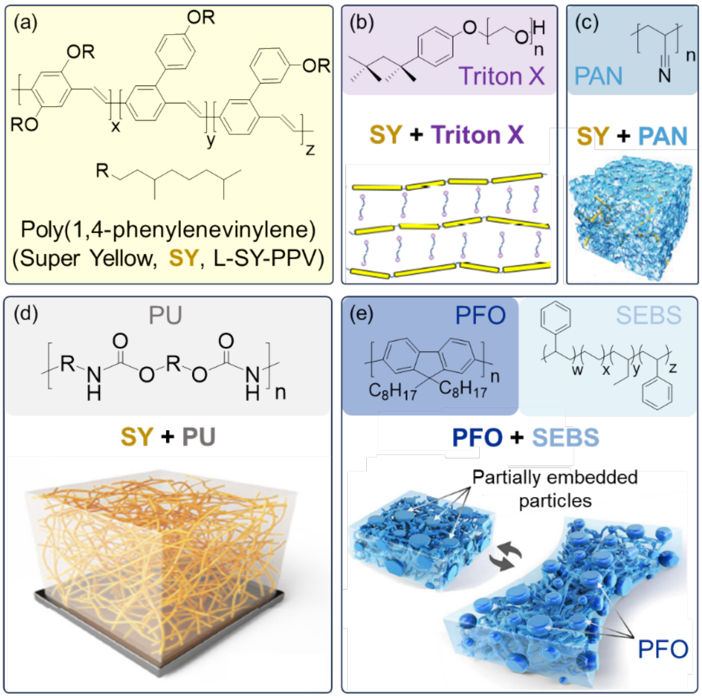

Figure 7 | Physical strategies of designing stretchable FL-EMLs. (a) Chemical structure of SY and its' full name and other abbreviations (b) Intrinsically stretchable EML by blending SY with plasticizer Triton-X. Reproduced with permission from ref [35]. Copyright 2021 AAAS. (c) Intrinsically stretchable EML by blending SY with polymer PAN. Reproduced with permission from ref [88]. Copyright 2022 Springer Nature Limited. (d) Intrinsically stretchable EML by blending SY with elastomer PU. Reproduced with permission from ref [16]. Copyright 2022 Wiley-VCH. (e) Intrinsically stretchable EML by blending PFO with elastomer SEBS. Reproduced with permission from ref [89]. Copyright 2023 AAAS.

3.3.2.2. Blending FL polymers with elastomers

Recent studies indicate that conventional EL polymers have the desired properties for blending with several types of insulating elastomers. The double-network morphologies achieved high stretchability and promising EL performance for fully stretchable OLED. In some of the reported designs, blending with certain elastomers could even benefit EL performance. One recent work using this strategy is through blending SY with elastomer polyacrylonitrile (PAN) (Figure 7b). Under the optimized blending ratio of 1:1, the composite EML forms a self-assembled 3D penetrating nanonetwork, which is beneficial for not only higher stretchability, but also better charge carrier mobility compared to the pristine SY. When testing in a rigid OLED design using a transfer-lamination method, for strained EMLs, the composite film achieves stable EL performance in the strain ranges of 0% to 80% with a turn-on voltage around 4.4-4.8 V, a maximum current efficiency of 8.13 cd A^{-1}, and an EQE of 2.64%. In the same year, Zhang et al. reported a design of blending SY with elastic matrix polyurethane (PU) (Figure 7c)[16]. Under the optimized SY/PU ratio, the composite EML shows stretchability greater than 100%, lower modulus, enhanced PLQY, and increased charge mobility than the original SY. These improvements reportedly arise from the charge-trapping dilution effect from the PU induced SY nanofibers. Besides, under stretching, the PLQE, charge carrier transport density (both electron and hole), as well as the luminance and current density were all enhanced, which is attributed to the alignment of the polymer and densifying of the film in the vertical direction under strain. It was also found that replacing PU with a non-polar polystyrene-block-poly(ethylene-ran-butylene)-block-polystyrene (SEBS) will cause severe phase separation with SY, which is unfavorable for charge transport and the EL properties. However, a recent study showed that when the EL polymer is changed to a more non-polar design, SEBS can be a suitable choice as the blended elastomer to achieve the desired morphology for both high stretchability and EL performance[89]. The results show that the blended composite films contain multidimensional interconnected nanodomains of EL polymers in the elastomer matrix (Figure 7d).

Those works clearly show that blending EL polymers with elastomers has emerged as a promising avenue for simultaneously enhancing the mechanical stretchability and EL performance by induing desired phase separation nanostructures. However, careful consideration must be given to factors such as the miscibility of the two polymers to minimize sacrifices to the EL performance.

