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Review

Electrochemiluminescent devices: Structural composition, preparation process and applications for flexible/stretchable displays

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ABSTRACT

Flexible and stretchable light-emitting devices are being developed to meet market needs, drive technological innovation, address application challenges, and improve user experience. With the improvement of living standards, there is a growing demand for stretchable light-emitting devices and wearable displays with user interaction features, a technology that ensures that electronic products work properly under bending, folding, curling, stretching, etc., and will play an even more important role in the future. Recently, electrochemiluminescent devices (ECLDs) have shown great potential in the field of flexible wearable displays due to their low driving voltage, high luminescence efficiency and excellent flexibility compatibility. In this paper, this review discusses ECLDs based on the research framework of "material-process-application" synergistic optimization. Based on the principle of electrochemiluminescence (ECL) with annihilation and co-reaction mechanisms, the material selection strategies of luminophores, ionic liquids (ILs) and conductive electrodes are discussed. The design strategies and preparation processes of ECLDs are summarized, including solution coating process, vapor deposition, patterning process, printing technology and electrospinning. In the end, focusing on multi-color displays, stretchable and flexible integration and wearable application scenarios, this review introduces the current research status of ECLD, and the future development direction is proposed. This review aims to provide a comprehensive reference for the research and application of ECLDs, and to promote its development from laboratory R&D to practical application.

1. Introduction

With the "Made in China 2025" and related policies to promote the Internet of Things, Industry 4.0, big data, artificial intelligence, robotics and digital health advances to make everything interconnected, the rapid development of the flexible electronics industry has entered the era of accelerated integration of technologies. The rapid penetration and influence of the mobile Internet into the relevant system is adding endogenous power to the development of the industry[1–3]. The rapid development of flexible electronics technology is profoundly changing the field of human-computer interaction and information display, wearable devices have become one of the research hotspots in the field of flexible electronics[4].

With the rapid growth of application scenarios such as wearable devices, e-skin and smart healthcare, traditional rigid light-emitting devices are difficult to meet the demand for dynamic deformation, long-term wearability and low energy consumption due to the defects of high-power consumption, mechanical brittleness and thick substrate. Flexible wearable devices have the characteristics of soft, lightweight, bendable, and adheres well to human skin[5–7]. With the continuous progress in the fields of nanotechnology and material science, the performance and functionality of flexible wearable devices have been greatly improved, and they have a wide range of applications in the fields of health monitoring[8–10], human-computer interaction[11,12], soft robotics[13], and haptic interfaces [14–16], etc. At the beginning of the 1990s, researchers began to integrate the electronic devices on flexible substrates. Foreign research has been relatively mature, and the industrial chain is relatively complete; while the relevant research in China started late, but has been rapidly developed in recent years, and some leading research teams and enterprises have emerged[1,17]. In flexible wearable devices, stretchable displays achieve instant response and signal feedback to external stimuli through rapid luminescence, and play an important role in the visualization field of the new generation of intelligent flexible electronics.

Traditional display technologies such as liquid crystal displays (LCDs), light-emitting diodes (LEDs), alternating current (AC) -driven excited

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flexible stretchable electroluminescent devices (ACELDs) have made significant progress, but still face many challenges in terms of flexibility and wearability. For example, the complexity of the multilayer structure requires high preparation requirements, and operation at high voltages may pose risks[18–21]. Synthesizing the existing conventional light-emitting devices and inspired by bioluminescence, a novel AC-driven electrochemiluminescent devices (ECLDs) have entered researchers' vision. Electrochemiluminescence (ECL), also known as electrogenerated chemiluminescence, refers to the generation of luminophores by electrochemical reactions on the surface of an electrode by applying an electric potential, followed by a series of redox processes. When these molecules or ions in the excited state return to the ground state, photons are released and luminescence occurs[22]. ECL is an ideal combination of electrochemistry and spectroscopy. It has the sensitivity and wide dynamic range of traditional chemiluminescence, while combining the advantages of electrochemical methods, such as simplicity and stability. It does not require the introduction of expensive laser light sources, no interference from the background light source and low cost. What's more, ECL has a shorter light emission time and better spatial control, and thus is widely used in luminescence display and sensing analysis.

To realize high-performance ECLDs, especially flexible/stretchable ECLDs, in-depth studies on device composition, preparation process, and applications development are required (Fig. 1). Firstly, starting from the principle of ECL (Chapter 2), we analyze the material selection principles of luminophores, ionic liquids (ILs), and conductive electrodes. Secondly, we compare and analyze the preparation processes such as solution coating, vapor deposition, patterning, printing technology, electrospinning (Chapter 3), and compare and analyze the applicable scenarios of different preparation processes. Subsequently, in combination with the multi-color displays, stretchable and flexible displays, and wearable scenarios (Chapter 4), then, we discuss the interface optimization and performance enhancement strategies of the devices in dynamic environments. Finally, the development and application prospects of ECLDs are summarized and outlooked (Chapter 5).

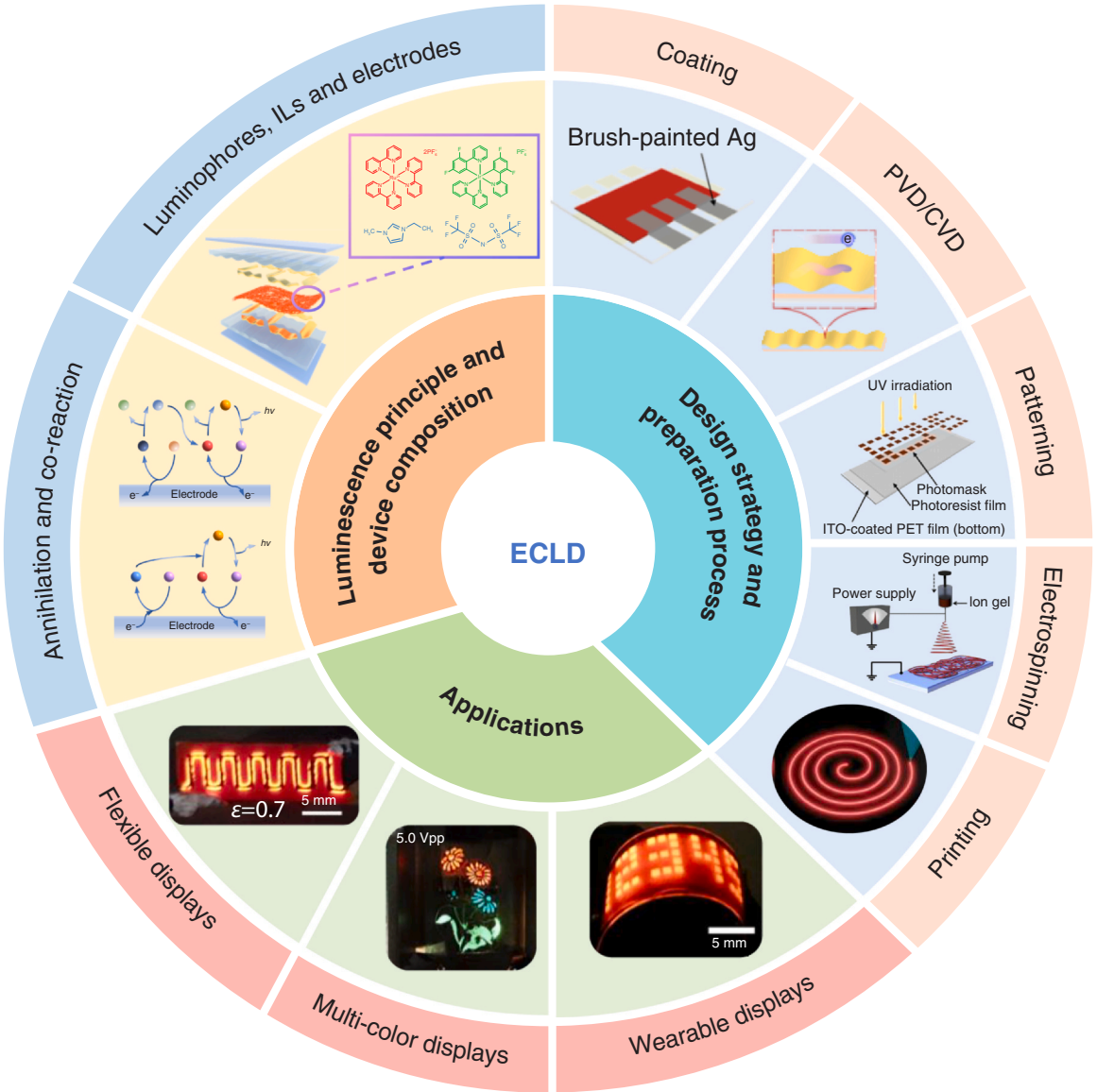

Fig. 1. Composition, preparation and applications of ECLDs. The luminescence principle and device composition include annihilation and co-reaction[23,24], luminophores, ILs and electrodes[25] (Copyright 2024, Wiley-VCH). The design strategy and preparation process include coating[26] (Copyright 2019, American Chemical Society), physical vapor deposition/chemical vapor deposition (PVD/CVD)[25] (Copyright 2024, Wiley-VCH), patterning[27] (Copyright 2015, American Chemical Society), electrospinning[28] (Copyright 2018, Elsevier) and printing[29] (Copyright 2024, Wiley-VCH). The applications include flexible[30] (Copyright 2017, American Chemical Society), multi-color[31] (Copyright 2020, Elsevier) and wearable displays[32] (Copyright 2014, American Chemical Society).

2. Principles, material selection and design strategies of ECLDs

In recent years, the market share of foldable and rollable electronic products has gradually increased, and demand for bendable and stretchable displays continues to grow. As a result, the development of flexible and stretchable LEDs that support a wide range of deformations has received significant attention. Most LED devices have a similar structure, where the semiconductor material generates photons using externally applied electron-hole pairs that undergo a compounding process. Electrons are injected from the cathode through the electron injection and transport layers, and holes are injected from the anode through the hole injection and transport layers, and subsequently enter the emitting layer, where they emit light through radiative compounding. Since the thickness of each active layer is very thin (only a few tens of nanometers), ultrathin LEDs can be prepared. Typical LEDs, such as organic light-emitting diodes (OLEDs) as well as quantum dot light-emitting diodes (QLEDs), will offer mechanically compliant elastic substrates combined with rigid LEDs[33]. Stretchable LEDs can be used as emitting pixels for optoelectronic sensors, which can be effectively applied in human-computer interaction, smart displays, and other fields by combining components such as sensor units, transistor arrays, and circuits[34–36]. Since the first flexible OLED was prepared in 1997 and the first flexible QLED in 2009, deformable LED display devices have been developed rapidly, and inorganic thin films, organics, polymers, quantum dots, and chalcogenides can be used as alternative materials for the emissive layer. Some studies have been devoted to increasing the degree of deformation freedom and further developing from flexible LEDs to stretchable. Lee et al.[37] reported a flexible and transparent electroluminescent (EL) skin based on QLEDs, which can display high-resolution pressure distribution images in real time. Choi et al.[38] reported an ultrathin, wearable colloidal QLED array. They prepared red, green, and blue arrays with uniformly aligned pixel sizes and resolutions up to 2460 ppi by gravure transfer printing. The technology is universally applicable and easily scalable, and shows high luminescence performance (14,000 cd/m^2) in low-pressure-driven white QLEDs and electronic tattoos. In addition, the devices maintain stable performance on flat, curved, and folded planes, and the high-performance arrays above demonstrate the possibility of integrating high-definition full-color displays in flexible wearable electronics. Uniform light emission, stable performance, and ultra-thin size are the advantages of OLED/QLED for applications such as pattern displays, pressure mapping, and implantable electronic systems [39,40]. However, this scheme may require additional transport layers and carrier injection to balance the carrier concentration, which is not conducive to low-cost and large-scale fabrication. Some components in the materials, such as Cd in CdSe quantum dots or Pb in chalcogenide materials are toxic and need to be replaced and improved. Researchers are actively studying the synthesis of new optoelectronic materials and their applications in flexible devices and system integration[41].

ACELDs are generally multilayer structures between EL functional layers and dielectric layers embedded in electrodes. Utilizing EL phosphor materials (e.g., ZnS:Cu, ZnS:Mn particles), they can be mixed with dielectric elastomers to confer intrinsic stretchability to the device. The rapid development of ACELDs has revolutionized the field of functional wearable devices and smart visual sensing and recognition. Compared with direct current (DC) actuators, ACELDs typically have lower energy consumption and longer lifetime, and are used in multi-color, high durability, stretchable and wearable displays[40,42]. Shi et al.[43] used “warp” and “weft” interlaced structures to realize large-area displays based on textile morphology, and employed micro-EL units to provide low-cost and highly flexible wearable displays for next-generation electronics. He et al.[44] used electrospinning silver nanofibers electrodes as the basis for the preparation of multi-color, high-resolution, large-area, and stretchable non-contact light-emitting interactive display devices. Zhang et al.[45] fabricated ACELDs with high compatibility and enhanced interlayer adhesion through customized ink and 3D printing techniques. The role of ACELDs in enabling digital visualization in next-generation electronic systems, including wearable electronics, soft robotics, and dermal bioelectronics, is becoming more and more apparent with the growing demands[46–48]. The shortcomings lie in the fact that high voltage operation may pose a risk and requires further development of the emitter layer to reduce the operating voltage. Moreover, it also consumes more energy to maintain operation.

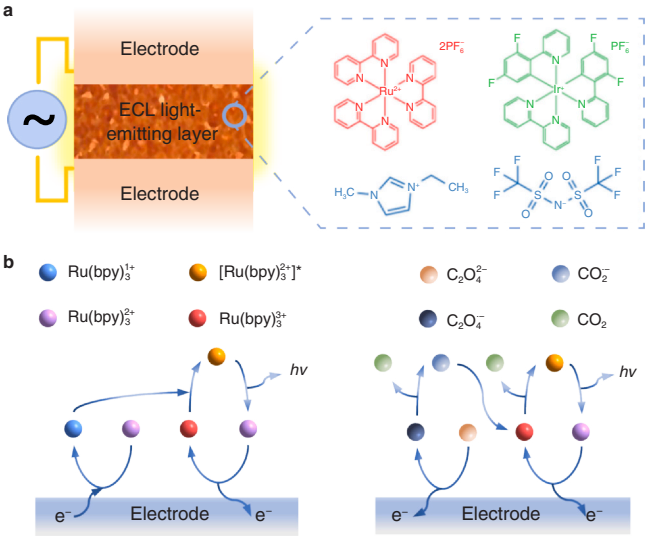

Fig. 2. Principles of ECL. (a) The structure of ECLD[23]. (b) Annihilation-type ECL and co-reactant-type ECL mechanism[23,24].

