

Review Article

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Flexible and stretchable electrochromic displays: strategies, recent advances, and prospects

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Abstract

Flexible and stretchable electrochromic displays (ECDs) perform a crucial function in Internet of Things (IoT) systems, as they have shown superior eye-friendly, energy-saving, mechanical (flexibility and stretchability) properties. They can be integrated with IoT devices and successfully applied as wearable and intelligent electronics. Flexible and stretchable ECD technology has shown promising potential but is still in the early stage of development. A systematic overview from comprehensive perspectives of materials selection and modification, structure design, and advanced fabrication methods of this technology is necessary. In this review, we concentrate on the strategies in substrates, active layers, and electrolyte aspects to fabricate high-performance flexible and stretchable ECDs. We have systematically summarized the materials selection/modification and device structure design in these strategies. We also outline recent advances in flexible and stretchable ECDs based on processing methods for electrode patterns, active layer patterns, electrolyte patterns, and ECD pixels. Moreover, the interactive visual displays integrating ECD pixels with different sensors have been elaborated. Finally, we outline the future directions for developing flexible and stretchable ECDs, focusing on materials, methods, and applications. These prospects aim to overcome the limitations in pattern resolution, electrolyte uniformity, and pixel size/number and realize the manufacturable, commercialized, scalable, and robust flexible ECDs. This review can further promote the basic research and advanced fabrication of flexible ECDs and facilitate the advancement of multifunctional displays to satisfy the increasing demand for next-generation flexible electronics.

Keywords: Electrochromic displays, flexible electronics, pattern design, pixelated displays

INTRODUCTION

During the rapid evolution of Internet of Things (IoT) systems, flexible display technologies have played a



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significant role in IoT, providing bendable and conformable display solutions to create versatile and adaptive interfaces that seamlessly integrate with interconnected devices^[1,2]. So far, these technologies have been widely studied and can be divided into emissive and reflective (non-emissive) displays^[3]. The flexible emissive displays, including liquid crystal (LCDs)^[4], light-emitting diode (LED)^[5], organic LED (OLED)^[6], and quantum-dot LED (QLED) displays^[7], have been extensively developed even with some commercial products as they have advantages of high resolution and vibrant colors. However, these displays strongly rely on internal backlight sources, suffer from limited lifespan issues and environmental concerns, and are susceptible to image retention with high voltage input and power consumption. In this case, the reflective (non-emissive) flexible displays, including electrophoretic (EPDs)^[8,9], electrowetting (EWDs)^[10], photonic crystal (PCDs)^[11], and electrochromic displays (ECDs)^[12], utilize the ambient light with lower voltage input and energy consumption would be a more advanced choice. They have shown superior eye-friendly properties, high suitability for outdoor use, and lower energy consumption^[13]. Among them, the flexible ECDs provide unique advantages compared to other reflective flexible displays. EWDs are known for fast response and good readability but face challenges with dielectric breakdown and high power consumption. ECDs are more energy-efficient than EWDs, which benefit from their inherent bistable nature allowing them to maintain the display content without continuous power input^[14]. Compared to EPDs, ECDs also offer faster response speed and better durability^[15]. PCDs provide high reflectivity and vibrant color but struggle with complex and costly preparation processes, while ECDs can be fabricated with facile and cost-effective methods. These advantages make ECDs stand out in functionalities and receive extensive attention, and successfully expand their applicability into wearable electronics, innovative packaging, flexible signage, and e-readers^[16,17]. Generally, the flexible ECD is constructed by the flexible electrode, active layers (electrochromic layer and ion storage layer), and ionic conducting electrolyte with a vertical or lateral structure^[18]. The underlying mechanism of the ECD is based on the redox reaction of electrochromic (EC) materials driven by external electricity input. Then, ECDs can present corresponding optical changes (absorbance/transmittance/reflectance) to display content and convey information^[15]. To have a systematical understanding of the flexible and stretchable ECDs, we have summarized the strategies for constructing these displays based on different device layers. Materials selection or modification and device structure design or distribution have been categorically introduced in this part. Besides, we have elaborated on recent advances in these flexible and stretchable ECDs from the perspectives of designing electrode patterns, active layer patterns, electrolyte patterns, and ECD pixels. Moreover, the versatile visual displays on the interactive system consisting of ECD pixel and sensor modules have also been summarized according to various signal types.

STRATEGIES FOR FABRICATING FLEXIBLE AND STRETCHABLE ELECTROCHROMIC DISPLAYS

When constructing flexible and stretchable ECDs, it is important to select the materials and consider the design and fabrication strategies for three aspects: the electrode, active layer, and electrolyte.

Flexible and stretchable electrode

The strategies aimed at developing flexible and stretchable electrodes for ECDs predominantly centered on the materials selection and integration of conductive layer and substrate to achieve desirable chemical, thermal, mechanical, and electrical characteristics^[17]. The prevalent choice for flexible electrodes is the commercially available indium tin oxide (ITO) deposited on polyethylene terephthalate (PET) or polyethylene naphthalate (PEN) substrates^[19-22]. However, the ITO exhibits a brittleness trait, constraining its chemical and mechanical performances^[23]. In this case, researchers have explored various alternative conductive materials onto conventional flexible PET substrates as flexible electrodes in ECDs, including poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS)^[24,25], silver nanowires (AgNWs)^[26,27],

metal grid (Au/Ag/Ni grid)^[28-30] and metal mesh (Ni/Ag mesh)^[31]. The inherent structural properties of metal nanowires (NWs)/mesh/grid and polymer confer the robust mechanical stability of these electrodes. Leveraging the outstanding conductivity of silver grids, our group fabricated flexible Ag grid-PET electrodes applied in WO₃-based ECD with high conductivity (1-5 Ω/sq) and transmittance (83% @400-800 nm) through direct preparation and chemical sintering [Figure 1A]^[30]. Besides, due to the susceptibility of some metals (Ag, Zn) to oxidation and corrosion, additional protective layers are often incorporated to bolster the chemical stability of flexible electrodes^[28,31-33]. For the In-coated Zn/Au grid-PET flexible electrode in multicolor ECD [Figure 1B]^[28], the Zn layer is added for involving electrochemical reaction and the In layer is additionally coated as a corrosion inhibitor to improve chemical stability and hydrogen evolution overpotential. For instance, an additional dense electrodeposited Ni mesh layer was prepared to fully cover the spikes of the Ag mesh layer, followed by the addition of a coated PEDOT:PSS PH1000 layer, resulting in an Ag/Ni mesh/PH1000 electrode with high electrochemical stability^[31]. Besides, considering that uneven electrical distribution of metal mesh/grids may cause the blooming effect of the ECDs, the additional polymeric layer can also homogenize the electrical field over the whole electrode, resulting in a faster switching speed and more uniform coloration state^[29,34]. Furthermore, some emerging materials have been selected to fabricate the flexible substrates of ECDs, including degradable and recyclable gelatin films [Figure 1C]^[35], commercial transparent tape^[36], biocompatible agarose/poly(ethylene glycol) (PEG) hydrogel substrates [Figure 1D]^[37]. The selection of these materials further broadens the ECD applications as green bioelectronics. In addition, fiber- and cellulose-based materials with high sustainability and biocompatibility have also been developed as substrates in the electrochromic field for intelligent wearable electronics^[38-44]. The single wall carbon nanotube (SWCNT) @AgNWs has been successfully transferred from the polycarbonate (PC) filter membrane to the nanopaper substrate owing to the strong hydrogen bonding between celluloses and conductive materials, ensuring the transparent conducting nanopaper's electrical and mechanical stability [Figure 1E]^[44]. Chen *et al.* reported an electrochromic fiber-shaped supercapacitor displaying a designed pattern, where this display is based on a carbon nanotube (CNT) sheet wound on the elastic rubber fiber^[43]. Besides, various fiber-based electrodes have been developed in flexible ECDs, such as the PEDOT:PSS/dimethyl sulfoxide (DMSO) on PET fabrics^[38] [Figure 1F], Au layers deposited on PET synthetic leather^[45], and metal wires (stainless wires^[46], Cu@Ni wires^[47]).

As for stretchable ECD, substrates such as polydimethylsiloxane (PDMS)^[48], styrene-ethylene-butylene-styrene (SEBS)^[49,50], and thermoplastic polyurethane (TPU)^[51] are commonly selected. To fabricate stretchable conductors with stable conductive properties under diverse mechanical and chemical conditions, our research group has developed various transparent electrodes by transferring AgNWs into PDMS substrates with an intimate combination, then followed by coating a PEDOT:PSS thin layer for further protection and improved electrical connection^[52-54]. Attributing to the deformability of PDMS, we further integrated the PDMS precursor with the phosphorescent materials as a phosphorescent substrate, which can function as a light source in dark conditions and successfully explore the application as a light-adaptable ECD [Figure 1G]^[53]. Besides, the WO₃/silver nanotrough network (AgNN)/PEDOT:PSS conductive layer with a well-encapsulated structure was successfully fabricated into various stretchable polyethylene (PE) cling wrap, PDMS, and Ecoflex substrates, exhibiting slight resistance variation upon stretching [Figure 1H]^[55]. Besides, the strategy of constructing semi-interpenetrating networks in stretchable and highly conductive polymers (ISHCP)-SEBS electrodes was well adopted, demonstrating minimal resistance loss even under 400% stretching [Figure 1I]^[50].