3.3.2.3. Backbone engineering for FL polymers

As a polymer's stretchability is largely determined by the flexibility of its backbone, synthetically incorporating flexible units into the backbones of FL polymers has also been implemented to improve stretchability.

One of the commonly used design architectures for such backbone engineering is block copolymers. In 2020, Jao et al. reported a rod-coil diblock copolymer design, poly[2,7-(9,9-

dioctylfluorene)]-block-poly(isoprene) (PF-b-PI), which combines soft units PI with conjugated fluorene structures PF in the backbone (Figure 8a)[90]. The polymer was synthesized through a new simplified procedure, combining Suzuki polycondensation for the PF rod and reversible addition−fragmentation chain transfer (RAFT) for a rubber-like PI coil. Even though the molecular weight for the PF part is low (M_n = 8.2 kDa), primarily limited by the suboptimal conditions for the Suzuki polycondensation, the RAFT enables tunable polymerization degrees to achieve a high molecular weight of the diblock copolymer, resulting in high stretchability up to 140% strain. However, these polymers exhibited poor EL performance in OLED, with a maximum luminance less than 200 cd m^{-2} at voltages higher than 15 V. The carbon-carbon double bonds in the PI units facilitate the polymer to be readily crosslinked through thiol-ene click reaction with the crosslinker 1,9-nonanedithiol initiated by azobisisobutyronitrile (AIBN)[91]. Although the crosslinking significantly boosts mechanical robustness, resulting stretchability up to 150% and high stability in PLQY over 1000 stretching cycles at 150% strain, there are, regrettably, no studies on the influence of the crosslinking process on either PL or EL properties.

More recently, Li et al. reported a random triblock copolymer consisting of pyrene, styrene, and butadiene through anion polymerization catalyzed by *n*-butyl lithium (Figure 8b)[92]. The polymerization method yields decent molecular weights (M_n > 10 kDa), and, notably, very high homogeneity (PDI ≈ 1). Continuous charge transport channels are maintained due to the self-aggregating connectivity between pyrene segments, while the flexible segments increase stretchability to as high as 806% strain. The EL performance in OLED based on this polymer gave a turn-on voltage of 9.3 V and a maximum EQE of 2.08%.

Other than block copolymer design, alternating copolymer designs have been another approach for introducing soft segments into backbones (Figure 8c)[93]. Based on polyfluorene structures, Ni et al. inserted the alkyl chains into the polymer backbone using Suzuki polycondensation. The resulting polymers exhibit enhanced stretchability compared with the fully conjugated polyfluorene-based polymers. However, the polymer demonstrates only moderate stretchability of around 15% strain, potentially limited by its relatively low molecular weight (M_n = 12 kDa). The OLED based on the polymer exhibited a turn-on voltage of 6.4 V, a maximum luminance of 117 cd m^{-2} at 11.5 V, and a maximum EQE of 0.25%.

Overall, the backbone engineering is a powerful strategy to enhance the stretchability of FL polymers. Even though block copolymers have achieved exceptional stretchability, the flexible, non-conjugated blocks significantly impede charge transport, leading to relatively high working voltages and low luminance in OLED devices. Further investigations should focus on resolving the trade-off between charge transport and stretchability in block copolymers. Moreover, new polymerization methods are needed to pursue higher molecular weights, ensuring sufficient entanglement in the polymer networks to enhance stretchability, particularly in alternating copolymer designs.

3.3.2.4. Side-chain engineering for FL polymers

In addition to modifying polymer backbones, side-chain engineering is another chemical strategy to tune the interaction and conformation of polymer chains to realize stretchable EL polymers. However, there is still limited research and knowledge available regarding side-chain engineering. In one representative work, Liu et al. chemically introduced amide groups onto the side chains of a polydiarylfluorens based polymer (Figure 8d)[94]. The strain dissipation mechanism provided by hydrogen bonds between amide groups helps to enhance stretchability to 30% strain. The OLED based on this polymer gave an EQE of 1.28%. In another work, bulky groups (i.e., conventional ester) were chemically bonded into the sidechains of conjugated FL polymers for weakening the interchain interactions, thereby increasing strtchability[95]. The freestanding film based on the synthesized polymer achieved a crack onset strain of 55%, representing a10-fold increase. No obvious influence on the PLQY was observed. The stretchable polymer exhibited a stable deep-blue emission with a maximum EQE of 1.06%. Overall, compared to backbone engineering, side-chain designs generally have weaker effects on stretchability.