In this context, ECLD stands out with its unique ECL mechanism-directly modulating the luminescence process through electrochemical technology, which has high sensitivity, fast response, and wide detection range, and thus has a wide range of applications in chemical analysis, biomedicine, and optoelectronic devices[49,50]. ECLD can realize high-brightness and high-contrast display at low voltage, and is also highly compatible with flexible substrates, which is expected to provide a new breakthrough solution for display technology. Compared to OLED/QLED and ACELD, ECLD has a simple structure consisting only of electrode layers and a light-emitting layer (Fig. 2a). It has lower operating voltage (excitation is possible with an AC voltage of 10 V or less), and the electrodes are not limited by the work function (upper and lower electrodes can be made of the same material). Even more appealing is the low-cost availability of large-area fabrication and fast response to luminescence. Based on the above advantages, ECLD has the potential to be applied in realizing flexible/stretchable light-emitting displays. Flexible electrodes and devices can provide greater freedom in design and operation, and flexible integrated arrays will reduce circuit complexity and wiring costs by reducing multiple types of wiring[51]. The structural design of ECLDs has a significant impact on their performance and applications. By optimizing the electrode materials, light-emitting layer materials, electrolyte materials, and preparation processes, efficient, long-lasting, and stable luminescence can be achieved[52–54].

2.1. Principles of ECLDs

ECL is an electrochemical-photophysical process in which the reaction occurs near the electrode surface in order to form an excited state and emit light. Since it is a luminescent phenomenon by bimolecular recombination of electrically excited generation radicals, the luminescence mechanism can be classified into annihilation-type and co-reaction-type mechanisms depending on the source of the radicals[23]. The principle of ECL is specified below using the representative red luminescent tris(2,2′-bipyridine) ruthenium(II) ($[Ru(bpy)_3]^{2+}$) as an example (Fig. 2b).

2.1.1. Annihilation-based mechanisms

The free radicals in the annihilation-type mechanism are generated from a single luminophore, and the luminescence process of ECLDs in the annihilation regime can be described as the stages of electron transfer, excited state generation, and radioluminescence. The luminophore undergoes direct electron transfer and is oxidized at the anode

or reduced at the cathode under the control of an AC voltage, resulting in the generation of excited or ground state luminophores. The left panel of Fig. 2b illustrates the distribution of luminophores near the electrode at one end of the applied bias voltage, as follows.

(i) Luminophores infiltrated in ionic liquid [EMIM][TFSI] are uniformly distributed before applying AC voltage;

(ii) Under the action of electric potential, $[Ru(bpy)_3]^{2+}$ was induced to be distributed at both electrodes to form a double electric layer (EDL), and the AC bias produced redox luminophores with the following reaction process:

$$\text{Reduction: } Ru(bpy)_3^{2+} + e^- \rightarrow Ru(bpy)_3^{1+} \text{ (Cathode)} \quad (2\text{-}1)$$

$$\text{Oxidation: } Ru(bpy)_3^{2+} - e^- \rightarrow Ru(bpy)_3^{3+} \text{ (Anode)} \quad (2\text{-}2)$$

(iii) Slow diffusion of reduced luminophores produced at the cathode. When the applied potential on the changing electrode is positive, the oxidized luminophores are produced much faster than the diffusion of the original reduced luminophores, and the two compounds to undergo electron transfer.

(iv) A pair of luminophores in the ground and excited states is produced by an annihilation reaction:

$$\text{Generation of excited state: } Ru(bpy)_3^{1+} + Ru(bpy)_3^{3+} \rightarrow Ru(bpy)_3^{2+} + [Ru(bpy)_3^{2+}]^* \quad (2\text{-}3)$$

Here, $[Ru(bpy)_3^{2+}]^*$ is the excited emitter. The excited luminophore has the following emission:

$$\text{Light emission: } [Ru(bpy)_3^{2+}]^* \rightarrow Ru(bpy)_3^{2+} + h\nu \quad (2\text{-}4)$$

Eventually the excited state luminophore returns to the ground state, accompanied by a luminescence process that releases energy $h\nu$. Thus, the luminescence is excited under AC bias without being limited by the electrode's work function. At the same time, due to a concentration difference, the reduced $Ru(bpy)_3^{1+}$ and the oxidized $Ru(bpy)_3^{3+}$ diffuse individually until relative equilibrium.

2.1.2. Co-reaction-based mechanisms

The co-reactive mechanism, on the other hand, involves a bimolecular electrochemical reaction between the luminophore and the co-reactant, in which an electrical signal applied to the electrode prompts the electrolyzed substance to become an oxidized or reduced intermediate. Subsequently, the intermediate further reacts with the oxidized or reduced co-reactant to form an unstable excited state. Once the substance returns from the excited state to the ground state, a light signal is emitted. The co-reaction method was first introduced by Bard et al. [24]. The electrochemical reaction of oxalate in the anodic region was studied in acetonitrile in the absence and presence of $[Ru(bpy)_3]^{2+}$, respectively. The right panel of Fig. 2b demonstrates the distribution of luminophores in the vicinity of the anode in the following procedure.

(i) Oxalic acid ($C_2O_4^{2-}$) and $[Ru(bpy)_3]^{2+}$ are oxidized simultaneously, $[Ru(bpy)_3]^{2+}$ is oxidized to form $Ru(bpy)_3^{3+}$, and oxalic acid oxidized produces $CO_2^{\bullet-}$ with high reducing capacity:

$$\text{Oxidation: } Ru(bpy)_3^{2+} - e^- \rightarrow Ru(bpy)_3^{3+} \quad (2\text{-}5)$$

$$C_2O_4^{2-} - e^- \rightarrow C_2O_4^{\bullet-} \quad (2\text{-}6)$$

$$\text{Generation of reducing agents: } C_2O_4^{\bullet-} \rightarrow CO_2^{\bullet-} + CO_2 \quad (2\text{-}7)$$

(ii) At this point $CO_2^{\bullet-}$ reacts with $Ru(bpy)_3^{3+}$ to produce an excited state luminophore:

$$\text{Generation of excited state: } Ru(bpy)_3^{3+} + CO_2^{\bullet-} \rightarrow [Ru(bpy)_3^{2+}]^* + CO_2 \quad (2\text{-}8)$$

(iii) Then it returns to the ground state with ECL:

$$\text{Light emission: } [Ru(bpy)_3^{2+}]^* \rightarrow Ru(bpy)_3^{2+} + h\nu \quad (2\text{-}9)$$

The above annihilation-based and co-reaction-based mechanisms are universal for other ECL light-emitting materials. For example, annihilation-based ECLs typically incorporate the reaction of aromatic compounds to produce radicals for stabilizing cations and radicals for anions under a wide potential window in organic solvents. Two common types of co-reactants used in co-reactive ECL processes are oxidizing-reducing co-reactants and reducing-oxidizing co-reactants. The reaction occurs at an electrode to which a positive bias voltage is applied and ultimately produces light is categorized as an anodic ECL or is referred to as oxidized-reduced ECL. The co-reactants involved in the process are also referred to as anodic co-reactants, and the main ones are oxalates ($C_2O_4^{2-}$), tripropylamine (TPrA), 2-(dibutylamino)ethanol (DBAE), and hydrogen peroxide (H_2O_2), etc. The most successfully commercialized reaction of this type is the $Ru(bpy)_3^{2+}$/TPrA system, known for its common use in ECL immunoassays. Typical of the $Ru(bpy)_3^{2+}/C_2O_4^{2-}$ system, the reaction process of which has been described in detail above, is that organic amines produce strong chemiluminescence by reacting with $Ru(bpy)_3^{2+}$, while these compounds can also be electrochemically oxidized to generate highly reactive intermediates. Another common combination is luminal (2,3-aminophenylhydrazine)/H_2O_2 In this combination, luminal is electrochemically oxidized in the presence of H_2O_2, leading to the generation of 3-aminobenzene dicarboxylic acid and the emission of blue light. If the charge transfer occurs between the reduced luminophore and the cathodically generated oxidant, the reaction is classified as a cathodic ECL (reduced-oxidized ECL). In $Ru(bpy)_3^{2+}/S_2O_8^{2-}$, a reduction-oxidation type system, persulfate is reduced to sulfate anion radicals ($SO_4^{\bullet-}$) and sulfate ions (SO_4^{2-}), and eventually $Ru(bpy)_3^{2+}$ emits a strong ECL in acetonitrile solution emits a strong ECL signal. The in-depth study of the above systems confirms that a large number of ECL-emitting materials follow the mechanism of annihilation and co-reaction. In addition, the two mechanisms can also make up for each other's deficiencies, e.g., when the free radicals generated in the annihilation pathway have short lifetimes, the use of co-reactants can capture the radical ions and be used to promote the reaction. After understanding the mechanism of ECL sensing, the materials that make up each part of the ECLD are detailed below.

2.2. Materials selection and electric field regulation of ECLDs

In the initial stages, ECLDs are usually driven using DC methods [26,55]. After switching on the applied voltage, it takes a long time (~1 s) to observe the cell's emission, which is due to the collision of cationic and anionic radicals at the cathode and anode, respectively. The oxidized and reduced state luminophores need to diffuse slowly over long distances and tend to react with impurities (e.g., oxygen) in the solution, leading to a decrease in the luminescence efficiency. In contrast, in the AC drive, the polarity of the electrode is switched rapidly in each half-cycle, which makes the oxidized and reduced state luminophores generate and collide with luminescence rapidly on the electrode surface, reducing the residence time in the solution, and thus reducing the reaction with impurities. At the same time, ECL can reduce the accumulation of ionic impurities on the electrode[55,56]. This driving method improves the slow conduction response, and by applying an AC voltage at 30 Hz achieves a fast response of about 10 ms [57] and a significant improvement in the luminous intensity[58], which is conducive to the improvement of luminescence efficiency (Fig. 3a). Therefore, we focus on ECLs driven by AC voltage.

2.2.1. Luminophores

Although there have been fruitful scientific results in ECL-related sensing, the high-throughput nature of ECL imaging still requires high luminophore luminescence efficiency. So far, only a few luminophore systems have been used for visual ECL analysis. Depending on the luminophore, electrochemical luminescence can be generally categorized into inorganic, organic and emerging systems such as nanoluminescence and aggregation-induced luminescence.

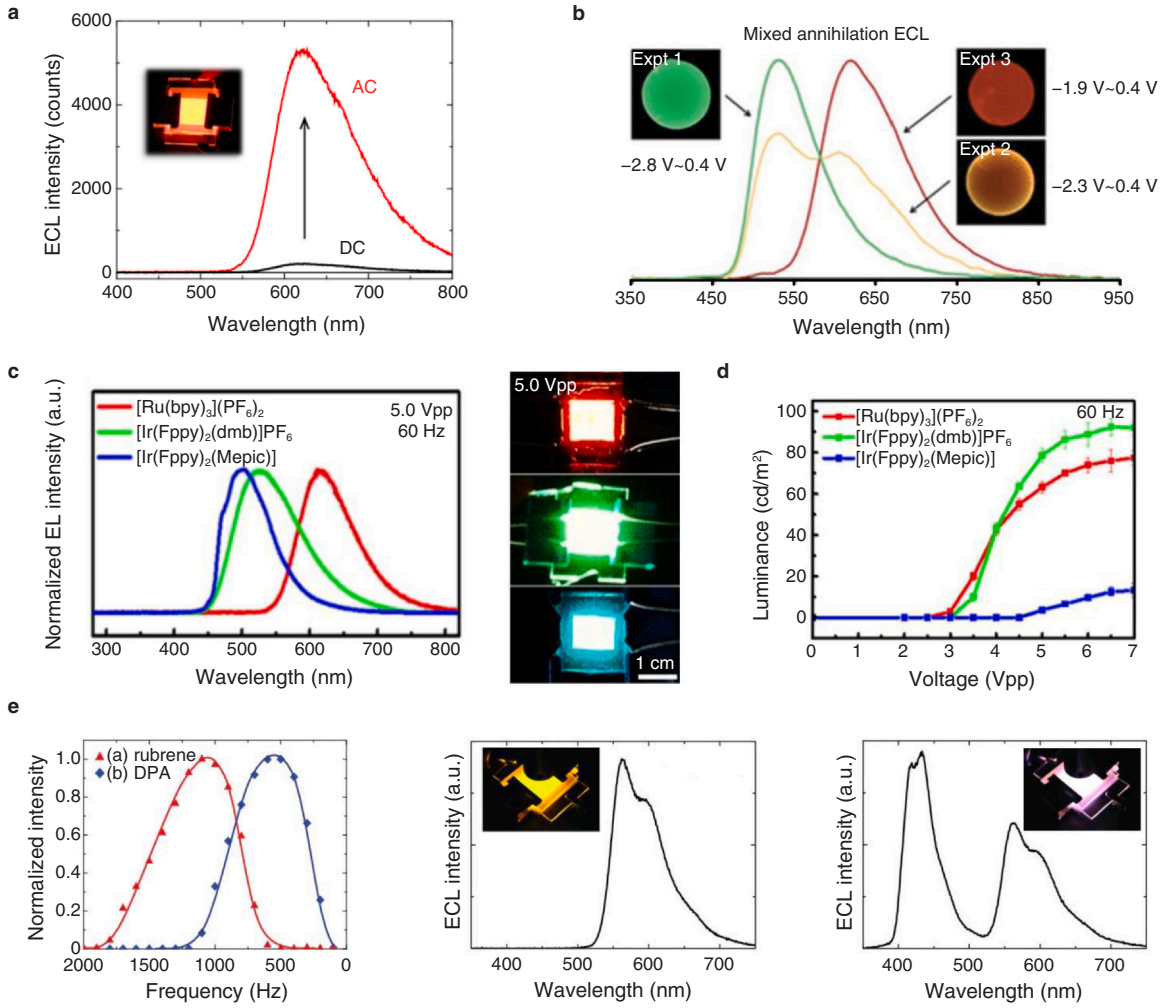

Fig. 3. Regulation of ECL luminous performance. (a) ECL intensity and photographs under AC/DC bias[55]. Copyright 2013, IOP Publishing. (b) Spectra and photographs of color-tunable ECL under different applied AC bias[61]. Copyright 2015, The Royal Society of Chemistry. (c) EL spectra and photographs of different light-emitting layer materials[31]. Copyright 2020, Elsevier. (d) Luminance of above materials under different applied AC bias[31]. Copyright 2020, Elsevier. (e) Spectra and photographs of color-tunable ECL under different applied frequency[62]. Copyright 2013, Wiley-VCH.