Active layers

The active layers are the key components of ECDs, which undergo the reversible electrochemical reaction upon applied bias to modulate the display's optical properties (transmittance, absorbance, and reflectance)^[56]. The critical performance indicators for constructing active layers in flexible electrochromic

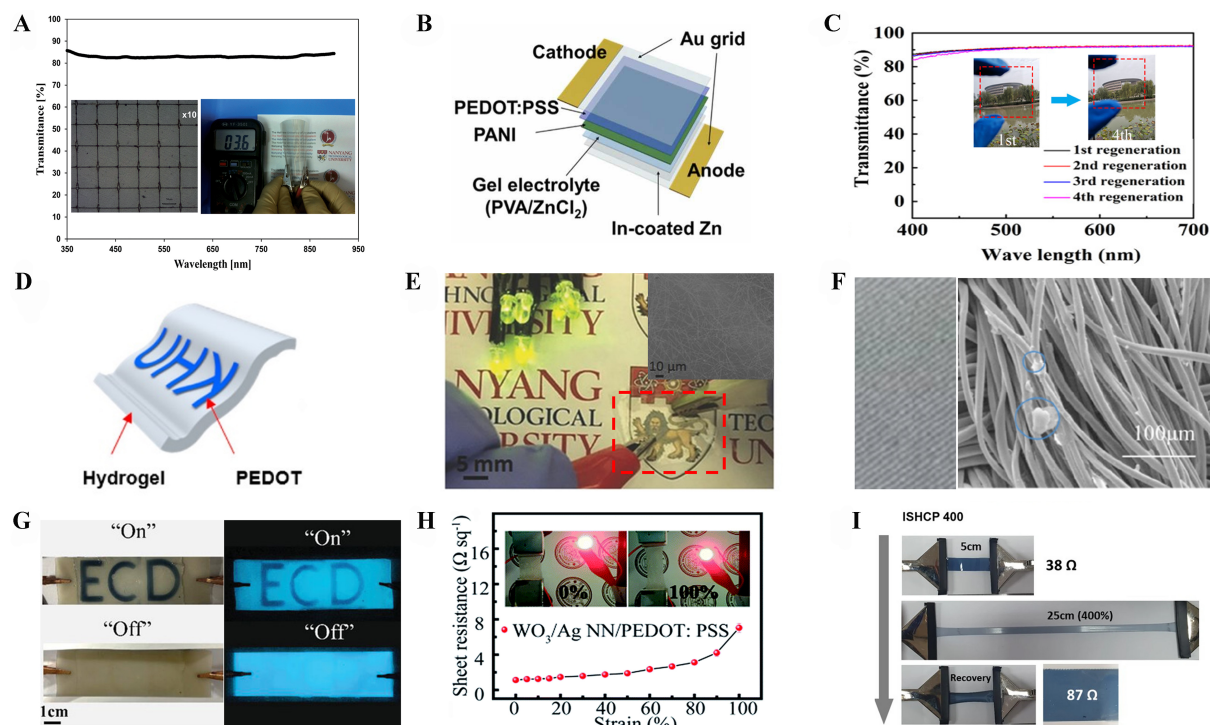


Figure 1. Strategies for fabricating flexible and stretchable electrodes. (A) Flexible Ag grid-PET electrodes with high conductivity ($1\text{--}5\ \Omega/\text{sq}$) and transmittance (83% @ $400\text{--}800\ \text{nm}$). Reproduced with permission^[30]. Copyright 2014, RSC Publications; (B) In-coated Zn/Au grid-PET electrode as the anode in the PANI-based ECD. Reproduced with permission^[28]. Copyright 2023, Elsevier; (C) The degradable and recyclable gelatin film as substrate in ECD. Reproduced with permission^[35]. Copyright 2022, ACS Publications; (D) The biocompatible agarose/PEG hydrogel substrate in PEDOT-based ECD. Reproduced with permission^[37]. Copyright 2016, ACS Publications; Nanocellulose-based electrodes of (E) foldable SWCNT@Ag nanopaper electrode and (F) PEDOT:PSS/DMSO-PET fabric electrode. Reproduced with permission^[38,44]. Copyright 2016, Wiley-VCH. Copyright 2023, Elsevier; (G) Stretchable phosphorescent PDMS-based substrate applied in the light-adaptable ECD. Reproduced with permission^[53]. Copyright 2023, Wiley-VCH; (H) The resistance change of a designed $\text{WO}_3/\text{AgNN}/\text{PEDOT:PSS}$ film on ecoflex with the strain change. Reproduced with permission^[55]. Copyright 2018, RSC Publications; (I) A ISHCP-SEBS electrode with stable conductivity upon stretching. Reproduced with permission^[50]. Copyright 2020, Springer Nature. PET: Polyethylene terephthalate; PANI: polyaniline; ECD: electrochromic display; PEG: poly(ethylene glycol); PEDOT: poly(3,4-ethylenedioxythiophene); SWCNT: single wall carbon nanotube; PEDOT:PSS: poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; DMSO: dimethyl sulfoxide; PDMS: polydimethylsiloxane; AgNN: silver nanotrough network; ISHCP: stretchable and highly conductive polymer; SEBS: styrene-ethylene-butylene-styrene.

devices are obtaining large optical modulation, fast switching speed, and stable cycling with various mechanical conditions. First, we summarize the electrochromic materials used in reported flexible and stretchable ECDs for the past ten years, which can be classified into organic and inorganic parts. The most studied inorganic materials are transition metal oxides (WO_3 , NiO , MnO_2 , and V_2O_5)^[20,57–59], Prussian blue $\{\text{PB}, \text{Fe}^{3+}_4[\text{Fe}^{2+}(\text{CN})_6]_3\}$ ^[21,60,61] and nickel hexacyanoferrate $\{(\text{KNI}^{2+}[\text{Fe}^{3+}(\text{CN})_6], \text{NiHCF})\}$ ^[21], which are based on the ion/electron dual insertion/extraction mechanism to realize the valence state variation with resulted electrochromic phenomenon of the film^[13]. State-of-the-art strategies, including nanostructure construction, doping, and component fabrication, are usually adopted to obtain satisfactory performances of inorganic electrochromic materials, which have been extensively reviewed in reported reviews^[12,17]. The commonly studied organic materials are viologen^[19,62,63], conducting polymers [polyaniline (PANI), PEDOT:PSS, polypyrrole (PPy), and poly(3,4-propylenedioxythiophene) (PProDOT)]^[31,35,38,64,65] and metal coordination complexes^[22,66]. The polymeric electrochromic material has attractive traits for flexible displays as it can provide superior mechanical flexibility, multicolor tunability, and fast switching. The previous review has comprehensively introduced representative examples, materials modifications, and corresponding mechanisms of the organic electrochromic materials for display applications^[67].

In addition to the material selection and modification of the electrochromic active layer, the design strategies for active layer structure should also be emphasized. With these structural design strategies, the ECDs are expected to realize multicolor, self-powered abilities and further enhance long-term stability. The first category is to fabricate the Fabry-Perot (F-P) type electrochromic electrodes^[68-70]. They have the structure of a WO_3 layer (varied thickness)/partially reflective metal W layer/conducting layer on the flexible substrate, which resulted in versatile structural colors of electrodes [Figure 2A]. The second category is to introduce a complementary electrochromic counter electrode, which functions as an ion storage layer for reversible electrochemical reactions and provides additional coloration ability. Examples of the working/counter electrochromic pair electrodes of MnO_2/PB ^[58], WO_3/NiO ^[20], poly(3-methylthiophene) (P3MT)/PB [Figure 2B]^[61], $\text{WO}_3/\text{P3MT}$ ^[41], PEDOT/PPy^[71], PANI/ V_2O_5 ^[59] have been extensively studied in high-performance ECDs. The third category is to introduce other types of active counter layers, such as the active metal electrode (Zn/Al) releasing ions and providing the build-in potential of the device [Figure 2C]^[21,28,53,72], and redox mediators [Ferrocene, dimethyl ferrocene (dmFc)] in the electrolyte as counter electrode material^[19,73].

Electrolyte

In ECDs, the electrolytes provide ionic conduction and prevent direct electrical contact between electrodes^[74,75]. The polymer matrix is usually chosen as part of the electrolyte component for the flexible and stretchable ECDs because of the superior mechanical flexibility. The selection of polymer materials is of great importance to electrochromic devices as it aims to ensure the electrolyte possesses superior mechanical strength, a wide potential window, and stable thermal, chemical, and electrochemical properties^[76,77]. To date, the extensively studied polymer materials are polyacrylamide (PAAM), polyethylene oxide (PEO)^[62], poly methyl methacrylate (PMMA)^[19], poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP)^[60], polyvinyl alcohol (PVA)^[53], and other natural and biodegradable polymers (agarose/gelatin)^[78,79]. The basic properties of each polymer used in the electrolytes of electrochromic devices have been illustrated in our previous published review in detail^[80]. Polymer electrolytes are usually composed of the dissolved salt in the polymer host structure, and the choice of salt greatly influences the ionic conductivity, potential window, and working temperature range of the electrolyte^[74]. Polymer-salt-solvent-based electrolytes are commonly developed in electrochromic devices such as the PMMA- LiClO_4 -PC organic gel and PAAM-LiCl hydrogel, which shows high ionic conductivity and good electrochemical stability at ambient temperature. To further improve the high/ultralow temperature adaption and ionic conductivity and widen the electrochemical window of the electrolyte, the emerging ionic liquid-based polymer electrolytes can achieve these targets^[81,82]. Santiago-Malagón *et al.* reported an ionogel comprising PVDF-HFP, 1-Ethyl-3-methylimidazolium trifluoromethanesulfonate (EMIM-Tf), and potassium trifluoromethane sulfonate (KTf) applied in the PB-based-ECD which provides a stable chemical environment for device operation^[60]. The deep eutectic solvents (DES), as the eutectic mixtures of Lewis, maintain similar favorable properties (wide tolerant temperature, high ionic conductivity, and large electrochemical window) as ionic liquids, which also have been successfully fabricated as DES-based gel in ECD. The Litfsi in N-methylacetamide (NMA) as the DES integrating with the copolymer poly(ethoxyethoxyethyl acrylate-co-isobornyl acrylate) [P(DEEA-co-IOBA)] exhibits high optical transmittance over 90% and ionic conductivity of 0.63 mS/cm and stretching strain over 2,000% [Figure 2D], resulting in the excellent electrochromic performances of the display^[20]. Furthermore, to increase the compatibility of polymer matrices and ionic liquids, our group has proposed a novel electrolyte in flexible ECD in which the poly[ionic liquid, 1-butyl-3-vinylimidazolium bis(trifluoromethanesulfonyl)imide (BVIMTFSI)] as the polymer backbone that anchors another ionic liquid (EMIMTFSI) with rapid photopolymerization method. The prepared ionogels have favorable physicochemical (thermal, electrochemical, mechanical) stability [Figure 2E], ensuring high performances of both WO_3 and iron-centered coordination polymer (FeCP)-based flexible ECD^[83]. In addition to the above strategies, a series of electrochromic ion gels were designed