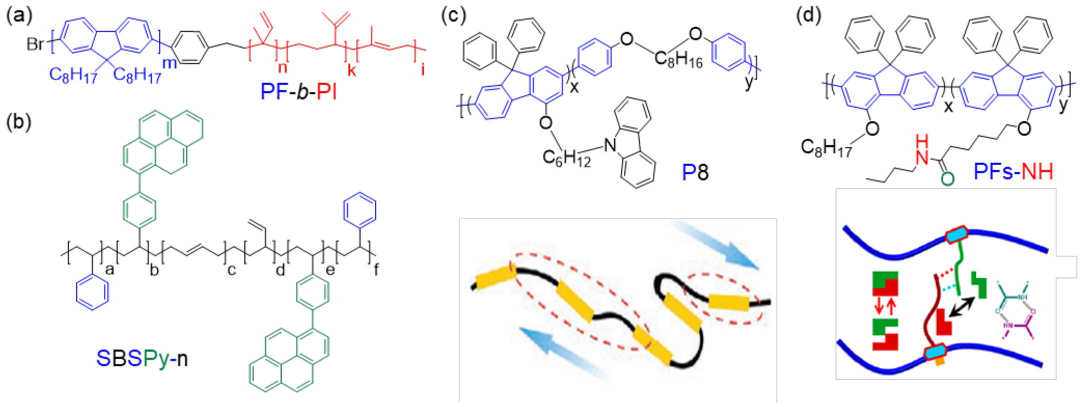

Figure 8 | Chemical strategies of designing stretchable FL-EMLs. (a) Chemical structure of PF-b-PI with a backbone engineering of rod–coil diblock copolymer design. Reproduced with permission from ref [90]. Copyright 2021 Wiley-VCH. (b) Chemical structure of SBSPy-n with a backbone engineering of random triblock copolymer design. Reproduced with permission from ref [92]. Copyright 2023 Wiley-VCH. (c) Chemical structure of P8 with a backbone engineering of alternative copolymer design. Reproduced with permission from ref [93]. Copyright 2021 Wiley-VCH. (d)

Chemical structure of PFs-NH with a side-chain engineering by incorporating H-bonding in the sidechains. Reproduced with permission from ref [94]. Copyright 2022 American Chemical Society.

3.3.3. Intrinsically stretchable PH-EMLs for LED

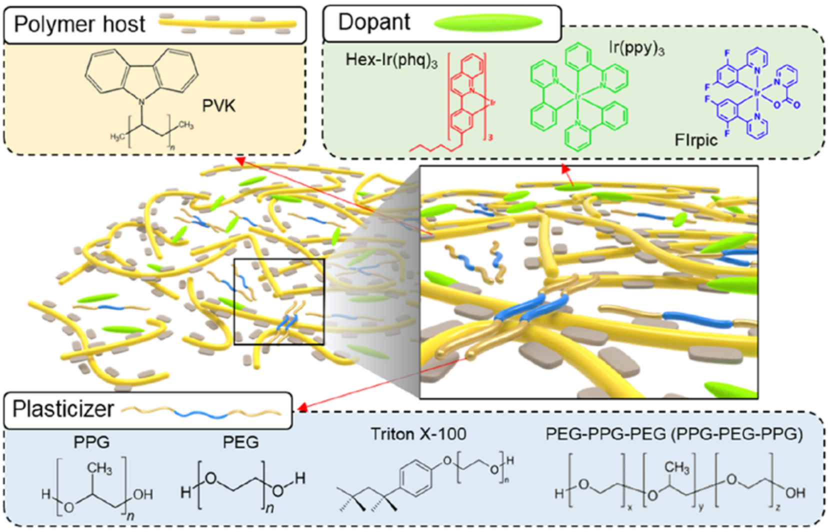

Figure 9 | Schematic illustration and components of stretchable EML based on PH emitters. Reproduced with permission from ref [96]. Copyright 2023 American Chemical Society.

PH emitters, because of a theoretical IQE as high as 100%, have been the dominating emitter design for commercial OLEDs. So far, very few endeavors have been made to realize stretchable PH-EMLs for OLEDs. Oh et al. reported the use of the plasticizing strategy on the emitter design with a polymer host and small-molecular PH dopants (Figure 9)[96]. It was found that the plasticizer poly(ethylene glycol)-block-poly(propylene glycol)-block-poly(ethylene glycol) (PEG−PPG−PEG) can significantly improve the stretchability of the conventional PH-based EML from 3% to about 100%, as well as enhance the brightness and the efficiency. The improvement in the EL properties was attributed to the charge balance between holes and electrons, achieved by the trap dilution and dipole effects of the PEG−PPG−PEG structure. The rigid OLED based on the newly developed intrinsically stretchable PH-EML shows a brightness of 5,400 cd/m^2, and a current efficiency of 25.3 cd/A. Recently, Ha et al. found that SEBS is a suitable elastic matrix to incorporate stretchability into the conventional PH EML based on a small-molecule host and PH dopants. The resulting blending EMLs are capable of withstanding strains of up to 200% with a minor reduction in charge-carrier mobility, and show the maximum current efficiency of 26.4 and 57.4 cd A^{-1} for red and green emissions[97]. Other than those two works, to the best of our knowledge, there haven't been any further attempts on the stretchable PH-EMLs.