Since the ECL phenomenon of silicon quantum dots has been reported, new materials such as quantum dots, noble metal nanoclusters, carbon nanomaterials, upconversion nanoparticles, and so on, have emerged, which have promoted the in-depth study of ECL. Among them, ruthenium(II) complexes are the most commonly used luminescent groups with good water solubility, stable chemical properties, reversible redox, and high luminescence efficiency, and are the most studied electrochemiluminescent active substances. And they have been successfully used in commercial luminescent displays and sample detection. In addition to ruthenium(II) complexes, iridium(III) complexes are also a typical representative of metal complexes in ECL. Compared with ruthenium(II) complexes, iridium(III) complexes have higher luminescence efficiencies and a wider range of luminescence wavelengths, and the joint use of the two is expected realize advantageous in the high-efficiency multi-color displays[59]. Hogan et al.[60] observed the luminescence spectra of a mixed annihilation ECL system containing ruthenium(II) and iridium(III) complex, in which multiple emitters emitted simultaneously, showing multi-color luminescence signals in blue and green. In addition, the overall color of the luminescence was adjustable by applying electrode potentials using redox reactions to achieve the luminescence color change from orange-red to green[61] (Fig. 3b), and the voltage-controllable luminescence color change provides the possibility of developing new ECL display technologies and sensors. Kwon et al.[31] achieved fine control of luminescence colors and intensities by adjusting the concentration of luminescence and mixing ratios of luminescent substances modulation. By mixing green and blue light emitters, a hybrid metal complex enhancement system was designed, which significantly improved the brightness and stability of blue light emission and helped to achieve red, green, and blue colors in the same ECLD (Fig. 3c).

The materials of ECLDs exhibit unique advantages and limitations in different systems. Ruthenium(II) complexes are the core materials for traditional ECL detection and monochromatic displays due to their high stability, good water solubility, and mature commercial applications, but their narrow luminescence wavelength range and single color limit the potential for multi-color displays. In contrast, iridium(III) complexes have higher luminescence efficiency and wider luminescence spectra, and can be color-adjusted by ligand design, but their high synthesis cost and poor environmental stability limit large-scale applications. Quantum dots and nanomaterials achieve full-spectrum luminescence and high brightness by virtue of size tuning, and are candidates for high-resolution displays and short-term sensing. However, they are susceptible to oxidation or photobleaching, and some of them carry the risk of biotoxicity, and their long-term stability still needs to be optimized. Hybrid systems (e.g., ruthenium/iridium composite systems) achieve multi-color luminescence and voltage-modulated color change through synergistic interactions, significantly enhancing

brightness and dynamic sensing capabilities, but complex process requirements and inter-material energy losses may reduce system efficiency. Emerging materials such as aggregation-induced luminescence and upconversion nanoparticles excel in solid-state anti-burst extinction and unique optical properties, which are particularly suitable for cutting-edge fields such as bioimaging, but most of them are still in the laboratory stage, with complex synthesis processes and insufficient commercialization validation. Overall, the optimization of ECL materials needs to weigh the performance and cost with specific scenarios. In the future, the existing bottlenecks can be broken through the development of multifunctional composite systems, the enhancement of nanomaterial stability, the optimization of green synthesis processes, and the integration of intelligent control technologies. The combination of hybrid systems and emerging materials may become a key path to realize highly efficient, multi-color and stable ECLDs.

2.2.2. Ionic liquids

Ionic liquids (ILs), as a kind of molten salt composed of organic cations and inorganic/organic anions, play a key role in ECLDs due to their unique physicochemical properties (e.g., low volatility, wide electrochemical window, high ionic conductivity, and designability) [63]. As the core electrolyte of ECLDs, ILs not only need to provide efficient ion transport channels to support electrochemical reactions, but also need to be compatible with the luminescent layer and electrode materials to ensure the stability of the devices under dynamic deformation. Here take a typical 1-ethyl-3-methylimidazole bis(trifluoromethanesulfonimide) salt ([EMIM][TFSI]) as an example, with a high ionic conductivity, a wide electrochemical window, and low volatility and thermal stability, compatible with the redox potentials of common luminophores, such as $Ru(bpy)_3^{2+}$, which belongs to the class of imidazole ILs. In addition, there are pyrrolidines, quaternary ammonium salts and functionalized ILs after functional group modification. The use of ILs as electrolytes not only improves the safety of the device, but also simplifies the packaging process. Currently, imidazoles and pyrrolidines ILs dominate the market due to high conductivity and wide electrochemical window, while functionalization and compounding strategies further enhance their mechanical and environmental adaptability. However, high cost, short lifetime and humidity sensitivity issues still need to be broken through[22,64].

Conventional ECL layers are generally liquid-phase materials, which inevitably encounter leakage problems in applications, affecting device integrity and luminescence, and liquid electrolytes are difficult to realize complex shapes and patterning. Solid-state electrolytes have low ionic conductivity, which makes it difficult to be applied to the luminescent layer. The emergence of ionic gel electrolytes may help solve the above. Based on this, in 2014, Frisbie et al.[32] synthesized the first block polymer gel-based ECLDs, which greatly promoted the research work in display. Ionic gel ECLDs have great ionic conductivity, fast response time, and bright emission characteristics, and the viscous and deformable gel helps to prepare light-emitting devices without the risk of leakage[27,65].

2.2.3. Conductive electrodes

Transparent conductive electrodes are essential components in a series of optoelectronic devices such as light-emitting diodes, photodetectors, solar cells, liquid crystals and flat panel displays. With the rise of flexible and stretchable displays, stretchable electrodes, as one of the key components, are required to maintain stability and conductivity under deformation states such as stretching, bending, and twisting, and advances in material selection, structural design, and preparation strategies for electronic products have laid the foundation for emerging wearable technologies[66,67]. For stretchable electrodes, the basic requirement to achieve high performance is to balance conductivity and transparency with oxidation resistance, tensile stability and long-term stability. In order to have equilibriums of these goals, transparent conductive materials are first needed to maintain carrier injection and to enable light transmission. Various conductive materials including metals, nonmetals, carbon nanomaterials, conductive polymers, and ionic conductors are widely used in the preparation of electrodes[68,69]. Metal nanowires serve as the core material system for the new generation of flexible transparent electrodes (FTEs). Their structural diversity (including silver, copper, and gold monolithic nanowires and core-shell composite configurations) endows the materials with multidimensional performance advantages, such as outstanding photovoltaic properties, excellent mechanical properties, solution processability, and good compatibility with semiconductors. It is therefore considered ideal for FTEs. Compared with conventional ITO-based electrodes, metal nanowire-based FTEs exhibit higher electrical conductivity and light transmission, along with high yield strength and excellent quality factor. In recent years, FTEs based on metal nanowires have made significant progress in the application of flexible and stretchable optoelectronic devices, and their performance has been improving in terms of efficiency and stability, showing promising applications in the fields of wearable power supplies, displays, and sensors. It is worth noting that traditional methods for preparing transparent electrodes usually involve vacuum evaporation or magnetron sputtering. These methods require expensive equipment, which increases the cost of preparing electrodes. In contrast, the solution processability of metal nanowires enables them to be mass-produced by preparative processes.Conductive polymer materials utilize their conjugated π-electron system to achieve the transition from insulating to conducting states through chemical or electrochemical doping strategies, and such materials have received widespread attention for their unique electro-mechanical coupling properties. Typical representatives include polyacetylene (PA), polypyrrole (PPY), polyaniline (PANI), polythiophene (PT), and their derivatives, which not only exhibit tunable electrical conductivity, but also have the advantages of chemical modifiability, biocompatibility, lightweight, and reversible redox properties. In recent years, electrospinning has become an important process for the preparation of conductive polymer nanofibers, but their industrial application still faces the key challenge of precursor solution engineering.Hydrogel has become the preferred material system for flexible electronics due to its mechanical properties that are similar to those of biological soft tissues. Its unique material properties include high fracture toughness, high water content, biomimetic ionic conductivity, and excellent biocompatibility, however, in comparison to metal conductors, there is a significant gap of 6–9 orders of magnitude in the intrinsic ionic conductivity of hydrogels, and in order to enhance their electrical properties, the researchers developed a conductive filler-enhanced composite hydrogel system. By introducing metallic nanostructures (nanowires/particles), carbon-based materials (carbon nanotubes/graphene), or intrinsically conductive polymers (PEDOT:PSS, PANI, etc.) into the polymer network. It is possible to enhance the material conductivity to the order of $10^{-5}\sim10^{1}$ S cm^{-1} while maintaining tissue-like mechanical compliance.

Indium tin oxide (ITO) is generally a transparent teal film or a yellowish gray mass that combines visible light transmittance with conductive applications, making it one of the most widely used transparent conductive materials. It can be prepared using a range of deposition techniques, including evaporation, sputtering and solid solution processing. There is a need to balance conductivity and transparency during film deposition, as high concentrations of charge carriers will increase the conductivity of the material, but continued deposition will reduce transparency. In addition, ITO has poor mechanical strength and flexibility and needs to be annealed to achieve high conductivity. Limited by the nature of the material itself and the preparation process, it is not suitable for flexible optoelectronic devices. From the perspective of resource utilization, indium resources are limited and expensive, and there are concerns about the cost and sustainability of ITO[70]. Further development of alternative materials to ITO for transparent conductive electrodes is needed.

Gold has a low surface resistance due to high free electron density. This results in excellent conductivity and low resistivity, which ensures efficient and stable current transfer. This stability is particularly important because ECL reactions usually involve complex electrochemical processes, and the stability of the electrode directly affects the performance and lifetime of the device. Deposited thin layers of gold have a

relatively high transparency, which is favorable for visible light escape. What's more, the stretchability of gold electrodes makes them important for applications in the field of flexible electronics, especially for scenarios requiring long-term stable operation, high-precision measurements and biomedical applications[69].

Carbon-based materials play a vital role in the survival and activities of living things. It has structures such as carbon nanotubes (CNTs, 1D) and graphene (Gr, 2D), and these materials have excellent electrical and thermal conductivity, low density, very high specific surface area, and great chemical stability. More importantly, they have the desired tunable properties, and the high specific surface area provides for their modification, favoring carrier migration, device integration, and wearable applications. CNTs are lightweight and connected in a hexagonal structure. When used as fillers in polymer substrates, CNTs can dramatically increase carrier mobility without sacrificing the original plasticity of the substrate[71]. Gr consists of carbon atoms with sp^2 hybridization orbitals tightly packed into a monolayer, which is structurally very stable. Gr and its derivative, reduced graphene oxide, have a single-atom-layer thickness, which leads to excellent transparency and electrical conductivity, as well as excellent chemical, mechanical, and thermal stability[72–74]. Nanowires (NWs), especially metallic NWs, exhibit high electrical conductivity. In addition, they also possess excellent light transmission and flexural resistance due to nanoscale size effects and large aspect ratios[75,76]. These materials are options for transparent conductive electrodes.

Hydrogel[77,78], polyethylene glycol terephthalate (PET)[79], polyimide (PI)[80], polydimethylsiloxane (PDMS)[81,82], polyurethane (PU), thermoplastic polyurethane (TPU)[83], styrene ethylene butylene styrene (SEBS)[84], self-healing elastomers[85,86] and other natural or synthetic polymers are often used as substrate materials due to their flexibility, high strength, and interfacial stability with metals or other inorganic materials. This makes them compatible with a variety of fabrication techniques, including PVD (e.g., sputter deposition and thermal evaporation deposition), CVD, and lithography. The modulus of elasticity of the materials allows them to bend and stretch without physical damage, enabling them to conform to the surface of the skin and effectively transmit physical signals such as shape, temperature and softness.

Due to the pursuit of high transmittance and conductivity, ECLDs generally use transparent and brittle materials such as ITO as electrodes, so that the device is only flexible or partially stretchable and can be curved, and it limits the fully stretchable of ECLDs [87]. In addition, in the case of different materials for the two electrodes, there may be asymmetry in the luminescence of the top and bottom sides, which is a disadvantage for two-sided displays. Fully stretchable devices contribute to better tensile stability and mechanical robustness. To realize high-performance ECLDs, issues such as material selection, structural design, preparation process, and system integration need to be addressed in an integrated manner.

2.2.4. Magnitude and frequency regulation of electric fields

The performance of ECLDs is highly dependent on the magnitude and frequency of the applied AC voltage. In terms of voltage, low voltage (1–5 V) is suitable for the low power consumption of wearable devices, but the brightness is limited; high voltage (> 5 V) can significantly improve the brightness, but it is easy to cause electrode corrosion and electrolyte decomposition, which will shorten the life of the device. By optimizing the combination of emitters and electrolytes, high-brightness ECL displays at low voltages (the voltage peak-to-peak V_{PP} is less than 3.5 V) were achieved, and the voltage dependent luminescence stability is provided for displaying at long time (Fig. 3d)[31].

The applied frequency also has a significant effect on the modulation of luminous brightness and color. Appropriate frequency increase leads to faster polarity switching of the electrodes, and more oxidized and reduced state luminophores can be generated and collide quickly. The frequency-dependent luminescence property allows fine tuning of the ECL brightness by simply adjusting the frequency (Fig. 3e)[62]. For two luminescent molecules, Rubrene (RUB) and 9,10-diphenylanthracene (DPA), the ECL intensity changes significantly with the applied AC frequency. When an AC bias of 1000 Hz is applied, the device emits yellow light (contributed by RUB); while when the frequency is reduced to 300 Hz, the device emits white light (contributed by both RUB and DPA). By adjusting the frequency, color mixing can be achieved by varying the luminous intensity of different light emitting molecules in the same device. Under low-frequency (< 1000 Hz) conditions, the ions in the ionic liquid have enough time to migrate to the electrode interface, and the redox reaction is fully carried out, resulting in high luminescence efficiency (e.g., the peak luminance of RUB- and DPA-based ECLDs exceeds 100 cd/m^2 at 300 Hz), but the response speed is limited. While high frequency (> 1000 Hz) drive can improve the response speed, it may cause a decrease in brightness due to the ion migration lag (more than 50 % brightness attenuation at > 1000 Hz), and the accumulation of Joule heat accelerates the aging of the materials[50,62].

The synergistic optimization strategy shows that low-frequency high-voltage pulses (50–300 Hz, 5 V) can balance the transient brightness and material stability, while high-frequency low-voltage continuous drive (1–5 kHz, 2–3 V) is suitable for dynamic display scenarios. The matching design of materials and electric field parameters (e.g., low-viscosity ionic liquids, nanoporous electrodes) can mitigate the interfacial degradation and enhance the device performance. By precisely tuning the frequency and magnitude of the AC voltage, combined with the development of broadband-responsive electrolytes and fast luminophores, the ECLDs are expected to realize a wider range of applications in the fields of flexible displays and wearable electronics.

