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Progress and Prospects in Stretchable Electroluminescent Devices

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Abstract: Stretchable electroluminescent (EL) devices are a new form of mechanically deformable electronics that are gaining increasing interests and believed to be one of the essential technologies for next generation lighting and display applications. Apart from the simple bending capability in flexible EL devices, the stretchable EL devices are required to withstand larger mechanical deformations and accommodate stretching strain beyond 10%. The excellent mechanical conformability in these devices enables their applications in rigorous mechanical conditions such as flexing, twisting, stretching, and folding. The stretchable EL devices can be conformably wrapped onto arbitrary curvilinear surface and respond seamlessly to the external or internal forces, leading to unprecedented applications that cannot be addressed with conventional technologies. For example, they are in demand for wide applications in biomedical-related devices or sensors and soft interactive display systems, including activating devices for photosensitive drug, imaging apparatus for internal tissues, electronic skins, interactive input and output devices, robotics, and volumetric displays. With increasingly stringent demand on the mechanical requirements, the fabrication of stretchable EL device is encountering many challenges that are difficult to resolve. In this review, recent progresses in the stretchable EL devices are covered with a focus on the approaches that are adopted to tackle materials and process challenges in stretchable EL devices and delineate the strategies in stretchable electronics. We first introduce the emission mechanisms that have been successfully demonstrated on stretchable EL devices. Limitations and advantages of the different mechanisms for stretchable EL devices are also discussed. Representative reports are reviewed based on different structural and material strategies. Unprecedented applications that have been enabled by the stretchable EL devices are reviewed. Finally, we summarize with our perspectives on the approaches for the stretchable EL devices and our proposals on the future development in these devices.

Keywords: Stretchable, Light-emitting device, electroluminescent, soft electronics, displays, nanowire, stretchable conductors, electronic skin

1 Introduction

Soft electronic devices are emerging as new technologies that push the boundaries and limitations in conventional planar and rigid devices. Deformable electroluminescent (EL) devices drive special interests, as they are expected to bring a plethora of new possibilities for the envisioned display-centric world in the near future. The idea to create flexible electronic devices dates back to the 1960s when flexible solar cell arrays on plastic substrates were first demonstrated [1, 2]. The first flexible organic light-emitting diode (OLED) devices on polyethyleneterephthalate (PET) substrate was demonstrated by Gustafsson *et al.* in 1992 using conductive polyaniline (PANI) as the flexible electrode [3]. To date, a number of organizations have been able to manufacture prototype flexible displays, such as flexible e-paper displays and rollable OLED panels with active endeavors dedicated to develop new technologies beyond the rigid and brittle devices. Success in the flexible EL devices has been proven with market share of the flexible displays increasing steadily and being predicted to become 50% by 2020.

Back in 1998, stretchable electrodes were reported based on thin, wrinkled, metal films on stretchable elastomers by Bowden and coworkers [4]. The mechanical conformability of stretchable devices is recognized to possess significant advantages over the simple bendability in flexible devices. With the special "soft" features, stretchable EL devices can be applied where demanding deformations such as volumetric 3D displays, which can present the

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objects in three physical dimensions, and interactive displays, which can provide users tactile interaction beside conventional planar graphic information, are required. Prototype applications were demonstrated by the MIT Tangible Media Groups by using 2D actuation arrays and a top projector to achieve dynamic shape displays [5]. Another example was created by Sif Khan, which is known as the MegaFaces in 2014. Unfortunately, these applications are still limited on the huge and bulky platforms without available technologies on stretchable EL devices. Though the technology difficulties in stretchable electronics may greatly exceed those in the flexible devices, increasing efforts are being channeled into this research area, trying to duplicate the success in the preceding flexible generation.

Based on the underlying idea that any materials in sufficiently thin form is flexible, Rogers and coworkers have developed a wide range of thin and flexible materials that can be patterned to achieve stretchable structures. These stretchable electronic systems are based on rigid device islands bridged by stretchable conductors [6–12]. With the structural strategy, stretchable light-emitting diodes (LEDs) were demonstrated by assembling ultra-small inorganic light-emitting diodes (ILEDs) onto rubber substrate with "wavy" and thus stretchable electrical interconnects, yielding stretchable devices [13]. New approaches and strategies were also developed to demonstrate stretchable EL devices with improved fabrication procedures and device performance. The successful demonstrations on stretchable EL devices can also be categorized into the two strategies defined as "Structure That Stretch" and "Materials That Stretch" [14]. By configuring the structures of the thin materials into "wavy" or buckling shapes and bonding them onto elastic substrates, the materials' flexibility can be used to accommodate stretching deformations. Most of the strain can be relaxed by the "wavy" or buckling structures. Advantage of the strategy is that conventional technologies in the LED fabrication can be exploited. Earlier works were reported by Rogers's group and Someya's group based on the strategy [13, 15–17]. Limitations in the structural strategy might be the challenge to develop effective approaches that can manipulate the stretchable structures and integrate all the active components with mismatched mechanical properties onto a single elastomer. By developing new emissive materials and conducting materials, fully stretchable light-emitting devices can also be achieved with the materials strategy. Different from the structural strategy, which used conventional rigid light-emitting elements (do not respond to strain) with stretchable conductor structures (respond to strain), the light-emitting elements and transparent conductors can be strained simultaneously under mechanical deformations in the fully stretchable EL devices. Pioneered by Pei's group and recently our group, successful approaches have been developed to fabricate intrinsically and fully stretchable EL devices based on different emission mechanisms [18–20]. However, these devices still need to tackle the challenges in their operating lifetime, switching speed, brightness, and operating voltage. In this review, the diverse strategies and approaches toward stretchable EL devices are discussed in detail including their advantages and drawbacks. New applications and progresses enabled by the stretchable EL devices will also be covered, followed by a summary.

2 Emission mechanisms for stretchable EL devices

2.1 Light-emitting diodes (LEDs)

The electroluminescent mechanism of LEDs is based on charge injection and radiative recombination at the $p - n$ junction. Figure 1a shows a schematic illustration of a typical LED device structure that comprises a p-type layer and an n-type layer with the $p - n$ junction at the interface (Figure 1b). Bottom and top electrodes are deposited on opposite sides of the device with at least one transparent electrode for light extraction. Under forward bias, charges will be injected from one layer into the opposite doped layers. The injected minority carriers will radiatively recombine with the majority carriers in the opposite side of the $p - n$ junction, leading to light emission.

Both organic materials and inorganic materials have been investigated for LED devices. Though the quantum efficiency and long-time stability in the OLEDs are still lower compared to the ILEDs, the OLEDs prevail over in certain features such as their good mechanical flexibility and low fabrication cost. OLEDs with stretchable structures have been successfully demonstrated using the excellent flexibility in ultrathin EL devices [16]. On the contrary, ILEDs generally require high-temperature deposition processes to deposit the single-crystalline inorganic layers, making it incompatible with most of the flexible or stretchable substrates. Alternative approaches are to develop effective methods to transfer the ILEDs from high-temperature compatible substrates onto flexible or stretchable substrates. Successful demonstrations were reported with transferred ILEDs onto flexible [21, 22] and stretchable conducting substrates [13, 23].

One-dimensional inorganic nanostructures have been intensively studied recently with the interesting proper-

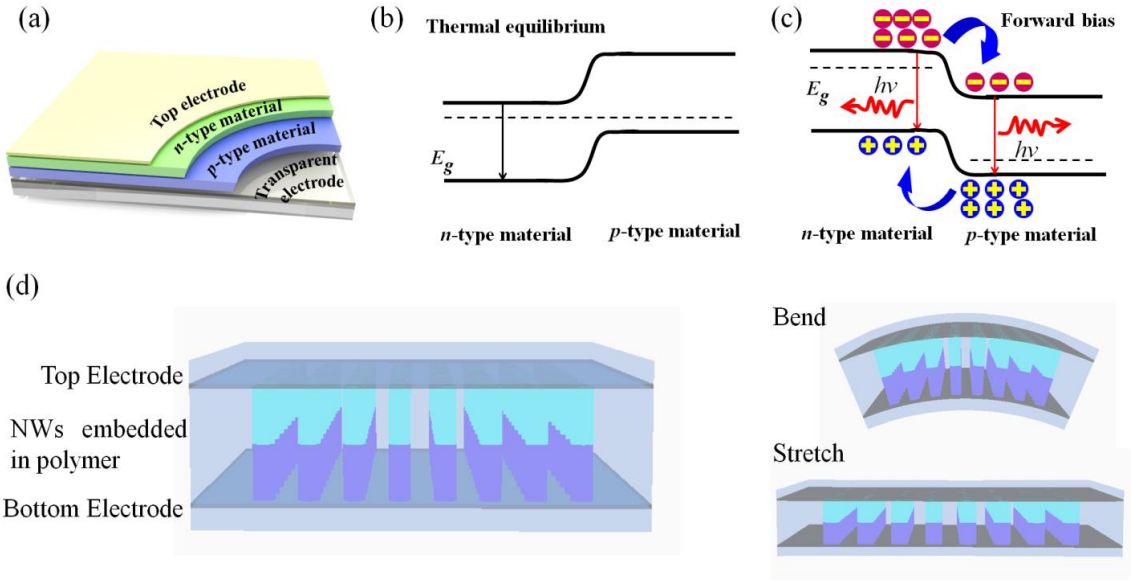

Figure 1: (a) A schematic showing the device configuration of an LED device. Energy band structures of the $p - n$ junction (b) under thermal equilibrium and (c) under forward bias. (d) A schematic design in the stretchable LED device using $p - n$ heterojunction nanowires (NWs) as the emissive materials. These devices are expected to possess excellent mechanical conformability and be able to comply with different deformations such as bending and stretching, as illustrated in the right images.