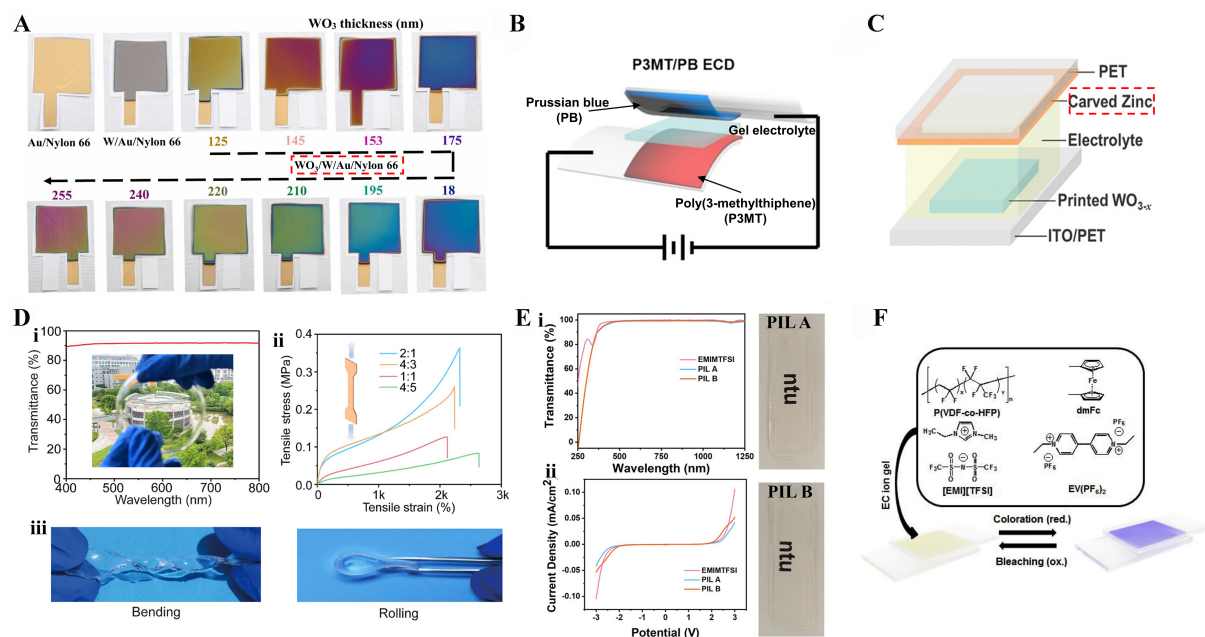


Figure 2. Strategies for fabricating active layers and electrolytes. (A) Optical photographs of WO₃ layers of varied thicknesses / W layer / Au (50 nm) on Nylon 66 substrate. Reproduced with permission^[68]. Copyright 2023, Elsevier; (B) The ECD structure is based on working/counter(P3MT/PB) EC pair electrodes. Reproduced with permission^[61]. Copyright 2019, Elsevier; (C) The ECD structure is based on the WO₃/Zn electrodes. Reproduced with permission^[72]. Copyright 2020, Wiley-VCH; (D) The excellent optical and mechanical performance of DES-based gel used in ECD. Reproduced with permission^[20]. Copyright 2022, Wiley-VCH; (E) The high transmittance, stretchable, and favorable electrochemical stability of ionogels used in ECD. Reproduced with permission^[83]. Copyright 2023, Wiley-VCH; (F) Chemical structures of components in EC electrolyte gel and structure of EC devices. Reproduced with permission^[32]. Copyright 2018, Elsevier. ECD: Electrochromic display; P3MT: poly(3-methylthiophene); PB: Prussian blue; EC: electrochromic; DES: deep eutectic solvents.

by cooperating with soluble electrochromic materials (mostly viologen type), electrolyte solution, and polymer skeleton, which act as both electrochromic layers and ionic conductors^[19,32,44,62,73,84-86]. The ECD with this multifunctional ion gel greatly simplified the device structure, resulting in rapid switching speed. Besides, the soluble anodic species (DMFc and Fc)^[32,73,86] are usually involved in the gel to supply electrons and balance charges during electrochemical reactions with desirable cycling performance [Figure 2F].

RECENT ADVANCES

This section will summarize the recent advances in reported flexible and stretchable ECDs over the last decade. Based on the device structure, we have clarified the following four design strategies [Figure 3]^[87].

Electrode pattern design

Designing electrode patterns is one of the commonly used strategies to fabricate a flexible ECD to convey information or content by electrochromic devices. As for commercial flexible electrodes such as ITO-PET and ultrathin ITO-glass, various etching methods were adopted to eliminate partially conductive layers, such as chemical/laser etching via cover photolithography/masking^[19,83,85,88-90]. In these ways, electrons only transfer in the active conductive part with the corresponding electrochemical redox process in the electrochromic layer, showing patterned information and realizing information encryption and transmission. Gu *et al.* successfully fabricated a bistable flexible ECD, which shows a Quick Response (QR) code pattern with high-resolution quality based on a laser-etching approach of PET-ITO substrates. This impressive bistable display, possessing eye-friendly and energy-saving properties, offers enhanced prospects for application in the field of human-computer interaction [Figure 4A]^[90]. Song *et al.* have demonstrated an

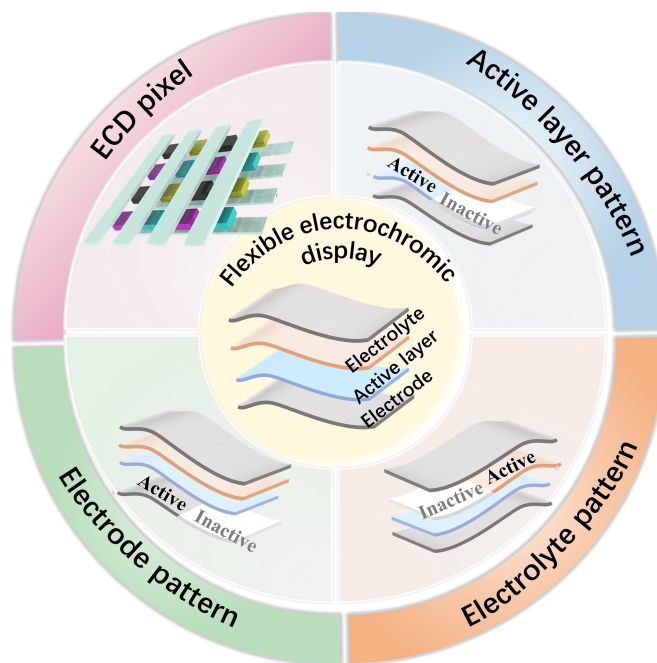


Figure 3. Four design strategies of flexible and stretchable ECDs, including electrode pattern, electrolyte pattern, active layer pattern, and ECD pixels (Reproduced with permission^[87]. Copyright 2022, ACS Publications). ECDs: Electrochromic displays.

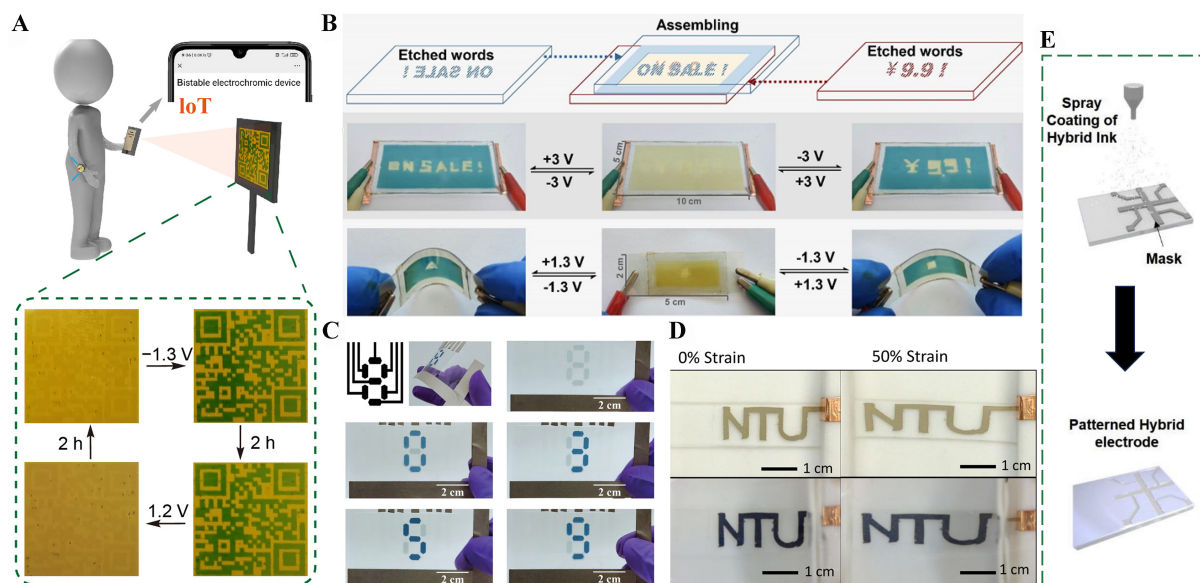


Figure 4. Recent advances in electrode pattern design. (A) A bistable ECD showing a QR code with laser-etched ITO-PET electrode. Reproduced with permission^[90]. Copyright 2022, CCS Publications; (B) Schematic and photographs of the flexible chalcogenoviologens-based display based on different etched patterns on ITO-PET electrodes. Reproduced with permission^[85]. Copyright 2021, Elsevier; (C) A patterned ECD with the etched digital array on ultrathin flexible ITO-glass electrode. Reproduced with permission^[83]. Copyright 2023, Wiley-VCH; (D) ECD based on the patterned AgNWs on the PDMS electrode. Reproduced with permission^[54]. Copyright 2014, ACS Publications; (E) Schematic illustration of template-assisted methods with spray coating to fabricate patterned AgNWs/PEDOT:PSS-PET electrode. Reproduced with permission^[32]. Copyright 2021, Elsevier. ECD: Electrochromic display; QR: Quick Response; ITO: indium tin oxide; PET: polyethylene terephthalate; AgNWs: silver nanowires; PDMS: polydimethylsiloxane; PEDOT:PSS: poly(3,4-ethylenedioxythiophene) polystyrene sulfonate.