2.3. Design strategies of ECLDs

With the increasing demand and the trend of miniaturization of stretchable components, new challenges arise in the complete stretchable devices from laboratory to industrial production: namely, the connection and integration of different stretchable components, and the integration of the parts is affected by mechanical mismatch and stress/strain concentration at the interface. To avoid functional failure of stretchable devices, improving the interfacial bonding strength between components is also a research hotspot[88], which is needed to ensure a stable connection between electrode assemblies and flexible substrates to realize the stable operation of electronic components in stretchable circuits. For most stretchable materials, especially inorganic materials, it is difficult to maintain a stable percolation network all the time during the stretching process, posing a challenge for the preparation of high-performance stretchable electrodes. Structural design has gained attention as an effective solution for developing stretchable electrodes. In order to achieve device stretchability and compliance, there are two main types of electrode designs: structural stretchable and intrinsic stretchable[40,89,90].

For structural stretchability, design strategies include structural engineering (such as flexural engineering, island-bridge structures, etc.) and modulus/stiffness engineering[91]. For flexible/stretchable electrodes, it is required that they follow deformation actions such as bending, stretching and twisting and still maintain electrical properties. Structural design not only improves the tensile properties of inherently stretchable materials, but also introduces stretchability to rigid materials. To achieve this goal, strain-resistant structures need to be precisely designed and fabricated at scale. Different microstructures, such as wrinkled[92,93], serpentine[94], mesh[95], kirigami[96,97], and island-bridge[98], impart tensile properties to rigid materials by dissipating the internal stresses between the electrodes and the substrate, thus reducing the mechanical damage to the functional materials. It maintains the integrity of the connection between the electrodes and the devices, and improve the electrical stability, which is a key design to realize the stretchability of the device.

The design of structural stretchability significantly improves the stretchability of electrodes and devices on the one hand. However, on the other hand, there are still many problems to be solved in applications. For example, pleated structure ultrathin devices are usually accompanied by complex film transfer processes, large periodic pleats of hundreds of micrometers are clearly visible which can lead to image display distortion, and the rigid materials of island-bridge structures as well as interconnecting areas are prone to be deformed during the cycling process[98]. Even if the area of the rigid material is small, localized mechanical mismatches can still occur when in contact with human skin or other flexible planes. In addition, the functional island area may be limited due to the interconnection area occupying most of the space. These are challenges that hinder further use and need to be optimized. In addition to structural stretchability, the development of intrinsically stretchable electrodes and devices is also an option.

Intrinsic stretchability refers to the use of intrinsically stretchable materials to obtain stretchable properties. The components of the stretchable/wearable devices thus prepared, including electrodes, functional layers, and substrates, are inseparable from intrinsic stretchability. Advantages of intrinsic stretchability include the versatility of soft materials as well as the ease and low cost of the preparation process[67,99]. Wearable electronics can be in close contact with the skin both in the case of dimensional changes and following motion, and in addition, the mechanical, electrical, and optical properties of the device should always remain stable even after mechanical deformation processes such as bending, twisting, and flipping, making it an ideal medium for human-machine interfaces and implantable applications.

For intrinsically stretchable electrodes, the components are a conductive medium and an elastomeric substrate that ensures mechanical compliance. Various intrinsically stretchable conductors, including nanomaterials (nanoparticles, nanowires, nanosheets, etc.), hydrogels, and conductive polymers, are preferred materials for stretchable electrodes. There are mainly the following typical electrode structures [100]: (1) Uniformly dispersed in an elastomeric substrate with functional fillers such as hydrogels and electrolyte gels. (2) A thin layer of conductive material coated on the surface of the elastomer substrate. (3) The conductive material is embedded in the surface of the elastomer substrate to form a percolation network. Preparation of polymers by combining soft main chains, side chains, and flexible crosslinked networks results in molecular structures that combine specific functionality and high stretchability.

Despite many advances, the development of intrinsically stretchable materials is still in the making. The mobility of the materials is generally lower than that of conventional inorganic and even organic materials, and the electrical properties of intrinsically stretchable semiconductor materials have much room for improvement. In addition, the integration of stretchable composites is an issue that needs to be considered, as differences in material properties and different preparation methods may lead to unstable device interfaces[43].

3. Preparation process of ECLDs

Optimization of processing methods and molecular engineering are also required to ensure the stability of the stretchable materials to maintain the long-term operation of the devices. After material selection and structural design, the preparation of the luminescent layers and electrodes is also worth discussing and optimizing. In the case of metal and oxide films, for example, structural design is required to give them transparency and mechanical deformability, considering the realization of high conductivity and transparency. Different preparation processes have their own advantages and disadvantages, and the selection of an appropriate method depends on the specific application needs, material properties, cost and accuracy requirements. This chapter gradually transitions from rigid to flexible devices, demonstrating the preparation processes for ECLD light-emitting layers, electrodes, and the overall devices.

3.1. Light-emitting layers deposition

As the typical and most frequently used preparation methods, solution processing techniques such as spin coating, scratch coating, drop coating, and brush painting have far-reaching impacts in luminescent layer and electrode preparation, which, in combination with the patterned template process, can be used for large-area preparation of ECLDs. As shown in Fig. 4a, the anode and cathode substrates were aligned and bonded under a contact pressure of 1.5 MPa at 140 ℃ for 5 min using a bonding machine. Heat bonding significantly enhances the interfacial force. It is utilized to soften or activate the surface of the material, and then tightly affix two surfaces together under pressure to, forming a strong connection that defines the space and location for microfluidic injection of the solution. The solution flowed into the channel through capillary force without externally applied power, and the encapsulation of the ECL solution was realized by PI-PI thermal bonding and epoxy sealing[101,102].

Spin coating can make the luminescent solution or conductive materials on the substrate quickly and uniformly unfolded to form a flat and uniform thickness of the film, which is very critical for the uniform distribution of luminophores and electrolytes in the ECL layer, applicable to ITO, plastics, metals and other substrate materials. However, for large areas or uneven and irregularly shaped substrates, spin coating is difficult to achieve uniform coating. For scratch coating, the thickness and uniformity of the film can be precisely controlled by adjusting parameters such as the angle, pressure and speed of the squeegee. Scraping requires skill and patience, and improper operation can easily lead to defects in the film, such as scratches and bubbles, which can lead to a reduction in performance and stability.

The drop coating process is simple and easy to perform and does not require complicated equipment and operation. Only the luminescent solution or conductive material is added dropwise to the substrate, and then the film can be formed by natural drying or heating drying. Compared to spin coating, less material is wasted in the drop coating process, which improves the material utilization. Hwang et al.[103] utilized a star-shaped block copolymer ($(MS)_6$) as a polymer gel agent to drop-coat an ECL gel onto patterned ITO, utilizing adhesive tape as the binder, and delineating four spaced areas (size: 2 mm × 2 mm, Fig. 4b). The shortcomings of drop coating mainly lie in the fact that the formed film is prone to problems such as uneven thickness and surface roughness, especially when there are defects or unevenness on the substrate surface, which can affect the performance of the ECL or electrode layer and lead to reduced luminescence efficiency and stability. In addition, drop coating often requires a longer drying time to ensure that the solvent is completely evaporated, which inadvertently extends the preparation cycle.

Brush coating is suitable for a wide range of substrate materials and shapes and requires relatively low surface flatness of the substrate. The thickness and uniformity of the film can be controlled to a certain extent by adjusting the amount of solution dipped and the brush painting speed. As shown in Fig. 4c, Moon et al.[26] realized patterned ITO electrodes by a combination of photolithography and wet etching, which resulted in significantly higher patterning accuracy and more complex and diverse shapes compared to the use of tape to delineate areas. ECL gel was spin-coated on top and silver paste was uniformly brushed onto the surface of the already prepared ECL gel to form a uniform electrode layer. Silver electrodes with good conductivity and adhesion can be easily prepared, and together with ITO electrodes, the ECL gel is clamped to form a complete ECLD structure. The film after brush painting is prone to localized incomplete drying or over-drying due to the uneven drying speed, which requires high requirements for brush painting techniques and experience.

3.2. Electrode fabrication

Ultra-thin metal-based transparent electrodes are often prepared by PVD methods (including thermal evaporation, magnetron sputtering, etc.), in which the solid phase is transformed into a gas phase, and

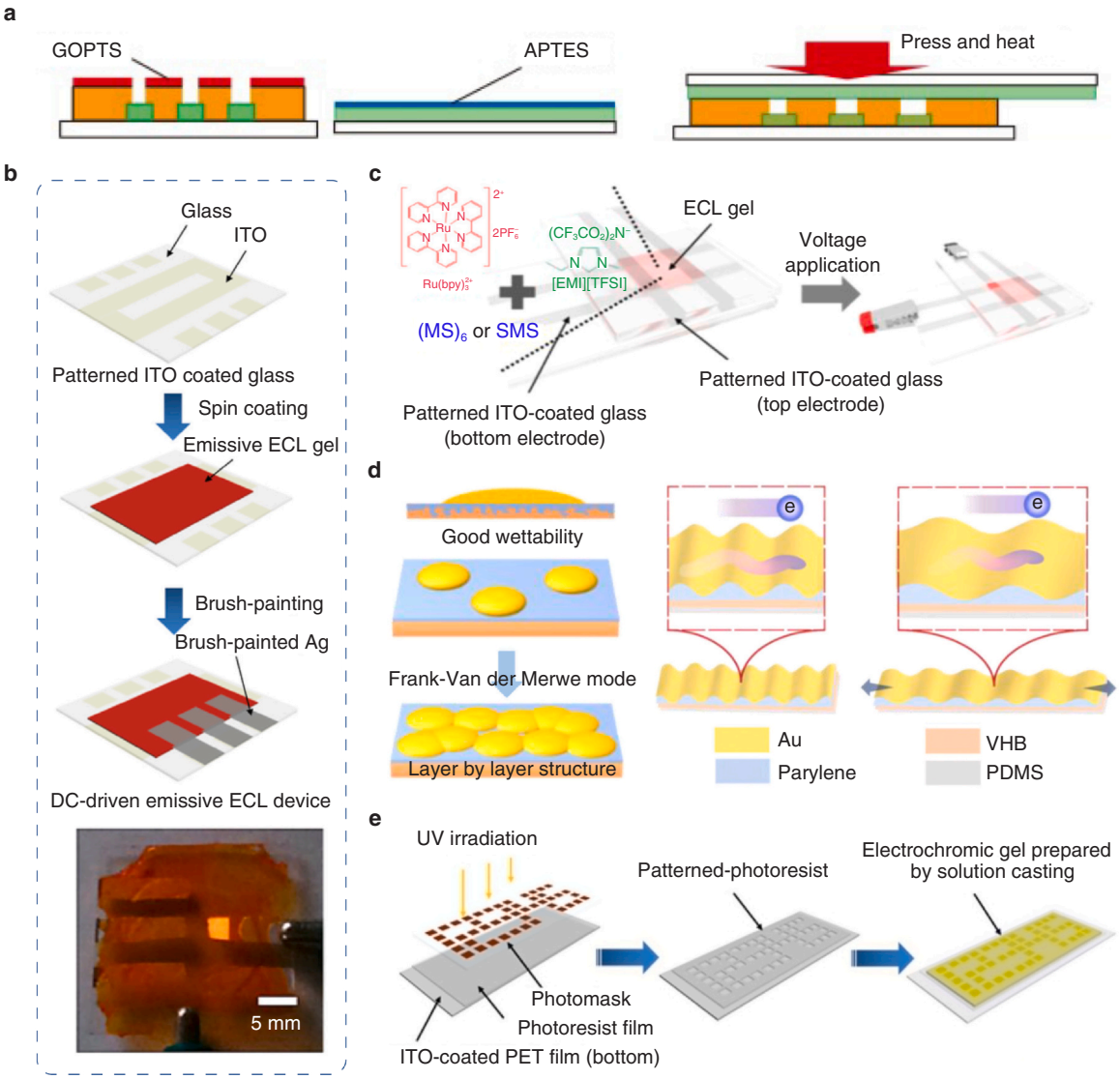

Fig. 4. Preparation technology of ECLD electrodes. (a) Patterned electrodes by etching and thermal bonding[101]. Copyright 2014, Elsevier. (b) Spin-coated and brush-painted ECL gel on patterned ITO/Ag electrodes[26]. Copyright 2014, American Chemical Society. (c) Drop coated ECL gel on patterned ITO electrodes[103]. Copyright 2019, American Chemical Society. (d) Pre-stretch-stabilized polymer/gold hybrid electrodes with parylene evaporation[25]. Copyright 2024, Wiley-VCH. (e) Flexible electrodes with photoetching and solution casting on plastic[27]. Copyright 2015, American Chemical Society.

atoms form a solid film by diffusion and condensation. Regarding the deposition of thin layers of gold, this is achieved through two stages: nucleation and nucleation-centered growth. Gold has a low surface resistance due to its high free electron density compared to conductive composites. In the initial stage of depositing thin films, many atoms and molecules coalesce and are deposited on the substrate. For the distribution of atoms, the effect depends on the forces of the metal atoms with neighboring atoms and with the substrate. Depending on the interactions, three modes of nucleation and growth of metal films exist [104,105]. By analyzing the principles of metal thin film deposition and growth, the nucleation of metal atoms usually follows the Volmer-Weber mechanism due to the surface energy of the substrate, and vacuum-deposited metals often nucleate randomly on the substrate surface. Various surface modification schemes, including doping, introduction of a wetting layer and use of surfactants, have been fruitful in suppressing the Volmer-Weber mode and lowering the permeation threshold. Metal island growth leads to poor electrode conductivity and transparency, which is usually attributed to weak adhesion of metal atoms to the substrate. To enhance adhesion, Volmer-Weber growth can be inhibited by pre-depositing an intermediate layer, such as a dielectric material or a metal with a high adhesion energy, on the substrate prior to depositing the metal. According to the thin film growth principle, this is expected to provide dense nucleation sites and suppress the appearance of discontinuous islands during deposition. In addition, the emergence of emerging technologies such as electrospinning, inkjet printing, and electro spraying has led to iterative upgrading of the preparation process and substantial improvement in mechanical and electrical properties[4,104]. It is not only applied to the preparation of electrodes, but also widely used in the overall design of ECLD.

He et al.[106] prepared strain-insensitive stretchable electrodes based on graphene-elastomer composites by the “pre-stretch-release method”. Using the self-assembly property of graphene nanosheets on water, the preparation of graphene thin films was realized, and the precise patterning of graphene was achieved by combining photolithography and masking techniques. The tensile properties of the material were optimized by tuning the pleated structure of the PDMS substrate. The pleated structure was able to accommodate external mechanical stresses in the normal state and disperse the stresses by unfolding the structure during stretching, thus maintaining the electrical conductivity of graphene. Subsequently, the group[25] utilized

this method to prepare pleated gold-polymer conformal composite electrodes (Fig. 4d). Aiming at the easy oxidization and brittleness of the electrodes, gold was chosen as the conductive material. The interfacial interaction between gold and polymer was enhanced by CVD of parylene interlayer to match the interlayer modulus, and the growth mode was changed to solve the problem of ultrathin continuous and stable growth of gold on the polymer surface, and thin films with excellent electrical conductivity (surface resistance < 10 Ω/sq) and transparency (light transmittance > 60 %) were prepared. Through the "pre-stretch-release method", the surface morphology of gold is regulated, and a pleated polymer-gold conformal composite structure is constructed, which converts the ductile strain into flexural strain at the microscopic scale, effectively disperses the mechanical strain, and forms a conductive network. The macroscale high tensile performance (> 50 %) and excellent mechanical robustness (withstanding more than 10,000 cycles of 40 % tensile cycling with only 6 % increase in relative resistance) of the gold film are realized. The electrodes are combined with rough porous structure of TPU fibers for ECLD, and the layers can be assembled into a device by their own adhesive properties without additional spacer layers, and no delamination occurs after stretching or bending. More importantly, the brightness of the ECLD remains stable.