ties of increased charge injection efficiency at the nanosize junctions and increased light-extraction efficiency in the wave-guide structures [24–31]. Compared to the thin-film ILEDs, the inorganic nanowire (NW) structures possess inherent advantages for "soft" LEDs with its one-dimensional configuration. For example, Lee *et al.* reported the fabrication of flexible ILEDs with core–shell NW structure on graphene film by using the excellent mechanical flexibility in the inorganic nanostructures [21]. Interestingly, the $p - n$ junctions can be fabricated with either a core–shell structure or an axial heterostructure in a single NW. Each of the NWs can be an independent light-emitting element. As shown in Figure 1d, the nanosize light-emitting elements can be embedded into elastic polymer matrix, imparting stretchability to the device structures. It is believed that with the unique structures and inherent flexibility, NW ILEDs will become an important part in the stretchable LEDs. NW ILEDs are promising to circumvent the bottlenecks in thin-film LEDs such as the limited stretchability and the complicated patterning processes required to attain stretchable structures (the thin-film LEDs with patterned device architectures are discussed in Section 3).

2.2 Light-emitting electrochemical cells (LEECs)

The light-emitting electrochemical cell (LEEC) was introduced by Pei and coworkers in 1995 by doping conjugated polymers with electrochemical reactions, forming $p - i - n$ junction in the emissive layers [32]. The LEEC devices have simple device structure with an EL layer sandwiched between two electrodes, as shown in Figure 2b. They have shown promising performance with rather simple and low-cost fabrication processes. Intensive study has been conducted in the field. A number of review papers on LEECs are available in the literatures [33–38]. The emission mechanism of LEEC devices is similar to that of LED devices that are based on charge injections and subsequent radiative recombination. Different from the conventional LEDs, the LEECs only consist of a single emissive layer with mixtures of dissolved salts, ionic conductors, and electroluminescent materials. The $p - i - n$ junctions can be *in situ* formed by external electrical bias that leads to electrochemical doping of the EL materials in the opposite sides, as illustrated in Figure 2a. Different approaches, such as temperature cooling and polymerization of the ionic dopants, are required to prevent redistributions of the dopants and preserve the $p - i - n$ junctions after the electrical bias is removed. Compared to the LEDs, LEECs possess advantages of reduced complexity in the device fabrication, efficient and balanced charge injections, and

applicable thick emissive layers with low-cost fabrication process [33]. These features make the LEEC an ideal candidate for stretchable EL devices. First of all, the single-layer device architecture circumvents the difficulties of the multilayer LED structures in which the stretchable materials for the different functional layers still need to be developed. The matching of mechanical properties between these layers will also need to be properly addressed in order to maintain good interfaces under mechanical deformations. Second, the highly doped polymers in the LEECs have good conductivity at the electrode and polymer interfaces, leaving the undoped polymer as the effective emission region. Consequently, the LEECs fabrication is not constrained by the stringent conditions of the electrode work function. It should be noted that LEDs require high work function anode and low work function cathode. In most cases, additional charge transport layers are required between the emissive layer and electrodes in order to achieve balanced charge transport, leading to more complicated device architectures. Finally, the thick polymer structure of the LEEC devices simplify the deposition process. The deposition can be accomplished by spin coating, drop casting, or screen printing. On the other hand, the thick polymer film improves the mechanical properties of the emissive layer with reduced defects and nonuniformities, which will induce stress concentrations and degrade the material stretchability in the thin film structure [33].

Collectively, the LEECs possess inherent structural advantages for stretchable EL devices compared to the conventional LEDs. Nevertheless, these devices still need improvements in the switching speed and long-term stability before wide applications are possible. Besides, the LEECs are similar to the LEDs in the emission mechanism. Performance of the devices is highly dependent on the electrode conductivity, which directly affects the current injection. The characteristic has become a limiting factor for the LEECs to achieve stable device performance under large strain. As it is still quite challenging to fabricate stretchable and transparent electrodes for EL devices with stable conductivity under high stretching strain [39, 40], the stretchable LEECs still suffer from dramatically degraded performance under large mechanical deformations [18, 41]. The problem will persist without the development of new materials or strategies for highly conductive electrodes that can achieve good transparency and mechanical stability.

2.3 Alternating current electroluminescence (ACEL)

Unlike the emission mechanism in the LEDs or LEECs, alternating current electroluminescence (ACEL) devices are based on different material systems with field-induced excitation for light emission. In these devices, the emission behavior includes multiple steps in sequence: charge injection into the phosphor layers, acceleration of the charge carriers under high electrical field, impact excitation of the luminescent centers by high-energy electrons, and radiative relaxation of the luminescent centers in the host materials. With the unique emission mechanism, the ACEL devices have the similar structural advantages of the LEECs with a single emissive layer sandwiched between two electrodes (Figure 2b). Besides, the ACEL devices also do not have harsh requirement on the electrode work functions for charge injection. The devices consist of thick emissive layers in the range of 50–100 μm (the current can be capacitively coupled into the device with displacement current). The ACEL materials have been developed for a few decades for display and lighting applications. The majority materials used for the ACEL devices are ZnS doped with different elements such as Cu, Al, and Mn to achieve different emission colors. A comprehensive overview on the materials with their preparations and properties are provided in the book edited by Vij [42].

Emission brightness of the ACEL devices using powder phosphors can be expressed by the following relation:

$$B = B_0 \exp(-b/V^{0.5}) \quad (1)$$

where B is the emission brightness of the device, V is the applied voltage, and B_0 and b are constants. The values of B_0 and b are determined by the particle size of the phosphors, concentration of the EL powders in the dielectric materials, dielectric constant of the embedding medium, and the device thickness [43]. Figure 2c is the energy band structure interpreting the emission model in the EL powders [44, 45]. Emission behavior of the device is mainly determined by the applied bias from the above equation. The resistance change in the transparent and stretchable electrodes is very small compared to the resistance of the emissive layer such that the EL performance will rarely be affected by the variations in the electrode conductivity under strain [20]. The property makes the ACEL devices superior to achieve stable EL performance compared to the LEDs or LEECs.

Existing challenges in the ACEL devices are their high operating voltage and low efficiency, which might impede their applications in wearable and portable electronics. Fortunately, studies are being carried out with

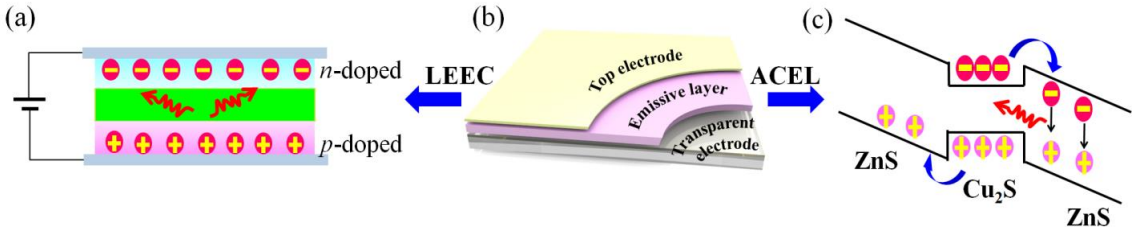

Figure 2: (a) Formation of a $p - i - n$ junction in the LEEC devices under external electrical bias. (b) Schematic illustration of the simple EL device architecture comprising a single emissive layer, top electrode, and transparent bottom electrode. (c) Band structure of the powder ACEL devices under electrical field excitation.

promising improvements of reduced operating voltages (< 50 V) and increased efficiency in the organic ACEL devices [46, 47]. Different approaches have also been conducted to effectively reduce the operating voltage in the inorganic ACEL devices. For instance, short carbon nanotube has been demonstrated to increase field strength in the inorganic powder EL devices and reduce operating voltage [48]. Similar performance enhancement was also reported in organic ACEL devices with reduced operating voltage and increased brightness (350 cd/m^2 at 25 V_{pp}) [49]. Other approaches include the use of fluorescent polymer or their hybrids with colloidal quantum dots (QDs) to achieve extremely bright emission at low operating voltage (~500 cd/m^2 at 30 V_{pp}) [50, 51]. These approaches are promising to tackle the weakness in the ACEL devices, enabling their applications in wearable and portable electronics.