easily bendable flexible chalcogenoviologens-based display composed of two ITO-PET electrodes with different etched patterns where the information of “¥ 9.9 !”/“ON SALE !” and “□”/“△” can alternatively exhibit under corresponding applied voltages [Figure 4B]^[85]. Moreover, our group fabricated a patterned digital array electrode by wet-etching the ultrathin flexible ITO glass, realizing dynamic switching of multiple number patterns and significantly improving the information capacity of WO₃-based ECDs [Figure 4C]^[83]. Apart from the etching methods on commercial flexible electrodes, there are other advanced approaches for mounting conductive patterns on flexible substrates. For example, the conductive PEDOT:PSS ink was directly screen-printed and inkjet-printed on flexible substrates (such as pre-engraved PET and paper substrates) as patterned electrodes. They can provide favorable electrical connections for the ECDs and are successfully applied as promising applications (electrochromic biosensors and paper-based ECDs)^[60,91]. Zhao *et al.* prepared a transparent Ni grid-PET electrode with multiple designed patterns through lithography and electrodeposition processes^[29]. The prepared Ni electrode has a high optical transmittance, electrical conductivity, and mechanical stability, which is advantageous for efficient electron interaction inside the flexible ECDs under various mechanical conditions and demonstrates its suitability for high-performance flexible ECD with fast switching speed and good bending capacity. Besides, our group utilizes the template-assisted filtration method and transfers the desired AgNWs patterns onto the PDMS elastomer substrate^[54]. This stretchable electrode was successfully assembled into the first stretchable and wearable patterned WO₃-based ECD, demonstrating its display function under 50% strain [Figure 4D]. Similar template-assisted technology with spray coating method has also been adopted to fabricate patterned electrodes^[26], such as AgNWs/PEDOT:PSS-PET electrodes [Figure 4E]^[32]. Overall, the strategies for designing electrode patterns have shown advanced progress on flexible and stretchable ECDs. The streamlined and feasible preparation techniques would always be preferentially considered for future displays. More importantly, the high information capacity and good display quality with high resolution will be promising targets for the electrode pattern design strategies.

Active layer pattern design

The strategies of designing active layer patterns in flexible and stretchable ECDs have also been most widely adopted over the last decade. These strategies are mainly classified into template-assisted and direct printing methods. With these strategies, only the active patterned electrochromic layer will process the electrochemical redox reaction and exhibit its optical properties variation upon electricity. In the following part, we will comprehensively explain how these methods achieve active layer patterning and attain the corresponding display functionalities of the fabricated electrochromic devices.

Template-assisted methods

As for the template-assisted methods, the common fabrication process is to deposit the active layer on the electrode with the mounted mask. So far, the most developed deposition methods are spray-coating, spin-coating, magnetron sputtering, electron beam thermal deposition, electrodeposition, and electrochemical polymerization. The deposition conditions and adhesion between the template and electrode determine the selection of the template. For example, a stable metal sheet was used as the shadow mask for thermally depositing a bowknot-like patterned WO₃ layer on a Cr/Ag-PET electrode under severe conditions (low vacuum). The prepared patterned electrochromic electrode was further assembled into the device, exhibiting dynamic multicolor under different potentials [Figure 5A]^[92]. Chen *et al.* have precisely fabricated the two-dimensional (2D) PEDOT micropatterns with a high fidelity of 97.3% compared to the patterned agarose template through the localized electrochemical polymerization method [Figure 5B]^[78]. It was realized by strong adhesion between the ITO electrode and agarose with electrolyte. The ionic transport kinetics of the electrochromic layer was boosted owing to the 2D structure of PEDOT which resulted in faster switching speed and electrochromic performances. This micropatterned PEDOT was further designed with the logo of Xiamen University and illustrated its application as a flexible ECD. Besides, a 2D ordered

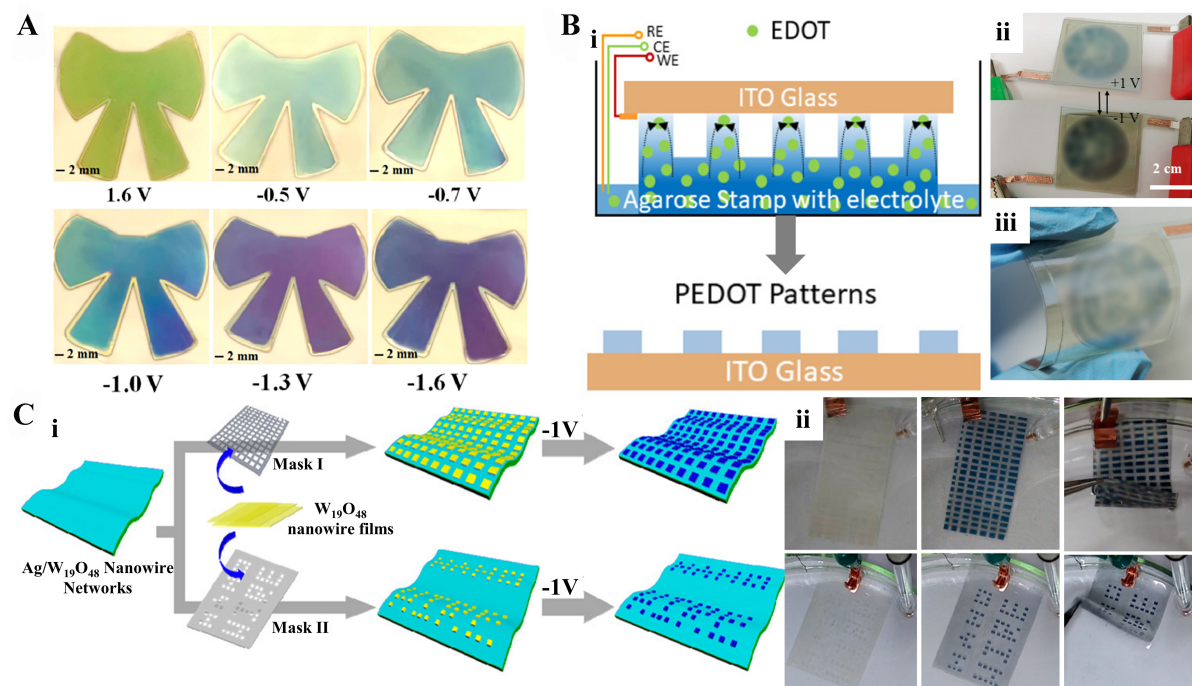


Figure 5. Recent advances in active layers pattern design. (A) A multicolor ECD based on the bowknot-like patterned WO₃ layer on the Cr/Ag-PET electrode with thermal deposition method. Reproduced with permission^[92]. Copyright 2024, Elsevier; (B) Flexible ECD with logo content based on the PEDOT micropattern with localized electrochemical polymerization method using agarose template. Reproduced with permission^[78]. Copyright 2021, ACS Publications; (C) Flexible ECD fabricating based on 2D ordered Ag/W₁₈O₄₉ nanowires network with LB technique. Reproduced with permission^[33]. Copyright 2017, ACS Publications. ECD: Electrochromic display; PET: polyethylene terephthalate; PEDOT: poly(3,4-ethylenedioxythiophene); 2D: two-dimensional; LB: Langmuir-Blodgett.

Ag/W₁₈O₄₉ NW network has been formed on the ITO-PET substrate by a novel Langmuir-Blodgett (LB) technique as the transparent electrode, following the patterned W₁₈O₄₉ NW film with different masks was assembled via the same technique for its application as electrochromic patterned or pixelated displays [Figure 5C]^[33]. The W₁₈O₄₉ NW film was firmly combined with the network electrode with this nanostructure construction, which protects the AgNWs layer for enhanced chemical stability and shows stable and satisfactory electrochromic performances with a large interface surface area. This strategy offers a novel micro-nano scalable approach to manufacturing a high-performance flexible ECD.

Interestingly, feasible ways are emerging, such as simply involving a transparent insulating template [PVA, polyvinyl chloride (PVC), photoresist film] in the electrochromic device forming the patterned content. The pre-patterned PVC sheet between the electrolyte and Fe-based metallo-supramolecular polymer (polyFe) electrochromic film on the ITO-PET substrate ensures that the film in contact with the electrolyte undergoes the electrochemical oxidation and shows the patterned content [Figure 6A]^[66]. Besides, a highly uniform and transparent patterned dielectric PVA layer has been closely screen-printed on the ITO-PET electrode and forms 1-methyl-4,4'-bipyridyl iodide (MBI) patterning via *in-situ* electrochemical processing on the uncovered area of the electrode [Figure 6B]^[62]. Besides, the patterned, insulated photoresist film has been firmly deposited on the ITO-PET electrode through ultraviolet (UV) irradiation with a photomask^[73,93]. The flexible ECD was fabricated by transferring three different viologen derivative gels on the ITO-PET substrate, showing red-green-blue (RGB) colors under -0.7 V [Figure 6C]^[73]. Moreover, the photolithography process of the viologen-based soluble electrochromic layer was simplified by integrating it with UV-curable ion gel, which can be cured on SWCNT/Pd-coated AgNWs (PCSNs) bilayer electrode-

