



Smart electrochromic devices based on reversible (non-)metal electrodeposition

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Abstract

Smart electrochromic devices based on reversible electrodeposition/dissolution have attracted increasing attention owing to their large optical modulation, simplified device configuration, and potential multifunctionality. Different from conventional electrochromic systems relying on ion insertion/extraction in pre-deposited films, these devices offer dynamic optical regulation through reversible electrochemical deposition and dissolution at the electrode/electrolyte interface. In this review, recent progress in reversible electrodeposition-based electrochromic devices is summarized with emphasis on three representative families: reversible metal, iodine, and MnO₂ electrodeposition. For metal-based devices, deposition morphology, alloy composition, electrolyte regulation, and electrode surface modification are key factors determining optical contrast, color tunability, and cycling stability. For iodine systems, the suppression of the polyiodide shuttle and dead iodine formation is central to achieving stable neutral-color modulation. For MnO₂ systems, reversible Mn²⁺/MnO₂ conversion enables wide ultraviolet (UV)-visible modulation and multicolor states, while dead manganese residues and non-uniform deposition remain major challenges. Finally, the common design principles and system-specific trade-offs are compared and discussed, followed by an outlook on practical reliability, large-area fabrication, adaptive thermal regulation, and multifunctional integration. This review aims at inspiring future endeavors towards implementation of reversible (non-)metal electrodeposition-based smart electrochromic devices.

Keywords: Electrochromic devices, reversible metal electrodeposition, reversible non-metal electrodeposition, smart windows, multicolor states

1. Introduction

Electrochromic devices (ECDs) can reversibly regulate optical transmittance, reflectance, or color under an external electrical stimulus, making them promising candidates for smart windows, adaptive displays, energy-saving buildings, and multifunctional optical systems^[1,2]. Conventional electrochromic materials (ECMs), such as transition metal oxides, conductive polymers, viologens, and Prussian blue analogues, have been widely studied, yet most of them depend on ion insertion/extraction in pre-deposited electrochromic (EC) films. Their optical performance is therefore strongly limited by the intrinsic properties, thickness, and ion transport behavior^[1,2]. In contrast, reversible electrodeposition-based ECDs operate through a different working principle: active species dissolved in the electrolyte are electrochemically deposited onto transparent electrodes during coloration and dissolved back during bleaching^[3-5]. This mechanism simplifies device fabrication and allows optical modulation to be controlled by the amount, morphology, composition, and spatial distribution of the deposited phase.

As schematically illustrated in [Figure 1](#), reversible electrodeposition-based ECDs generally share a similar sandwich-type configuration composed of two transparent conductive electrodes (TCEs) and an ion-conducting electrolyte layer containing dissolved electroactive species. Under an external voltage, these active species migrate through the electrolyte and undergo reversible interfacial redox reactions at the electrode/electrolyte interface, leading to the in-situ formation or dissolution of optically active deposits on the TCE surface and thereby enabling dynamic optical switching. For reversible metal electrodeposition (RME), metal cations such as Cu²⁺, Zn²⁺, or Bi³⁺ are reduced and deposited according to $M^{n+} + ne^- \rightleftharpoons M$, and the resulting metallic films,



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nanoparticles, or alloyed deposits can induce strong light blocking, reflection, neutral-color tinting, or even localized surface plasmon resonance (LSPR)-related multicolor modulation depending on their morphology, size, and composition^[3,4]. In contrast, reversible iodine electrodeposition is based on the interconversion among I^- , I_3^- , and I_2 species, in which iodide ions are electrochemically oxidized to surface-confined iodine species during coloration and reduced back during bleaching^[6,7]. Owing to the broadband visible-light absorption of iodine/polyiodide species, such systems are particularly suitable for neutral-color optical modulation and complementary EC windows. Different from both metal and iodine systems, reversible MnO_2 electrodeposition proceeds through Mn^{2+}/MnO_2 conversion, where Mn^{2+} ions are oxidized to form MnO_2 deposits during coloration and dissolved back during bleaching^[8,9]. The gradually formed MnO_2 layer can provide broad ultraviolet (UV)-visible modulation and multiple optical states, and can also be integrated with other EC or battery-like redox processes to realize multifunctional devices. Therefore, although these three representative mechanisms share the common device-level feature of voltage-driven reversible deposition/dissolution within a sandwich-structured ECD, they differ substantially in deposited species, optical regulation mechanisms, and application scenarios. RME-based ECDs are attractive for high-contrast smart windows, privacy protection, reflective regulation, and multicolor displays^[3-5]. Iodine-based ECDs are advantageous for color-neutral modulation and simplified complementary architectures^[6,7]. MnO_2 -based ECDs show promise in multilevel spectral regulation, multicolor electrochromism, and EC energy-storage integration^[8-10].

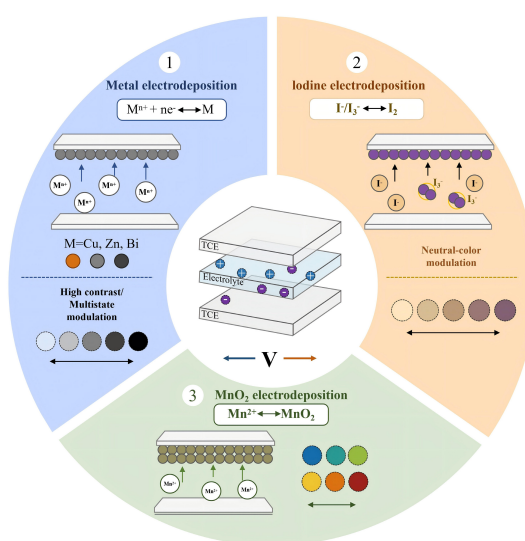


Figure 1. Schematic illustration of three representative reversible electrodeposition-based electrochromic mechanisms, including reversible metal electrodeposition, iodine electrodeposition, and MnO_2 electrodeposition. The central panel shows the typical sandwich-type device structure composed of two transparent conductive electrodes and an electrolyte layer. The surrounding sections summarize the corresponding redox reactions and optical modulation features of the three mechanisms. TCE: transparent conductive electrode.

ECDs based on related routes, including reversible Br^-/Br_3^- conversion and electrically regulated dynamic metal-ligand interactions (DMLI), have also been reported^[11,12]. However, these two types of ECDs are not included in this review due to the limited performance of Br^-/Br_3^- -based ECD and the complicated working process of DMLI-based ECD. Accordingly, the following discussion concentrates on RME, reversible iodine electrodeposition (RIE), and reversible MnO_2 electrodeposition^[6-10].

From the development roadmap point of view, this field has progressed from early neutral-color transparent-to-opaque metal windows^[3,4], to alloy-assisted and LSPR-based multicolor regulation^[13-15], and subsequently to large-area electrolyte and interface engineering^[16-18]. More recently, iodine- and MnO_2 -based systems have expanded reversible electrodeposition toward complementary electrochromism, multilevel spectral regulation, and electrochromic energy storage^[6-10], while adaptive radiative cooling and solar-heating devices have further extended its application scope toward dynamic thermal management^[19,20].

Although these systems differ in chemical reactions and optical responses, their performance is governed by ion transport, interfacial nucleation or redox conversion, deposit growth, and reverse dissolution. Consequently, non-uniform deposition, incomplete dissolution, irreversible residues, slow switching kinetics, and large-area coloration inhomogeneity recur across the three systems, although they originate from different chemical and interfacial processes^[6,9,16,17]. This review therefore compares the three representative families within a unified mechanism-challenge-strategy framework, with particular attention to electrolyte regulation, interfacial reaction control, dissolution reversibility, device architecture, balanced performance evaluation, and practical reliability. It is envisioned that reversible (non-)metal electrodeposition will bring new possibilities in ECDs towards reversible, durable, scalable and multifunctional photothermal regulations.