3.3.4. Intrinsically stretchable TADF-EMLs for LED

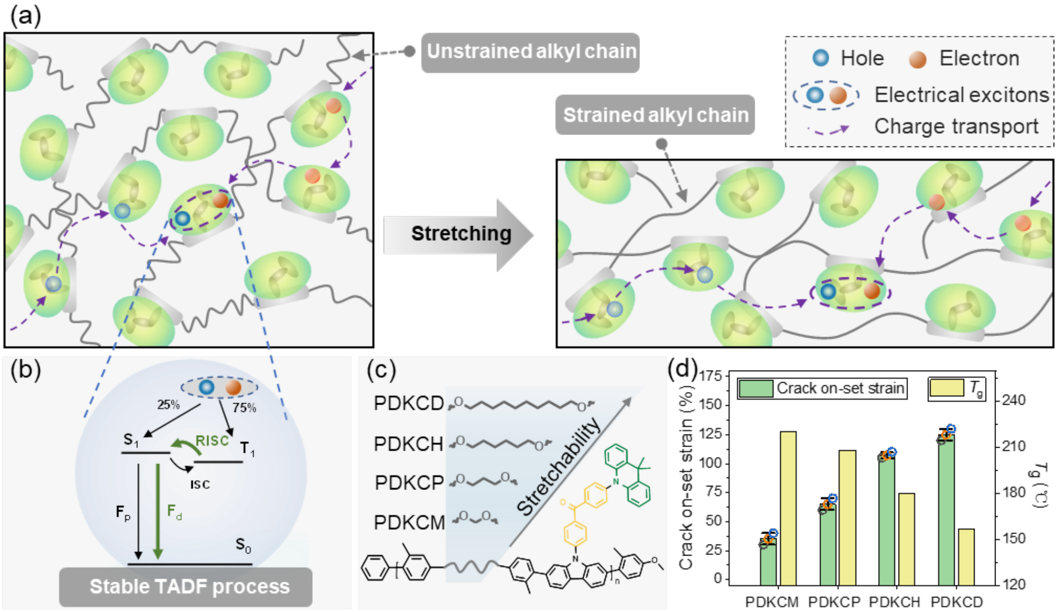

Figure 10 | Intrinsically stretchable TADF-EMLs for LED. (a) Design principle for realizing stretchable TADF polymers via incorporation of soft alkyl chains into the polymer backbone. (b) Light-emitting mechanisms of the stretchable TADF polymer. (c) Chemical structures of stretchable TADF polymers having different alkyl chains in the polymer backbone. (d) T_g and crack on-set strains of stretchable TADF polymers. Reproduced with permission from ref [17]. Copyright 2023 Springer Nature Limited.

TADF emitters, as the third-generation emitter, can also achieve a theoretical IQE of 100%. As a distinct advantage over PH emitters, TADF emitters don't need to incorporate heavy metals, which renders TADF emitters less expensive, more environmentally friendly, and more biocompatible. As the first effort in introducing stretchability to TADF emitters, our group recently reported the designs of stretchable TADF polymers by copolymerization of the TADF segments alkyl chains with different chain lengths (Figure 10)[17]. Suzuki polycondensation was unitized under optimized condition to obtain the relatively high molecular weights ($M_n \approx 30$ kDa) for the polymers. With increasing alkyl chain length, the T_g of the polymer significantly decreases, which means increased polymer chain dynamics. The polymer with a 10-carbon alkyl chain achieved a maximum stretchability of 125%.

Comprehensive investigations from both experiments and theoretical simulations suggested that neither inserting an alkyl chain nor strain had a notable influence on TADF properties. The stretchable EMLs maintained PLQYs of c.a. 60% and EQEs around 10%, even under stretching to 100% strain. While the advancement of stretchable TADF-EMLs presents a highly promising direction (Table 1), further improvements are needed on multiple fronts. Central to everything, the EL performance of stretchable TADF emitters needs to better approach the state-of-the-art small-molecule emitters. Also, incorporating different emission colors (in particular, red and blue) with enhanced color purity[98] into stretchable TADF emitters is imperative for the applications of both displays and bio-stimulations.

Table 1. | Performance comparison of the intrinsically stretchable EMLs for LED.