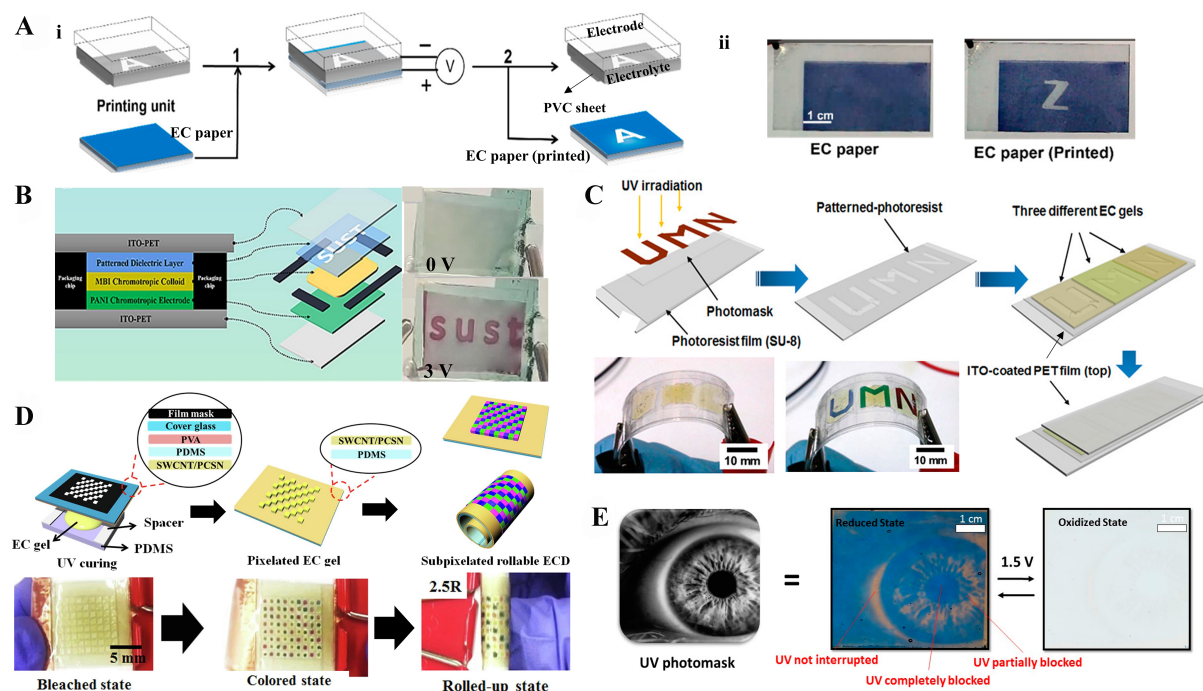


Figure 6. Recent advances in active layers pattern design. (A) The printing process of flexible ECD using a patterned PVC sheet. Reproduced with permission^[66]. Copyright 2023, ACS Publications; (B) Device structure with a screen-printed PVA layer template for forming the MBI patterning with *in-situ* electrochemical processing. Reproduced with permission^[62]. Copyright 2024, Elsevier; (C) Schematic illustration of flexible ECD based on the patterned EC gel using a patterned photoresist template. Reproduced with permission^[73]. Copyright 2016, ACS Publications; (D) Schematic illustration of fabricating the multicolor display with pixelated EC gel. Reproduced with permission^[94]. Copyright 2020, Elsevier; (E) PEDOT:Tos pattern with gradients by using the grayscale picture as the UV photomask. Reproduced with permission^[95]. Copyright 2019, MDPI. ECD: Electrochromic display; PVC: polyvinyl chloride; PVA: polyvinyl alcohol; MBI: 1-methyl-4,4'-bipyridyl iodide; EC: electrochromic; PEDOT:Tos: poly(3,4-ethylenedioxythiophene) with tosylate; UV: ultraviolet.

coated PDMS substrate simultaneously realizing the multicolor pixilation and enhances the display capacity and quantity with microscale size pixels^[86,94]. The specific fabrication process is shown in Figure 6D^[94]. Taking advantage of these flexible EC gels, this assembled ECD showed stable mechanical stability under repeated rolling tests for 500 cycles, which provides a promising strategy for future wearable electronics. Brooke *et al.* have consistently studied the novel active layer patterning techniques based on UV irradiation and vapor phase polymerization for conductive polymers (PEDOT, PPy) using grayscale photomask^[71,95]. The grayscale photomask controlled the amount of UV exposure on the oxidant-coated ITO-PET substrate, resulting in the electrochromic performances with different degrees in the corresponding region of poly(3,4-ethylenedioxythiophene) with tosylate (PEDOT:Tos) film [Figure 6E]^[95]. It realized the electrochromic patterns with gradients and showed a picture-to-picture switching function in the flexible ECD. This technique is an evolutionary progress that significantly enhances the information capacity of the ECD with a high resolution of less than 10 μm . So far, the patterning resolution of the active layer prepared by template-assisted methods is mostly restricted to the millimeter level due to the precision of the template. In short, advanced lithography technologies with nanoscale patterning are urgently needed. After improving the template resolution and the adhesion between electrodes/templates, the display quality of flexible ECDs can be greatly improved. Considering these two factors, more research ideas can be inspired to design active layer fine patterns to promote the advancement of flexible ECDs.

Direct printing methods

The direct printing methods for fabricating active layer patterns in flexible ECDs can be classified into four main categories: screen printing, inkjet printing, bespoke dispenser printing, and brush printing. The screen printing method allows for high-yield and large-scale manufacturing with high resolution ($\approx 70 \mu\text{m}$). The screen-printed ink dispersing the electrochromic materials in solvents and surfactants with a suitable viscosity (500-5,000 cP) and surface tension (25-40 dyn/cm) can be uniformly and closely deposited on various substrates with patterns^[96]. Recent studies reported that the patterned electrochromic electrodes have been screen-printed on PEDOT:PSS paper, ITO-PET substrates, and PEDOT-PET substrates^[96-98]. Brooke *et al.* also developed a screen printing technology combined with vapor phase polymerization to deposit various patterned polymer [poly(dimethylpropylene-dioxythiophene) (PProDOT-Me₂), PPy] films on the PEDOT-PET substrate. This work demonstrates all screen-printed flexible ECDs with various patterns, highlighting their potential for multicolor displays [Figure 7A]^[99]. Besides, inkjet printing is the most commonly used technique because of its advantages of cost-efficiency and ingredient-saving, simplified processing, and fabricating of high-resolution patterns^[31]. This technology has been widely used in rigid and flexible electronics and can print large-scale products^[72,100,101]. The crucial point for high-quality inkjet printing is to ensure the ink has satisfactory rheological properties such as optimum viscosity (1-10 cP), surface tension (28-35 dyn/cm), and appropriate contact angle on the electrode. Besides, the particles with nano-size ($< 200 \text{ nm}$) in the ink should be compatible with the solvent system, forming uniform and stable particle dispersion, which ensures smooth and consistent ink droplets through nozzles^[102]. Our group has put continuous effort into inkjet printing various electrochromic ink on rigid, flexible, and stretchable substrates, achieving versatile inkjet-printed electrochromic devices^[30,52,103-107]. For example, the inkjet-printed WO₃-PEDOT:PSS layer on a flexible Ag grid-PET electrode with various patterns [Figure 7B]^[103], the inkjet-printed WO₃ electrode on a stretchable PEDOT:PSS-AgNWs-PDMS electrode with "NTU" patterns^[52]. Besides, Chen *et al.* have precisely inkjet-printed the multicolor patterned electrochromic film at target positions of the ITO-PEN substrate. The multicolor phenomenon exhibiting the coloration from red to blue is realized by regulating the ratio of Fe/Ru-Metallo-supramolecular polymers (MEPE) solution as a series of inkjet inks. It was further fabricated as a flexible ECD, switching from bleaching to multicolor states at different voltages [Figure 7C]^[22]. Besides, the inkjet printing technique can be extendedly used for both the electrochromic layer and electrode with a simplified process, and the individual printed PEDOT:PSS pixels can be realized with a printed connected Au electrode on cellulose diacetate foil. By attaching the gelatin electrolyte, the assembled biodegradable ECD prototype can present quantities of display information, including battery level, weather conditions, and time [Figure 7D]^[79]. Moreover, a newly water-based additive-free ink has been optimized for inkjet printing patterned ammonium-functionalized viologen derivatives film with high resolutions, exhibiting excellent photo- and electrochromic performances [Figure 7E]^[63]. The bespoke dispenser printing is another reported direct printing method for active layer patterning. This method can allow higher viscosity ink (10-1,000 cP) and larger particle size. Moreover, the ink should exhibit thixotropic behavior to accommodate the printer trait, reducing viscosity under shear during dispensing and staying stable at rest^[108]. For instance, a WO₃-based electrochromic film on the ITO-PET flexible substrate was fabricated by the modified electrostatic force-assisted dispensing printing with an adjustable pattern width from 693 to 256 μm . By using the dmFc as the soluble anodic species, two WO₃ flexible electrodes with different patterns have been fabricated as the symmetric electrochromic device, alternately showing dual images with applied bias under bending conditions [Figure 7F]^[109]. For the brush printing method, a higher viscosity ink (100-10,000 cP) is required to ensure it adheres to the brush and substrate without dripping^[110]. Currently, the brush printing method applied in flexible ECDs is only reported on one paper, where the patterned PEDOT film is painted on the flexible conductive tape with a brush^[36]. The above direct printing methods provide more efficient and facile ways of fabricating the active layer patterns. To ensure high-quality printing, factors of ink formulation, printing parameters, and post-printing treatments should be carefully considered and optimized.