In the cutting-edge research exploring electrical properties of stretchable materials, from material selection to structural design, lays a solid foundation for high-performance. To transform these high-performance materials into high-efficiency electronic devices for applications, electrode patterning process is an indispensable link[107]. Electrode patterning, as one of the core preparation processes of ECLDs, directly affects the sensitivity, spatial resolution and integration of the device. For example, photolithography with etching and stripping processes are suitable for high-precision patterning, screen printing and inkjet printing are suitable for the preparation of large-area and thick-film electrodes, and laser direct-writing and 3D printing provide higher flexibility and the ability to prepare complex structures.

Photolithography is a micro-nanofabrication method based on the selective exposure and development of photosensitive materials (photoresist), which is used to selectively expose the photoresist through a photomask, transfer specific patterns to the substrate surface, and combine with the etching process to realize the electrode microstructure processing. In ECLD, photolithography is mainly used to prepare high-precision electrode arrays, microchannel structures, and surface functionalized regions. Moon et al.[27] used photolithography to define electrode patterns on an ITO/PET substrate, followed by solution casting to fill the ECL gel into the patterned regions, and successfully integrated patterned ECL units on a plastic substrate (Fig. 4e). The combination of photolithography and solution processing achieves high-resolution, flexible-compatible electrode patterning and significantly improves the spatial resolution and sensitivity of the ECL signal.

3.3. Patterned technology for ECLDs

Electrode patterning not only requires that the conductive material can be precisely deposited on the substrate to form a specific pattern, but also involves how to optimize the patterning process to further enhance the overall performance and stability of the device. After the electrodes are prepared by patterning, various processes are successively introduced to obtain complete ECLDs[32,56,101]. ECL solutions (or even precursors of ECL gels) can be applied to various solution processes, including patterning methods. As shown in Fig. 5a, Itoh[56] demonstrated the process of mask printing ECLDs using gel light-emitting layers. The ECL gel was printed on a fabric by means of a metal mask and a rubber squeegee and clearly displayed high-contrast characters when a voltage of 4.0 V at 60 Hz was applied. The process is simple, easy, and low-cost, and is suitable for the preparation of large areas and complex patterns.

Adopting a more convenient and faster printing technique, Lee et al. [108] combined an ion gel capacitor with the triboelectric nanogenerator (TENG) to achieve effective modulation of the TENG output voltage (Fig. 5b). It was able to modulate the spike-like voltage into a square wave and adjust the output voltage to a suitable range. Self-powered light emission was realized by directly driving the ECLD with the power generated by the TENG. By adjusting the parameters of the ion gel capacitor (e.g., area, connection method), the luminous intensity and voltage range of the ECLD can be precisely controlled. By patterning and customizing 20 ECL units and connecting multiple ECL light-emitting points in series, the output voltage is significantly increased, resulting in enhanced light-emitting intensity; whereas parallel connection reduces the output voltage but enables a more uniform voltage distribution, which is suitable for uniform light-emitting over large areas. This integration not only solves the problem of high and spike-like output voltage of TENG, but also realizes low-cost, high-efficiency preparation and flexible patterning of ion-gel capacitors by printing technology. Several printing methods are also described more specifically below.

Inkjet/3D printing technologies are widely used for the preparation of stretchable electrodes and flexible devices due to their advantages of high automation, precise positioning, fast preparation and high resolution. The inks are printable non-Newtonian fluids containing organic solvents and additives, which are additively manufactured by direct ink writing (DIW). In the DIW pneumatic extrusion process, the non-Newtonian fluid is extruded from a micro-nozzle by shear thinning and deposited as a customized pattern under the motion of the substrate [109,110].

Conventional electrode preparation methods usually print conductive materials on the surface of elastomeric substrates or embed them inside. However, surface-printed electrodes are prone to failure due to interfacial weakening during stretching, while fully embedded electrodes are limited by their conductive properties, making it difficult to use them as electrodes directly. As analyzed in the introduction section of this chapter, the structure of conductive materials surface-embedded in elastomers can help to solve the problems. Song et al.[87] utilized the DIW technique to print a precursor solution on an elastomer substrate and converted it into metal nanoparticles by chemical reduction (Fig. 5c). These nanoparticles formed effective connections inside and on the surface of the substrate, thus realizing a "surface-embedded" electrode structure. This ensures strong adhesion between the electrodes and the substrate and allows the electrodes to withstand large mechanical deformations while maintaining electrical conductivity. The study utilizes the viscoelastic properties of the composite material at high temperatures, and the electrode film can be spontaneously adhered to the surface of the 3D structure through thermal annealing treatment.

Kim et al.[29] proposed a fully 3D-printed flexible ECLD preparation process by DIW printing of ECL luminescent materials and electrode materials of various complex shapes (e.g., comb and helix). In this work, the ECLs belonged to the annihilation-type mechanism in order to take advantage of the long-term stability of the emission on the two electrodes (Fig. 5d). They demonstrated ECL based on a fully printed process by directly 3D printing the luminophore solution as an ink. To improve the poor printability and structural integrity of the ECL luminophore ink, SiO_2 nanoparticles were doped to improve the rheological properties. The ink exhibits a solid-like behavior in the normal state and a liquid-like behavior under sufficiently high shear stress, which is conducive to maintaining the shape integrity during the printing process. Meanwhile, in order to enhance the electrochemical stability and conductivity, graphene is introduced onto the Ag electrode to ensure the stability of the ECLD in the electrochemical reaction and the efficient transmission of the electric current, thus realizing the uniform and long-time stable luminescence of the whole device. By applying printable ECL ink, patterns with high structural integrity were designed to realize electrically driven illumination based on the full printing process on flexible substrates.

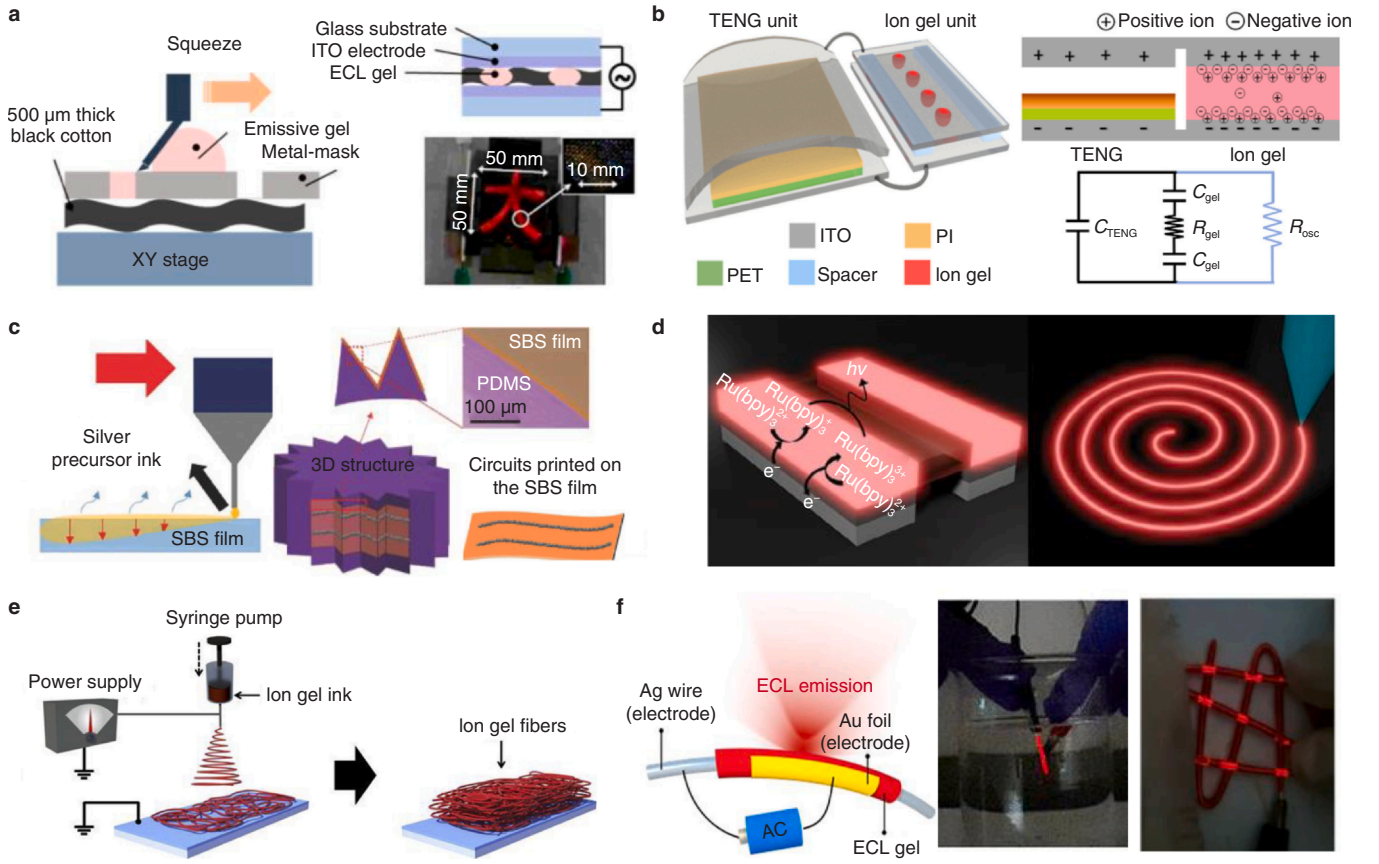

Fig. 5. Preparation technology of ECLDs. (a) Metal-mask-printed ECLDs[49,56]. Copyright 2018, American Chemical Society. (b) ECL ion gel unit for TENG[108]. Copyright 2018, Elsevier. (c) Direct-printed surf-embedded structures for stretchable ECLDs[87]. Copyright 2017, Wiley-VCH. (d) All-printed ECLDs via DIW[29]. Copyright 2024, Wiley-VCH. (e) ECLDs based on electrospinning microfibers[28]. Copyright 2018, Elsevier. (f) Core-shell fibers for washable ECLDs[111,112]. Copyright 2022, American Chemical Society and Copyright 2023, American Chemical Society.

Overall, the rheological and electrical properties of the modulated ink particles can be tuned to combine with the dispensing process to realize the possibility of high precision patterning, large scale printing, and printing on a variety of substrates. The precursor inks of the materials are compatible with a variety of preparation processes such as spin coating, inkjet printing, dispensing and brush painting, further illustrating the potential of the emerging technology in complex stretchable systems. On the other hand, there are shortcomings in the printing technology, such as inks requiring special handling, difficulty in obtaining uniform pixels, and suboptimal stability and consistency in large-scale production, which also require technology iteration and upgrading.

In recent years, wearable e-textiles have been developing rapidly in the field of flexible electronics[113,114]. As a basic component of e-textiles, spun fibers have received much attention due to their flexibility, stretchability, and scalability[115]. Among the many preparation techniques, electrospinning is a classical spinning process based on the principles of electrohydrodynamics, which is modified by physicochemical means to fabricate micro- and nanomaterials[116]. The technique involves the application of high voltages (> 10 kV) to a polymer solution to create a charge imbalance and form micro-nanofibers with high specific surface area and tunable morphology and composition. Electrospinning in the laboratory consists of the following processes: pressurization and deformation, jet formation, jet thinning, curing and collection of fibers. The electrospinning equipment consists of a syringe with a metal needle, a controller to adjust the solution flow rate and spinning duration, a drum receiver to receive the electrospinning fibers, and a power supply to generate an electric field between the jet and the receiver. The polymer is dissolved in a solvent and delivered to the metal needle through the syringe. When the high voltage is turned on, a conical solution (i.e., a Taylor cone) forms at the tip of the metal needle. As the solution continues to be pushed, an electrically charged jet is ejected from the Taylor cone, and the diameter of the jet becomes progressively finer as it approaches the collector, accompanied by the evaporation of the organic solvent, and is ultimately deposited as solid fibers in the receiver for the preparation of smart textile products.

Hong et al.[28] prepared ionogels using poly(vinylidene fluoride-hexafluoropropylene) (P(VDF-HFP)) as a polymer mixed with [EMIM][TFSI]. The ionic gels were prepared by electrospinning into porous reticulated microfibers with diameters ranging from 1.4 to 2.3 μm and thicknesses of about 50 μm. ECL luminophores imparted luminescent properties to the microfibers, and the fiber devices emitted an orange-red light with a maximum luminance of 50 cd/m^2 at a low operating voltage (< 2.5 V), and demonstrated mechanical flexibility (Fig. 5e). On this basis, the group[111] prepared ECL fibers by a simple one-step dip-coating method at room temperature, and the fibers consisted of inner electrodes (silver wires), ECL gel, and outer electrodes (gold foils), forming a cylindrical and stable core-shell structure (Fig. 5f). The ECL fibers could be easily bent, deformed, and hand-knitted with commercial knitwear, and retained luminescent properties after multiple washes. It provides a new idea for the development of wearable EL fabrics. Further, they proposed a fiber module assembly method similar to Legos[112]. Multiple luminescent fibers can achieve point emission (perpendicular contact) and line emission (parallel contact) by changing the contact mode. Thus, it is possible to assemble two or more fiber modules into luminescent pixels by simple physical contact without external pressure or heating. Pan's group[25] prepared fully stretchable ECLDs based on electrospinning fiber luminescent layers impregnated with luminescent and ILs. The luminescent solution consisted of red

Table 1
Summary of materials, structures, flexibility and stretchability of ECLDs.