EL mechanism	Stretchability design principle	EL materials	Stretchability (%)	Emission color	Rigid OLED performance			Ref.
					V_{on} (V)	CE_{max} (cd/A^{-1})	EQE_{max} (%)	
Perovskite	Perovskite + elastomer	$MAPbBr_3$	~ 30	green	-	-	-	82
			~ 35	green	2.5	1.77	-	83
FL	EL polymer + plasticizer	SY	80	yellow	-	-	-	35
	EL polymer + elastomer	SY	80	yellow	4.6	8.13	2.64	88
	EL polymer + elastomer	SY	100	yellow	~ 4,5	~ 6.5	-	16
	EL polymer + elastomer	PFO	110	blue	3.71	0.6	-	89
	Polymer mainchain engineering	SBSPy-n	806	blue	9.3	5.23	2.08	92
	Polymer mainchain engineering	P8	15	blue	6.4	~0.4	0.25	93
	Polymer sidechain engineering	PFs-NH-30%	30	blue	5.0	-	1.28	94
	Polymer sidechain (internal plasticizer)	PF-MC8	55	blue	5.5	0.97	1.06	95
PH	PH emitter + host polymer + plasticizer	$Ir(ppy)_3$	100	green	6	25.3	6.8	96
	PH emitter + host + elastomer	$Ir(mppy)_3$	200	green	4.2	57.4	16	97
		$Ir(mphmq)_2(tmd)$	200	red	4.2	26.4	17.6	
TADF	Polymer mainchain engineering	PDKCD	125	green	3.5	31.6	10.8	17

3.3.5. Fully stretchable LED

LED typically has multiple-layer device structures to achieve efficient and balanced injections of electrons and holes from two electrodes, and also confinement of the formed excitons in the EML. A typically LED structure (Figure 2c) can include the following layers: (1) an anode electrode with relatively high work function, (2) a hole injection layer (HIL) and hole transporting layer (HTL), which are usually combined into one layer (HIL/HTL) with relatively deep highest occupied molecular orbitals (HOMO) and efficient hole transport, (3) an EML, (4) an electron-transport layer (ETL) with relatively high lowest unoccupied molecular orbitals (LUMO) level and efficient electron transport, (5) an electron-injection layer (EIL), and (6) a cathode electrode with relatively low work function. In the past few years, along with the developments of stretchable EMLs, several design strategies for fully stretchable OLEDs have been reported. In this section, we review the different designs for each functional layer, as well as the device fabrication techniques.

3.3.5.1. Intrinsically stretchable electrodes for LED

It is worth noting that LEEC and LED typically share the same principles for electrode designs. Adapted from stretchable LEEC, the most prevalent electrodes used in fully stretchable LED are created by embedding AgNWs in elastomers (Figure 11a). In these composite electrodes, the percolated AgNW network provides high conductivity, while the large openings among the nanowires allow visible light to pass through and enable the infiltration of the elastic matrix, such as poly(urethane acrylate)[44], polyurethane[17], PDMS[35, 88], SEBS[87], and polyethyleneimine ethoxylated (PEIE)[35].

However, AgNW-based electrodes still face a dilemma between conductivity and transparency, particularly when aiming for high stretchability. Furthermore, the percolated AgNW networks often result in a rough electrode surface, potentially leading to device shorting. To overcome those challenges, Zhang et al. utilized a polyrotaxane (PR) modified PEDOT:PSS as the electrodes for fully stretchable OLED (Figure 11b)[16]. Adding PR in PEDOT:PSS reduced the overall elastic modulus and increased the stretchability to over 100% strain. The resulting electrode achieved high stretchability, conductivity (700 S cm^{-1}), transparency (92% at 550 nm), solvent-resistivity, and device processability. This design has demonstrated excellent performance in the fabrication of fully stretchable OLED.

Electrodes based on AgNW or PEDOT:PSS typically exhibit a work function around 4.7 eV[16, 17], which is not well-suited for efficient charge injection, especially for the electron injection at the cathode side, in fully stretchable OLED. However, there has been little effort in addressing this issue. Zhou et al. recently reported 2D-contact stretchable electrodes (TCSEs) by modifying the AgNWs with 2D graphene (Figure 11c)[87]. TCSEs can be further modified to form p-type and n-type TCSEs with tunable work functions ranging from 5.69 to 3.57 eV, enabling efficient hole and electron injection, respectively.

Nevertheless, realizing electrodes with low surface resistance, high optical transparency, and appropriate work functions for efficient charge injection, particularly for electrons, is still challenging and needs a leap of progress.