Apart from the emissive devices (light is generated from the device) discussed earlier, the nonemissive devices (light is generated from the background lighting and modulated by the device), such as electrophoretic devices, which change between the white and black contrast on the movement of the charge pigments under electrical bias [52–54], and electrochromic devices, which use materials that change their optical properties in the oxidizing and reducing states [55, 56], have rarely been investigated for stretchable devices. The advantages of extremely low power consumption, simple device architecture, and solution-processable fabrication in these devices will promote their applications in stretchable electronics. It is believed that the nonemissive devices will also become an important part in the stretchable display.

3 Partially stretchable EL devices

3.1 Electrodes with stretchable structure

Stretchable EL devices assembled with rigid ILEDs and stretchable electrical interconnects on elastomers are representative examples that conventional techniques can be combined to achieve exceptional applications by implementing new underling strategies [13, 17, 23, 57–59]. In these cases, the light-emitting elements are bonded device islands that can sustain the significantly reduced local strain under stretching (the maximum local strain in the ILEDs is ~0.17% with a 24% universal strain on the whole device). Most of the strain is relaxed by the stretchable interconnects designed to isolate the rigid light-emitting elements from strain. The challenges in these devices are confined to the fabrication of stretchable electrodes and integration of the rigid light-emitting elements. Successful demonstrations were reported by Rogers and coworkers [57]. Stretchable electrodes were fabricated by designing thin metals in their serpentine or “wavy” structures bonded onto elastic substrates, as shown in Figure 3a and 3b. The wavelength and amplitude of these “wavy” structures can change accordingly in response to the applied strain. Most of the strain will be relaxed by the stretchable structures so that the flexible thin films can easily accommodate the reduced deformations. Multiple-step lithography and etching processes were required to pattern the anchor points (for easy device transfer after fabrication on rigid substrates) and “wavy” electrode structures (for stretchable structures). After metal electrodes were evaporated, the stretchable electrodes were transferred onto an Ecoflex elastomer. LED devices (dimensions of 1 mm × 0.6 mm × 0.2 mm, length × width × thickness) were then carefully aligned to the electrical contacts and bonded onto the elastic substrate by transfer printing. The EL device arrays can be uniaxially stretched to a peak strain of 200% with

no significant variation in the performance of the EL devices.

Similar idea was also demonstrated to fabricate biaxially stretchable EL devices with different patterning processes. The final structure is shown in Figure 3c [17]. Different from the previous design with a "wavy" structure, the devices used an arch-shape interconnect, which can buckle out of plane from the underlying substrate. Instead of being transferred onto a relaxed substrate, the devices were bonded onto a prestrained poly(dimethylsiloxane) (PDMS) balloon after the device structures were patterned. When the substrate was released, the thin electrical interconnects resulted in buckling structures with noncoplanar layouts in response to the compressing strain. The compressing strain was converted into flexing strain with the buckling structure in the thin metal films, which can be easily accommodated. As long as the deformation was limited below the prestrain values, the interconnections could be repetitively stretched and released with the device performance maintaining invariant even after 100,000 deformation cycles. Compared to the in-plane "wavy" structures, the out-plane buckling interconnects can achieve larger stretchability for a given space between the device islands, leading to improved active device coverage on the substrates. Moreover, with the stretchable interconnects designed in the orthogonal directions, the devices can be subjected to different modes of deformations such as biaxial strain, twisting, shear, and related strain.

3.2 Electrodes with stretchable materials.

Although the stretchable electrodes based on patterned structures have been demonstrated as successful approaches to achieve stretchable EL devices, their fabrication required multiple steps of patterning to accomplish the designed structures on rigid supporting platforms. Efficient transfer methods were required to tether the fabricated devices on elastic substrates after the complicated patterning processes. To avoid the difficulties, different strategies were reported.

The direct usage of stretchable electrode materials was demonstrated to be a straightforward method that can circumvent the fabrication difficulties in the stretchable structures. Single-wall carbon nanotube (SWNT) paste was reported by Sekitani *et al.* as a promising material for stretchable electrodes [15]. The conductive inks were first prepared by mixing SWNT and ionic liquid in 4-methly-2-pentanone to form black paste-like gels. After mixing with a fluorinated copolymer (vinylidenefluoride–tetrafluoroethylene–hexafluoropropylene), which was also dissolved in 4-methly-2-pentanone, the resultant composite gel became conductive rubber once dried. The composite gel could be directly patterned onto elastic substrate by screen printing through shadow masks, forming intrinsically stretchable electrical interconnections. Conductivity of the rubber could be tailored by varying the SWNT content in the composite. Increasing the SWNT amount could significantly improve conductivity of the rubber. The stretchability of the conductive rubber would be decreased with a trade-off effect. With the SWNT content higher than 6 wt%, conductivity of the rubber was above 50 S/cm, while the stretchability would decrease below 40%. Figure 4a shows the stretchable conductors printed on an elastic PDMS substrate. OLEDs fabricated separately with the conventional process were laminated onto the stretchable conductors to achieve a rubber-like stretchable LED display, as demonstrated in Figure 4b. Compared to stretchable thin-film structures, the idea of printable conductive gel on elastic substrate was proven to significantly reduce the fabrication difficulties. Inspired by the material strategy, different ideas have been reported to fabricate stretchable electrodes with new stretchable conductive materials. For example, Kim *et al.* had fabricated stretchable electrodes with reduced electrical conductivity under increased stretching strain, as shown in Figure 4c [59]. Nickel powders mixed in PDMS were used as the conductive ink, which was exposed to high magnetic field to align the nickel powders vertically to the polymer surface before the PDMS curing. The conductor showed unique property with rapidly decreased resistance under increasing tensile strain, attributed to the increased conductive paths with more nickel powders in contacts under strain. Many other fillers, such as nanotubes, nanoplates, nanoparticles, and textiles, were also demonstrated to be promising materials that could be embedded into the elastic polymer matrix for stretchable electrodes [60–66].

4 Fully stretchable EL devices

The devices discussed earlier depend on stretchable conductors with either structural or material strategies. The objective of achieving stretchable light-emitting elements remains unresolved. To achieve fully stretchable EL devices, the challenges of fabricating stretchable light-emitting elements need to be tackled. As discussed in Section 2.2, because of the structure complexity, it is rather demanding to achieve stretchable LED devices in their multilayer structures. Nevertheless, the structural strategy that has been exploited to achieve stretchable con-