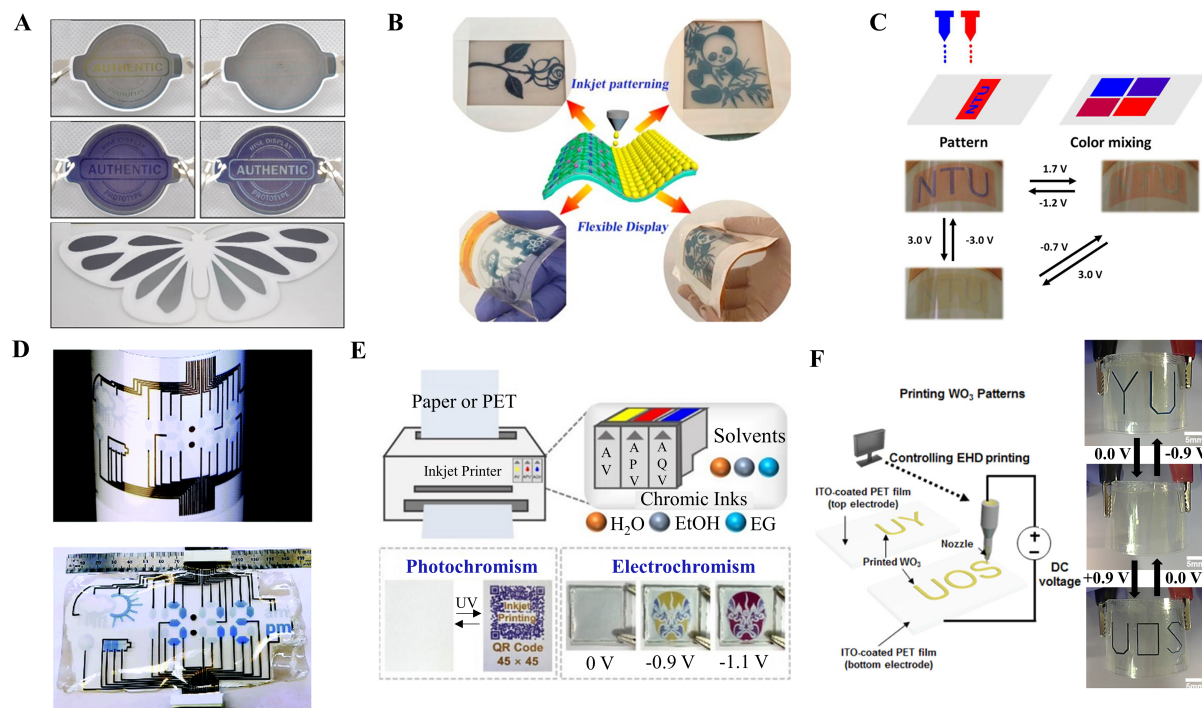


Figure 7. Recent advances in active layers pattern design with direct printing methods. (A) PEDOT/PPy and PEDOT/PProDOT-Me₂ in an authentic pattern showing reduced and oxidized states, and PEDOT/PProDOT-Me₂ with butterfly pattern with top half reduced state and lower half oxidized state. Reproduced with permission^[99]. Copyright 2023, Wiley-VCH; (B) Flexible ECD with patterns based on the inkjet-printed WO₃-PEDOT:PSS film on Ag grid-PET substrate. Reproduced with permission^[103]. Copyright 2018, Elsevier; (C) A multicolor flexible ECD based on the patterned film fabricating by different ratios of Fe/Ru-MEPE solution as a series of inkjet inks. Reproduced with permission^[22]. Copyright 2015, ACS Publications; (D) ECD presents quantities of display information with the inkjet-printed PEDOT:PSS pixels and Au electrode. Reproduced with permission^[79]. Copyright 2020, RSC Publications; (E) Flexible photochromic and ECD based on the inkjet-printed film using water-based additive-free chromic inks. Reproduced with permission^[63]. Copyright 2023, ACS Publications; (F) The fabrication process of patterned WO₃ film using electrostatic force-assisted dispensing printing methods and assembled as the voltage-tunable dual-image ECD. Reproduced with permission^[109]. Copyright 2020, ACS Publications. PEDOT: Poly(3,4-ethylenedioxythiophene); PPy: polypyrrole; PProDOT-Me₂: poly(dimethylpropylene-dioxythiophene); ECD: electrochromic display; PEDOT:PSS: poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; PET: polyethylene terephthalate; MEPE: metallo-supramolecular polymers.

Apart from the template-assisted and direct printing methods to design active layer patterns for display function, the emerging flexible electrochromic sensors combined with the biosensor and electrochromic electrodes as active layers also deliver optical information display to respond to the electrochemical sensing process. The two approaches for electrochromic sensors displaying information are showing color tone or color gradient [Figure 8A]^[111]. Limited literature reports the flexible electrochromic sensor, which comprises a lactate oxidase and osmium-polymer-based anode connected to a coplanar PB cathode printed on a PEDOT:PSS-PET flexible electrode. This flexible electrochromic sensor can display different lactate concentrations (0-10 mM) in the color length of the PB electrode [Figure 8B]^[60].

Electrolyte pattern design

Designing electrolyte patterns is a newly developed strategy for preparing flexible ECDs, which focuses on constructing free-standing patterned electrolyte gels on electrochromic films^[20,21,25,50]. An innovative self-powered flexible ECD with an electrolyte pattern has been designed by connecting the LiCl/PAM hydrogel-based pen with electrochromic PB or NiHCF film on ITO-PET substrate by the Al wire. The writing letters of “H/T” are simultaneously displayed (from colored to bleached) when the electrolyte pen contacts with Al wire and PB/NiHCF flexible electrodes, owing to the potential differences between electrodes driving for the

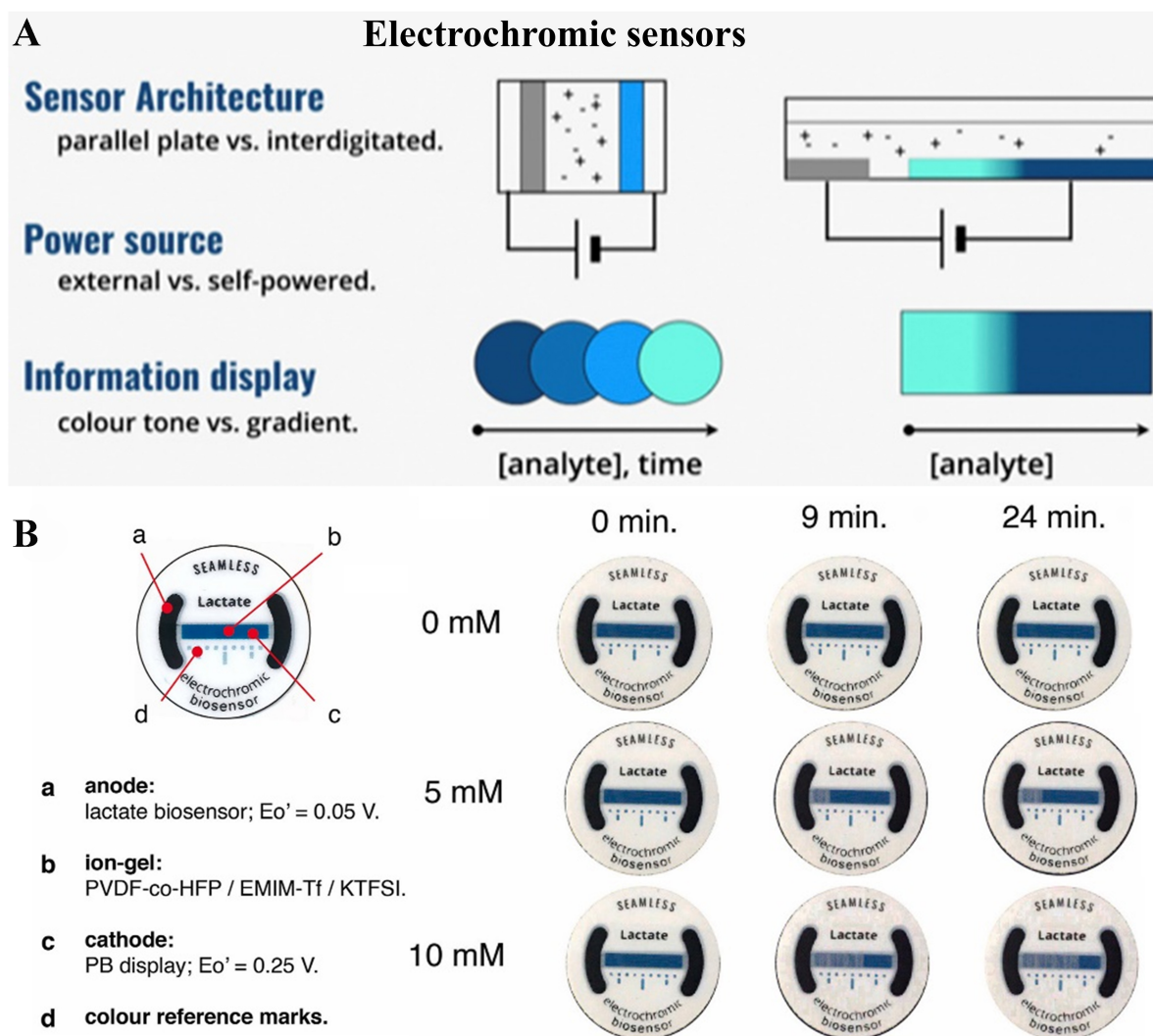


Figure 8. Recent advances in active layers design in electrochromic sensor displays. (A) The electrochromic sensors architecture, power source, and information display. Reproduced with permission^[11]. Copyright 2019, Elsevier; (B) Flexible electrochromic sensor's structure and evolution of the display color change as a function of lactate concentrations^[60]. Reproduced with permission. Copyright 2021, Elsevier.

self-bleached electrochromic redox reaction. After that, the information can be restored by the oxidation reaction in air [Figure 9A]^[21]. The demonstrated display with a repeatable display function and energy-efficient properties can provide new insights into developing future flexible electronics. UV-curable electrolytes enable a feasible and rapid transformation from precursor to gel, offering tremendous possibilities for using direct printing methods to fabricate complex patterned electrolytes in flexible ECDs. Figure 9B exhibited the electrolyte patterns with “K, Y, N” letters by printing H_2SO_4 -based UV-curable electrolytes on ISHCP-coated SEBS stretchable substrate, where the stretchable ECDs showed stable electrochromic performances under repeat stretching conditions of 100% strain^[50]. Sun *et al.* creatively proposed a simplified 3D direct printing technique for fabricating patterned DES-based gel electrolytes. By assembling with flexible WO_3 and NiO film on ITO-PEN substrates, the fabricated ECD showed a delicate blue-colored pattern under -3.0 V [Figure 9C]^[20]. The above strategies significantly increase the processibility of electrolytes and promote the advancement of flexible electrochromic devices. However,