2. Reversible Electrodeposition-Based Electrochromic Devices

To facilitate cross-system comparison, [Section 2.1](#), [Section 2.2](#) and [Section 2.3](#) follow a common analytical sequence covering the working mechanism and optical characteristics, principal mechanistic challenges, regulation strategies, and application-oriented assessment. The shared design principles and system-specific trade-offs are subsequently compared in [Section 2.4](#).

2.1 Reversible metal electrodeposition-based electrochromic devices

RME-based devices (RMEDs) have attracted broad attention owing to their large optical modulation achieved by the reversible plating/stripping of metals^[3,4]. Unlike conventional ECDs that rely on ion insertion/extraction mechanisms and are inherently constrained by the optical properties of prefabricated EC films, the optical responses of RMEDs are predominantly governed by the morphology, chemical composition, and spatial distribution of in-situ electrodeposited metal layers^[21-23]. Thanks to this unique working principle, RMEDs exhibit remarkable application potential in broadband solar spectral regulation, neutral-color tinting, high-contrast dynamic smart windows, and advanced building thermal management^[3,5,20,24].

To further elucidate the working mechanisms of RMEDs, it is essential to examine the mechanisms of RMEDs based on deposited metal species and their corresponding optical modulation principles. For single-metal RMED systems, reversible optical switching is achieved via electrochemical redox reactions: metal ions in the electrolyte are reduced and electrodeposited onto the electrode surface to form opaque metal films during coloration, while the deposited metal is reversibly oxidized and stripped back into the electrolyte during bleaching. As illustrated in [Figure 2a,b](#), Zn-based RMEDs are typical single-metal RMED systems that enable rapid and reversible switching between a highly transparent state and an opaque dark state via controllable Zn plating/stripping. For such chemically simplified single-metal systems, electrolyte formulations play a decisive role in device performance, as they profoundly modulate Zn nucleation behavior, deposited film compactness, parasitic side reactions, and long-term cycling durability^[25-27].

When metal electrodeposition is confined to the nanoscale, single-metal RMEDs can achieve sophisticated color modulation via LSPR effects^[28]. As depicted in [Figure 2c](#), Ag-deposition-based RMEDs can realize multiple optical states, including mirror, black, magenta, cyan, and yellow, by precisely tailoring the size and morphology of Ag nanoparticles through voltage programming and electrode surface engineering^[13,29]. In this case, the optical response is no longer solely dependent on the total amount of electrodeposited metal, but is further dominated by the LSPR absorption bands of nanostructured metal films. Accordingly, single-metal LSPR-type RMEDs bridge the performance gap between conventional single-opacity modulation and high-value multi-color electrochromism, offering a viable strategy for advanced optical regulation.

Beyond single-metal systems, dual-/multi-metal RMEDs introduce additional regulation freedom through redox sequence, alloy composition, nucleation guidance, and metal-metal interaction. Cu/Zn dual-metal electrolyte systems outperform pure Zn single-metal RMEDs by enabling gradient CuZn reversible alloy electrodeposition (RAE), which delivers improved deposition uniformity ([Figure 2d](#)), faster switching response, reduced desolvation energy barriers, and enhanced cycling stability^[15]. Benefiting from the higher standard redox potential of Cu^{2+}/Cu relative to Zn^{2+}/Zn and favorable lattice compatibility between Cu and Zn, pre-deposited Cu nanostructures act as uniform nucleation sites to guide subsequent homogeneous Zn growth. Enabled by this staged electrodeposition mechanism, the optimized CuZn-RAE system exhibits a set of improved EC performances, including an ultra-low transmittance of 0.01% after 120 s deposition, neutral-color tinting (chroma $C^* = 5.8$), a large visible transmittance modulation of approximately 82%, fast switching at 550 nm ($t_c = 8$ s and $t_b = 18$ s), and a solar heat gain coefficient (ΔSHGC) modulation of 0.448^[15].

In addition to broadband opacity or mirror-like regulation, dual-metal systems can also enable LSPR-related multicolor optical states. As shown in [Figure 2e](#), Bi-Cu RMEDs can achieve purple transparent, purple mirror, yellow transparent, and yellow mirror states through controlled bimetal electrodeposition^[14]. These diverse chromatic states originate from tunable LSPR effects, which are determined by the morphological features and elemental ratio of electrodeposited Bi-Cu composite films ([Figure 2f](#)). Notably, dual-metal electrolytes are not merely simple mixtures of two metal ion species; their device performance is governed by the coupled modulation of redox sequences, alloy phase composition, nucleation dynamics, film morphological evolution, and stripping reversibility. Together with Cu/Ni and Cu/Sn bimetallic systems, the well-established Bi/Cu and Cu/Zn RMED platforms verify that dual- and multi-metal strategies have become effective approaches to optimize color neutrality, enrich multi-color modulation capability, extend memory effects, and improve long-term cycling durability^[14,15,30-32].

Despite their prominent advantages, the practical deployment of RMEDs remains hindered by several bottlenecks. First, precise control over metal electrodeposition on conductive electrodes is challenging. The synergistic effects of interfacial electric field heterogeneity, nucleation energy barriers, and ion concentration gradients frequently trigger uneven metal deposition, resulting in the formation of rough, isolated metal islands or dendritic microstructures^[21,25]. Such dendritic defects not only degrade optical uniformity but also impair the electrochemical reversibility of repeated plating/stripping^[26,27]. This morphology-dependent performance characteristic is even more critical for LSPR-type RMEDs, where the optical resonance bands and visualized color output are precisely determined by the size, geometry, interparticle spacing, and composition of electrodeposited metal nanostructures^[13,14,29,33]. Consequently, morphological regulation is pivotal for guaranteeing deposition uniformity, electrochemical

reversibility, and reproducible chromatic output in multi-color RMEDs. Second, the repeated deposition/dissolution of metal species may cause irreversible residues, resulting in slower switching kinetics and gradual optical decay, particularly in long-term cycling^[25–26]. Third, when RMEDs are scaled up, uniform metal deposition across the whole electrode becomes more difficult. The voltage drops between edge and center regions cause inhomogeneous electric field distribution and thereby non-uniform tinting. Overall, the practical implementation of high-performance RMEDs necessitates the simultaneous realization of excellent deposition uniformity, robust electrochemical reversibility, long-term operational stability, and reliable scalability.