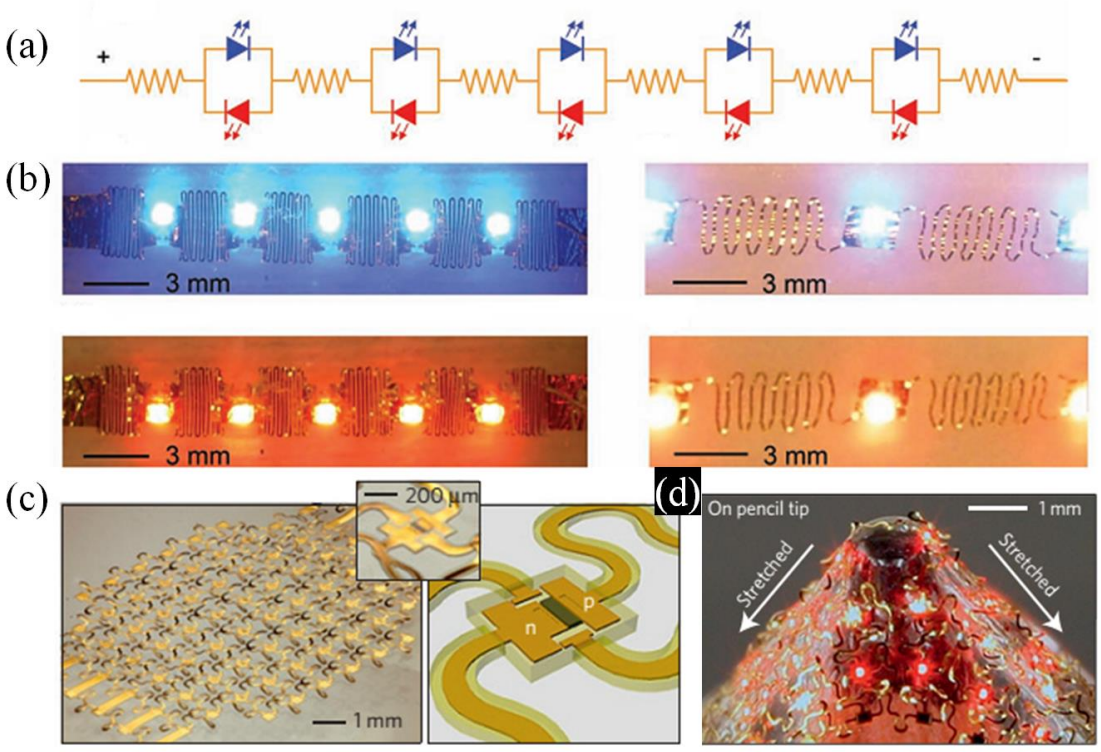

Figure 3: (a) The circuit model of the LED arrays for stretchable EL devices, consisting of blue and red LEDs connected reversibly. (b) The blue or red LEDs were controlled to turn on or off by reversing the applied bias under relax states (left photographs) and under 140% strain (right photographs). Reproduced with permission from [57]. (c) An optical image of an array of μ-ILEDs connected by non-coplanar serpentine metal bridges on a PDMS substrate. The right image is a schematic illustration of a single device with a representative device shown in the inset photograph. (d) An optical image of the stretchable EL device conformably attached onto a sharp pencil tip. Reprinted with permission from Macmillan Publishers Ltd: [Nature Materials] [17], copyright (2010).

ductors can also be applied on the thin-film LED devices. As shown in Figure 5a, to meet the extreme flexing conditions that will be induced in the stretchable structures, ultra-thin PLEDs were fabricated on a 1.4-μm PET foil [16]. The ultrathin PLEDs were fabricated using similar procedures as the conventional PLED devices. First, poly(3,4-ethylenedioxythiophene):poly(styrenesulphonate) (PEDOT:PSS) was deposited as the transparent electrodes. Poly(p-phenylene-ethynylene)-alt-poly(p-phenylene-vinylene) (PPE-PPV) derivative or poly[2-methoxy-5(3,7-dimethyloctyloxy)]-1,4-phenylenevinylene (MDMO-PPV) thin film was subsequently prepared as the light-emitting polymer by spin coating. LiF/Al was thermally evaporated as the back contact. Finally, the device was peeled off from the supporting substrate (glass) after fabrication, yielding a device thickness of around 2 μm. With the significant reduced thickness, the device could allow a bending radius below 10 μm. The ultrathin PLEDs were then pressed and adhered to a prestrained elastomer tapes. Similar to the arch-shape electrode structures in Section 3.1, the thin devices could form buckling structures to relax the compressing strain when the prestrained tapes were released. The final devices could be crumpled (Figure 5b) or stretched up to the prestrained state (100% stretching strain, Figure 5c).

In a different way, the material strategies were also demonstrated to achieve fully stretchable EL devices with much simpler device architectures. Pei's group has been developing the technologies in stretchable LEECs and reported leading works in the area [18, 19, 41, 67–69]. Figure 6a illustrates the fabrication procedure of the stretchable LEEC device. The transparent and stretchable electrodes were fabricated by embedding thin film of silver nanowire (AgNW) network into stretchable polymers. Briefly, AgNWs were first spray coated onto a glass substrate and formed random NW network. To achieve better conductivity and transparency in the NW networks, AgNWs with larger length are beneficial to reduce the number of junctions and thus to reduce the junction resistance in the networks [63, 70]. After the coating process, copolymer of siliconized urethane acrylate oligomer (UA) and ethoxylated bisphenol A dimethacrylate (EBA) were

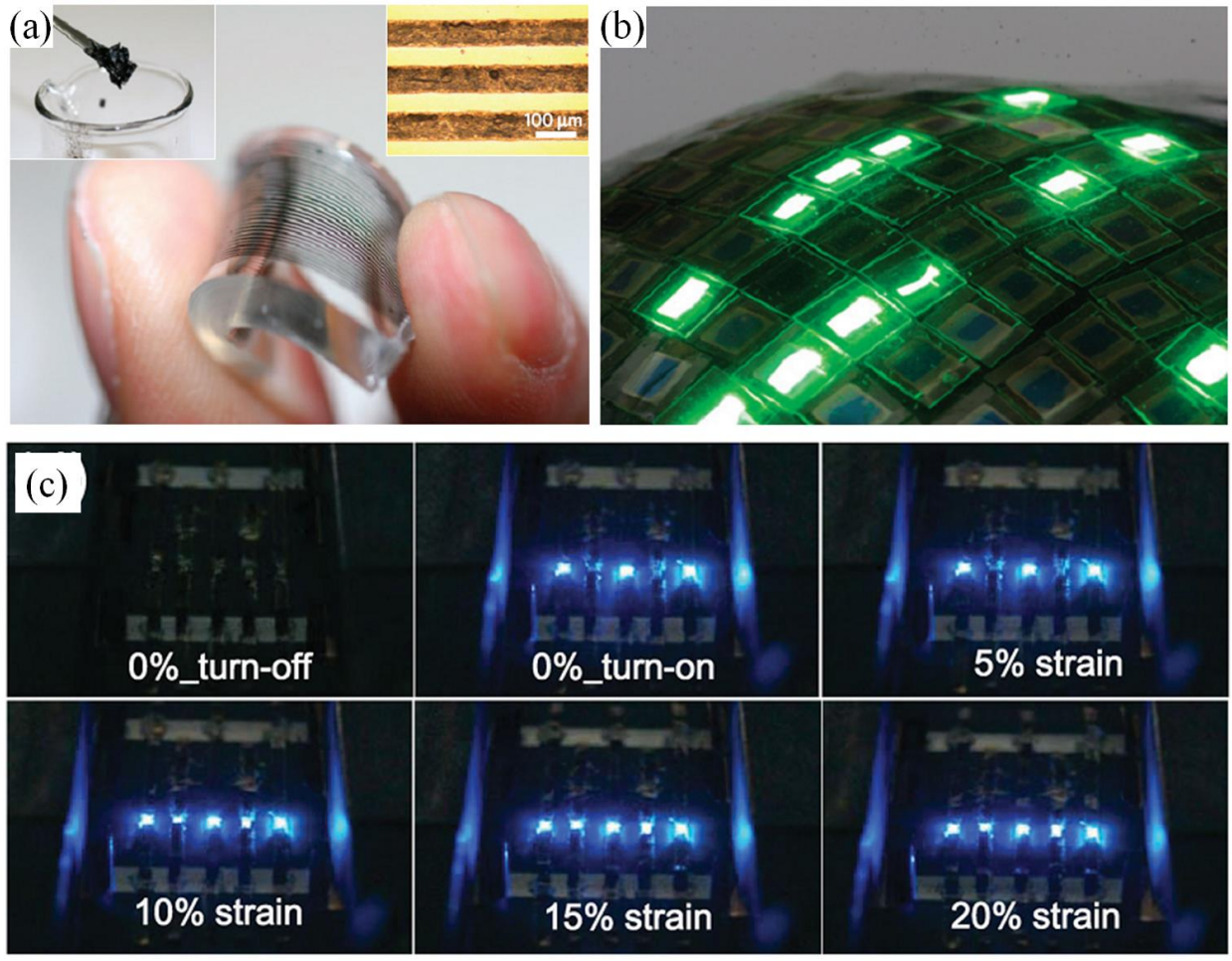

Figure 4: (a) A photograph showing the stretchable elastic conductors printed on a PDMS substrate using the SWCNT paste (left inset photograph) with linewidth of 100 μm (micrograph on the right inset). (b) A photograph of the stretchable display fabricated by assembling active-matrix OLED arrays. The stretchable display can be attached onto arbitrarily curved surfaces. Reprinted with permission from Macmillan Publishers Ltd: [Nature Materials] [15], copyright (2009). (c) Blue LED devices connected by elastic conductor with negatively strain-dependent resistance. Reproduced with permission from [59].

poured onto the percolating NW network, followed by subsequent curing with ultraviolent light. The embedding polymers might not be limited to PUA polymer. Properties of the polymers such as the glass-transition temperature, stretchability, transparency, and bonding force with the network to be transferred should be considered. If the bonding interactions between the polymers and conductive networks are weak, insufficient transfer will lead to dramatic conductivity degradation in the thin and transparent layers. Additives in the polymers will help to increase the bonding force between the polymers and NWs, assisting the transfer process. For instance, EBA was added into UA and Zonyl was added into PDMS before curing to improve the transferring process [39, 40]. After peeled off from the supporting substrate, stretchable and transparent electrodes were achieved. Subsequently, the EL layer consisting of a blend of yellow light-emitting polymer (a poly(p-phenylenevinylene) copolymer known as SuperYellow), ethoxylated trimethylolpropanetriacrylate (ETPTA), polyethylene oxide (PEO), and lithium trifluoromethane sulphonate (LiTf) was spun after the stretchable electrodes were protected by a thin layer of poly(3,4-ethylenedioxythiophene):poly(styrenesulphonate) (PEDOT:PSS). SuperYellow is a light-emitting material with high molecular weight, which can retain its functionality under large strain. ETPTA and PEO are the stretchable ionic conductors that transport ionics from the LiTf. The prepared stretchable electrodes were laminated on top of the emissive layer to complete the device fabrication.