Electrode materials	Flexibility/ Stretchability of electrodes	Materials of light-emitting layer	Form of light-emitting layer	Ionic liquid/ Electrolyte/ Solvent	Main preparation technology	Flexibility/ Stretchability of ECLDs	Applied voltage	Ref.
ITO	Brittle	$Ru(d_8\text{-}bpy_3)(PF_6)_2$	Liquid/Gel	[ABIm][TFSI]	Drop casting/ Etching/ Photolithography	Brittle	AC	[56]
ITO	Brittle/ Flexible	$Ru(bpy)_3Cl_2$/ DPA/RUB	Liquid	[EMIM][TFSI]	Etching/ Drop casting	Brittle/ Flexible	AC	[102]
ITO	Brittle	$Ru(bpy)_3(PF_6)_2$	Liquid	TBAP/PC	Drop casting	Brittle	DC/AC	[121]
ITO	Brittle	$Ru(bpy)_3(PF_6)_2$	Liquid	TBAP/PC	Drop casting	Brittle	DC/AC	[55]
ITO	Brittle	DPA/RUB	Liquid	TBAP/NMP	Drop casting	Brittle	AC	[62]
ITO	Brittle	$Ru(bpy)_3Cl_2$	Liquid	TBAP/PC/ DMSO/THF	Spin coating/ Drop casting	Brittle	DC/AC	[58]
ITO	Brittle	$Ru(bpy)_3Cl_2$	Gel	[EMIM][TFSI]	Spin coating/ Brush painting/ Photolithography	Brittle	DC	[26]
ITO	Flexible	$Ru(bpy)_3Cl_2$/ Ir $(diFppy)_2(bpy)PF_6$	Gel	[EMIM][TFSI]	Spin coating/ Drop casting/ Brush painting/ Photolithography	Flexible	AC	[32]
ITO	Brittle	PLQ/DPT/ RUB/DBP	Liquid	[TMP][TFSI]	Spin coating/ Template/ Wet etching/ Photolithography	Brittle	AC	[101]
ITO	Flexible	$Ru(bpy)_3Cl_2$	Gel	[EMIM][TFSI]	Spin coating/ Drop casting	Flexible	AC	[122]
ITO/ Ag NWs/ Au	Flexible/ Stretchable/ Stretchable	$Ru(bpy)_3Cl_2$	Gel	[EMIM][TFSI]	Spin coating/ Brush painting/ Blade coating	Flexible	AC	[123]
ITO	Brittle	$Ru(bpy)_3(PF_6)_2$/ $Ir(diFppy)_2(bpy)PF_6$	Gel	[EMIM][TFSI]	Brush painting/ Blade coating	Brittle	AC	[65]
ITO/Ag	Flexible/ Stretchable	$Ru(bpy)_3(PF_6)_2$	Gel	[EMIM][TFSI]	Spin coating/ Dot printing/ Drop casting	Partially stretchable	AC	[87]
ITO/Au	Brittle/ Stretchable	$Ru(bpy)_3(PF_6)_2$	Gel	[EMIM][TFSI]	Drop casting	Brittle	DC/AC	[49]
Au	Stretchable	$Ru(bpy)_3(PF_6)_2$	Gel	[EMIM][TFSI]	Automatic coating/ Dot printing	Fully stretchable	AC	[30]
ITO	Flexible	I-DPA	Viscoelastic solid	TBAP/NMP	Drop casting	Flexible	AC	[124]
ITO	Brittle	$Ru(bpy)_3(PF_6)_2$/ $Ir(diFppy)_2(bpy)PF_6$/DPA	Viscoelastic solid	[EMIM][TFSI]	Dot printing	Brittle	DC/AC	[125]
ITO	Flexible	$Ru(bpy)_3(PF_6)_2$	Gel	[EMIM][TFSI]	Spin coating/ Drop casting/ Template	Flexible	AC	[108]
ITO/Ag	Brittle/ Stretchable	$Ru(bpy)_3Cl_2$	Electrospinning fibers	[EMIM][TFSI]	Electrospinning/ Evaporation	Brittle	AC	[28]
ITO	Brittle	$Ru(bpy)_3(PF_6)_2$	Liquid	[BMIM][TFSI]	Drop casting	Brittle	AC	[126]
ITO	Flexible	$Ru(bpy)_3(PF_6)_2$	Gel	[EMIM][TFSI]	Drop casting	Flexible	AC	[103]
ITO	Brittle	$Ru(bpy)_3(PF_6)_2$/ $Ir(diFppy)_2(bpy)PF_6$	Viscoelastic solid	[EMIM][TFSI]	Drop casting	Brittle	AC	[127]
ITO	Flexible	$Ru(bpy)_3(PF_6)_2$/ $Ir(Fppy)_2(dmb)PF_6$/ $Ir(Fppy)_2(Mepic)$	Viscoelastic solid	[EMIM][TFSI]	Spin coating/ Template	Flexible	AC	[31]
ITO/ CNT	Flexible/ Stretchable	$Ru(bpy)_3(PF_6)_2$/ Ir $(Fppy)_2(dmb)PF_6$/ DPA	Viscoelastic solid	[EMIM][TFSI]	Spin coating/ Template	Partially stretchable	AC	[128]
ITO	Flexible	$Ru(bpy)_3(PF_6)_2$	Gel	[EMIM][TFSI]	Drop casting/ Template	Flexible	AC	[129]

(*continued on next page*)

Table 1 (*continued*)

Electrode materials	Flexibility/ Stretchability of electrodes	Materials of light-emitting layer	Form of light-emitting layer	Ionic liquid/ Electrolyte/ Solvent	Main preparation technology	Flexibility/ Stretchability of ECLDs	Applied voltage	Ref.
ITO/ZnO	Brittle	$Ru(bpy)_3(PF_6)_2$/ $Ir(Fppy)_2(dmb)PF_6$/ $Ir(Fppy)_2$(Mepic)	Viscoelastic solid	[EMIM][TFSI]	Spin coating/ Template	Brittle	AC	[130]
ITO/Au	Brittle/ Stretchable	$Ru(bpy)_3(PF_6)_2$	Visco-poroelastic solid	[EMIM][TFSI]	Spin coating	Partially stretchable	AC	[131]
PEDOT: PSS/Ag/ Au	Flexible	$Ru(bpy)_3Cl_2$	Gel	[EMIM][TFSI]	Brush painting/ Dip coating	Flexible	AC	[111]
Au	Stretchable	$Ru(bpy)_3(PF_6)_2$/ $Ir(diFppy)_2(bpy)PF_6$	Electrospinning fibers	[EMIM][TFSI]	Magnetron sputtering/ Electrospinning/ Evaporation	Fully stretchable	AC	[25]
ITO/Ag/ Graphene	Brittle/ Stretchable/ Stretchable	$Ru(bpy)_3Cl_2$	Ink	[EMIM][TFSI]/ PVAc	3D printing	Flexible	AC	[29]

luminescent $[Ru(bpy)_3](PF_6)_2$ dissolved in acetone, green luminescent $[Ir(diFppy)_2(bpy)]PF_6$, and [EMIM][TFSI] induced to produce ECL. Further, electrolyte gel-based ECLDs were prepared by spin coating the same formulated solutions as a comparison, and it was found that the luminophores were more uniformly distributed in the spinning layer, and the electrospinning ECLDs were far superior to the electrolyte gel-based ECLDs in terms of their optoelectronic and mechanical properties. the liquid luminescent layer was fluid, and the morphology was prone to leakage during stretching. The electrolyte gel may have uneven and uncontrolled thickness: the thin gel layer is easy to break and not easy to peel off. And the thick gel layer is difficult to firmly adhere to the electrode layer, which affects the conformal contact between the two. The porous structure of the TPU film provides the "backbone" support, and the highly stretchable fibers promote the uniform penetration of luminescent solution.

In summary, various design strategies and preparation processes for devices have their own scope of applications. In actual use, it is necessary to select the appropriate preparation method according to specific material properties, substrate requirements, device structure and performance requirements. It is worth noting that the above preparation methods are generally not used independently, and need to be used in combination with several of them or other processes, which is conducive to the realization of better performance[117,118].

4. Flexible/stretchable ECLDs for display applications

Advances in display technology have not only enhanced the visual experience, but also promoted the thinness, portability and high performance of electronic devices. The rise of flexible electronics has brought new changes to display technology. The new generation of light-emitting display devices should be stretchable, flexible, and bright. As a component of them, high demands are placed on both the electrode layers and the light-emitting functional layers[104,119,120]. The percolation threshold determines the formation of interconnecting conductive networks, and the deposition of ultrathin metals contributes to the formation of efficient carrier injection[105], whereas the electrode thickness capable of reaching the percolation threshold creates a contradiction with high transparency[38]. There is also a need to optimize the optoelectronic properties of the luminescent functional layers, as well as the modulus matching between layers, so as to prevent delamination. With the development of flexible electronics technology, flexible/stretchable ECLDs have gradually become a research hotspot. Flexible/stretchable ECLDs are not only able to achieve stable luminescence under complex deformations such as bending, folding, and even stretching, but also have the characteristics of thin, light, transparent, and low power consumption, which are promising for the application in wearable light-emitting devices, optical fibers, and represent the expansion of electrochemical- and optics-based wearable sensing devices. Table 1 summarizes the electrode materials and stretchability, light-emitting layer materials and morphology, ILs, electrolytes, and solvents used in ECLDs in chronological order, as well as the device preparation process, device stretchability, and applied operating voltage, which to a certain extent can reflect the development trend in recent years.

4.1. Multi-color displays

By choosing different luminescent materials, flexible/stretchable ECLDs can achieve full-spectrum luminescence from the UV to the IR, which opens up a wide range of applications in fields such as multi-color displays and imaging. Kasahara et al.[101] demonstrated multi-color microfluidic OLEDs based on pyrene-based liquid organic semiconductors. They utilized a microfluidic structure to achieve multi-color ECLDs by introducing $Ru(bpy)_3Cl_2$ (red), RUB (yellow), and DPA (blue) luminescent solutions to achieve multi-color ECLDs in the channel. The device can constantly replenish the luminophores to make

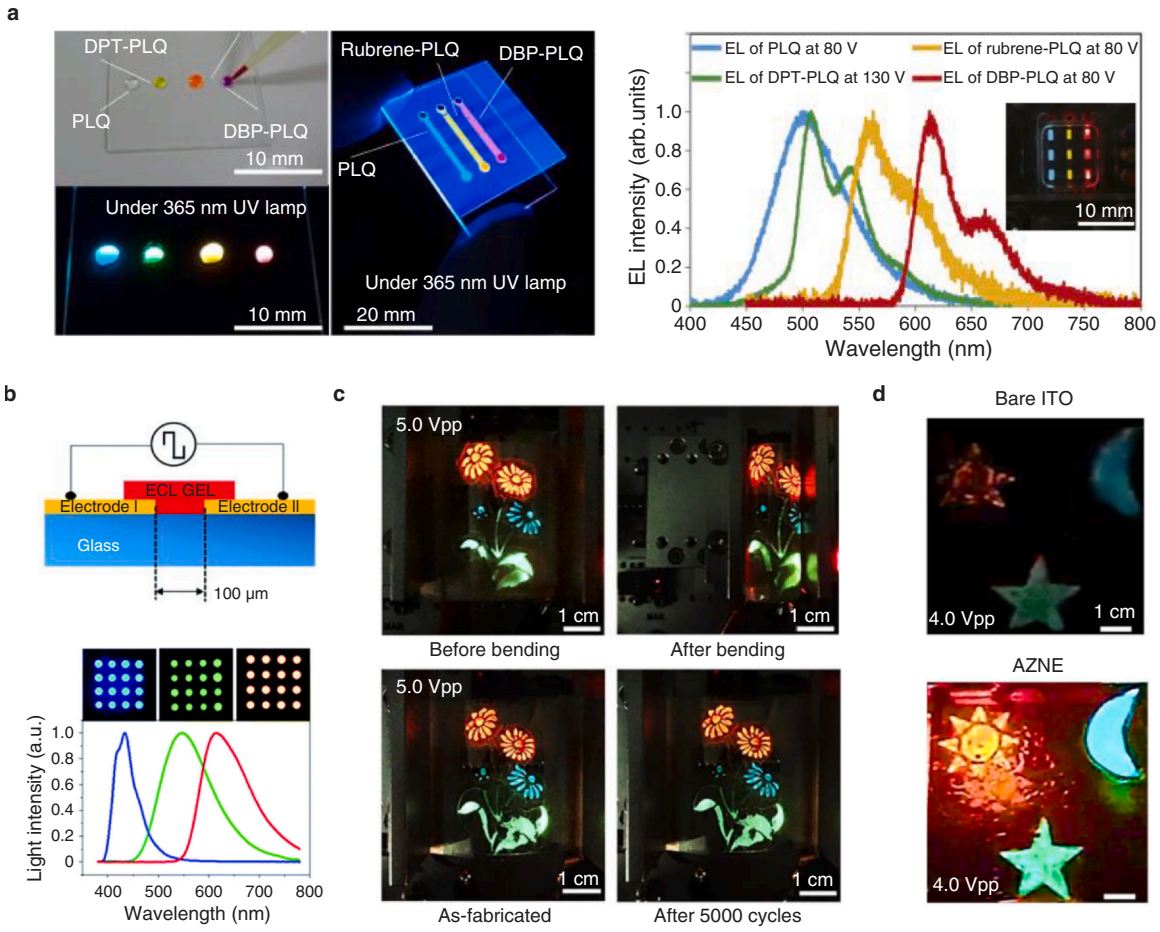

Fig. 6. ECLDs for multi-color displays. (a) Pyrene-based liquid organic semiconductors for multi-color microfluidic ECLDs[101]. Copyright 2014, Elsevier. (b) Viscoelastic solid displays depend on effect of ion migration[125]. Copyright 2018, The Royal Society of Chemistry. (c) Multi-color and long-time stable bending displays of ion gel-based flexible ECLDs[31]. Copyright 2020, Elsevier. (d) Enhanced light-emission efficiency of multi-color displays[130]. Copyright 2021, Elsevier.

up for their loss in the cyclic redox reaction. In addition, multiple ECL luminophores can be precisely mixed by the microfluidic structure, which enables tunable luminescence of a series of ECL solutions (Fig. 6a). On top of the luminescent solutions, the emergence of gel-based devices presents new opportunities. Free radical ions in gel-based devices significantly affect the luminescence behavior of ECLs, which is closely related to factors such as ion species, operating frequency and voltage. Shin et al.[125] determined the optimal operating conditions for $Ru(bpy)_3(PF_6)_2$, $Ir(diFppy)_2(bpy)PF_6$ and DPA luminophores by studying the migration behavior of different types of luminophores (e.g. voltage and frequency) were tuned to realize more efficient and stable multi-pixel ECL display arrays with solid gel-based ECLDs (Fig. 6b).