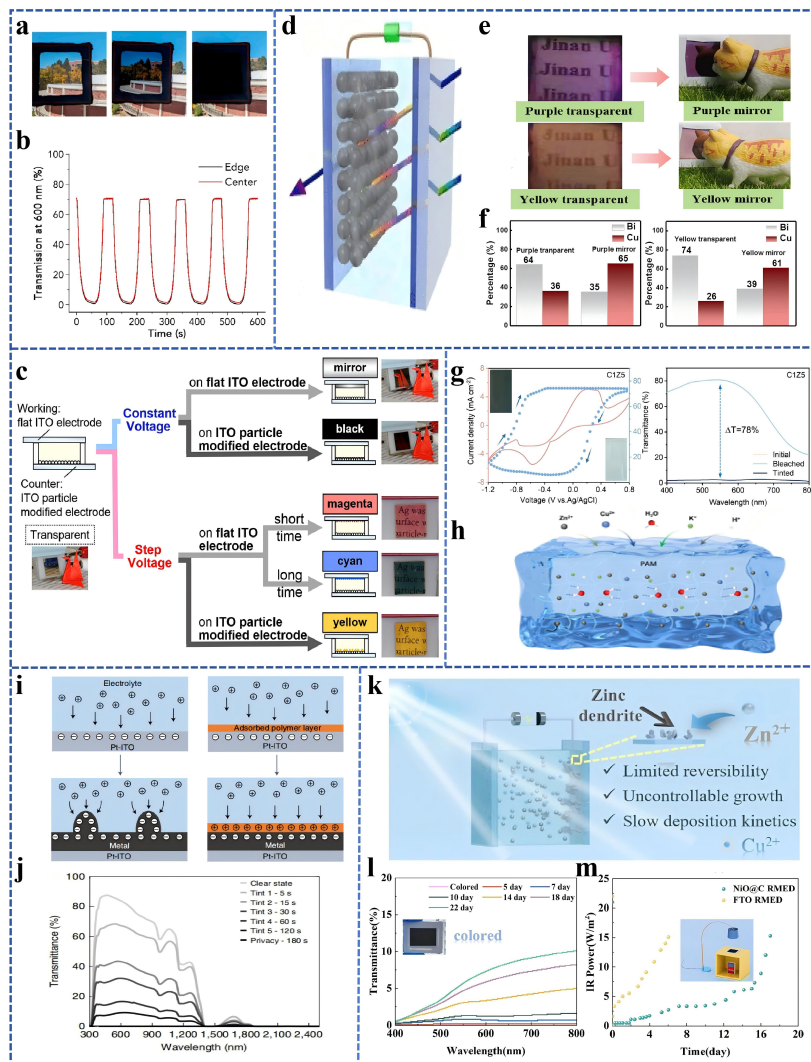


Figure 2. Representative RMED systems and regulation strategies for electrochromic smart windows. (a) Photographs of a Zn-based RMED showing reversible optical switching from transparent to dark states; (b) In situ transmittance evolution at 600 nm measured at the edge and center regions during repeated switching cycles. Reproduced from reference^[3]. CC BY 4.0; (c) Schematic illustration of an Ag deposition-based LSPR electrochromic device with mirror, black, magenta, cyan, and yellow optical states enabled by voltage-controlled Ag nanoparticle growth. Reproduced with permission from reference^[13]. Copyright © 2024 American Chemical Society; (d) Schematic illustration of a CuZn-based reversible alloy electrodeposition device. Reproduced with permission from reference^[15]. Copyright © 2014 American Chemical Society; (e) Photographs of Bi-Cu multicolor RMEDs showing purple transparent, purple mirror, yellow transparent, and yellow mirror states; (f) Atomic percentages of Bi and Cu in different Bi-Cu optical states. Reproduced with permission from reference^[14]. Copyright © 2022 Elsevier; (g) Electrochemical and optical properties of the CuZn hydrogel RMED, including cyclic voltammetry curves and transmittance spectra in the initial, bleached, and tinted states; (h) Schematic illustration of the PAM-based dual-metal CuZn hydrogel electrolyte containing Zn^{2+} , Cu^{2+} , H_2O , K^+ , and H^+ . Reproduced from reference^[18]. CC BY 4.0; (i) Schematic comparison of dendritic metal deposition on a bare Pt-ITO electrode and uniform metal deposition regulated by an adsorbed polymer layer; (j) Transmittance spectra of a polymer-regulated RMED from the clear state to different tinted and privacy states. Reproduced with permission from reference^[16]. Copyright © 2021 Springer Nature; (k) Schematic illustration of Zn dendrite formation, limited reversibility, uncontrollable growth, and slow deposition kinetics in conventional FTO-based RMEDs; (l) Optical memory behavior of the NiO@C-modified RMED in the colored state over different storage times; (m) Infrared power comparison between NiO@C RMED and FTO RMED, showing the thermal regulation effect of electrode surface modification. Reproduced with permission from reference^[17]. Copyright © 2026 Royal Society of Chemistry. RMED: reversible metal electrodeposition-based electrochromic device; LSPR: localized surface plasmon resonance; PAM: polyacrylamide; FTO: fluorine-doped tin oxide; ITO: indium tin oxide.

In addition to metal species engineering and LSPR-based multi-color regulation, electrolyte engineering serves as another critical strategy to modulate ion transport behavior, interfacial electrochemical reactions, and metal deposition/dissolution kinetics. Quasi-solid-state hydrogel electrolytes have been widely adopted to resolve the inherent electrolyte leakage and packaging challenges of liquid-based RMEDs for practical applications. Hydrogel electrolytes maintain efficient ion conduction while significantly enhancing the mechanical robustness and operational safety of devices. In particular, electrolyte pH regulation, though often underemphasized, is a key optimization parameter. The H^+ concentration directly affects metal-ion deposition kinetics, metal dissolution efficiency, byproduct generation, and parasitic side reactions such as hydrogen evolution^[18,25,26,34]. As shown in Figure 2g,h, after Cu^{2+} : Zn^{2+} ratio optimization and pH tuning, a hydrogel-based CuZn-RMED achieved a reduced deposition activation energy of 6.7 kJ mol^{-1} , large transmittance modulation of $\Delta T = 78\%$ at 550 nm, fast switching kinetics ($t_c = 15.0 \text{ s}$, $t_b = 24.0 \text{ s}$), over 90% ΔT retention after 3,000 cycles, and 18–33% simulated building energy savings across different climatic regions^[18]. Furthermore, the well-regulated hydrogen bonding networks bring anti-freezing capability (-20°C), while the free-standing hydrogel electrolyte offers patternability and recyclability in the CuZn-RMED, demonstrating an embodiment of cost-effective, sustainable, and energy-efficient RMED smart windows^[18]. Therefore, hydrogel and pH regulation should also be considered as electrolyte design factors, because they simultaneously affect ion transport, deposition/dissolution behavior, and side reactions. However, the balance among mechanical integrity, ionic conductivity, and side-reaction suppression remains central to quasi-solid-state RMEDs.

Polymer additives are also widely used in electrolyte engineering to suppress dendritic growth and improve deposition morphology. During metal electrodeposition, localized uneven ion flux can induce the growth of protrusions that evolve into dendrites^[16,26,35]. In this regard, polymer inhibitors such as polyvinyl alcohol (PVA) have been introduced into the electrolyte. As shown in Figure 2i, PVA can adsorb on the electrode surface, homogenize ion flux, and promote smooth metal deposition^[16]. As a result, the PVA-regulated RMED can achieve multiple tint states across the solar spectrum, a privacy state with visible transmittance $< 0.001\%$ within 3 min (Figure 2j), high infrared reflectance $> 70\%$, $\Delta T_{vis} = 0.76$, and $\Delta SHGC = 0.56$ ^[16]. More importantly, this polymer inhibitor strategy can increase the viscosity of electrolyte and reduce the voltage-drop between edge and center regions, facilitating the demonstration of a large-area (900 cm^2) RMED-based dynamic smart window^[16]. Polymer additives therefore provide a practical route for suppressing dendritic growth and improving large-area uniformity, although excessive adsorption or viscosity may also slow ion transport and compromise rapid switching.

Besides electrolyte engineering, electrode surface modification provides a direct way to regulate metal nucleation. Since electrodeposition begins at the interface between the transparent conductive substrate and the electrolyte, the surface properties of the electrode strongly affect the following growth behavior^[17,21]. On bare fluoride-doped tin oxide (FTO), metal nucleation may be non-uniform because of poor interfacial compatibility and large nucleation barriers. Bare FTO-based RMEDs can suffer from Zn dendrite formation, limited reversibility, uncontrollable growth, and slow deposition kinetics^[17].