With all the device structures constituting of stretchable materials, the polymer light-emitting electrochemical cell (PLEC) device can accommodate large mechanical de-

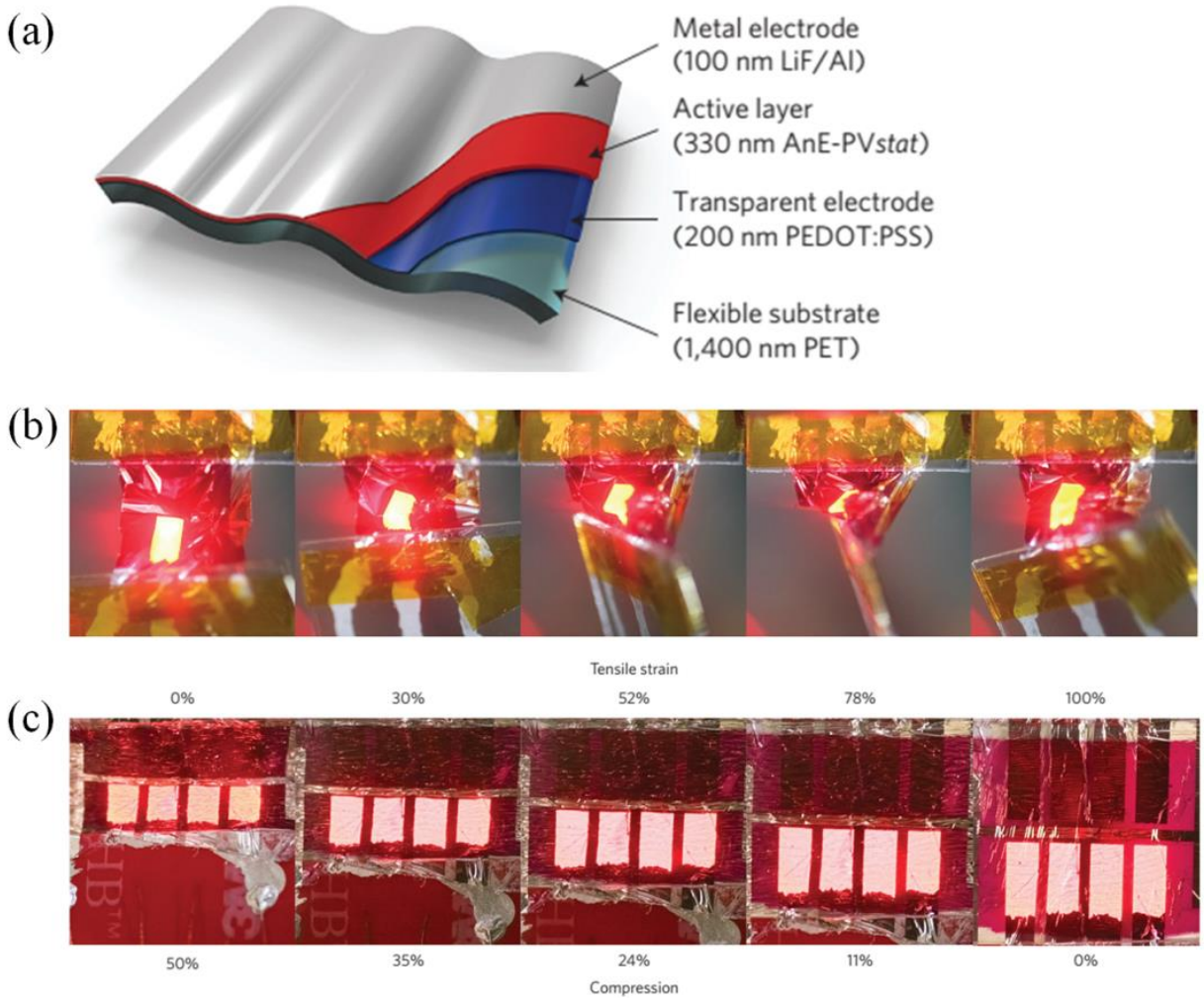

Figure 5: (a) Schematic drawing of the ultrathin PLEDs. (b) Photographs of the ultrathin PLEDs under crumpling test. (c) Photographs of the ultrathin PLEDs adhered onto prestrained elastomer tape. The PLEDs buckled to accommodate the strain after the elastomer was relaxed and could be stretched up to the prestrained state. Reprinted with permission from Macmillan Publishers Ltd: [Nature Photonics] [16], copyright (2013).

formations with stretching strain up to 120% before device failure, as shown in Figure 6b. The PLEC could achieve the emission intensity of 320 cd/m^2 at the applied voltage of 16 V (Figure 6c). The external quantum efficiency of the PLEC devices was calculated to be 4%, comparable to the state-of-the-art performance of EL devices based on SuperYellow fabricated with other electrodes. Emission intensity of the device reduced with the increased stretching strain, as illustrated in Figure 6d. The brightness reduced for ~85% at 120% strain (compared to the initial brightness at 0% strain), which could be attributed to the increased resistance in the stretchable electrodes. Mechanical cycling of the devices was limited at 30% strain. The device performance deteriorated when the stretching strain is 40% or higher. First of all, the electrical conductivity of the electrodes gradually degraded under the large stretching strain, reducing the current injection into the device. Second, pinholes were observed in the EL layers after the stretching and the density of the pinholes increased with the increasing stretching cycles, indicating the limited stretchability of the EL layers.

While the stretchable LEECs will need to overcome the problems of reduced conductivity in the transparent electrodes and limited stretchability in the emissive polymers, stretchable ACEL devices were reported to possess significantly improved stretchability [20]. As schematically shown in Figure 7a, the ACEL device consists of a stretchable emissive layer with the phosphor powders mixed in stretchable polymer matrix. The emissive layer is sandwiched between two stretchable and transparent elec-