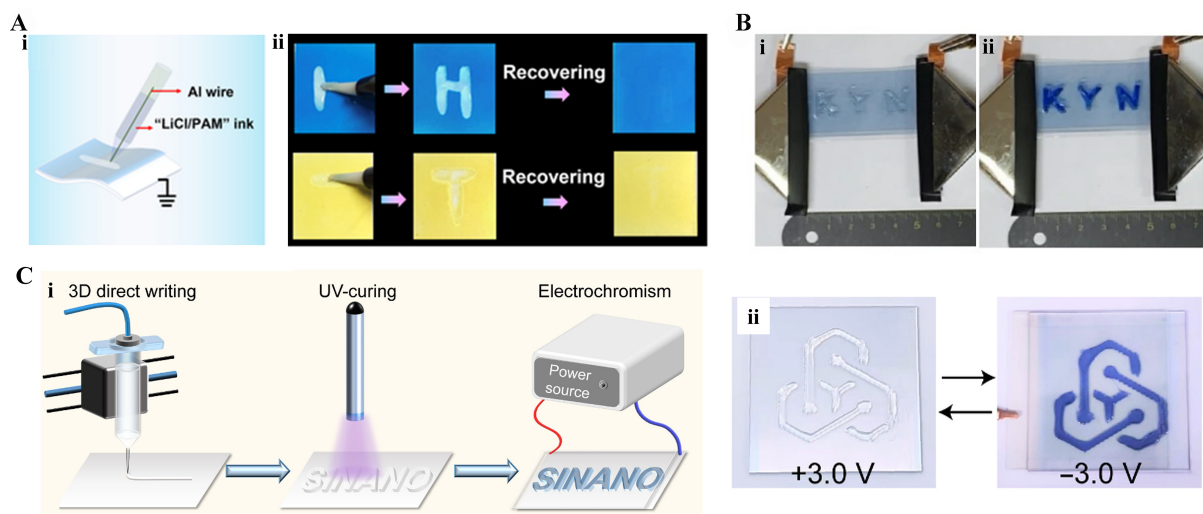


Figure 9. Recent advances in electrolyte pattern design. (A) Flexible and self-powered ECD with electrolyte pattern by connecting the hydrogel pen with EC film by Al wire. Reproduced with permission^[21]. Copyright 2023, SPJ; (B) Stretchable ECD with screen-printed patterned electrolyte. Reproduced with permission^[50]. Copyright 2020, Springer Nature; (C) Flexible ECD with 3D direct printed patterned UV-curable electrolyte. Reproduced with permission^[20]. Copyright 2022, Wiley-VCH. ECD: Electrochromic display; EC: electrochromic; UV: ultraviolet.

current electrolyte patterning design mainly relies on UV curing-based methods, which has potential issues with precise controllability of pattern resolution due to the dimensional changes after curing. Besides, the limited penetration depth of UV light may also influence the uniformity of the patterned electrolyte, especially for the thicker one.

ECD pixel design

Generally, the pixelated ECDs are composed of one or more subpixels with the designed arrangement and the controlled external circuit. According to the types of external circuits, the pixelated ECDs can have passive matrix, active matrix, and point-to-point driving modes^[87]. Based on three driving modes, we will focus on the design strategies of pixel arrangement and subpixel device structure construction in flexible and stretchable pixelated ECDs. Subsequently, we will elucidate the multifunctionality of these highly integrated flexible ECD pixelated displays.

Firstly, the passive matrix-based ECDs are constructed by the row and column electrodes, where the electrochromic subpixels are located in the intersection. With this matrix architecture, achieving a high color contrast while mitigating cross-talk remains a primary objective^[112]. In current flexible and stretchable ECDs, researchers have adopted the approaches for fabricating the polyelectrolyte pixels and using the V/3 addressing protocols to prevent the unwanted electrochromic phenomenon for inactive subpixels. In this case, the flexible [Figure 10A]^[113] and stretchable [Figure 10B]^[114] PEDOT:PSS-based passive matrix-based ECDs were successfully prepared with addressability and low cross-talk under mechanical conditions. Secondly, the active matrix-based ECDs greatly solve the cross-talk issues because each electrochromic pixel is combined with one transistor, which can turn off the unwanted current of the inactive subpixel^[24,112,115]. Kim *et al.* have successfully fabricated a 6×6 stretchable active matrix electrochromic array by introducing an n-type field-effect transistor (FET) with the P3MT/PB ECD pixel using liquid metal interconnection on the ITO-PET substrate, which showed a high-resolution display capability even under bending conditions [Figure 10C]^[61]. To further improve the manufacturing feasibility of the display owing to the additional transistor component, a fully screen-printed effective process was adopted to prepare the SWCNT thin film

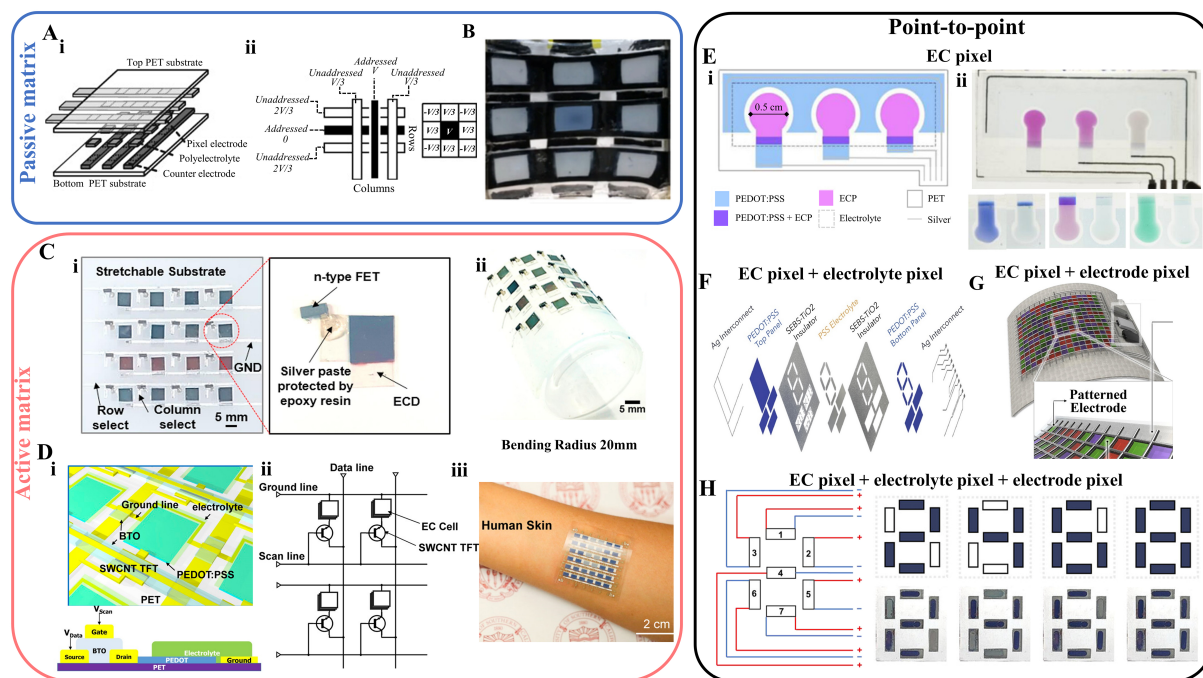


Figure 10. Recent advances in ECD pixels design. Device demonstration of (A) flexible and (B) stretchable PEDOT:PSS-based passive matrix ECDs. Reproduced with permission^[113,114]. Copyright 2023, ACS Publications. Copyright 2013, Elsevier; (C) 6 × 6 stretchable active matrix ECD by integrating n-type FET with the P3MT/PB ECD pixel using liquid metal interconnection. Reproduced with permission^[61]. Copyright 2019, Elsevier; (D) A fully screen-printed active-matrix PEDOT:PSS-based ECD fabricating with SWCNT thin film transistor. Reproduced with permission^[116]. Copyright 2016, ACS Publications; (E) Lateral ECD pixels based on three electrochromic polymers with the same PEDOT:PSS counter electrode. Reproduced with permission^[119]. Copyright 2021, ACS Publications; (F) numerical electrochromic pixelated display constructed by the same PEDOT:PSS counter electrode and EC and electrolyte pixels. Reproduced with permission^[65]. Copyright 2021, Elsevier; (G) multicolor ECD with electrochromic polymer pixels and patterned Au-PET electrode. Reproduced with permission^[117]. Copyright 2020, Elsevier; (H) Eight digital flexible ECDs with seven separated ECD pixels. Reproduced with permission^[84]. Copyright 2023, Wiley-VCH. ECD: Electrochromic display; PEDOT:PSS: poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; FET: field-effect transistor; P3MT: poly(3-methylthiophene); PB: Prussian blue; SWCNT: single wall carbon nanotube; EC: electrochromic; PET: polyethylene terephthalate.