To solve this problem, researchers have introduced interface layers such as NiO-based modification layers onto the electrode surface. A suitable interfacial layer can reduce lattice mismatch, improve nucleation uniformity, and guide more stable metal deposition^[17]. The NiO@C-FTO electrode showed a low metal plating/stripping activation energy of 7.32 kJ mol^{-1} , large optical modulation of $\Delta T = 65.1\%$, fast switching times of $t_c = 10.2 \text{ s}$ and $t_b = 7.2 \text{ s}$, and stable operation over 3,100 cycles^[17]. In contrast, Figure 2k shows that a low-lattice-mismatch NiO@C interface layer can guide more uniform metal deposition and improve plating/stripping reversibility. The assembled NiO@C-RMED also maintained $7\% < 10\%$ after 22 days in the tinted state and achieved a $\Delta SHGC$ of 0.38, confirming its advantage in bistable optical memory and thermal regulation (Figure 2l,m)^[17]. Therefore, surface modification is especially important for improving cycling durability, optical memory, and long-term device stability^[17]. Compared with electrolyte regulation, electrode surface modification acts more directly on the initial nucleation interface, while its long-term effectiveness is still constrained by the adhesion, chemical stability, and optical transparency of the introduced interfacial layer.

Overall, RMED smart windows have evolved from simple metal plating systems into highly engineered electrochemical devices. Their key challenges include uneven deposition, dendrite growth, limited reversibility, slow deposition kinetics, and poor large-area uniformity. Electrolyte engineering addresses these problems by regulating the ion environment, metal composition, polymer adsorption, hydrogel confinement, solvent structure, and pH conditions. Electrode surface modification addresses these problems by controlling the nucleation interface and improving interfacial compatibility. These two strategies are complementary rather than isolated. In future RMED development, the combination of optimized electrolytes and rationally modified electrodes will be essential for achieving large-area uniform coloration, long-term cycling stability, high solar modulation, and practical building-level energy control^[17,18,36].

2.2 Reversible iodine electrodeposition-based electrochromic devices

Recent years have witnessed the development of reversible non-metal electrodeposition (RnME)-based devices, including RIE systems. RIE-based devices (RIEDs) achieve efficient optical switching between a transparent state and a deep brown or near-black state through the reversible electrochemical conversion of the I^-/I_2 or $I^-/\text{polyiodide}$ redox couples. In contrast to conventional ion insertion/extraction-based ECDs, RIEDs eliminate the need for pre-deposited EC films, thereby substantially simplifying the device architecture. Moreover, owing to the broadband optical absorption characteristics of iodine species, an ideal neutral chromatic state

can be attained in the colored state. In terms of operating mechanism, RIEDs rely on the redox behavior of iodide ions at the electrode/electrolyte interface: under a positive bias, I^- is oxidized at the working electrode surface to form solid iodine, which is deposited and blocks the transmission of visible light; upon application of a reverse bias, the deposited iodine is reduced back to I^- and re-dissolves into the electrolyte, restoring the device to its transparent state.

Despite their high optical modulation and color neutrality, RIEDs remain limited by the polyiodide shuttle, dead iodine accumulation, and large-area coloration inhomogeneity^[6,37,38]. In aqueous electrolytes, oxidized I_2 can complex with excess I^- to form soluble I_3^- , which migrates away from the working electrode and promotes parasitic self-discharge; this speciation and shuttle pathway is well established in Zn- I_2 electrochemistry^[39–43]. In RIEDs, the resulting loss of surface-confined iodine weakens optical memory and cycling reversibility^[6,37]. In addition, the low electrical conductivity and weak interfacial contact of solid I_2 can generate electrically isolated dead iodine during repeated deposition/dissolution^[38,44]. At larger device areas, frame-type counter electrodes impose long lateral ion-transport paths and non-uniform electric-field distributions, leading to differences between edge and center coloration commonly found in different types of ECDs^[7,45].

To address the aforementioned challenges, researchers have undertaken systematic efforts spanning electrolyte engineering, synergistic optimization of the electrode/electrolyte interface, and complementary device design.

In the realm of electrolyte engineering, the suppression of I_3^- formation and diffusion constitutes the central strategy for achieving high reversibility. Li and coworkers^[6] pioneered the introduction of a highly concentrated $ZnCl_2$ water-in-salt electrolyte (WiSE, 29.5 M) into iodine-based EC systems. This approach substantially reduces the content of free water molecules, thereby inhibiting iodine dissolution and I_3^- generation (Figure 3a). Spectroscopic characterization and theoretical calculations have confirmed that I^- predominantly exists in the form of complex ions such as $[ZnI_3]^-$ and $[ZnCl_2 \cdot H_2O]^-$ within the WiSE environment. Consequently, the Gibbs free energy change for the conversion of I_2 to I_3^- is shifted from -0.27 eV in conventional dilute solutions to $+0.21$ eV, thermodynamically suppressing the formation of shuttle species (Figure 3b,c). Benefiting from the stable deposition of I_2 monomers, this system achieves a high optical contrast of 76.0% and exhibits a certain degree of self-healing capability. Nevertheless, the cycling stability remains constrained by the inherent low electrical conductivity of solid I_2 . To further overcome the interfacial deficiencies of solid I_2 , researchers have developed coordination-anchoring strategies based on ionic liquids. Meng *et al.*^[37] introduced 1-ethyl-3-methylimidazolium ($[EMIM]^+$) cations into the I^- -containing electrolyte (Figure 3d), leveraging the strong coordination interaction between $[EMIM]^+$ and I_3^- to generate insoluble $[EMIM]I_3$ complexes that anchor to the electrode surface, thereby fundamentally blocking the shuttle effect. Devices constructed using this approach exhibit negligible performance degradation after 10,000 cycles, unequivocally demonstrating the efficacy of coordination anchoring in suppressing the shuttle phenomenon (Figure 3e). Building upon this foundation, Chen *et al.*^[38] further introduced 1-methyl-3-propylimidazolium cation ($[MPI]^+$), which enables the concurrent generation of solid I_2 and liquid 1-methyl-3-propylimidazolium triiodide ($MPII_3$) during the oxidation process (Figure 3f). In contrast to the $[EMIM]^+$ system, liquid $MPII_3$ not only possesses markedly superior electrical conductivity relative to solid I_2 but also imparts an interfacial self-healing effect by filling the interstitial voids among solid I_2 particles, thereby significantly enhancing the electrical contact between the deposited layer and the FTO substrate and effectively eliminating the accumulation of dead iodine. Devices based on $MPII_3$ -WiSE exhibit coloring and bleaching response times of merely 6.9 s and 11.9 s (Figure 3g), respectively, and retain over 100% of their initial optical contrast after 20,000 cycles (Figure 3h), achieving an excellent balance between cycling stability and switching speed. Together, these studies demonstrate that regulating iodine speciation, deposit conductivity, and interfacial adhesion is central to simultaneously suppressing shuttle, dead iodine formation, and kinetic degradation in RIEDs.