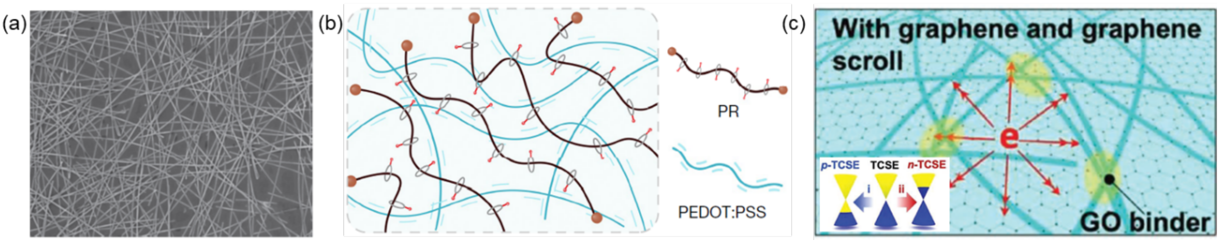

Figure 11 | Stretchable electrode designs for fully stretchable LED. (a) SEM images of AgNW-PUA composite electrode. Reproduced with permission from ref [43]. Copyright 2013 Springer Nature Limited. (b) Schematic morphology of a PEDOT:PSS/PR film. Reproduced with permission from ref [16]. Copyright 2022 Springer Nature Limited. (c) Conceptual illustration of the fully stretchable OLED based on p- and n-doped 2D-contact stretchable electrodes (TCSEs). Reproduced with permission from ref [87]. Copyright 2022 Wiley-VCH.

3.3.5.2. Intrinsically stretchable HTL/HIL for LED

PEDOT:PSS is widely used as HTL/HIL due to its suitable HOMO level for hole injection, high hole mobility, solution processability, and resistance to organic solvents for orthogonal layer-by-layer fabrication. To create stretchable HTL/HILs for LED, research has primarily focused on developing stretchable versions of PEDOT:PSS through physical blending techniques. As of our knowledge, two designs, namely PEDOT:PSS_Triton X composite and PEDOT:PSS_PFI composite, have been prominent in this context.

The majority of fully stretchable LEDs employ the first approach, involving Triton X, which prevents strong electrostatic interactions between PEDOT and PSS and alters the conformation of PEDOT chains (Figure 12a). This benefits both hole transport and mechanical stretchability, with the resulting composite remaining crack-free even under high strains of up to 160%.[35]

In rigid LED designs, PFI has already been used as an additive in PEDOT:PSS HTL/HIL to deepen the HOMO level, thereby enhancing hole injection. Our group recently discovered that blending PFI into PEDOT:PSS results in a highly stretchable film due to two complementary mechanisms: the soft and stretchable PFI serving as the stretchable matrix and the weakening of interchain interactions among PEDOT chains (Figure 12b)[17].

Moving forward, beyond these PEDOT:PSS-based composite designs for HTL/HILs in fully stretchable LED, innovations are still needed for better hole injection/transport and electron/exciton blocking abilities.