transistor (TFT) and PEDOT:PSS-based electrochromic cell for an active matrix-based ECD [Figure 10D]^[116]. This flexible ECD exhibited satisfactory electrochromic properties and can be well laminated in human skin, which shows promising possibilities for low-cost and scalable flexible displays. Thirdly, point-to-point driving modes are the most prevalent way in flexible and stretchable pixelated ECDs due to their limited pixel number and small subpixel size in current development status. Using this method, every electrochromic pixel is individually controlled with corresponding external bridged circuits, which can provide more possibility of constructing various subpixel device structures without considering the cross-talk issues^[49,84,117,118]. For example, the lateral ECD pixels of three electrochromic polymers controlled by the Ag circuit on a PET substrate have been designed with the same screen-printed PEDOT:PSS counter electrode and Li⁺ electrolyte. This flexible pixelated ECD can switch the corresponding pixel's optical properties in seconds to minutes [Figure 10E]^[119]. Besides, the numerical electrochromic pixelated display with layered structure has been fabricated with top PEDOT:PSS panel electrode, where the EC and electrolyte pixels were individually separated by SEBS-TiO₂ insulator matrix. The interconnection of each electrochromic pixel used the screen-printed Ag circuit to achieve the point-to-point controlling mode [Figure 10F]^[65]. Koo *et al.* prepared a dynamic multicolor ECD where the electrochromic polymer pixels and ionic-TPU solid electrolyte film were sandwiched between the patterned Au-PET and ITO-PET electrodes. The user can control the selection of independent active pixels for arbitrary information display, paving new ways for adaptive camouflage applications [Figure 10G]^[117]. Moreover, seven separated ECD

pixels fabricated an eight-digital flexible ECD, where the electrochromic layer, electrolyte, and electrode were in the same pixel arrangement. Each flexible ECD subpixel's positive and negative electrodes were connected with copper foil so that they were controlled individually showing different numbers [Figure 10H]^[84]. Besides, a novel multicolor fiber-based ECD has been fabricated by independently woven 1,1'-Diethyl-4,4'-bipyridilium dibromide (EtVio)-, 1-heptyl-4-(4-pyridyl)pyridinium bromide (HVio)- and 1,1'-bis(p-cyanophenyl)-4,4'-bipyridilium dichloride (p-CVio)-based electrochromic fibers into nylon yarns forming patterns. This display as a smart wearable application has shown its promising potential for adaptive camouflage in response to changes in the surrounding environment^[47]. In conclusion, the above design strategies of device structures provide an adequate research foundation for high-performance flexible and stretchable ECDs. Due to their easier processibility, flexible and stretchable pixelated ECDs are primarily developed with point-to-point modes. This approach arises from significant limitations related to pixel size and number. To improve the current situation, the processing difficulty of active/passive matrix-based ECDs with flexible substrates should be addressed from three aspects: (1) The flexible interconnect should be firmly prepared to accommodate the bending/stretching of the substrate without losing electrical conductivity; (2) A thinner patterned electrolyte with precise alignment of the patterned flexible electrode is expected to address the signal crosswalk issue; (3) Develop reliable, high-performance transistors on a flexible substrate to integrate well with electrochromic pixels. Addressing these limitations requires ongoing research and development in materials science, device engineering, and fabrication technologies to enhance the performance, durability, and scalability of flexible and stretchable ECDs.

Featured by the point-to-point driving mode, these flexible ECDs, which exhibit dynamic information, are promising to be integrated with various sensors to visualize corresponding signals through color changes of resultant electrochromic pixels. These signals, including the temperature^[28], strain^[28,51], Na⁺ and Vitamin C concentration^[49,65], pressure^[120], and energy storage^[27], have been demonstrated in current flexible ECDs. For instance, the flexible ECD pixels function as energy storage devices integrated with the temperature and strain sensors, which power the sensors and detect the bio-signals of skin temperature and wrist bending angle with the help of a microcontroller [Figure 11A]^[28]. Besides, Yin *et al.* have successfully integrated various chemical sensors with individual flexible electrochromic pixels, such as the PH sensor, Na⁺ and Vitamin C sensors [Figure 11B]^[65]. The corresponding working voltage of the chemical sensors obtained by different chemical substance concentrations was converted into a logic output. After programming by the microcontroller, the obtained voltage value of the electrochromic pixelated display is associated with the display content, visualizing the corresponding chemical signals. Moreover, the pressure of the pen tip can be detected using the novel vertically integrated device with the PANI-based pixelated ECD module on top and PVDF-based patterned piezo electrical pressure sensor module on bottom. Specifically, the piezoelectric signal collected by the PVDF-based pressure sensor was amplified by an amplifier circuit and converted into a direct current (DC) signal by a bridge rectifier, which was then applied to the pressed PANI pixels for electrochemical reaction and showed corresponding color change [Figure 11C]^[120]. Similarly, the resistance change upon stretching in the strain sensor can trigger the "on/off" of the stretchable ECD pixel, showing the corresponding display response [Figure 11D]^[51]. Moreover, a voltage level can be directly visualized when the battery is connected with the multicolor electrochromic pixels showing the corresponding color of relevant voltage [Figure 11E]^[27]. The above interactive visual displays have greatly provided new avenues in future flexible IoT systems and intelligent robot applications.

CONCLUSIONS AND PROSPECTS

In this review, the latest research on flexible and stretchable ECDs has been summarized. We have provided an overview of the strategies for fabricating high-performance flexible and stretchable ECDs from three aspects, including flexible and stretchable electrodes, active layers, and electrolytes. For these strategies, the

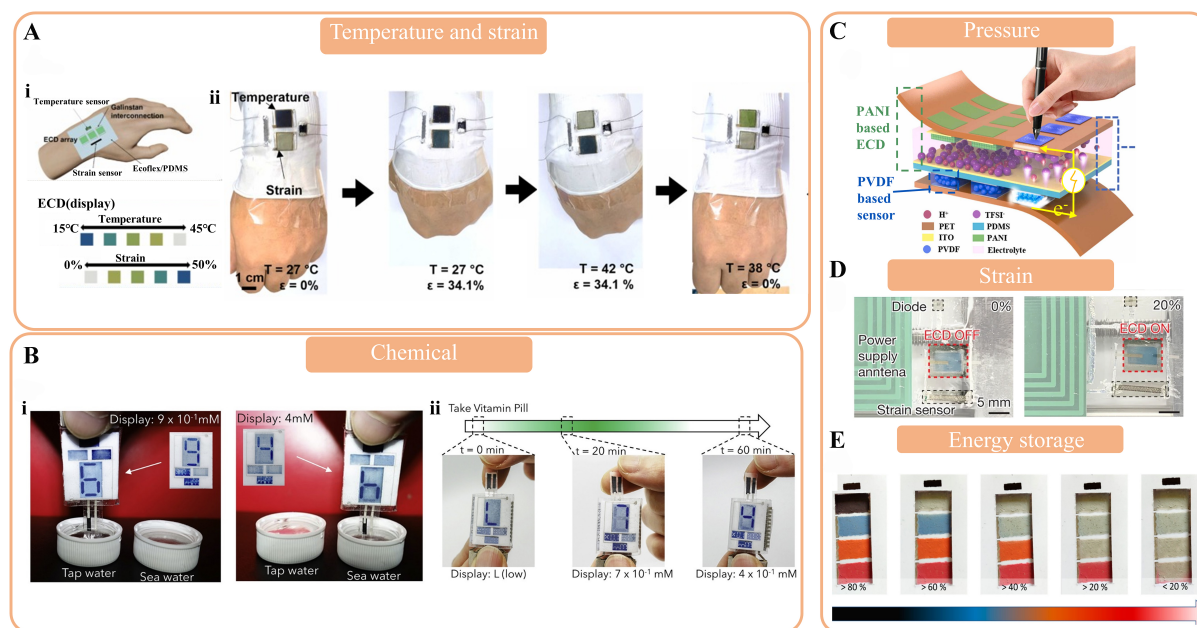


Figure 11. Recent advances in ECD pixels integrated with various sensors based on point-to-point mode. (A) ECD pixels integrate with the temperature and strain sensors detecting the skin's temperature and bending of wrist bending. Reproduced with permission^[28]. Copyright 2023, Elsevier; (B) ECD pixels integrate with various chemical (Na^+ and Vitamin C) sensors visualizing corresponding chemical concentrations. Reproduced with permission^[65]. Copyright 2021, Elsevier; (C) Schematic diagram of a device integrating PANI-based ECD pixels with PVDF-based piezo electrical pressure sensor. Reproduced with permission^[120]. Copyright 2022, Elsevier; (D) ECD pixels triggered by the stretching state because of the resistance change. Reproduced with permission^[51]. Copyright 2021, Springer Nature; (E) Multicolor ECD pixels indicate a battery meter with corresponding color states. Reproduced with permission^[27]. Copyright 2021, ACS Publications. ECD: Electrochromic display; PANI: polyaniline; PVDF: polyvinylidene fluoride.

streamlined and feasible preparation methods are preferentially considered for constructing every layer of the device to ensure high compatibility between layers and obtain excellent mechanical, chemical, and thermal stability of devices. The selection of corresponding materials for each layer has also been comprehensively elaborated. In addition, recent advances in these flexible and stretchable ECDs have been systemically classified into four types: electrode pattern design, active layer pattern design, electrolyte pattern design, and electrochromic devices pixels design. The relevant processing methods for constructing the above patterns and device structure for corresponding driving modes (passive matrix, active matrix, and point-to-point) in pixelated ECDs have been summarized. The highly integrated flexible ECD pixelated displays with different sensors for visualizing various signals are concluded. Besides, some limitations have been pointed out: the limited resolution of the active layer pattern due to the precision of the template, the difficulty controlling electrolyte uniformity and resolution due to the UV curing method, and the limited pixel size and number of the pixelated ECDs. Despite remarkable progress and advancements achieved by numerous researchers over the last few years, gaps remain in realizing manufacturable, commercialized, scalable, and robust flexible ECDs that meet market demands. To further promote the progress of the flexible and stretchable ECD technology and address the current limitations and challenges, researchers need to make great efforts in the following aspects: First, from the perspective of materials selection, the emphasis lies on the preference for low-cost, low-power, green, and biodegradable materials. This focus addresses environmental concerns and contributes to cost-effectiveness and sustainability in production processes. Second, in terms of processing methods, facile approaches that simultaneously achieve display patterning and materials microstructure construction with simplified structures are desirable. Such methods streamline manufacturing processes, leading to increased efficiency and scalability in the production of flexible displays. Third, regarding the applications, improving scalability and increasing the matrix/pixel

number of flexible ECD electronics is necessary, thereby enhancing their information capacity. Moreover, there is a growing demand for full-color displaying capabilities in flexible display electronics, alongside the integration of energy-saving traits to improve overall efficiency and sustainability. These prospects can significantly contribute to the evolution of flexible and stretchable ECDs, making them increasingly viable for diverse applications in various industries.

DECLARATIONS

Authors' contributions

Conceptualization and supervision: Lee PS

Prepared the figures and wrote the article: Wu W

Revised and corrected the article: Lee PS, Wu W

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Not applicable.

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Conflicts of interest

Both authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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