Beyond electrolyte regulation, synergistic optimization of the electrode/electrolyte interface represents an equally effective approach for reducing reaction overpotentials and improving kinetic reversibility. Building upon the use of the ionic liquid $MPII_3$ as the electrolyte, Guo and coworkers^[44] further introduced a RuO_2 catalytic modification layer onto the surface of the FTO working electrode (Figure 3i). In this synergistic configuration, the $MPII_3$ electrolyte provides a stable chemical environment for the I^-/I_3^- redox reaction by virtue of its excellent ionic transport properties and coordination-anchoring effect on iodine species, while the RuO_2 modification layer substantially lowers the energy barrier for interfacial charge transfer through catalytic pathways. Density functional theory calculations revealed that RuO_2 reduces the energy barrier for $I^- \rightarrow I_3^-$ oxidation from 0.82 eV to 0.55 eV (Figure 3j), and linear sweep voltammetry measurements demonstrated a corresponding overpotential reduction of 0.42 V (Figure 3k). Owing to the synergistic interplay between the electrolyte and the electrode, the onset oxidation potential of the device is lowered to 1.52 V (vs. Zn^{2+}/Zn), effectively circumventing deleterious oxygen evolution side reactions in the high-voltage regime while concurrently enhancing both energy efficiency and cycling durability. This work underscores that the rational integration of electrolyte engineering with deliberate electrode surface modification can synergistically unlock the potential of iodine electrodeposition from both thermodynamic and kinetic perspectives.

At the device architecture level, Guo and coworkers^[7] proposed a WO_3 - I_2 complementary ECD (Figure 3l). In this design, a WO_3 thin film prepared by magnetron sputtering serves as the working electrode, while bare FTO glass functions as the counter electrode, with a $ZnCl_2$ water-in-salt electrolyte containing ZnI_2 interposed in between. During the coloring process, the WO_3 electrode undergoes reduction concomitant with Zn^{2+} intercalation, transitioning from transparent to blue, while I^0 in the electrolyte is simultaneously

oxidized at the counter electrode FTO surface to form I_2 , which deposits stably thereon. The spectral complementarity of these two coloring effects endows the device with a near-zero transmittance, fully black appearance in the colored state, thereby achieving ideal color neutrality. This complementary architecture eliminates the need for a separately prepared counter electrode EC layer, substantially simplifying the device configuration. Furthermore, since I_2 deposition occurs directly on the counter electrode surface, the ion transport distance is effectively shortened, facilitating uniform modulation over large areas. The WO_3 - I_2 complementary device achieves an average optical modulation amplitude of 70.7% across the visible spectrum, with nearly identical modulation values at the edge and center regions (Figure 3m). After 100 cycles, the retention of optical modulation reaches 88.5% (Figure 3n), and uniform coloration has been successfully demonstrated in a large-area device measuring $10 \times 10 \text{ cm}^2$. This work provides a novel paradigm for the construction of practical complementary EC smart windows.

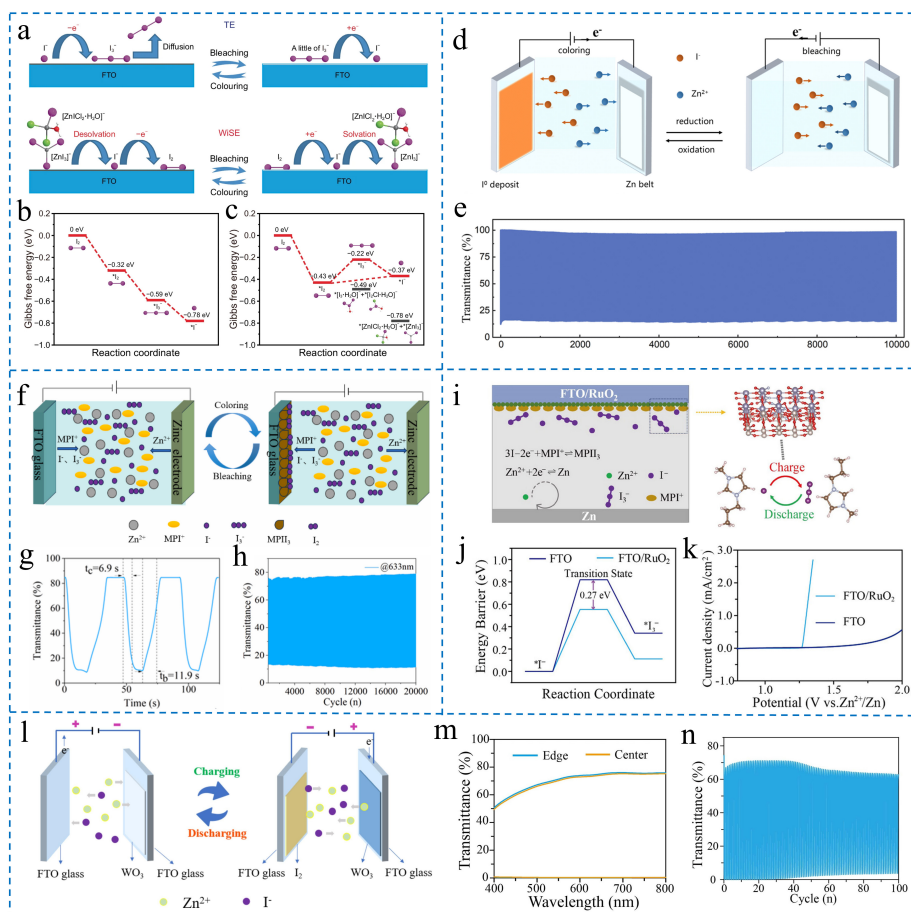


Figure 3. (a) Schematic diagram depicting the EC reaction mechanism in the TE and WiSE. The Gibbs free energy diagrams of I_2 reduction reaction in the (b) TE and (c) WiSE. The asterisk "*" represents the active site. Reproduced from reference^[6]. CC BY 4.0; (d) Schematic diagrams showing the device's working principle; (e) Cycling performance within 10,000 cycles of the I^-/I_2 conversion system in the ZIE electrolyte. Reproduced with permission from reference^[37]. Copyright © 2025 Royal Society of Chemistry; (f) Schematic diagram of the operating principle of an electrochromic device; (g) In-situ transmittance at 633 nm under alternating coloring/bleaching voltages of 2.3 V and 0.1 V; (h) In-situ transmittance curve after 20,000 coloring/bleaching cycles at 633 nm. Reproduced with permission from reference^[38]. Copyright © 2026 Elsevier; (i) Schematic illustration of the Zn-MPII₃ electrochromic battery; (j) Calculated reaction energy barriers for the I^-/I_3^- redox conversion on bare FTO and FTO/RuO₂ current collectors; (k) LSV comparison of Zn-MPII₃ batteries constructed with FTO and FTO/RuO₂ current collectors. Reproduced with permission from reference^[44]. Copyright © 2025 Wiley-VCH; (l) Schematic of the operating mechanism of the WO_3 - I_2 complementary electrochromic device; (m) Optical transmittance of the device measured at the center and edge positions in both colored and bleached states; (n) In situ transmittance spectra of the WO_3 - I_2 electrochromic device at 633 nm over 100 coloring/bleaching cycles. Reproduced with permission from reference^[7]. Copyright © 2025 Elsevier. EC: electrochromic; WiSE: water-in-salt electrolyte; MPII₃: 1-methyl-3-propylimidazolium triiodide; FTO: fluoride-doped tin oxide; LSV: linear sweep voltammetry.

Overall, RIEDs offer high optical modulation and neutral-color output through chemically simple I^-/I_2 or I^-/I_3^- conversion, but their practical performance is controlled by iodine speciation, deposit conductivity, interfacial adhesion, and transport uniformity. WiSEs suppress soluble polyiodide formation, coordinating cations immobilize I_3^- , conductive liquid iodine phases improve interfacial contact, catalytic layers accelerate charge transfer, and complementary architectures shorten ion-transport paths. These strategies are effective but introduce trade-offs in electrolyte cost, viscosity, chemical compatibility, and device encapsulation, which should be considered together with wide-temperature operation and long-term sealing reliability.