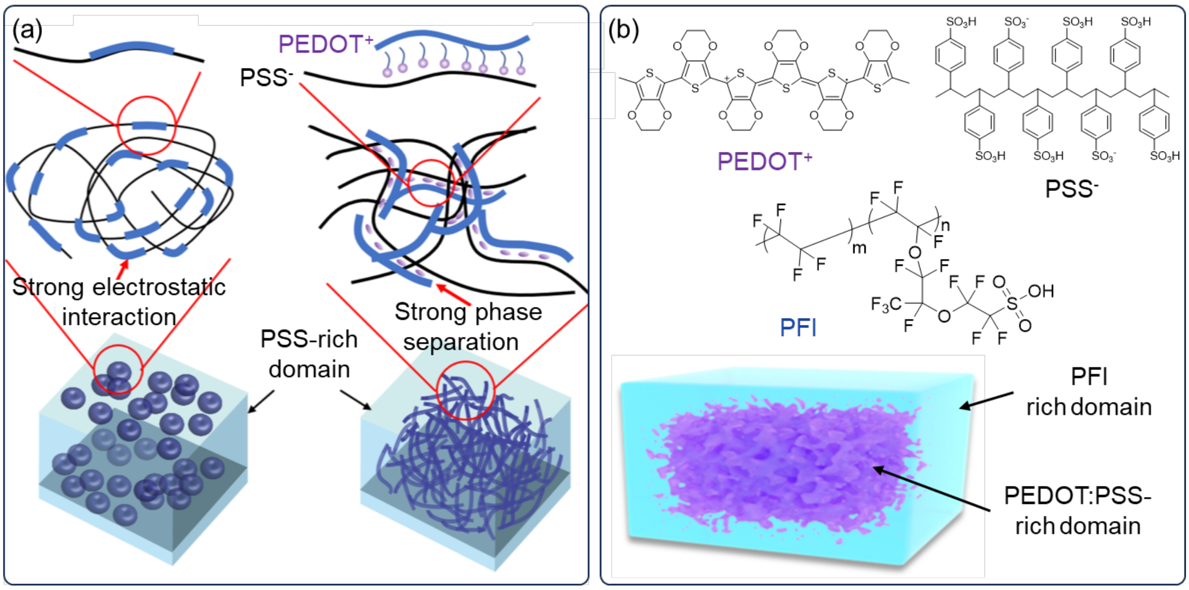

Figure 12 | HTL/HIL for fully stretchable LED. (a) Schematics of the microstructures of pristine HTL and intrinsically stretchable HTL based on PEDOT:PSS_Triton X composite. Reproduced with permission from ref [35]. Copyright 2021 AAAS. (b) Chemical structures of PEDOT:PSS and PFI. Proposed 3D illustration of the morphology of the PEDOT:PSS_PFI composite film. Reproduced with permission from ref [17]. Copyright 2023, Springer Nature Limited.

3.3.5.3. Intrinsically stretchable ETL/EIL for LED

Polymers containing aliphatic amine groups, such as PEIE and polyethyleneimine (PEI), serve as universal surface modifiers to reduce the work function of an electrode[99], which helps electron injection. In the meanwhile, the low T_gs of PEIE and PEI make them highly promising candidates for ETL/EIL in fully stretchable LED. However, pure PEIE/PEI cannot compete with conventional alkali metal ion-based materials (e.g., LiF, Cs_2CO_3) when used as EIL. This results in low electron injection efficiency and high turn-on voltage, primarily due to their insulating nature. To fabricate ETL/EILs with better electron injection and transport properties for fully stretchable LED, PEIE/PEI are typically modified to form EIL/ETL composites by blending them with materials possessing high electron mobility, such as ZnO[35], crown-CPE[87], 1,10-phenanthroline monohydrate (pBphen)[88], and poly[(9,9-bis(3′-((N,N-dimethyl)-N-ethylammonium)-propyl)-2,7-fluorene)-alt-2,7-(9,9 dioctylfluorene)] (PFN-Br)[16, 17]. These reported composite designs for EIL/ETLs show high stretchability and decent electron injection/transport abilities in fully stretchable OLED. However, compared to ETLs/EILs in conventional rigid OLEDs, those stretchable EIL/ETLs still show inferior electron injection/transport and limited hole-blocking properties, thereby resulting in higher turn-on voltages and lower device efficiencies. Moving forward, there is a pressing need for better stretchable ETL/EIL materials with desired properties, including high electron mobility, alignment of LUMO levels with both the cathode and the EML, deep HOMO levels for hole blocking, high triplet energy states for exciton blocking, and high stretchability, etc.

3.3.5.4. Device designs for fully stretchable LED

Liang et al. reported the first fully stretchable OLED in 2014, featuring a device structure of AgNW/PEDOT:PSS/EML/PEI/ AgNW[44]. The HTL/HIL/EML and ETL/EIL layers are separately spin-coated onto two substrates, one serving as the anode and the other as the cathode. The device fabrication was completed by laminating the two electrodes. The resulting device has a turn-on voltage of 7 V, a maximum brightness of 1100 cd m^{-2} at 21 V, and a maximum current efficiency of 4 cd A^{-1}. The device achieved stretchability up to 130% strain and could endure 100 cycles of repeated stretching to 40% strain. Similar device fabrication techniques are employed in most of the subsequent works with new generations of improved material designs (Table 2). It's noteworthy that it's possible to create a fully stretchable OLED[35] by consecutively spin-coating each layer, including all functional layers and the cathode, directly onto a pre-prepared anode substrate. This approach eliminates the need for a second substrate and the lamination process, potentially avoiding shorting effects when the device is subjected to compressed strain.

Similar to LEECs, as discussed in Section 3.1.2, hermetically sealing fully stretchable OLEDs is essential to achieve enhanced air stability. However, the development of encapsulation techniques for fully stretchable OLEDs has been largely overlooked. For most of these devices, the functional layers, including semiconducting layers and electrodes, are merely sealed with electrode-substrates made of common elastomers, such as PDMS, SEBS, TPU, and PVDF-HFP, which are usually suboptimal barriers for moisture and oxygen. Therefore, investigations of device stability are rarely included in reported studies, possibly due to the extremely short lifetime of those devices. To the best of my knowledge, the only reported T_{50} for a fully stretchable OLED is as short as 27 seconds in ambient[35].