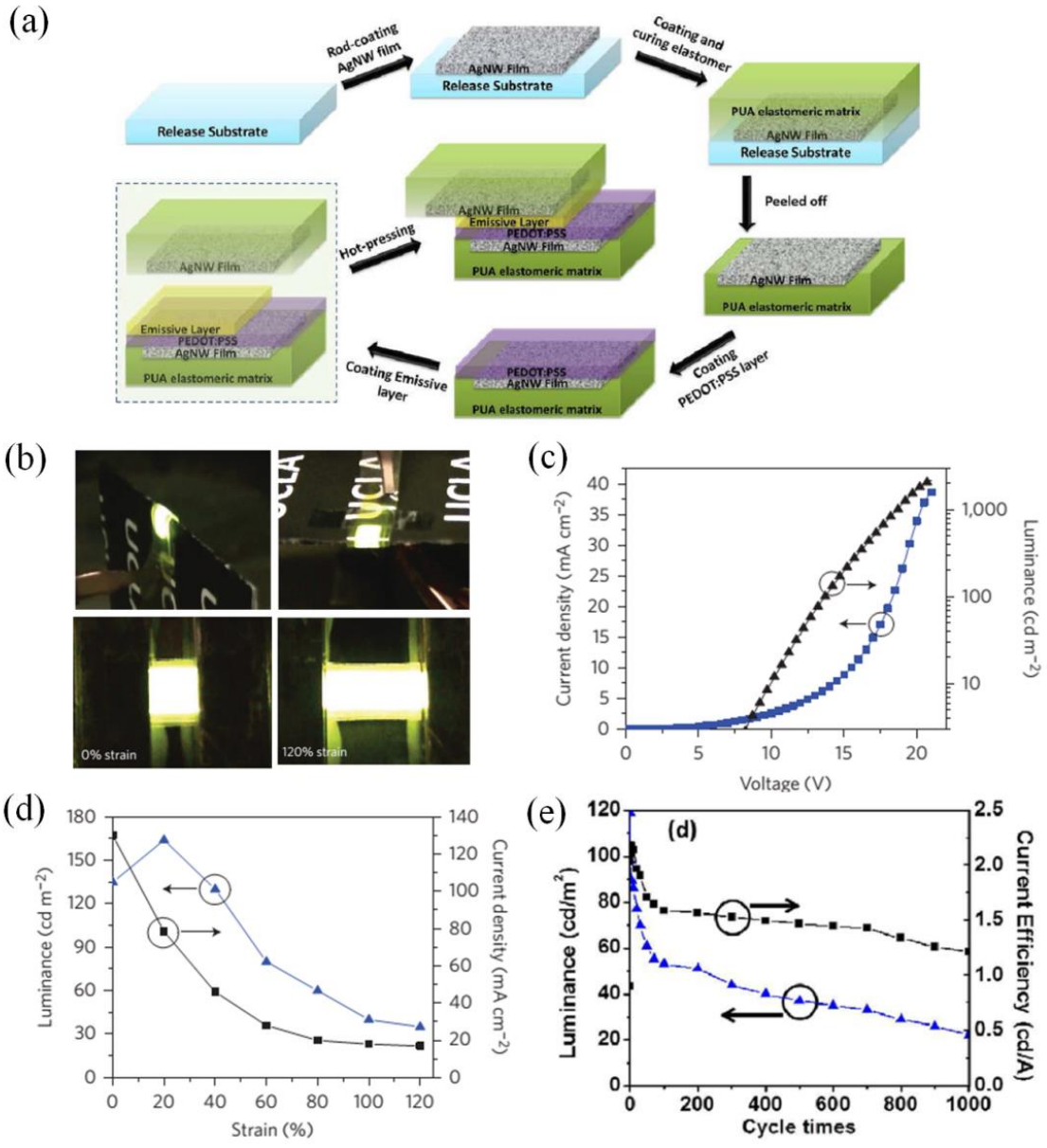

Figure 6: (a) A schematic illustration showing the fabrication process of the stretchable PLECs. (b) Images of a PLEC device wrapped around a cardboard with 400-μm thick and stretched to a maximum strain of 120%. (c) Current density and luminance characteristics of the PLEC under different driving voltages. (d) Change of the luminance and current density of the PLEC with increased strain (biased at 12 V). (e) Mechanical cycling of the PLEC device at 30% strain. Reprinted with permission from Macmillan Publishers Ltd: [Nature Photonics] [18], copyright (2013).

trodes. The fabrication of stretchable ACEL devices starts with the stretchable and transparent electrodes. Instead of using the embedding and transferring method, the AgNW network was directly spray coated onto an elastic substrate and simultaneously embedded into the polymer matrix by subsequent deposition of the EL layer. With this method, damage to the conductive percolating networks during the transfer process can be avoided to maintain high conductivity in the transparent layer. PDMS was the stretchable polymer used for the device fabrication. During the subsequent layers deposition, the PDMS liquid would partially dissolve the underneath PDMS layer, leading to "*in situ*" assembling of the different layers. With the "*in situ*" assembly, strong bonding strength was established in the different layers that was crucial to achieve the highly stretchable device structures. As the emission property is mainly decided by the voltage applied on the EL layers, the stretchable ACEL devices can achieve stable performance under

mechanical deformations as discussed in Section 2.3. The ACEL devices are required to operate under alternating current and the emission intensity are affected by frequencies of the bias. As shown in Figure 7c, emission luminance of the device significantly increased with increased bias frequencies. The luminance reached 225 cd/cm^2 with the bias field of 7.5 V/μm at 50 kHz. The estimated luminance efficiency was ~5 lumens/W. Significant improved stretching stability was observed in the ACEL devices. As presented in Figure 7d and 7e, emission intensity of the device remains quite stable. The variation is below 13% between the maximum and minimum emission intensity. The emission intensity remained at ~62% after stretched to 80% strain for 1,000 cycles.

A relative thick emissive layer (~40 μm) was used for the devices fabrication. As a result, the operating voltage of the device at 200 cd/m^2 is around 250 V. Their applications in portable or wearable electronics may be limited because of the high voltage operation. Stretchable ACEL devices with organic phosphors or structural strategies to reduce the operating voltage will need to be developed before the devices could be used in these areas.

5 Unprecedented applications with stretchable EL devices

The excellent mechanical conformability in the stretchable EL devices makes them feasible to keep conformable contacts with static or dynamic curvilinear surfaces, easily comply to external deformations with maintained functionality, driven to arbitrary shapes with internal forces, and so on, leading to unprecedented applications. Here, we review some of these exciting applications.

5.1 Wirelessly powered light-emitting systems

Biomedical device is one of the important applications with stretchable EL devices that can be implanted onto parts of the human body to accelerate wound healing, activate photosensitive drugs, or image the internal tissues. A system with the stretchable EL device connected to a spiral inductor coil was demonstrated by Kim *et al.*, as shown in Figure 8a [58]. The inductor coil can inductively couple the electrical field in radio frequencies and convert it into voltage output to power the connected ILEDs. As shown in Figure 8b, with the "soft" nature, the wireless ILED system can be laminated conformably onto a mouse model and covered with dermis. The ILEDs can be lighted up by the primary coil through inductive coupling. The stretchable wireless devices will have potential applications in implantable and biocompatible systems.

5.2 User-interactive electronic skin

By integrating the OLED arrays with thin-film transistors and pressure sensors, Wang *et al.* had demonstrated a user-interactive electronic skin that could provide direct and quantified visualization of the applied pressure on the devices [71]. Figure 9a shows the schematic illustration of the integrated devices. The OLED device was connected between a nanotube thin-film transistor (TFT, independently control the on and off states of the OLED) and a pressure-sensitive rubber (PSR). Conductivity of the PSR increased with the applied pressures, leading to the increase in the emission intensity from the OLED, which will provide instant quantified visualization on the applied pressures, Figure 9c and 9d. The user-interactive system will find interesting applications in automotive control panels, interactive input devices, robotics, and health monitoring devices.

5.3 Active self-deformable EL devices

Integrated with other functional components, the stretchable EL devices can be imparted with combined functionalities that are beyond the capability of conventional devices. For example, with the simple device fabrication, the ACEL devices could be directly integrated with dielectric elastomer actuators (DEAs) to achieve self-deformable EL devices [20]. The working mechanism of the DEAs is based on the Maxwell stress on the dielectric elastomer when the electrical bias was applied. Under electrical bias, opposite charges on the top and bottom electrodes of the elastomer will generate attraction force, while the similar charge on the same electrodes give repulsion force. The resultant Maxwell stress ($p = \varepsilon\varepsilon_0 E^2$, where ε is the relative permittivity of the elastomer, ε_0 is the permittivity of free space, and E is the applied electrical filed) on the elastomer compresses it and leads to lateral expansion. Fabrication procedures of the device are illustrated in Figure 10a. The DEAs were fabricated by spray-coated AgNWs network on both sides of the prestrained 3M VHB elastomer. The stretchable EL device was subsequently fabricated on the DEAs. Figure 10b shows a cross-sectional view of the self-deformable ACEL devices. Figure 10c shows that the device can be dynamically driven into different area ex-