2.3 Reversible MnO₂ electrodeposition-based electrochromic devices

Reversible MnO₂ electrodeposition has emerged as another electrochromic pathway in which Mn²⁺ is oxidized to MnO₂ during coloration and the deposited oxide is reduced and dissolved during bleaching. Unlike conventional ion-insertion electrodes, the optically active layer is generated in situ, simplifying device fabrication and allowing its thickness to be regulated by the applied voltage and deposition time^[8–10]. MnO₂-based reversible oxide electrodeposition devices (ROEDs) can provide broad UV-visible modulation, multilevel coloration, and compatibility with battery-like redox processes, although their response and durability remain strongly dependent on electrolyte acidity, manganese-intermediate chemistry, and device architecture.

Mechanistic guidance for reversible MnO₂ deposition has also been obtained from aqueous Zn-MnO₂ batteries. Zhuang *et al.*^[46] developed an acidic hydrogel electrolyte composed of poly(2-acrylamido-2-methylpropanesulfonic acid) and polyacrylamide, in which sulfonic-acid groups stabilized proton activity and suppressed Mn³⁺ disproportionation. Although this work was not an ECD, it provided useful electrolyte-design principles for subsequent MnO₂-based ROEDs. Inspired by the advances in Zn-MnO₂ batteries, Wang *et al.*^[8] employed an optimized aqueous electrolyte containing 0.5 M H⁺, 80 mM Mn²⁺, and 80 mM Cu²⁺ ions, and a copper frame counter electrode to assemble ROEDs (Figure 4a). By applying a voltage, the deposited MnO₂ was dissolved in the acidic solution, thereby achieving bleaching on the FTO substrate. Subsequently, a multifunctional ROED was successfully realized, enabling independent modulation of the UV-VIS dual band. At wavelengths of 400 nm and 550 nm, the optical modulation amplitude (ΔT) reached 93.2% and 93.6%, respectively. The device exhibited multicolor changes including transparent, yellow, light brown, and dark brown states (Figure 4b), along with excellent optical memory and high energy storage characteristics.

However, ROEDs using copper-frame counter electrodes face several limitations. First, incomplete MnO₂ dissolution and electrochemically inactive manganese-containing residues gradually reduce cycling reversibility^[9,47]. In strongly acidic electrolytes, Mn³⁺ intermediates may undergo disproportionation or remain kinetically trapped at the electrode surface, producing residual MnO₂/MnOOH species that hinder complete bleaching and accelerate optical degradation. Second, because the counter electrode is confined to the device edge, ions must travel laterally toward the center, resulting in preferential edge deposition and prolonged switching times for large-area coloration. Third, many devices provide only fully bleached and fully colored states, with limited control over stable intermediate optical states.

To address the three issues mentioned above, researchers have carried out modification studies from two aspects: electrolyte optimization and device structure optimization.

With regard to the “dead manganese” residue problem, improvements can be achieved through the synergistic effect of chemical and electrochemical methods. Liu *et al.*^[9] applied the mechanism of Fe³⁺ mediator-facilitated MnO₂ dissolution, where 15 mM FeCl₃ was added as a mediator to a base electrolyte containing 0.5 M H₂SO₄, 1 M MnSO₄, and 15 mM CuSO₄. The redox potential of the Fe³⁺/Fe²⁺ couple is lower than that of the Mn⁴⁺/Mn³⁺ couple, thus accelerating the dissolution of “dead manganese” species (MnO₂ and MnOOH). During bleaching, Fe³⁺ is first reduced to Fe²⁺, and then Fe²⁺ accelerates the dissolution of MnO₂ and MnOOH by reducing them to Mn²⁺. Morphology observations showed that after 150 cycles in this modified electrolyte, no residual MnOOH deposits were detected on the working electrode surface (Figure 4c). Meanwhile, this electrolyte completely eliminated the decay of optical modulation amplitude. After 1,000 cycles, ΔT at 550 nm slightly increased from 44.8% to 46.9% (Figure 4d). Building on the introduction of Fe³⁺/Fe²⁺ to accelerate “dead manganese” dissolution, Zhang *et al.*^[47] focused on the effect of the mediator addition in the electrolyte on solution acidity. Highly acidic electrolytes may corrode FTO, and the reversibility remains limited. By comparing the coloration time, bleaching time, and transmittance variation under different H₂SO₄ and FeSO₄ concentrations, the study found that adding 10 mM Fe²⁺ to an electrolyte containing 0.5 M MnSO₄ allowed the required H₂SO₄ concentration to be reduced to 50 mM, while achieving high transmittance contrast (ΔT = 99.85% at 450 nm) and fast response (coloration time 24 s, bleaching time 26 s) (Figure 4e,f), with stable performance maintained after 500 cycles (Figure 4g). Lv *et al.*^[48] introduced a Br[−]/Br₃[−] redox couple into the CuBi/MnO₂ synergistic electrodeposition system (Figure 4h), which directly accelerated the deposition and dissolution processes of MnO₂ and shortened the device response time. The standard potential of the Br[−]/Br₃[−] couple is approximately 1.05 V (vs. standard hydrogen electrode [SHE]), slightly lower than that of the MnO₂/Mn²⁺ couple. This potential difference enables the Br[−]/Br₃[−] couple to activate the MnO₂ residues on the electrode surface. Cyclic voltammogram analysis confirms that in the presence of Br[−], the original single reduction peak split into two peaks, corresponding to faster ion-transport kinetics (Figure 4i). The dynamic transmittance spectra further confirmed that in the electrolyte containing Br[−], the response times for both coloration and bleaching processes of MnO₂ were shorter (Figure 4j).

To address the challenge of achieving stable and reproducible intermediate states between fully transparent and fully colored states, Ma *et al.*^[49] introduced an organic small-molecule viologen derivative, N,N'-bis(3-sulfonatopropyl)-4,4'-bipyridinium (SV), into the MnO₂ reversible electrodeposition system. By exploiting the complementary electrochemical behaviors of MnO₂/Mn²⁺ and SV, they achieved stepwise and precise regulation of the optical states. In a single electrolyte containing both Mn²⁺ and SV, the device could attain four stable optical states: transparent state, heat state (L-H state, low visible (VIS) transmittance, high near-infrared (NIR) transmittance), cool state (L-L state, low VIS/NIR transmittance), and block state (VIS/NIR transmittance nearly zero) by varying the applied external voltage (Figure 4k,l). This enables multilevel optothermal modulation from high transparency, heat insulation, and solar shading to full privacy, balancing energy efficiency and indoor comfort.