According to the literature summary, the luminescence of blue light displays is often unsatisfactory. An important reason is that the blue light-emitting materials in such studies generally use aromatic organic semiconductor compounds, which are fluorescent materials, and the emission efficiency itself is relatively low compared with that of red and green light-emitting materials (metal chelates of phosphorescent materials). In order to enhance the blue light-emitting effect, a higher operating voltage is required, which is contrary to the starting point of the requirement that the device works stably and efficiently at low AC voltage. Therefore, to realize red, green, and blue and even full-color displays simultaneously, it is necessary to solve the problem of blue light emission and improve its luminous efficiency at the same time. In 2020, Myoung's group[31] solved the problem of previous ECL gel-based displays that had difficulty in realizing the RGB three-color luminescence at the same time. They designed a mixed-metal chelate system to achieve a significantly improved sky-blue display by mixing green and blue emitters and utilizing the interaction between oxidized and reduced state emitters, demonstrating a floral display pattern based on a gel ECL layer (Fig. 6c), which remained stable after more than 5000 stretching and bending cycles and still maintained luminescence performance. However, the luminescence efficiency of this gel-based ECLD remains low. For this reason, in 2021, they used Au nanoparticles as catalysts to significantly promote the redox reaction[130], and the 3D-structured ZnO nanorods greatly increased the surface area of the electrodes, enhanced the catalytic activity of Au, and also improved the electron transport pathway, enabling more luminophores to participate in the reaction, thus effectively enhancing the luminescence efficiency of the multi-color ECLD (Fig. 6d).

4.2. Stretchable and flexible displays

Flexible and stretchable ECLDs are able to maintain consistent luminescent properties under high strains. This high stretchability makes them suitable for dynamic environments such as motion monitoring devices or implantable medical devices; able to adapt to complex mechanical deformations such as bending, folding and twisting without compromising their performance. In the initial period, the liquid state dominated the luminophores for such light-emitting devices. Okumura et al.[102] developed a flexible device based on an electrochemical luminescent solution consisting of an electrostatic ruthenium complex as the luminescent material and an ionic liquid as the nonvolatile solvent (Fig. 7a). Specifically, the solution was patterned and sealed by

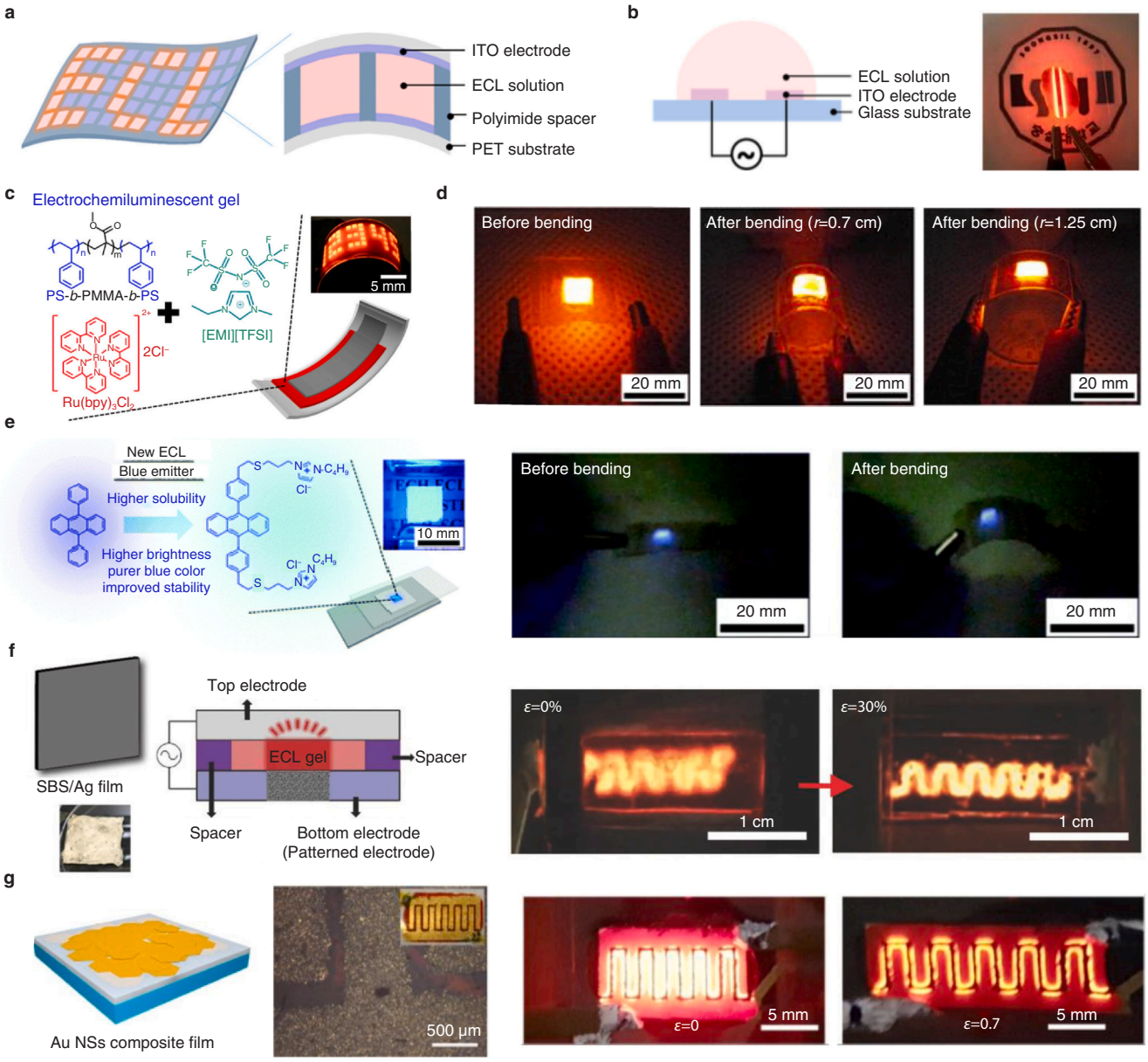

Fig. 7. Flexible/stretchable ECLDs. (a) Microfluidic injection solution and package of ECLDs adapted to flexible substrate[49,102]. Copyright 2018, American Chemical Society. (b) Solution-based ECLDs in an unconventional parallel-electrode structure[49,121]. Copyright 2018, American Chemical Society. (c) Solution-processable ECLDs for multi-pixel patterns[32]. Copyright 2014, American Chemical Society. (d) Gel-based ECLDs with full-surface luminescence subjected to bending deformation[103]. Copyright 2019, American Chemical Society. (e) Flexible ECLDs with blue color[124]. Copyright 2017, Royal Society of Chemistry. (f) Partially stretchable ECLDs[87]. Copyright 2017, Wiley-VCH. (g) Fully stretchable ECLDs[30]. Copyright 2017, American Chemical Society.

microfluidically injecting and sealing the solution between two PET films. The liquid luminescent material is able to withstand large bending deformations as it is free to change its shape and does not degrade by bending. In Fig. 7b, the electrolytic nature of the active layer of the ECL allows researchers to fabricate ECLDs with parallel electrode structures instead of the traditional “sandwich” structures [49,121]. The parallel electrode structure allows free radicals to be generated near the electrode surface and react quickly, reducing the diffusion loss of free radicals in the solution. This structure not only improves the luminescence efficiency, but also reduces the ineffective current consumption and improves the current efficiency. Fig. 7c demonstrates the multi-pixel image display capability of ECLD prepared on a plastic substrate after bending[103]. The ECLDs were prepared by solution casting ECL gels and brush painting Ag electrodes, realizing a simple two-step solution handling process. In addition, patterned ECLDs were prepared by photolithography, which enables precise control of the shape and size of the light-emitting region. The fabricated devices emit light only in the patterned area, so that they can be viewed as macroscopic digital patterns consisting of small square pixel dots of 1 mm × 1 mm, and maintain stable light emission even after bending. Similarly, the form of the luminous body is liquid. The above devices may pose a problem of chemical degradation or changes in physical properties over time during operation, especially during repeated mechanical deformation and electrochemical reactions, and lack of long-term stability. In addition, the liquid material may undergo uneven distribution due to gravity, capillary action, or other physical effects, resulting in inconsistent brightness and color in the light emitting region.

The above problems were significantly improved when solution-based devices were replaced with gel-based devices. Hwang et al.

prepared flexible ECLDs using six-armed star-shaped block copolymers [32], which enabled the separation of the entire polymer chain from the network structure to be minimized under mechanical deformation, even if one arm was pulled out, and the other five were connected through the core (Fig. 7d). Compared with conventional linear block copolymers (e.g., SMS), it has higher mechanical strength and thermal stability. The ionogel has more than two times higher modulus of elasticity and remains in a stable gel state at temperatures up to 180 °C without gel-solution transition. Unlike Fig. 7c, this work is unpatterned, the luminescence of the entire contact surface is achieved by simple bonding of the electrode layer to the luminescent layer. When the bending radii were 1.25 cm and 0.7 cm, corresponding to strains of 0.5 % and 0.9 %, respectively, the latter was able to maintain 72 % of its original luminance after 3000 bending cycles, while the device before gel modification maintained only about 47 % of its initial luminescence intensity at the same strain. While flexible ECLDs are usually dominated by red or green emitting materials, blue emitting materials are an essential component in full-color displays. The above research group achieved blue luminescence through I-DPA and outperformed conventional DPA materials in terms of color purity and brightness[124], which is an important impetus to realize diversification and high performance of flexible displays (Fig. 7e).

The core of flexible ECLDs lies in the use of flexible substrates (e.g., plastics, thin films) and flexible electrode materials (e.g., conductive polymers, metal nanowires), which allow the device to adapt to various shape changes without destroying the internal structure. Stretchable ECLDs are further required to maintain functionality under large strains. Such devices require not only flexible substrates, but also special structural designs (e.g., wave-pleat structures, serpentine interconnections) or intrinsically stretchable materials (e.g., conductive networks in elastomers) to achieve electrical and luminescence stability under large deformations[132–134]. Song et al.[87] prepared stretchable ECL displays by directly printed surface-embedded stretchable electrodes (Fig. 7f). Serpentine electrodes were designed to disperse the strain and thus maintain the electrical performance under large strains. As the bottom electrode of the stretchable ECLD, the device was realized to have no brightness degradation or even slight enhancement at a stretch rate of 30 %.

However, brittle ITO is still used for the top electrode in order to ensure the light transmittance, so the device here is only partially stretchable for the bottom electrode and the light-emitting layer, and the overall full stretchability of the device has not yet been realized, and finding a suitable alternative material for the ITO electrode can help to solve the above problem. Replacing ITO with ductile gold as the conductive electrode by sacrificing part of the light transmittance is a feasible solution[135]. Cho et al.[30] prepared fully stretchable ECLDs using Au-polystyrene-block-polybutadiene-block-polystyrene-PDMS fully elastic conductive films that achieved more complex display functions and greater elongation by comb electrode design (Fig. 7g). Compared with rigid ECLDs, flexible and stretchable ECLDs have significant advantages in terms of mechanical properties, application flexibility, performance, manufacturing cost and environmental adaptability.

4.3. Pressure-sensitive and wearable displays

The above advances indicate that many excellent characteristics of ECLDs are increasingly recognized compared with traditional light-emitting devices, and they have become a strong contender for the new generation of stretchable wearable devices[136–139]. Among them, pressure-sensitive displays and wearable displays show potential as cutting-edge application directions of ECLDs.

Pressure-sensitive displays enable dynamic responsive interaction by converting mechanical stimuli (e.g., pressure, stretching) into light signal changes. Its core lies in the conversion of mechanical pressure signals into modulation of luminous intensity. Its working mechanism can be categorized into interface contact modulation type and ion transport modulation type[140]. The former modulates the charge injection efficiency by changing the contact area between the electrode and the luminescent layer through pressure. For example, the contact area of microstructured electrodes (e.g., pyramidal arrays) increases when they are pressurized, the current density rises, and the luminescence brightness is enhanced. The latter utilizes pressure to change the mobility or distribution of the electrolyte, affecting the rate of electrochemical reactions. For example, the porosity of porous electrolyte layers decreases under pressure, shortening the ion migration path and accelerating the reaction kinetics, and Lee et al.[131] utilized the so-called “piezoelectric ion effect” to enhance luminescence intensity by inducing changes in the ion distribution through mechanical stress. This effect allows the material to significantly enhance the ECL signal when subjected to stress, resulting in highly sensitive detection of stress (Fig. 8a). Significant enhancement of luminescence intensity was exhibited when subjected to both positive and tensile stresses. For example, the luminescence intensity increased about 8-fold when the positive stress was increased from 0 to 60 kPa, and the luminescence intensity increased 1.5-fold when the tensile strain was increased from 0 % to 100 %. This stress-dependent luminescence property allows the ECLD to directly convert mechanical stimuli into visual signals for spatial discrimination of locally applied stresses without the need for complex pressure sensor arrays.

Wearable displays, on the other hand, rely on the thin, lightweight, and stretchable properties of ECLDs to promote the seamless integration of display devices with the human body. Kwon et al.[128] reported a wearable pressure-sensitive tactile sensor (PLS), in which they embedded CNTs in PDMS to prepare semi-transparent electrodes, and used ECL gel as the luminescent layer to fabricate a structurally simple sensor device, which can immediately reflect external stimuli (Fig. 8b). Despite the low operating voltage, the PLS was able to achieve a high luminance output. For example, the red, green, and blue-emitting PLSs have a maximum luminance of 68.2 cd/m^2, 84.7 cd/m^2, and 35.4 cd/m^2, respectively, at 6.5 V_{PP}, which allows for the clear display of information even in darker environments. As a wearable device, it is capable of displaying dynamic information of continuous external stimuli in real time, such as the motion trajectory of an object or pressure changes. Chen et al.[25] prepared strain-insensitive wearable ECLDs (Fig. 8c) by researching the electrode design, electrochemical luminescent layer, device structure, and luminescence performance. Specifically, the pleated polymer-gold conformal composite structure was able to convert ductile strain into flexural strain at the microscopic scale, realizing high tensile performance and excellent mechanical robustness of the electrode at the macroscopic scale. The luminescent layer is realized by electrospinning, and the luminescent layer has a rough and porous structure, which is conducive to the uniform infiltration and penetration of the luminescent solution, and its optoelectronic and mechanical properties are superior to those of the conventional electrolyte gels. In addition, the interface between the luminescent layer and the electrodes maintains close contact without delamination during stretching or bending, and the ECLD forms conformal contact with the skin and maintains great performance under large deformation, and the device with multiple patterns and multi-pixel arrays can follow the skin bending, twisting, and stretching to maintain stable display.