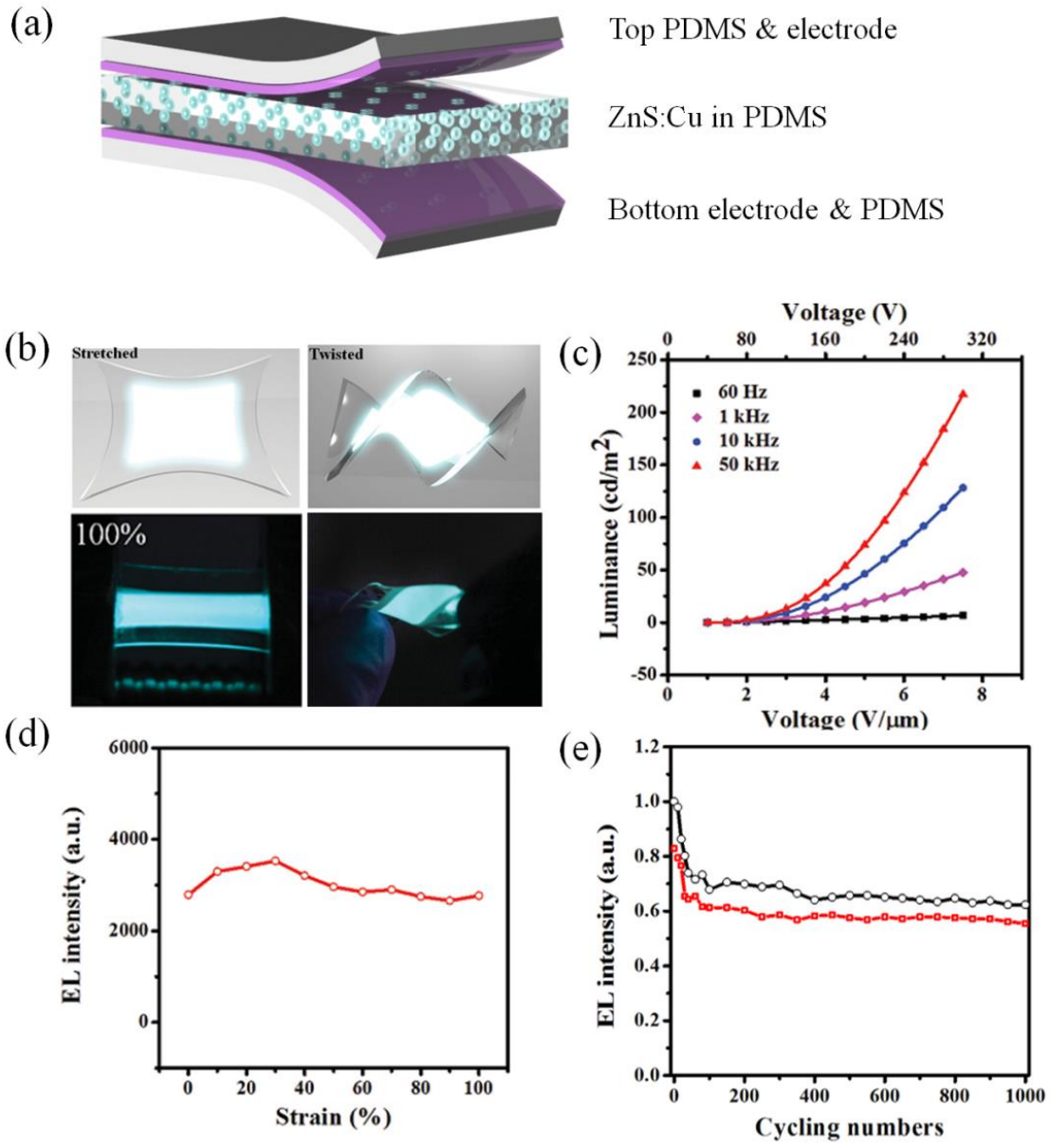

Figure 7: (a) A schematic illustration of the stretchable ACEL device architecture, which included the ZnS:Cu powders mixed in PMDS as the emissive layer and AgNW networks embedded in PDMS as the top and bottom electrodes. (b) Schematic images and photographs of the stretchable ACEL device under stretching and twisting. (c) Luminance characteristic against the applied voltages. (d) Stretching stability test of the ACEL device with the maximum strain of 100%. (e) Mechanical cycling test of the ACEL device at the stretching strain of 80%. Reproduced with permission from [20].

pansion under electrical bias. Figure 10d is the area expansion under the corresponding applied voltages. The novel idea demonstrated in this device may inspire a plethora of new applications such as volumetric displays, which can represent the objects virtually in three physical dimensions, and interactive display systems, which can provide users tactile interaction beside conventional planar graphic information.

6 Summary and Outlook

To conclude, we have reviewed the recent progress of stretchable EL devices that provide new technologies and solutions for the next-generation soft lighting and display applications. Stretchable EL devices are still in their nascent stage compared to their preceding flexible generation. Many challenges and hurdles still exist in the en-

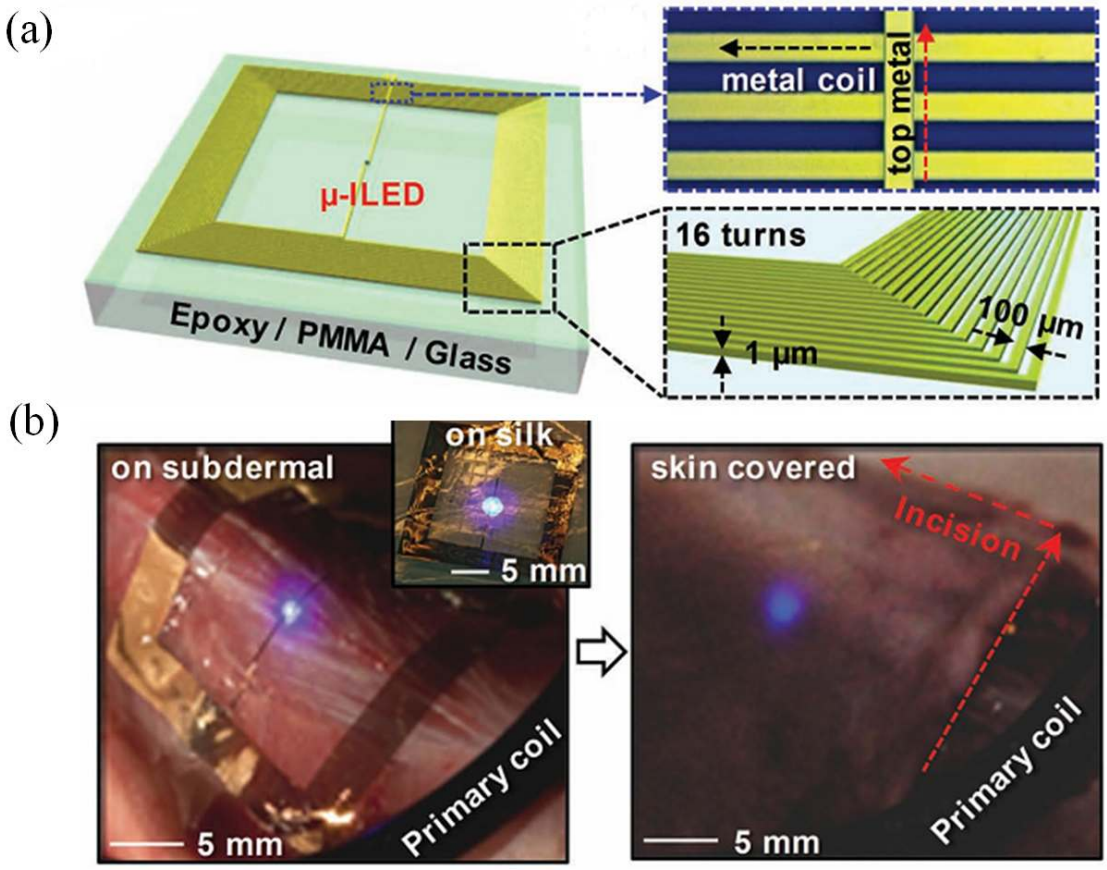

Figure 8: (a) Schematic illustration of the wireless μ-LED device design. The spiral inductor coils are used to power the connected μ-LED wirelessly. Inset pictures show the enlarged top interconnects and wireless coil design. (b) Optical images of the wirelessly powered μ-LED laminated onto a mouse model (left) and covered with dermis (right). Reproduced with permission from [58].

abling technologies. Stretchable ILEDs based on the assembled rigid and discrete light-emitting elements will encounter difficulty in the trade-off between active component coverage (larger component coverage will increase the pixel density) and bridge distance of the devices islands (longer bridge distance will increase the stretchability). Many fundamental issues need to be investigated, such as the mechanical matching between materials with largely varied modulus, adhesion, and interface between the different layers for endurable device structures. More importantly, a cost-effective solution is required to overcome the complicated patterning, transferring, and bonding processes. The unexplored charter for stretchable ILED devices is the emerging light-emitting NWs. NWs possess structural advantages with improved crystallinity and unique waveguide property that are beneficial to increase both the internal and external quantum efficiencies. In addition, the light-emitting NWs are significantly miniaturized emissive elements that can be easily embedded into stretchable polymers and harvest stretchability from the composite materials. They have been widely studied in flexible EL devices, showing advantages over the thin-film counterparts [21, 31, 72–75]. Although no study has been reported with demonstration on stretchable NW EL devices, it is believed that the overwhelming advantages in these devices will be fully used once effective approaches have been developed to transfer the NWs from high-temperature substrates onto elastic platforms.