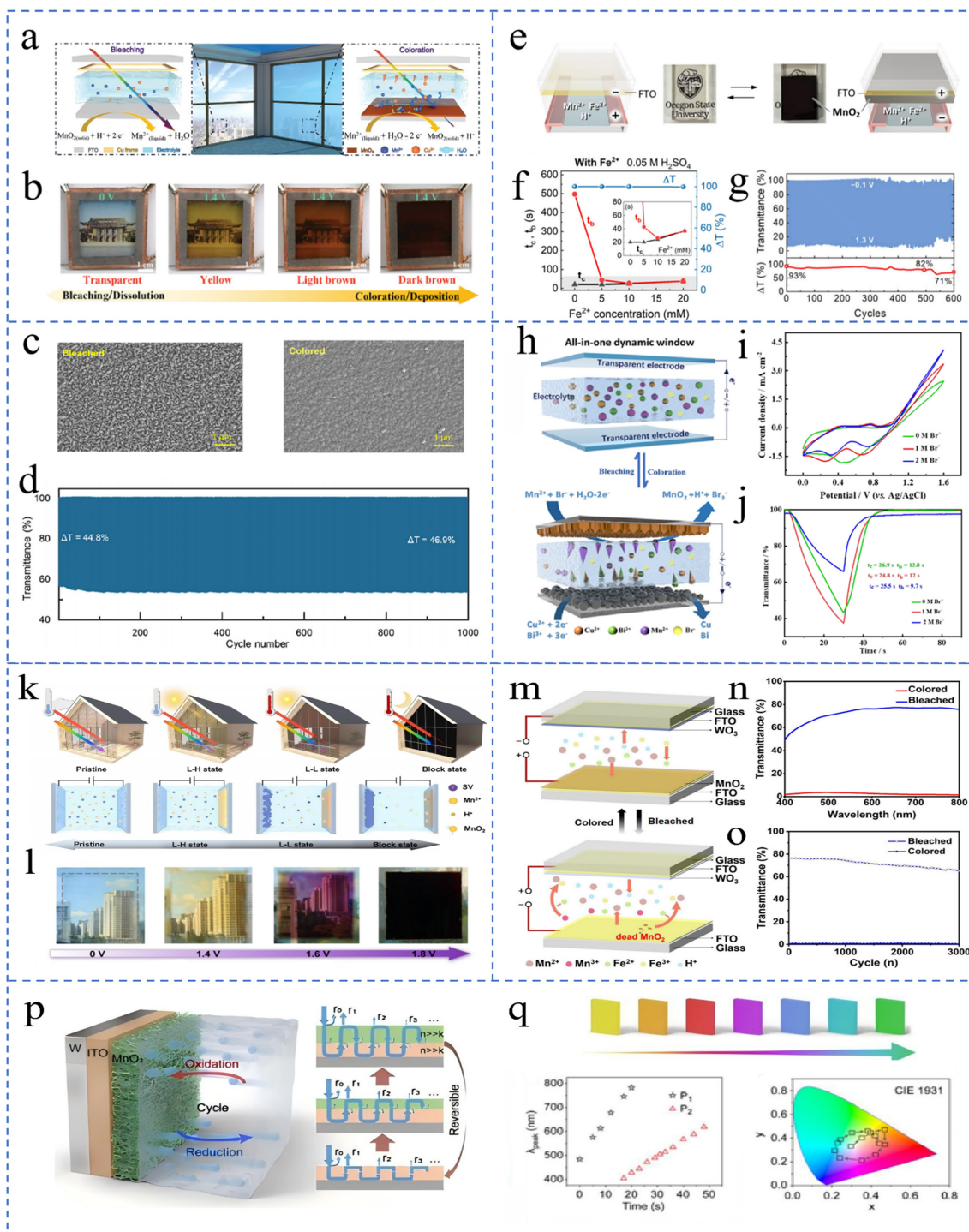


Figure 4. (a) Schematic illustration of the working principle of MnO_2 -based RED electrochromic process and the application scenario of smart windows in the building; (b) Digital photos of the device in the bleached state (transparent) and coloration states (yellow, light brown, and dark brown). Reproduced with permission from reference^[6]. Copyright © 2024 Wiley-VCH; (c) SEM images of the colored and bleached states; (d) Electrochromic performances of the ROE- MnO_2 system in the FeCl_3 -modified electrolyte: transmittance evolution curves at 550 nm during continuous color switching. Reproduced from reference^[49]. CC BY 4.0; (e) Electrochromic switching based on reversible electrodeposition and dissolution of $\text{MnO}_2/\text{Mn}^{2+}$ on an FTO electrode; (f) t_c , t_b , and ΔT vs. FeSO_4 concentration in the electrolyte with 0.5 M MnSO_4 and 0.05 M H_2SO_4 ; (g) Transmittance and transmittance change (ΔT) versus cycle number for long-term stability tests of the MnO_2 -based EC device. Reproduced with permission from reference^[47]. Copyright © 2024 American Chemical Society; (h) Schematic illustration of the working mechanism of the all-in-one dynamic window. (i-j) Electrochemical and electrochromic behavior of MnO_2 in different Br^- electrolytes; (i) CV curves in the potential range of 0 to 1.6 V (scan rate, 20 mV s^{-1}); (j) In situ dynamic transmittance spectra at 500 nm by applying the potentials of 1.4 V for 30 s and 0 V for 60 s. Reproduced with permission from reference^[48]. Copyright © 2026 Wiley-VCH; (k) Transmittance modulation of VIS and NIR light by four states. Changes in the amount of viologen derivative coloring and the amount of MnO_2 deposited in the four states; (l) Physical diagrams of four typical states of SV- MnO_2 devices and applied voltages. Reproduced

with permission from reference^[49]. Copyright © 2026 Wiley-VCH; (m) Schematic diagram illustrating the working principle of a complementary electrochromic window via proton insertion/extraction and ionic electrodeposition; (n) Transmittance spectra of the MnO_2 - WO_3 complementary device in bleached and colored states; (o) Transmittance variation at 670 nm over 3,000 cycles of periodic bleaching (0 V) and coloring (2 V) voltages. Reproduced with permission from reference^[10]. Copyright © 2024 American Chemical Society; (p) Structural diagram of the super-wide color tunable EC device. In situ formation and dynamic evolution of MnO_2 dielectric layer on the W/ITO electrode surface. The reconstruction process of optical cavity; (q) The achievement of seven primary colors. Variation of peak reflection wavelength λ_{peak} as a function of time. And each reflection spectrum's corresponding positions in CIE 1931 color space. Reproduced with permission from reference^[50]. Copyright © 2025 Wiley-VCH. FTO: fluoride-doped tin oxide; ITO: indium tin oxide; SV: N,N'-bis(3-sulfonatopropyl)-4,4'-bipyridinium; SEM: scanning electron microscope; ROE: reversible oxide electro-deposition/dissolution; RED: reversible electrodeposition; CV: cyclic voltammetry.

To address the issue of non-uniform MnO_2 deposition, device structure improvement can be adopted. Zhang *et al.*^[10] replaced the conventional copper-frame counter electrode in the reversible MnO_2 electrodeposition system with a facing parallel WO_3 thin-film electrode (Figure 4m), and used an acidic electrolyte containing Mn^{2+} and $\text{Fe}^{3+}/\text{Fe}^{2+}$ ions to suppress the Jahn-Teller distortion. This design combines the conventional ion-intercalation/deintercalation with the MnO_2 deposition/dissolution mechanism, realizing a complementary EC window. The facing electrode configuration facilitates proton intercalation/deintercalation into/from WO_3 , promotes vertical ion diffusion, shortens the ion diffusion path, and thus ensures uniform MnO_2 deposition. Consequently, large-area electrochromic devices ($10 \times 10 \text{ cm}^2$) were fabricated. The device achieves full modulation in the visible spectral range (400–800 nm) (Figure 4n), maintains a neutral color in the colored state, and exhibits high optical contrast ($\Delta T = 67.3\%$), fast response, and excellent long-term cycling stability (85.0% retention after 3,000 cycles) (Figure 4o). Moreover, because the complementary-structure electrolyte contains no copper ions, the initial transmittance of the device is higher.