Combining pressure-sensitive and wearable applications, Moon et al.[129] proposed an ionogel with dual electrical and optical signal outputs (Fig. 8d). The porous structure with a sugar cube as the “skeleton” provides a large deformation space, and the change of the optical signal reflects the change of the contact area and the bending motion of the fingertip during the compression process. Even under very low pressure, the ionic gel can deform through the compressed pores and

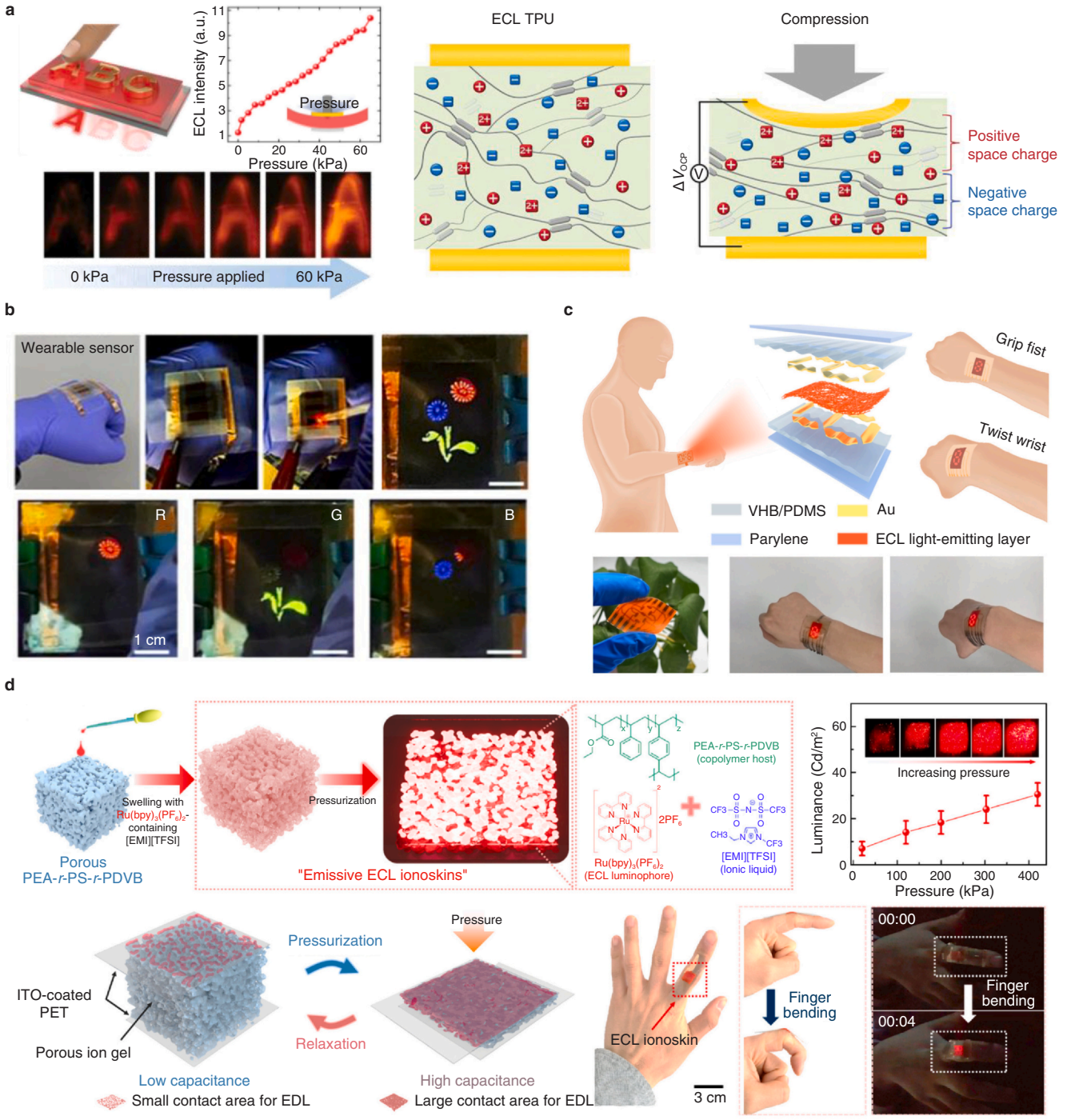

Fig. 8. Pressure-sensitive and wearable ECLDs. (a) Visco-poroelastic ECLDs with piezo-ionic effect[131]. Copyright 2021, Wiley-VCH. (b) Wearable and semi-transparent pressure-sensitive ECLDs[128]. Copyright 2020, American Chemical Society. (c) Wearable ECLDs with multiple patterns and multi-pixel arrays displayed [25]. Copyright 2024, Wiley-VCH. (d) Porous ionoskins with electrical and optical dual output[129]. Copyright 2021, American Chemical Society.

cause a large change in the contact area between the gel and the electrode, resulting in a significant difference in dual capacitance. The porous ionogel was applied to wearable sensing by optimizing mechanical and electro-optical properties. The ionogel exhibits excellent stability over 6000 compression-release cycles and maintains stable sensing performance over 15,000 seconds of continuous operation. Its combination of high sensitivity and wide detection range, pressure-dependent ECL, real-time pressure monitoring capability, excellent stability and durability provide new solutions for the development of next-generation wearable devices.

However, the application of ECLDs also faces some challenges, such as material stability, device lifetime and further optimization of the fabrication process[141]. Future research will focus on the development of higher performance materials, optimization of the device structure, and improvement of the precision and efficiency of the fabrication process.

Finally, we show the applications of ECLDs for displays in recent years, especially multi-color, stretchable/flexible, and wearable displays, in a timeline sequence (Fig. 9). It is believed that more high-performance and low-cost ECLDs for light-emitting displays will emerge with the continuous optimization of materials, design and preparation.

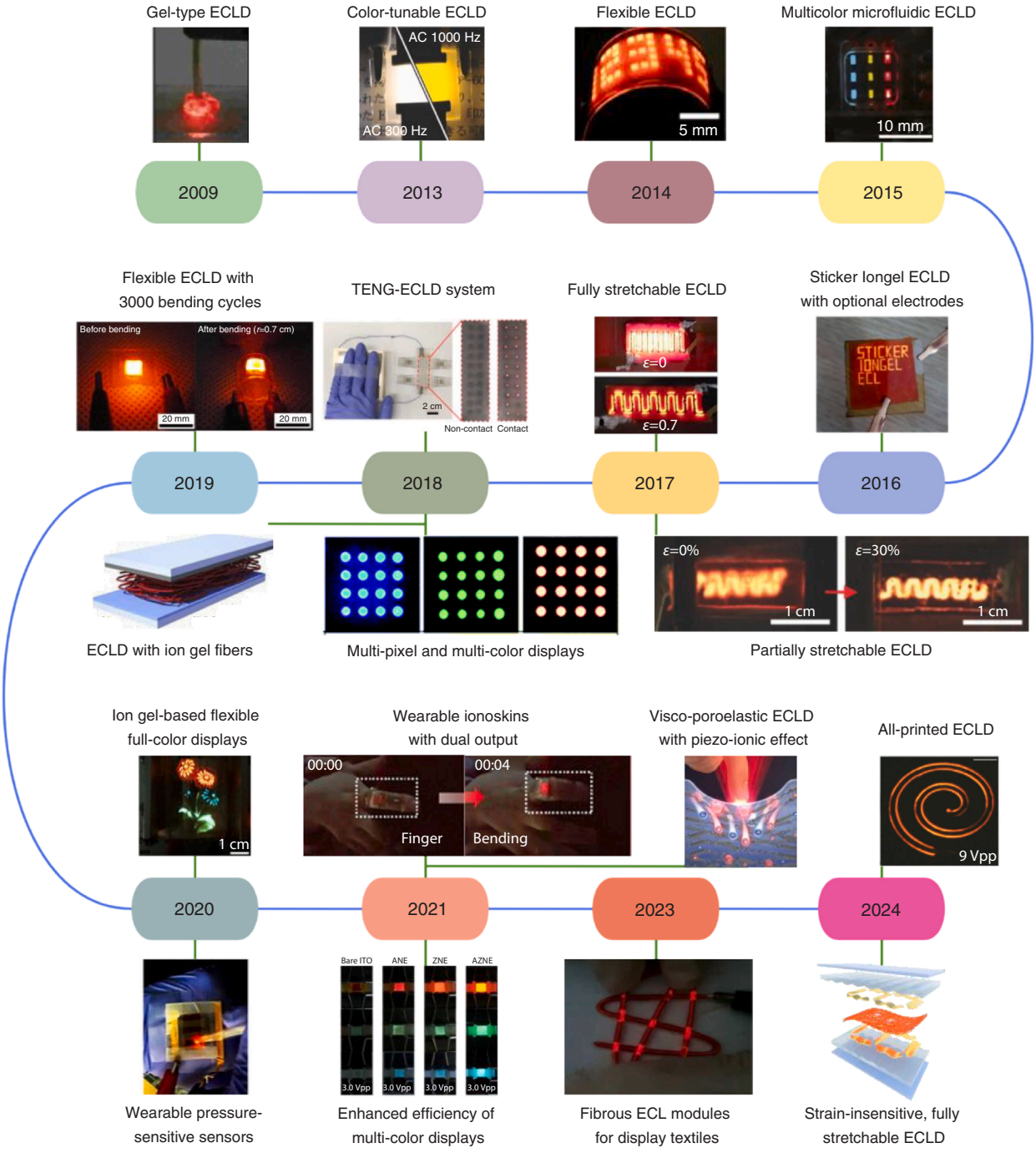

Fig. 9. Time line of the ECLDs for dispaly. In chronological order, it is as follows: Gel-type ECLD[56]. Copyright 2009, IOP Publishing. Color-tunable ECLD[62]. Copyright 2013, Wiley-VCH. Flexible ECLD[32]. Copyright 2014, American Chemical Society. Multi-color microfluidic ECLD[101]. Copyright 2014, Elsevier. Sticker Iongel ECLD with optional electrodes[123]. Copyright 2016, The Author(s). Partially stretchable ECLD[87]. Copyright 2017, Wiley-VCH. Fully stretchable ECLD[30]. Copyright 2017, American Chemical Society. Multi-pixel and multi-color displays[125]. Copyright 2018, The Royal Society of Chemistry. TENG-ECLD system[108]. Copyright 2018, Elsevier. ECLD with ion gel fibers[28]. Copyright 2018, Elsevier. Flexible ECLD with 3000 bending cycles[103]. Copyright 2019, American Chemical Society. Ion gel-based flexible full-color displays[31]. Copyright 2020, Elsevier. Wearable pressure-sensitive sensors[128]. Copyright 2020, American Chemical Society. Wearable ionoskins with dual output[129]. Copyright 2021, American Chemical Society. Enhanced efficiency of multi-color displays[130]. Copyright 2021, Elsevier. Visco-poroelastic ECLD with piezo-ionic effect[131]. Copyright 2021, Wiley-VCH. Fibrous ECL modules for display textiles[112]. Copyright 2023, American Chemical Society. Strain-insensitive, fully stretchable ECLD[25]. Copyright 2024, Wiley-VCH. All-printed ECLD[29]. Copyright 2024, Wiley-VCH.

5. Conclusions and outlook

In general, this paper systematically summarizes the research progress of ECLDs in material system, preparation process and flexible displays application. Through the in-depth analysis of the ECL principle, the luminescence mechanism of ECLD is clarified, and the key points of material selection for the electrochemical luminescence layer and electrode layer are discussed. At the material design, the preferred selection and combination of luminophores, ILs and conductive electrode materials significantly enhance the luminescence efficiency and interface stability of ECLD. At the process optimization, solution coating, vapor deposition, as well as precision patterning, printing technology, and electrospinning technology, provide diverse options for the efficient preparation of ECLDs. As for the application, this review

discusses the latest progress of ECLDs in the fields of multi-color, flexible/stretchable, and wearable displays, demonstrating the great potential in the field of flexible electronics. Future work needs to focus on the following directions.

5.1. Synergistic optimization of material systems

Currently, ECLD still suffers from the lack of material system stability and compatibility. Most ECL luminescent materials (e.g., ruthenium complexes, quantum dots) face irreversible reactions in long-term electrochemical cycling and are prone to redox reaction-induced structural decomposition. The electrolyte ion migration efficiency is low, and traditional ILs or gel electrolytes are prone to uneven ion distribution under dynamic deformation and concentration polarization under high-frequency AC drive, which leads to a decrease in the reaction rate and affects the charge transfer rate and reaction kinetics. Therefore, the redox electrochemical reaction-based light-emitting devices have shortcomings such as brightness attenuation and shorter life span during long-term use, and maintaining uniform and stable brightness over a large area is the goal that must be pursued to realize fully stretchable displays. It is undeniable that brightness of some ECLDs has reached a high level, however, the overall luminous efficiency is still lower than that of OLEDs and other mature technologies. It is necessary to explore new efficient and stable luminophores and electrolyte systems and high-throughput, low-cost fabrication processes to further increase the brightness and efficiency, and improve the stability and lifetime.

5.2. Multiple innovations in preparation processes

The innovation of ECLD preparation process needs to shift from single technology breakthrough to multi-process synergistic optimization. In terms of preparation, traditional preparation methods such as spin coating and drop coating are still the mainstream technology for luminescent layer deposition by virtue of their low cost and convenience, but it is difficult to realize the preparation of large areas and complex patterns. Metal mask printing, inkjet printing, have achieved micron-level patterning, but the finer process accuracy is still facing challenges, complex structure device uniformity control and high-resolution patterning is difficult to consider, the printing technology of the ink rheological properties of the stringent requirements of the limitations of the material universality. There is a need to further improve the precision of the preparation process, such as the development of hybrid processes based on nanoimprinting or photolithography, which are expected to achieve higher-precision patterning. While improving the performance, the issue of cost is also a must for moving towards industrialization. It is necessary to reduce the preparation cost of ECLDs through material and process optimization in order to improve their market competitiveness.

5.3. Continuous expansion of application scenarios

Despite the potential applications of ECLDs in display technology and lighting, the range of applications is still relatively limited. Applications in flexible displays and wearable devices are still in the laboratory research stage and have not yet been commercialized on a large scale. In addition, the application of ECLDs in other fields such as biomedicine and optical communication is also in the exploratory stage. Future research requires in-depth exploration in material selection, device structure optimization, and preparation process improvement to promote the wide application of ECLDs. In terms of display control, the dynamic display of patterns can be controlled using a program, considering the interface between the circuit and the flexible interface, while solving the crosstalk between the light-emitting pixels, and combining with the back-end circuits to ultimately realize large-scale, multi-patterned, and integrated design of the device. Deepen the integration of ECLD with intelligent sensing and wearable technology, and promote its application in dynamic display, real-time sensing and other scenes.

CRediT authorship contribution statement

Ziyu Chen: Writing – original draft. **Huichen Xu:** Writing – original draft. **Caofeng Pan:** Writing – review & editing. **Rongrong Bao:** Writing – review & editing.

Consent to participate

Non applicable

Ethical approval

Non applicable

Declaration of Competing Interest

Caofeng Pan is the Editorial Board Member of the journal Wearable Electronics and was not involved in the editorial review or the decision to publish this article. The other authors declare that there are no competing interests.

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