On the other hand, intrinsically stretchable EL devices based on PLECs and ACEL devices show significantly simplified fabrication procedures with all-solution deliverable approaches. One of the challenges in the PLECs is their dramatically affected device stability under stretching states. To overcome the problem, stable conductors with good conductivity and transparency are required. Further improvements in the PLECs performance also require systematic study, especially stabilization of the dynamic junctions after the $p - i - n$ junctions are electrochemically formed. Stability of the junctions is directly related to the key figures of merit of the devices in their

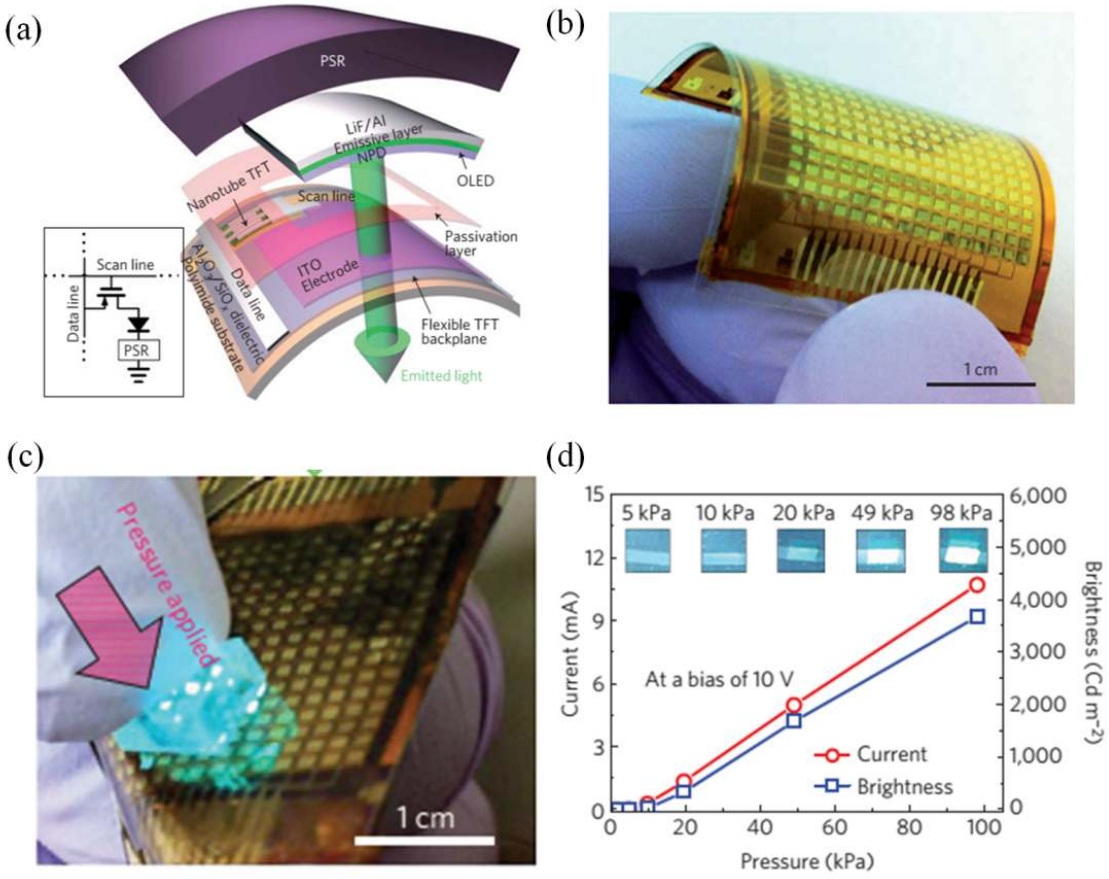

Figure 9: (a) Schematic layout of a single device on the user-interactive e-skin, including a TFT, OLED, and pressure sensor integrated in the whole system. (b) Optical image of a fabricated user-interactive e-skin device under bending. (c) A photograph of the device responding to the external pressure. The OLED turned on at the pressing location. (d) Device performance of the e-skin under different pressures. Reprinted with permission from Macmillan Publishers Ltd: [Nature Materials] [71], copyright (2013).

response speed, efficiency, and long-time stability. ACEL devices have been developed for a few decays. Systematical study with their emission behavior, device performance, and material systems have been well established. They prevail over with their low production cost and stable performance under severe environment conditions. The unique emission mechanism in the ACEL devices provides the advantage to achieve stable EL performance under large strain compared to the LEDs and PLECs. It also accommodates a wide choice of binding polymers. Polymer matrix with high dielectric constant will help to concentrate the electrical field on the ACEL particles, while polymer matrix with large stretchability will help to improve the stretching limitation in the ACEL devices. Future improvement in the ACEL devices should emphasize on reducing the operating voltage and increasing the quantum efficiency.

Apart from the emissive devices, the nonemissive devices also hold great potentials for the stretchable display applications. For example, the electrophoretic and electrochromic devices are of great interests. However, more research endeavors are required to develop corresponding technologies such as electrochemically stable conductors, stretchable electrodes with good transparency, and stretchable materials for the functional layers.

It is undoubted that the stretchable EL devices will meet another great success in the near future with functionality and versatility exceeding their preceding flexible generation. Before that, the enabling and supporting technologies will need time and efforts to establish. Successful outcomes will lead to revolution in the way the information is delivered and communicated.

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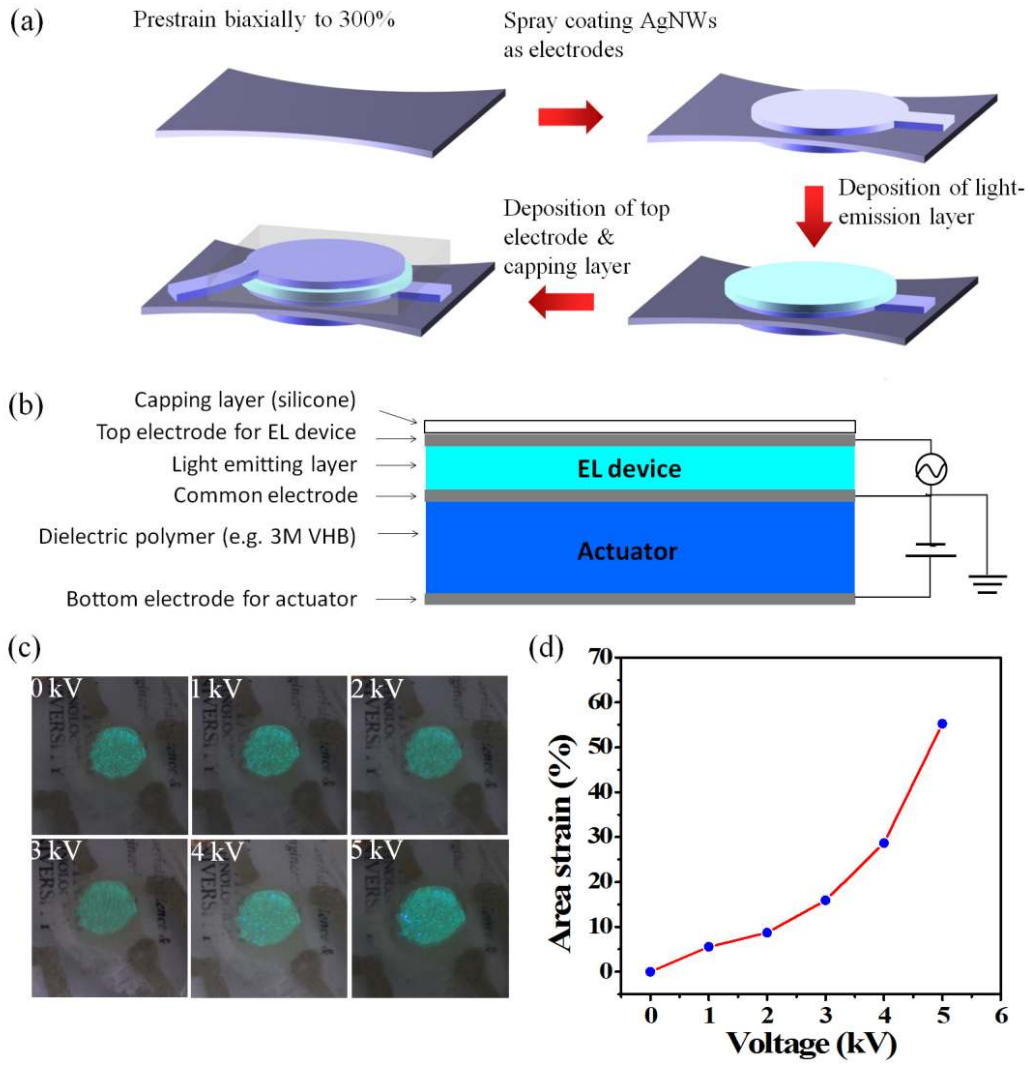

Figure 10: Schematic images of the self-deformable ACEL device: (a) the device fabrication procedure and (b) the cross-sectional view of the device. (c) Photographs of the self-deformable ACEL devices actuated under different bias. (d) Electrical actuating performance of the device. Reproduced with permission from [20].

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