The above strategies focus on optimizing the reversibility and uniformity of MnO_2 electrodeposition, yet the color change remains confined to the transparent-dark brown range. Tang *et al.*^[50] combined EC materials with optical resonant cavities. By exploiting the dynamic reconstruction of MnO_2 under electrical stimulation, they constructed Fabry-Pérot cavities on W/indium tin oxide (ITO) electrode surfaces, achieving a wide-gamut color change spanning yellow, orange, red, purple, blue, cyan, and green (Figure 4p,q). The electrodeposited MnO_2 acts as the dielectric spacer layer. As the duration of the applied voltage increases, the thickness of the MnO_2 layer gradually increases, altering the resonance condition of the optical cavity and causing the reflection spectral peak wavelength to continuously blue-shift or red-shift across the entire visible region. This device opens up a new direction for the application of MnO_2 electrodeposition in reflective displays.

MnO_2 -based ROEDs therefore offer a distinct combination of broad spectral modulation, multilevel color control, and energy-storage compatibility. Their principal advantage lies in the versatility of $\text{Mn}^{2+}/\text{MnO}_2$ conversion, whereas the main limitations originate from Mn^{3+} -related side reactions, acidic corrosion, residual deposits, and non-uniform ion transport. Redox mediators improve dissolution kinetics but increase electrolyte complexity; complementary electrodes shorten transport distances but add fabrication steps; and optical cavities enrich color output while requiring precise control of deposit thickness. These trade-offs should be considered together when selecting MnO_2 -based architectures for smart windows or multifunctional energy devices.

2.4 Comparison

Although RMEDs, RIEDs, and MnO_2 -based ROEDs involve different redox couples, their performance is governed by the same sequence of ion transport, interfacial reaction, deposit growth, and reverse dissolution. Electrolyte regulation controls solvation, free-active-species concentration, viscosity, acidity, and mediator chemistry; interface engineering controls nucleation, adhesion, and charge-transfer barriers; and device architecture determines transport distance, electric-field uniformity, and sealing requirements. Their mechanistic failures are nevertheless system-specific: dendritic or compositionally non-uniform metal growth limits RMEDs^[16,17], soluble polyiodides and electrically isolated iodine limit RIEDs^[6,37,38], and Mn^{3+} -related disproportionation and residual manganese oxides limit ROEDs^[9,47].

These differences define complementary application scenarios. RMEDs provide strong broadband attenuation, reflection regulation, and LSPR-enabled multicolor states, but require strict control of morphology and large-area current distribution. RIEDs offer neutral-color absorption and simplified complementary designs, but shuttle suppression and chemical compatibility are critical. MnO_2 -based ROEDs enable broad UV-visible modulation, multilevel states, and coupling with energy-storage reactions, although acidic electrolytes and residual deposits challenge durability. Table 1 summarizes these relationships and the corresponding mechanism-challenge-strategy pathways.

Performance should therefore be evaluated as a multidimensional balance rather than by optical contrast alone. In situ transmittance monitoring is essential for evaluating switching kinetics, optical-memory retention, cycling degradation, and edge-center uniformity. Ex situ scanning electron microscopy reveals deposit morphology, dendritic growth, and residual phases, whereas X-ray photoelectron spectroscopy can identify oxidation-state changes and irreversible surface species. For practical devices, these measurements should be combined with device-area, energy-consumption, temperature, humidity, UV-exposure, long-term storage, electrolyte-leakage, and sealing tests.

Table 1. Comparison of representative reversible electrodeposition-based electrochromic systems.

System	Reversible process	Optical characteristics	Principal advantages	Main challenges	Representative strategies	Representative reported performance	Application focus
RMED	M^{n+}/M plating and stripping	Broadband absorption or reflection; LSPR-dependent multicolor response	High optical contrast, neutral-color tinting, reflective regulation, optical memory	Dendritic growth, non-uniform nucleation, irreversible metal residues, large-area potential gradients	Alloy regulation, polymer additives, hydrogel electrolytes, pH control, interface modification	$T_{\min} = 0.01\%$, $\Delta T \approx 82\%$, $t_c/t_b = 8/18$ s ^[15] ; > 900 cm ² devices ^[16] ; $> 3,000$ cycles ^[17,18]	Smart windows, privacy control, displays, adaptive thermal management
RIED	I^-/I_2 and I^-/I_3^- conversion	Broadband iodine/polyiodide absorption and neutral-color darkening	Film-free configuration, high optical modulation, complementary coloration	Polyiodide shuttle, dead iodine, low deposit conductivity, sealing compatibility	WiSE, coordinating cations, liquid active adhesion phases, catalytic interfaces, complementary electrodes	$\Delta T = 76.0\%$ ^[6] ; $t_c/t_b = 6.9/11.9$ s and 20,000 cycles ^[38] ; 10×10 cm ² device ^[7]	Neutral-color windows and electrochromic energy-storage devices
MnO ₂ -based ROED	Mn^{2+}/MnO_2 deposition and dissolution	Broad UV-visible absorption, multilevel coloration, cavity-dependent reflective colors	Wide spectral modulation, multiple optical states, compatibility with energy storage	Mn^{3+} disproportionation, residual MnO ₂ /MnOOH, acidic corrosion, non-uniform lateral transport	Fe^{3+}/Fe^{2+} mediation, acidity regulation, Br^-/Br_3^- mediation, complementary electrodes, optical cavities	$\Delta T = 93.2\%/93.6\%$ at 400/550 nm ^[8] ; 10×10 cm ² and 85% retention after 3,000 cycles ^[10]	Smart windows, multicolor displays, electrochromic energy storage

The reported values were obtained under different wavelengths, voltages, switching protocols, electrolyte compositions, and device areas; direct numerical comparison should therefore be treated with caution. RMED: reversible metal electrodeposition-based device; RIED: reversible iodine electrodeposition-based device; ROED: reversible oxide electrodeposition device; UV: ultraviolet; WiSE: water-in-salt electrolyte; LSPR: localized surface plasmon resonance.

3. Conclusion

Reversible electrodeposition-based ECDs, including RMEDs, RIEDs, and MnO₂-based ROEDs, combine large optical modulation with simplified device configurations and diverse optical functions. Despite their different redox chemistries, their practical limitations arise from a common sequence of non-uniform interfacial reaction, incomplete dissolution, and transport inhomogeneity. In RMEDs, dendritic growth and uneven nucleation degrade optical uniformity and reversibility; in RIEDs, polyiodide migration and dead iodine weaken optical memory; and in ROEDs, Mn^{3+} -related reactions and residual manganese oxides impede complete bleaching. These problems are further amplified in large-area devices by electric-field gradients, long ion-transport paths, electrolyte leakage, and sealing failure. Practical evaluation should therefore balance optical contrast and color diversity against switching speed, cycling life, energy consumption, device area, wide-temperature operation, long-term storage, UV and humidity tolerance, and encapsulation reliability.

Future opportunities lie in coordinated electrolyte, interface, and architecture design. Solvation regulation, polymer or hydrogel confinement, coordinating ions, redox mediators, and catalytic interfaces can jointly improve deposition morphology and dissolution kinetics, while facing-electrode and complementary configurations can enhance large-area uniformity. At the application level, reversible electrodeposition is expanding from smart windows and displays toward adaptive radiative cooling/solar heating, electrochromic energy storage, and programmable spectral regulation. The integration of environmental sensing, feedback control, reliable encapsulation, and scalable manufacturing will be essential for transforming electrically switchable devices into practical intelligent and smart optical and thermal-management systems.

Declarations

Authors contribution

Ji Y, Xu Q: Writing-original draft.

Liu S: Writing-review & editing.

Xu B: Writing-review & editing, conceptualization, supervision.

Chen J: Writing-review & editing, conceptualization, supervision, project administration, funding acquisition.

Conflicts of interest

The authors declare no conflicts of interest.

Ethical approval

Not applicable.

Consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

Not applicable.

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