All in all, with the overarching goal of achieving high EL performance, the further development of fully stretchable OLED should mainly focus on TADF and PH-type EMLs, emphasizing high efficiency, color purity, and stability. So far, the device structures of fully stretchable OLED still yield significantly lower performance than what can be offered by the utilized EMLs. To bridge this gap, the main challenges lie in the following aspects. First, for the cathode, there has been no development of a stretchable cathode with a sufficiently low work function for efficient electron injection. Second, it still lacks stretchable EIL/ETL polymers with relatively low LUMO, deep HOMO, and high energy state (i.e., triplet state, T_1). Third, for such multi-layer stacked device structures built by solution-processed polymers, the fabrication method must ensure compatible stacking of different layers without encountering solvent compatibility issues. Additionally, reliable encapsulation techniques and fabrication of high-density arrays are essential for real applications, such as skin display.

Table 2. | Performance comparison of representative fully stretchable EL devices based on different device and emitter mechanisms.

EL type	EL materials	Turn-on voltage (V)	Voltage @1000 cd/m^2 (V)	CE_{max} (cd/A^{-1})	EQE_{max} (%)	Stretchability (%)	Turn-on speed	Ref.
LEEC	PF-B	4.8	-	1.24	-	45	~ min	42
	SY	6.8	18.0	5.7 + 5.7[c]	2.0 + 2.0[c]	120	~ min	43
	SY	3.7	7.0	3.14[c]	-	30	~ min	73
LEC	ZnS	>1000[b]	-	-	-	~ 500	-	46
	ZnS:Cu	23, 1k Hz[b]	-	-	-	~ 800	~ ms	47
FL-OLED	WLEP[a]	7.0	10.5	2.0 + 2.0[c]	-	130	-	44
	SY	13.0	-	-	-	50	-	71
	SY	8.0	10.5	1.6	-	80	-	35
	SY	5.0	10.5	5.3 + 5.2[c]	-	100	-	16
	SY	5.3	11.0	~ 20.3[d]		20	-	87
TADF-OLED	PDKCD	4.75	8.1	5.3 + 4.9[c]	1.7 + 1.6[c]	60	~ ms	17

[a] WLEP, white-light-emitting polymer, which was provided by Cambridge Display Company but without specific named offered from the literature[44].

[b] AC model.

[c] Measured from both anode and cathode sides.

[d] Total value from both anode and cathode sides.

4. Conclusions and perspectives

In conclusion, we have summarized the main strategies utilized to obtain intrinsically stretchable EL materials for three types of stretchable light-emitting devices, including LEEC, LEC, and LED. During the early stages of intrinsically stretchable EL development, LEEC and LEC made significant progress owing to their simpler device architectures compared to LED. More recently, the research focus has shifted substantially to OLED, driven by their superior efficiency, brightness, operational speed, and lower voltage requirements.

For the EML materials, as the key component for OLED, the primary development goal is to combine skin-like stretchability with high EL efficiency and brightness. So far, the design approaches for achieving stretchability mainly fall under two categories - the physical blending of active EL materials with elastomers or small-molecule additives, and the chemical synthesis of new polymer structures that combine EL-active moieties with strain-dissipation moieties. Therefore, the realization of high EL efficiency mainly relies on two aspects: utilizing EL materials/moieties with high-efficiency EL mechanisms (such as multi-resonance TADF, perovskite, and quantum dots) and selecting stretchable additives/units that do not compromise, and, ideally, enhance the EL performance. Therefore, a major opportunity in future research lies in leveraging advanced EL mechanisms and designs that can provide high efficiency and stability, a broad range of emission colors, and improved color purity.

To unlock the full potential of excellent stretchable EML in fully stretchable OLED, it is essential to concurrently develop other functional layers in OLED, including semiconducting layers and electrodes. Firstly, stretchable semiconducting layers need to be combined with desired energy levels for efficient charge injection/blocking and exciton blocking abilities, efficient electron/hole transport, and high transparency. Secondly, electrodes with low surface resistance, high transparency, and appropriate work functions for efficient charge injection, particularly for electrons, are also under the spotlight.

Moreover, for stretchable display applications, innovations in fabrication processes for high-density arrays are crucial, which also requires new material developments to achieve the patterning of individual layers and also compatible deposition of different layers. Besides, the significance of addressing interfacial adhesion between adjacent layers within the device cannot be overstated, robust mechanical performance under various deformations hinges on this aspect. Last but not least, the pursuit of encapsulating materials with robust moisture and gas barrier properties, coupled with high optical transparency and compatibility with other device layers, is of paramount importance to enhance the device stability.

Overall, the development of stretchable EL technologies is predominantly steered by material developments. These developments hinge on the expertise of chemists in designing and synthesizing novel materials, amalgamating insight from photophysics, materials chemistry, polymer sciences, and mechanics. This interdisciplinary direction offers a lot of opportunities for innovations at both fundamental and technological levels. Collectively, these research outcomes will significantly impact various domains such as consumer electronics, robotics, medical devices, bioengineering.

Conflict of Interest

There is no conflict of interest to report.

Acknowledgments

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